AIR QUALITY EXPERT GROUP

Nitrogen Dioxide in the United Kingdom

Prepared for:
Department for Environment, Food and Rural Affairs;
Scottish Executive; Welsh Assembly Government; and
Department of the Environment in Northern Ireland

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This is the first report produced by the Air Quality Expert Group.

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Terms of Reference

The Air Quality Expert Group was set-up in 2001, to provide independent scientific advice on air quality, in particular the air pollutants contained in the Air Quality Strategy (AQS) for England, Scotland, Wales and Northern Ireland and those covered by the EU Directive on ambient air quality assessment and management (the Air Quality Framework Directive). AQEG replaces the Airborne Particles Expert Group, who published their report on 'Source apportionment of airborne particulate matter in the UK' in January 1999.

AQEG reports to the Secretary of State for Environment, Food and Rural Affairs, Scottish Ministers, the National Assembly for Wales and the Department of the Environment in Northern Ireland (the Government and Devolved Administrations). AQEG is an advisory non-departmental public body in England, Wales and Northern Ireland. In terms of the Scotland Act 1998, the Group is a jointly established body.

AQEG's main functions are:

- to give advice to ministers on levels, sources and characteristics of air pollutants in the UK;
- to assess the extent of exceedences of Air Quality Strategy objectives and proposed objectives, EU limit values and proposed or possible limit values, where monitoring data is not available;
- to analyse trends in pollutant concentrations;
- to assess current and future ambient concentrations of air pollutants in the UK; and
- to suggest potential priority areas for future research aimed at providing a better understanding of the issues that need to be addressed in setting air quality objectives.

The Group will not give approval for products or equipment.

Further information on AQEG can be found on the Group's website at: http://www.defra.gov.uk/environment/airquality/aqeg/index.htm Information on these pages includes the dates, agendas, and minutes of meetings as they become available, a list of the members, the Register of Interests, and draft and final reports as they become available.

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Executive summary

This report provides a detailed analysis of nitrogen dioxide in the UK. It addresses:

- Emissions sources and their past and projected trends.
- Meteorological and chemical processes that affect ambient nitrogen dioxide concentrations.
- Measurement methods and networks.
- Modelling and mapping methods and their coupling with measurements at monitoring sites to predict future concentrations of nitrogen dioxide.
- Analysis of monitoring data and projections of future concentrations and likely exceedences of air quality objectives and limit values.

This report will not address health effects, indoor air pollution or effects on ecosystems or vegetation. The later was dealt with in the National Expert Group on Transboundary Air Pollution (NEGTAP) Report on Transboundary Air Pollution: Acidification, Eutrophication and Ground-Level Ozone in the UK, published in 2001.

EU Limit values and Air Quality Strategy Objectives for nitrogen dioxide (NO_2) are shown in the table below. They are primarily based on health effects. At relatively high concentrations, NO_2 causes inflammation of the airways. There is evidence to show that long-term exposure to NO_2 may affect lung function and that exposure to NO_2 enhances the response to allergens in sensitised individuals. The effects of NO_2 are related to the total exposure to the pollutant over the relevant averaging period, both the 1-hour and annual periods. This exposure will take place in a whole range of micro-environments, each with its own concentration pattern. UK objectives only apply where there is exposure (or likely future exposure) that relates to the relevant averaging period. For instance, for the annual mean NO_2 objective the relevant roadside location is the façade of a residential building, while for the 1-hour objective it would be the pavement of a street where people might regularly spend up to an hour, for instance a street with pavement cafes.

Legislation	**Hourly (µg m^{-3}) objective**	**Annual (µg m^{-3})**	**Achieve by**
EU First Daughter Directive (99/30/EC)	200 µg m^{-3} with up to 18 exceedences per year	40	2010
Air Quality Strategy (2000)	200 µg m^{-3} with up to 18 exceedences per year	40	2005

In addition, NO_2 contributes to the formation of ozone (O_3) and of secondary particles and is implicated in acidification and eutrophication. The National Emission Ceilings Directive establishes a national limit for each Member State for emissions of nitrogen oxides (NO_x) and other atmospheric pollutants. It incorporates (and in some cases goes beyond) ceilings established earlier under the *UNECE Convention on Long-Range Transboundary Air Pollution Gothenburg Protocol*. Under the Directive, the UK is committed to reducing its national annual emission of NO_x from 1728 kt as NO_2 in 2000 to below 1167 kt as NO_2 by 2010.

Emissions

- All combustion processes in air produce NO_x. NO_2 and nitric oxide (NO) are both oxides of nitrogen and together are referred to as NO_x. It is NO_2 which is subject to health based air quality standards.
- Overall emissions of NO_x have fallen by 37% from 2744 kt as NO_2 in 1990 to 1728 kt as NO_2 in 2000 due mainly to reductions in emissions from road transport and public power generation.
- Road transport is the largest source of NO_x emissions in the UK, contributing 49% of total emissions in 2000. However, emissions from road transport have fallen by 34% between 1990 and 2000 following improvements in engine design and the fitting of three-way catalysts to petrol cars, as a result of increasingly tight European vehicle emission standards.
- The contribution of road transport to NO_x emissions in urban areas is generally higher than the national average. Thus, total urban UK (and London only) road transport emissions of NO_x in 2010 are predicted to have declined by 49% (and 53%) from 1999 levels, respectively.
- UK NO_x emissions from all sources are projected to fall by a further 25% from 2000 levels by 2010. This is largely driven by a continuing decline in emissions from road transport as vehicles meeting tighter emission standards penetrate the UK fleet.
- NO_x emissions are primarily in the form of NO, but there is evidence for significant amounts of NO_2 emitted directly from the tailpipe of diesel vehicles, especially when slow moving, with levels possibly as high as 25% of total NO_x emissions in mass terms. These primary emissions have a significant impact on roadside NO_2 concentrations in areas where there is considerable diesel vehicle activity. The effect of new exhaust after-treatment technologies on primary NO_2 emissions from diesel exhaust needs to be monitored, especially catalytically regenerating traps used to reduce particulate emissions.
- Current evidence suggests that increasing the diesel car penetration rate in the UK fleet would lead to a small increase in NO_x emissions from road traffic in urban areas, but a more significant increase in primary NO_2 emissions. An increase in diesel car sales in 2010 from 22% of new car sales (close to the current rate) to 30% could increase urban UK road transport emissions of NO_x in 2010 by 0.7% and NO_2 emissions by 3%.
- The contribution to annual mean NO_x concentrations in 2001 from domestic and commercial emissions (primarily natural gas space heating) at background locations in central London is estimated to be about a quarter. Emissions from these sectors are expected to increase somewhat over the next 10 years or so across the UK, in contrast to the decline of emissions from traffic.
- Uncertainties in national emission estimates need to be continuously re-evaluated. Current assessments suggest a ±7% uncertainty in total UK NO_x emission estimates for 2000 at the 95% confidence level. However, uncertainties of emissions from specific sources and their spatial distribution are much higher than this and need to be carefully considered in any inventories developed for local air quality modelling.

Meteorological and chemical processes

- The concentration of NO_2 at a given location is determined by a combination of emissions, meteorology and chemistry.
- In addition to primary emissions sources, secondary NO_2 is formed in a rapid chemical reaction between primary NO and O_3. The availability of O_3 close to the emission source is a key factor determining the concentration of NO_2. Source receptor relations differ for primary and secondary pollutants and the distinction between primary and secondary NO_2 is an important policy issue.
- Meteorological processes operating on scales from a few metres to hundreds of kilometres play an important role in controlling dispersion and accumulation of NO_2 and the availability of O_3.
- Very low wind speeds, temperature inversions and a shallow, stable boundary layer are necessary conditions for winter NO_2 episodes. Summer episodes of NO_2 are associated with ozone episodes, that is with hot, still, sunny days.

Measurements

- The chemiluminescent analyser is widely used for continuous monitoring of NO_2 concentrations in the UK. It is the reference method specified in the EU First Air Quality Daughter Directive and is based on the chemiluminescent reaction between NO and O_3.
- Monitoring of NO_2 concentrations using the chemiluminescent analyser is carried out on a national scale, on behalf of Defra and the Devolved Administrations, and at a regional or sector-specific scale, on behalf of local authorities, the Highways Agency, the power generation industries, and other organisations. These networks are subject to rigorous quality assurance and quality control procedures, which ensure the reliability of the data. This report draws heavily on data from these networks.
- No measurement is exact, and all ambient concentration data has a degree of uncertainty, which is quantified using information about the calibration and other quality assurance/quality control (QA/QC) procedures applied. All the data used in this report is expected to meet the uncertainty requirements currently being set out in detail for the purposes of European legislation. This uncertainty will generally be less than for emission estimates, which rely on imperfect knowledge of other factors as well as actual measurements of emissions.

Modelling and mapping

- Monitored data represent measurements that are made only at specific locations at specific times. As such, these data alone provide limited information about the spatial extent of pollutant concentrations and how pollutant concentrations may change in the future. Policy requirements therefore necessitate some form of modelling to be undertaken to address these limitations. Modelling methodologies can be categorised as (i) empirically-based statistical models and (ii) physical process-based deterministic models. Deterministic models are based on mathematical formulations which describe the physical and chemical processes operating within the atmosphere, and which can be expressed at different levels of complexity. Some models combine both approaches. All of these model types have been used in this report, especially in the interpretation of monitoring data and in the projection of future concentrations of NO_2.

- There are a number of uncertainties that should be taken into account in assessing the policy conclusions concerning NO_2 and NO_x based on empirical, dispersion and other physically-based process models. Emission estimates are a crucial source of uncertainty, particularly the mass fraction of NO_x emitted as NO_2 and the likely future split between vehicle-kilometres travelled by petrol and diesel cars. Difficulties remain in representing urban influences on dispersion of primary NO_2 and in representing the 3-dimensional distribution of O_3 in urban areas which drives secondary NO_2 formation. Extra uncertainties occur when estimating concentrations at roadside.

NO_2 concentrations and recent trends

- Measured NO_2 concentrations during 2001 have been assessed for a total of 212 automatic monitoring sites. Data from 15 of these sites have been analysed in detail in this report. Annual mean NO_2 concentrations above 40 μg m^{-3} (the annual mean objective for 2005 and limit value for 2010) were measured at the majority of roadside and kerbside sites in London and at about half of the background sites. Exceedences outside London were largely confined to roadside and kerbside monitoring sites. The measurements show that the annual mean is the key assessment objective for NO_2.
- Annual mean concentrations of NO_x at urban background and urban centre sites have declined over the past decade, with NO_2 showing a far more modest fall at most sites. An assessment of rolling annual mean concentrations at 11 background and roadside sites in London since 1996 shows a decrease in NO_x concentrations to 70% of the 1996 values. NO_2 concentrations in 2001 were 88% of their 1996 values. These trends can be reconciled with the changes in emissions by a combination of the source apportionment of current measured concentrations and the trends in emissions in published emission inventories.
- An analysis of the roadside NO_2 concentrations has shown the key role of the emissions of primary NO_2 from traffic sources in determining annual mean NO_2 concentrations at such locations. Higher primary NO_2 is associated with a greater proportion of the emissions being from diesel vehicles, particularly in slow moving congested traffic.
- Annual mean NO_2 concentrations monitored at urban background sites in the United Kingdom are broadly comparable with those monitored across Europe. Proximity to traffic appears to be the major determining influence on the annual mean NO_2 concentrations monitored at over 1600 sites across Europe. Annual mean exceedences of 40 μg m^{-3} NO_2 are widespread in urban centres across Europe.
- The use of models enables the extension of an analysis based on results of monitoring data to the whole country. The results of dispersion modelling studies for 2001 and 1999 are generally consistent with measurements in terms of exceedences of 40 μg m^{-3}. Exceedences are currently widespread throughout the London conurbation in the immediate vicinity of heavily-trafficked roads. These areas of exceedence spread further into urban background locations and fill much of the space between the major road links (A roads and motorways) in the centre of London.
- Detailed dispersion modelling studies indicate that in 1999 annual mean NO_2 concentrations exceeded 40 μg m^{-3} in London over 34-68% of the total area and alongside 95-98% of major roads (at about 5–10 m from the edge of the road) . The national models suggest that in 2001 concentrations were above this level alongside 90% of major roads in London, 33% in the rest of England, 17% in Scotland and 5% in Wales and Northern Ireland.

- Episodes when hourly mean NO_2 concentrations exceed 200 µg m^{-3} can be classified into two types. Winter episodes are associated with poor dispersion of primary NO_x emissions. Under these circumstances, elevated NO_2 concentrations are derived from a combination of NO_x emitted as primary NO_2 and emitted as NO followed by reaction with oxygen (O_2), superimposed on the regional background oxidant level which results from the reaction of emitted NO with O_3. Summer episodes are characterised by the elevated secondary NO_2 concentrations associated with regional scale photochemical oxidant episodes.
- An analysis of the occurrence of NO_2 episodes since 1988 shows that such episodes have become less common, with summer episodes becoming relatively less important than winter episodes in terms of their contribution to the total number of hours with concentrations in excess of 200 µg m^{-3}.
- Northern hemisphere mean O_3 baseline concentrations have shown an upward trend at the Mace Head site in Ireland of 1 µg m^{-3} (0.5 ppb) per year averaged over the period from 1987–2001. An increase in the oxidising capacity of this magnitude of rural air entering urban areas could lead to increased urban NO_2 concentrations.

Projected NO_2 concentrations in 2005 and 2010

- Empirical and dispersion model studies indicate that emissions reductions will lead to a significant reduction in exceedences of 40 µg m^{-3} at the roadside and in background locations, both nationally and in London.
- The national modelling projections, based on current national policies, suggest that in 2005 an annual mean concentration of 40 µg m^{-3} is likely to be exceeded alongside 65% of major roads in London, 18% in the rest of England, 8% in Scotland, 3% in Northern Ireland and 2% in Wales. By 2010 this is expected to reduce to 23% of major roads in London, 5% in the rest of England, 2% in Scotland and zero in Northern Ireland and Wales. These projections are broadly confirmed by detailed dispersion modelling studies for London. Projections of the extent of exceedence in background locations are less consistent between different models, which predict exceedence areas in Greater London of between 5-32% for 2005 and 3-13% for 2010.
- Traffic sources and the total of domestic and commercial emissions are predicted to each contribute roughly one third of the total annual mean NO_x concentration by 2010 in central London. The remaining third includes contributions from regional rural concentrations, industry, other area and point sources.
- NO_x emissions reductions in 2005 and 2010 to levels well below those expected to be delivered by current national policies will be required if an annual average concentration of 40 µg m^{-3} is to be met at *all* locations including the most highly polluted hot-spots.
- The current relationships between NO_x and NO_2 concentrations will not hold into the future if baseline O_3 concentrations increase and/or if the mass fraction of NO_x emitted directly as NO_2 also increases, leading in either case to increased difficulties with achieving an annual mean concentration of 40 µg m^{-3}. These issues highlight the complexities of predicting future NO_2 concentrations, involving the interactions of influences at global, regional and local scales.

Conclusions

- Widespread exceedences of 40 µg m^{-3} NO_2 as an annual mean remain, despite a decade of decreasing NO_x emissions. Annual mean NO_2 concentrations show significantly smaller downward trends than NO_x concentrations because of both limitations on chemical production and increased direct emissions of NO_2.
- With current national policies some exceedences of the annual mean objectives and limit values for NO_2 will remain in the years 2005 and 2010, respectively. These will be less widespread than at present, but nonetheless still appreciable. If future traffic flows exceed expectations, or emission control technologies fail to deliver anticipated reductions, continued exceedences will be more widespread.
- There is strong evidence that heavy duty diesel vehicles make a major contribution to NO_x emissions from UK road transport and to direct NO_2 emissions. In the context of meeting the National Emissions Ceiling Directive target for NO_x emissions in the UK, attention will also need to be given to forecasts of activity in the power generating sector, particularly in the fuel mix used for electricity generation.
- There are reasons to believe that the current projections for future urban NO_2 concentrations may be optimistic. If northern hemisphere baseline O_3 concentrations continue to rise and influence rural O_3 concentrations in the UK, then the relationships between urban NO_2 and NO_x concentrations will alter resulting in higher than expected future annual mean NO_2 concentrations. Furthermore, if catalytically-regenerative particulate traps that are being retrofitted to diesel powered vehicles dramatically increase direct emissions of NO_2, as indicated by studies carried out in the USA, there will be further breaches of the air quality objective and limit value.
- NO_2 should not be considered in isolation from other pollutants. In developing air pollution controls, there may therefore be trade-offs between different pollutants, and it is a matter of concern that much of local air quality management is currently driven by exceedences of one air quality objective: the annual mean of 40 µg m^{-3} for NO_2. The Expert Group feels that a more flexible and holistic approach to air quality management might deliver more effective control strategies.

Chapter 1

Introduction

1. The Air Quality Expert Group, AQEG, was established to help the Government and Devolved Administrations tackle air pollution by examining sources and concentrations of pollutants in the UK. It was set up following the publication of the *Air Quality Strategy for England, Scotland, Wales and Northern Ireland* with the following functions:

- To advise the Government on levels, sources, and characteristics of air pollutants in the UK;
- To assess the extent of exceedences of existing and proposed Air Quality Strategy objectives and EU limit values, where monitoring data are not available;
- To analyse trends in pollutant concentrations;
- To assess current and future ambient concentrations of air pollutants in the UK;
- To suggest potential priority areas for research aimed at providing a better understanding of the issues that need to be addressed in setting air quality objectives.

2. The aim of this report, the first by the Group, is to address these issues with respect to nitrogen dioxide (NO_2).

1.1 Effects of NO_2

3. The Air Quality Strategy objectives for NO_2 are primarily based on health effects. At relatively high concentrations, NO_2 causes inflammation of the airways. There is evidence to show that long-term exposure to NO_2 may affect lung function and that exposure to NO_2 enhances the response to allergens in sensitised individuals. This report will not address the health effects of NO_2 or indoor air pollution. In addition, NO_2 contributes to the formation of ozone (O_3) and of secondary particles and is implicated in acidification and eutrophication. Nitrogen oxides (NO_x) are associated with damage to vegetation and an EU limit value has been set to protect vegetation[1]. However, no further considerations given to this aspect of NO_x in this report. The subject was dealt with in the National Expert Group on Transboundary Air Pollution (NEGTAP) Report on Transboundary Air Pollution: Acidification, Eutrophication and Ground-Level Ozone in the UK, published in 2001.

1.2 Emissions

4. All combustion processes in air produce oxides of nitrogen. Nitrogen dioxide (NO_2) and nitric oxide (NO) are both oxides of nitrogen and together are referred to as NO_x. It is NO_2 which is associated with adverse effects upon human health. Road transport accounts for about half of total UK emissions of NO_x, with further major contributions from the electricity supply industry and the industrial and commercial sectors. In London, road transport accounts for over two thirds (68%) of emissions. UK NO_x emissions have declined substantially since a peak in 1989, with overall reductions of 38% to 2000. The decline is mainly due to reductions from road transport and electric power generation, through the use of catalysts and low NO_x burners, respectively. Emissions are likely to decline further over the coming years as new technology and emissions standards continue to be introduced. Estimates of current and future emissions

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[1] Applies to vegetation more than 20 km from an agglomeration; and more than 5 km away from industrial sources regulated under Part A of the 1990 Environment Act, motorways and built up areas of more than 5,000 people.

are provided via emissions inventories, which allocate emissions to categories, for example, road transport and power generation, and also provide spatial maps. The National Atmospheric Emissions Inventory (NAEI) covers the whole of the UK, mapping emissions on a 1 km x 1 km spatial grid. There are, in addition, several local inventories for example, the London Atmospheric Emissions Inventory (LAEI).

5. NO forms the principal component of emitted NO_x, although primary emissions of NO_2 are significant. NO is converted into NO_2 in the atmosphere, mainly by reaction with O_3. The conversion depends on the amount of O_3 available, so that the NO_2 concentration, and its dependence on NO_x, depends on both meteorology (transport of O_3-containing air to the point of emission and of NO_x-containing air into the surroundings) and chemistry. As a result, the estimation of future concentrations of NO_2 based on predictions of NO_x emissions is not easy and relies on models. These models are largely based on either previous measurements of NO and NO_2 and the development of relationships between NO_2 and NO_x, for a range of locations (roadside, urban background, etc) or on simple chemical relationships.

1.3 Measurements of NO_2 and NO_x

6. The UK has an extensive national network of 85 automatic monitoring sites measuring NO_x, comprising the Automatic Urban and Rural Network (AURN) and some affiliated sites from the London Air Quality Network (LAQN). The AURN sites include both those operated for Defra and the Devolved Administrations and some owned and operated by other organisations, mainly local authorities. These sites all use chemiluminescent analysers to measure both NO and NO_2. In addition, there are several regional networks operated, for example, by the Highways Agency and the power generators. Extensive measurements have also been made using diffusion tubes. The collective measurements, some extending over more than 10 years, provide an invaluable resource with which to assess the temporal and spatial variations in NO_2 and NO_x, including exceedences of the air quality objectives. These data have been used extensively in this report.

Box 1.1. Units of measurement

Regulations in the United Kingdom have, until recently, expressed concentrations of nitrogen oxides (NO_x) in units of parts per billion (1,000,000,000) – ppb. This is the ratio of the number of nitrogen dioxide (NO_2) molecules, for example, to the total number of molecules in the volume of air of interest. These units have the advantage that if the temperature or pressure of the air changes, the ratio remains the same.

European legislation sets limit value concentrations as micrograms of pollutant per cubic metre of air ($\mu g\ m^{-3}$). When these units are used without correcting to a standard temperature and pressure, the same 'packet' of air will have a different concentration as these properties of the air change. The legislation specifies that the air volume must be corrected to be at a standard temperature of 293 K (about 20°C), and a pressure of 101.3 kPa (about average atmospheric pressure at sea level).

For these conditions the conversion factors between the two sets of units are:

for nitrogen dioxide (NO_2)	1 ppb = 1.913 $\mu g\ m^{-3}$
for nitrogen oxide (NO)	1 ppb = 1.248 $\mu g\ m^{-3}$.

When considering the sum of NO and NO_2 (defined as NO_x), it is desirable to express the concentration in a way that remains constant if any of the NO becomes oxidised to NO_2. One way to achieve this is to use ppb units, where the NO_x concentration in ppb is the sum of the NO and NO_2 concentrations in ppb. However, this is not the case if NO_x is given in $\mu g\ m^{-3}$ as the sum of the NO and NO_2 concentrations in $\mu g\ m^{-3}$.

When NO_x is given in units of $\mu g\ m^{-3}$ in this report, it is calculated as if all the NO has been oxidised to NO_2. This is done by adding the concentrations of NO and NO_2 as ppb, and converting the sum to $\mu g\ m^{-3}$ using the conversion factor for NO_2 given above. The unit is written *$\mu g\ m^{-3}$, as NO_2*. The same convention is required when reporting NO_x concentrations for the European legislation. Throughout this report NO_x in $\mu g\ m^{-3}$ is expressed as NO_2.

1.4 Modelling

7. While the monitoring sites have a wide spatial coverage, they can clearly provide only a scattered representation of pollutant concentrations – they do not, in themselves, provide a complete UK map of NO_2 concentrations. In addition, they cannot be used in a simple way to project future concentrations and to assess future exceedences, because of the complex nature of both the likely future emissions and the relationship between NO_x and NO_2. As a result, modelling is essential to assess the linkage between estimates of emissions and of atmospheric concentrations, to interpolate between monitoring sites to provide national maps, and to estimate future concentrations and exceedences. A fundamental approach, in which the meteorology and chemistry are modelled on all distance scales, from metres to many kilometres, using mathematical formulations, is not yet used for assessment purposes. An empirical approach, utilising statistical analysis of the large amount of monitoring data, coupled in some circumstances with modelling of dispersion, has been widely used in the UK and forms a central component of the analysis and projections presented in this report. It is coupled with surface concentration modelling in the generation of maps of NO_2 concentrations across the UK. More fundamentally-based or deterministic models also have an important role to play and are used in this report, particularly to test the assumptions implicit in the empirical projections.

1.5 Policy background

8. NO_2 emissions and concentrations in the UK are subject to a number of national and international agreements and initiatives. These are outlined below and are described in more detail in individual chapters, where appropriate.

1.5.1 International legislation and agreements

9. There are three EU Directives that relate directly to concentrations of NO_x in the air:

a) The *First Air Quality Daughter Directive* (1999/30/EC) which sets limit values for hourly and annual average NO_2 to be achieved throughout the Community by 1 January 2010.

b) The *National Emission Ceilings Directive* (2001/81/EC) (NECD) which establishes a national limit for each Member State for emissions of certain atmospheric pollutants including NO_x. The NECD incorporates (and in some cases goes beyond) ceilings established earlier under the *UNECE Convention on Long-Range Transboundary Air Pollution Gothenberg Protocol.* Under the Directive the UK is committed to reducing its national annual emission of NO_x to below 1167 kt by 2010.

c) The *Third Air Quality Daughter Directive* (2002/3/EC) which primarily establishes a target (rather than limit) value for O_3 but also requires monitoring for NO_2 and for O_3 precursors.

Table 1.1 EU Limit values and air quality strategy objectives for NO_2.

Legislation	Hourly (μg m^{-3}) objective	Annual (μg m^{-3})	Achieve by
EU First Daughter Directive (99/30/EC)	200 μg m^{-3} with up to 18 exceedences per year	40	2010
Air Quality Strategy (2000)	200 μg m^{-3} with up to 18 exceedences per year	40	2005

10. The *Auto-Oil Programme* introduced tighter European vehicle emission and fuel quality standards. In 1998 three Auto-Oil proposals for further standards from 2000 were agreed. The key elements to this agreement – now consolidated into two directives – are:

- stringent series of emission standards for cars and light vans, (known as the Euro III standard) to apply to all new vehicles sold from January 2001, and for heavy-duty vehicles from October 2001;
- more stringent standards applying from 1 January 2006 (Euro IV standards) and for heavy-duty vehicles from October 2006;
- tighter fuel quality specifications which apply to all petrol and diesel sold from 1 January 2000 and 2005; and
- a ban on the general marketing of leaded petrol from 1 January 2000.

11. For industrial plants, two additional directives provide (either directly or indirectly) controls on emissions of NO_x: The *Large Combustion Plant Directive* (2001/80/EC) establishes controls on emissions from large combustion plants, which include power stations, oil refineries and large energy producers in industry, particularly the metals sector.

12. The *Integrated Pollution Prevention and Control (IPPC) Directive* (Directive 96/61/EC) requires site specific permits, that take account of the characteristics of each installation, its location and the state of the local environment. A wide range of installations and environmental impacts are taken into account by the regulator in considering a permit application. Conditions must be included to address any transboundary pollution from the installation. The Directive requires individual industrial plants to take any measures necessary to comply with relevant European Community legislation.

1.5.2 National framework

13. *Part IV of the Environment Act 1995* required the Government to produce a *National Air Quality Strategy* containing standards and objectives and measures to achieve the objectives. The first edition of the Strategy, published in 1997, fulfilled that requirement. This was reviewed in 1999 and the *Air Quality Strategy for England*, *Scotland*, *Wales and Northern Ireland* was published in January 2000. The Act also laid the foundations for the system of local air quality management.

14. *Local Air Quality Management* (LAQM) forms an important part of the Government's strategy to meet both the UK air quality objectives and EU limit values. The strategy recognises that national measures, for example, controls on emissions from new motor vehicles, will not always be the most appropriate way to deal with localised hot-spots. LAQM requires all local authorities to carry out regular reviews and assessments of air quality in their area, with a view to identifying all those locations where one or more of the objectives is unlikely to be met by the relevant date, taking account of relevant exposure (Defra, 2003). Such locations have to be formally designated as Air Quality Management Areas (AQMAs) and an Action Plan prepared to move towards meeting the objectives. The LAQM process, now in its fifth year, has identified the need for 132 local authorities, out of 407 in England, Scotland and Wales (Northern Ireland is working to a different timetable) to declare air quality management areas. Of these, 92% are for NO_2, 45% for PM_{10}, mostly in combination with NO_2, and 4% for sulphur dioxide. The NO_2 declarations are all for road traffic, although in some cases in combination with a contribution from industry. NO_2 is thus the principal pollutant driving the LAQM process. These local studies of NO_2 have generated information that supplements the findings of the national studies, and both are drawn upon in this report. For instance, while the national studies are inevitably focused on London and other major conurbations, as well as on the most heavily trafficked roads, the LAQM studies have shown that exceedences can also occur in smaller towns, where narrow streets coincide with congested traffic.

15. The Greater London Authority Act received Royal Assent in November 1999. It provides for the Mayor of London to prepare and publish a *Mayor's Air Quality Strategy* (MAQS) for Greater London. London local authorities must take the MAQS into account when exercising their functions under Part IV of the Environment Act 1995.

16. The MAQS, published in September 2002, sets out how the Mayor intends to implement the national Strategy in Greater London and achieve the objectives in the relevant regulations. It also contains:

- information about the current and likely future air quality in Greater London;
- the measures which are to be taken by the Greater London Authority, Transport for London and the London Development Agency;
- information about measures the Mayor will encourage other people and organisations to take; and
- any other proposals and policies the Mayor considers appropriate.

17. *Part I of the Environmental Protection Act 1990* has been the main mechanism for minimising air pollution from industrial sources for a number of years. This Act established two pollution control regimes: the *Local Air Pollution Control* (LAPC) regime and the *Integrated Pollution Control* (IPC) regime, which not only took account of releases to air, but also to land and water.

18. IPC and LAPC are being succeeded by the new pollution prevention and control (PPC) regimes implementing the Integrated Pollution Prevention and Control (IPPC) EC Directive 96/61. There are separate regulations in Scotland and it will be for the new Executive to determine the arrangements in Northern Ireland. Although the terminology used differs slightly, the principles underpinning both IPC and PPC are similar. Regulators are required to ensure that pollution from industry is minimised through the use of the best available techniques, which takes into account the balance between the costs and environmental benefits. Both systems also require regulators to take account of the characteristics of each installation, its geographical location and local environmental conditions. In England and Wales, PPC is regulated by the Environment Agency and local authorities, in Scotland, by Scottish Environment Protection Agency (SEPA). In Northern Ireland, PPC is the responsibility of the Industrial Pollution and Radiochemical Inspectorate of the Environment and Heritage Service and district councils.

Box 1.2. Exposure

The effects of an air pollutant are related to the total exposure to the pollutant over the relevant averaging period. In the case of NO_2 these are both 1-hour and annual periods. This exposure will take place in a whole range of micro-environments, each with its own concentration pattern. Epidemiological health effects studies inevitably simplify the measure of exposure, usually characterising the exposure of a large population group using a relatively small number of monitoring sites. These studies form the basis, in large part, of the air quality standards to protect human health. The standards, when translated into legislation, usually make clear where they are to be applied. Thus the air quality regulations that bring into force the air quality objectives in the UK state that they are to be applied at locations:

a) which are situated outside of buildings or other natural or man-made structures above or below ground; and

b) where members of the public are regularly present.

This text still leaves the relevant locations at which to apply the objectives open to interpretation. To help deal with this, further guidance has been provided on where the objectives should be applied (Defra, 2003). Thus for the annual mean NO_2 objective the relevant roadside location is the façade of a residential building, while for the 1-hour objective it would be the pavement of a street where people might regularly spend up to an hour, for instance a street with pavement cafes. The guidance in particular makes clear that the UK objectives only apply where there is exposure (or likely future exposure) that relates to the relevant averaging period.

The EU Directive setting limit values for NO_2 specifies that the siting of monitoring stations should be in areas where 'the highest concentrations occur to which the population is likely to be directly or indirectly exposed for a period which is significant in relation to the averaging period of the limit value(s)'. There is further detail in the Annexes as to where to monitor, for instance, traffic orientated samplers should be at least 25 m from the edge of major junctions, with the sampler no more than 5 m from the kerb and no closer than 4 m from the centre of the nearest traffic lane, i.e. ~2 m from the kerb. Also, the sampling points should represent air quality in a surrounding area of no less than 200 m^2. It should be noted though that draft Guidance from the Commission suggests that there may be some ambiguity between the Framework and Daughter Directives as to where limit values apply, although it is assumed in the document that a pragmatic approach will be applied to assessing compliance, taking exposure into account (Guidance on Assessment under the EU Air Quality Directive, available at: www.europa.eu.int/comm/environment/air/ambient.htm)

The issue of where objectives and limit values apply may ultimately be clarified by way of a legal ruling. In the absence of this, the Expert Group adopts the pragmatic view that exposure is a key factor when assessing the results of monitoring and modelling in relation to compliance with both objectives and limit values.

1.6 Structure of the report

19. This report aims, for the specific case of NO_2, to fulfil the tasks placed on AQEG and outlined in the Introduction to this chapter. Chapters 2–5 review the main issues relating to (i) the construction of emissions inventories, (ii) the measurement of NO and NO_2, (iii) the impact of meteorology and chemistry on NO_2 concentrations and (iv) modelling and mapping of NO_2 concentrations. Where possible and appropriate, we have included a discussion of fundamental as well as practical issues.

20. Chapter 6 provides a detailed analysis of data and an examination of historic trends. Data on annual mean NO_2 were assembled for 212 sites and are provided, with appropriate additional data, on the AQEG website (http://www.defra.gov.uk/environment/airquality/aqeg/index.htm). The data are used to examine exceedences and to construct both background and roadside national maps of NO_2 concentrations for 2001. In addition, 15 sites, covering all site-types, were selected for more detailed examination of hourly data. This analysis facilitates an optimisation of the empirical models and the development of a more detailed understanding of, for example, the linkage between NO_2 and traffic flow and the effects of meteorology. The chapter also includes an analysis of both wintertime and summertime episodes and an examination of European data to provide a European context.

21. In Chapter 7 projected concentrations for 2005 and 2010 are calculated and examined. Site specific projections are made for the 15 sites selected for detailed analysis, while simple projections of annual means are made for all of the sites and national maps constructed. Particular attention is focused on London, because of the wealth of data and the comparatively high incidence of exceedences of the annual mean limit value. This analysis also facilitates an understanding of the implications of national projections in other cities. Trajectory models are employed to examine the impact of policy measures and to test the effects of global issues, such as the increase observed in background tropospheric O_3 and the likely impact of climate change. Each chapter provides a list of key points and conclusions.

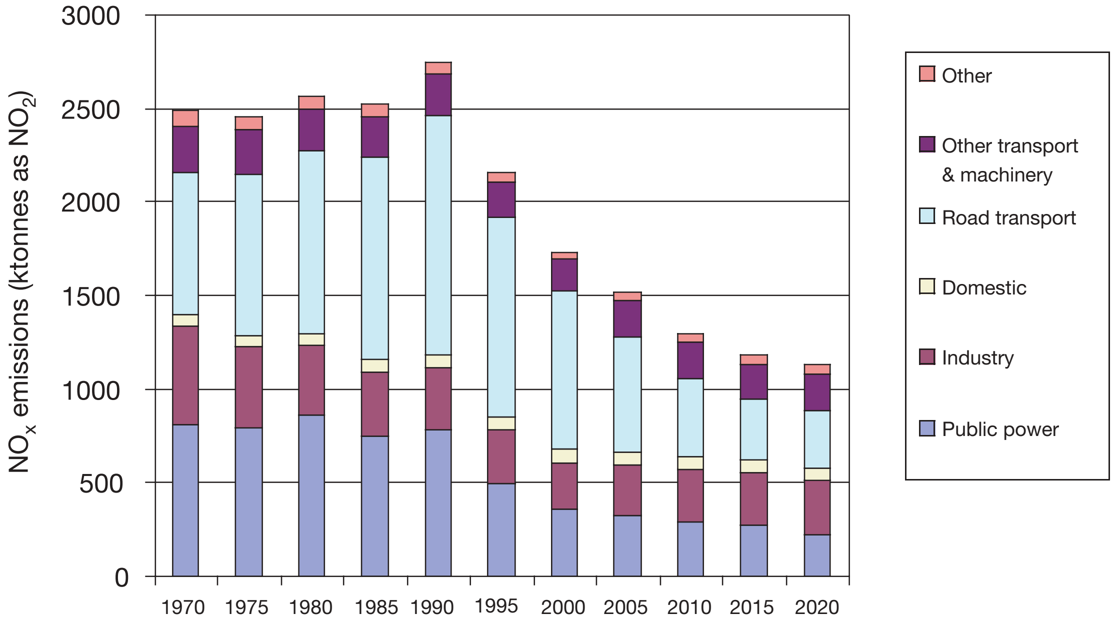
Chapter 2

NO$_x$ emissions and emission inventories

Key points

Figure 2.1 shows the time series of NO$_x$ emissions in the UK from 1970 projected to 2020.

Figure 2.1 NO$_x$ emissions in the UK by source from 1970–2020.



- Emissions of NO$_x$ have fallen by 37% from 1990 to 2000. This has been mainly due to reductions in emissions from road transport and public power generation.
- Road transport is the largest source of NO$_x$ emissions in the UK, contributing 49% of total emissions in 2000. However, emissions from road transport have fallen by 34% between 1990 and 2000 due to improvements in engine design and fitting of three-way catalysts to petrol cars driven by increasingly tighter European vehicle emission standards.
- The contribution of road transport to NO$_x$ emissions in urban areas is generally higher than the national average. In London, 68% of NO$_x$ emissions come from road transport.
- Total UK NO$_x$ emissions are projected to fall by a further 25% from 2000 levels by 2010. This is largely driven by a continuing decline in emissions from road transport as vehicles meeting tighter emission standards penetrate the UK fleet.
- Emissions of NO$_x$ from road transport in London are predicted to decline by 53% from 1999 levels by 2010. This compares with a predicted decline of 49% in emissions from total urban UK road transport emissions over the same period. Little, if any, decline is expected in emissions from other sources in London.

- The total UK NO_x emission projections depend critically on assumptions based on traffic growth, energy demand and fuel mix for electricity generation, as well as emission performance of new vehicles, especially heavy duty diesel vehicles, and implementation of the EC's Large Combustion Plant Directive. The assumptions underlying the emission projections will need to be continuously monitored in order to ensure the UK complies with the emission targets for 2010 set in the Gothenburg Protocol and National Emissions Ceilings Directive.
- Heavy duty diesel vehicles currently emit 43% of NO_x emissions from UK road transport, but these figures are based on relatively few real world emission tests on these vehicles. Evidence suggesting that real-world NO_x emissions from HGVs and buses have changed little during the 1990s from pre-Euro I to Euro II standards needs verification with further tests.
- Local and spatially resolved inventories help identify pollution 'hot spots' and provide vital input data to atmospheric dispersion models. Considerable advances have been made in spatially resolving emission estimates. Local inventories need to be regularly updated and developed using a consistent approach.
- Uncertainties in national emission estimates need to be continuously re-evaluated. Current assessments suggest a ±7% uncertainty in total UK NO_x emission estimates for 2000 at the 95% confidence level. However, uncertainties of emissions from specific sources and their spatial distribution are much higher than this and need to be carefully considered in any inventories developed for local air quality modelling.
- There is evidence for significant amounts of NO_2 emitted directly from the tailpipe of diesel vehicles, with levels possibly as high as 25% of total NO_x emissions in mass terms. This will have a significant impact on roadside NO_2 concentrations in areas where there is considerable diesel vehicle activity and this needs to be taken into account in modelling of ambient concentrations. The effect of new exhaust after-treatment technologies on primary NO_2 emissions from diesel exhaust needs to be monitored, especially catalytically regenerating traps used to reduce particulate emissions.
- Current evidence suggests that increasing the diesel car penetration rate in the UK fleet would lead to a small increase in NO_x emissions from road traffic in urban areas, but a more significant increase in primary NO_2 emissions. An increase in diesel car sales in 2010 from 22% of new car sales (close to the current rate) to 30% would increase urban UK road transport emissions of NO_x in 2010 by 0.7% and NO_2 emissions by 3%.

2.1 Introduction

22. Almost all of the NO_x emitted to air in the form of NO and NO_2 are from combustion sources. Collectively, these two species are referred to as NO_x ($NO_x = NO + NO_2$). Combustion sources may also emit small quantities of nitrous acid (HONO) which is also photochemically active in the atmosphere. This chapter discusses the main sources of NO_x emissions and the fundamentals of NO_x formation in combustion systems. It also discusses some of the main abatement options that are used in the UK to control NO_x emissions from different sources. Different technologies for reducing NO_x emissions are available for both stationary and mobile sources. These are discussed in the context of the regulatory framework setting limits on emissions from specific sources in the UK and Europe as well as the national emission targets which countries in Europe must achieve by the year 2010.

23. Emission inventories are an important means of quantifying emissions of NO_x from different sources in different locations. These provide the necessary input data for atmospheric models predicting NO_2 concentrations and photochemical activity in general at different geographical scales. Emission inventories are produced at European, national and regional and local levels using common methodologies, but using information at different levels of geographical detail. The principles and methodologies behind the inventories compiled in the UK are discussed, with particular attention given to the sources of information used to compile the inventories from mobile and stationary combustion sources. The time series of NO_x emissions in the UK from 1970 to 2000 is shown, broken down by emission source sector. This information is vital for explaining the trends in emissions over the past 30 years and for directing future policy making in areas that will affect emissions and air quality in future years.

24. Some of the principles behind the development of spatially resolved local inventories and differences that occur with the national emission maps are discussed. The national inventory for the UK is currently mapped on a 1x1 km spatial scale. Both this and more detailed local inventories play a key role in local policy making and understanding of ambient concentrations of NO_x and NO_2. Local inventories produced for different areas of the UK are discussed, with particular attention given to the London Atmospheric Emissions Inventory. These play a key role in local scale modelling of air quality in the UK.

25. It is important to understand the nature and extent of uncertainties in the NO_x emission estimates, both in individual sources and in the overall UK inventory, if the results are to be compared each year and the potential impact of measures to reduce emissions are to be understood in terms of their impact on ambient concentrations. A rigorous, quantitative analysis of the uncertainties in the overall national emission estimates has been undertaken using statistical methods. The results will help to prioritise future work by pointing to those areas where further information is required to improve the reliability of the inventory. Uncertainties in emission estimates in local areas are of more consequence to local air quality modelling of pollutant concentrations and are usually of greater magnitude than the uncertainties in the national emission totals. Consideration is given to the factors influencing uncertainties in modelling the spatial distribution of emissions.

26. Emission projections are used to inform Government policy and to forecast the likelihood of attaining the UK's emissions targets for 2010. They are one of the principal drivers in forecasting the UK's air pollution climate and achievement of air quality objectives. The UK's base emission projections for NO_x take account of current Government policies, European Directives and Regulations on emissions, technology improvements and current understanding of future growth in energy demand, transport and industrial activity. The base projections for UK emissions in 2005, 2010, 2015 and 2020 are shown, as well as emissions forecasts for specific urban areas like London. The UK projections for 2010 are put into context with the country's emission targets set in terms of the National Emissions Ceilings Directive. Road transport is a major contributor of NO_x emissions in urban areas and illustrative transport scenarios have been assessed to highlight the sensitivity of projected urban NO_x emissions to key transport factors.

27. Combustion sources emit NO_x mostly in the form of NO. However, for some sources, especially road vehicles, direct emissions of NO_2 appear to be significant. This chapter points to evidence for direct NO_2 emissions from road vehicles based on tailpipe emission measurements. There may also be evidence for direct emissions of HONO from vehicles. The conditions and technologies that favour NO_2 emissions from vehicle exhausts are discussed. The extent of primary NO_2 emissions from other sources such as stationary combustion has not been considered as no new evidence of these as significant sources of primary NO_2 has emerged.

28. Combustion sources can also emit NO_x in the form of nitrous oxide (N_2O). This is an inert and relatively harmless compound which takes no part in ground-level photochemical activity. However, there are concerns over its emissions because it is a potent greenhouse gas implicated in potential global climatic changes. Some abatement technologies designed to reduce NO_x emissions can lead to higher emissions of N_2O as a by-product of the NO_x control process. This report is not primarily concerned with N_2O, but the secondary effects of NO_x control measures on emission projections of N_2O in the UK are briefly discussed. Similarly, the possible secondary effects that certain NO_x abatement technologies may have on ammonia (NH_3) and primary NO_2 emissions are also briefly discussed.

2.2 NO_x formation in combustion sources

29. Combustion of fossil fuels is by far the dominant source of NO_x emissions. There are three main mechanisms by which NO_x is formed in combustion systems (Miller and Bowman, 1989). Thermal-NO and prompt-NO mechanisms produce NO_x by the high temperature oxidation of elemental nitrogen present in the combustion air. Fuel-NO is formed from nitrogen chemically bound in certain fuels. High temperatures and oxidation-rich conditions generally favour NO_x formation in combustion.

2.2.1 Thermal-NO mechanism

30. Formation of NO by the thermal-NO mechanism is initiated by the reaction of O atoms with N_2:

$$O + N_2 \rightarrow N + NO$$

$$N + O_2 \rightarrow O + NO$$

$$N + OH \rightarrow H + NO$$

31. This process is also known as the Zel'dovich mechanism (Zel'dovich, 1946). The initial step is the rate determining step in the mechanism, influencing the amount of NO which gets formed. O atoms are abundant in combustion systems under oxygen rich and near-stoichiometric conditions, but the rate of the reaction with N_2 is highly dependent on temperature, reflecting the high activation energy of the $O + N_2$ reaction. The reaction is extremely slow at low combustion temperatures, but increasing the temperature rapidly increases the rate of the reaction. For example, increasing temperature from 1200°C to 2000°C increases the rate of this reaction by a factor of ten thousand (Baulch *et al*., 1994). Hence, controlling combustion temperature in the flame is one means of controlling the amount of NO_x which is produced. NO forms by this process in both the flame front itself and in the postflame gases (Heywood, 1988). However, NO formation in the hot postflame gases usually dominates any flamefront-produced NO.

2.2.2 Prompt-NO mechanism

32. The prompt-NO mechanism forms NO from nitrogen much earlier in the flame than the thermal-NO mechanism, as its name suggests. The mechanism is initiated by the reaction of CH radicals with N_2:

$$CH + N_2 \rightarrow HCN + N$$

33. Both N and HCN react rapidly with oxidant to form NO in the flame. CH radicals are formed as intermediates in the combustion of hydrocarbons, but compared with O atoms, only small concentrations of CH radicals are formed. However, the reaction of CH with N_2 is much faster at low temperatures and the rate shows a much lower dependence on temperature due to its lower activation energy. Hence, they lead to NO being formed earlier in the flame (Miller and Bowman, 1989; Fenimore, 1971; Hayhurst and Vince, 1980).

34. Overall, the amount of NO formed by the prompt-NO mechanism is small compared with the amount formed by the thermal-NO route because of the lower abundance of CH radicals in the flame. However, it makes a significant contribution under fuel-rich conditions when higher concentrations of hydrocarbon radical species like CH are present.

2.2.3 Fuel-NO mechanism

35. Certain fuels contain appreciable amounts of nitrogen chemically bound in the fuel. For example, coal contains 0.5 to 2% nitrogen by weight chemically bound in large heterocyclic compounds with pyridine and pyrrole-type structures and aromatic amines (Solomon and Colket, 1978). A fraction of the nitrogen compounds is released into the gas-phase through the process of devolatilisation when the coal particles are heated. As they are released, the fuel-nitrogen undergoes rapid pyrolysis to form small nitrogen compounds such as hydrogen cyanide (HCN), CN radicals and ammonia (NH_3). These species are then rapidly converted to NO_x by a complex sequence of reactions involving the small gas-phase radicals (O, OH, H) produced in the main combustion process (Miller and Bowman, 1989). Additional NO_x is produced by heterogeneous oxidation of the nitrogen remaining in the coal char particles (Pershing and Wendt, 1977; Wendt, 1980). Unless combustion conditions are controlled, the fuel-NO mechanism leads to very significant amounts of NO_x being formed in the flame. The nitrogen content of petroleum-based fuels is usually lower then in coal, but varies significantly from less than 0.05% in light distillates (for example, diesel) to 1.5% in some heavy fuel oils. Fuel nitrogen is virtually absent in gaseous fuels. Consequently, the amount of fuel-NO produced in gas combustion is negligible compared with the amounts produced in coal combustion.

2.2.4 Formation of NO_2 in combustion

36. Nitrogen oxides formed in combustion systems by these mechanisms are mainly released in the form of NO. As a general rule of thumb, it is assumed that about 5% of NO_x released from combustion is in the form of NO_2, although this assumption for many sources still remains poorly quantified. While this is usually the case in many systems, there are certain combustion conditions that can lead to higher proportions of NO_x emitted as NO_2. There is also evidence that small amounts of nitrous oxide (N_2O) and nitrous acid (HONO) are emitted from combustion systems (Miller and Bowman, 1989, and references therein; see also Section 2.6.2.7).

37. NO_2 is readily formed in combustion systems if the conditions are favourable. A combustion system that has pockets of gas at low temperatures under oxygen rich conditions are those conditions which favour formation of NO_2.

38. NO formed in the high temperature flame zone can be rapidly converted to NO_2 via the reaction:

$$NO + HO_2 \rightarrow NO_2 + OH$$

39. However, HO_2 is only abundant in low temperature combustion regions and any NO_2 which is formed is readily removed by reaction with other combustion radicals, for example, O and H, and converted back to NO:

$$NO_2 + O \rightarrow NO + O_2$$

40. Hence, in combustion systems that are well-mixed, most of the NO_x remains as NO. This is generally the case for burners and spark-ignition engines. However, in compression-ignition diesel engines, combustion does not generally occur in such a well-mixed environment and cooler regions exist which quench the combustion chemistry (Heywood, 1988). Then, NO formed in the flame may react to form NO_2, but the conversion of NO_2 back to NO is quenched in the cooler regions. It has been previously found that the highest NO_2/NO ratio occurs in a diesel engine at light load, where cooler regions are more widespread. Early engine research showed that as much as 30% of NO_x is emitted as NO_2 from a diesel engine at low engine speed and light load (Hilliard and Wheeler, 1979). The oxygen rich environment of combustion in a diesel engine also aids formation of NO_2.

41. If NO concentrations are high in the presence of excess oxygen downstream of the engine or burner in the tailpipe or flue gas, NO_2 can be formed by the reaction:

$$NO + NO + O_2 \rightarrow 2\ NO_2$$

42. The rate of this reaction is quite slow at the NO concentration levels found in the exhaust pipe and has a weak negative temperature dependence (Atkinson and Lloyd, 1984). To illustrate this point, for typical NO concentrations of 500 ppm found in the presence of excess oxygen in the tailpipe of a heavy duty diesel vehicle, it would take at least 5 seconds for 1% of the NO to be converted to NO_2 by this reaction. This is a conservative lower limit on the NO conversion time. It should also be noted that the thermodynamics favours NO over NO_2 at higher temperatures.

2.3 Emission sources and regulatory framework

43. The major emission sources of NO_x in the UK are combustion for public power generation, combustion in industry and road transport. These were responsible for 82% of all UK NO_x emissions in 2000 and it is in these areas where most effort has been undertaken in reducing emissions (Goodwin *et al*., 2002).

44. Reductions in emissions of NO_x from stationary combustion and mobile sources have been driven by various national regulations and European Directives covering different emission sources combined with international protocols aimed at achieving emission targets in each country for future years.

45. Emissions from stationary combustion plant in the UK have been regulated under the Environmental Protection Act 1990. Integrated Pollution Control (IPC) is a system established under Part I of the Act to control pollution from industry. It applies to Part A processes which are potentially the most polluting or technologically complex processes in England and Wales and is enforced by the Environment Agency. A parallel, but separate, system of IPC is used in Scotland and enforced by the Scottish Environment Protection Agency, SEPA. IPC is concerned with the release of polluting substances to air, land and water. Part B processes, also covered under the Environmental Protection Act, refer to less polluting, less complex processes and these are regulated by local government. In Scotland, Part B processes are being regulated by SEPA under the Pollution Prevention and Control (Scotland) Regulations 2000 (PPC). Although less important in a national context, these processes can be influential in a local context.

46. Directives regulating emissions from new petrol passenger cars have been around since the 1970's, but these focused on CO and hydrocarbon emissions. It is only since the early 1990's that tough standards on NO_x emissions from new cars sold in Europe were introduced. This first came about with EU Directive 91/441/EC which effectively mandated the fitting of three-way catalysts to all new petrol cars to significantly reduce emissions of CO, hydrocarbons and

NO_x (Official Journal, 1991). This Directive set limits on the sum of hydrocarbon and NO_x emissions, rather than NO_x itself, but it did lead to very substantial decreases in NO_x emissions. Standards for this Directive, frequently referred to as Euro I, were followed by Euro II standards implemented by Directive 94/12/EC during the mid-90s. Yet more stringent EU Directives have been put in place to reduce NO_x emissions further, the most recent of these (98/69/EC) setting emission limits on NO_x itself for petrol cars sold after 2000 and then after 2005 (Euro III and Euro IV standards, respectively (Official Journal, 1998)).

47. NO_x emissions from diesel vehicles have also been regulated since the early 1990's (since 1988 for heavy duty diesel vehicles). These have been tightened up with the introduction of a succession of more stringent EU Directives, currently extending to tougher limits on emissions from heavy duty vehicles sold after 2008 (Official Journal, 1999). However, none of the emission reductions required to meet the tighter standards on diesel vehicles have been as dramatic as the 91/441/EC Directive was on petrol cars in the early 1990s.

48. Table 2.1 shows the limit values on NO_x, hydrocarbons (HC) and particulate matter (PM) set by the various Directives for the different vehicle types since 1990. The limit values for each Euro standard refer to tests over specified vehicle or engine test cycles, some of which have changed in recent years in order to make the test more representative of the performance of the vehicle on real road conditions.

49. As well as regulations on emissions from specific sources, the UK is subject to certain national emission targets to be achieved by 2010. In the mid-1990s, the UNECE started negotiating a multieffect, multipollutant protocol on NO_x and related substances. This was aimed at addressing photochemical pollution, acidification and eutrophication. The Protocol to Abate Acidification, Eutrophication and Ground-Level Ozone was adopted in Gothenburg in December 1999, where it was signed by the UK. The Gothenburg Protocol set emissions ceilings for nitrogen and sulphur oxides, ammonia and NMVOCs to be achieved by each country by 2010. The Gothenburg Protocol forms a part of the Convention on Long-Range Transboundary Air Pollution (UNECE, 1999).

50. Within the EU, the National Emissions Ceilings Directive (NECD) set emissions ceilings for 2010 for each Member State for the same four pollutants as in the Gothenburg Protocol (Official Journal, 2001). A number of Member States reduced their ceilings somewhat below the levels included in the Protocol. The UK reduced its ceiling for NO_x emissions from 1181 ktonnes (as NO_2) set in the Gothenburg Protocol to 1167 ktonnes for the NECD.

51. Within the UK, the implementation of the EC's Large Combustion Plant Directive and other associated policy measures have led to a substantial reduction in NO_x from power plant and industrial sources. This, combined with further stringent vehicle emission and fuel quality Directives being implemented over the next 10 years, will help the UK towards meeting its emission target. Integrated Pollution Prevention and Control (IPPC) is a system following the European Community Directive (96/91) which will introduce a more integrated approach to controlling pollution from industrial sources across the UK.

52. Section 2.7 provides a more detailed account of the inventory of NO_x emissions in the UK by sector and trends over time. Since 1990, emissions from public power generation, combustion in industry and road transport have been reduced by 49%. In the power generation and industrial combustion sectors, the reductions have been partially achieved through the switch from coal to gas. However, emission abatement technologies in remaining coal plant, gas turbines and road vehicles have made a significant contribution to these emission reductions.

Table 2.1 Emission limit values for different vehicle types in Europe since 1990. Dates of implementation refer to new registrations.

	Directive		Date of implementation		Test cycle	Units	NO$_x$*	HC*	NO$_x$ + HC*	PM*
Petrol cars	91/441/EEC	Euro I	1/7/1992		ECE15 + EUDC	gkm^{-1}			0.97	
	94/12/EC	Euro II	1/1/1997		ECE15 + EUDC	gkm^{-1}			0.50	
	98/69/EC	Euro III	1/1/2001		ECE15 + EUDC	gkm^{-1}	0.15	0.20		
	98/69/EC	Euro IV	1/1/2006		ECE15 + EUDC	gkm^{-1}	0.08	0.10		
Diesel cars	91/441/EEC	Euro I	1/7/1992		ECE15 + EUDC	gkm^{-1}				0.14
	94/12/EC	Euro II	1/1/1997	IDI	ECE15 + EUDC	gkm^{-1}			0.70	0.08
				DI	ECE15 + EUDC				0.90	0.10
	98/69/EC	Euro III	1/1/2001		ECE15 + EUDC	gkm^{-1}	0.50		0.56	0.05
	98/69/EC	Euro IV	1/1/2006		ECE15 + EUDC	gkm^{-1}	0.25		0.30	0.025
Petrol vans	96/69/EEC	Euro II	1/1/1998	<1305kg	ECE15 + EUDC	gkm^{-1}			0.50	
			1/1/1998	1305 – 1760kg					0.65	
			1/1/1998	>1760kg					0.80	
	98/69/EC	Euro III	1/1/2001	<1305kg	ECE15 + EUDC	gkm^{-1}	0.15	0.20		
			1/1/2002	1305 – 1760kg			0.18	0.25		
			1/1/2002	>1760kg			0.21	0.29		
	98/69/EC	Euro IV	1/1/2006	<1305kg	ECE15 + EUDC	gkm^{-1}	0.08	0.10		
			1/1/2006	1305 – 1760kg			0.10	0.13		
			1/1/2006	>1760kg			0.11	0.16		
Diesel vans	93/59/EEC	Euro I	1/10/1994	<1305kg	ECE15 + EUDC	gkm^{-1}			0.97	0.14
			1/10/1994	1305 – 1760kg					1.40	0.19
			1/10/1994	>1760kg					1.70	0.25
	96/69/EEC	Euro II	1/1/1998	<1305kg	ECE15 + EUDC	gkm^{-1}			0.60	0.10
			1/1/1998	1305 – 1760kg					1.10	0.15
			1/1/1998	>1760kg					1.30	0.20
	98/69/EC	Euro III	1/1/2001	<1305kg	ECE15 + EUDC	gkm^{-1}	0.50		0.56	0.05
			1/1/2002	1305 – 1760kg			0.65		0.72	0.07
			1/1/2002	>1760kg			0.78		0.86	0.10
	98/69/EC	Euro IV	1/1/2006	<1305kg	ECE15 + EUDC	gkm^{-1}	0.25		0.30	0.025
			1/1/2006	1305 – 1760kg			0.33		0.39	0.04
			1/1/2006	>1760kg			0.39		0.46	0.06
Heavy-duty vehicles	91/542/EEC	Euro I	1/10/1993		ECE R49	gkW^{-1}h^{-1}	8.0	1.10		0.36
	91/542/EEC	Euro II	1/10/1996		ECE R49	gkW^{-1}h^{-1}	7.0	1.10		0.15
	99/96/EC	Euro III	1/10/2001		ESC + ELR	gkW^{-1}h^{-1}	5.0	0.66		0.10
					ETC	gkW^{-1}h^{-1}	5.0	0.78		0.16
	99/96/EC	Euro IV	1/10/2006		ESC + ELR	gkW^{-1}h^{-1}	3.5	0.46		0.02
					ETC	gkW^{-1}h^{-1}	3.5	0.55		0.03
	99/96/EC	Euro IV (Pt 2)	1/10/2008		ESC + ELR	gkW^{-1}h-^{1}	2.0	0.46		0.02
					ETC	gkW^{-1}h^{-1}	2.0	0.55		0.03

*Where no data is shown no limit values apply to these pollutants.

2.4 Emission controls and abatement technologies

53. Emissions of NO_x from both stationary and mobile combustion sources can be reduced by careful control of the combustion conditions in the burner or engine and by after-treatment of the exhaust gases. Control over the combustion environment usually entails manipulating the mixing of fuel and air to affect the temperature and fuel/air ratio in the burner or engine. The aim is to minimise the formation of NO_x in the first place. This is the concept behind the operation of low-NO_x burners for stationary combustion and the effectiveness of exhaust gas recirculation (EGR) in internal combustion engines. After-treatment of exhaust gases generally relies on catalyst technologies to reduce NO_x concentrations in the exhaust, although other methods have been developed which do not involve catalysts.

54. This section provides a brief description of three of the main techniques used to control NO_x emissions from combustion sources in the UK. These are low-NO_x burners for stationary combustors and exhaust gas recirculation and three-way catalysts for mobile combustion sources. A description of other technologies showing promise for NO_x abatement currently available and still under development is provided in the Technical Annexe to this chapter (Appendix 1). These include Selective Catalytic Reduction (SCR) methods for NO_x reduction in stationary and mobile combustion sources, non-catalytic reduction technologies and NO_x traps for mobile sources.

2.4.1 Emission abatement for stationary combustion sources

55. Low-NO_x burners work on the principle of staged combustion in which the fuel and air are mixed in the burner in stages to create regions of different temperature and fuel/air ratio in the flame. Some regions of the flame are fuel-rich and it is here that chemical reactions take place converting NO and its precursors to N_2.

56. Low-NO_x burners have been fitted to all coal-fired power stations in the UK with generating sets of 500 MW or more. They are currently considered the most cost-effective method of achieving NO_x reduction from existing coal-fired plant, with around 30-60% reduction in emissions being achieved (IEA, 2001). Although after-treatment methods such as SCR and other, non-catalytic processes offer potentially greater NO_x reduction efficiencies, the capital and operating costs of these technologies are considerably higher and there are still some operational concerns over the lifetime and costs of the catalyst. SCR and other after-treatment methods are not operational on a large scale in the UK, but are in commercial use on large combustion plant overseas, especially in Japan and Germany. Demonstration and full scale systems are also being installed in US coal-fired plant. These abatement technologies are believed to become more economically competitive over longer operating periods (IEA, 2002).

2.4.2 Emission abatement for mobile combustion sources

57. Road vehicles are a major source of NO_x emissions in the UK. Emissions of NO_x are different for different types and sizes of vehicle and they depend on the type of fuel the vehicle runs on (petrol or diesel) and the load on the engine.

58. In order to achieve increasingly stringent emission standards placed on road vehicles, reductions in NO_x emissions have been and continue to be achieved through a combination of engine design improvements, engine management systems, exhaust after-treatment systems and on-board diagnostics.

59. For petrol vehicles, the main achievements have been through the introduction and refinement of three-way catalyst technology, supplemented by Exhaust Gas Recirculation (EGR). EGR works by recycling a fraction of the exhaust gases through a control valve from the exhaust to the engine intake system where it is mixed with fresh fuel-air mixture. EGR gases act as a diluent, thereby reducing the peak flame temperature and hence the rate of NO formation in the burned gases through the thermal-NO mechanism (Heywood, 1988). Substantial reductions in NO_x emissions (60-80%) can be achieved by EGR.

60. Three-way catalysts have been fitted on all new petrol cars and vans sold in Europe since 1992 in order to meet the stringent emission standards set in Directive 91/441/EEC. It is effectively the only way of meeting the challenge of reducing emissions of CO, hydrocarbons (HC) and NO_x from vehicle exhausts simultaneously. A single catalyst bed can reduce NO and oxidise CO and HC if the fuel/air ratio is maintained close to stoichiometric (i.e. neither fuel or oxygen are in excess). The catalyst effectively brings the exhaust gas composition to a near-equilibrium state: enough reducing gases are present to reduce NO to N_2 and enough O_2 to oxidise CO and HC to CO_2 and H_2O. Around 80-90% reduction in NO_x emissions have been achieved with the three-way catalyst. These figures are based on tests on in-service cars under real-world drive cycles when the vehicle is at normal operating temperature (Euro II relative to pre-Euro I cars, see Section 2.6.2.1). For the most modern cars, the reductions in emissions may be higher than this, including the time when the vehicle is started cold, thanks to improvements in the time it takes for the catalyst to warm up to its normal efficiency (see Section 2.6 and Technical Annexe in Appendix 1). Further details on three-way catalyst technology are given in the Technical Annexe (Appendix 1).

61. Reductions in NO_x emissions are harder to achieve for diesel vehicles, due in part to the oxygen rich regime of the engine. Improved engine design and management systems and EGR have led to the reductions in emissions seen so far, but exhaust after-treatment methods may be necessary to achieve further, more stringent standards on diesel exhaust emissions in the future. A number of these are being developed (see the Appendix 1).

62. Since the introduction of three-way catalysts, emissions of NO_x from petrol cars have decreased below levels from diesel cars of a similar size. Before this, petrol car emissions of NO_x were rather higher than for diesel cars.

63. In parallel with vehicle technology developments, improvements in the quality of petrol and diesel fuels have been made. Fuel quality has little effect on emissions itself, but improvements, especially through reductions in the sulphur content, have opened the gate to emerging exhaust emission after-treatment technologies and better catalyst performance. The EU Directive on fuel quality 98/70/EC has driven the improvements being made to the quality of petrol and diesel fuels sold in the UK.

2.4.3 Formation of NO_2 in diesel exhaust after-treatment systems

64. After-treatment catalyst systems on diesel exhausts can enhance the proportion of NO_x emitted as NO_2. This may be particularly true in an oxidising environment above the catalyst surface where the catalyst may promote the oxidation of NO to NO_2.

65. There is also some concern that certain particulate emission abatement devices may increase the proportion of NO_x emitted as NO_2. Diesel particulate filters (DPF) are increasingly being fitted to buses and other heavy duty diesel vehicles and are very much seen as the solution to the challenge of reducing particulate emissions from these vehicles. There are a number of designs of particulate reduction technologies on the market. The most common of these in the UK are the type that work on the principle of deliberately converting NO in the exhaust stream to NO_2 over an oxidation catalyst unit and using the NO_2 to oxidise the particulates held on the filter, thus regenerating the trap. Continuously Regenerating Particulate Traps (CRTs) which use these techniques do also lead to a small decrease (5-10%) in NO_x emissions, but if no additional measures are put in place, a greater proportion of the NO_x can be expected to be emitted as NO_2. Some studies in the U.S. have suggested as much as 50% of NO_x emissions from a diesel vehicle with this type of DPF may be emitted as NO_2 (CARB, 2002; DaMassa, 2002; IDRAC, 2002), although systems there may not be as well optimised.

66. However, this may only be a temporary problem perhaps restricted to retrofit applications because to comply with tighter Euro IV standards on NO_x emissions, manufacturers of new heavy duty diesel engines will need to find ways of reducing total mass of NO_x emissions substantially. Then, even though the proportion of NO_x emitted as NO_2 is higher, the total mass emissions of primary NO_2 will be reduced compared with levels from existing vehicles. Furthermore, the proportion emitted as NO_2 may depend on duty cycle and could be low at low speeds (for example, in urban areas) when the exhaust temperature is low and the catalyst is not at its working temperature. It is also the case that a number of catalyst optimisation and thermal management strategies exist to minimise the emissions of NO_2 from these types of catalyst-based diesel particulate filter systems (McKinnon, 2002; IDRAC, 2002). These have reduced levels of NO_2 in the exhaust to less than 20% of total NO_x emissions. Certain fuel additives may assist in this. With the increasing need to simultaneously reduce both particulate and total NO_x emissions from diesel exhausts to comply with tighter emission legislation, some manufacturers of particulate traps are developing combined CRT/SCR systems. These work on the principle of using a Selective Catalytic Reduction system with urea injection downstream of the CRT unit to reduce NO and NO_2 (CARB, 2002; IDRAC, 2002).

67. The 50% figure for the proportion of NO_x emitted as NO_2 from these types of DPFs based on studies in the U.S. is almost certainly a 'worst case' estimate and figures for CRTs used in the UK may be lower than this where systems are better optimised. However, the lack of data on primary NO_2 emissions from these types of particulate traps does need to be addressed through further independent research on in-service vehicles over real-world cycles and operating conditions. It is understood that the CleanUp programme, run by the Energy Saving Trust to promote and fund grants towards the cost of fitting emission abatement equipment to commercial and public service vehicles in the UK, is addressing this issue by supporting emission testing programmes that will lead to results in the near future. Other types of particulate reduction technologies are not widespread in the UK at the moment, but may show a greater market share in the future. The effectiveness of these and their penetration (and that of combined NO_x reduction technologies) into the UK market will need to be closely monitored. A number of combined particulate and NO_x reduction systems for diesel vehicles are becoming available or are under development for both original equipment and retrofit applications.

2.4.4 By-product emissions of N_2O and NH_3

68. It is known that some of the NO_x abatement technologies discussed above can lead to increased emissions of N_2O and NH_3 formed as by-products of the NO_x emission reduction process. This is particularly the case for after-treatment systems using catalyst technology. Emissions of N_2O and NH_3 are not regulated, but there are wider environmental concerns about the impact that increased emissions of these pollutants will have. N_2O is a greenhouse gas implicated in global climate change (DETR, 2000a); ammonia plays an important role in the long range transport of acidifying pollutants and is also involved in formation of secondary particulate matter aerosols in the atmosphere (NEGTAP, 2001; APEG, 1999).

69. N_2O is formed in combustion systems, but is removed rapidly in flames by reaction with small combustion radicals (H, O atoms), so under normal conditions, only small amounts of N_2O are actually emitted (Miller and Bowman, 1989). However, N_2O can be formed on catalyst surfaces as a by-product of the catalyst chemistry that reduces the NO_x to N_2. N_2O can be formed in stationary and mobile SCR systems.

70. Cars fitted with three-way catalysts may emit as much as ten times more N_2O than cars without catalysts, leading to a growth in overall emissions from road transport as catalyst cars penetrate the UK fleet (EEA, 2000; Pringent and de Soete, 1989). It is possible that advanced catalyst systems may address the problem of N_2O emissions otherwise increased emissions from the road transport sector in particular will counteract the downward trend in N_2O emissions being achieved with other sectors, for example, industry. There is some evidence to suggest that the enhanced emissions of N_2O from catalyst vehicles occur in just the first few minutes after engine start up when the catalyst has not yet reached its optimum operating temperature. This may be addressed in modern cars (Euro II-IV) equipped with close-coupled catalyst technologies reducing the time it takes for the catalyst to warm up, thereby reducing emissions of a number of different pollutants at engine start-up.

71. Increased NH_3 emissions have also been observed from cars with three-way catalysts. Early catalyst cars (Euro I models) may emit as much as 50 times more NH_3 than non-catalyst cars (pre-Euro I) (EEA, 2000; Baum *et al.*, 2000; Baum *et al.*, 2001; Färnlund and Kågeson, 1998). Measurements on more recent models (Euro II and III) have suggested a fall in NH_3 emissions probably due to better engine management systems and catalyst technology, although emissions still remain higher than levels for non-catalyst cars (Barlow *et al.*, 2001). Formation of NH_3 on the catalyst is a by-product of the NO_x reduction mechanism and depends on the amount of time the engine strays off running at stoichiometric into the fuel rich condition. These are the conditions which favour NH_3 formation. Improvements in engine control systems are significantly reducing the time combustion occurs under fuel rich conditions by narrowing the width of the fuel/air ratio fluctuations (see Technical Annexe, Appendix 1). This is likely to lead to further reductions in NH_3 emissions from advanced three-way catalyst cars (for example, Euro IV).

72. SCR deNO_x systems use NH_3 or a derivative such as urea to reduce NO_x emissions in stationary combustors and diesel engines (see Technical Annexe, Appendix 1). Without effective controls, NH_3 slip can occur in the exhaust leading to higher emissions (CONCAWE, 1999; Koebel *et al*., 2000). This has been a concern for urea-based SCR deNO_x systems being developed for heavy duty diesel vehicles. However, a combination of NO_x sensors with feedback to carefully meter the amounts of urea being injected and downstream oxidation catalyst units to oxidise any NH_3 slip which does occur are expected to address this problem and prevent any excess NH_3 emissions.

2.5 Emission inventories – general principles

2.5.1 General requirements

73. Emission inventories provide an estimate of the mass release rate of emissions of NO_x (expressed as tonnes of NO_2 equivalent) from different sectors at national, regional and local scales. Inventories are compiled on a regular basis so that trends in emissions can be seen. They also have other applications in that emission estimates provide an essential input into air quality modelling activities.

74. The UK's National Atmospheric Emissions Inventory (NAEI) is compiled each year by netcen and provides the time-series in annual emissions of a large number of pollutants by source sector from 1970 to the most recent inventory year, currently 2000 (Goodwin *et al*., 2002). The information is provided in various formats to ensure the UK meets its international reporting commitments to bodies such as IPCC and UNECE. The trends in UK emissions over time and the contribution made by different sectors are used by the national and regional Governments to see the effect of national policies, European directives and economic and energy trends.

75. The Environment Agency compiles a Pollution Inventory of air emissions from around 2,000 major point sources in England and Wales. The Pollution Inventory (PI) database is compiled from a large number of different source sectors. The PI can be accessed through the Environment Agency website at www.environment-agency.gov.uk/pi. Information from the PI is fed into the NAEI either directly as emissions data from specific point sources or used in the NAEI as surrogate data to derive emission factors for particular source sectors. Data from the PI also feeds into the European Pollutant Emission Register (EPER) which provides useful information establishing compliance with international treaties and conventions. Under the IPPC Directive the UK Government must report emissions from PPC permitted sites to EPER. The UK PI has been adapted to meet this reporting requirement and the Environment Agency is responsible for gathering the data. SEPA is required under the Directive to report emissions from PPC permitted sites in Scotland. The requirement to report to EPER is statutory on the UK Government and the Environment Agency can use a legal notice to gather the necessary data. Processes are contacted when the Environment Agency has reason to believe that they will be permitted under the PPC Regulations in future. All Integrated Pollution Control (IPC) authorisations and certain waste management licenses will be subject to PPC. Inventory data are also being collected for EPER purposes for those installations falling under the IPPC Directive which are being regulated by local government.

76. A number of local inventories have been compiled for different areas of the country, usually for the major conurbations. These take advantage of local knowledge and usually more detailed activity data, but they generally use the same emission factors and overall methodology that is used for the national inventory. Local inventories may be able to use emissions or activity data held by Local Authorities for small combustion plant and industrial processes under their jurisdiction (Part B processes regulated by Local Authorities under the 1990 Environment Act, Part I) and also local domestic and transport data. In Scotland, SEPA regulate both Part A and Part B processes. Local inventories can provide data at a fine spatial and temporal resolution and incorporate additional information such as release heights which are required for dispersion modelling exercises, such as those associated with local air quality management activities.

2.5.2 Basic inventory methodological approaches

77. The general approach in compiling an emission inventory is to use where possible measured and reported emission rates for a particular point source, for example, a power station, refinery, or major chemical or industrial plant. These may be held in the PI or registers held by SEPA and local authorities. However, the bulk of emissions are estimated using source-specific emission factors and activity statistics. For combustion sources which are the source of NO_x emissions, emission factors are usually fuel-related and expressed in grammes NO_x emitted per kg fuel consumed. They are then combined with fuel consumption statistics for the relevant source. For road transport, emission factors are traffic-related and expressed in grammes NO_x emitted per kilometre travelled and combined with vehicle kilometre figures from transport statistics publications. Other statistics like population and employment might be used for area-type emission sources such as domestic and commercial combustion at a local level.

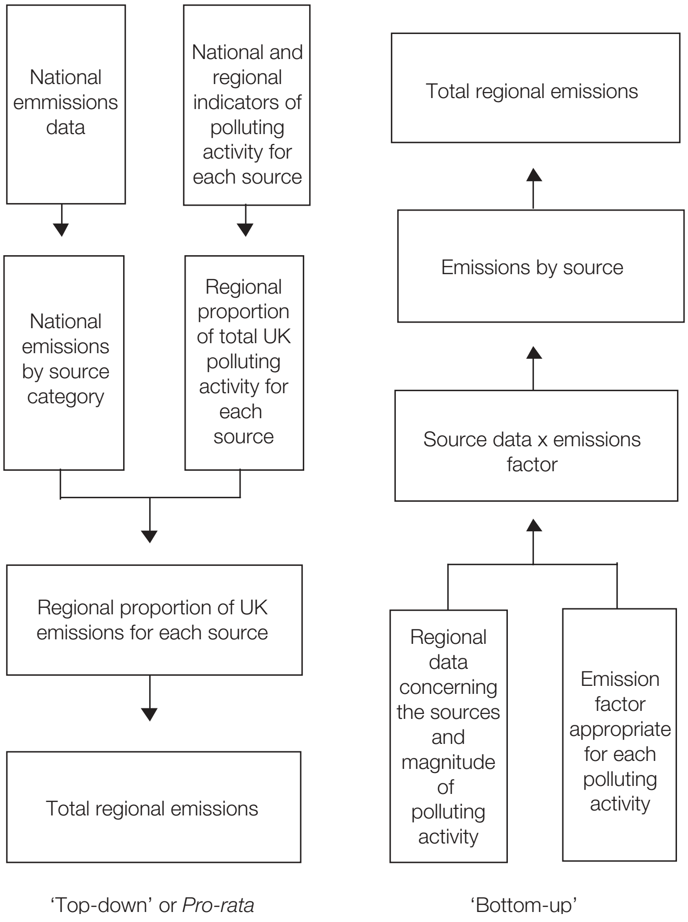
78. Section 2.6 describes the methodologies for estimating emissions from specific source sectors in the national inventory, in most local inventories and in inventories focused on specific local sources (for example, airports).

2.5.3 Spatially resolving emission inventories

79. The national emissions inventory for NO_x refers to emissions in the whole of the UK over an entire year. For many sources, the UK inventory is compiled using national fuel consumption and transport statistics. There will be areas in the country, as well as times of the day and year, where the contribution from certain sources (traffic for example) will be higher than indicated proportionally by the national inventory figures. Emissions from different sources also vary in height of release.

80. Spatially resolved emission estimates are required for ambient NO_x and NO_2 concentration modelling. The development of spatially resolved emission inventories requires information about the geographical location of sources as well as an estimate of mass emissions. There are two approaches (Figure 2.2) to the generation of spatially resolved emission inventories which have been termed ‘top-down’ and ‘bottom-up’. The ‘bottom-up’ approach provides an estimate of emissions for a particular area by utilising geographical source data with local datasets and appropriate emission factors. This provides the most reliable indication of the magnitude and spatial distribution of emissions. The ‘top-down’ or pro-rata approach involves disaggregating national emission estimations to a local level through the use of geographical data and indicators of the proportion of a particular polluting activity occurring in the specified region. It is particularly useful when there is no direct measure of polluting activity available. In practice, individual inventories may make use of both ‘bottom-up’ and ‘top down’ methodologies for different source sectors.

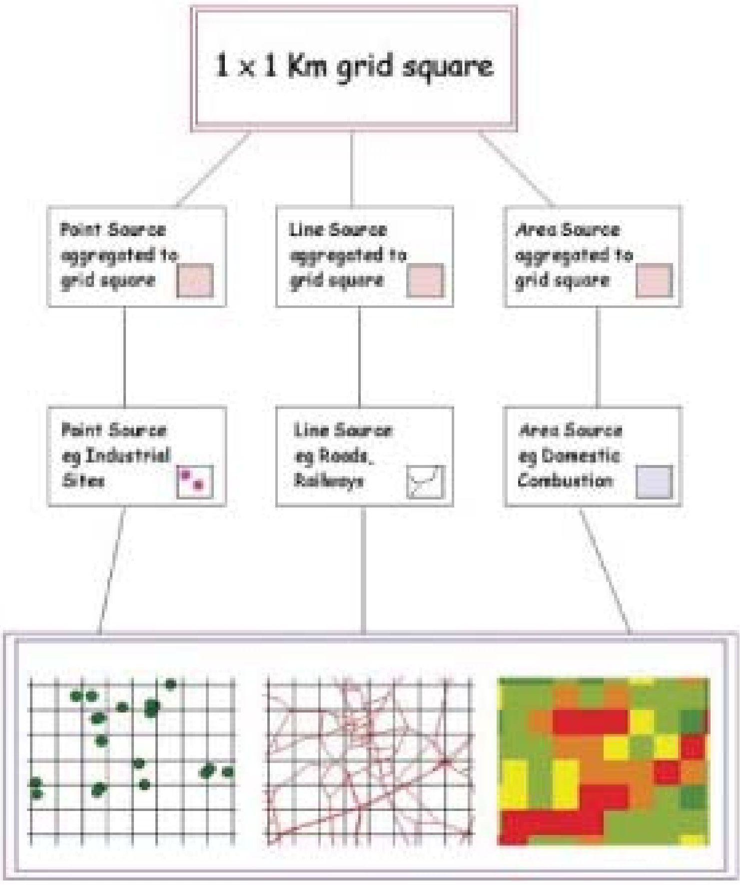
Figure 2.2 The ‘top-down’ and ‘bottom-up’ approaches to emission estimation (Lindley *et al.*, 1996).



81. Geographical Information Systems (GIS) have become increasing important tools for the development of spatially resolved emissions inventories and the mapping of national estimates. The use of GIS allows the application of complex emission estimation models to large spatially referenced datasets which can be relatively easily updated and manipulated. GIS technology is also useful in relation to the presentation or further application of resultant emissions data. This can provide a suitable platform for the generation of spatially resolved emissions data for particular time periods and the incorporation of contextual data which may be required for dispersion modelling such as stack heights, topography and surface roughness.

82. Vector-based GIS packages lend themselves to emissions inventory development since the representation of spatial entities as point, line and polygon (area) features can be readily applied to a characterisation of pollution sources as stationary (point), mobile (line) and diffuse (area) sources. Area-based representations are also used to make emission estimates for sources where individual treatment is either not desirable due to the quantities of sources and their relatively small contribution to overall emissions, such as with domestic combustion sources or not possible due to a paucity of appropriate spatial or attribute data. Within a GIS, data layers or themes are generated corresponding to individual sources which can then be aggregated to a uniform spatial unit for display or further use (Figure 2.3). The point, line and area representations may be used as a direct input to certain dispersion models such as ADMS Urban, which has its own emissions estimation module operating on a vector based environment.

Figure 2.3 Organisation of point, line and area emission sources in a GIS to produce a spatially resolved emission inventory (aric, 2000).



83. Local emission inventories generated from a ‘bottom-up’ approach and national emission maps generated from a ‘top-down’ approach should give broadly the same trends in terms of emissions per square kilometre and the contribution of different sources to total emissions in an area over the same time period. However, there will inevitably be differences in detail in the results between the two inventory approaches. These differences can result from variations in the source of spatial data used to represent the location of sources. For some sectors, for example, emissions from major roads and power stations, the emission maps and local inventories should provide the same emissions in each grid square because both approaches will use the same source of data, for example, major road traffic flows provided by the DfT traffic census. However, in deriving the national maps, emissions from some sources in a particular grid square will be estimated using surrogate data to spatially disaggregate national emission totals that might be derived from national fuel consumption data. Examples of this are emissions from domestic combustion, small industrial boilers and traffic on minor roads. In local inventories, detailed data may be available for these sources from local authorities, local traffic surveys and other local sources. Such detailed knowledge would be impossible to obtain and use in a consistent manner across the whole of the country when generating the national emission maps. Even on major roads which use the same traffic flow data, local inventories may take account of local differences in the age of the fleet from traffic camera information, local speed data or efforts made by local fleet operators (for example, buses) to reduce emissions from their fleet by, for example, retrofitting or engine upgrading.

84. Details of methods used to provide the 1x1 km map of UK NO_x emissions and local area inventories are described in Section 2.8 with particular attention given to the inventory for London.

2.6 Estimating emissions from different sources

85. The following sections describe the methodologies used for quantifying emissions of NO_x in the UK from each of the main sectors. Further details of the methods used for the national inventory can be found in the NAEI annual report (Goodwin *et al*., 2001) and website at www.naei.org.uk/index.php. Emission factors for specific sectors can be found in the Emission Factor Database also available on this website.

86. The measurement of NO_x and primary NO_2 emissions from road vehicles, the influencing factors and the derivation of emission factors are discussed in detail in this section.

87. Estimating emissions from a specific source at a finer temporal or spatial resolution may require more detailed approaches and emissions data to be used than is required for the national inventory. Some particular considerations related to this are discussed and examples shown where these have been applied to calculating emissions from different transport sectors.

2.6.1 Emissions from stationary combustion sources

88. Emissions of NO_x from public power generation are based on reported emissions data for individual power stations provided by the Pollution Inventory and station operators. These include all power stations running on coal, oil and gas, as well as Municipal Solid Waste. For the small amount of electricity generated from other fuels, such as landfill gas, emissions were estimated from fuel-based emission factors taken from the USEPA's Compilation of Air Pollution Emission Factors AP-42 (USEPA, 1997) and DTI figures on fuel consumption for this source.

89. Estimates are made of emissions of NO_x occurring from combustion in a number of industrial sectors. These include combustion at petroleum refineries, in the manufacture of solid fuels (for example, coke production), in iron and steel production and for processes in a variety of other industries. They are based on reported emissions data for individual plant operations provided by the Pollution Inventory and plant operators and also on fuel consumption data for different industries from DTI (DTI, 2002) combined with emission factors for each fuel type. Emission factors are taken from several sources, including CORINAIR (1999), USEPA (1997) and IPCC

(1997) manuals. Emissions data reported for large combustion plant in Scotland collected by SEPA are also used.

90. NO_x emissions from domestic consumption of coal, oil and gas and small boilers in the commercial and public sector are estimated from DTI fuel consumption data for these sectors by fuel type (DTI, 2002) and emission factors from USEPA and CORINAIR.

2.6.2 Emissions from road transport

91. The importance of road transport as a ground-level source of emissions in urban areas and the availability of appropriate emissions and transport data has meant that road transport emissions of NO_x are relatively well quantified. Exhaust measurement programmes within the UK and elsewhere have been on-going since the late 1960s. With the availability of fairly detailed traffic and fleet composition data, fairly sophisticated methods can be used to estimate emissions from this sector in contrast to other transport modes.

92. All vehicles and engines entering the UK market are subject to type approval and as part of this approval process, engines are required to comply with emission limits (Table 2.1). Compliance with these limits is established through the measurement of the vehicle (for light-duty vehicles) on a chassis dynamometer where it is driven over a regulated test cycle. The exhaust emissions over this test cycle are sampled using a constant volume sampler (CVS) and analysed either on a continuous basis, or through the use of bag samples, using conventional gas analysers. The total emissions recorded over this test cycle are then compared to the legislative limit value, routinely expressed in $gtest^{-1}$ or gkm^{-1}. In the case of heavy-duty vehicles, the existing legislative test is conducted on the engine alone, using an engine dynamometer, with the measurement of exhaust emissions at the sum of 13 engine load and speed conditions.

93. A comparison of driving characteristics demonstrates that the legislative test conditions poorly represent existing in-service conditions. In general, these test conditions are characterised by a passive driving style with constant and relatively slow changes in vehicle speed. A comparison of steady-state and transient driving conditions (rapidly varying speed, load and acceleration), demonstrate that emissions are considerably higher under conditions of transient operation (Joumard *et al*., 1995). Therefore, emission factors generated through the Type Approval procedure are not used within the derivation of standard emission databases used for emission modelling.

94. Emission factors for quantifying real world emissions are generated through the measurement of emissions from a selection of vehicles, driven over a range of real world driving cycles. Each of these driving cycles or associated sub-cycles are characterised by a specific average speed. It is this average speed that is used as a surrogate for vehicle operation, with emission factors allocated to a range of average speeds. The results from tests over a range of average speeds may then be used to derive an average speed-emission relationship. It is this approach that remains most widely used with average speed-emission curves generated for various vehicle types, engine sizes, fuel types, legislation classes and pollutants.

95. Emissions of NO_x from road transport are derived using these speed-related emission factors. Within the UK, emission factors for vehicles at their normal operating temperature come from a large database of emission measurements held by TRL. These are combined with fleet composition and traffic activity data for different years on the national road network provided by DfT. From this, the hot exhaust emissions are derived for each vehicle and road type.

96. Emission factors are intended to be representative of the vehicle fleet. Thus the selection of test vehicles routinely attempts to reflect the real fleet composition. This is obviously easier with larger sample sizes, and indeed large samples can not only reflect the vehicle type, fuel and engine size, but may also reflect various states of maintenance. This level of maintenance is difficult to assess in smaller samples, and whereas before this level of maintenance was built into the basic emission factors (Euro I and earlier), this factor is increasing handled separately.

97. Furthermore, because the emission database is dominated by results from conventional vehicles, these data will undoubtedly have the lowest uncertainty. For these conventional vehicles and fuels the repeat measurements of the same vehicle over the legislative test cycle can vary by approximately ±7% of the mean. For real-world cycles this repeatability can be considerably higher (±30% for the new ARTEMIS high speed driving cycle).

98. Vehicles generally emit more when the vehicle is started with the engine cold. This is partly due to the poor combustion efficiency when the engine is cold and the need to run the engine slightly fuel rich to achieve combustion stability. It is also due to the low efficiency of the three-way catalyst during the time it takes to reach the light-off temperature (see Technical Annexe, Appendix 1). The excess cold start emissions are calculated in the inventory using standard emission equations and vehicle trip start data. Cold start emissions of NO_x are relatively minor compared with hot exhaust emissions. This is not the case for CO and HC where cold start emissions are considerably more important.

2.6.2.1 Exhaust emission factors for NO_x

99. Surveys of road vehicle exhaust emission factors are on-going within the UK and elsewhere. The last revision to the UK database was undertaken by TRL on behalf of the DfT (Barlow *et al*., 2001). Historically these emission test programmes have involved relatively large vehicle samples (in excess of 200 vehicles), but more recently this sample size has been reduced, in favour of an increase in the number of pollutants and conditions analysed. Statistically this can result in an increase in the uncertainty associated with the basic emission factors. The NAEI incorporates the TRL emission factor database, but it should be recognised that this database is dominated by test data associated with conventional fuels and that the latest revision supplements the existing emission factors with data for Euro I and Euro II vehicles. Previously, with the exception of petrol cars, emission factors for these classes of vehicles were estimated based on changes in the emission directive’s type-approval limit values. A substantial part of the new measurements of Euro I and Euro II emission factors come from several test programmes funded by DfT and Defra and carried out at UK test laboratories between 1999 and 2001 (Barlow *et al*., 2001). The measurements were made on dynamometer test facilities under various simulated real-road drive cycles.

100. Table 2.2 shows NO_x emission factors for each class of vehicles calculated from the emission factor equations at typical speeds on urban, rural and motorway roads. These emission factors are currently used in the NAEI. The vehicle types match the traffic activity data that are used in the national emission calculations.

101. Emissions from future emission classes yet to be introduced, must again be estimated through an examination of the proposed future legislation and the technical ability to meet these new limits. This estimation process can result in significant errors for future emissions factors. This was demonstrated with the previous over-estimation of the reduction in NO_x emission factors for Euro I and II diesel vehicles, when in fact the latest series of tests showed little difference between the emission factors for the pre-Euro I, Euro I and Euro II classes of vehicles. In some cases, the Euro I factors were higher than the pre-Euro I factors, or Euro II higher than Euro I, indicating that improved technologies during the 1990’s had delivered little or no benefits to NO_x emissions from diesel vehicles. There are two possible explanations for this. One is the possibility of ‘cycle beating’, at least in the case of heavy duty vehicles (UBA, 2003; Kågeson, 1998). Modern engine management systems are tightly tuned and there is the possibility that legitimate NO_x emission reductions over the specified cycle for type-approval testing can be achieved, but when the vehicle is driven under real road conditions (as in the emission factor tests), the engine is frequently operating outside its ‘tuned’ condition and the emission reductions are not realised. The risk of cycle beating has been recognised by the EU by revising the test cycle for Euro III and Euro IV engines for heavy duty vehicles.

102. Another possibility that applies to the light duty diesel vehicles is the degree of tolerance that the regulations provide in the permitted levels of NO_x emissions, allowing the vehicle manufacturers to trade-off a certain amount of NO_x in order to achieve the more stringent limits set on particulate emissions. Examination of the NO_x emission factors for pre-Euro I diesel cars and vans at average speeds of the test cycle shows that they were close to or already below the limits set in the Euro II standards. Thus, manufacturers could concentrate their attention on methods to reduce particulate emissions, a more challenging problem at the time. It should be emphasised, however, that the new emission factors for Euro II diesel vehicles (and even Euro I factors for heavy duty diesel vehicles) are still based on a very small number of vehicles tested. An increase in this sample size will assist in assessing these conclusions, although it should be noted that these are already supported through other national test programmes, including those sponsored by the German and Austrian Environmental Protection Agencies (UBA, 2003).

103. Table 2.2 also provides emission factors for Euro III and Euro IV vehicles, the latter referring to standards which will come into effect in 2006 (and 2008 for heavy duty vehicles). These are based on scaling factors assumed to apply across all speeds relative to emission factors for Euro II vehicles. For light duty vehicles (petrol and diesel), the scaling factors were estimated by the NAEI on the basis of current emission levels for Euro II vehicles and the amount emissions will need to be reduced to achieve the type-approval limit values, taking into account differences in the drive cycles used and the contribution that cold start emissions will make to emission over the new 98/69/EC test cycle. A similar approach was used to estimate emissions from new motorcycles, using factors for pre-2000 models from TRL and scaling factors based on recently agreed emission limits (Official Journal, 2002). For heavy duty vehicles, it was assumed that Euro II vehicles are currently at their emission limits and will be reduced no more than required to achieve the new emission limits, using scaling factors from the COPERT III database (EEA, 2000). COPERT III is the methodology and database of road transport emission factors developed by a group of experts for the European Environment Agency and recommended for national emission inventory reporting under CORINAIR.

Table 2.2 Exhaust emission factors for NO_x (as NO_2) for different road types used in the National Atmospheric Emissions Inventory, based on data from TRL (Barlow *et al.*, 2001).

$gNO_x(\text{as } NO_2)km^{-1}$		Urban	Rural	Motorway
Petrol cars	ECE 15.04	1.644	2.211	3.164
	Euro I	0.257	0.368	0.663
	Euro II	0.229	0.245	0.370
	Euro III	0.137	0.147	0.222
	Euro IV	0.073	0.078	0.118
Diesel cars	Pre-Euro I	0.623	0.570	0.718
	Euro I	0.537	0.465	0.693
	Euro II	0.547	0.505	0.815
	Euro III	0.547	0.505	0.815
	Euro IV	0.273	0.253	0.407

gNO_x(as NO_2)km^{-1}		Urban	Rural	Motorway
Petrol LGVs	Pre-Euro I	1.543	1.783	2.351
	Euro I	0.361	0.356	0.531
	Euro II	0.319	0.385	0.567
	Euro III	0.192	0.231	0.340
	Euro IV	0.105	0.127	0.187
Diesel LGV	Pre-Euro I	1.332	1.254	1.549
	Euro I	1.035	0.892	1.384
	Euro II	0.983	0.848	1.315
	Euro III	0.735	0.634	0.982
	Euro IV	0.383	0.330	0.512
Rigid HGVs	Pre-1988	13.53	13.53	13.53
	88/77/EEC	6.02	4.96	5.91
	Euro I	7.63	6.87	7.23
	Euro II	6.51	5.78	6.02
	Euro III	4.49	3.99	4.15
	Euro IV (2006)	3.19	2.83	2.95
	Euro IV (2008)	1.82	1.62	1.69
Artic HGVs	Pre-1988	20.70	20.70	20.70
	88/77/EEC	16.93	12.93	11.52
	Euro I	20.25	18.25	19.22
	Euro II	13.96	12.40	12.91
	Euro III	9.63	8.56	8.90
	Euro IV (2006)	6.84	6.08	6.32
	Euro IV (2008)	3.91	3.47	3.61
Buses	Pre-1988	16.74	13.83	13.36
	88/77/EEC	13.62	5.45	6.13
	Euro I	10.93	6.18	6.51
	Euro II	9.78	5.52	5.75
	Euro III	6.75	3.81	3.97
	Euro IV (2006)	4.79	2.71	2.82
	Euro IV (2008)	2.74	1.55	1.61
Mopeds, <50cc, 2st	Pre-2000	0.030	0.030	0.030
	Euro I	0.010	0.010	0.010
Motorcycles, >50cc, 2st	Pre-2000	0.032	0.066	0.126
	Euro I	0.025	0.029	0.052
	Euro II	0.025	0.029	0.052
	Euro III	0.006	0.007	0.012
Motorcycles, >50cc, 4st	Pre-2000	0.156	0.229	0.385
	Euro I	0.210	0.279	0.448
	Euro II	0.210	0.279	0.448
	Euro III	0.048	0.064	0.103

2.6.2.2 Road transport activity data

104. To calculate UK emissions from road transport in the NAEI, the hot exhaust emission factors are combined with annual UK vehicle kilometre data for different vehicle types on different roads. For Great Britain, vehicle kilometre data are taken from the DfT annual traffic census for the national road network (DfT, 2002a). Vehicle kilometre data for Northern Ireland are derived from the survey by the Transportation Unit, Road Services Headquarters (NI Road Services, 2001).

105. Data are also required on the composition of the fleet for each type of vehicle and the petrol vs. diesel mix for cars and LGVs (light goods vehicles less than 3.5 tonnes). The fleet composition data required are the vehicle size mix (engine cc and gross vehicle weight) and the proportion of vehicle kilometres travelled in a year by each Euro standard. The latter is related to the age profile of the fleet, using year-of-first registration data from DfT's vehicle licensing statistics published each year (DfT, 2002b). Account is also taken of the fact that newer vehicles travel more miles in a year than older ones, using survey data from DfT.

106. It is assumed in the NAEI that 5% of cars fitted with three-way catalysts fail each year, but 95% of failed catalysts are repaired after the vehicle is 3 years old and reaches the age of MOT testing. This failure rate assumption was first adopted for early generation catalyst systems (Euro I) to reflect accidental damage and poisoning of the catalyst system by drivers. However, the application of the 5% failure assumption to more modern and future car models (Euro II-IV) is currently under review, in light of better system durability required as a condition of Type-Approval and advanced on-board diagnostic systems on cars which should be able to detect problems when they arise. It is indeed the case that although there are no definitive sets of data on failure rates, emissions information from MoT test returns and road-side testing, and data from car leasing companies, equipment suppliers and manufacturers' warranty returns are all pointing towards lower failure rates (and in some cases much lower) than 5% in recent years. For catalyst cars, a slow deterioration in emissions with vehicle mileage is taken into account in the NAEI.

107. Further details on the emission calculation methodology are provided in the NAEI report (Goodwin *et al.*, 2001).

2.6.2.3 Cold start emissions

108. The excess emissions that arise when the vehicle is started with its engine cold or below its optimum operating temperature are calculated using a method taken from COPERT III. The procedure uses equations that take account of the effects of ambient temperature on the excess emissions and its effect on the distances travelled with the engine cold. The distances travelled with cold engines also depend on average trip lengths: the shorter the average trip lengths are, the greater the contribution of cold start emissions. The method calculates the ratio of cold to hot exhaust emissions for light duty vehicles which are used to calculate the overall cold start emissions from these vehicles. Further details on the methodology used for cold starts are provided in the NAEI report (Goodwin *et al.*, 2001).

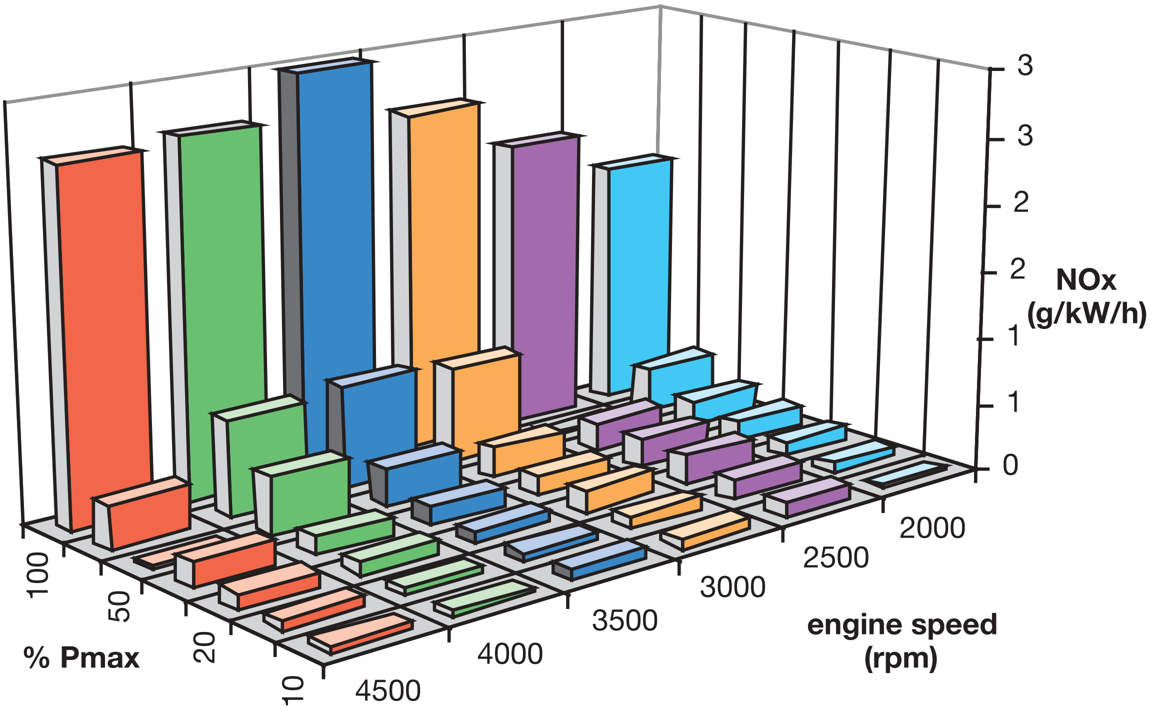
2.6.2.4 Emissions from instantaneous operations of road vehicles

109. Modelling and mapping emissions from road transport on a local scale involves simplifying the road network as a series of line sources subdivided into links (with uniform flows and speeds) and allocating fleet corrected average-speed emission rates in gkm^{-1}. These average speed emission rates are used for simplicity, as it has been considered that the average speed of a road link may be readily determined. However, this average speed emission factor has actually been a surrogate for vehicle operation, and in reality the emissions would be better described through the use of a number of other variables such as relative positive acceleration and engine load, rather than road speed itself. However, in practice these variables would be extremely difficult to determine for each vehicle.

110. For pre-Euro I vehicles, the average speed approach appears to be sufficient to simulate variations in emissions and thus roadside air pollution on a geographic and temporal scale. With the introduction of the three-way catalyst and its associated large reduction in exhaust emissions, the average speed approach appears to be becoming less appropriate.

111. For example, evidence suggests that catalysts tend to exhibit on/off control, and emission levels from catalyst-equipped vehicles are much more sensitive to operating conditions than those from non-catalyst vehicles. Under particular operating conditions the catalyst may be working at its maximum efficiency, but for slightly different conditions the conversion efficiency may be low. For example, measurements by Joumard *et al*. (1998) have shown that for engine loads (the actual power divided by the maximum power at a given engine speed) greater than 75%, instantaneous CO emissions can be 20,000 times higher, and NO_x emissions 10 times higher, than for lower loads (Figure 2.4). Over an entire motorway driving cycle around 90% of the total CO emissions occurred during only 15% of the time.

Figure 2.4 NO_x instantaneous emission versus engine speed and load over the motorway IM modem cycle for a catalyst-equipped light-duty vehicle (Joumard *et al*., 1998).



112. An examination of instantaneous exhaust emissions against vehicle speed (for a Euro III petrol vehicle) indicates that during the majority of a typical test cycle emissions remain very low, but are subject to substantial elevation when the engine is subjected to changes in engine load, such as gear change events. This type of observation would suggest that the use of an average speed based modelling approach in a typical urban street could over-estimate the emissions along the majority of the link, but underestimate them at situations where vehicles interact, such as junctions and during conditions characterised by stop-start driving. This has obvious implications for emissions and subsequent air quality modelling of NO_2 concentrations at specific points near a road junction or areas of congestion.

113. Research into the development and use of instantaneous emission models has been on-going for many years and includes studies by INRETS, TUEV Rheinland, LAT and TRL within the MODEM project (EU 4th framework project), and more recent studies by the Technical University of Graz and EMPA within the ARTEMIS project (EU 5th framework project) and separate studies by MIT. However, these studies do not fully address the most fundamental problem relating to instantaneous emission modelling: it is extremely difficult to measure emissions on a continuous basis with a high degree of precision, and the emissions and fuel-consumption values recorded in the one-second steps might not be successfully allocated to the associated operating conditions. For example, because of the time required to transport the exhaust gas to the analysers, and the actual response time of the analysers themselves, the emission signals are delayed relative to the driving cycle. Furthermore, the exhaust gas is mixed in the exhaust system. This results in a general flattening of instantaneous emission peaks over a period of more than one second. The dynamics of mixing also depend on the gas flow rate, and the situation is even worse when dilute exhaust gas is being sampled using a CVS (Boulter, 2002).

114. These results have obvious implications for the development of modal emission models based on instantaneous vehicle operation. Therefore, even if modal emission models were constructed using raw exhaust measurements, such results indicate that there would clearly be problems matching an emission signal in any given second to the appropriate speed or acceleration measurement.

115. Clearly, advances in the field of modal emission modelling will not be forthcoming until realistic continuous emission data are available. Efforts are now underway to reduce the dynamic distortion of the emission data (Weilenmann *et al.*, 2001).

116. In summary, it may be concluded that the transfer of instantaneous modelling to real-world conditions is considerably more complex than that of the average speed approach.

2.6.2.5 Impact of traffic management schemes

117. Much of the progress to date in the control and reduction of the environmental impact of traffic has been through the use of improved vehicle technology and fuels, often prompted by increasingly stringent legal limits and design standards. In addition, the way in which a vehicle is operated has a significant influence on its environmental performance. Traffic congestion and the associated driving pattern of stops and starts, accelerations and decelerations can produce excessive emissions. Thus, traffic management schemes that modify the behaviour of the traffic, as well as the volume of traffic, can supplement the effects of improving technology. In general traffic management that encourages drivers to drive in a more passive style are beneficial (Abbot *et al*., 1995).

118. In the mid-1990s the DfT commissioned a programme of research investigating the links between traffic management and air quality. This programme (TRAMAQ) is on-going, and comprises a number of inter-related studies largely focusing on urban traffic control (Abbot *et al*., 1995; Boulter *et al*., 2001; McCrae *et al*., 2000).

119. A review of the various traffic management case studies suggests that lower speed limits and traffic calming schemes usually reduce NO_x emissions on the affected roads.

120. The potential of Low Emission Zones (LEZ), restricting access of vehicles in a zone to those meeting a certain emission standard, has been assessed for London. An initial study carried out by TRL on behalf of Westminster Council (Cloke *et al*., 2000) concluded that restricting

HDVs to only those complying with Euro III limits could reduce NO_x emissions by up to 20%. A further feasibility study funded by the GLA, Transport for London, Defra and the Association of London Government has so far considered many of the issues of importance to an LEZ in London. These have included: emission change, impact on atmospheric concentrations of NO_2 and PM_{10}, implementation issues and the cost-effectiveness of any scheme considered. Initial results from the study (www.london-lez.org) suggest that the benefits of an LEZ in London would be less than those identified in the previous study carried out by TRL. The principal reason for a reduced impact is the use of the latest emissions factors for NO_x in the latter study, which for diesel vehicles show less benefits for new technologies than previously thought (Barlow *et al*., 2001). Most of the scenarios considered by the LEZ team have focussed on HGVs, taxis and LGVs, which are almost all diesel-engined.

121. Other traffic management schemes assessed include dedicated bus lanes in Copenhagan which were estimated to have reduced NO_x emissions by between 5-15%. A pilot study on the introduction of red-routes within London estimated that NO_x emissions would be reduced by between 1-3%. Finally, the introduction of variable speed limits on the M25 motorway, while demonstrating improvements in CO and CO_2 emissions, were judged to have a neutral effect on NO_x emissions (Barlow, 1997).

122. Traffic management may only be used to improve air quality if the area of influence of the management scheme is large. In the case of NO_2, roadside concentrations of NO_x are large and thus the limit on NO_2 is not necessarily NO_x emissions, but frequently the availability of oxidant to convert NO to NO_2 in the urban atmosphere, as will be discussed later in this report. Therefore, even if traffic management regimes are shown to reduce the emissions of NO_x, the resulting impact on NO_2 concentrations is always lower (McCrae et. al. 2000).

2.6.2.6 Primary NO_2 emissions from road vehicles

123. On behalf of the Highways Agency and DfT, TRL carried out a small investigation to determine the proportion of NO_x emitted as NO_2 from a range of road vehicles (Latham *et al*., 2001; McCrae *et al*., 2002). In order to determine the proportion of NO_x emitted in the form of NO_2, a vehicle test matrix was developed to provide a representative sample of vehicles in the UK vehicle fleet. A resulting small sample of eighteen vehicles was subsequently subjected to testing, covering 9 petrol cars, 3 diesel vans, 3 diesel heavy goods vehicles and 3 buses, meeting the pre-Euro I, Euro I or Euro II legislative standards. All testing was undertaken on vehicles taken from the in-service fleet and tested in the ‘as-received’ condition. The study did not include any diesel vehicles equipped with particulate traps or any other specific exhaust after-treatment devices.

124. The vehicles were subjected to tests on a chassis dynamometer over a series of legislative and real world driving cycles, while emissions were recorded on a continuous basis. These included the Warren Spring Laboratory (WSL) congested urban, urban, suburban and rural cycles and legislative 96/69/EC test and for heavy goods vehicles and buses over the simulated 13-mode FIGE cycle followed by the Millbrook bus cycle.

125. For the light duty vehicles (the petrol cars and diesel vans), the study examined for any statistically significant dependence of the measured NO_2/NO_x emission ratio on the sampling method used; the fuel type (petrol or diesel); average speed of the test cycle; and the emission standard of the vehicle. A smaller range of tests was carried out on the heavy duty vehicles.

2.6.2.6.1 The measurement techniques and sampling strategy for NO_2 emissions

126. The standard legislative method for measuring NO_x emissions from vehicles depends on the type of vehicle being measured. In the case of light-duty vehicles, NO_x measurements are made on diluted exhaust gas, whereas those for heavy-duty engines are made on raw exhaust. In addition, for the NO_x measurements from HGVs, the sample may be pre-conditioned by passing the gas stream to a chiller unit via a heated line, prior to analysis in an unheated NO_x analyser, in order to remove water from the exhaust sample. Alternatively, the legislation also allows for the measurement of NO_x in a heated NO_x analyser, delivered directly through a heated sample line. These various techniques, while not having a significant impact on total NO_x emission, could affect the fraction of NO_x that is measured as NO_2.

127. Conventionally, NO_x emissions are measured using chemiluminescence techniques (see Chapter 4). To achieve continuous measurements of both NO and NO_x, using a reducing agent to convert all NO_x to NO, requires either a switchable unit or the use of two separate analysers. Either way, the resultant NO_2 emissions are subject to a relatively large potential error, since the indirect NO_2 measurements are derived through the difference between two concentrations (NO_x and NO) of a similar magnitude.

128. Infra-red absorption spectroscopy relies on the principle that different chemical groups absorb radiation at different frequencies. The absorption spectra for NO and NO_2 are at different frequencies allowing for their direct, independent and continuous measurement, without introducing potential errors derived from the difference of two measurements. Fourier Transform Infrared Spectroscopy (FTIR) and standard chemiluminscence techniques were used to measure NO_2 in this study.

129. The sampling strategy may have a significant impact on the proportion of NO_2 measured in the NO_x sample, due to variations in the sample residence times, temperatures, and its subsequent dilution and reaction in the diluent air. Emissions of NO, NO_2 and NO_x were recorded using five different sampling and analysis approaches:

- raw exhaust with unheated chemiluminescent analyser,
- raw exhaust gas with heated chemiluminescent analyser,
- diluted exhaust gas with heated chemiluminescent analyser,
- diluted exhaust gas with a heated FTIR analyser, and
- raw exhaust gas with a heated FTIR analyser.

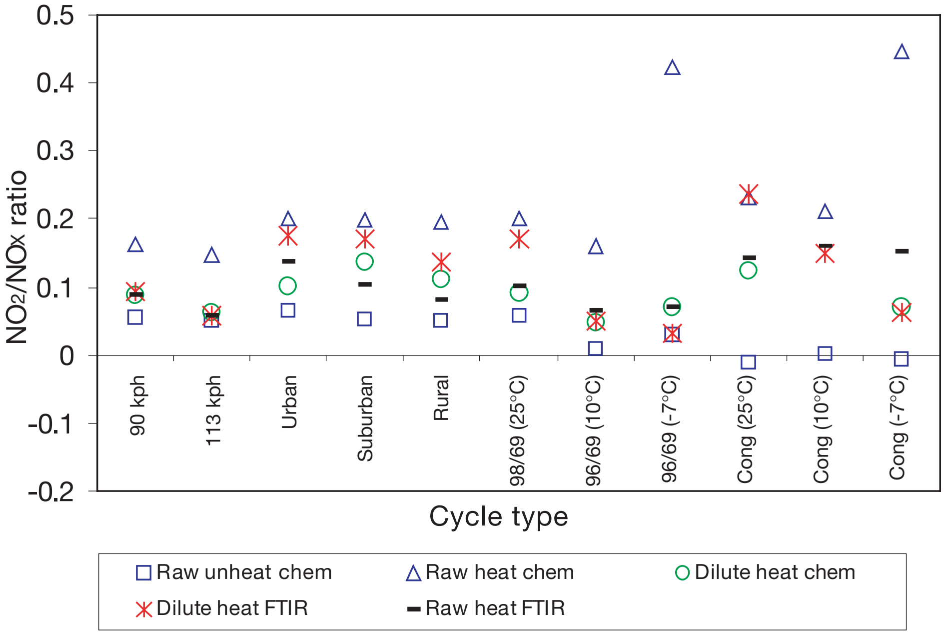
130. Different levels of NO_2 detected could be influenced by reaction of NO with O_3 in the diluent air and through the slow NO + NO + O_2 reaction which would occur to varying degrees in each sampling regime.

2.6.2.6.2 Emissions from light duty vehicles

131. The NO_2/NO_x ratios measured for light duty vehicles covered a broad range over the test matrix covered, with values as low as 0.03 to as high as 0.46. In general, the highest ratios were found for tests carried out using the raw exhaust with heated chemiluminescent analyser over the congested urban cycle. Figure 2.5 shows the average NO_2/NO_x ratios measured using the different sampling and analysis methods over the various vehicle test cycles.

132. With so may potentially influencing variables in the vehicle test matrix used, a detailed statistical analysis of the results was carried out to identify those factors that showed a statistically significant effect on the NO_2/NO_x ratio. The analysis was carried out on the results obtained using the FTIR detection method.

Figure 2.5 NO_2/NO_x ratios for light duty vehicles determined using different driving cycles and measurement techniques (Latham *et al*., 2001).



133. The results of the analysis showed there was a statistically significant relationship between the NO_2/NO_x ratio and the fuel type, the average cycle speed and the method of sampling used, but not with the vehicle's emission standard. The results indicated:

- The NO_2/NO_x ratio was significantly higher for diesel vehicles than for petrol vehicles. The difference in the ratio for the different fuels was statistically highly significant and estimated to be 0.25.
- The ratio increased with decreasing average speed of the drive cycle. The effect was small compared with fuel type, but the analysis suggested the effect was statistically significant and that the difference in the NO_2/NO_x ratio at speeds of 7 and 120 kph was 0.11.
- The ratio was higher when a dilute sample was used than for the raw exhaust.
- There was no statistically significant dependence of the ratio on the Euro standard of the vehicle. This implies that the effect of the legislation in reducing total NO_x emissions has been equally effective in reducing primary NO_2 emissions.

2.6.3 Emissions from heavy duty vehicles

134. A less detailed study was made on factors influencing the ratio of NO_2 to NO_x emissions from heavy duty diesel vehicles and the study did not quantify the magnitude of the NO_2/NO_x ratio for this class of vehicles. The study concluded that NO_2 emissions were not significantly affected by Euro standard and since NO_x emissions from pre-Euro I, Euro I and Euro II classes of HDVs are not significantly different, it has to be concluded that the NO_2/NO_x ratio is also similar for all the Euro standards.

2.6.4 Conclusions concerning primary NO_2 emissions from road vehicles

135. The results from this study have to be viewed in the context of the relatively small sample size investigated. The study has confirmed the higher fraction of NO_x emissions as NO_2 in the exhausts of diesel vehicles compared with petrol vehicles, but has also indicated the importance of sampling and measurement method used as well as average speed or drive cycle of the vehicle. Further measurements are needed to define and quantify the average speed dependence of the NO_2/NO_x ratio on all types of vehicles, but especially for heavy duty vehicles. The speed-dependence noted on emissions from light duty vehicles is significant as it implies a greater proportion of NO_x emitted as NO_2 in urban areas, where average speeds are lower, compared with rural areas and motorways. The possible implications this could have on primary NO_2 emissions from slow-moving buses and large HGVs, as well as possibly diesel vans and taxis, in the centre of towns and cities are significant.

136. It is hard to provide a firm and unambiguous value on the NO_2/NO_x ratio for emissions from road vehicles. On the basis of the results from this investigation carried out by TRL, an average value of 0.05 ± 0.03 is suggested for petrol vehicles and an average value of 0.25 ± 0.10 is suggested for diesel vehicles. For diesel vehicles, the value could be higher than this average value in urban areas and lower than this in rural areas and motorways. Further research is required to confirm these estimates at different speeds or cycles.

137. The possibility of higher primary NO_2 emissions from heavy duty diesel vehicles equipped with CRTs, relying on the deliberate formation of NO_2 in the exhaust for regeneration of the particulate trap, also needs to be investigated, especially for operation of the vehicles over urban drive cycles.

2.6.2.7 Primary emissions of nitrous acid (HONO) from road vehicles

138. A number of studies have demonstrated that nitrous acid (HONO) is emitted in vehicle exhaust in conjunction with NO and NO_2 (for example, Pitts *et al*., 1984; Kirchstetter *et al*., 1996; Martinez-Villa, 2001; Kurtenbach *et al*., 2001). Emissions of HONO have also been quantified from a diesel-fuelled electric power generator (Gutzwiller *et al*., 2002). The results of recent studies are summarised in Table 2.3. The vehicle exhaust studies have been made at roadside locations and in road tunnels, and have generally distinguished primary emissions of HONO from its secondary formation from subsequent reactions of NO and NO_2. The European studies generally indicate that HONO accounts for approximately 0.5–1.0% of emitted NO_x. Although this is a small fraction, it is potentially significant from a chemistry point of view, because HONO photolyses efficiently to generate free radicals, possibly leading to additional NO-to-NO_2 conversion (see Section 3.3). The measurements of Gutzwiller *et al*. (2002) indicate a similar proportion of emitted HONO from a diesel-fuelled electric power generator.

Table 2.3 Summary of reported information on primary emissions of HONO.

Study	HONO/NO_x (% v/v)	emission index (mg HONO)/kg fuel	location	note
Kirchstetter *et al.* (1996)	0.29 ± 0.05	–	Caldecott tunnel, San Francisco Bay, USA	f
Martinez-Villa (2001)	1.10 ± 0.09[a]	-	Marylebone Rd, London, UK	g
Kurtenbach *et al.* (2001)	0.80 ± 0.10	88 ± 18[b]	Kiesbergtunnel, Wuppertal, Germany	h
	0.53 ± 0.08	115 ± 10[c]		i
	0.66 ± 0.20	94 ± 27[d]		j
	0.65 ± 0.24	101 ± 37[e]		k
Gutzwiller *et al.* (2002)	0.65	71	Laboratory facility	l

Notes

[a] may include a secondary contribution;

[b] corresponding emissions of 7700 ± 1100 mg NO/kg fuel and 590 ± 80 mg NO_2/kg fuel reported (⋗ 4.7 ± 0.6% v/v primary NO_2);

[c] corresponding emissions of 11900 ± 700 mg NO/kg fuel and 2000 ± 200 mg NO_2/kg fuel reported (⋗ 9.8 ± 1.0% v/v primary NO_2);

[d] corresponding emissions of 8600 ± 1700 mg NO/kg fuel and <30 mg NO_2/kg fuel reported (⋗ < 0.2% v/v primary NO_2);

[e] corresponding emissions of 9500 ± 1700 mg NO/kg fuel and 860 ± 230 mg NO_2/kg fuel reported (⋗ 5.5 ± 1.5% v/v primary NO_2);

[f] >99% petrol vehicles; <0.2% heavy duty trucks;

[g] Measurements took place in October 1999; ca. 10% HGV;

[h] 74.7% petrol cars; 12.3% diesel cars; 6% commercial vans; 6% heavy duty trucks; 1% motorcycles;

[i] MAN truck (diesel) 0-40 km hr^{-1} over 3500 m;

[j] VW Golf (diesel) 0-40 km hr^{-1} over 3500 m;

[k] VW Golf (petrol) 0-40 km hr^{-1} over 3500 m;

[l] Kubota GL-4500s diesel electric generator, maximum load 4.5 kW.

2.6.3 Emissions from aviation and airports

139. Emissions from aviation are estimated within a 1000 metre ceiling of takeoff and landing. In the NAEI, emissions are calculated from the number of aircraft movements at all the British airports (DfT, 2002a) and emission factors expressed as emissions per landing and takeoff cycle. The emission factors are taken from the local inventory studies carried out for different conurbations by London Research Centre in the 1990s (discussed in section 2.8.3), with additional aircraft emission factors taken from the 1998 inventory for Heathrow Airport compiled by AEA Technology (Underwood and Walker, 1999).

140. The Heathrow Inventory is an example of a spatially resolved inventory centred on a specific source. This section focuses on the methods used in the 1998 Heathrow Inventory to calculate emissions from the airport and surrounding area rather than on the results from the inventory itself. Annual emission rates and their spatial distribution were prepared in a form suitable for input to an atmospheric dispersion model with a view to estimating the airport contribution to ambient concentrations in the area. It is these rather than the emission figures in their own right which provide a better measure of the impact of the airport on local air quality. All the modelling and assessments discussed in this report have been based on the 1998 version of the Heathrow Inventory, but it should be pointed out that an updated version of the inventory has just been completed for 2000 which incorporates a number of significant improvements to the methodology. However, the 2000 Heathrow Inventory was not made available in time for this report.

141. The Heathrow Inventory covered emissions from the aircraft during the landing and take-off (LTO) cycle up to 1000 m in height, emissions from road traffic on the surrounding road network, emissions from airside support vehicles and emissions from heating plant used for the airport.

142. To enable pollutant concentrations at the airport to be calculated by dispersion models, the emissions from ***all*** road traffic on the surrounding road network were estimated as these make a contribution to the background concentrations at the airport. However, only a portion of the traffic is airport-related.

143. Aircraft emissions were estimated from data on the number of aircraft movements during 1998, supplied by Heathrow Airport Ltd and broken down by aircraft type and airline, together with emission factors mainly given in the International Civil Aviation Organisation Engine Emissions Data Bank (ICAO, 1999). Emissions were estimated for eight phases of the LTO cycle:

1) taxi-out;
2) hold;
3) take-off roll;
4) initial climb (i.e. wheels-off to throttleback, assumed to occur at 450 m);
5) climb out (450 m to 1000 m);
6) approach (1000 m to touchdown);
7) landing roll;
8) taxi-in.

144. Emission factors for a given engine type vary with thrust setting. The ICAO database gives emission test results for different engines in service at four thrust settings: 7%, 30%, 85% and 100%. In general, each of the LTO cycle phases was assigned one of these thrust settings. A limited amount of information was used on operational procedures where aircraft do not use full thrust during take-off. It should be noted that emissions data in the ICAO database were not primarily intended for use in emissions modelling, as they may not be representative of real-world aircraft operations. However, these were the best data available for the Heathrow Inventory. There is a paucity of emissions data from aircraft engines operating over real-world cycles and clearly this is an area that needs attention. Further research is needed into actual aircraft operations, their impact on emissions and into the way aircraft emissions are modelled.

145. Emissions from road traffic on and around the airport were calculated from traffic volumes and speeds derived from the Heathrow Road Traffic Model (HRTM). This covered both airport- and non-airport-related traffic. Emissions from road traffic inside and outside the airport perimeter roads were calculated. The traffic model covered an extensive road network on and around the airport including sections of the M4 and M25 (Underwood *et al*., 1994). The traffic data were used in conjunction with NAEI road vehicle emission factors.

146. Emissions were spatially distributed in three dimensions by representing different sources as point, line and area sources. For aircraft emissions, taxi and hold routes to and from runway were used. Emissions for initial climb, climb-out and approach from and to runway were spatially distributed according to representative angles of climb and descent for each aircraft weight category. Road vehicle emissions from free-flowing traffic were distributed on a road link-by-link basis as line sources. Car park emissions were treated as area sources.

147. A portion of the aircraft and road traffic emissions included in the inventory occur at some distance horizontally away from the airport. Estimates of annual emission totals for the airport therefore reflect the fairly arbitrary choice of spatial boundaries used in the inventory, for example, the extent of the road network covered. The Heathrow Inventory study did produce emission totals by source category for sources designated as 'airport-related' even though much of these occur outside the physical domain of the airport. The 1998 Heathrow Inventory showed that over half of the NO_x emissions designated as airport-related occur from the aircraft during take-off and landing, but much of this will be at some distance from airport ground-level. Around a third of all NO_x emissions from the aircraft (including ground-level emissions from auxiliary power units, engine testing etc, as well as take-off and landing) occur below 100 m in height. The remaining two-thirds occur between 100 and 1000 m and contribute little to ground-level concentrations. Receptor modelling studies based on dispersion models and the 3-dimensionally resolved emission inventory show the impact of airport activities on ground-level NO_2 concentrations. Studies have shown that although emissions associated with road traffic are smaller than those associated with aircraft, their impact on population exposure at locations around the airport are larger (Farias and ApSimon, 2003).

148. An updated version of the Heathrow emissions inventory has just been compiled by AEA Technology for the 2000 calendar year, using more detailed aircraft activity information. The new inventory is based on a number of improvements to the data and methods used which have led to a reduction in the estimates of the amount of NO_x emitted from aircraft at Heathrow compared with the estimates made in the 1998 inventory. The new inventory has accounted for the usage of reduced thrust during take-off more fully than had been possible for the 1998 inventory and assumptions about reverse-thrust usage have been updated. Other improvements have included information on Auxiliary Power Unit running times. The improvements have led to an overall 38% reduction in the estimate of NO_x emissions from aircraft at Heathrow relative to the estimates made in the 1998 inventory. This reduction was made up of a 19% reduction in aircraft emissions at ground-level and 46% reduction in elevated emissions. It should be emphasised that these reductions mark improvements in the inventory estimate due to methodological improvements, not to actual reductions in emissions over this time period.

2.6.4 Emissions from shipping

149. A recent exercise to quantify and map emissions of NO_x from shipping across the EU (including emissions in the North Sea, English Channel, Irish Sea and in-port sources) has been undertaken by ENTEC for DG Environment for the year 2000 (ENTEC, 2002). This used activity data from the Lloyds Maritime Intelligence Unit on ship movements and considered seasonal variations in activity through selecting data for January, April, July and October indicative of winter, spring summer and autumn operations. Other sources of information used were: vessel characteristics (vessel type, engine size and type), a detailed shipping emission factor database developed by the Swedish Environmental Research Institute (IVL) (ENTEC, 2002), data reflecting the time spent in port from a questionnaire survey and data on fuel consumption from fishing vessels, fishing grounds and the size of catches. The study covered all ships greater than 500 gross tonnes. Typical NO_x emission factors for these vessels currently range from 13 to 19 $gkWh^{-1}$, approximately 3 to 4 times higher than those associated with a Euro III heavy-duty road vehicle.

150. Estimates were made using a GIS methodology and the results presented on the 50 x 50 km EMEP grid. The study showed that two British ports were ranked in the top 10 highest emitters of NO_x in the EU, Milford Haven and Immingham, with estimated emissions of 2 ktonnes and 1.3 ktonnes, respectivelty, in 2000. Within the ENTEC study area covered (North Sea, Irish Sea, English Channel, Baltic Sea, Black Sea and the Mediterranean), it was estimated that NO_x

emissions are likely to equal two thirds of land emissions by 2010. The European Union is thus working with member states to encourage tighter emission standards through the International Maritime Organisation. The model for these proposed emission reductions may already be seen in the Baltic area with fiscal benefits awarded to ships meeting low SOx and NO_x standards. With respect to shipping the dominant technology for NO_x reduction is currently SCR with urea injection, as being developed for heavy duty road diesel engines (see the Technical Annexe, Appendix 1).

151. In the NAEI, total UK emissions from shipping cover fishing and coastal shipping in UK waters. NO_x emissions are estimated from DTI fuel consumption data for this sector and emission factors from CORINAIR (1999).

2.6.5 Emissions from rail transport

152. Limited information exists for the emissions associated with rail locomotives. In the UK, less than 50% of the rail network is electrified and thus the use of diesel locomotives is common. With respect to NO_x, diesel locomotives have an emission rate of between 6 and 16 $gkWh^{-1}$. The most recent studies (ARTEMIS) on these emissions have been undertaken by the Danish Technical University and involve the development of a methodology that attempts to incorporate fundamental physical factors for determining driving resistance of the train as a function of speed, load and weight. This is subsequently used to calculate the energy required to move the train, and propulsion efficiencies are applied for diesel engines and transmissions. Operation is divided into elements in an acceleration-velocity matrix, and the driving resistance calculated for each condition in the matrix. Based on a distribution function for the frequency of occurrence of each condition, the total energy consumption of the train for a given driving pattern is evaluated. Since the frequency of driving conditions is used, the method can be applied to a wide range of conditions, from a single train on a single route to an entire fleet of trains of a given type averaged over a large geographical scale. In order to facilitate the use of available operation data, the model can use either temporal or spatial distribution of the operating conditions. The results of this method have been favourably compared to measurements from a small number of Danish trains (Hickman, 1999).

153. In the NAEI, emissions from railways cover emissions from the stationary combustion of oil and gas by the railway sector and emissions from diesel trains. Emissions from generation of power consumed by the railway sector for electric traction is reported under the ‘Public Power’ sector, following emission reporting guidelines. Emissions from stationary combustion are based on emission factors and fuel consumption data for the sector. Emissions from diesel trains are estimated for Intercity, regional and freight trains using railway kilometre data, gas oil consumption data for the sector and emission factors for different types of diesel locomotives used for each type of journey (Goodwin *et al*., 2001). All the fuel consumption data are from DTI (2002).

154. The NAEI maps UK emissions from the rail sector according to rail track data. No specific or detailed inventories of emissions from local rail operations have been generated. Emissions from diesel locomotives represent 0.6% of the total UK inventory of NO_x emissions and are not likely to pose significant urban air quality problems except possibly in the immediate vicinity of major rail terminii. Without improvements in the emission performance of diesel locomotives, it is conceivable that emissions from the rail sector will become a more important issue in transport inter-modal comparisons as emissions from road vehicles continue to decline. However, greater electrification of the rail network will lead to reduced emissions of NO_x from trains at the point of use.

2.6.6 Emissions from other mobile machinery

155. Emissions from off-road sources and mobile machinery cover a range of portable or mobile equipment powered by diesel or petrol engines. They include agricultural and forestry equipment such as tractors and combine harvesters; construction equipment such as bulldozers and excavators; industrial machines such as portable generators, compressors and forklift trucks; aircraft support vehicles; and domestic house and garden equipment such as lawn mowers. In the NAEI, emissions and fuel consumption are estimated from population and machinery lifetime data from a DETR sponsored survey covering around 70 classes of machinery, power rating, load and annual usage data for each class of machinery and emission factors (Goodwin *et al*., 2001; Murrells and Salway, 2000). The emission factors for each machinery class mostly come from CORINAIR (1999) and Samaras (1996) and are expressed in g NO_x (as NO_2).kWh^{-1}. Where possible, the emission estimates are normalised according to fuel consumption data available from DTI for certain groups of machinery or estimated from other surrogate activity factors.

2.6.7 Primary emissions of NO_2 from non-road transport combustion sources

156. Whilst there is no evidence for significant emissions of primary NO_2 from other combustion sources, the presence of primary NO_2 emissions in diesel vehicle exhausts suggests this should be verified in certain other key combustion sources where ground-level exposure could be significant. For example, it is conceivable that diesel locomotives could emit significant amounts of NO_x as primary NO_2 while their engines are idling at low load in major rail termini which are often in enclosed spaces. Levels of primary NO_2 emitted in domestic burners and combustion appliances should be monitored to determine whether these sources, which can be significant sources of total NO_x emissions in urban areas, produce significant amounts of primary NO_2.

2.7 Time series of NO_x emissions in the UK

2.7.1 UK emissions of NO_x from 1970 to 2000

157. The NAEI time series of UK emissions of NO_x are shown in Table 2.4 and were depicted in Figure 2.1. The sector 'other' includes combustion in agriculture, waste incineration, but is mainly emissions from small boilers in the commercial and public sector.

158. Based on the 2000 NAEI figures (Goodwin *et al*., 2002) using the latest vehicle emission factors (Table 2.2), road transport is the largest source of NO_x emissions in the UK, contributing 49% of total emissions. Public power generation is the next largest source, contributing 21% of UK emissions, followed by industry (14%) and other transport and machinery (10%).

Table 2.4 UK emissions of NO_x (ktonnes as NO_2).

ktonnes NO_x	1970	1980	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000
Public power	812	861	781	683	671	567	527	495	449	372	364	338	358
Industry	524	372	337	328	323	319	339	291	283	285	283	266	250
Domestic	62	64	64	71	69	72	69	66	75	69	71	71	72
Road transport	762	978	1286	1257	1208	1141	1111	1063	1062	1020	953	910	844
Other transport & machinery	241	220	220	224	218	212	199	194	204	197	187	176	169
Other	90	73	56	57	53	44	47	46	46	40	38	37	35
TOTAL	**2490**	**2568**	**2744**	**2620**	**2543**	**2355**	**2290**	**2154**	**2120**	**1983**	**1897**	**1799**	**1728**
Road transport – using old vehicle emission factors			*1305*	*1274*	*1224*	*1147*	*1084*	*997*	*956*	*881*	*788*	*716*	*629*
Totals submitted to UNECE			*2763*	*2637*	*2558*	*2361*	*2263*	*2088*	*2014*	*1844*	*1732*	*1604*	*1512*

Source: 2000 National Atmospheric Emissions Inventory. The road transport figures and emission totals shown in bold are based on the latest set of vehicle emission factors which were not available at the time of formal submission of the UK 2000 emission estimates to UNECE. The figures shown in italics are based on the old emission factors used for the UK submission of the 2000 NAEI data.

159. Emissions have fallen steadily since their peak levels in 1989, with a reduction of around 38% since then. The decline has mainly been due to reductions in emissions from road transport and public power generation; these fell by 34% and 54%, respectively, from 1990 to 2000. The reductions in road transport emissions have been mainly due to the penetration of cars fitted with three-way catalysts in the fleet. Emissions from power stations were fairly constant in the 1970s, but have been declining since 1979. In the 1980s this was due to the increased use of nuclear power and an increase in average efficiency of thermal power stations. However, since 1988, the electricity generators have adopted a programme of progressively fitting low-NO_x burners to their large coal-fired stations. The introduction of modern CCGT plant burning natural gas in the early 1990s and the decreased share of coal for power generation in favour of gas, further reduced NO_x emissions from this sector up until 1999. Emissions of low-NO_x gas turbines are lower than those of pulverised coal fired plant even when these are fitted with low-NO_x burners. However, this trend was broken in 2000 when output from coal-fired stations grew again at the expense of nuclear power production. Further growth in electricity output from coal-fired stations occurred in 2001, this time at the expense of gas possibly due to increased price of natural gas supplies.

160. Emissions from industry have declined by 26% since 1990, again mainly due to the decline in coal use in favour of gas.

161. It should be noted that the figures in Table 2.4 are based on the latest vehicle emission factors which were not available at the time the 2000 reported version of the National Atmospheric Emissions Inventory (2000 NAEI) was compiled in 2002. Using the new vehicle emission factors increased the emission estimates for this sector due to the higher factors for Euro I and Euro II diesel vehicles than had been used previously (Section 2.6.2.1). Table 2.4 also shows the road transport emission estimates and total UK NO_x emissions from 1990 to 2000 that were reported in the 2000 NAEI derived from the old vehicle emission factors (emissions from all other sectors are unchanged).

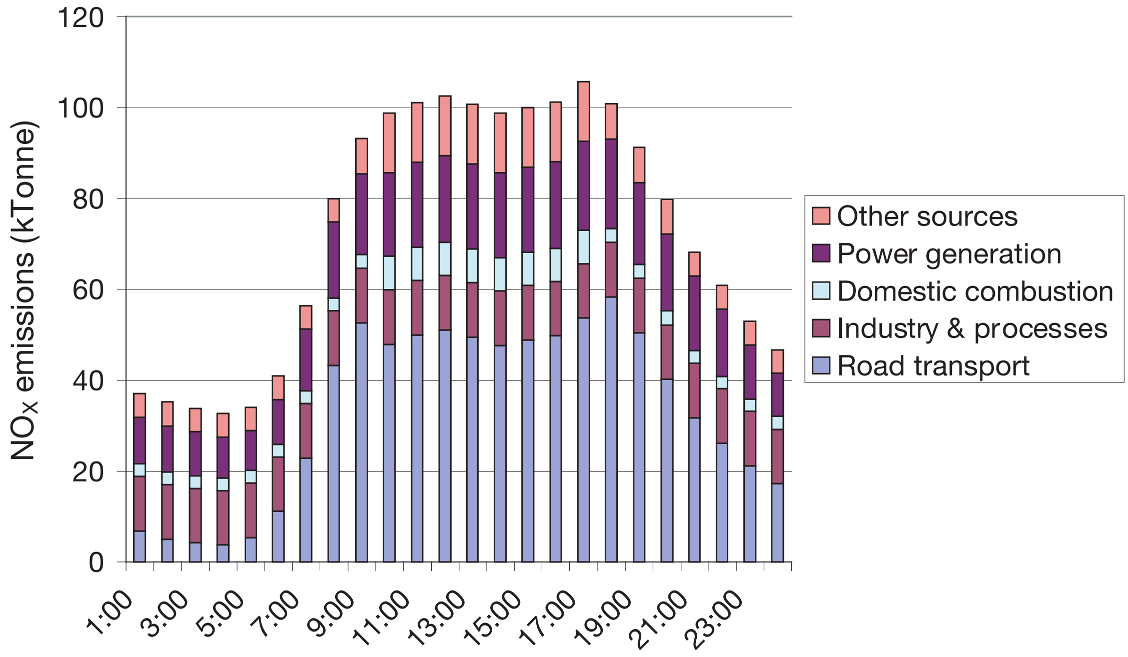
2.7.2 Temporal dependence of NO_x emissions

162. The emission figures shown in Table 2.4 refer to annual emission rates (ktonnes per year). Emissions vary with time of day and, for some sectors, with time of year and day of the week. The temporal dependence of emissions of NO_x and other O_3 precursors has been estimated by Jenkin *et al*. (2000) using the temporal activity profiles for key sectors like road transport and power generation. For road transport, hourly and daily average traffic flow data were used, taken from the DfT's Road Traffic Statistics publications. For power generation, hour of day and day of week profiles were based on activity data from the National Grid.

163. The diurnal variation in NO_x emissions is particularly significant, reflecting the varying nature of combustion activity associated with industrial, domestic and transport activity during the course of the day. Figure 2.6 shows the annual rates of NO_x emissions for each hour of the day broken down by sector. Emissions rise rapidly between 6 and 9am, remain fairly stable during the course of the day and fall off rapidly after 6pm. This diurnal behaviour is mainly because of the increase in road traffic activity between 6am and 6pm, but increased domestic combustion and power generation during the day time also contribute to the diurnal emission trends.

164. Emissions tend to be slightly lower in the summer months, owing to a decrease in the contribution made by space heating. The emissions are substantially lower (20-30%) at weekends compared with weekdays, owing to reduced contributions from a number of sources, but especially road transport. The results from this study were used by Jenkin *et al*. (2000; 2002) to model the formation of O_3 along trajectories arriving at rural sites in the UK at different days of the week during the conditions of an O_3 episode.

Figure 2.6 Diurnal variation in UK NO_x emissions (from Jenkin *et al*. (2000)).



2.8 Spatially resolved inventories of NO_x emissions

165. The NAEI produces national maps of NO_x emissions for the UK. While these will not replace detailed local inventories, they can provide a valuable basis from which a local inventory can be built and validated. They can also be used in assessments required for national policy making.

166. This section outlines the methods used to map national emissions data from the NAEI. There are many examples of local inventories that have been developed for specific areas of the UK, but one of the most recent and comprehensive local inventory is the London Atmospheric Emission Inventory (LAEI). A comparison between mapped estimates of the LAEI and the NAEI is provided in this section as a means of illustrating the effects of the different spatial inventory modelling approaches. Some indicative examples of the range of other local inventory activity in the UK are provided in this section.

2.8.1 The NAEI 1x1 km UK emission maps for NO_x

167. The NAEI national emissions inventory for NO_x is mapped over the UK on a 1x1 km grid. Figure 2.7 shows the map for UK NO_x emissions in 2000. Emissions are represented in tonnes of NO_x (as NO_2) per 1 km grid square. Details of the methods used for mapping emissions in the NAEI are given in Goodwin *et al*. (2001).

168. Sectors such as power stations, refineries and large industrial plant can be represented as point sources because their locations are known and reported emissions data or data to estimate emissions are available for the plant. Fuel consumption data for individual plants may be used as a surrogate to distribute national emissions from industrial combustion if emissions data are not directly available.

169. Emissions from road transport, rail and shipping are represented by lines where data are available, while source sectors such as domestic and commercial combustion are treated as areas. Emissions from road transport are mapped by treating major and minor roads separately. Major roads are treated as line sources using traffic flow data on road links in the national road network recorded each year. The traffic flow data come from the DfT 3 year rolling survey and the Annual Traffic Census Report for Northern Ireland. Emissions are calculated on each link using emission factors, traffic flows for the different vehicle types and average speeds appropriate for the road or area type. Emissions are mapped according to the Ordnance Survey grid reference for the road link. Minor roads are treated as area sources because centralised traffic flow data are not available for these roads. Emissions on minor roads are estimated at a national level and then distributed using surrogates derived from OS minor road maps, land cover data and regional average traffic flows on these types of roads.

170. Emissions from rail are mapped using track and train kilometre data available for each rail link. UK shipping emission totals are distributed according to port areas and routes defined by shipping arrival data from DfT. Aircraft emissions are treated as area sources for the 1x1 km grids covering the airports where take-off and landing occurs. The methods incorporate airport emission estimates from the London Research Centre (LRC) urban inventory studies and aircraft emission estimates from the Heathrow Inventory study. The number of aircraft movements at individual UK airports is used.

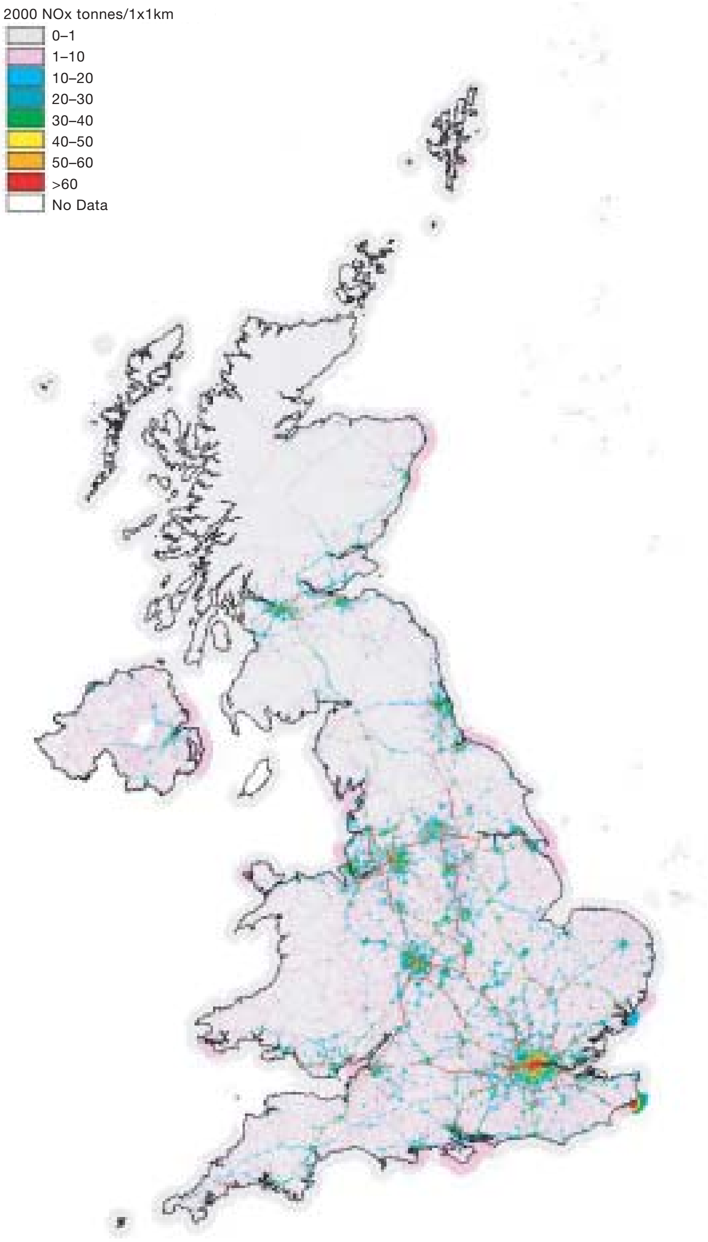
171. Emissions from domestic, small industrial and commercial combustion sources and off-road vehicles and machinery are treated as area sources. National totals for these sectors are distributed over the UK based on surrogate statistics represented in area grids. Area grids are compiled from geographical statistical data available on land use, population and employment.

172. The emission map shown in Figure 2.7 is available on the NAEI website (www.naei.org.uk/index.php) where the user can extract emissions for a particular area.

173. The frequency distribution of NO_x emission values is strongly positively skewed and it is important to note that a large fraction (around 30%) of the total emissions of NO_x are concentrated in a few grid squares which contain major combustion-related point sources. Road transport dominates total NO_x emissions with around one-third of total NO_x emissions occurring on major sections of road. Vehicles travelling at high speed contribute most and as a result the major motorways and primary routes are clearly defined on the map. Conurbations and city centres show high emissions resulting from large volumes of traffic, residential and commercial combustion. A combination of relatively high national shipping emission estimates and relatively few large ports result in significant localised emissions from shipping in port areas.

174. For a particular grid square, the difference in emissions derived from the national map and a detailed local inventory where available give a measure of uncertainty in the UK emission maps.

Figure 2.7 UK NO_x emission map from the NAEI.



2.8.2 NO_x emissions in London

175. The London Atmospheric Emissions Inventory (LAEI) was developed for 1999 by the Greater London Authority with Transport for London (TfL) and King's College, London (GLA, 2002). Recognising the importance of traffic as a source of emissions in London, this inventory used a great deal of detailed traffic activity data, but also other London-specific data for the inventory covering an area within and including the M25.

2.8.2.1 Overview of the London Atmospheric Emissions Inventory

176. The methodology for estimating road transport emissions in the LAEI recognises the importance of road traffic as a source of air pollution in London. Since, it is known that exceedences of air pollution objectives in London will increasingly be restricted to the near-road environment, the LAEI places considerable importance on the calculation of individual link emissions. It has therefore been produced primarily with dispersion modellers in mind and as a source of data for air pollution modelling runs in London. London is also unique in the UK in many respects:

- It is the largest urban area in the UK;
- Road traffic emissions are relatively more important than in many other urban areas in the UK;
- London has a unique vehicle stock, for example, TfL buses and taxis;
- There is a considerable amount of information available from which calculations of emissions can be made.

2.8.2.2 Calculating emissions from traffic in the LAEI

177. Most emphasis has been placed on observed manual counts of traffic in London, since these are more reliable on a link-by-link basis than traffic models or ad-hoc measurements. However, it is strongly linked with the LTS traffic model to assist with future traffic scenarios for London, for example, redistribution of traffic as a result of road user charging.[1] The LAEI uses count data for 11 different vehicle categories including taxis.

178. The analysis of on-road vehicle age data highlights significant variations in vehicle age by road type in London compared to the UK fleet represented in the NAEI model. It was found that in London, there is a slightly newer vehicle stock on motorways on average and older vehicle stock on minor roads compared with the national fleet. This was therefore accounted for in the inventory, although the overall effect was very small. Further investigations will take place into the age profiles of vehicles in London and will be included in future inventories.

179. The LAEI uses the same speed-dependent emission factors as the NAEI, but combines them with realistic speeds in London. The current inventory uses detailed speed data from actual measurements of speed derived from the 'floating-car' technique (Roland, 1998).

180. London buses and taxis make an important contribution to traffic flows in London, particularly central London, and are therefore important contributors to local NO_x emissions. Bus flow data for links were provided by DfT, but TfL Buses gave additional vehicle kilometre information (by location in London) as well as vehicle technology profiles of the TfL bus fleet. The bus vehicle stock was broken into two parts, central London (defined by LTS) and other London stock representing all other locations in London. LT bus services were assumed to represent 90% of the bus vehicle

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[1] This feature is retained because traffic models can be very useful in suggesting the likely future distribution of traffic, for example, greater growth in outer London compared with central London.

kilometres in London (TfL London Buses). Bus vehicle stock was then broken down by Euro class and included those vehicles fitted with an oxidation catalyst or particulate trap.

181. Taxi movements are important in London and particularly central London. Data relating to taxi activity in London has been analysed from several different sources including count data from London boroughs and specific studies in London concerned with taxi activity. Information provided by the Public Carriage Office enabled a specific breakdown of the taxi fleet by Euro class, the number of alternatively fuelled vehicles in the fleet, and an estimate of vehicle stock turnover year-on-year. An analysis of these data suggested that in central London taxis made up approximately 20% of the total car plus taxi total. In inner London the proportion was assumed to be 4% and 1% in outer London.

2.8.2.3 Calculating emissions from non-traffic sources in the LAEI

182. These calculations have mostly been made by the GLA, based on the original LRC inventory for London. Some modifications have been made to update these emissions.

183. For Heathrow, the inventory compiled by AEA Technology has been used (Underwood and Walker, 1999). Any overlap in emissions with the rest of the LAEI, i.e. some road emissions, were removed from the Heathrow Inventory. Various simplistic reductions were applied to some emission sources for future years.

184. Updates were also made to emissions of NO_x from natural gas combustion (domestic, commercial and small industrial). These emissions were increased in line with projected increases in natural gas use in London. These projections suggest that natural gas use continues to increase significantly year-on-year in London.

2.8.2.4 Results from the LAEI in comparison with data for London from the NAEI

185. Table 2.5 shows the amount of NO_x emitted in London calculated in the LAEI and the contribution from road traffic. Also shown are the results from the NAEI UK emission maps derived by putting a boundary around the M25, so including the same geographical area. The NAEI data refers to emissions in 2000, while the LAEI data is for 1999. It can be seen that the LAEI estimates 14% less NO_x than the NAEI, but the contribution from road traffic is not so different in each inventory. The LAEI estimates 64.7 kt of NO_x are emitted from traffic in London (in 1999); the figure from the NAEI is 73.0 kt (in 2000).

Table 2.5 Emissions of NO_x in London. Comparison of data from the LAEI and the NAEI. The LAEI data are for 1999; the NAEI data are for 2000. Both inventories are based on the new vehicle emission factors.

	Total emissions in London (ktonnes)		**% road transport emissions in London**		**% road transport emissions in UK**
	LAEI 1999	NAEI 2000	LAEI 1999	NAEI 2000	NAEI 2000
NO_x	94.7	110.3	68.3	66.2	49

186. However, comparing results from the two inventories is complicated by the fact they refer to different years. According to the NAEI, urban UK road transport emissions in 1999 were 7.5% higher than in 2000 due to the lower proportion of vehicles meeting the latest Euro standards in the fleet. If it is assumed this also applied to London, then the NAEI estimate for NO_x emissions from road transport in London in 1999 would be 78.5 kt, an increase of 5.5 kt above

2000 levels. If it is also assumed that emissions from other sources are unchanged from 1999 to 2000, then an NAEI estimate for total NO_x emissions in London in 1999 would be 115.8 kt. The LAEI estimate for 1999 would then be 18% lower than the NAEI estimate. However, the percentage contribution from road traffic based on the NAEI estimate for 1999 would be 67.8%, very close to the LAEI estimate.

187. Given the differences in the inventory approaches, with the LAEI using much more detailed and localised activity data than the NAEI map, the differences in emission estimates are not disturbing. In both cases, the contribution from road transport is higher in London than it is for the UK, as would be expected. It is worth noting that the road traffic contribution estimated for London of around 68% in 1999, is slightly down on the contribution previously estimated for 1996 of around 75% in the inventory developed by London Research Centre (Buckingham *et al*., 1998a). Comparing the LAEI and the earlier LRC inventory for London is complicated by the fact that different vehicle emission factors were used in each inventory. However, a significant drop in road transport emissions from 1996 to 1999 would have been expected due to the fleet penetration of newer and cleaner vehicles, accompanied by smaller reductions in emissions from other sources. This would lead to a decline in the traffic contribution to overall NO_x emissions in London. The LRC estimate for non-road transport emissions in London in 1996 was 36.7 kt compared with the LAEI figure for 1999 of 30.0 kt.

188. The contributions of different sources to emissions in London in 1999 are compared with the corresponding figures for the UK derived in the NAEI for the same year in Table 2.6. All the estimates are based on the new vehicle emission factors.

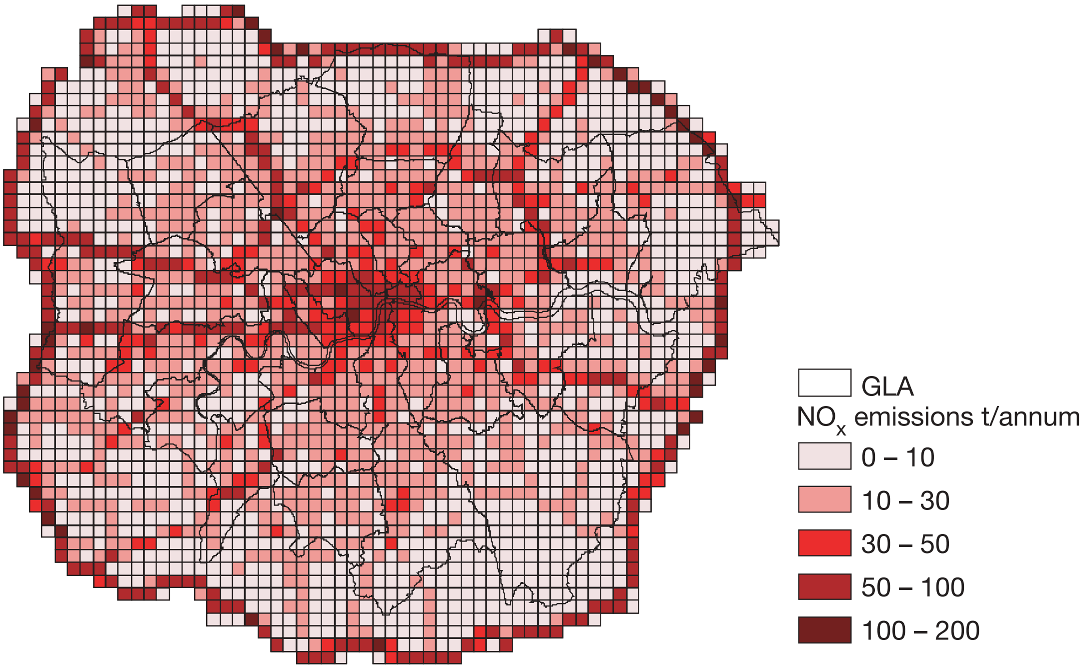
Table 2.6 Contribution of sources to NO_x emissions in London and the UK in 1999.

	% emissions in London LAEI	% emissions in UK NAEI
Road transport	68	50
Domestic and commercial combustion	17	6
Industry and public power	8	34
Other transport	6	10
Other	1	0

189. It can be seen that road transport and domestic and commercial combustion make a much bigger contribution to NO_x emissions in London than they do in the UK as a whole, as would be expected in a large conurbation. Industry and public power generation make a much smaller contribution to NO_x emissions in London than in the UK due to the lack of major industrial combustion plant in London. It should be noted though that the LAEI and the NAEI make slightly different definitions in terms of what is classified as ‘commercial’ and ‘industrial’ combustion processes, but this does not change the overall pattern of sector contributions to emissions shown in Table 2.6.

190. Figure 2.8 shows the contribution of road transport to total NO_x emissions in each grid square in London in 1999, according to the LAEI. It can be seen how much higher the contribution from traffic is to emissions in central London compared with outer parts of London, although the relatively high emissions from important radial roads and the M25 orbital can also be clearly seen.

Figure 2.8 NO_x emissions from road transport in London (1999).



191. The LAEI also addressed the contribution of traffic to emissions in central and Greater London separately and also the contribution made by different vehicle types. Overall, traffic made a similar (~60%) contribution to NO_x emissions in both central (defined by the inner ring road) and Greater London, but the contribution from different vehicle types was different in each area. In central London, buses and taxis made a larger contribution to NO_x emissions (~21% and 11%, respectively) than they did in London as a whole (7% and 1%), with a smaller proportion of emissions coming from cars. The contribution from buses is significant in spite of the fact that the bus fleet in London is among the cleanest in the UK. Cars are responsible for nearly a half of all NO_x emissions from road transport in London, with around 40% coming from vans and HGVs. Figures 2.9 and 2.10 summarise the percentage contribution to total emissions by vehicle type in central London and all of London (up to and including the M25).

192. The geographic variation in emissions might have important consequences for how emissions are expected to reduce. Section 2.6.2.1 showed that NO_x emissions from diesel vehicles have been declining more slowly with the introduction of the tighter emission standards than has been occurring with petrol vehicles. In central London, where there is a high proportion of diesel buses and taxis, the decline in emissions of NO_x in this area will not be as significant compared with other locations in London.

193. Any further emission inventories compiled for London would need to consider the effect of the recently introduced congestion charging scheme on traffic emissions. A report ‘Road Charging Options for London: A Technical Assessment’ was published in 2000 and concluded that the overall air quality benefits from fewer private cars would be offset by the impact of larger numbers of heavy duty vehicles. This assessment would need to be verified using real traffic flow data in the areas effected once the scheme has been operating long enough. Similarly, current work has investigated the ***potential*** emission benefits of a London Low Emission Zone of the type discussed earlier, restricting access to low emitting vehicles (see the London LEZ website, www.london-lez.org). However, emission inventories developed for London in the future would be able to show the actual benefits a scheme has had on emissions using actual traffic flow data available once the scheme has been implemented.

Figure 2.9 Percentage of total NO_x emissions from road transport in central London by vehicle type in 1999 (LAEI).

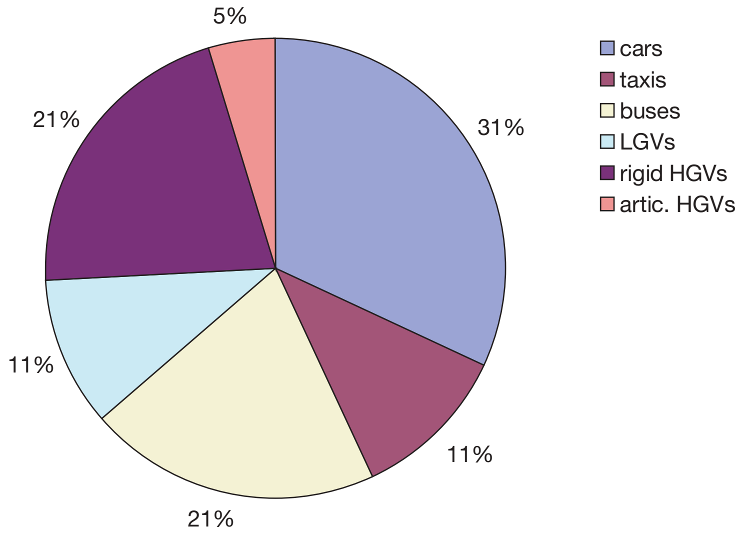
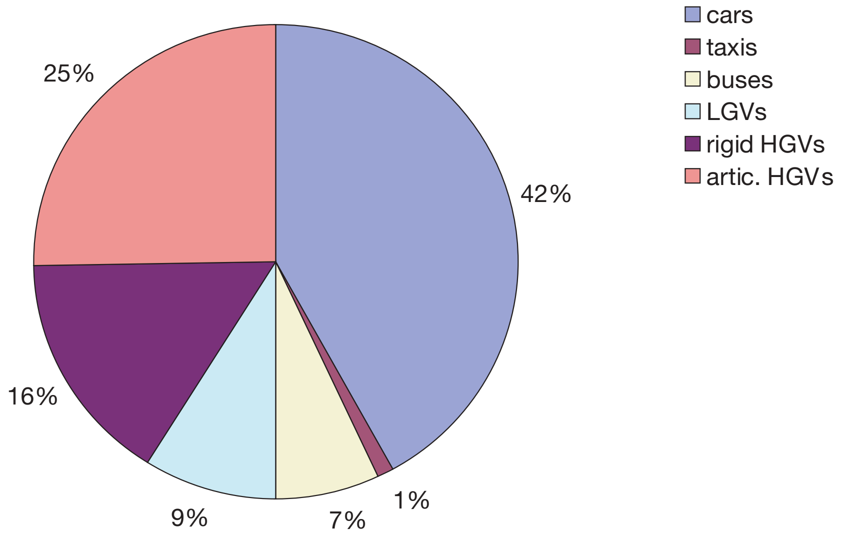


Figure 2.10 Percentage of total NO_x emissions from road transport in the whole of London by vehicle type in 1999 (LAEI).



2.8.3 Other local emissions inventories

194. A number of urban inventories have been compiled over the years, notably the ones produced by London Research Centre for several conurbations in the UK during 1995 and 1996: the West Midlands 1994; Merseyside 1995; Greater Manchester and Warrington 1995, Portsmouth and Southampton 1995; Bristol and Avonmouth 1995; Port Talbot and Swansea 1996; Middlesbrough 1996; City of Glasgow 1996; and Urban West Yorkshire 1996 (Hutchinson and Clewley, 1996; Buckingham *et al.,* 1997a; b; 1998b). As part of the same 'Ten Cities' initiative a further inventory was subsequently developed for the Greater Belfast area (1999). All of these inventories used 'bottom-up' methodologies for key sources, such as road transport and industry.

195. Greater Manchester, like a number of the urban areas for which emission inventories were produced in the 1990s, has continued to update and further develop inventory methodologies. The latest emission estimates available are for 1999 (aric, 2000) and these have been used for several local air quality management tasks including: Stage 3 and 4 Review and Assessment (Peace *et al.,* 2002a; b); air quality management planning; land use planning and the development of transportation policies.

196. Other detailed emission inventories have been produced independently of the Ten Cities initiative. Although not an exhaustive list, some examples include Sussex, Kent, Nottinghamshire (for 2001), Slough (for 2000), Cheshire (for 1998) and Stoke-on-Trent & Newcastle-under-Lyme. In addition to providing a foundation for dispersion modelling, the main benefit of compiling local inventories is the detailed examination of particular sources, such as specific industrial processes, that are known to be influential in a local context but which may not be well treated in the national inventory due to a lack of consistent national data.

197. In view of the importance of specific local sources, detailed emission inventories are also produced for discrete source sectors. Examples of spatially resolved airport inventories, include both major international airports such as Heathrow and Manchester (for 2000) and also regional airports such as Birmingham, Newcastle, East Midlands and Bristol (all for 2000). Emissions from shipping have been mapped across the EU, including the coasts and ports around the UK. These were discussed earlier.

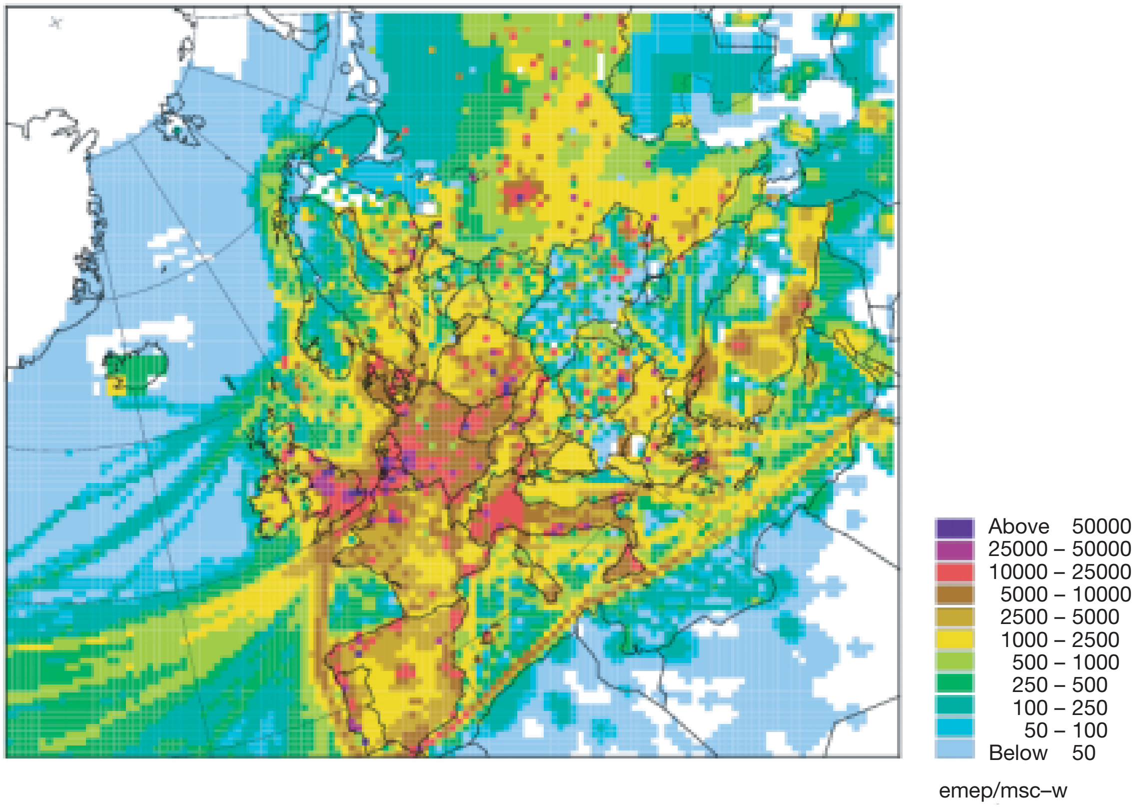
198. Local Authorities should refer to the Local Air Quality Management Technical Guidance Notes (LAQM, 2003) available on the Defra and Devolved Administration's websites when considering the requirements of an emissions inventory for their area. In the UK, the master source of information on emissions is the National Atmospheric Emissions Inventory (NAEI) providing emission maps on a 1x1 km grid. Local Authorities should use the emissions data within the NAEI first, and only compile their own inventory if these data are likely to be inadequate.

199. In many cases, compiling a complete local inventory from start is inappropriate and can be extremely time consuming and difficult without previous experience. This can lead to the generation of local inventories of varying degrees of quality. Using a common approach and methodology is crucial if inventories for different areas are to be comparable. However, where air quality exceedences are likely to occur as a cumulative result of a large number of different sources then the compilation of a more complete, local inventory may be useful.

2.9 NO_x emissions in Europe

200. Emission inventories for NO_x are compiled for other countries in Europe as required under UNECE/EMEP and IPCC reporting protocols. Table 2.7 shows the latest reported emission totals for NO_x in each of the EU15 countries, plus Norway and Switzerland and four central and Eastern European countries for which recent data are available (EMEP, 2002). Also shown are the emissions ceilings that apply to each country in 2010 (Official Journal, 2001). Table 2.7 also shows the data expressed as emissions per capita and per GDP in each country. Figure 2.11 shows the EMEP 50x50 km map of NO_x emissions in Europe for 2000.

Figure 2.11 Map of NO$_x$ emissions in Europe in 2000 on a 50x50 km grid (EMEP 2002). Units are in tonnes NO$_x$ as NO$_2$.



201. The emission figures in Table 2.7 refer to emissions in 1999, the most recent year data are available for most countries. For Greece and Portugal, the data are for 1998. The UK reports emissions data from the NAEI, but the figures shown are corrected for the latest vehicle emission factors. The table shows that all countries (except Bulgaria) need to reduce their emissions to achieve their emission ceilings, typically by around a third from 1999 values across all the EU15 countries. Emissions vary quite significantly on a per capita and per GDP basis. Emissions per capita vary from 13.9 kg NO$_x$ per capita in Switzerland to 51.9 kg NO$_x$ per capita in Norway. Emissions per GDP in western European countries vary from 0.41 tonnes NO$_x$ per million ECU in Switzerland to 3.5 tonnes NO$_x$ per million ECU in Portugal. The variation in these figures by country reflects the amount of economic, industrial and transport activity, fuel mix for energy production and amounts of emission abatement in each country. The low levels in Switzerland may be due to the amount of power generated by hydroelectric sources. The UK figure of 30.3 kg NO$_x$ per capita is fairly close to the EU15 average value of 27.5 kg NO$_x$ per capita. The emissions per GDP figure for the UK is also close to the EU15 average.

Table 2.7 Emissions of NO_x in Europe

	NO_x emissions (ktonnes as NO_2) in 1999	Emissions ceilings for 2010 (ktonnes as NO_2)	Emissions per capita (kg/capita)	Emissions/ GDP (tonnes/ million ecu)	% emissions from road transport sector	% emissions from energy production sector
Austria	172	103	21.3	0.87	50	5
Belgium	291	176	28.5	1.24	51	12
Denmark	210	127	39.6	1.27	33	27
Finland	247	170	47.8	2.05	46	13
France	1533	810	26.0	1.14	47	9
Germany	1637	1051	20.0	0.83	51	15
Greece	382	344	36.3	3.26	27	20
Ireland	119	65	31.7	1.33	42	33
Italy	1486	990	25.8	1.34	50	13
Luxembourg	16	11	37.5	0.87	42	1
Netherlands	409	260	25.9	1.09	42	12
Portugal	381	250	38.2	3.52	43	43
Spain	1379	847	35.0	2.44	-	-
Sweden	261	148	29.5	1.15	42	5
United Kingdom	1799	1167	30.3	1.31	51	19
EU 15	10321	6519	27.5	1.29	41	13
Norway	231	156	51.9	1.60	24	12
Switzerland	99	79	13.9	0.41	54	1
Bulgaria	203	266	25.3	-	26	31
Czech republic	391	286	38.0	-	43	21
Hungary	221	198	22.1	-	52	22
Poland	953	879	24.7	-	30	28

Emissions data are from EMEP (2002) and are for 1999, except for Greece and Portugal which are for 1998. The figures for the UK are taken from the NAEI, but corrected for the latest vehicle emission factors.

Emissions ceilings for all the EU15 countries are the NECD ceiling levels (Official Journal, 2001). For all other countries, the ceilings refer to limits agreed under the Gothenburg Protocol (UNECE, 1999)

202. It is also interesting to look at the breakdown in NO_x emissions by sector in each country. These are shown in Table 2.7 for the main sectors of emissions from road transport and energy production. Again, there is fairly significant variation in the sectorial split. For most countries in western Europe, the contribution made by road transport is in the region 40-50%, although there are some outliers like Denmark, Greece and Norway where the contributions are lower (33%, 27% and 24%, respectively). The contribution from the energy production sector is far more variable, probably reflecting the fuel mix and levels of emission abatement used for power generation, as well as variable contributions from other sectors, for example, industrial combustion. Particularly low contributions to emissions from power generation in Austria and Switzerland are probably due to the amount of power generated by hydroelectric sources, while low levels in France may reflect the use of nuclear energy for power generation.

2.10 Uncertainties in emission estimates

203. An appreciation of the uncertainties in emission estimates is important for different reasons, depending on the geographical scale of the inventory, detail in the emission sector split and intended application of the inventory results. For a particular area, an inventory is usually reported as tonnes emitted over a certain period of time, normally a calendar year. An inventory calculated for more than one year is useful because it demonstrates how emissions have been changing over a period of time. Such emission trends demonstrate the effectiveness of national or local policies in curbing emissions. However, emissions calculated over more than one year can only be interpreted if the uncertainties in the estimates for each year can be understood. For example, emissions calculated for two different years may be different because emissions really have changed; or because new sources were found in the inventory for the second year which were missing from the inventory for the first; or because different methodologies were used in calculating them. An uncertainty analysis would help to demonstrate whether any changes in emissions between years were significant.

204. In the NAEI, a complete time series of UK emissions from 1970 to the most recent year that data are available are calculated each year. In other words, emissions from earlier years are recalculated each year. This may lead to changes in historic emission estimates due to new emission factors becoming available, new emission calculation methodologies being used or changes to national activity statistics (for example, fuel consumption) in earlier years being made by government or industry sources. However, this does mean that emissions calculated for a stream of different calendar years in a particular inventory compilation are comparable and the emission trends probably robust. In the NAEI, it is possible that relative changes in emissions each year shown in a particular year's inventory are more accurate than the absolute emission rates in tonnes.

205. Local inventories are generally more difficult to calculate than the national figures because they have to rely on different sources of information which are generally more uncertain, incomplete or difficult to obtain. They are not generally calculated every year and emissions in an area in a previous year are not usually recalculated when an inventory for a new year is compiled. Because of this, inventories compiled at different times for a particular area covering different calendar years may not be comparable. New sources may be discovered and methodologies improved for the more recent inventory. An uncertainty analysis for each inventory would help to put this into context.

206. The uncertainties in emission estimates need to be quantified at different geographical levels and different levels of sector detail. The results are required for different purposes. Uncertainty estimates for the national totals are required for interpreting the effect of national policies on emission trends and for appreciating the emission levels in the context of national emission targets and emissions ceilings (for example, the NECD targets).

207. Uncertainties in emissions from individual sectors may be quite different to the overall uncertainties in the national totals, but may still be required when considering the direction of government policies towards specific sectors, for example, power station emissions, road transport emissions and emissions from specific vehicle types.

208. Uncertainties in emissions in local areas are of more consequence to air quality modelling of local pollutant concentrations than uncertainties in the national totals and they will usually be of greater magnitude. Uncertainties in local inventories are affected by different factors to those affecting uncertainties in the national inventory estimates. For example, the national emission estimates are often based on national statistics like fuel consumption which are generally quite reliable. Local inventories require more local knowledge on activity drivers, which, while more detailed, may be considerably more uncertain than national figures. The inventories are also affected by knowledge of emission location and GIS mapping techniques.

209. This section considers levels of uncertainties in the national totals, in individual sectors and in local inventories and issues relating to methods for estimating them.

2.10.1 Uncertainties in national emission estimates

210. Quantifying the uncertainties in the national NO_x emission estimates helps to prioritise future work by pointing to those areas where further information is required to improve the reliability of the inventory. The National Atmospheric Emissions Inventory has made a detailed and quantified assessment of the uncertainties in the national emission estimates for NO_x and other pollutants in 2000 using a direct simulation approach (Passant, 2003).

211. The procedure used corresponds to the IPCC Tier 2 approach discussed in the Good Practice Guidance (IPCC, 2000), as well as the Tier 2 method proposed in the draft 'Good Practice Guidance for CLRTAP Emission Inventories', produced for inclusion in the EMEP/CORINAIR Guidebook on Emission Inventories. The same approach has also been applied to the UK greenhouse gas inventory (Salway *et al*., 2001) and has been described in detail by Charles *et al*. (1998).

212. In the approach used, an uncertainty distribution was allocated to each emission factor and each activity rate. The distribution types used were drawn from a limited set of either uniform, normal, triangular, beta, or log-normal. The parameters such as mean value, standard deviation etc. of the distributions for each emission factor or activity rate were set either by analysing the available data on emission factors and activity data or by expert judgement. A calculation was set up to estimate the emission of each pollutant by sampling individual data values from each of the emission factor and activity rate distributions on the basis of probability density and evaluating the resulting emission. Using a Monte Carlo type software tool, this process could be repeated many times in order to build up an output distribution of emission estimates both for individual sources but also for total UK emissions of each pollutant. The mean value for each emission estimate and the national total was recorded, as well as the standard deviation and the 95% confidence limits.

213. Uncertainties in the emission estimates arise from uncertainty in both the emission factor and activity rate used in the calculations. In some cases, emission estimates are supplied directly by process operators or regulators (for example, the Pollution Inventory) and are converted into an overall emission factor using national activity data. In this case, the uncertainty in the activity data is ignored and an uncertainty distribution is selected for the emission factor reflecting the uncertainty in the supplied emission estimate. Where this is not known, there is a need to improve the understanding of uncertainties in emissions data provided directly by industry or regulators.

214. An important assumption made in this statistical approach for estimating uncertainties in national emissions is that uncertainties in each source are not correlated. If there are cross-correlations in emission estimates for different sources, the analysis becomes much more complex.

215. Fuel consumption is one of the main activity datasets used. The uncertainties and distribution parameters in these were taken from Salway *et al*. (2001) and were based on data from DTI. However, the main sources of uncertainty in emission estimates are in the emission factors used rather than the fuel consumption data. Uncertainty estimates for these are largely based on expert judgement, guided by the range in emission factors given by literature sources and elsewhere. Industrial trade associations and industrial process operators provided a final source of expert opinion. The report by Barlow *et al*. (2001) provided an indication of the range of measured emission factors for road vehicles.

216. Uncertainties in UK road transport emissions were based on estimated levels of uncertainty in the emission factor for each category of vehicle and in the vehicle kilometre activity data used. For NO_x, the emission factor uncertainties used ranged from 25% for non-catalyst petrol cars to 60% for heavy duty diesel vehicles, all at the 95% confidence level. These largely reflect the scatter in the emission factor results at different vehicle speeds which were statistically interpreted in the report by Barlow *et al*. (2001). It is assumed that the vehicles tested are representative of those in the UK fleet. However, the changes in the latest set of vehicle emission factors, discussed in Section 2.6.2.1, from earlier figures that were used in the NAEI, are consistent with the emission factor uncertainty estimates used in this analysis. Uncertainties in the national vehicle kilometre figures were estimated to be 5–10% for each vehicle category. Road tunnel exhaust measurements can be used to validate vehicle exhaust emission models and provide further insight into levels of uncertainties in road transport emission estimates (Sjodin and Cooper, 2001).

217. As a consequence of the way uncertainties in emissions from different sources overlap and cancel out in this type of statistical procedure, the uncertainty in total UK emissions are smaller than might be expected from the levels of uncertainties in emission factors and activity data for individual sources. The analysis gave an overall uncertainty in UK emissions from road transport of 12% (95% confidence limit). For power stations, the overall uncertainty estimate in UK emissions was 10% and for domestic combustion 17%.

218. The overall uncertainty in the annual total for UK NO_x emissions (all sources) in 2000 was estimated to be 7% at the 95% confidence level for the mean value emission figure of 1512 ktonnes (using the old vehicle emission factors). The relatively low uncertainty is a result of statistical interpretation of the contributions from the main emission sources each with moderately high uncertainties and a degree of cancelling out.

219. The uncertainty estimate quoted here only refers to uncertainty in the national annual emission totals, not to emissions from particular sources. It also does not reflect uncertainties in emissions at a particular time or location. However, it is worth noting that IIASA concluded a very similar level of uncertainty (±10%) in their independently-derived estimate of UK emissions of NO_x using a similar statistical approach (Suutari *et al.*, 2001).

220. Uncertainties in UK emissions for all other air quality pollutants were estimated and found to be higher than the 7% figure for NO_x. The exception was for SO_2 which had an estimated level of uncertainty in UK emissions of ±3%.

221. A ‘key source’ analysis was also undertaken following IPCC Tier 2 methodology to identify the major contributors to inventory uncertainty (Passant, 2003). Key sources will often, but not always, be those sources which contribute most to national emissions. For NO_x, a number of key sources were identified. These were:

- Road vehicle emissions
- Domestic and industrial gas consumption
- Cement production
- Use of coal and landfill gas for electricity generation
- Off-road vehicles and machinery
- Gas oil consumption for coastal shipping
- Offshore oil and gas installations

222. The results of the uncertainty analysis apply to the estimates for the year 2000 only. An issue to consider is that since emissions are believed to be declining from many sectors as a results of legislation, it is vital that the emission factors and other data used in the NAEI are updated regularly to reflect any reductions in emissions that are occurring. This will require continuous and detailed research be carried out to provide evidence for any reductions, thereby allowing the emission factors used for the annual NAEI emission estimates to continue being up-to-date. A case in point is the road transport emission factors.

223. The statistical method used in the uncertainty analysis is useful for investigating the likely impact of uncertainties in emission estimates. However, it is not helpful for identifying missing sources in the inventory and the impact this has on uncertainties. A few sources of NO_x emissions not currently included in the NAEI have been identified in Passant (2003), but none of these are considered significant. These included emissions from natural and accidental fires; chemical waste incineration; and flares at refineries, chemical sites and landfills. Estimates for these sources will nevertheless be added to the NAEI over the next few years.

224. The uncertainties in inventories are a reflection of the quality and quantity of data which are obtained when compiling the inventory. The current approach for analysing uncertainty in the inventory allows the most critical parameters to be identified, thus priority areas for research can be set and the national inventory improved in the most cost-effective way.

2.10.1.1 Uncertainty in local emission estimates and emission mapping

225. Although there has been some progress in making a quantitative assessment of uncertainty in national emission estimate totals and for individual source categories, the production of statistical error estimates on spatially resolved inventories is still problematic. The most robust uncertainty estimates would account for uncertainty in activity data, emission factors, the assumptions required to apply emission factors and the spatial data used to represent sources. There are several aspects of spatial data quality to be taken into account such as positional accuracy, logical consistency, completeness and temporal accuracy. As with any other source of data, the spatial data that are used in an inventory project will influence the results obtained, for example, through varying road lengths or influencing the accuracy with which sources are located. This is particularly important where data are to form an input into dispersion modelling activities.

226. In addition to classical statistical uncertainty tests, there are various verification techniques that can be undertaken to help assess the reliability of inventory data. These include clear and transparent documentation of data and procedures; basic quality checks through cross reference with other sources; comparison with alternative inventories and ground truth verification. A comparison of two spatially resolved inventories for the Greater Manchester and Warrington areas, one produced by the London Research Centre (Buckingham *et al*., 1997a) and the other as a research project, indicated broad agreement in overall totals. Despite some methodological and source data differences, estimates were within ~12% for road transport sources (47.5 ktonnes (LRC) compared to 42.2 ktonnes (Lindley *et al*., 1996)) and ~21% for all sources (75.4 ktonnes (LRC) compared to 62.3 ktonnes (Lindley *et al*., 1996)). The nominal base years were 1995 (LRC) and 1994 (Lindley *et al*., 1996) which will have accounted for some of the differences in emission totals and their spatial distribution and it should be noted that the LRC inventory covered a wider range of sources in total. Figure 2.12 illustrates the differences in the spatial distributions of emissions which were found to be due to differences in spatial data sources, completeness and consistency (Lindley *et al.*, 2000).

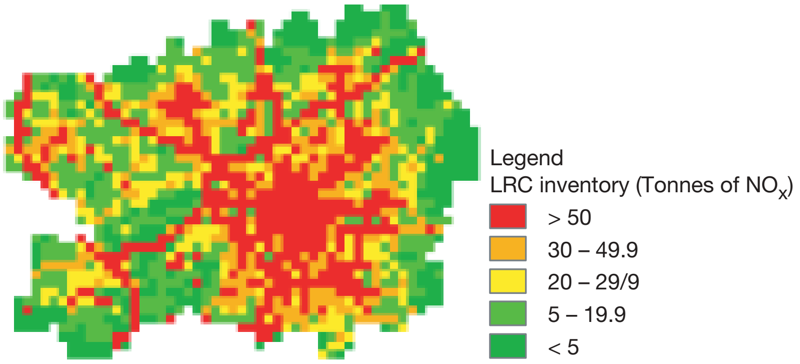
227. The comparison between the emission estimates for London from the LAEI and the NAEI maps also give a measure of uncertainties in an inventory for a local area based on the inventory technique used: ‘top-down’ or ‘bottom-up’. Overall NO_x emission estimates for London in the LAEI are 18% lower than the ‘top-down’ estimates of the NAEI for the same inventory year (Section 2.8.2.4).

228. One can also consider uncertainty in NO_x emissions on individual road links, again using a Monte Carlo analysis, where the range of speeds is taken into account. Consider the uncertainty in emissions for a typical traffic flow of 1000 vehicles per hour along a 1 km length of road, with a heavy duty vehicle mix of 10%. To illustrate this and assuming uncertainties in vehicle flow, speed and emission factors of ±10%, ±6 kph and ±15-25% (light and heavy-duty vehicles), respectively, the road link emissions have a calculated uncertainty of ±16% for an average speed of 30 kph. For an average speed of 15 kph, the road link emissions have a calculated uncertainty of ±28%. The relative uncertainty in emissions is considerably greater for lower vehicle speeds, as expected from the shape of the speed-emission curves which show emissions rising steeply at low speeds. This highlights the importance of good vehicle speed estimates.

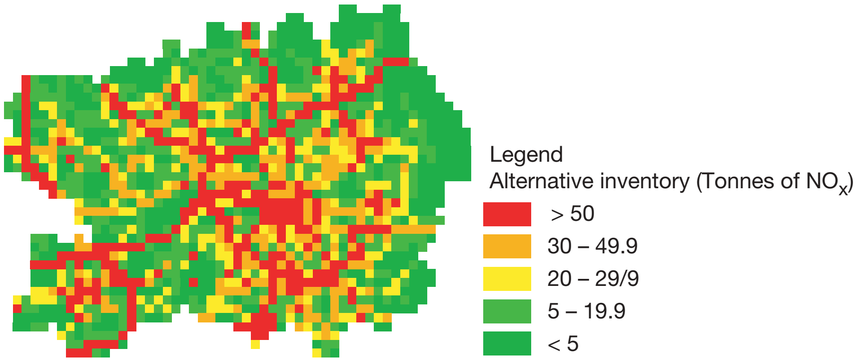
229. This simple uncertainty analysis illustrates that the uncertainty in individual road link emissions is significant when the potential range of vehicle speeds and driving patterns are considered.

Figure 2.12 Comparison of the results from two spatially resolved emissions inventories for Greater Manchester and Warrington (a) London Research Centre (Buckingham *et al.*, 1997a) (b) North West emissions inventory (Lindley *et al.*, 2000).

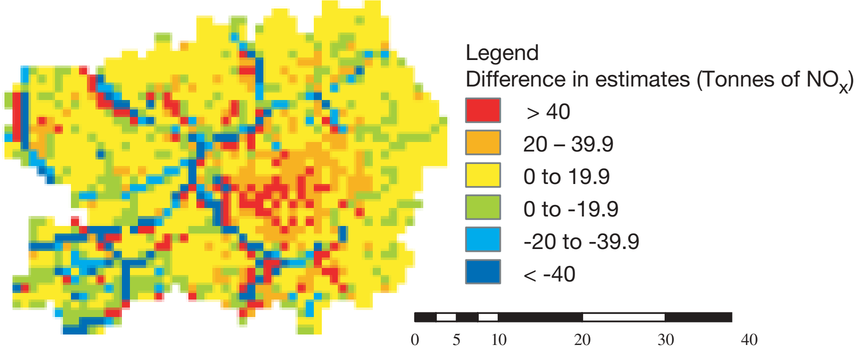
(a) Local emissions inventory produced by the London Research Centre



(b) Extract from an alternative local inventory



(c) Difference in estimates at the 1 x 1 km grid square resolution



2.11 NO_x emission projections

2.11.1 UK emission projections

230. The NAEI provides projections in UK emissions of all the major air quality pollutants by source sector. The projections are based on a number of different activity drivers, the most common one being fuel consumption forecasts for many of the sectors. Account is also taken of emissions legislation, fuel quality directives, abatement technology penetration and other factors which are likely to affect unit emissions. For road transport, a fairly detailed emission forecasting approach is used, adopting the latest traffic forecasts reflecting current Government policies on transport, fleet turnover and the penetration of vehicles meeting the tougher European vehicle emission Directives and the impact these will have on emissions from in-service vehicles.

231. The national projections are used by the Government to check against progress in achieving agreed emission reductions targets (for example, the National Emissions Ceiling Directive) and also in development of national strategies and policies. The emission projections are used for local air quality Review and Assessments.

2.11.1.1 Assumptions for non-traffic sources

232. For most of the sectors other than road transport, the main activity driver is forecasts in fuel consumption according to the central growth/high fuel price scenario in Energy Paper 68 (EP68), provided by the DTI. These estimates incorporate an assumed growth in economic activity of about 2.5% per year and the continuation of current trends towards greater use of natural gas and cleaner technologies (DTI, 2000). Industrial production activity projections from EP68 are also taken into account.

233. The projections take account of the impact, on emissions, of abatement in the electricity supply industry from EP68, the impact of the Large Combustion Plant Directive and improvements in efficiency in the cement industry and refineries.

2.11.1.2 Assumptions for road transport

234. The base emission projections for road transport are currently from a 2000 base year and are calculated from a combination of road traffic activity projections and knowledge of the expected emission characteristics of the vehicle fleet in the future.

235. The changes in emission factors for vehicles in the projections are largely driven by the legislative emission standards set in the European Directives for Euro III and Euro IV vehicles applying to conventional petrol and diesel powered vehicles. This means that no further improvements in new vehicle emissions occur beyond the limits set for introduction in 2006 for light duty vehicles and 2008 for heavy duty vehicles. In other words, the penetration of advanced vehicle types and technologies currently under development, such as fuel cell, hybrids and other electric powered vehicles are not considered in the base projections, neither are alternative fuels such as CNG.

236. Table 2.2 provided the vehicle emission factors for Euro III and Euro IV vehicles, based on scaling factors assumed to apply across all speeds relative to emission factors for Euro II vehicles. The basis for these factors was discussed in Section 2.6.2.1. In addition to this, the impact of current vehicles in the fleet (pre-2000 standards up to Euro II) running on ultra-low sulphur fuels meeting the 2005 Fuel Quality standards was accounted for. This grade of fuel is already widely available in the UK, but the effect on NO_x emissions (from 500 ppm to 50 ppm sulphur (ULS) grade) is quite small: reductions are typically less than 15%. The effects of fuel quality on emissions were based on empirical relationships taken from the European EPEFE Programme (EPEFE, 1995). It is assumed that 100% market penetration of 2005 standard fuels (ULSP and ULSD) is achieved by 2001.

237. The turnover rate for vehicles in the national fleet combined with new vehicle sales projections define the penetration rate for new vehicles meeting the tighter emission standards and the phasing out of older, higher emitting vehicles in the fleet (Goodwin *et al*., 2001). The fleet projections are calculated from survival rate functions implied by historic vehicle licensing statistics (number of new registrations and numbers still registered by age in each year). Growth in new car sales was taken from DfT's Vehicle Market Model. For other vehicle types, new sales and growth in the vehicle stock were related to the growth in number of vehicle kilometres. A number of assumptions relating to the composition of the national vehicle fleet were specified by the DfT in 2001 and are shown in Table 2.8. Note that the number of heavy duty vehicles fitted with particulate traps shown in the table is being reviewed in light of data from the Energy Saving Trust, on the number of grants provided under the Government's CleanUp programme, which suggest the actual figures may be higher than shown.

Table 2.8 Assumptions affecting the composition of the UK fleet provided for the NAEI base projections by DfT in 2001.

Year	2000	2001	2002	2003	2004	2005	2006	2007	2008+
Diesel penetration of new car market (%)	14	16	17	18	19	20	21	22	22
Proportion of Euro IV cars in the new petrol car market (%)	0	20	40	60	80	100	100	100	100
Proportion of new diesel cars with particulate traps (%)	0	5	10	15	20	25	25	25	25
Cumulative number of heavy duty vehicles retrofitted with particulate traps	4000	6000	8000	10000	12000	14000	14000	14000	14000

238. The current NAEI projections are from a 2000 base year which means that 2001 is treated as a 'projection year'. One of the key assumptions in the base projections is the rate of penetration of diesel cars in the fleet. This is driven by the percentage sales of new diesel cars. The 2000 NAEI base projections assumed only a modest growth in diesel car sales, as defined by DfT in 2001 and shown in Table 2.8. However, an accelerated growth in diesel car sales appeared to take place in 2001 so, whereas the predicted rate used in the projections was 16%, the actual rate was 18% for the year as a whole, but rising upwards during the course of 2001. This led DfT to re-forecast the rate of diesel car sales and the effect this has on NO_x emission projections for road transport will be discussed in Section 2.11.3.

239. DfT assumptions on the early introduction of Euro IV petrol cars in the UK market are shown in Table 2.8 expressed as the percentage of new petrol cars sold which meet these standards. Table 2.8 also shows the proportion of new diesel cars sold which are fitted with particulate traps. However, the fitting of a particulate trap on a diesel car does not have a significant effect on total NO_x emissions. A small number of heavy duty vehicles are also assumed to be retrofitted with particulate traps from 2000, the numbers growing to 2005. There is some evidence that CRT traps may lead to a small decrease in NO_x emissions from heavy duty vehicles.

240. In addition to these, the following assumptions are made concerning the introduction of new vehicle emission standards:

Petrol cars:
Euro III (98/69/EC) – 10% in 2000 (balance are Euro II's)
Euro IV (98/69/EC) – as in Table 2.8; balance from 2001 are Euro III

Diesel cars:
Euro III (98/69/EC) – 10% in 2000 (balance are Euro II's). 100% from 1 January 2001 to 2005
Euro IV (98/69/EC) – 100% from 1 January 2006

LGVs (petrol and diesel):
Euro III (98/69/EC) – 100% from 1 January 2002 (2001 for small LGVs)
Euro IV (98/69/EC) – 100% from 1 January 2006

HGVs and buses:
EURO III (1999/96/EC) – 100% FROM 1 OCTOBER 2001
Euro IV (1999/96/EC) – 100% from 1 October 2006 (standards for NO_x introduced in 2 stages, second stage from 1 October 2008)

Motorcycles:
97/24/EC – 100% from 1 January 2000

All these assumptions are factored into the fleet composition data.

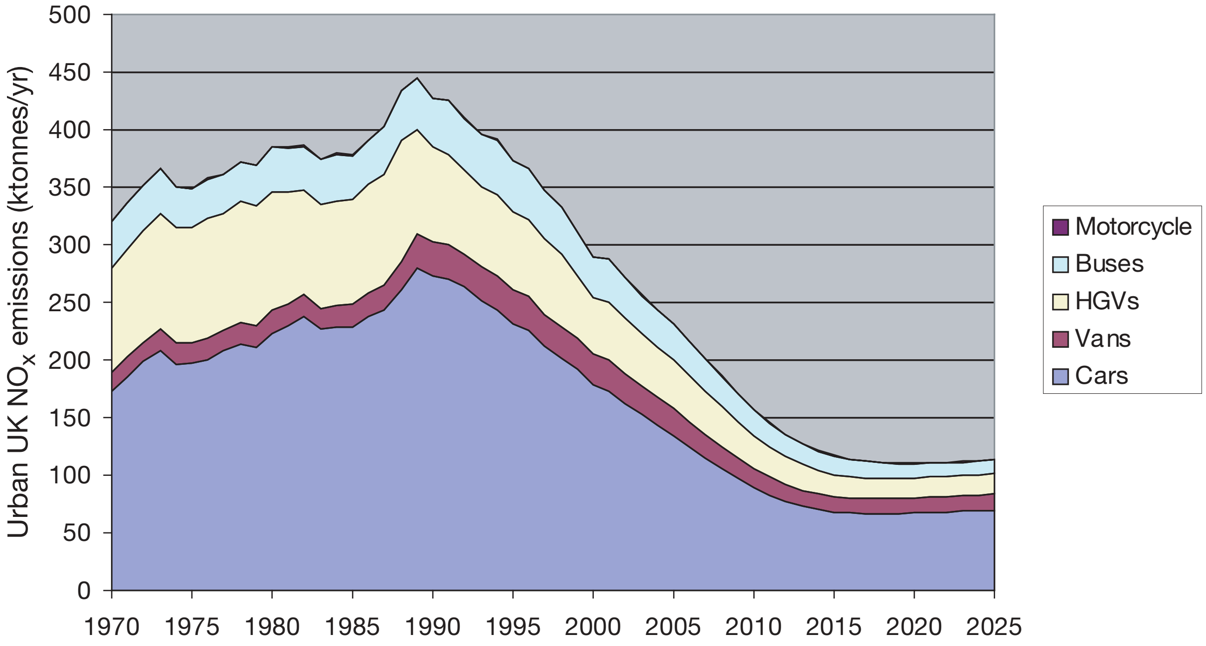
241. The other key driver in the road transport emission projections is the growth in traffic activity. The growth in UK vehicle kilometres is forecast from 2000 for each vehicle, road and urban area type, using separate forecasts for traffic in England, Scotland, Wales and N Ireland. A combination of data sources are used, including the Ten Year Plan for Transport 'Plan' scenario (DETR, 2000b), the 1997 version of the National Road Traffic Forecasts from DfT and the Central Scotland Transport Model (CSTM3) from the Scottish Executive.

2.11.1.3 Baseline NO_x emission projections for road transport

242. Tables showing UK and urban UK road transport emissions from 1990 to 2025 by vehicle type are provided in the Technical Annexe to this report (Appendix 1). All the figures are based on the new vehicle emission factors (not originally used for the 2000 NAEI base inventory, see section 2.7.1) and are projections from the year 2000.

243. Figure 2.13 demonstrates the trends in urban UK road transport emissions by vehicle type from 1970 to 2025. NO_x emissions from road transport are expected to continue to decline from 2000 until around 2015, due to the penetration of cleaner vehicles of all types in the fleet. After 2015, without any further improvements in vehicle emissions beyond those required by the Euro IV standards, emissions start to rise again as the continued growth in traffic starts to offset the gains achieved by penetration of cleaner vehicles in the fleet. Passenger cars are the dominant source of road transport emissions in 2000 contributing to 49% of road transport emissions nationally and 62% in urban areas. This dominance continues although it is slightly reduced to 44% of road transport emissions nationally and 57% in urban areas by 2010.

Figure 2.13 Urban UK road transport emissions of NO_x.



2.11.1.4 NO_x emission projections for all sources

244. The projections of UK NO_x emissions from all sources were displayed in Figure 2.1 which showed the complete time-series in emissions by sector from 1970 to 2020. These are based on the latest vehicle emission factors.

245. Emissions are predicted to fall by 25% from 2000 to 2010 and by 34% from 2000 to 2020. The fall is mainly due to the decline in road transport emissions and emissions from public power generation. From 2000 to 2010, road transport emissions fall by 50%, while emissions from public power generation fall by 18%. The contribution from road transport to total UK NO_x emissions also falls from 49% in 2000 to 32% in 2010.

246. In spite of this encouraging decline in emissions, without any further measures put in place the fall does not appear to be enough to reach the NECD emission ceiling of 1167 ktonnes which the UK must achieve by 2010. Based on the NAEI's current projections from 2000, emissions will be 1293 ktonnes in 2010, still 126 ktonnes short of the target. A further 10% reduction in emissions in 2010 will be required to meet this target. This requires the UK to find a cost-effective means of further reducing emissions to achieve the NECD ceiling. However, it also highlights the need to fully understand the uncertainty in the national emission projections and their sensitivity to key parameters such as the emission factors for current and future vehicles, forecasts in transport and industrial activity, effectiveness of abatement technologies and demand for energy and likely fuel mix for future electricity generation. The Government has recently proposed a National Emission Reduction Plan for implementation of the Large Combustion Plant Directive which is expected to deliver a greater degree of certainty in the outcome of the Directive by offering operators greater flexibility in achieving reductions cost effectively.

2.11.2 NO_x emission projections in specific urban conurbations

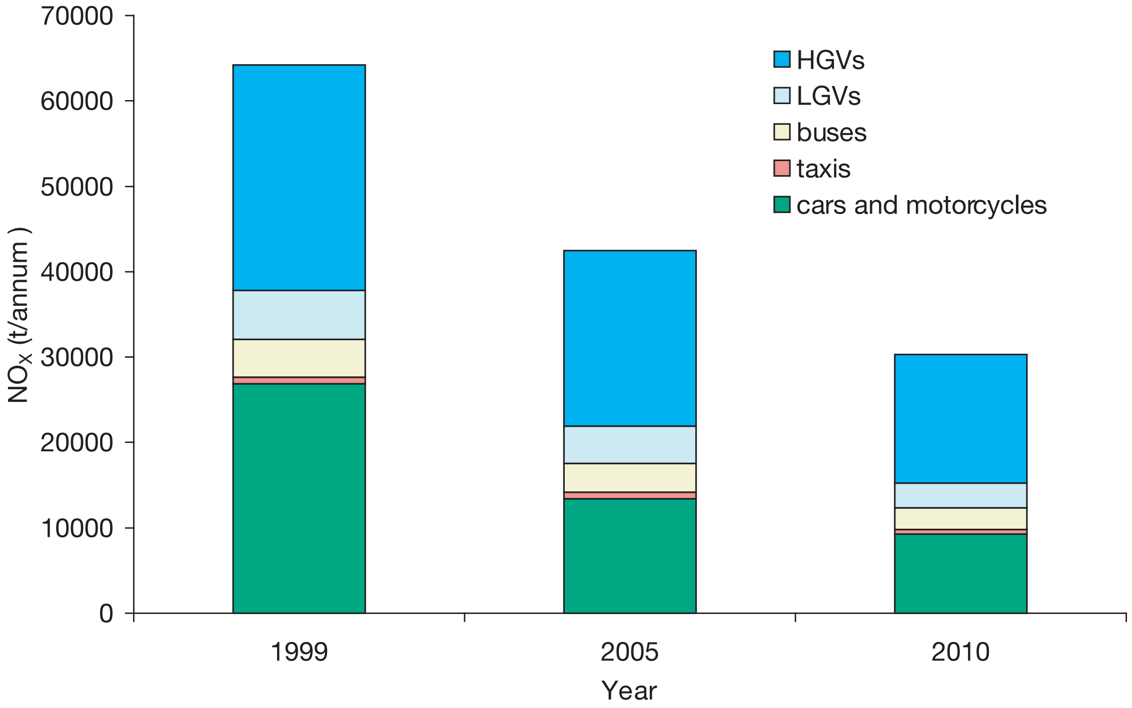
2.11.2.1 London

246. The LAEI has been developed to provide projected estimates of NO_x emissions in London.

247. A simple approach has been taken to the change in vehicle flows between 1999 and 2004/5, based on information from the GLA. The following traffic growth figures have been assumed: central London 0% per annum, inner London 0.2% per annum and outer London 1% per annum. These growth factors are assumed to apply equally to all vehicle types. Recent information from the GLA suggests that all TfL buses will be fitted with particulate traps by 2005. The NAEI emission factors are used for forecasting vehicle emissions in the LAEI projections.

248. Projections of road transport emissions of NO_x in London are shown in Figure 2.14. Between 1999 and 2010 total NO_x emissions from road transport are projected to decrease by 53%. This compares with a decrease of 49% in total urban UK road transport emissions over the same period according to the NAEI. The most significant decline is for passenger cars (65% reduction). HGV emissions only decline by 43%.

Figure 2.14 Projected emissions of NO_x from road transport in London (LAEI).

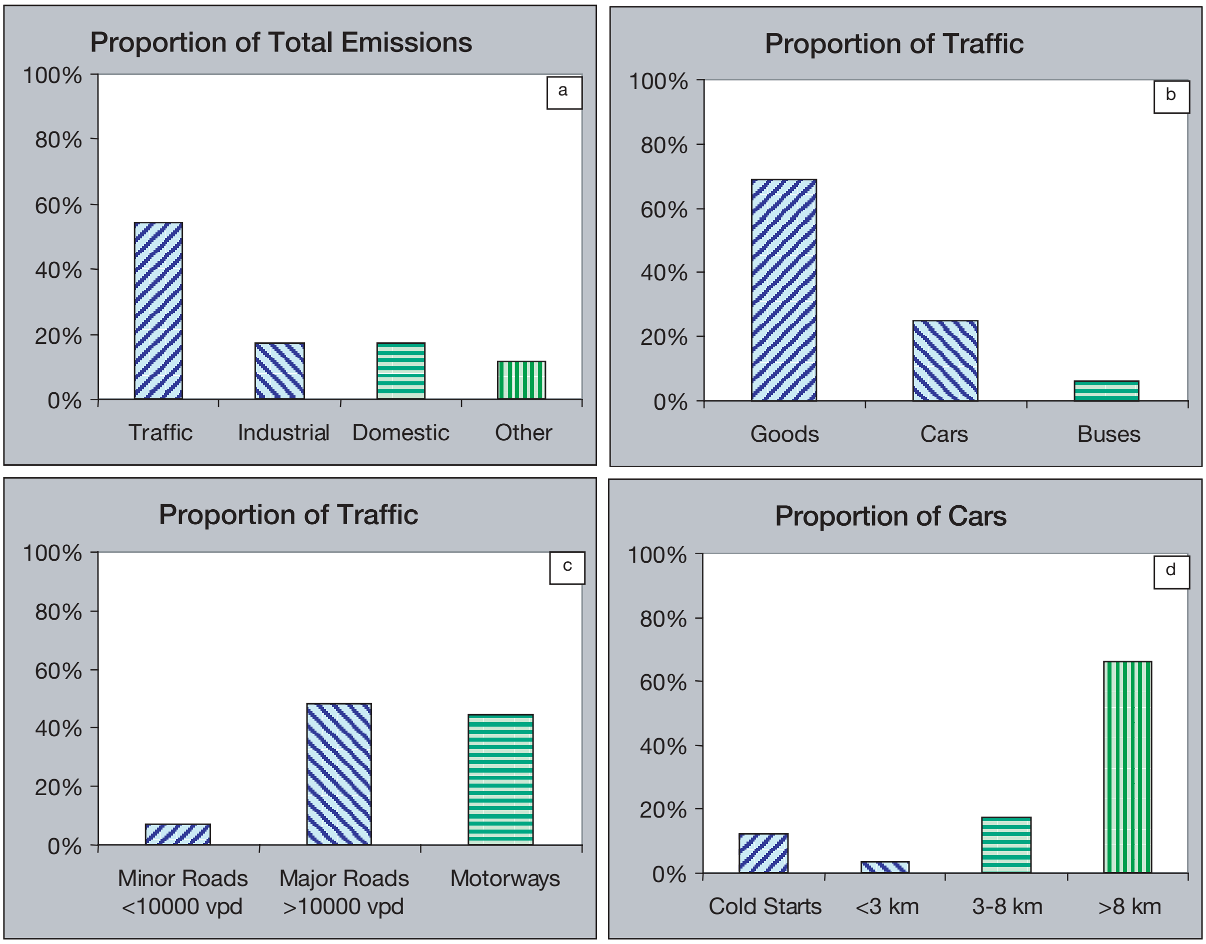


249. The change in non-road transport sources of NO_x is less certain. NO_x emissions from these sources are projected to increase slightly in London between 1999 and 2010, mostly as a result of a significant increase in natural gas use for domestic premises and commercial buildings. However, no account has been taken of the potential decrease in NO_x emissions through improvements in boiler technology. At present, emission factors for these sources are based on a single fuel-based factor and it is recognised that refinements are needed for these emission estimates. In the LAEI projections, emissions at Heathrow are expected to decline significantly for non-aircraft emissions, whereas aircraft emissions are assumed to remain at 1999 levels. The updated Heathrow emissions inventory currently being compiled by AEA Technology should lead to improved estimates of NO_x emissions from Heathrow.

2.11.2.2 Greater Manchester

250. An inventory recently completed by Aric for the Greater Manchester Area shows that traffic emissions will continue to dominate in 2005 (Figure 2.15a), with most of these emissions being due to goods vehicles rather than cars (Figure 2.15b). Traffic on motorways will account for over 40% of the emissions, with minor roads playing a small role (Figure 2.15c). Interestingly, the car emissions are dominated by vehicles on longer journeys >8 km, with short journeys playing an insignificant role (Figure 2.15d). These car journey emissions do not include cold start emissions, which would apply to all categories to some extent. However, the pattern would not be changed significantly, as cold start emissions only represent around 10% of car emissions (Figure 2.15d).

Figure 2.15 Estimated emissions of NO_x from the Greater Manchester area in 2005; a) Total emissions by source, b) traffic emissions by vehicle type, c) traffic emissions on different road types, d) car emissions by journey length and cold start emissions.



2.11.3 Road transport emissions: evaluation of current policies and directives and illustrative projection scenarios

251. The reduction in emissions from road transport that has occurred over the past decade and is expected to continue over the next decade is as a net result of technology improvements required to meet the European vehicle emissions Directives. Domestic transport policies will also have had a bearing on recent and future emission trends. The effect of individual policies and Directives has been evaluated by modelling UK road transport emissions in past and future years with the policy or measure excluded. The effect of the measure can be found by difference with the base projection figures. This procedure can be used, for example, to demonstrate the effect that the introduction of the three-way catalyst to new petrol cars and vans has made to the emission reductions achieved over the past 10 years. Emissions can also be further broken down by Euro standard to illustrate the contribution that vehicles meeting recent and future standards make to the inventory and how much remains from older vehicles in the fleet.

252. Studies of this nature have been carried out previously by netcen on behalf of Defra, as a policy evaluation exercise on measures to reduce air pollution from road traffic (Bush *et al.*, 2001a). Results are shown in this section from a revised study on a selected number of measures based on the new emission factors.

253. Projected emissions from road transport can be sensitive to the assumptions made about the penetration of diesel cars in the national fleet. Since the 2000 NAEI projections were compiled, DfT have revised their opinion on the likely growth in diesel car sales in response to a surge in sales which took place in 2001. The effect this has on the UK projections of NO_x emissions from road transport is shown. In effect, this also amounts to a form of sensitivity analysis, in this case showing the sensitivity of NO_x emissions to diesel car penetration rates.

254. The Air Quality Strategy consultation document published by Defra and the devolved administrations in 2001 showed the impact of a number of additional national policy measures on UK PM_{10} emission projections for road transport (AQS, 2001). These included the fitting of particulate traps to all new diesel vehicles, the early introduction of sulphur-free diesel fuel and a short-term retrofitting programme for older diesel vehicles, targeted at particular conurbations. These are not discussed further in this report, but illustrate the types of national transport measures that can be assessed.

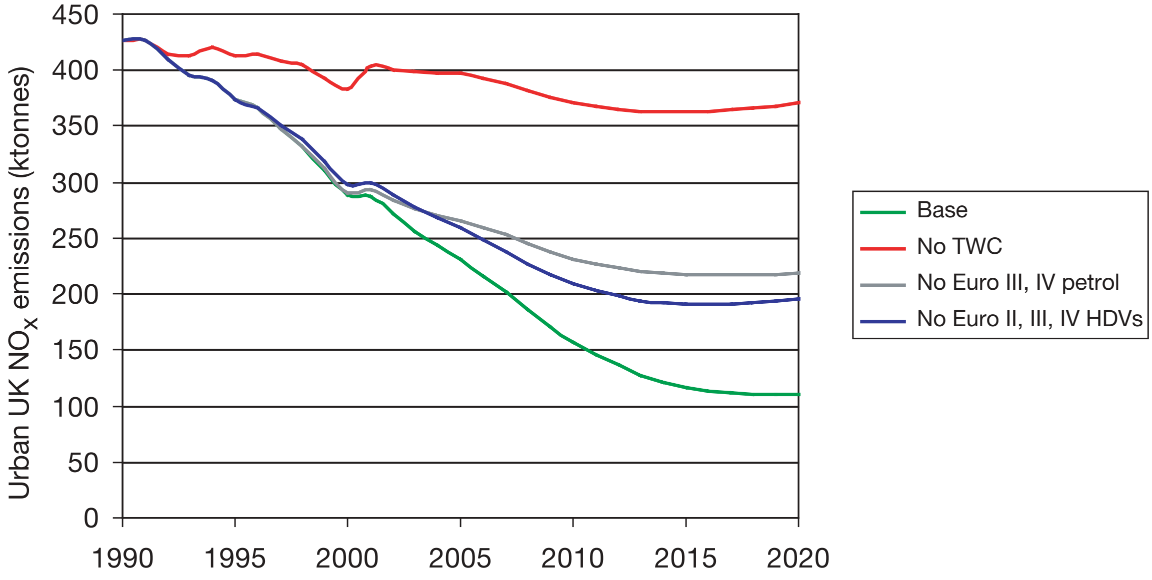
2.11.3.1 The impact of the introduction of the three-way catalyst on NO_x emissions from road transport

255. Emissions from road transport were calculated assuming Euro I standards (91/441/EEC) and beyond had not been introduced requiring petrol cars and light vans to be fitted with three-way catalysts. This meant that emission factors for these vehicles were retained at pre-Euro I levels. The effect this would have had on urban UK NO_x emissions from road transport is shown in Figure 2.16. All other assumptions remained as for the base case, including the implementation of standards for diesel vehicles (light and heavy duty vehicles). The difference in the results for this scenario and the base case (i.e. the effect of introducing three-way catalysts) are reflected by the difference in emissions.

256. Urban UK road transport emissions were reduced by 94 ktonnes (25%) from what they would have been in 2000 had three-way catalysts not been introduced. Without their introduction, NO_x emissions would have decreased at a much slower rate. The base inventory suggest urban UK NO_x emissions decreased by 32% from 1990 to 2000, but without the introduction of catalysts on petrol cars and vans, this would only have led to a 10% reduction.

257. Figure 2.16 shows the projected effects of having introduced the three-way catalyst and tighter Euro standards on petrol cars and vans. Emissions would continue to decrease at a much slower rate without catalyst introduction. By 2010, urban UK road transport emissions would have been only 13% less than 1990 levels, whereas a 63% reduction is anticipated with catalyst introduction and the progressive tightening up of emission standards down to Euro IV levels.

Figure 2.16 Urban UK road transport emissions of NO_x for different Euro standard and technology uptake scenarios.



2.11.3.2 The impact of the introduction of Euro III and IV standards for petrol cars on NO_x emissions from road transport

258. Emissions from petrol cars and light vans have been further tightened up with the introduction of Euro III and IV standards. The Directive 98/69/EC requires all new cars and light vans to meet Euro III standards by January 2001 (2002 for heavier vans) and Euro IV standards by January 2006. Some models already met Euro III standards before 2000 and some car models now on the market already meet Euro IV standards.

259. Emissions were calculated assuming Euro III and IV standards had not been introduced. This meant that emission factors for these vehicles were retained at Euro II levels from 1997 onwards. The effect this would have had on urban UK NO_x emissions from road transport is shown in Figure 2.16. All other assumptions remained as for the base case, including the implementation of all the standards for diesel vehicles (light and heavy duty vehicles). The difference in the results for this scenario and the base case (i.e. the effect of introducing Euro III and IV standards for petrol cars and vans) are reflected by the difference in emissions.

260. Emissions would continue to decrease, but at a slower rate without the introduction of the Euro III and IV standards for these petrol vehicles. By 2010, urban UK road transport emissions would be only 20% less than 2000 levels, even with all the standards for diesel vehicles up to Euro IV included. It is anticipated that a 46% reduction in urban NO_x emissions from road transport will actually occur with the introduction of standards for all vehicle types.

2.11.3.3 The impact of the introduction of Euro II – IV standards for heavy duty vehicles on NO_x emissions from road transport

261. Euro II standards for heavy duty diesel vehicles were introduced in 1996. Emissions were calculated assuming neither Euro II standards nor the successive Euro III and IV standards for Directive 99/96/EC (2001 and 2006) were introduced. This meant that emission factors for these vehicles were retained at Euro I levels from 1993 onwards. The effect this would have had on urban UK NO_x emissions from road transport is shown in Figure 2.16. All other assumptions remained as for the base case, including the implementation of all the standards for petrol vehicles. The difference in the results for this scenario and the base case (i.e. the effect of introducing Euro II-IV standards on HDVs) are reflected by the difference in emissions.

262. It can be seen that urban UK road transport emissions were reduced by 9 ktonnes (3%) from what they would have been in 2000 had the Euro II standards not been introduced. The effect of the Euro II standards on HDVs in reducing urban road transport NO_x emissions has been small during years up to 2000. However, the effect of these and tougher Euro III and IV standards becomes much more significant in future years. Without these standards, NO_x emissions would continue to decrease, but at a slower rate. By 2010, urban UK road transport emissions would be 30% less than 2000 levels, with all the standards for petrol vehicles up to Euro IV included. This compares with the 46% reduction that is actually expected.

2.11.3.4 The impact of higher penetration rates for diesel cars in the new car fleet on NO_x and NO_2 emissions from road transport

263. DfT have recently revised their opinion on the likely growth in diesel car sales in the UK in response to a surge in sales which took place in 2001. Sales were originally forecast to be 16% of all new car sales in 2001, but the actual figure was 18% for the year as a whole, growing to a level of around 23% in the early part of 2002.

264. The original growth estimates were designed to reflect the way car manufacturers would meet the voluntary agreement on CO_2 new car emissions by increasing diesel car sales. The recent rapid increase in diesel car sales is thought to be due to new company car taxation rules, but the view of DfT is that this trend may continue. The view now is that diesel car sales will increase to 25% in 2003 and increase linearly to 30% by 2010. As diesel cars emit more NO_x than petrol cars, this will lead to an increase in the overall emission projection estimates for road transport.

265. The effect of the higher diesel car penetration rate on NO_x emissions from traffic was modelled leaving all other assumptions the same. Emission projections were calculated based on the high diesel car growth assumption, reaching 30% of sales by 2010, and compared with the original baseline projections assuming a growth to 22% in sales by 2007, remaining constant thereafter (Table 2.8).

266. The amount of increase in urban UK road transport emissions of NO_x was found to be small. Urban UK road transport emissions of NO_x are increased by 0.7% in 2005 and 0.7% in 2010 relative to the baseline projections. The increase in total UK emissions of NO_x in 2010 is 3.5 ktonnes, or 0.3% relative to the baseline emission projections for all sources.

267. The effect on primary NO_2 emissions in urban areas would be more significant, however, if diesel cars emit a higher proportion of NO_x as NO_2, as suggested in Section 2.6.2.6. On the basis that diesel cars emit 25% NO_x as NO_2 (by mass) and with petrol cars emitting only 5% NO_x as NO_2, as concluded in Section 2.6.2.6, then increasing the diesel car penetration rate from 22% to 30% by 2010 would increase urban road transport emissions of primary NO_2 in 2010 by 3%.

268. Sales of diesel cars are currently on average around 35% of all new car sales in other European countries. The European car industry group ACEA predicts this will rise to 40% of new car sales to meet the voluntary agreement on CO_2 emissions. DfT consider that with current fuel duty policy, diesel car sales are unlikely to reach this level in the UK. However, in an extreme case where sales of new diesel cars rise to 50% of new car sales by 2010, then urban UK road transport emissions of NO_x in 2010 would increase by 1.2% and primary NO_2 emissions by 6% compared with the low (22%) penetration rate assumption.

Chapter 3

Meteorological and chemical processes

Key points

- The concentration of NO_2 at a given location is determined by a combination of emissions, meteorology and chemistry.
- NO_2 may be dominated by primary sources (i.e. direct emission), secondary sources (i.e. chemical formation), or may have significant contributions from both. It is a vital issue for policy whether the origin of the NO_2 at a given location is primary or secondary, because source-receptor relationships are dramatically different for primary and secondary pollutants.
- Meteorological processes operating on scales from a few metres to hundreds of kilometres play an important role in controlling dispersion and accumulation of NO_2.
- Very low wind speeds, temperature inversions and a shallow stable boundary layer are necessary conditions for winter NO_2 episodes.
- Limited meteorological observations in complex environments, such as in urban areas, leads to questions over sampling representativeness and model parameterisations which are not fully tested.
- Model simulations of NO_2 concentrations are sensitive to inter-annual variations in meteorological conditions.
- The chemical coupling of NO_x with ozone (O_3) plays a central role in determining the ambient level of secondary NO_2. Because of this chemical coupling, it is often convenient to define NO_2 and O_3 collectively as oxidant (OX).
- Other chemical processes may lead to additional NO to NO_2 conversion under specific conditions such as urban episodes during stagnation events.
- Chemical transformations of NO_x can generate a variety of inorganic and organic oxidised nitrogen compounds. Of these, HONO and PAN potentially contribute to measurements of NO_2 made using chemiluminescent analysers with thermal converters. The level of interference is likely to be small under the majority of ambient conditions.

3.1. Introduction

269. The concentration of NO_2 at a given location is determined by a number of factors. These include the magnitude and proximity of NO_x emissions sources, the rate at which these emissions are dispersed and the area over which they are transported (i.e. meteorology of winds, turbulent mixing and dilution), and the processes which determine what proportion of NO_x is in the form of NO_2 (that is, in addition to the proportion of NO_x directly emitted as NO_2, the chemistry leading to subsequent generation and destruction of NO_2). Accordingly, models which attempt to describe the concentration distribution of NO_2 in air must ideally contain an adequate representation of emissions, meteorology and chemistry.

270. A detailed description of the sources of NO_x and how the emissions density varies spatially and temporally is provided in Chapter 2. The purpose of this chapter, therefore, is to provide a summary of current understanding of the fundamental meteorological and chemical processes that play a role in controlling the ambient level of NO_2. To put these processes into context, the section concludes with a description of how emissions, dispersion and chemistry combine to determine the distribution of NO_2 in the vicinity of roads in rural and urban environments.

3.2. Meteorology and NO_2

3.2.1. Introduction

271. Variations in weather conditions on scales of metres to thousands of kilometres can all be shown to impact upon spatial and temporal variations in NO_2 concentration. For example:

- increases in hemispheric scale background O_3 concentration may, if transported down into the urban boundary and canopy layers, become available as additional oxidant in urban environments.
- the familiar weather patterns which appear on television and newspaper weather maps, with their associated wind fields and stability conditions, control the degree of accumulation of air pollutants and the advection of primary and secondary pollutants from upwind sources and provide a context for local meteorological variations.
- local wind circulations, such as land-sea breezes and urban-country breezes, help to maintain ventilation and minimise the accumulation of NO_2 and its precursors. Conversely urban areas may act to block and divert the larger scale flows, and within urban areas topographical features such as rivers and hills may channel and modify the local wind patterns.
- microscale local meteorological variations occur in heterogeneous environments such as towns and cities as a result of roughness variability, varying degree of shade, anthropogenic energy sources, and modified moisture budgets. Vehicle movements also initiate localised turbulence which can be important in the local dispersion process.

272. The importance of all these meteorological scales are highlighted in this chapter, with an emphasis on their relevance to air pollution modelling and to the meteorological conditions which have accompanied important observed NO_2 episodes in the UK over the last decade. The section ends with some commentary on relevant meteorological trends over recent decades together with projections over the next century which have been simulated using climate models.

3.2.2. The atmospheric boundary layer

273. Pollutants emitted into the atmosphere are generally transported, dispersed and deposited in the lowest part of the atmosphere – the atmospheric boundary layer. A description of the airflow and turbulent motions within this layer is therefore helpful in understanding the dispersion of pollutants and this forms the subject matter of this section. Factors affecting the boundary layer structure and common features of the boundary layer over flat terrain are briefly described. There then follows a section on dispersion over flat terrain before a discussion of meteorology and dispersion in more complex situations such as the urban environment.

3.2.2.1 Factors determining boundary layer structure

274. The mean flow, turbulence and the temperature distribution in the atmospheric boundary layer are broadly governed by three groups of factors which are as follows:

- Heating at the surface which generates convective turbulence. This is determined by such factors as the solar radiation reaching the surface and the absorption and release of latent heat by water vapour. Urban surfaces and buildings modify the absorption and reflection of solar radiation and sensible heat fluxes. They also enhance run-off following rain, and affect evaporation.
- The roughness and changes in elevation of the earth's surface near and for some distance upwind of the region of interest.
- The roughness effect is caused by surface obstacles or roughness elements obstructing and hence decelerating the airflow; these may range in scale from grass to buildings and even ranges of hills. A 'roughness length' can be used to characterise the effect of these elements on the flow above them with large elements generally causing more deceleration and greater turbulence levels and hence greater mixing of pollution. Within the obstacles, as occurs in an urban area, the flow is more complex as discussed in Section 3.2.3.
- The effects of changes in elevation on the airflow are characterised both by the heights and the length scales of the terrain features and may be very complex. Typically the flow accelerates over hill summits and decelerates up and downwind. As the hill slopes increase there can be regions of separation (downstream) and blocking (upstream) with the flow direction reversing, resulting in very complex dispersion of pollution. Local solar heating effects or cooling effects can result in anabatic (upslope) and katabatic (downslope) winds, or local circulations over water or park areas.
- Finally the airflow at the top boundary layer determined by the large scale pressure gradient broadly determines wind speeds. However in light or calm winds the airflow patterns are strongly affected by local heating effects for example from buildings and by local features such as slopes or valleys.

3.2.2.2 Typical boundary layer structure over flat terrain

275. Within the atmospheric boundary layer there are three broad types of structure corresponding to (i) unstable/convective boundary layers, (ii) neutral boundary layers and (iii) stable boundary layers. Typical conditions characterising these structures are for (i) light winds, and strong surface heating and consequently strong vertical mixing; for (ii) high winds and/or weak surface heating or cooling; and for (iii) night time conditions of light winds and negative surface heat flux (cooling) respectively.

276. In the neutral boundary layer the mean flow and turbulence are determined by the drag of the flow over the surface. The increase of mean flow U with height z can be described by a simple logarithmic formulation as

$$U(z)=\frac{u_*}{\kappa}\ln\left(\frac{z}{z_0}\right)$$

where u_* is the friction velocity related to the surface shear stress and turbulence levels and z_0 a representative surface roughness typically a few centimetres in rural areas and over 1 m in the most built up urban centres. (κ is von Karman's constant (0.4)).

277. In built up areas the flow above the buildings reacts as if the surface is significantly higher than z_0, typically over 50% of the building height. This effective height is often referred to as the zero plane displacement.

278. Both lengthscales and timescales of turbulence are important in determining the rate of spread or mixing of a dispersion plume and the chemical reaction rates. For instance chemical reaction rates are limited by turbulent mixing rates if the turbulent mixing timescale is greater than chemical reaction timescales (i.e. the reaction can proceed only after the reactive species have been mixed together). In a neutral boundary layer over flat terrain this timescale is approximately 0.3 z/u_*, ranging from a few to hundreds of seconds; in the urban canopy the timescale can be determined by building height (h_B) and is roughly h_B/u_* (tens of seconds).

279. In the convective boundary layer the surface heat flux $F_{\vartheta u}$ provides an additional input of turbulent energy and the vertical component of turbulence can increase with height which is very important in the dispersion of elevated sources of pollutant. A useful parameter is the Monin Obukhov length, L_{MO}, which is a length scale used to denote the relative importance on turbulence generation of surface drag and surface heating

$$L_{MO} = \frac{-u_*^3}{\kappa g F_{\theta 0} / (\rho c_\rho T_0)}$$

where g, h, ρ, c_p and T_0 are the acceleration due to gravity, boundary layer height, density of air, specific heat of air and temperature respectively.

280. In convective conditions the L_{MO} is negative (implying an unstable layer) and decreases with increasing heat flux being a few tens of metres for strongly convective conditions. For a given L_{MO}, scales larger than $|L_{MO}|$ are dominated by convectively produced turbulence.

281. In the stable boundary layer surface cooling stably stratifies the surface layer which in turn inhibits the production of turbulence. Turbulence levels, lengthscales and mixing are smaller than those occurring in the neutral boundary layer and consequently the boundary layer height h is smaller. In stable layers the Monin Obukhov length (L_{MO}) corresponds approximately to the largest turbulent length scales with scales decreasing as the cooling increases and hence L_{MO} decreases.

282. Over urban areas the general description of the idealised atmospheric boundary layer structure given above only applies to the flow above the urban canopy with the bulk impact of the urban canopy represented by a higher surface roughness (for example, z_0 = 1m) and with allowance made for anthropogenic inputs to the heat flux. Some discussion of effects within the urban canopy appears in Section 3.2.3.

3.2.2.3 Dispersion over flat terrain

283. In Figures 3.1.1 and 3.1.2 a dispersion model based on many of the principles described in this chapter, ADMS 3 (Carruthers *et al*., 1994), has been used to illustrate how in general terms the ground level concentration (glc) of a pollutant emitted from a point source varies with distance downwind from the source over flat terrain. Figure 3.1.1 is for a ground level source whilst Figure 3.1.2 is for an elevated source. The concentration is for an averaging time of one hour which is generally smaller than the timescale of typical changes in meteorology, but larger than the timescales of turbulence fluctuations in concentration. The weather conditions range from Case 1 (very unstable), through to Case 2 (neutral), through to Case 3 (very stable).

284. For the ground level source the maximum surface concentration occurs at the release point and then decays away from the source with the rate of decay being highest for the most unstable conditions, because of rapid turbulent mixing, and lowest for the most stable case, because of very weak turbulence. Thus ground level concentrations remain quite high well downwind of sources in stable conditions.

285. For the elevated source, Figure 3.2, the position of maximum surface concentration is downstream of the source since the pollutant has to be mixed down to the surface. The highest surface concentrations occur in the most unstable conditions because of rapid vertical mixing and also because the skewed nature of the vertical component of turbulence brings the plume centreline down towards the surface. This also results in the position of the surface maximum being closest to the source for unstable conditions. As conditions become less convective and finally stable the maximum surface concentration decreases whilst the downwind distance of greater impact at ground level increases significantly.

Figure 3.1 Variations of ground level concentration (µg m^{-3}) with downwind distance for a ground level point source. Emission rate 0.01 g s^{-1}, wind speeds for unstable, neutral and stable flow are 2, 4 and 2 ms^{-1} respectively.

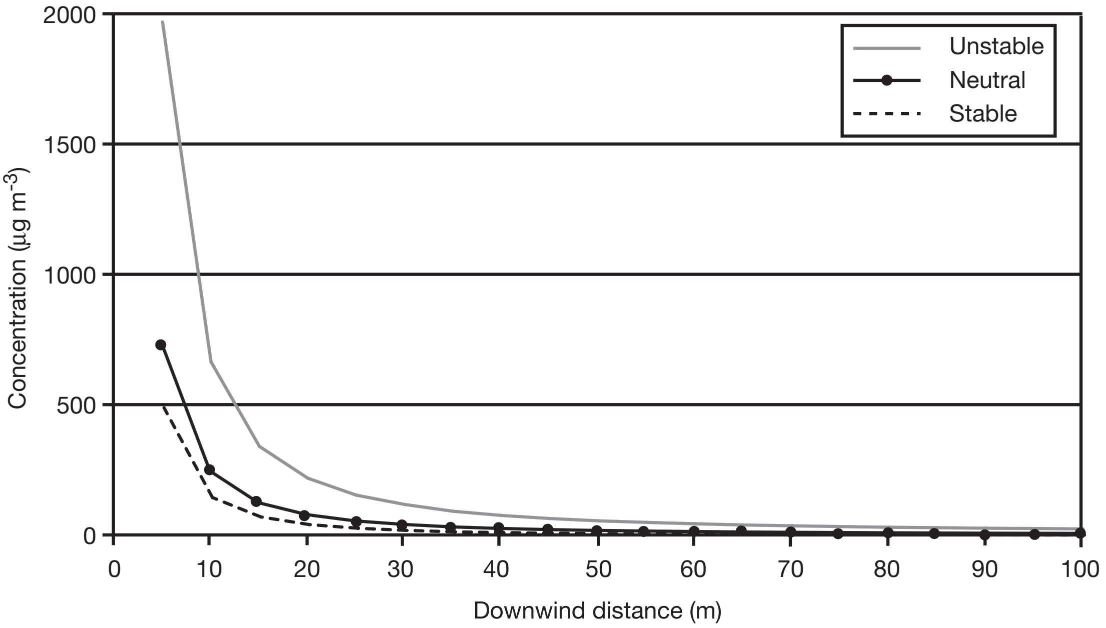
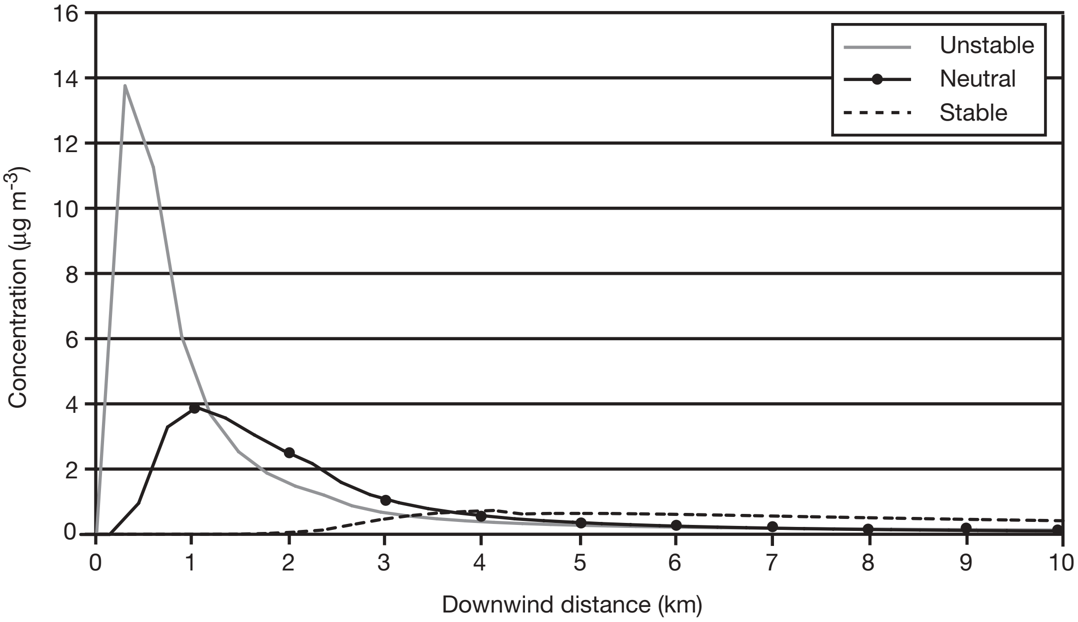


Figure 3.2 Variation of ground level concentration with distance for an elevated point source of height 99 m. The emission rate is 2 g s^{-1}. Wind speeds are as for Figure 3.1.



3.2.3. Urban meteorology

286. Exceedences of NO_2 objective limits are likely to be associated with urban areas, caused primarily by the NO_x emissions within urban areas. Winter episodes of very high concentrations of primary pollution, such as NO_x in urban areas, are associated with a high temperature-wind index according to Middleton and Dixon (2001), but secondary episodes of NO_2, for example, are more dependent on regional meteorology.

287. A purely deterministic description of the levels of NO_2 within an urban area would require a full description of emissions, dispersion and chemistry over very short spatial and temporal scales, which current models do not treat explicitly. Instead existing NO_2 models are either semi-empirical relying on a large set of relevant data, or attempt to describe these complex processes in an idealised way. The strong dependence of NO_2 on oxidant which tends to have a smoother, regional distribution, attenuates local concentration gradients. The calculation of primary pollution concentrations, such as NO_x, depends much more strongly on factors associated with dispersion. NO_2 tends to be a small fraction of NO_x determined by oxidant levels except when NO_x concentrations are low. Therefore the sensitivity of NO_2 concentrations to uncertainty in the NO_2-NO_x relationship is more important than the large uncertainties in urban meteorological factors. In this report a number of such relationships are used but none appear to depend strongly on urban meteorological factors.

288. It is therefore unclear how exceedences of the long-term NO_2 objective depend on urban meteorological factors. However since many measurement sites are within urban areas, it is important to review qualitatively the factors in an urban area which could influence dispersion. Differences between wind measurements made at a meteorological site within and outside a city illustrate climatological differences between urban and rural areas, but do provide explicit information about the local, site-specific factors which influence urban dispersion. These factors are reviewed below, in which the dynamical and thermal effects of urban areas are discussed separately.

3.2.3.1 Urban dynamical and thermal effects

289. In contrast to rural areas, the urban boundary layer is more complex, as a roughness sublayer of much larger vertical extension than found in typical rural areas occupies the first tens of metres above the surface, with the remainder of the surface layer (the inertial sublayer) above. The roughness sublayer includes the urban canopy layer, which is composed of individual street canyons and other roughness elements (see Figure 3.3). This figure illustrates the widely varying scales of motion affecting urban pollution concentrations. These motions and the turbulence associated with them determine the rate of mixing of pollution within an urban area. Pollution measurements are made between buildings, whose presence on aggregate change wind profiles and structure in an urban area compared with surrounding rural areas. Several simplifying approaches to describing urban wind and turbulence profiles have been proposed recently, such as Rotach (2002), Martilli *et al.* (2002) and Soulhac *et al.* (2002). Figure 3.4 illustrates schematically complex dispersion produced by the re-circulating flow within a street canyon.

290. The WMO-guideline for rural stations considers wind measurements to be representative if placed 10 m above ground without close obstacles; temperature and humidity measurements should be conducted at 2 m. For urban areas, no WMO guidelines for proper siting exist (Oke, 1999). Only in the upper part of or above the surface layer are contributions from individual surface roughness elements blended into possible representative averages.

Figure 3.3 Schematic description of the urban boundary layer including its vertical layers and scales. (Oke, 1999).

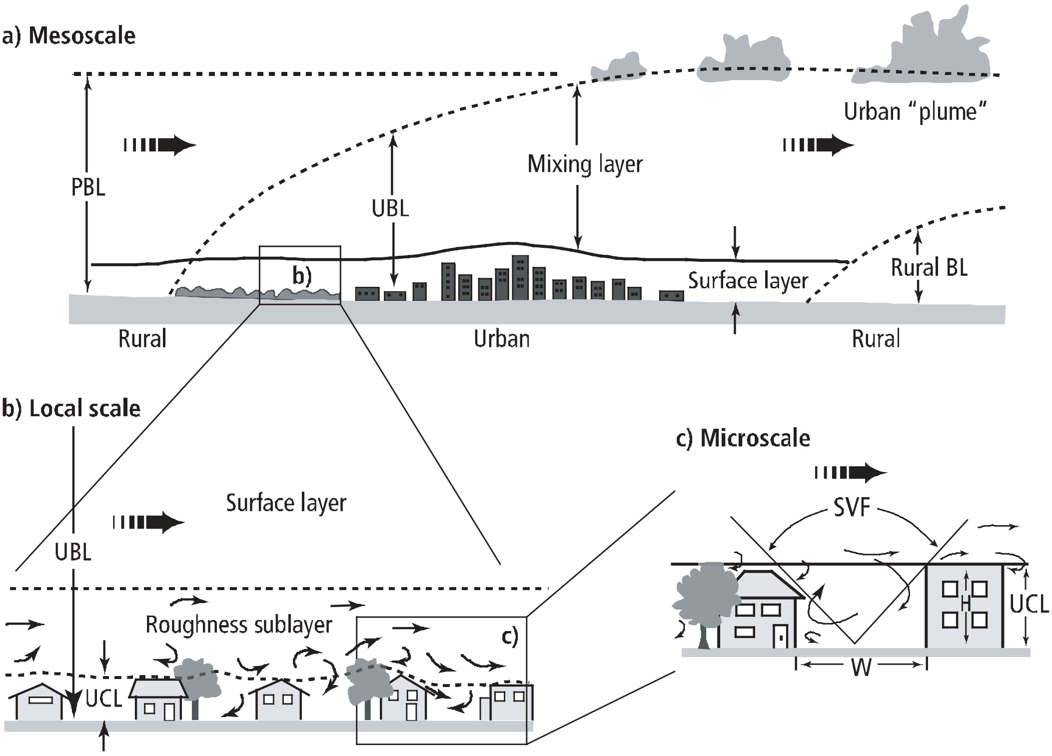
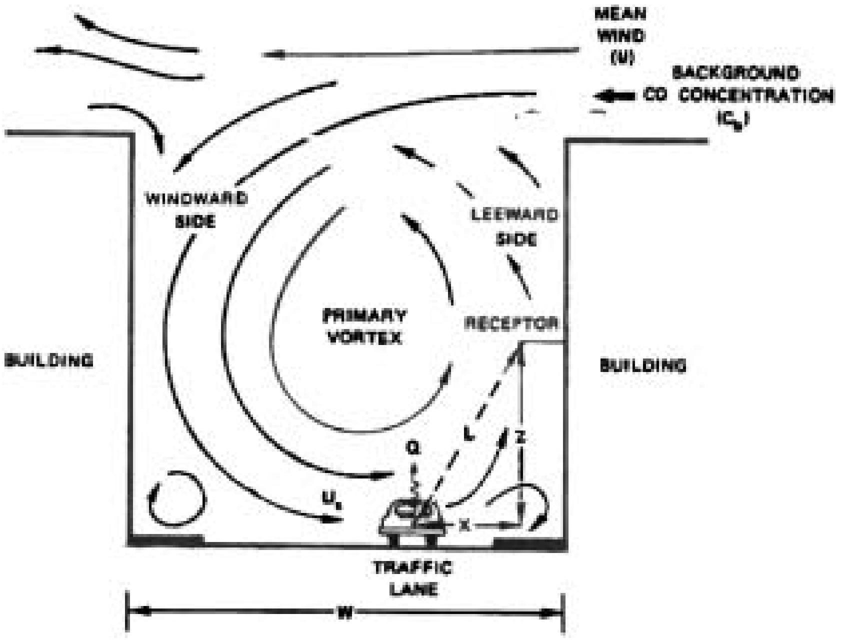


Figure 3.4 Schematic representation of the air flow within a street canyon.



291. The heating of air near the surface of urban areas is an important factor determining dispersion in lower layers of the urban atmosphere and is determined by the energy balance. This is the key component of any model aiming to simulate the partitioning of dynamical and thermodynamical patterns (Piringer, 2001) between net radiation, the turbulent sensible and latent heat fluxes, respectively, the storage heat flux and additional sources of energy due to human activities.

292. In urban areas there are marked differences in energy partitioning compared to rural conditions, where most measurements have been performed. Knowledge of surface heat fluxes as well as atmospheric stability and surface roughness is essential for air pollution dispersion models. Normally the components of the surface energy balance, or its components, are not directly measured at meteorological stations. In the last decade, a series of local-scale energy balance observations have been conducted to test practical schemes for estimating surface heat fluxes. Middleton (2002) gives extra detail on the experimental studies currently under way within Europe. Assessing methods of determining the height of the urban boundary layer, which is dependent on the surface heat fluxes, is another important task.

293. Schemes have been developed to estimate net radiation, sensible heat flux and other urban boundary layer parameters from hourly standard meteorological data. Such meteorological pre-processing models are needed to calculate dispersion parameters related to meteorology, before a dispersion modelling calculation can be performed. The goal of recent research is an urban pre-processor scheme, which makes use of parameterisations that require standard meteorological observations, supplemented by basic knowledge of the surface character of the target urban area. One such scheme, LUMPS, has been shown to perform well when evaluated using data from North American cities (Grimmond and Oke, 2001). However, a review (Schatzmann *et al.*, 2001) of current methods of obtaining urban meteorological data in Europe for dispersion calculations concluded that many of the operational approaches to the pre-processing of meteorological data in urban areas have little justification.

3.2.3.2 Reduction in complexity

294. The basic premise of the meteorological pre-processing schemes is that heat fluxes can be modelled using net radiation, simple information on surface cover (area of vegetation, buildings and impervious materials), surface geometry (surface element roughness and density) and standard weather observations (air temperature, humidity, wind speed and pressure). The aim of these methods is to reduce the complexity in the description of the urban surface heat flux with a minimum increase in uncertainty

295. Of course there are situations where these methods do not apply. Since the methods are based on a one-dimensional energy balance, it is unlikely to perform well in areas where there is significant spatial variability in land cover or surface geometry, for example, at the urban-rural interface.

296. Simplification methods have also been applied to the aerodynamic properties of urban areas. The roughness length and zero plane displacement are the two main properties influencing the flow. Two broad methods have been proposed: the geometric method which uses parameters which broadly describe the geometric form, or micrometeorological methods that use observations of wind and turbulence to derive parameters from the logarithmic wind profile. The later method requires tall instrumented towers. Sodar and meteorological masts have been used to derive the roughness length and displacement length for Lille (Wrobleski *et al.*, 2001). The former method requires knowledge of certain average geometric factors describing urban roughness elements, such as the average height of roughness elements (buildings or trees), fractional plan area, fractional frontal area etc. Relationships can be derived from idealised flows over simplified arrays in wind tunnels, but need to be tested in real situations (Grimmond and Oke, 1999). A classification of effective terrain roughness is often valuable in working situations (Davenport *et al.*, 2000).

297. For an air quality calculation one needs to derive a number of dispersion parameters from the meteorological factors, such as roughness length, zero plane displacement, surface heat flux, boundary layer height. Some may be derived from routine urban measurements and some from formulae based on idealised conditions (usually by applying some scaling rule). There are other parameters which are related to dispersion rather than to local meteorology, such as the velocity in the street (or equivalently within the urban canopy u_C), the exchange velocity w_E between the street and the flow above, and the canopy length Lc equal to the distance required for the flow to adjust to the step change between the urban and rural roughness. Study of methods for deriving these parameters is still a matter of research (Hanna *et al.*, 2002).

298. u_C, w_E and Lc are parameters which should be specified if assessments of pollutant concentrations in and near streets are to be made. They are required input parameters if well-known models, such as CALINE4 for roadside concentrations, and OSPM for street canyon concentrations (street canyon models are reviewed by Vardoulakis *et al.,* 2002a) are to be applied. Such models are necessary if roadside concentrations are to be estimated.

299. Vardoulakis *et al.* (2003) have shown that the prediction of concentrations in street canyons is subject to considerable uncertainty, arising from various causes (such as the uncertainties in emissions, local meteorology and the description of processes within a model). Vardoulakis *et al.* (2002b) recommend an approach based on a mixture of measurements and models. The semi-empirical approaches used in this report to assess roadside concentrations avoid these difficulties. However, uncertainties then remain as to what is meant by a roadside concentration, and how the exposure to high concentrations (Vardoulakis *et al.,* 2002a) depends on local factors, such as street geometry, distance from road etc. For example the site in London Bloomsbury is under trees within a square, so that mixing of nearby traffic emissions within the square will be different between summer and winter. The London Senator House site is located on the roof of an office block.

300. The London Marylebone Road site is within a street canyon, but the street does not consist of buildings of uniform height, nor are these continuous without gaps between them. Concentration gradients near to traffic emissions are steep, so that the estimated concentration along a road link, based on the aggregation of measurements made at various distances and locations near to different roads, represents an average which is difficult to define rigorously. One of the consequences of NO_x chemistry is that variations between different locations are likely to be attenuated reducing the seriousness of the problem. However, this is just one example of the difficulty of assessing what measurements at a site in an urban area represent.

3.2.3.3 Blending height

301. The problem of deciding what measurements at a single point within a region of spatially varying surface characteristics means, applies to both meteorological and pollution measurements. Applying a blending height, taken approximately to be 2.5 times the height of buildings, is a useful simplification. Above the blending height, effectively constant turbulence levels (described by the friction velocity) and roughness lengths can be defined for surfaces with inhomogeneous surface characteristics (spatially varying roughnesses or surface heat fluxes). Associated with the blending height are aggregation formulae, or weighted averages, of the surface characteristics. For the regional heat flux, a simple weighted average over the sub-areas of patches with different characteristics is used (Gryning and Batchvarova, 2001). For the effective roughness length a more complex averaging is needed. Such methods need to be tested, but are clearly essential for meso-scale models in which surface characteristics are averaged over some grid square, within which some effective exchange of heat or momentum is visualised to take place.

302. Meso-scale models are the main tool by which episodes of high pollution in urban areas are predicted, as shown by a review of European approaches conducted by Kukkonen (2001). In meso-scale models with scale of order 1 km, the main urban modification is to increase the roughness using a roughness length of order 1 m. This is defined by taking a logarithmic wind profile measured in the inertial sublayer, above the roughness sublayer. In an urban area the building density changes, and the urban boundary layer evolves and it is not obvious that a local roughness length can be defined. As described above pollution transport within the roughness sublayer is a further complication, but needs to be described in order to assess human exposure.

303. The so-called ‘urbanisation’ of meso-scale models is an important issue for improving urban pollution forecasts. Present methods include numerical weather prediction models, which may not see the urban area because the smallest horizontal length scale is too large. Finer scale models may have urban adjusted surface boundary conditions, or may have detailed descriptions of the urban neighbourhood nested within them (Baklanov *et al.*, 2002).

3.2.3.4 Inclusion of meteorology in urban dispersion calculations

304. The urban boundary layer height is the height to which the urban pollution mixes. Persistent low boundary layer heights are associated with pollution episodes as dilution is restricted. The blended surface heat flux is the appropriate quantity to use in estimating the height of the urban boundary layer (Baklanov, 2002). The literature is full of formulae (for example, Baklanov (2001)) for mixing height under stable conditions.

305. There are other features of urban areas for which there are at present no suggested practical formulae, for example on the relationship between the wind speed at a standard 10 m height outside a city to the wind speed on a mast, or above roof level within the city. Experimental results have been reported, for example:

- Roof-top wind (Leek U.K.) = 0.63 (airport wind at 10 m);
- Urban wind at 32 m (Lisbon) = 0.65 (rural wind at 10 m) + 1.24;
- Urban wind at 30 m (Copenhagen) = 0.51 (airport wind at 10 m);
- Urban wind at 30 m (Birmingham) = rural wind at 10 m.

306. Bezpalcova *et al.* (2002) have used numerical simulations to assess deviations from idealised wind profiles for simple cases of the urban atmospheric boundary layer. Thus the interpretation of the differences between measurements at Heathrow and the London Weather Centre is difficult.

307. Methods developed to describe dynamic and thermodynamic aspects of urban meteorology have application in the practical methods for predicting urban pollution levels based on source-receptor relationships. However within the roughness layer of the urban atmosphere which contains all the measurement sites, it is difficult to make recommendations. Belcher and Coceal (2002) introduce an additional spatial averaging term into the momentum equations describing the flow within the lower layers of the urban atmosphere. Similarly Carissimo and Macdonald (2001) introduce effective drag, and turbulence terms to describe the averaged flow equations within the urban canopy.

308. Even in situations when very detailed turbulence measurements are available above the roof canopy, for example, Basel, Hannover (Mueller *et al.*, 2002) and Nantes (Mestayer *et al.*, 2002) there is a need to extrapolate to other locations or positions. Paschke *et al.* (2002) argue that the use of systematic wind tunnel experiments is the way to close gaps in field data. It is essential to identify urban meteorological issues relating to urban pollution assessments. In addition for practical use, advice needs to be given on urban correction factors. The starting point for this has been the inventory of urban meteorological stations, which now comprises over 300 sites from all countries active in COST 715 (Mueller *et al.*, 2002).

309. However the influence of meteorology is indirect. It is only one of many factors, which can influence urban pollution predictions. The outcomes of a pollution assessment are predictions of ground-level concentrations and the success of an assessment is judged against measurements of concentrations, not meteorological variables. Rotach (2001) has, for example, carried out this process using two urban simulations, one with and one without the roughness sublayer (as would normally be adopted in a dispersion calculation assuming surface characteristics prevail down to the ground). For limited measured data sets, the urban parameterisation performs better. A similar approach has been used to produce an urban meteorological pre-processor for Helsinki (Karppinen *et al.*, 2000), dividing the surface layer into an inertial and roughness sublayer and adopting an upper bound on surface cooling in stable atmospheric conditions, again showing differences in dispersion. These approaches have been applied to whole cities, namely Zürich (de Haan *et al.*, 2001) and Helsinki (Karppinen *et al.*, 2000). In the U.K. the application of ADMS Urban is a comparable approach.

3.2.3.5 Role of urban meteorology in NO_2 assessments

311. Urban meteorology is of major importance in assessing concentrations of primary pollutants, such as NO_x. A set of empirical curves may be used to represent the relationship between NO_2 and NO_x. The curves vary according to the type of source and the amount of oxidant available.

311. Thus the semi-empirical approaches to urban NO_2 concentration assessments provide an assessment method without the need to include urban meteorology explicitly. Given the current difficulties of describing urban meteorology this has some advantages. However the empirical approaches have the limitation that impact of certain factors cannot be evaluated. Therefore process models such as ADMS Urban, have been applied widely, with the more complex aspects of urban meteorology neglected They provide reasonably adequate assessments within a reasonable degree of uncertainty. Future work should be directed to including the more detailed influences of urban meteorology.

3.2.4. Station meteorological data for use in air pollution modelling

312. Since exposure to NO_2 is almost exclusively an urban problem in the UK, much attention is paid to the availability and quality of meteorological data from urban meteorological stations for use as input to modelling studies. Unfortunately:

- there are a declining number of urban weather stations as a result of recent changes to the monitoring network;
- it is difficult to collect meteorological data which sufficiently characterises the important range of environments which is typical of urban regions (Oke, 1999) and compromises in instrument exposure commonly have to be made;
- insufficient studies have been carried out comparing those urban measurements which do exist against upwind and downwind rural stations.

313. As a result there is a very incomplete understanding of the sensitivity of model results to the weather station selected as being most representative and to the particular year of data used. Rotach *et al*. (2001) are addressing this issue by designing monitoring strategies which consider the variation in meteorological conditions across a city both in the horizontal and the vertical plane.

314. The selection of meteorological data to be used as input to air pollution models is a crucial step and the following section highlights spatial and temporal issues which should be considered.

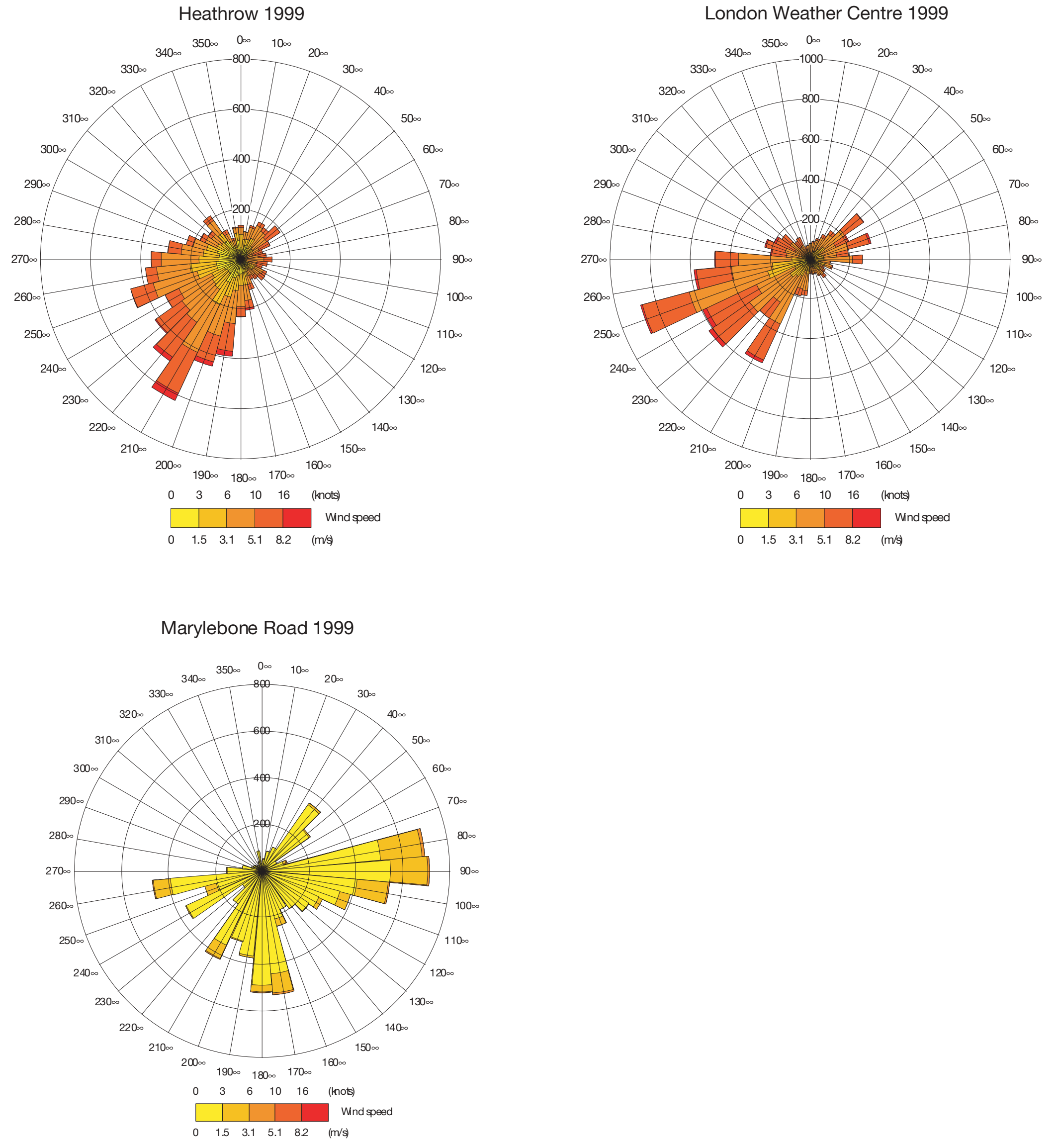
3.2.4.1 Variations between neighbouring stations

315. Table 3.1 and Figure 3.5 illustrate the difference between meteorological data recorded at Heathrow, London Weather Centre (LWC) and Marylebone Road. Note the lower wind speed at Heathrow and that the prevailing direction is backed relative to LWC. The LWC site is two metres above a 29 m building in Central London. The wind speed is retarded by the high roughness of Central London, but is greater than Heathrow because of the height of observation and flow speed-up over building top. The wind speeds measured in the Marylebone Road canyon can be seen to be significantly lower than at the other two sites.

Table 3.1 Summary of 1999 meteorological data.

	Heathrow			London Weather Centre		
Location and type	(507700, 176700) in the Heathrow airport/flat terrace			(530200, 180000) 2 m above building; Central London		
Height above ground	10 m			31 m		
Roughness length	0.2 m			1 m		
Data Capture	99.8%			95.9%		
Statistics	Mean	Minimum	Maximum	Mean	Minimum	Maximum
Temperature (°C)	11.8	-4.6	32.7	12.5	-1.3	31.9
Wind speed (m s^{-1})	3.1	0.0	12.9	4.0	0.5	12.9
Cloud cover (oktas)	5.6	0.0	8.0	5.5	0.0	8.0

Figure 3.5 Windroses for meteorological data from London for 1999.



3.2.4.2 Inter-annual variations

316. Inter-annual variations in meteorology are thought to impact annual average concentrations by no more than ±15% (Defra, 2003). Middleton and Dixon (2001) show how significant inter-annual differences in wind roses and frequency of 'calm' conditions can be observed at a single monitoring station (Heathrow Airport). Sensitivity studies are recommended which take account of this potential, including 'worst case' scenarios.

3.2.5. NO_2 episodes

3.2.5.1 Classification methods

317. A number of tools have been and continue to be used in analysis of weather conditions accompanying poor air quality conditions:

- Station wind roses (for example, Middleton and Dixon, 2001)
- Isobaric, geopotential height and windfield analyses from numerical weather prediction models (for example, Dorling and Doyle, 2003)
- Back trajectories (for example, Stohl, 1998; Jenkin *et al.*, 2002)
- Subjective and Objective Weather Type Classifications (for example, Davies *et al.*, 1991; Buchanan *et al.*, 2002)
- Stagnation Indices (for example, Middleton and Dixon, 2001; Wang and Angell, 1999; Dorling *et al.*, 2003)

318. In many cases a number of these are used in combination in order to add further confidence to the interpretation.

319. Wintertime NO_2 episodes are strongly related to the incidence of periods of air stagnation and the accumulation of pollution from local sources. Summertime episodes, meanwhile, are affected by the advection of O_3 from upwind sources. Since the frequency of these important episode-related weather conditions varies from year to year, trends in precursor emissions can be masked by inter-annual variations in meteorology.

320. Dorling and Doyle (2003) have attempted to capture the variability in seasonal synoptic scale weather patterns by clustering daily fields of the height of the 850 hPa pressure surface , over the period 1991-2001, using the NCEP re-analysis dataset (Kalnay *et al*., 1996). Such an approach is arguably best suited to reviewing variation in weather conditions over an extended period. The dataset is divided into 5-day non-overlapping periods and ten clusters are retained in each season (DJF, MAM, JJA, SON). As examples, Figures 3.6 and 3.7 show the geographical domain considered and the middle day of each cluster for winter and for summer.

Figure 3.6 Winter 850 hPa pressure surface height (metres) clusters (cluster 1 top left, cluster 2 bottom left, cluster 10 bottom right).

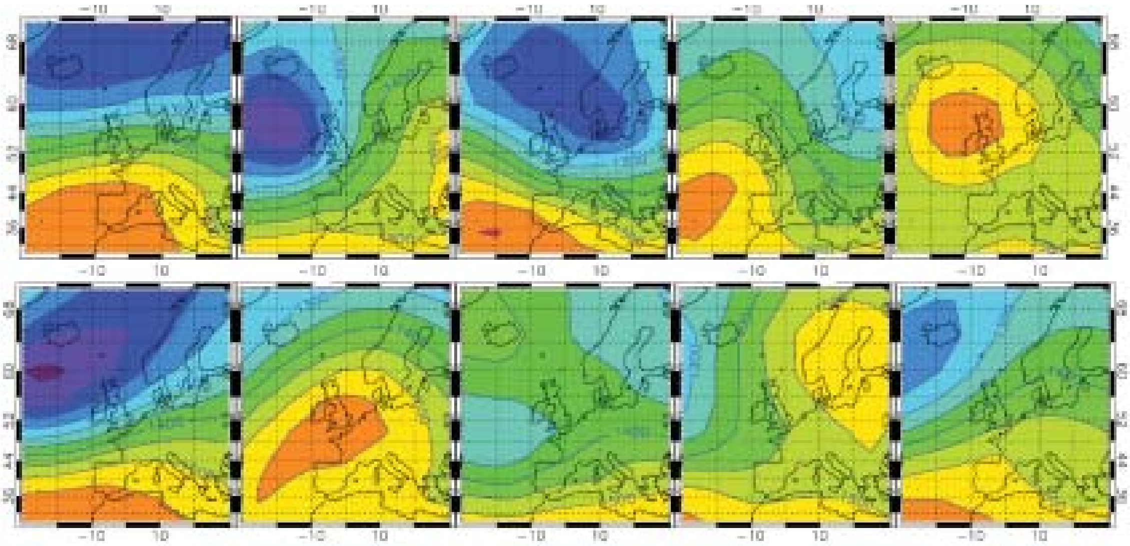
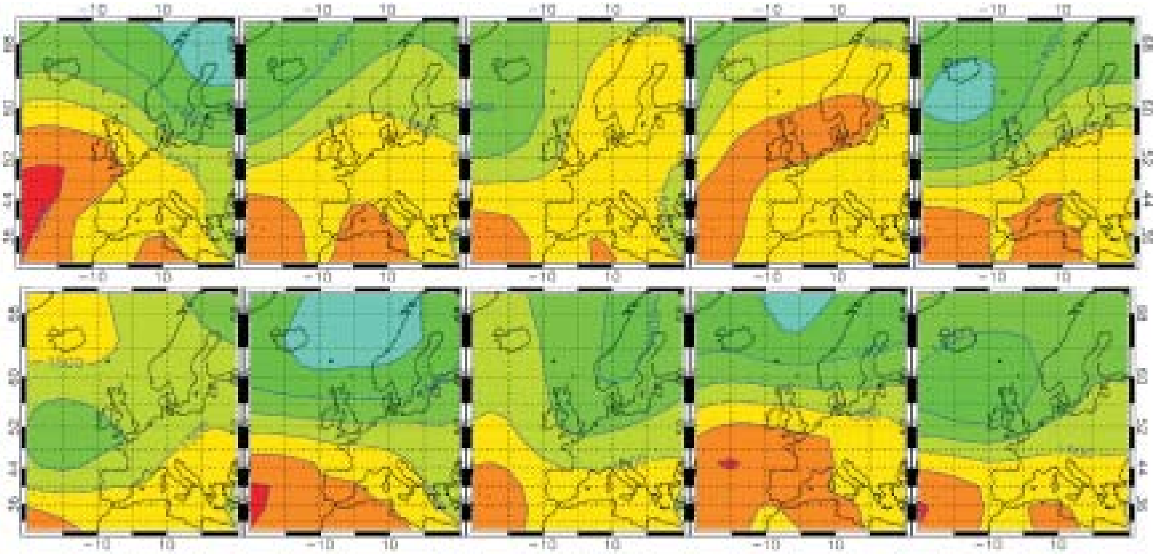


Figure 3.7 Summer 850 hPa pressure surface height (metres) clusters (cluster 1 top left, cluster 2 bottom left, cluster 10 bottom right).



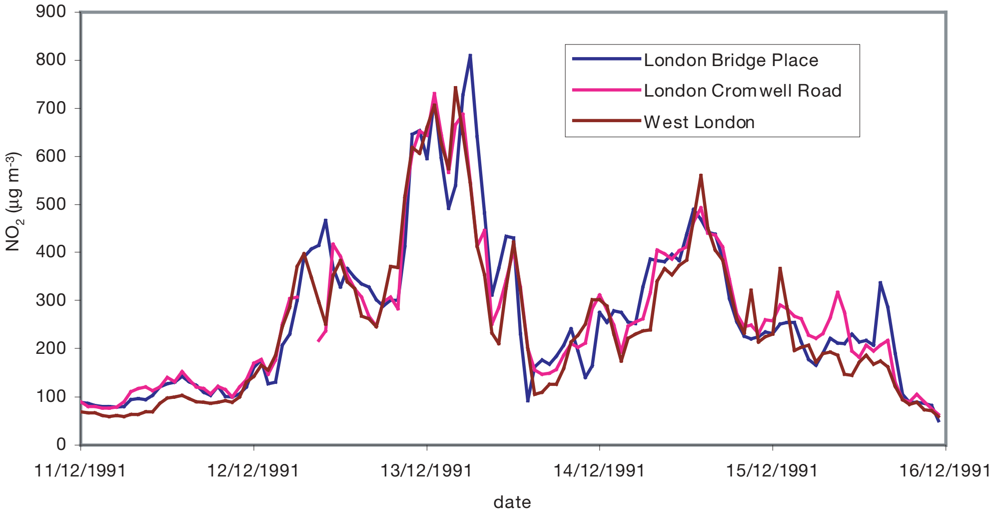
321. Figure 3.6 shows that winter clusters 4 and 9 represent situations where anticyclones (and poor dispersion) dominate UK winter weather most. As the month of lowest incidence of solar radiation (and hence maximum incidence of temperature inversions and poor dispersion) and of minimum vehicle engine efficiency (associated with ambient temperatures), December is especially exposed to conditions of poor dispersion. December 1991, 1992 and 2001 stand out in the recent observational record in terms of the occurrence of consecutive periods of anticyclonic winter clusters 4 and 9. December 1991 and 2001 are also especially prominent in the high NO_2 concentration observational record.

322. The usefulness of this approach is apparent in the analyses of the following high UK NO_2 concentration episodes. The approach also lends itself to an analysis over an extended historical period and can also be applied to climate model output.

3.2.5.2 11th-15th December 1991 – London and South-East England

323. Bower *et al.* (1994) and Derwent *et al.* (1995) provide a detailed analysis of the meteorological conditions which accompanied this noteworthy NO_2 episode. Figure 3.8 presents a timeseries of the NO_2 concentration during this episode.

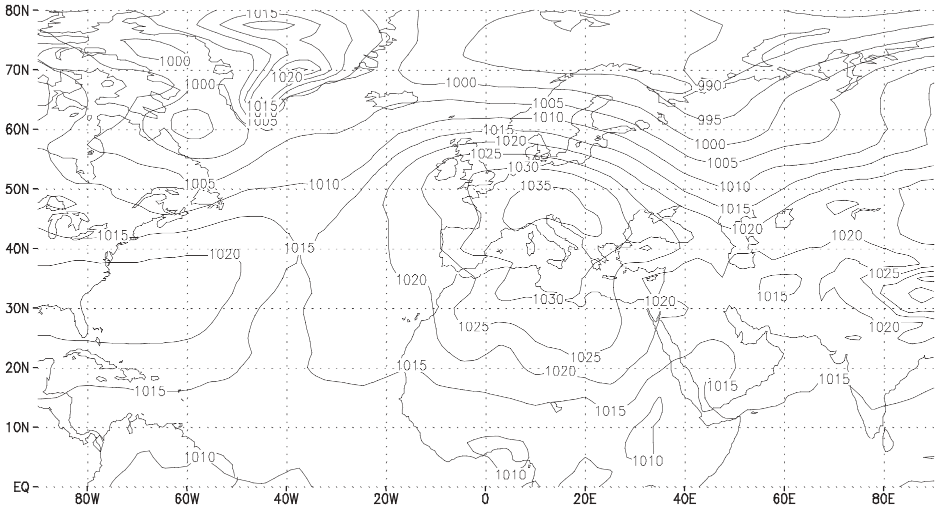
Figure 3.8 NO_2 concentrations in London 11th–15th December 1991 (µg m^{-3}).



324. The period 2nd-6th December 1991 was classified as being of Winter cluster type 9, followed by a spell of cluster 4 over the period 7th-16th December (Figure 3.6). This was an extended period of light winds, low insolation levels (exacerbated by periods of fog), temperature inversions and poor dispersion generally.

325. Figure 3.9 presents a composite sea-level pressure chart for the duration of the main period of the episode, 12th-15th December 1991, demonstrating the relatively weak pressure gradients and the influence of an anticyclone centred over the near continent.

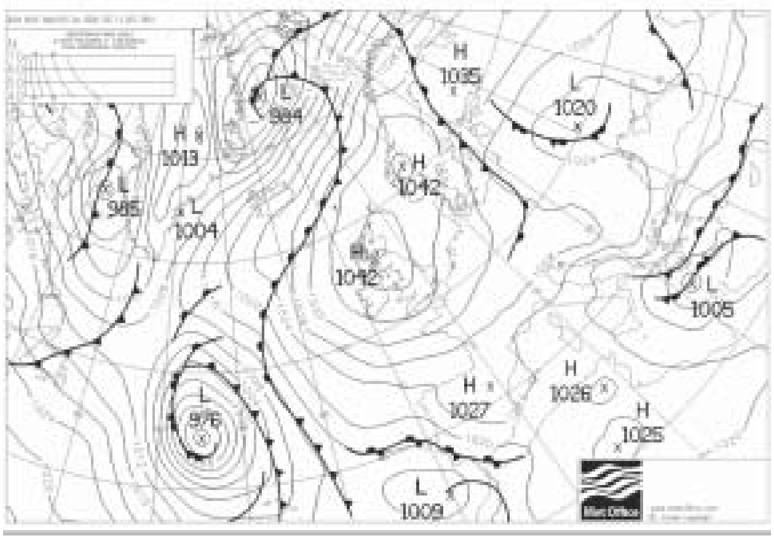
Figure 3.9 Composite sea-level pressure analysis for the period 12th-15th December 1991.



3.2.5.3 11th-12th December 2001 – North-West Britain

326. A timeseries of NO_2 concentrations during this episode is presented in Chapter 6. Figure 3.10 presents the sea-level pressure analysis for the 12th December 2001 by way of example. On this occasion an example of winter cluster 4 occurred between 4th and 8th December, followed by consecutive examples of cluster 9 between the 9th and the 18th (Figure 3.6). This sequence was the reverse of the conditions accompanying the December 1991 episode considered previously, and led to cities in NW Britain (Glasgow, Manchester, Belfast) being more severely affected this time. However, once again, an extended period of relatively calm weather was shown to be necessary.

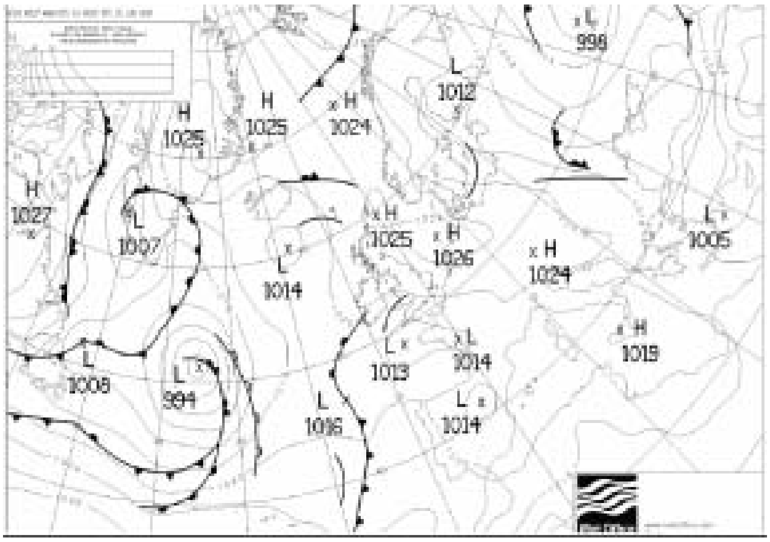
Figure 3.10 Sea-level pressure analysis for 00z 12th December 2001.



3.2.5.4 26th June 2001 – London and the South-East of England

327. A timeseries of NO_2 concentrations during this episode is presented in Chapter 6. Maximum temperatures on this day rose to 30°C and the sea-level pressure analysis chart is shown in Figure 3.11.

Figure 3.11 Sea-level pressure analysis for 00z 26th June 2001.



328. Summer Cluster 6 (Figure 3.7) best represents the situation where a ridge of high pressure over Western Europe is leading to advection over Southern Britain from the European continent. It is not surprising that this cluster was indeed found to best describe conditions over the period 17th-26th June 2001.

329. High concentrations of European-derived O_3 are likely to have been available to promote NO_2 formation in polluted environments, especially in SE England. Jenkin *et al.* (2002) have shown that there is a tendency in the UK for O_3 episodes to preferentially occur later in the working week as a result of the accumulation of precursor pollutants and the timescales involved in their advection from source regions.

3.2.5.5 Trends

(a) Historical

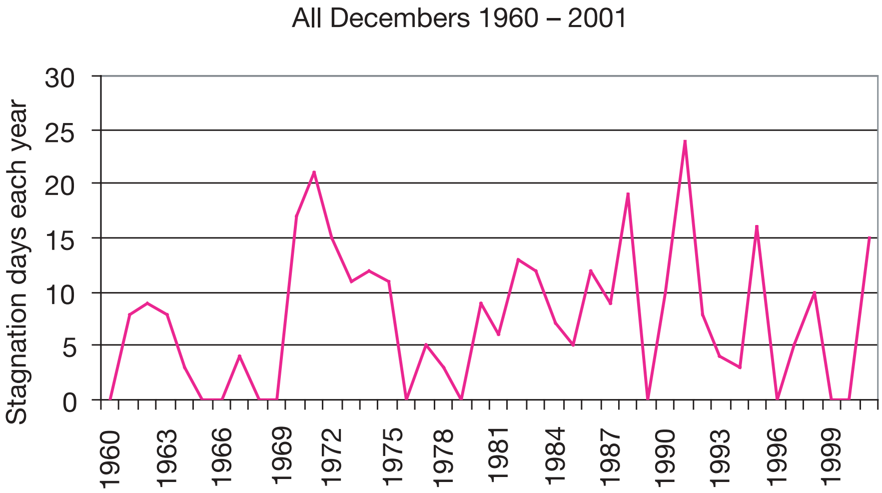
330. Middleton and Dixon (2001) show that a significant declining frequency of 'calm' conditions appears to be evident in the station wind data collected at Heathrow Airport over the period 1949-1997. This same trend is also evident when considering December alone, although December 1991 stands out as a very high frequency 'calm' month, in keeping with the episode discussion above.

331. A regional scale quantification of poor dispersion conditions can be derived through the use of a stagnation index. After analysing the meteorological characteristics of recent historical NO_2 episodes, Dorling *et al.* (2003) defined a stagnation index over the UK where the criteria for stagnation are the persistence for three days or more of:

- a high pressure feature over the UK and/or immediate surroundings with central pressure greater than 1030 hPa;
- a sea-level pressure gradient over the UK of < 2.5hPa/250km (equivalent to a surface windspeed of <3.2 m s^{-1}).

332. Application of these criteria to the 2.5°x2.5° resolution NCEP re-analysis dataset (Kalnay *et al.,* 1996) over the period 1960-2001 generates the stagnation frequency results for the month of December shown in Figure 3.12. The episodes in December 1991 and 2001 discussed in sections 3.2.5.2 and 3.2.5.3 clearly affect the frequency data shown. Also interesting, however, is the significant interannual variability and relatively low frequency of stagnation events in the 1960's.

Figure 3.12 December Stagnation Day frequency over the UK derived from the NCEP re-analysis dataset.



(b) Future

333. The stagnation index definition described above can also be applied to the output of the UK Meteorological Office 50 km resolution Regional Climate Model, HadRM3 (Dorling *et al.*, 2003). The HadRM3 control climate (1960-90) simulation of the seasonal cycle of stagnation frequency over the UK is very similar to that observed in the NCEP re-analysis dataset. This result encourages consideration of the HadRM3 simulation of stagnation frequency in the future climate period, 2070-2100. Table 3.2 presents the stagnation frequencies simulated in the winter months in the HadRM3 control climate (H3B2a_achgi) and in two 2070-2100 climate change scenarios (Hulme *et al.*, 2002) known as 'Mitigation' (Medium-Low H3B2a_ackdd) and 'Business as Usual' (Medium-High H3A2a_ackda).

Table 3.2 Percentage Frequency of Total Stagnation Days for the HadRM3 Control Climate (achgi) and the 'Mitigation' (ackdd) and 'Business as Usual' (ackda) 2070-2100 simulations.

% Days	Dec	Jan	Feb	Winter	Year
1960-1990 Control Climate	10.97	14.62	15.48	13.69	9.43
2070-2100 Business as Usual	7.74	5.48	8.06	7.10	6.31
2070-2100 Mitigation	11.29	7.96	12.04	10.43	7.81

334. Taking the winter as a whole, both of the future scenarios show a substantial fall in stagnation day frequency over the UK relative to the control climate period. However, the 'Business as Usual' emission scenario leads to the more dramatic frequency reduction of almost 50%. These findings are consistent with the more general trends in winter windspeeds reported in Hulme *et al.* (2002) and Anderson *et al.* (2000).

3.3. Chemical processes

335. NO_2 is predominantly a secondary pollutant, its major atmospheric source being the oxidation of emitted NO. An understanding of the chemical processes which produce and destroy NO_2 is therefore central to interpretation of ambient measurements, and to the prediction of how its concentration is likely to vary with implementation of NO_x emissions controls. In this section, a description of the main chemical and photochemical reactions which influence the level of NO_2 is provided.

336. In this section (and in section 5.2.4), graphs which present data for either O_3 or oxidant (defined below), in addition to NO_2, exceptionally make use of ppb units in preference to μg m^{-3}. This is because the ideas and relationships which are being presented relate directly to the chemical coupling of NO, NO_2 and O_3, as described by reactions (1) – (3). The reactions occur on a molecular basis, such that one molecule (or ppb) of NO in reaction (1) reacts with one molecule (or ppb) of O_3, to generate one molecule (or ppb) of NO_2 (in mass terms 1 μg m^{-3} NO reacts with 1.60 μg m^{-3} O_3 to generate 1.53 μg m^{-3} NO_2). It is therefore more logical and instructive to present the data in the molecular units in these instances.

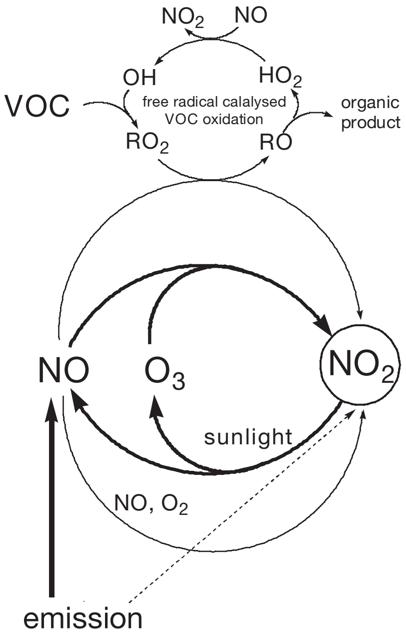
3.3.1. Interconversion of NO and NO_2

337. Under the majority of atmospheric conditions, the dominant pathway by which NO is converted to NO_2 is via the reaction with O_3 (see Figure 3.13):

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (1)$$

338. At polluted locations comparatively close to sources of NO_x, NO is in large excess and the availability of O_3 provides a limit to the quantity of NO_2 that can be produced by this reaction. The timescale for consumption of O_3 depends on the concentration of NO. At the high end of the range of hourly-averaged concentrations generally observed at polluted roadside locations (ca. 1 ppm), the time constant for O_3 consumption is ca. 2 seconds, with this lifetime increasing at lower levels of NO, in inverse proportion. At unpolluted locations, when O_3 is present in excess, the timescale for conversion of NO to NO_2 by reaction (1) is ca. 90 seconds, at a typical boundary layer background concentration of 30 ppb O_3. At intermediate levels of NO_x, when neither NO nor O_3 is in large excess, reaction (1) progressively depletes both reagents, and the reaction time constant can become very long. For example, in an airmass initially containing 15 ppb of both NO and O_3, it takes ca. 30 minutes for 90% conversion of NO (or O_3) to NO_2 by reaction (1).

Figure 3.13 Schematic representation of the main pathways interconverting NO and NO_2 in the atmosphere. Sources of free radicals which promote the free radical catalysed VOC oxidation (and NO-to-NO_2 conversion) are discussed in the text.



339. During daylight hours, NO_2 is converted back to NO as a result of photolysis, which also leads to the regeneration of O_3 as follows,

$$NO_2 + \text{sunlight} \rightarrow NO + O \qquad (2)$$

$$O + O_2\ (+M) \rightarrow O_3\ (+M) \qquad (3)$$

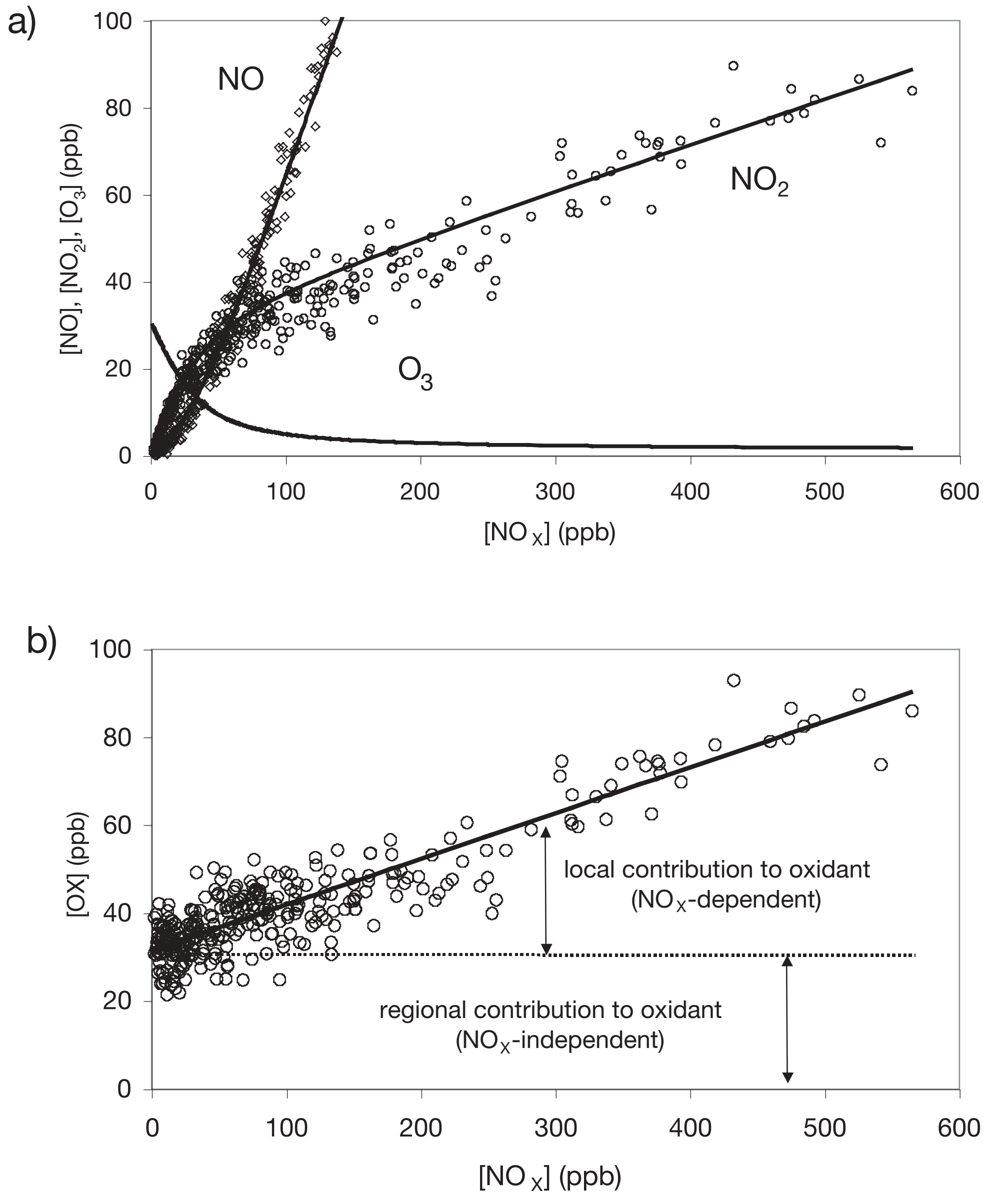
where M is a third body, most commonly N_2. As a result of this efficient interconversion by reactions (1) – (3), the behaviour of NO and NO_2 is highly coupled under atmospheric conditions, and it is convenient to refer to these species collectively as NO_x. Because this coupling also involves O_3, however, NO_2 and O_3 are also often collectively defined as ‘oxidant (OX)’. Consequently, reactions (1) – (3) are a cycle with no net chemistry, which has the overall effect of partitioning NO_x between its component forms of NO and NO_2, and OX between its component forms of O_3 and NO_2, but leaving the total concentration of both NO_x and OX unchanged.

340. The lifetime of NO_2 with respect to photolysis in the boundary layer depends on latitude, season and time of day. As shown in PORG (1997), summertime measurements indicate that the minimum lifetime of NO_2 under conditions typical of the UK is of the order of 100 seconds, with a mean daylight lifetime of ca. 3 minutes. In the wintertime, this is typically a factor of two or three longer. Under conditions when photolysis is sufficiently rapid, NO, NO_2 and O_3 are potentially in chemical equilibrium, a condition usually referred to as 'photostationary state' (Leighton, 1961). The above cycle predicts that the photostationary state concentrations of the three species are related by the expression $[NO].[O_3]/[NO_2] = J_2/k_1$, where J_2 is the rate coefficient of NO_2 photolysis, and k_1 is the rate coefficient for the reaction of NO with O_3.

341. For a variety of reasons, this relationship does not always hold precisely (for example, see Calvert and Stockwell, 1983; Carpenter *et al.*, 1998). As shown in Figure 3.14a, however, it does provide a good semi-quantitative description of how daylight ambient concentrations of NO, NO_2 and O_3 vary with concentration of NO_x, consistent with the chemical coupling of these species being dominated by reactions (1) – (3). These data indicate that the NO_x crossover point (i.e., when $[NO] = [NO_2]$) typically occurs at about 115 μg m^{-3} NO_x, as NO_2 (60 ppb NO_x). At lower levels, NO_2 is the major component of NO_x, whereas NO dominates at higher concentrations. The OX crossover point (i.e., when $[O_3] = [NO_2]$) typically occurs at about 25 ppb NO_x, with O_3 being the dominant form at lower levels, and NO_2 dominating at higher levels. This pattern is typical, although the precise crossover points vary with conditions (for example, with the value of J_2).

342. Figure 3.14a thus clearly shows how the decrease in O_3 concentration as NO_x increases, is accompanied by an approximately corresponding increase in NO_2 concentration. It also shows, however, that NO_2 levels continue to increase with NO_x even when O_3 levels are close to zero (i.e. at > ca. 100 ppb NO_x). Indeed, Figure 3.14b shows that total OX appears to increase approximately linearly with NO_x over the entire range, such that the level of OX at a given location has a NO_x-independent contribution, and a NO_x-dependent contribution. The former is effectively a 'regional' contribution which equates to the regional background O_3 level, whereas the latter is effectively a 'local' contribution which correlates with the level of primary pollution. Elevated levels of OX at urban locations (i.e. mainly NO_2) may therefore be generated either by having a large local source of oxidant (i.e. high NO_x levels), or by having a large regional source of oxidant in the presence of moderate NO_x levels. The former conditions are most likely to occur in wintertime episodes, when levels of NO_x ca. 700 ppb are typically required for the hourly threshold of 105 ppb (200 μg m^{-3}) to be exceeded for NO_2. During summertime, when boundary layer depths are much greater, such levels of NO_x are less common and NO_2 exceedences are only likely to occur when there is a large regional input of oxidant resulting from elevated background O_3 levels during photochemical pollution episodes (for example, as described by Jenkin *et al.*, 2002).

Figure 3.14 Example of variation of daylight average concentrations of (a) O_3, NO and NO_2, and (b) oxidant (OX = O_3 + NO_2) with level of NO_x. Data are presented for each day of November 1998 and 1999 at Marylebone Rd., Bloomsbury, Hillingdon, Teddington, Reading and Harwell. The lines in (a) were calculated with the assumption of photostationary state, using a daylight-averaged value of $J_2 = 2.9 \times 10^{-3}\ s^{-1}$, with OX constrained to values described by $[OX] = 31.1 + 0.104.[NO_x]$, based on a linear regression of the data presented in (b) (adapted from Clapp and Jenkin, 2001).



343. A significant proportion of the local contribution is likely to result from primary emission of NO_2, this is variable, but is believed to account for ca. 5% of emitted NO_x, on average. Additional thermal and photochemical sources also potentially contribute, as shown schematically in Figure 3.13. These are all processes which convert NO to NO_2 without associated removal of O_3, and are therefore net sources of OX. Some conversion of NO to NO_2 can potentially occur from the termolecular reaction of NO with O_2:

$$2\ NO + O_2 \rightarrow 2\ NO_2 \qquad (4)$$

344. The rate of this reaction is strongly dependent on the NO concentration, such that it is much more rapid at the elevated levels typical of those close to points of emission. For example, the time for 1% conversion of NO to NO_2 by this reaction is ca. 20 seconds at 100 ppm NO in air, but increases dramatically as NO is diluted. At 1 ppm NO (i.e. the high end of the hourly averaged values observed at roadside locations) the time for 1% conversion is ca. 30 minutes. The extent to which this reaction can contribute is therefore strongly influenced by the rate at which NO is diluted following emission. It is probable that only limited NO to NO_2 conversion by reaction (4) occurs under typical ambient conditions. However, under wintertime pollution episode conditions, when a shallow inversion layer can lead to a combination of high NO_x levels and stagnant air for periods of a day or more (for example, QUARG, 1993; Bower *et al.*, 1994), reaction (4) can potentially make a substantial contribution to OX generation.

345. Other chemical processes which convert NO to NO_2 involve the formation of free radicals, which can catalyse the oxidation of emitted volatile organic compounds, VOC. As shown in Figure 3.13, the reactions of the hydroperoxy radical (HO_2) and organic peroxy radicals (RO_2) with NO,

$$HO_2 + NO \rightarrow OH + NO_2 \qquad (5)$$

$$RO_2 + NO \rightarrow RO + NO_2 \qquad (6)$$

contribute to the catalytic VOC oxidation cycles, and provide the necessary coupling between the VOC and NO_x chemistry. For these cycles to contribute significantly to NO to NO_2 conversion (and therefore OX formation) under urban conditions, significant sources of free radicals (hereafter referred to as 'HOx') are required to initiate the process. A number of thermochemical and photochemical sources of potential significance have been identified, which involve species emitted concurrently with NO_x. The formation of HOx from the thermal reactions of alkenes with O_3 has received particular attention (Paulson and Orlando, 1996; Bey *et al.*, 1997), and these reactions potentially contribute at moderate NO_x levels when O_3 is also available. The thermal reactions of NO_2 with some conjugated dienes emitted from road transport have also been found to generate significant yields of HOx radicals (Atkinson *et al.*, 1984; Shi and Harrison, 1997; Jenkin *et al.*, 2003). These reactions are comparatively slow, however, such that they are only likely to contribute under wintertime pollution episode conditions when boundary layer levels of NO_x and VOC are significantly elevated (Harrison *et al.*, 1998).

346. A detailed discussion of the comparative importance of photochemical sources of HOx radicals for a variety of conditions is available elsewhere (for example, PORG, 1997; Jenkin and Clemitshaw, 2000). Under polluted conditions, the photolysis of formaldehyde, HCHO (and possibly other carbonyls), and nitrous acid, HONO, have been identified as potentially important:

$$HCHO + \text{sunlight} \ (+2O_2) \rightarrow CO + HO_2 + HO_2 \qquad (7)$$

$$HONO + \text{sunlight} \rightarrow OH + NO \qquad (8)$$

347. Both species are known to be emitted in road traffic exhaust (PORG, 1993), such that their emissions correlate with NO_x. HONO is particularly photolabile, its photolysis lifetime being typically 15 and 40 minutes under daylight-averaged midsummer and midwinter conditions, respectively (PORG, 1997).

3.3.2. Chemical removal of NO_x

348. NO and NO_2 are efficiently interconverted during daylight hours by the processes described above. Other atmospheric chemical transformations of NO_x typically occur on a longer timescale, leading to the generation of a variety of inorganic and organic oxidised nitrogen compounds. Together with NO and NO_2, these species are usually collectively referred to as 'NOY'. The various compounds make variable contributions to NOY, and their formation and removal chemistry is described in detail elsewhere (for example, PORG, 1997; Jenkin and Clemitshaw, 2000). The predominant loss process for NO_x results from its conversion to 'nitrate', either in the form of gaseous nitric acid (HNO_3) or nitrate aerosol, followed by physical removal by dry or wet deposition. The formation of other oxidised nitrogen compounds, such as HONO and peroxyacetyl nitrate (PAN), are only temporary loss processes for NO_x under boundary layer conditions, since these species are broken down comparatively efficiently to regenerate NO_x. However, they potentially have additional significance through interfering with measurement of NO_2 by widely applied techniques. In the present section, attention is therefore given to describing the chemical processes which lead to the formation of nitrate, HONO and PAN in the boundary layer.

3.3.2.1 Formation of nitric acid (HNO_3) and nitrate aerosol

349. The predominant chemical removal route for NO_x during daylight is the reaction of OH with NO_2, which can therefore play a major role in controlling the ambient NO_x concentration:

$$OH + NO_2\ (+M) \rightarrow HNO_3\ (+M) \qquad (9)$$

350. At a typical background OH concentration of 0.04 ppt, NO_2 is converted to HNO_3 at about 5% hr^{-1} by this reaction, although the conversion rate is correspondingly more rapid at elevated OH concentrations consistent with a photochemical episode (i.e. up to ca. 0.1 ppt). HNO_3 therefore becomes an increasingly important component of NOY downwind of source regions (for example, Harrison *et al.*, 1999). It is removed comparatively efficiently from the troposphere by both wet and dry deposition (for example, Huebert and Robert, 1985; Derwent *et al.*, 1988), and also by adsorption on, or reaction with, the tropospheric aerosol (for example, Cox, 1988; Fenter *et al.*, 1995), leading to the formation of nitrate aerosol. Reaction of HNO_3 with gaseous ammonia also leads to ammonium nitrate aerosol formation, which is a reversible temperature and humidity-dependent process (PORG,1997).

351. Nitrate aerosol is also generated via the formation of the higher oxides, NO_3 and N2O5, as follows:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad (10)$$

$$NO_3 + NO_2\ (+M) \rightleftharpoons N_2O_5\ (+M) \qquad (11,-11)$$

$$N_2O_5 + H_2O\ (+\text{aerosol}) \rightarrow 2\ HNO_3\ (\text{ads}) \qquad (12)$$

352. This reaction sequence is very inefficient during the day, and at higher levels of NO, because NO_3 photolyses and reacts with NO rapidly, leading to NO_x regeneration. During the night, however, conversion of NO_2 to nitrate aerosol by this route potentially occurs on the timescale of a few hours, although the efficiency can be reduced by the reaction of NO_3 with residual NO or with some organic compounds. The latter may either regenerate NO_x, or form a variety of oxidised organic nitrogen compounds or gaseous HNO_3 (for example, Jenkin and Clemitshaw, 2000).

3.3.2.2 Formation of nitrous acid (HONO)

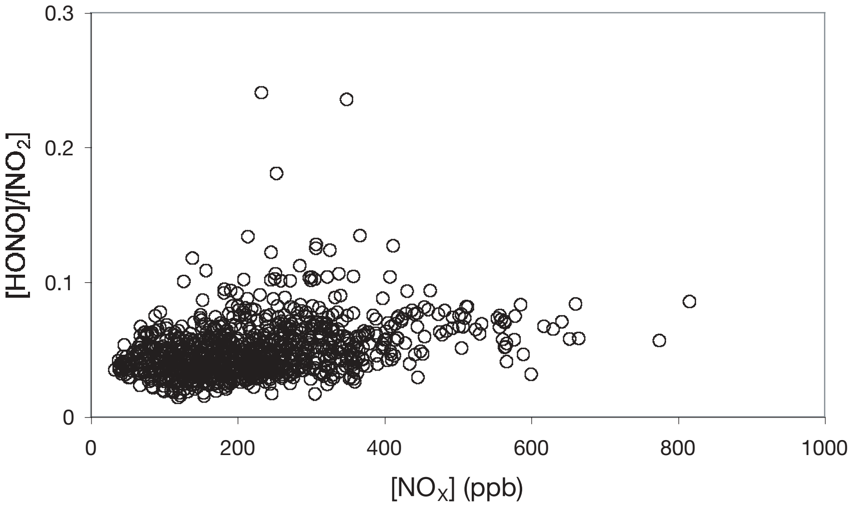
353. In addition to its direct emission from combustion sources, HONO is generated from chemical processing of NO_x During daylight, the reaction of OH with NO leads to the production of HONO:

$$OH + NO\ (+M) \rightarrow HONO\ (+M) \qquad (13)$$

354. At a typical background OH concentration of 0.04 ppt, NO is converted to HONO at about 4% hr^{-1} by this reaction. However, HONO only acts as a temporary reservoir for NO_x during daylight because of its comparatively efficient photolysis (reaction (8)), such that HONO cannot accumulate as a result of reaction (13). Observational data are consistent with the existence of additional thermal sources of HONO, which may operate throughout the diurnal cycle, leading to an accumulation of HONO during the night, followed by photolysis at sunrise (for example, Harris *et al.*, 1982; Kessler and Platt, 1984; Harrison *et al.*, 1996). The precise formation mechanism is unknown, although published information is consistent with HONO production from heterogeneous reactions of NO_2 on land or aerosol surfaces (for example, Lammel and Cape, 1996). Recent studies have demonstrated that diesel exhaust particulates are particularly reactive, and may represent an important substrate for HONO formation (Gutzwiller *et al.*, 2002).

355. Ambient $[HONO]/[NO_x]$ ratios as high as 0.04 have been observed at urban locations (for example, Lammel and Cape, 1996), with contributions to HONO potentially being made by both emissions and secondary reactions of NO_x. Reported measurements of HONO at Marylebone Rd., London (Martinez-Villa *et al.*, 2003) indicated $[HONO]/[NO_x]$ ratios of about 0.01, with the major contribution to the observed HONO being attributed to road transport emissions. As shown in Figure 3.15, $[HONO]/[NO_2]$ therefore tended to increase at higher NO_x (i.e. as $[NO_2]/[NO_x]$ decreased), but typically fell in the range 0.01-0.1 (mean ≈ 0.05). HONO at this level potentially makes a small, but significant contribution to measurements of NO_2 made by chemiluminescent analysers with thermal converters.

Figure 3.15 Observed $[HONO]/[NO_2]$ ratios as a function of NO_x at Marylebone Rd, 11-24th October 1999 (Martinez-Villa *et al.*, 2003). Each data point is based on 15 minute averaged concentrations of HONO and NO_2. Raw data kindly supplied by K. Clemitshaw, Imperial College, London.



3.3.2.3 Formation of peroxyacetyl nitrate, PAN ($CH_3C(O)O_2NO_2$)

356. The reactions of NO_2 with organic peroxy radicals (RO_2) in general lead to the production of organic peroxy nitrates (RO_2NO_2). The majority of such compounds are thermally very unstable under boundary layer conditions. However, those peroxy radicals containing a carbonyl (C=O) group adjacent to the peroxy radical centre form peroxy nitrates which possess significantly greater stability. The simplest example of this class of compound, peroxy acetyl nitrate or PAN ($CH_3C(O)O_2NO_2$), is thus generated as follows,

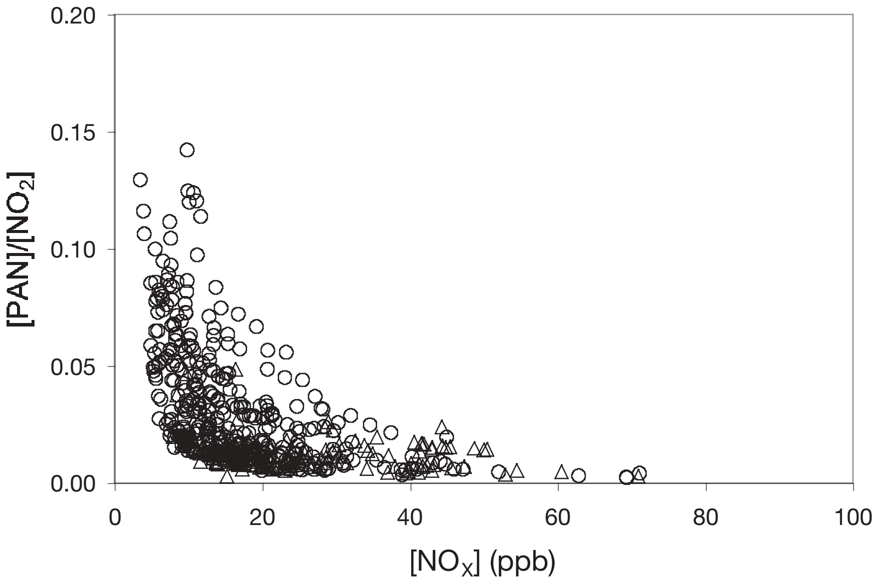
$$RC(O)O_2 + NO_2 \ (+M) \rightleftharpoons RC(O)O_2NO_2 \ (+M) \qquad (14,-14)$$

where the organic group R is CH_3. Higher homologues are also formed in the atmosphere, but are generally far less abundant than PAN (for example, Altschuller, 1993). This is mainly because the precursor peroxy radical, $CH_3C(O)O_2$, is potentially produced from the degradation of a large number of emitted VOC $\geq C_2$, whereas the abundance of potential source compounds systematically diminishes for the larger $RC(O)O_2$ radicals.

357. The ambient concentration of PAN is determined by a balance between its production by reaction (14) and loss by thermal decomposition (the reverse reaction (-14)). Its production rate is highest under conditions when efficient photochemical processing of VOC occurs, because ambient levels of free radicals, including $CH_3C(O)O_2$, are elevated under such conditions. However, the thermal decomposition rate of PAN also tends to be greater under such conditions, because the temperature is higher: the thermal decomposition lifetime of PAN is strongly dependent on temperature, varying from about 1 hour at 298 K to about 2.5 days at 273 K. As a result, historical concentration data measured at rural sites at Harwell (Oxfordshire) and Bush (Midlothian) demonstrate significant levels of PAN throughout the year, with the maximum values (on a weekly-averaged basis) typically occurring in the springtime (PORG, 1993, 1997; Cape and McFadyen, 2001).

358. In addition to the above factors, formation of PAN is also favoured at higher $[NO_2]/[NO]$ ratios which, as shown in Figure 3.14a, are observed at lower NO_x levels. This is because $CH_3C(O)O_2$ also reacts rapidly with NO in competition with reaction (14). This is clearly illustrated in Figure 3.16, which presents data measured in Ascot (Berkshire) in the summer of 1999 (Andres-Hernandez and Burrows, private communication). The observed $[PAN]/[NO_2]$ ratio thus varies from as high as 0.17 at low NO_x (i.e. high $[NO_2]/[NO]$) on a photochemical episode day, to essentially zero at the high end of the NO_x range (i.e. high $[NO]/[NO_2]$) under non-episodic conditions. Over the entire 3-week period of the measurements, the average $[PAN]/[NO_2]$ ratio was ca. 0.02, with the mean $[NO_2] \approx 16$ ppb. PAN therefore potentially makes a small, but significant contribution to measurements of NO_2 made by chemiluminescent analysers with thermal converters, at lower NO_x levels.

Figure 3.16 Observed [PAN]/[NO_2] ratios as a function of NO_x at Silwood Park, Ascot, Berkshire 24th July – 12th August 1999. Each data point is based on 30 minute averaged concentrations of PAN and NO_2. Squares are daylight data on photochemical episode days (defined by mean O_3 > 50 ppb); circles are daylight data on other days; triangles are nighttime data. Data kindly supplied by M. Andres-Hernandez and J. Burrows, IUP, Bremen.



3.3.3 Deposition of NO_2

359. The removal of NO_2 by dry deposition occurs slowly, and represents only a minor loss process. Studies of the rates of deposition onto terrestrial surfaces have generally focused on uptake by vegetation, and have demonstrated that this is dominated by stomatal absorption (for example, Hargreaves *et al.*, 1992). The reported deposition velocities (typically 0.5 – 2 mm s^{-1}) are consistent with NO_2 lifetimes of several days or greater with respect to removal by deposition.

360. As indicated above, NO_2 may also be removed by reaction on non-vegetated land surfaces, with the probable generation of HONO. Reported studies suggest that the reaction may involve adsorbed water, leading to the disproportionation of NO_2 into gaseous HONO and adsorbed HNO_3 (Lammel and Cape, 1996),

$$2\ NO_2(g) + H_2O(ads) \rightleftharpoons HONO(g) + HNO_3(ads) \qquad (15)$$

such that oxidised nitrogen is lost to the ground as HNO_3. Kitto and Harrison (1992) reported evidence for a surface source of HONO by reaction of NO_2, and Harrison *et al.* (1996) derived an effective conversion rate of (5.6 x 10^{-6} x 100/h) s^{-1} (where h is the mixing height in m) in the suburban boundary layer. This suggests a typical lifetime of NO_2 of at least several days with respect to conversion to HONO at the ground. If reaction (15) is an adequate representation of the chemistry occurring, this is accompanied by simultaneous loss of oxidised nitrogen from the air.

3.4. Factors and processes controlling the distribution of NO_2 in the ambient environment

361. The majority of the NO_x emissions by mass enter the atmosphere as NO and are converted there to form NO_2. A small fraction of the NO_x is emitted to the atmosphere as NO_2. At any point in space and time, then, the NO_2 that is present in the atmosphere may have been directly emitted, that is to say primary NO_2, and some may have been formed in the atmosphere, that is to say is secondary NO_2. More often than not it will be a mixture of both primary and secondary NO_2. It is a vital issue for policy whether the origin of the NO_2 is primary or secondary, because source-receptor relationships are dramatically different for primary and secondary pollutants. The different source-receptor relationships influence the different control policy actions that should be taken so that satisfactory NO_2 air quality can be reached efficiently and effectively in the future.

362. In this section some illustrative situations are reviewed where elevated NO_2 concentrations have been observed and describe how atmospheric processes influence the balance between the primary and secondary sources of NO_2. These situations include:

- an isolated major road in a rural environment;
- an isolated major road in an urban environment.

3.4.1. Distribution of NO_2 downwind of an isolated major road in a rural environment

363. Here air is advected from a rural environment across a major double carriageway road carrying heavy traffic that acts as a large source of NO_x, mainly as NO but with a small percentage of its emissions as NO_2. The air is initially assumed to have pollutant levels typical of a relatively unpolluted rural environment, that is to say containing ppb levels of NO and NO_2 and baseline levels of O_3. Since O_3 concentrations generally increase with height above rural surfaces, the air close to the ground may contain O_3 concentrations of typically 30 ppb, whereas aloft, O_3 concentrations may be 40-45 ppb at heights of 100 m or so. Details of an illustrative calculation are given in Box 3.1, with the results presented in Figure 3.17. The scenario represents the case of a very busy dual carriageway under low wind speed conditions.

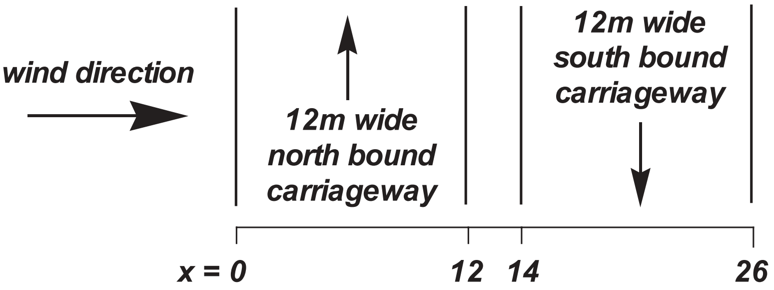
364. The air that arrives close to the ground at the upwind carriageway encounters immediately the emissions of NO. At the resultant concentrations of several hundred ppb NO, O_3 reacts quickly with a time constant of a few seconds (see section 3.3.1), which corresponds to a spatial scale of up to a few tens of metres under the conditions of this illustration. This reaction:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad (1)$$

goes to near-completion so that by the time the air close to the surface has crossed the dual carriageway NO_2 formation has largely ceased. At this point, secondary NO_2 concentrations reach a maximum which approaches initial baseline O_3 concentration in the upwind rural environment. Vertical dispersion acts to reduce secondary NO_2 concentrations, which fall off with height and distance downwind from the dual carriageway. Air that crosses the upwind carriageway at some height above the surface encounters the traffic plume slightly later than air close to the surface. The region of maximum secondary NO_2 concentrations is therefore located further downwind with increasing height above the surface.

Box 3.1: NO_x, NO_2 and O_3 pollution concentrations in the vicinity of a major road

A dual carriageway has been modelled using ADMS-Roads. The road has been modelled as two line sources, each 12m wide with a 2m wide central reservation between them, as shown in the following layout:



It has been assumed that both sides of the carriageway have equal traffic flows, with 8000 light vehicles per hour driving at 100 km hr^{-1} and 1000 heavy vehicles per hour driving at 90 km hr^{-1}. 1999 DMRB emission factors for the year 2003 have been used to calculate the emission rates on the road, as follows:

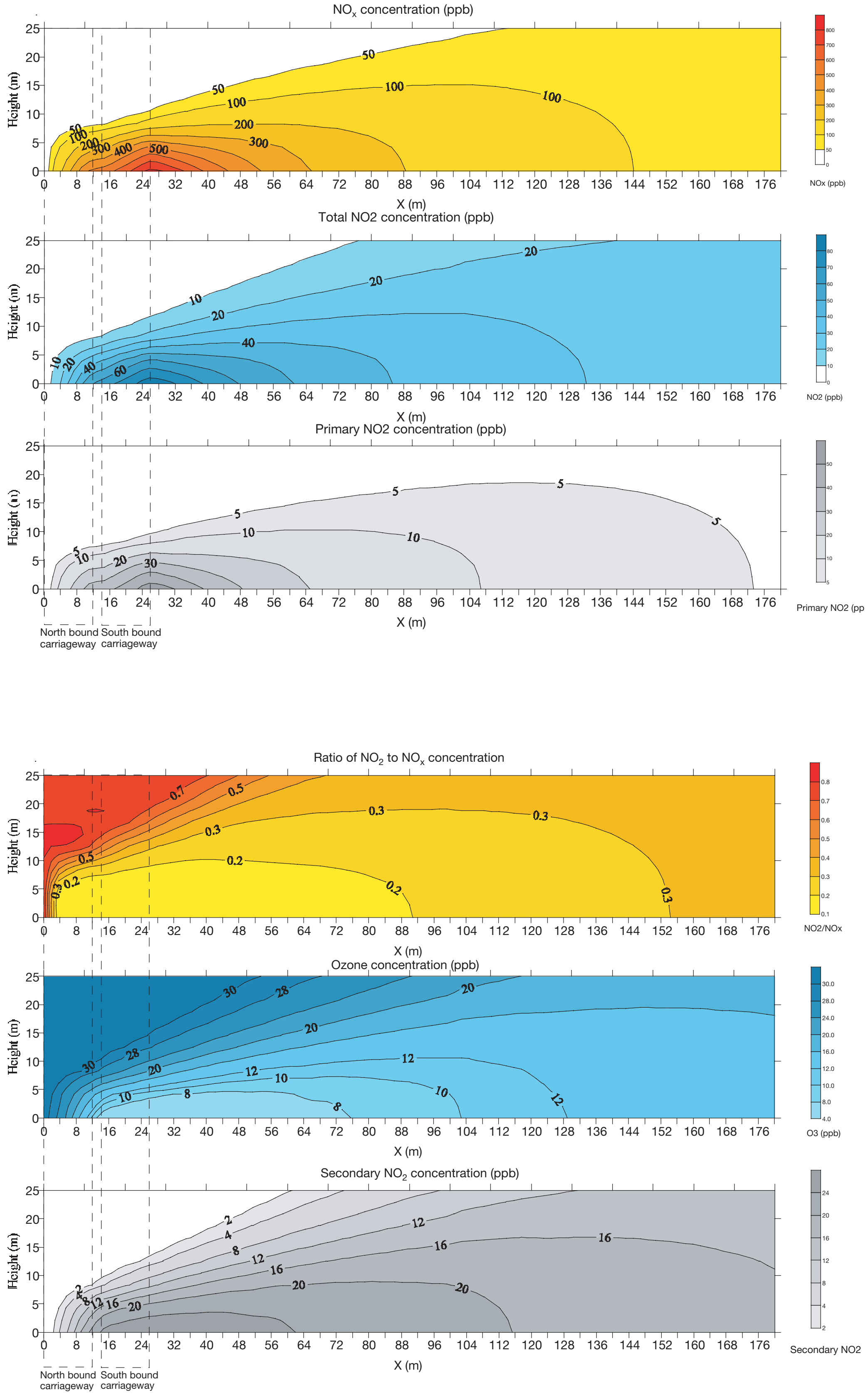
Pollutant	Emission per carriageway	Total emission
NO_x (g km^{-1} s^{-1})	4.07	8.13
NO_2 (g km^{-1} s^{-1})	0.41	0.81
VOC (g km^{-1} s^{-1})	0.51	1.02

A background O_3 concentration of 31 ppb has been applied with background NO_x and NO_2 concentrations of 5 ppb and 4 ppb respectively.

Meteorological conditions are as follows: wind speed 2 m s^{-1}, direction of 270 degrees. The modelled hour is 16:00 on May 1st, with a cloud cover of four oktas and a temperature of 15ºC. The resulting boundary layer height is 998 m and the Monin-Obukhov length is -43 m.

Concentrations of NO_x, NO_2 and O_3 have been calculated up to 180 m from the western edge of the road, and up to 25 m in height, as shown in Figure 3.17.

Figure 3.17 Variation of pollutants with distance and height from a 26 m wide road. Concentrations calculated as described in Box 3.1.



365. Secondary NO_2 concentrations do not fall off as quickly with downwind distance from a major road compared with primary pollutants. This is because NO_2 is still being formed aloft at the top of the traffic plume by reaction between O_3 entrained from the upwind rural environment with NO carried up from the surface by turbulence. This secondary NO_2 is brought down to the surface by turbulent exchange in the downwind environment. Because of the elevated height of this secondary NO_2 source, this downwards turbulent transport has a significantly longer time constant compared with the upwards turbulent transport of the secondary NO_2 formed near the surface. This elevated secondary NO_2 source controls the concentration of secondary NO_2 in the environment downwind of the road edge. However, with increasing downwind distance, secondary NO_2 concentrations eventually begin to fall as vertical exchange depletes the NO concentrations throughout the traffic plume, reducing secondary NO_2 formation.

366. If all the NO_x from traffic was emitted as NO, then the sequence of processes outlined above would entirely control the distribution of NO_2 in the environment downwind of the major road and broadly speaking NO_2 concentrations would never exceed nor even approach the O_3 concentrations in the rural environment upwind of the major road. Direct emissions of NO_2 occur with a small percentage by mass and these generate a distribution of primary NO_2 that is superimposed upon the distribution of secondary NO_2 described above.

367. As air reaches the upwind carriageway, close to the ground, it immediately encounters the direct emissions of NO_2 from the traffic. Primary NO_2 concentrations steadily rise across the upwind carriageway as direct emission competes against vehicle-generated turbulence and vertical dispersion. Somewhere over the downwind carriageway, vertical dispersion starts to dominate over direct emission and primary NO_2 concentrations begin to level off and subsequently start to decrease. At the edge of the downwind carriageway, direct emission of NO_2 ceases and primary NO_2 concentrations fall off steadily with increasing downwind distance. The higher the windspeed, the sooner the primary NO_2 concentration maximum is reached and the lower in magnitude it is. Since traffic emissions have little buoyancy, the concentration contours of this primary NO_2 have their maximum concentrations close to the ground and concentrations fall off rapidly with both height above the surface and downwind distance.

368. If there was no O_3 in the upwind environment, then this direct NO_2 emission together with vertical dispersion and vehicle-generated turbulence would control the distribution of NO_2 downwind of the major road. Inevitably, air parcels arriving at the upwind carriageway of the major road contain O_3 at concentrations close to baseline levels and so account must always be taken of secondary NO_2 formed through the reaction of primary NO with O_3. The distribution of NO_2 downwind of a major road is therefore the sum of the primary and secondary NO_2 distributions and a complex three-dimensional spatial pattern results. The interaction between the processes is illustrated for a two-dimensional section in Figure 3.17.

3.4.2. Distribution of NO_2 downwind of an isolated major road in an urban environment

369. Here air is advected across an urban environment and over a major road carrying heavy traffic that acts as a large source of NO mainly but with a small percentage of its NO_x emissions as NO_2. If the oxidising capacity of the atmosphere has been largely exhausted before the air reaches the upwind carriageway then the air will contain a mixture of NO and NO_2, but O_3 levels may well be a few ppb or less. Under these conditions, little additional secondary NO_2 will be formed close to the road surface and the distribution of NO_2 downwind of the major road will be determined by the primary emissions of NO_2 and the NO_2 distribution upwind of the major road in question.

370. As air reaches the upwind carriageway, NO_2 concentrations will begin to rise as direct emission competes against vertical dispersion and vehicle-generated turbulence. The distribution of primary NO_2 will follow closely that given above for the rural major road under the conditions where no O_3 was present. In the urban case, the distribution of NO_2 downwind of the major road would follow that of any other primary motor vehicle pollutant. NO_2 concentrations would peak close to the kerb of the downwind carriageway and concentrations would fall off rapidly with height and inversely with increasing downwind distance.

371. Secondary NO_2 still continues to be formed aloft in urban areas long after O_3 is depleted close to the surface. At some height, say tens to hundreds of metres above the surface, O_3 will still be present at concentration levels approaching those in the upwind rural environment. NO transported up to this level by turbulent exchange will react with O_3 in a thin layer just below the bottom of the O_3-rich layer. The secondary NO_2 formed here can then be transported down towards the surface by turbulent exchange and mix with the primary NO_2 found close to the surface.

372. During a photochemical O_3 episode, there may be several times more O_3 in the upwind rural environment compared with baseline conditions, for a few hours during mid-afternoon on each of the episode days. This increased O_3 will lead to increased O_3 concentrations within urban areas as NO is converted increasingly to NO_2. Under these conditions, the case of the major road within an urban environment may be changed to look more like the rural case with the overlapping distributions of both primary and secondary NO_2 downwind of the major road.

373. Inevitably in urban areas, the distribution of NO_2 is controlled either by the availability of O_3 from the upwind rural environment or by the availability of NO_x emissions and their split between NO and NO_2. As emission density increases, secondary formation of NO_2 becomes of diminished significance and NO_2 concentrations become increasingly dominated by direct emissions of NO_2. So, for example, the traffic excess of NO_2 found by subtracting the simultaneous NO_2 concentrations observed at the Bloomsbury urban background site from the roadside Marylebone Road site, can be accounted for largely by the direct emissions of NO_2 without any requirement for additional urban conversion of NO to NO_2.

Chapter 4

Measurement methods and UK monitoring networks for NO_2

Key points

- Several techniques are available to measure NO_2 concentrations in the atmosphere.
- The chemiluminescent analyser is widely used for continuous monitoring of NO_2 concentrations in the UK. It is the reference method specified in the EU First Air Quality Daughter Directive and is based on the chemiluminescent reaction between NO and O_3.
- Other methods of measurement, such as diffusion tube samplers, electrochemical sensors, thick film sensors and open path optical methods are also available, and can provide useful data in many circumstances and for different applications. However, the data from these other measurement techniques may not be directly comparable with the chemiluminescent reference method for a number of reasons.
- Monitoring of NO_2 concentrations using the chemiluminescent analyser is carried out on a national scale, on behalf of Defra and the Devolved Administrations, and at a regional or sector-specific scale, on behalf of local authorities, the Highways Agency, the power generation industries, and other organisations. These networks are subject to rigorous quality assurance and quality control procedures, which ensure the reliability of the data.
- Data from 212 chemiluminescent monitoring sites, including kerbside, roadside, urban background, industrial and rural locations, have been considered in this report. A number of these sites have co-located O_3 analysers.
- A CEN standard for making NO_2 measurements using chemiluminescent analysers is being drafted. This will provide performance specifications for instruments, and operational procedures, to ensure that data meet the uncertainty requirements of the EU First Daughter Directive.

4.1 Introduction

374. There are a variety of monitoring methods available for the measurement of ambient NO_2 concentrations. These include:

- Chemiluminescent analysers
- Diffusive samplers
- Electrochemical sensors
- Thick film sensors
- Differential optical absorption spectroscopy (DOAS).

The various advantages and disadvantages of each of these methods are summarised in Table 4.1.

375. Whilst a number of wet chemical methods have also been developed, these are no longer widely used, and have been almost entirely replaced by the techniques described above. Fourier-transform infrared (FTIR) samplers are also available for NO_2 measurements, although their use has, to date, largely been restricted to the laboratory and to emissions testing.

Table 4.1 Summary of advantages and disadvantages of NO_2 measurement methods.

Technique	Advantages	Disadvantages
Chemiluminescence[a]	The reference method specified in the EU First Daughter Directive. Lower detection limit of ~ 1 μg m^{-3}. Provides real-time data with short time resolution (<1hr) that can be used for public information.	Relatively high capital cost. High operating costs.
Diffusion tubes	Low capital and operating costs. Possible to carry out surveys over wide geographical areas to provide information of the spatial distribution of NO_2 concentrations. Require no power supply, and minimal training of site staff. Site calibrations are not required. Lower detection limit of ~2-3 μg m^{-3} for a 4 week exposure period.	Only provide concentrations averaged over the exposure period (typically 4 weeks). Accuracy of the method, and bias relative to the reference sampler, is dependent upon the method of tube preparation and the laboratory completing the analysis. Results need to be ‘bias-corrected’ before comparison with limit values and objectives.
Electrochemical Sensors[a]	Portable samplers that can be easily deployed in the field.	Lower detection limit of some samplers (~200 μg m^{-3}) makes then unsuitable for ambient monitoring.
Thick-film sensors[a]	Portable samplers that can be easily deployed in the field. Potential to measure a number of pollutants simultaneously. Provides real-time data with short time resolution (<1 hr) that can be used for public information. Lower detection limit of ~4 μg m^3.	At this time, the sensor for NO_2 has not been commercially released. Only measurements of NO_x may currently be carried out.
DOAS[a]	Concentration integrated over the length of the light path. Gives an ‘average’ concentration that can be useful to assess public exposure. Potential to measure a number of pollutants simultaneously. No inlet manifold required. Provides real-time data with short time resolution (<1 hr) that can be used for public information. Lower detection limit of ~ 1 μg m^{-3}.	The integrated measurement cannot be directly compared with the EU limit values or the air quality objectives (which are set for a single point measurement). Relatively high capital cost. Unfavourable weather conditions, such as fog or snow, can affect the instrument performance.

[a] Methods require regular calibration

4.2 Chemiluminescent analysers

376. The chemiluminescent analyser is widely used for continuous monitoring of NO_2 concentrations in the UK, and throughout the World. It is the reference method specified in the EU First Air Quality Daughter Directive. A typical specification is provided below.

- Lower Detection Limit: < 2 mg m^{-3}
- Precision: <1% (Full Scale)
- Linearity error: <2%
- 95% response time: <180 seconds (a typical specification is 20 seconds lag + 60 seconds rise to full scale)

377. Chemiluminescence describes the process of fluorescence resulting from a chemical reaction. The chemiluminescent sampler for the measurement of NO_2 relies on the reaction of NO with O_3 to produce an 'excited' form of NO_2. As the excited molecule returns to its ground state, fluorescent radiation is emitted, the intensity of which is proportional to the concentration of NO. This is expressed by the following reactions:

$$NO + O_3 \rightarrow NO_2^* + O_2$$

$$NO_2^* \rightarrow NO_2 + hv$$

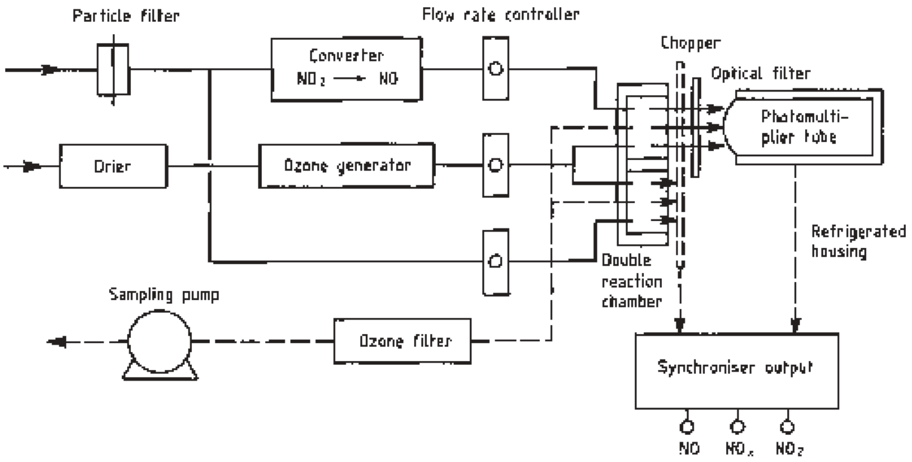
Where *hv* represents the emitted radiation.

378. It is important to note that chemiluminescent analysers do not directly measure concentrations of NO_2. This is achieved by reducing the NO_2 component of the ambient air sample to NO (in a NO_2-to-NO converter[1]), which gives the concentration of total NO_x and then subtracting NO:

$$NO_2 = NO_x - NO$$

It is therefore necessary for the analyser to make two independent measurements, of NO and NO_x ($NO+NO_2$). Within some analyser types, this is achieved by having a dual-chamber configuration that measures NO_x and NO simultaneously in individual reaction chambers. A typical dual-chamber configuration is shown in Figure 4.1. Some dual-chamber instruments have dual detectors and some have only a single detector.

Figure 4.1 Schematic diagram of a dual-chamber NO_x analyser.



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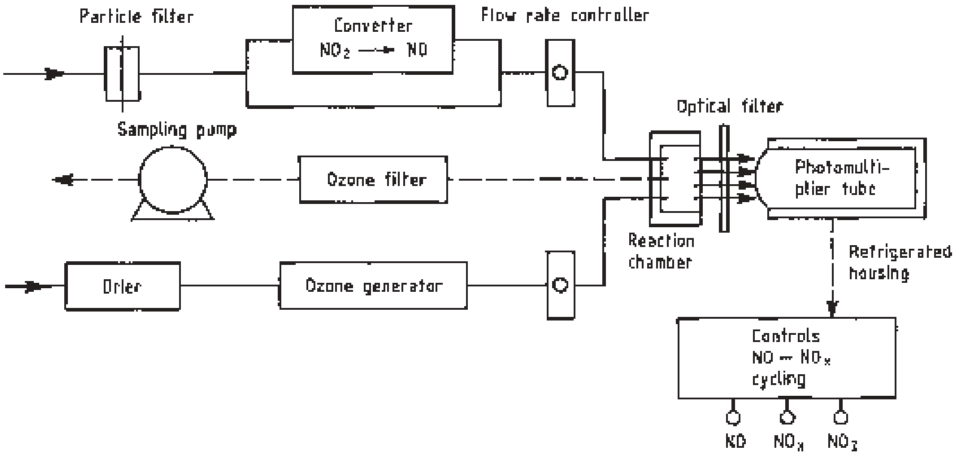
[1] Typically, these converters are comprised of a high-temperature (≈350ºC) molybdenum catalyst cell, although low-temperature converters (≈200ºC) are now also available.

379. In recent years, many manufacturers have moved to the production of single-chamber instruments, mainly to save on the costs of plumbing and electronics. In this case, the measurements of NO_x and NO_2 must be made sequentially, by switching the air sample either directly to the reaction chamber or via the NO_2-to-NO converter. A typical single-chamber configuration is shown in Figure 4.2.

Figure 4.2 Schematic diagram of a single-chamber NO_x analyser.



4.2.1 Single- vs dual-chamber analysers

380. In the majority of cases, single-chamber instruments perform without difficulty. To overcome problems that may occur at sites where the NO_x and NO concentrations fluctuate rapidly, for example at kerbside or roadside sites, manufacturers have devised a number of solutions. These mostly involve increased cycle frequency, or the introduction of delay loops (see below). Experience to date within the operation of the UK national networks has not indicated concerns regarding the performance of single-chamber instruments at roadside and kerbside sites. However, at the London Marylebone Road kerbside, which represents an extreme monitoring environment, a dual-chamber sampler is installed. Care in the installation and configuration of single-chamber instruments at roadside and kerbside sites is recognised as an important aspect of site commissioning.

381. There is a paucity of peer-reviewed studies comparing the performance of single- and dual-chamber samplers and reliance has largely been based on manufacturers' reports. There is a need for more independent research in this area. The new CEN standard for chemiluminescent analysers (see Section 4.10) will introduce a specific test to evaluate the ability of analysers to respond correctly under conditions of rapidly changing concentrations. This will provide a valuable benchmark against which to assess all analysers in future.

4.2.2 Increased cycle frequency

382. Single-chamber chemiluminescence analysers are typically set to operate with 15-second cycle times i.e. the switching between the NO_x and NO measurement takes place every 15 seconds. In environments where the NO_x concentration is expected to fluctuate on a shorter timescale than this, then the cycle time can be reduced to 3 or 4 seconds. An investigation into this approach was performed at a kerbside location in Oxford for a 3-month period in 1996, when a single-chamber and a dual-chamber instrument were operated in parallel. The single-chamber instrument was modified to allow operation at high switching speeds by reducing the cycle time to 3 seconds per mode, increasing the response time of the input amplifier, adjusting the final signal processing, and fitting a reservoir volume to the sample inlet to reduce gas transients to within the overall cycle time of 6 seconds. The single-chamber instrument showed no negative NO_2 15-minute averages, which would appear if rapid fluctuations were a significant problem, and good long-term agreement with the dual-chamber instrument.

383. More recently, single-chamber analysers have been developed with low-temperature NO-to-NO_2 converters and fast-response photodiode detectors. The system allows switching between the NO_x and NO channels at 0.5 second intervals, and provides a virtually concurrent analysis.

4.2.3 Delay loops

384. An alternative approach to increased cycle frequency has been the introduction of a delay loop into the sample stream. This effectively allows the instrument to sample NO_x and NO simultaneously, but to measure them sequentially. Both the NO_x and NO sample streams are collected at the same time. Whilst NO_x is measured, the NO sample stream is sent round a precise volume loop, delaying its arrival at the reaction chamber until the NO_x cycle is completed. Typical cycle times of 4 seconds are used.

4.2.4 Interferences

385. An assessment of overall uncertainty budgets, including interferences, has been reported (Gerboles *et al.,* 2002). Two forms of interference may occur in the chemiluminescent sampler; a reduction in the chemiluminescence intensity by quenching in the reaction chamber and, bias due to conversion of various N-species to NO taking place in the NO_2-to-NO converter. Whilst a variety of compounds (including H_2O, O_2, CO_2, CO, Ar and H_2) may give rise to quenching effects, Gerboles *et al* concluded that only water vapour is likely to give rise to significant effects. The installation of permeation driers at the sample inlet should prevent such problems occurring and are a common feature of most modern samplers.

386. The conversion of a number of additional oxidised nitrogen species to NO has been reported to occur with high efficiency, under the conditions in the NO_2-to-NO converter (for example, Winer *et al*., 1974; Grosjean and Harrison, 1985). These species include HNO_3, HONO, peroxyacetyl nitrate (PAN) and alkyl nitrates, which therefore potentially make some contribution to the apparent NO_2 signal. As described further in Section 3.3.2, only PAN and HONO are believed to contribute significant interference at ambient concentrations observed in the UK. For high temperature converters, these interferences are likely to account for ≤ 2% and ≤ 5% of the NO_2 signal, respectively, under typical urban background conditions, although larger contributions potentially occur under certain conditions.

4.3 Diffusive samplers

387. Passive diffusive samplers for NO_2 are simple and inexpensive devices that have been used over many years for the measurement of ambient NO_2 concentrations. The samplers widely used in the UK are based on the design introduced by Palmes (Palmes *et al.,* 1976) and are comprised of an acrylic tube that can be sealed at both ends. One end of the tube contains stainless steel grids coated with triethanolamine (TEA) that adsorbs NO_2 to produce a nitrite salt that is subsequently determined by colorimetry (analysis by ion chromatography has also been used (Gair *et al.,* 1991) to improve the detection limit for monitoring in remote rural areas). Exposure commences when the inlet cap is removed, which sets up a concentration gradient within the tube, such that molecular diffusion occurs towards the TEA-coated grid. The rate of diffusion occurs according to Fick's Law:

$$F = -D\frac{dC}{dZ}$$

Where F is the molar flux (mol cm^{-2} s^{-1}), D is the diffusion coefficient (cm^2 s^{-1}), C is the concentration (mol cm^{-3}) and Z is the diffusion path coordinate (cm).

388. After the period of exposure (t), the total quantity of gas transferred along the tube (Q) is determined by chemical analysis. Since the rate of gas transfer is governed by the tube geometry (length (l) and internal area (Πr^2)), and the gas concentration at the closed, absorber, end of the tube is zero, the average gas concentration, C, at the open end of the tube during the exposure can be calculated from:

$$C = \frac{Ql}{D\pi r^2 t}$$

389. Whilst Palmes tubes are used almost exclusively in the UK, other passive diffusion samplers have been developed in other countries. These include the Passam sampler (Switzerland), Analyst (Italy), IVL (Sweden), and Radiello (Italy).

390. It is important to note that there are a number of different tube preparation methods in use, which may have a significant effect on the performance of the method. The difference principally relates to the way in which the metal grids are coated with the TEA solution. The methods currently in common use are based on 50% TEA in acetone, 50% TEA in water, and 20% TEA in water, with a few laboratories using 10% TEA in water.

4.3.1 Performance of diffusion tubes

391. A number of studies have examined the performance of diffusion tubes, since their first application to ambient sampling in the early 1980s. Most have involved exposing tubes alongside chemiluminescence monitors. Several factors, listed below, have been identified that appear to affect tube performance, although the findings are often confusing and contradictory.

- the laboratory preparing and analysing the tubes
- the exposure interval – weekly, fortnightly or monthly
- the time of year
- the exposure setting – sheltered or exposed
- the exposure location – roadside or background
- the tube preparation method
- the exposure concentration and NO_2/NO_x ratio.

392. A number of mechanisms have been identified to explain some of the observations. This section identifies those factors that clearly influence tube performance. It is based on a more detailed, but not exhaustive, review presented in Appendix 2. It is important to note that the majority of NO_2 data used in compiling this report are derived from continuous analysers, rather than diffusion tubes, although the latter remain important for general assessment of NO_2 air quality.

393. It is clear that attempts to identify and/or quantify the influence of a given factor may be confounded by the influence of other factors. For example, the statement commonly made that diffusion tubes tend to over-estimate NO_2 concentrations, for example, Heal *et al.* (1999), Heal *et al.* (2000), Kirby *et al.* (2001) and Bush *et al.* (2001), applies to specific circumstances. In practice there are circumstances when diffusion tubes can systematically over-estimate or under-estimate concentrations.

394. The key factor influencing the performance of diffusion tubes is the laboratory carrying out the preparation and analysis. For instance, in a recent comparison of 7 laboratories, using one tube preparation method, 50% TEA in acetone, with tubes exposed for a month at a time, the annual mean bias (as quantified against collocated chemiluminescence analysers) ranged from +20% to -26% from one laboratory to another. The reason for the differences between the laboratories is currently not understood. Variability in performance may also be related to the tube preparation method, in particular the method of preparation of the adsorbent coating on the grids. Four TEA/solvent preparations are currently used in the UK: 50% TEA in acetone, 50% TEA in water, 20% TEA in water, and 10% TEA in water. Preparation methods also differ in whether grids are coated in advance of tube assembly by dipping into TEA/solvent mixtures, or whether an aliquot of TEA/solvent is pipetted onto grids after the tubes have been assembled. Various studies have examined whether preparation method affects performance but, as yet, no consistent pattern has emerged.

395. Diffusion tube performance is clearly influenced by the exposure interval, which ranges from 1 week to 1 month, with tubes exposed for longer intervals producing lower concentrations. Although no convincing mechanism has been put forward to account for lower measured concentrations, a light-dependent loss of nitrite from the adsorbent, or a temperature dependency in the diffusion coefficient that is not allowed for, have been postulated. Time of year also influences performance, with TEA-in-water tubes showing higher concentrations in the August to November period. This seasonal pattern is not seen in results for tubes prepared with TEA-in-acetone. There is currently no explanation for this pattern.

396. There is some evidence that the exposure setting may influence concentrations, with greater uptake in more exposed locations. The explanatory mechanism is believed to be a shortening of the effective diffusion length as a result of wind induced turbulence at the tube entrance.

397. There is some evidence that tube performance is affected by ambient concentrations of NO_2, NO_x and O_3 at the exposure site. The postulated mechanism is that a greater uptake of NO_2 arises from production of additional NO_2 along the diffusion path within the tube because of reaction between co-diffusing NO and O_3 and a disruption of the photochemical equilibrium between NO_2 – NO – O_3 (see Section 3.2.1). (The photolytic component is switched off within the tube). Evidence for any systematic difference in diffusion tube performance between roadside and background location is equivocal and will be related to the above mechanism. Clearly, any relationship between diffusion tube performance and ambient concentrations is complex and may be masked by effect on performance arising from preparation method, exposure interval, etc.

398. Defra has recently initiated an annual intercomparison exercise for diffusion tubes. This will involve exposure of triplicate tubes, from the full range of laboratories supplying and analysing tubes in the UK, alongside a chemiluminescence monitor at a background location at Leigh, in Greater Manchester. Unlike previous such studies, this scheme will run using monthly exposures for a full year (although some smaller laboratories have opted to participate on a quarterly basis). It will provide useful additional information on tube performance.

399. The precision of diffusion tube samplers, as determined by the standard deviation of results from groups of 6 tubes exposed simultaneously as part of the UK NO_2 Diffusion Tube Network Annual Intercomparison (AEA Technology, 2002), has been found to be below 6 $\mu g\ m^{-3}$ in most cases

4.3.2 Recommendations for use of diffusion tubes

400. The analysis of factors influencing diffusion tube performance identifies a range of factors that seem to be related to tube performance: the laboratory preparing and analysing the tubes; the exposure interval; the time of year; the relationship between NO, NO_2 and O_3 at the exposure site; possibly a difference between exposed and more sheltered settings; and the method of tube preparation. Because there are so many factors affecting tube performance, it is crucial for any particular survey that:

- the variables that can be controlled are held constant, especially laboratory, tube preparation method and exposure interval;
- the performance of the tubes is determined specifically for the fixed set of variables being used, and the results adjusted for any bias identified.

401. Diffusion tube measurements would benefit from specific, validated procedures for their implementation. The preferred route would be for these to emerge through the work of the European standardisation body, CEN, mentioned further in Sections 4.8.3 and 4.10. If this does not happen in the near future, the UK should consider adopting a national standardised method.

402. As the principal value of diffusion tubes is to measure annual mean concentrations, the assessment of tube performance should be determined over a full year, with a minimum of 9 months collocation data being necessary to give a reasonable estimate of annual performance (Laxen & Wilson, 2002). It seems reasonable to conclude, on the evidence available, that when used in an appropriate way, as described above, diffusion tubes should be capable of measuring the annual mean concentration to within ±20% (95% confidence interval).

4.4 Electrochemical cells

403. There are a variety of portable samplers available, based on the use of electrochemical cells. In its simplest form, the principal of operation depends on the electrochemical reduction of NO_2 between two electrodes immersed in an electrolyte reservoir. NO_2 present in the sample air passes through a capillary diffusion barrier into the reaction cell, where it is reduced at the electrode. The migration of electrons produced by the reaction results in a net current flow, which is proportional to the NO_2 concentration.

404. Whilst these types of samplers continue to be widely used for the assessment of occupational exposure, their application to ambient monitoring is limited due to the high limit of detection of the electrochemical sensor. As an example, the stated lower limit of detection for NO_2 quoted for one commercially available electrochemical analyser is 200 $\mu g\ m^{-3}$.

405. There is evidence that some or all varieties of electrochemical cell intended to measure NO_2 actually measure a combination of NO_2 and O_3. This may be useful at certain times in some environments, for example at city centre roadside locations where NO_2 concentrations are often higher than O_3. Elsewhere, interpretation of the data can be difficult.

406. A growing application of analysers based on electrochemical cells is as part of urban traffic control systems or urban traffic management and control systems. Because of the problems with NO_2 measurement by electrochemical cells, the air quality data used in these situations tends to focus on CO, not NO_2, as an indicator of levels of traffic pollution, since CO is easily measured by electrochemical cells (Ropkins *et al.,* 2000).

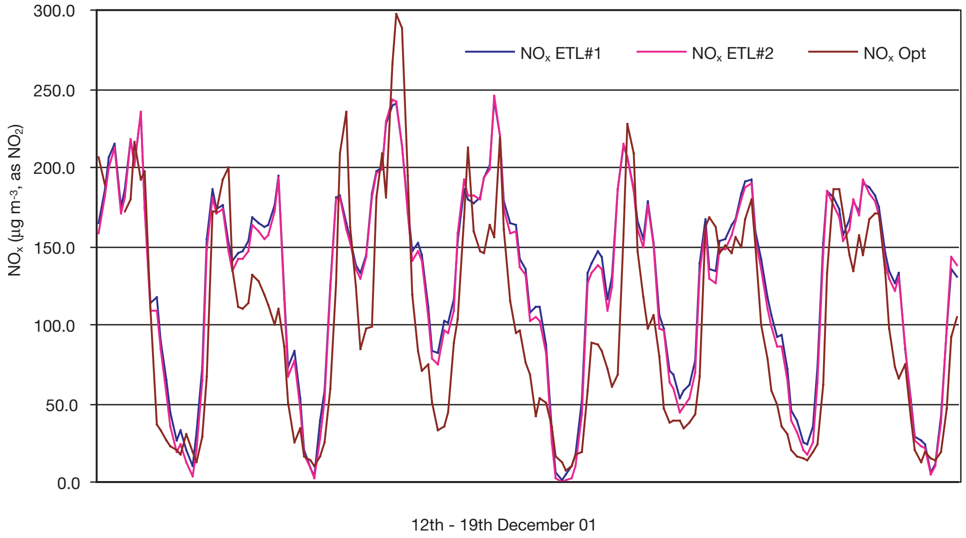
4.5 Thick film sensors

407. The development of solid-state thick film gas sensors based on semi-conducting oxides has provided a new range of samplers for environmental monitoring (Carotta *et al.,* 2000). These sensors are constructed from nano-structured semi-conducting metal oxides, which are maintained at an operating temperature of between 250 and 400°C. As current is passed through the sensor, an electrical response is produced in proportion to a specific gas concentration.

408. Currently, these thick film sensors have been produced for a range of common pollutants, including CO, O_3, NO and NO_2, and commercially available instruments are becoming available. Whilst the NO_2 sensor is still undergoing development, a thick film NO_x sensor has been trialed in the UK and shown to demonstrate reasonable comparison with a co-located chemiluminescent analyser (see Figure 4.3).

409. Whilst is it not envisaged that these samplers will replace the chemiluminescent method, they offer a number of potential benefits, such as use in screening surveys in urban locations, the identification of pollution ‘hot spots’; and mobile monitoring in buses and cars etc.

Figure 4.3 Comparison of co-located thick film sensors and chemiluminescence analyser at a roadside site (12-19 December 2001). NO_xETL#1 and NO_xETL#2 are thick film sensors and NO_xOpt is a chemiluminescent analyser.



4.6 Differential optical absorption spectroscopy (DOAS)

410. DOAS is a technique that can be used for the measurement of a wide range of air pollution species. In the usual configuration, light (visible, non-laser light) from a light source passes through a fixed path in the atmosphere, typically 100 – 1000 m in length. At the end of this path, the light received is analysed in an opto-analyser system. The amount of a specific gaseous substance in the atmosphere follows the principle of the Beer Lambert Law:

$$I_1 = I_0 e^{-\alpha LC}$$

where: I_1 is the light intensity after absorption, I_0 is the light intensity before absorption, α is the absorption co-efficient at the selected wavelength, L is the optical path length and C is the concentration of the gaseous substance.

411. In commercial analysers, sophisticated signal processing is undertaken to account for interfering species and variability in atmospheric optical transmission conditions.

412. Many species can be measured by the DOAS technique, but the most common configuration for ambient air monitoring is to measure NO_2, SO_2, O_3 and benzene. NO can be measured, but requires a different lamp type. At least one commercial DOAS instrument has EPA approval for the measurement of NO_2 (and SO_2 and O_3) provided certain operational and calibration requirements are followed.

413. Many intercomparison studies have demonstrated that DOAS can provide comparable NO_2 data to chemiluminescent point analysers, within the constraint that the DOAS analyser is averaging concentration measurements along the path length of measurement, rather than measuring at a single point.

414. As with all air pollution analysers DOAS instruments require regular calibration and other QA/QC checks to ensure reliable, high quality data.

4.7 The UK monitoring networks

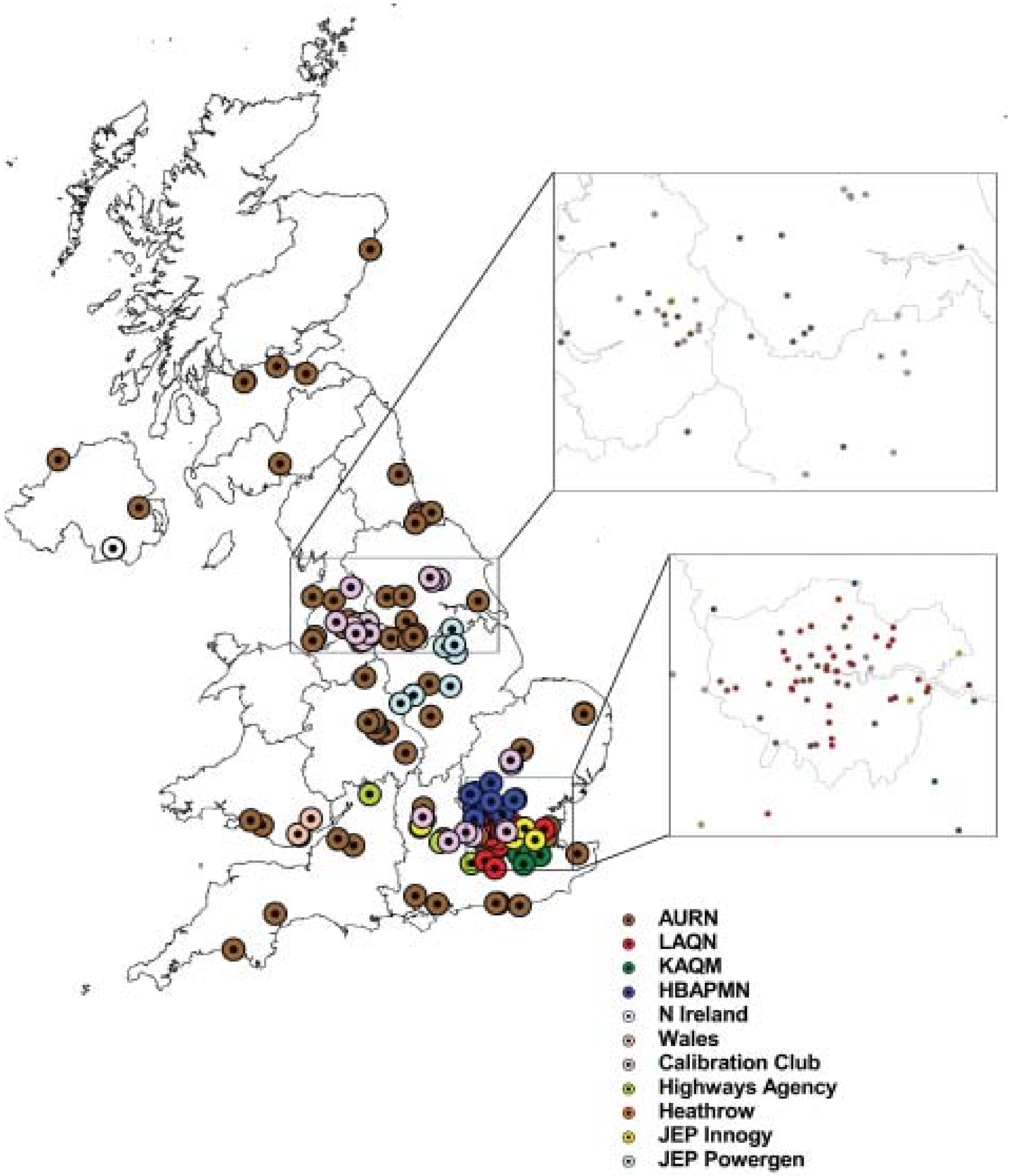
415. Monitoring of NO_2 concentrations is carried out at both a national and regional scale using both chemiluminescence and diffusion tube samplers. These networks are described in the following sections.

4.7.1 Automatic monitoring networks

416. Automatic networks are operated at both the national level, on behalf of Defra and the Devolved Administrations, and on a regional or sector-specific level, on behalf of local authorities, the Highways Agency, the power generation industries and other organisations.

417. The locations of all sites which have provided NO_2 data for this study, for 2001, are shown in Figure 4.4.

Figure 4.4 Location of NO_2 monitoring sites (2001) used in this study.



418. The structures of the various monitoring networks are summarised in Table 4.2.

Table 4.2 Summary of automatic NO$_2$ monitoring networks

[Note – this table summarises information for the NO$_2$ monitoring networks used in this report – it is not a complete list of all networks in the UK]

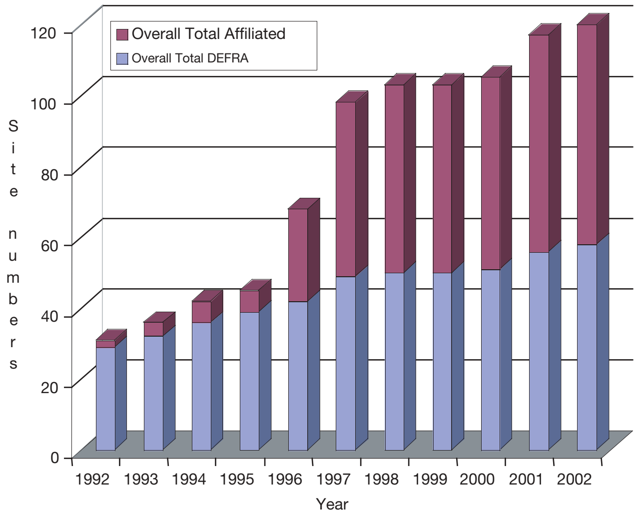
Network responsibility	Management unit	QA/QC responsibility	Number of NO$_x$ monitoring locations
UK Networks			
● AURN[1]	Casella Stanger	netcen	85
● LAQN[2]	Kings College ERG	netcen	15
Regional Networks			
● LAQN[2]	Kings College ERG	NPL/ERG	54
● KAQM[3]	Kings College ERG	NPL/ERG	15
● HBAPMN[4]	Kings College ERG	NPL/ERG	13
● N. Ireland	netcen (from 01/03/02)	netcen (from 01/03/02)	2
● Wales	LA or Powergen[5]	LA or Powergen[5]	3
Other Networks			
● Calibration Club	netcen	netcen	23
● Highways Agency	TRL	TRL	5
● Heathrow Airport[6]	netcen	netcen	1
● JEP/Innogy	Casella Stanger	Casella Stanger	7
	PowerTechnology		8
● JEP/Powergen[5]		PowerTechnology	

1 Automatic Urban and Rural Network
2 London Air Quality Network. A number of sites in this Network have been affiliated into the national networks
3 Kent Air Quality Network
4 Hertfordshire and Bedfordshire Air Pollution Monitoring Network
5 The Powergen, Sealand, Deeside site is also part of the Wales network
6 The Heathrow Airport site is also part of the LAQN

4.7.1.1 UK networks

419. The UK networks are comprised of monitoring sites within both the Automatic Urban and Rural Network (AURN) and the London Air Quality Network (LAQN). The AURN contains a number of monitoring sites that are directly funded by Defra and the Devolved Administrations, together with additional sites that are owned and operated by other organisations (mainly local authorities) and which have been affiliated into the network. The LAQN consists solely of affiliated local authority monitoring sites. The national networks have expanded significantly over the past 8 years, most recently in order to meet the requirements of the First Daughter Directive (see Figure 4.5). Thus, whilst some sites have been operating for 10 years or more, and can provide invaluable information on long-term trends over this period, data are limited to a considerably shorter period at the majority of monitoring sites.

Figure 4.5 Number of monitoring sites in the UK National monitoring network.



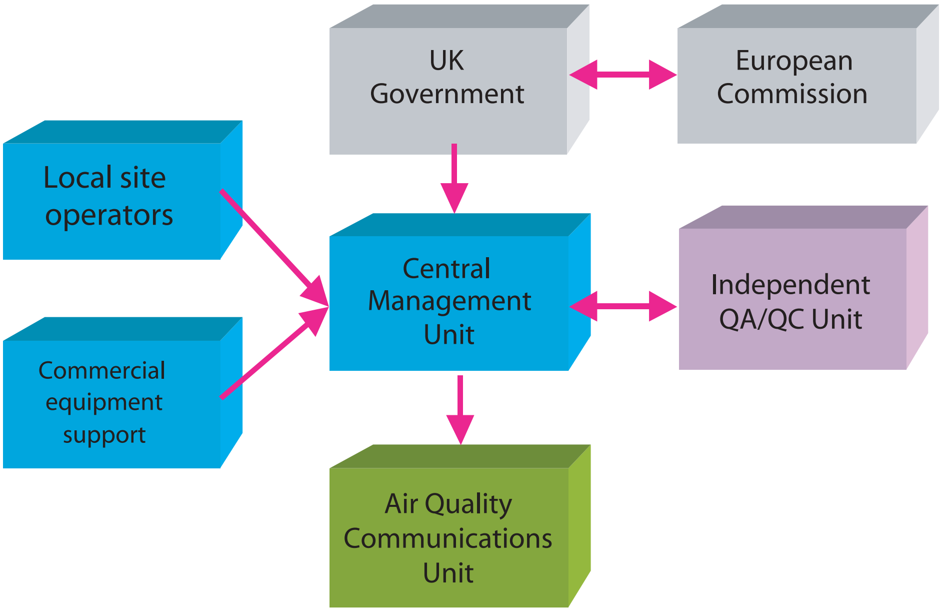
420. Monitoring stations are classified according to their location; a summary of the NO_x monitoring sites is provided in Table 4.3. Details of the site type definitions used in the UK are provided in Appendix 3. Further details on the national monitoring network sites, and measurement data, can be found at www.airquality.co.uk.

Table 4.3 Summary of NO_x monitoring stations in the UK national networks (number of sites with co-located O_3 analysers shown in parentheses).

Site Type	AURN	LAQN
Kerbside	1 (1)	2 (1)
Roadside	15 (2)	5 (0)
Urban Background	29 (18)	6 (6)
Urban Centre	23 (23)	—
Suburban	5 (4)	—
Industrial	5 (2)	—
Rural	7 (7)	—
TOTAL	**85 (56)**	**13 (7)**

421. The basic operating structure of the AURN and LAQN sites in the national networks is identical, and is represented in Figure 4.6. Data are collected from the network sites on an hourly basis and, after initial validation, are disseminated to public and the media via the Air Quality Communications Unit.

Figure 4.6 Outline structure of the National Air Quality Monitoring Networks.



4.7.1.2 Regional networks

422. Regional networks are operated or co-ordinated by Kings' College ERG, DoE Northern Ireland and the Welsh Assembly Government, on behalf of local authorities. The operation of the ERG networks mirrors that of the national networks, with routine data polling and validation, and subsequent QA/QC to provide ratified datasets. A summary of the site types within these networks is provided in Table 4.4.

Table 4.4 Summary of NO_x monitoring stations in other UK networks.

Site Type	LAQN[a]	KAQN	HABAPMN	N. Ireland	Wales
Kerbside	1	–	–	–	–
Roadside	32	7	4	1	1
Urban Background	14	4	9	1	1
Urban Centre	–	–	–	–	–
Suburban	7	1	–	–	–
Industrial	–	1	–	–	1[b]
Rural	–	2	–	–	–
TOTAL	54	15	13	2	3

a These sites not part of the UK network (Table 4.3)
b Also part of the JEP Powergen network (Table 4.5)

4.7.1.3 Other networks

423. Some Local Authority monitoring stations, throughout the UK, are part of the Calibration Club network within which data handling and data QA/QC are undertaken centrally by netcen, on behalf of the individual authorities.

424. Other monitoring networks are orientated towards monitoring air quality close to particular emission sources. Examples of such monitoring networks used in this report are those operated by the power generators, Heathrow Airport Ltd and the Highways Agency (monitoring sites close to very busy roads). As with national and regional networks, these other networks have recognised procedures for data quality assurance. A summary of the site types within these other networks is given in Table 4.5.

Table 4.5 Summary of NO_x monitoring stations in other UK networks.

Site Type	Calibration Club	Highways Agency	Heathrow Airport Ltd	JEP/ Innogy	JEP/ Powergen
Kerbside	–	–	–	–	–
Roadside	9	2	–	–	–
Urban Background	11	–	–	–	–
Urban Centre	1	–	–	–	–
Suburban	–	–	–	–	–
Industrial	–	–	–	–	1[b]
Rural	2	–		7	7
Special					
rural kerbside	–	2	–	–	
suburban roadside	–	1	–	–	–
airport	–	–	1[a]	–	–
TOTAL	**23**	**5**	**1**	**7**	**8**

a also part of the LAQN (Table 4.4)
b also part of the Wales network (Table 4.4)

4.7.2 The UK NO_2 diffusion tube network

425. The Department for Environment, Food and Rural Affairs (Defra) and the Devolved Administrations fund a large-scale survey of NO_2 concentrations throughout the UK, using passive diffusion tubes operated by local authorities. The principal aims of this survey are to:

- Identify high concentrations of NO_2 that may warrant more detailed investigation;
- Determine the spatial variation of NO_2 concentrations in the UK;
- Determine trends in NO_2 concentrations over a number of years.

426. The survey commenced in 1993 and currently comprises over 1300 sites operated by more than 300 local authorities. Up to December 2000, each authority operated 4 sites, including one kerbside location (1 to 5 metres from a busy road), one intermediate site (20 to 30 metres away from a busy road) and two background sites (in residential areas, and more than 50 metres from a busy road). From January 2001, sampling at intermediate sites has been discontinued and kerbside sites have been renamed as roadside sites to be consistent with the site classification system used for automatic monitoring. Hence, since January 2000, most authorities now operate 2 roadside sites and 2 urban background sites.

427. Tubes are supplied and analysed by a variety of analytical laboratories, and exposed over 4 or 5 week periods. Data are collated and processed centrally by netcen. Detailed information on the monitoring sites and measured data can be found at www.airquality.co.uk. Annual network reports are published (Loader *et al*., 2002) and a 5-year summary has been prepared (Stevenson, 2001).

4.8 Measurement uncertainty

4.8.1 The uncertainty of measurements – general principles

428. No measurement is exact, however sophisticated the method used. A measurement can only determine that the measured quantity lies within a certain range, and when interpreting data it can be as important to know the size of this range as it is to know the values of the data. This range is called the uncertainty of the measurement, and it is typically expressed as the range within which the true value is likely to lie with a probability of 95%.

429. Historically, uncertainties have been determined by different groups of scientists in quite different ways, so that the results are not necessarily comparable. Since 1993 there has been wide agreement to follow the principles of the ISO Guide to the Uncertainty of Measurement (ISO, 1993). In formal terms this requires an evaluation of all the factors linking the measurement to the definition of the units in which it is expressed[2].

430. The field of air quality in Europe has needed to take a close look at measurement uncertainties, because the Air Quality Directives specify an upper limit for the uncertainty with which different types of measurements must be made. This has resulted in a Report by CEN (the European Centre for Standardisation) entitled: ‘Air quality – approach to uncertainty estimation for ambient air reference measurement methods’ (CEN, 2002). This provides a common approach to determining measurement uncertainty for all pollutants, using various relevant measurement techniques such as automatic analysis, or manual sampling (pumped or diffusive) with subsequent laboratory analysis. The uncertainty limits for NO_x and NO_2, as specified in the EU Directive 1999/30/EC are listed in Table 4.6.

Table 4.6 Measurement uncertainty objectives for NO_x and NO_2 given in EU Directive 1999/30/EC.

	Uncertainty (listed as accuracy in the Directive)
Continuous Measurement (for example, chemiluminescent analyser)	15%
Indicative Measurement (for example, diffusion tubes)	25%

Note: The percentages given in the table are for individual measurements averaged over the period considered by the limit value, at concentrations close to the limit value, for a 95% confidence interval.

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[2] To take a simple example, we might find that using a metre ruler allowed us to measure consistently to the nearest 0.5 mm. However, the GUM approach also requires us to compare our ruler at regular intervals with the definition of the metre – in practice using another ruler that has ultimately been calibrated against a national standard – and to evaluate how our ruler may change in length between calibrations, for example with changes in temperature, in order to assign the correct uncertainty. 0.5 mm is a measure of the repeatability of the measurement, while the uncertainty requires the other factors to be taken into account as well.

4.8.2 The uncertainty of chemiluminescence measurements

431. CEN has produced a draft standard (CEN, 2003a) setting out how National Networks should operate chemiluminescent analysers in order to meet the required uncertainty of ±15% (at the 95% confidence level). This includes a set of performance characteristics against which analysers need to be assessed for official approval, and activities required for ongoing QA/QC. Although the situation is not entirely clear (for example, no analysers commonly used in the UK have been put through a complete set of performance tests), it is likely that the great majority of UK National Network measurements will meet the uncertainty requirement.

432. Uncertainty in measurements with chemiluminescent analysers can arise from many factors relating to the instrument and to its operation and calibration. For example:

Instrument:

- linearity;
- repeatability;
- drift in zero and span;
- dependence on sampling pressure, air temperature, sample gas temperature, voltage;
- averaging error;
- interferences;
- converter efficiency.

Instrument calibration:

- uncertainty in calibration gas concentration;
- incorrect procedure for calibration.

Instrument operation:

- losses in sample line;
- operation outside of specified temperature, line voltage etc.

433. Careful testing of analyser performance and close control of calibration and operational conditions is required to ensure minimum data uncertainty.

4.8.3 The uncertainty of diffusively sampled measurements

434. Although the procedure for calculating the uncertainty of measurements made with diffusive samplers is given within CR 14377 (CEN, 2002), there are as yet no clear results for NO_2 samplers. Properly conducted diffusive measurements of benzene have been found to have uncertainties of around 20 to 25%, but the physical mechanisms for absorbing and analysing hydrocarbons with diffusive samplers are much simpler than those for NO_2. There is a CEN Working Group looking at diffusive samplers for NO_2 measurement. Samplers of several different designs used across Europe – the Palmes tube used in the UK Diffusion Tube Network, Passam (Swiss), Analyst (Italian), IVL (Swedish), and Radiello (Italian) – have been exposed simultaneously alongside automatic analysers. The results give an indication of the variability within individual samplers of the same design, analysed at the same laboratory, and between different designs of sampler, but the study so far is limited in scale and the results are not published.

435. The recent analysis of collocation studies carried out by local authorities throughout the UK has demonstrated that the annual mean concentrations for 50% TEA-in-acetone diffusion tubes, when adjusted for bias, have an uncertainty of ± 20% (at the 95% confidence level). The uncertainty was greater for tubes prepared with 50% TEA-in-water.

4.9 QA/QC of NO_2 air quality monitoring data

4.9.1 Introduction

436. In order to make a reliable assessment of air quality from measurement data, it is essential that the data are of sufficient quality. The overall aim of any quality assurance programmes should be to ensure that the measurement data fulfil the aims and objectives of the monitoring programme.

437. In this report, data from several monitoring networks are utilised. The aims and objectives of these networks may differ slightly, but the following general objectives will apply in most cases:

- meeting statutory requirements;
- providing necessary information on air quality for the public, regulators and/or the scientific community;
- providing information for local authority air quality review and assessment;
- identifying long term trends on air pollutant concentrations;
- assessment of policy effectiveness.

438. ***Quality assurance*** refers to the overall management of the process involved in obtaining the data; whilst ***quality control*** refers to the activities undertaken to check and optimise data accuracy and precision after collection. Hence, quality assurance relates to the measurement **process**, whilst quality control is concerned primarily with **outputs**.

439. This section will summarise QA/QC activities in relation to both the automatic NO_2 monitoring data and the diffusion tube data presented in this report.

4.9.2 QA/QC activities for automatic air monitoring networks

440. There will, inevitably, be differences in the detailed QA/QC activities undertaken in each network supplying data for this report. However, this section provides a general overview of activities which will be applied to all networks, perhaps to a greater or lesser extent, depending on the specific objective of the monitoring undertaken.

441. Quality assurance activities include:

- network design;
- station siting;
- instrument selection;
- instrument calibration;
- instrument service and repair;
- operations manual;
- operator training;
- correct operation of the on-site equipment.

442. Quality control activities include:

- information management;
- data ratification;
- quality circle review and feedback.

443. It is important that all factors that may affect the quality of the data are considered. For example, the sample inlet line to the gas monitor needs to be inert and have no, or negligible, effect on the gases monitored. The sample line needs to be carefully designed, cleaned and maintained and regularly tested for its integrity.

4.9.3 Quality assurance

444. The design of the Automatic Monitoring Network and information on general station siting is provided in the Automatic Urban Monitoring Network Site Operator's Manual (http://www.aeat.co.uk/netcen/airqual/reports/lsoman/lsoman.html) (AEA Technology, 1998). In recent years, some new sites have been added to ensure that the network conforms to the network design and monitoring criteria specified in the Air Quality Framework Directive and subsequent Daughter Directives (96/62/EC, 1999/30/EC, 2000/69/EC). A classification system for UK air quality monitoring site locations has been devised and is presented in Appendix 3. The locations of all monitoring sites used in this report have been classified according to this system.

445. The Site Operator's Manual provides information on the procedures adopted in the network for instrument service and repair and general background information about the network. All site operators have a copy of the manual and are trained by the network QA/QC Unit on all of the network operation procedures.

4.9.3.1 Instrument selection

446. All of the automatic NO_2 data presented in this report have been obtained from chemiluminescent analysers. This is the EU reference method of measurements and described in Section 4.2 of this report. Provided that the operational and QA/QC requirements discussed in this section are followed, measurements from chemiluminescent analysers will be robust and reliable.

4.9.3.2 Calibration

447. In the national network and in the majority of local networks a 3-stage approach to calibration is adopted:

- daily span and zero check with the analysers internal gas source (permeation tube for NO_2 analysers)
- weekly or fortnightly manual calibration with certified calibration gas cylinder
- 6-monthly site audit check with an independent gas standard to check the calibration of the analyser and the stability of the on-site gas calibration cylinder.

448. The daily autocalibration is used only to identify rapidly any possible analyser faults that may require attention. The gas from internal permeation devices is not considered sufficiently stable or reliable to be considered as a calibration source.

449. The weekly or fortnightly manual calibrations with accredited gas standards provide the main calibration data to scale the analyser output to correct concentration units for the gas species being monitored. Detailed records are kept for all calibrations undertaken and, during the calibration visits, any consumables (such as filters etc.) are replaced and the general fabric of the site inspected.

450. A major tool for quality assurance checking in the national networks is the detailed site, instrument, calibration gas and operator audit carried out at 6-monthly intervals. A single independent body visits every site in the network to undertake a full site check. All analysers on site are checked; in relation to the NO_2 analyser, the following checks are undertaken:

- linearity;
- noise;
- response time;
- leaks and flow check;
- converter efficiency;
- analyser calibration.

451. The sample inlet and manifold system and the on-site gas calibration standards are also checked to ensure correct operation of the full measurement system. The bodies undertaking these tests have the appropriate UKAS accreditation and a UKAS certificate of calibration is produced documenting the results of the checks.

452. These quality assurance tasks ensure, as far as possible, that data collected from the analysers are correct and accurate at the time of collection. However, there may be faults and problems that only become apparent when a large time series of data or calibrations are examined together. The Quality Control system therefore provides a further check on the data.

4.9.4 Quality control

453. Quality control of air pollution data is the process of checking, accepting, rejecting or adjusting the data, on the basis of all available information. This operation is usually referred to as data ratification and is generally carried out on 3 or 6-month data blocks, or annually, so that a reasonable amount of data and several calibrations are available to assess consistency and long-term performance. The ratification procedure consists of collating all of the available data, calibration records, service records and any other information relevant to the analyser or site operation. All of this information, together with meteorological data and information from other analysers or other sites, may be used in the ratification process. An important principle of the data ratification procedures is that data are always retained unless there is a specific reason for rejection.

454. The final task of ratification, within the national automatic network, is to hold a quality circle to identify any generic issues arising from the data ratification process and to use these as a basis for recommendations to improve network performance.

4.9.5 Diffusion tubes

455. Diffusion tube data in the UK Nitrogen Dioxide Diffusion Tube Network undergo similar QA/QC procedures with the same aims as those for the automatic data, but with differences appropriate to the method.

456. The monitoring sites are classified using the same system as for the automatic sites, although all diffusion tube sites are now either roadside or urban background. All monitoring locations selected by local authorities are checked from maps and photographs supplied. A manual for the survey (AEA Technology, 2003) has been prepared as can be found at: http://www.aeat.co.uk/netcen/airqual/reports/NO_2man/NO_2instr.pdf

457. Data are manually checked as they are received from the local authorities on a monthly basis. Any problems identified at this stage are discussed with the local authority concerned.

458. Two quality assurance schemes are operated to control analytical laboratories contributing data to the network. The Health and Safety Executive operate a laboratory proficiency test scheme as part of the Workplace Analytical Scheme for Proficiency (WASP) and, in addition, a field intercalibration of diffusion tubes from all laboratories takes place annually. The latest report has been published (AEA Technology, 2002).

459. At the end of each calendar year, the dataset is checked in its entirety and also fed back to the local authorities for final checking and any additional comments on their own data. When these checks have been completed, the data are formally ratified.

4.9.6 Provisional and ratified data

460. Data from the UK national air quality monitoring networks are marked as 'provisional' or 'ratified' depending on what stage they are at in the process of ratification. When data are first collected from the analyser on site, they are scaled with calibration data from the last calibration. In many cases, this will produce satisfactory data, with problems only arising if the instrument is unstable or drifting rapidly. The data are then automatically screened using computer algorithms to highlight suspect data, but no data are deleted at this stage. This process occurs in real time and all data that are not marked as suspect are sent to the National Air Quality Information Archive within about 1 hour of being collected. Data sent to the archive at this stage are marked as provisional.

461. All of the provisional data, together with all the identified suspect data, then undergo full ratification, as described in Section 4.9.4 in 3-month blocks. As part of the ratification process, the suspect data may be reinstated if deemed to be genuine. Following the ratification process, the fully ratified dataset are re-sent to the Air Quality Archive (in 3-monthly blocks, 3 months in arrears) and the provisional data are overwritten with the ratified data.

462. A similar procedure is adopted for the NO_2 diffusion tube data except that the provisional data are not collected automatically but on manually returned forms from the many network participants. The initial screening is undertaken manually and the provisional data are updated to the Archive every 3 months. These provisional data are then ratified annually to produce the final ratified dataset for the year, using the procedures outlined in Section 4.9.5. This ratified data set then overwrites and updates the Archive once per year, approximately 6 months after the year ends.

463. All data provided in this report have undergone quality assurance and quality control checking and are therefore regarded as ratified.

4.10 Future developments relating to European standardisation

464. As mentioned in Section 4.8.2, one of the elements of the European Air Quality legislation is a set of written standards that are currently being produced by the European Centre for Standardisation (CEN). These are designed to ensure that all Member States are making their measurements with the required level of accuracy, and Member States must follow these standards unless they can demonstrate that their own methods give equivalent results.

465. The draft standard for chemiluminescent NO and NO_2 measurements, prEN 14211 (CEN, 2003a), has been written by a CEN Working Group of technical experts from the Member States. As this AQEG report is being written, comments are being sought on the draft from the wider measurement community through national standardisation bodies, for example BSI in the UK. These comments are expected to lead to changes in at least the detail of the standard before it comes into force. It would therefore be premature to predict the ways in which National Network measurements will need to change to comply with European requirements in future. They will depend on the results of tests on analysers as they are assessed against the final set of performance characteristics, and on the detailed requirements for daily calibration checks and longer term Quality Assurance/Quality Control, for example. When published, this CEN standard will legally replace the existing British standard covering ambient NO and NO_2 measurements, ISO 7996:1985. The CEN standard will place more stringent requirements on the measurement process than the ISO standard.

466. A formally accredited route for certifying instruments against the performance characteristics is already in place within the Environment Agency's MCERTS scheme.

467. Regarding diffusive sampling, a series of CEN standards has recently been published (CEN, 2003b) as EN 13528 Ambient Air Quality – Diffusive samplers for the determination of concentrations of gases and vapours: Part 1 – General Requirements; Part 2 – Specific Requirements and Test Methods; and Part 3 – Guide to selection, use and maintenance. These standards have a different approach to that for chemiluminescent measurement in that they were not specifically written to support the EU legislation, and so are not obligatory, and they do not go into a similar level of detail as to how measurements should be made. The scope of these standards includes samplers for NO_2, NO, SO_2, NH_3, O_3, formaldehyde, and volatile organic compounds. Further CEN standards covering individual pollutants such as NO_2 in detail may be produced, but the position at present is not clear.

Chapter 5

Modelling

Key points

- Empirical models have been developed to address the relationship between hourly mean NO_2 and NO_x concentrations and annual mean NO_2 and NO_x concentrations. The latter have had major application in air quality policy development with respect to achieving the annual mean air quality objective for NO_2.
- The validity of the cause and effect relationships implied in the national scale empirical model have been verified in physically-based process models and shown to result from the conversion of NO to NO_2 by reaction with O_3 and from the direct primary emission of NO_2.
- Comparisons between the national empirical model and dispersion models have been undertaken for locations in London. However, London is a special case in terms of geographical extent, emissions, high concentrations of NO_2 and number of monitoring sites which has allowed optimum development of empirical schemes. Thus the conclusions of such comparisons may not be valid in other major cities where the empirical formulations may be based on very few monitoring sites. It is recommended that further comparisons should be performed in major cities with the highest annual average NO_2 concentrations. It is recognised that such comparisons would be enhanced by more rural sites measuring both NO_x and O_3.
- There are a number of uncertainties that should be taken into account in assessing the policy conclusions concerning NO_2 and NO_x based on empirical, dispersion and other physically-based process models. Emission estimates are a crucial source of uncertainty, particularly the mass fraction of NO_x emitted as NO_2 and the likely future split between vehicle-kilometres travelled by petrol and diesel cars. Difficulties remain in representing urban influences on dispersion of primary NO_2 and in representing the 3-dimensional distribution of O_3 in urban areas which drives secondary NO_2 formation.
- There remains uncertainty as to the detailed spatial distribution of NO_x and NO_2 close to streets or junctions adjacent to buildings. There is a need for improved parameterisations in practical models which may be based on advanced building-resolving numerical models and measurements. There remains a basic incompatibility between the outputs derived from traffic counts and models, and the basic traffic input requirements of both emission and dispersion models. Emission and dispersion models require disaggregated data not just by type of vehicle (for example, passenger car, vans, heavy goods vehicles etc), but by fuel type, engine size and most significantly by vehicle age. It is these parameters, combined with vehicle speed, that are used to characterise the emissions associated with individual vehicles. Existing automated traffic counts and assignment models routinely provide traffic data in terms of axle counts and vehicle length subdivided into a number of vehicle length bins. The translation of these counts into corresponding emission categories undoubtedly introduces additional uncertainty.

- Advanced Eulerian and computational fluid dynamics (CFD) models complement the simpler deterministic and empirical models. The advanced models require very much greater computational resources, time, and data, but assist in the understanding of the complex interactions between processes that determine ground-level concentrations of NO_2. For example, CFD examines the effect of the shape and separation of buildings, while the larger-scale models allow study of regional processes, the response of ground-level concentrations to vertical mixing and the development of the urban boundary layer.

5.1 Introduction

468. This chapter describes techniques for modelling and mapping pollutant concentrations at local, urban and regional scales. Although air quality monitoring generally provides reliable data on ambient concentrations of pollutants, monitored data only represent measurements that are made at specific locations at specific times. As such, these data alone provide limited information about the spatial extent of pollutant concentrations and how pollutant concentrations may change in the future. Policy requirements therefore necessitate some form of modelling to be undertaken to address these limitations and to fulfil the following roles:

- providing increased understanding of the relative importance of the different physical and chemical processes that result in NO_2 pollution levels. This includes both increased understanding of transport and dispersion and of the NO_2 to NO_x relationships identified in the data;
- providing an assessment of NO_2 air quality through the generation of pollutant concentration maps for screening purposes for planned road and other development schemes, for example, using the approach in the Design Manual for Roads and Bridges (DMRB);
- interpolating NO_2 air quality data between monitoring sites and to map NO_2 air quality data over extended local, urban and regional scales;
- providing modelling tools to project NO_2 concentrations into the future and to study policy options and management activities for future NO_x emission reductions;
- providing indirect verification or otherwise of emission inventories for NO_x.

5.1.1 Modelling approaches

469. The Expert Group has not relied on one particular modelling approach but has used a variety of approaches and techniques depending on specific requirements. These requirements relate to the purpose of the exercise in terms of the level of accuracy required and the temporal and spatial scales under investigation.

470. Modelling methodologies can be categorised into two general types, namely empirically-based statistical models and physical process-based deterministic models. Deterministic models are based on mathematical formulations which describe the physical and chemical processes operating within the atmosphere, and which can be expressed at different levels of complexity. Some models combine both approaches; for example using a deterministic approach for the transport and dispersion of pollution, but statistical or empirically-based formulations to derive NO_2 concentrations from NO_x concentrations. Each modelling approach is associated with a set of advantages and disadvantages which are discussed in greater detail in the individual sections within this chapter.

471. This diversity of models provides a pragmatic route forward because it is not practical in one model to cover all spatial scales from as small as a metre up to the national scale, and all timescales from less than an hour to annual. There is neither the UK-specific input data necessary to drive such a model, nor are there computers approaching the required capacity to run it. In the paragraphs below, a short introduction is given to a range of different models, followed by a more detailed description of their features and application in the UK to the development of air quality policy on NO_x.

5.1.2 Empirically-based statistical models

472. Empirical models are based largely on air quality data. With the dramatic increase in the number of automatic continuous NO_x monitoring sites during the 1990s, empirical models have become more robust and their coverage of the urban environments has become more extensive. They have been used extensively for policy support through their application to mapping current NO_2 levels and the exceedences of air quality targets and to the projections into the future of such levels and exceedences. The assumptions that correlations represent cause and effect and that regression parameters remain constant into the future, are their main weaknesses.

473. Although empirical models can be used for short-term predictions based on correlations between pollutant concentrations and meteorological variables these types of models are most reliable for longer term averages (for example, annual averages). In view of speed of calculation and reasonable accuracy they have been used extensively to model annual average concentrations of NO_2 across the UK. Specifically, for annual average NO_x concentrations the large number of monitoring sites now available has allowed the contribution to the NO_x concentration at a specific point to be modelled as three components:

(i) a background based on rural data;

(ii) an urban background component based on a regression analysis of dispersion modelling of relatively local emissions together urban background monitoring data;

(iii) a roadside component which can be based on empirically derived formulations.

474. An alternative approach has also used dispersion modelling for the roadside component of the NO_x, from which annual average NO_2 are then derived using empirically based formulae as discussed in section 5.2.2. This is the approach used by the screening model in the Design Manual for Roads and Bridges (DMRB) (Highways Agency, 2003).

475. Surface concentration modelling is a particular form of empirically-based modelling referring to the process of mapping continuous surface data solely from point-based measurements using interpolation procedures. This approach enables monitoring data to be used in exposure assessments in relation to both long-term patterns and short-term events, such as air pollution episodes. Interpolation procedures are also implicit in a number of other empirical models and dispersion models. As with the other modelling procedures outlined in this chapter, there is no universally agreed procedure to estimate concentration surfaces and the robustness of the method for a particular parameter depends on the nature of that parameter and the characteristics of the measurements made (for example, siting characteristics, sample locations and sample numbers). However, there are a number of techniques that are commonly employed to map the spatial distributions of air quality data and these are outlined later in the chapter.

5.1.3 Deterministic models

476. Deterministic models range enormously in sophistication, type and application and include Gaussian plume models, Eulerian grid models, Lagrangian trajectory models and computational fluid dynamics (CFD) models.

477. Gaussian plume models based on simple solutions of the diffusion equation are commonly used within the UK. They offer a practical and physically-based treatment of the dispersion of NO_x emitted from large point sources, such as power stations; from area-based sources, such as commercial, industrial and domestic sources; and from linear sources, such as motor vehicle traffic, using fixed plume geometries. They can represent many of the important chemical conversion processes and can treat most of the important spatial and temporal scales. Furthermore, their output in the form of maps provides a ready visualisation of the impacts of NO_x control policies on NO_2 levels and air quality target exceedences. They are used extensively for local policy applications in the UK but have limited utility at the national scale due to computational and data requirements.

478. Lagrangian trajectory models simulate the advection of individual air parcels by a three-dimensional wind flow. These models offer the prospect of a more detailed representation of the turbulence, convection and chemical processes influencing urban air quality. They share many of the advantages and disadvantages of complex deterministic models. They require large amounts of input data and the results of their model validation and verification studies are difficult to interpret. Most studies conducted so far with this type of model have represented motor vehicle sources at the 1 km x 1 km scale and this scale is too coarse to represent the detailed structure in the spatial distribution of NO_2.

479. The most commonly used model type for urban air quality calculations in North America are numerical Eulerian grid models. Such models are also used for policy related assessment in Europe. These generally include both meteorological models to calculate mean flow and turbulence, and transport and diffusion models. These models generally require large amounts of input data and computer time and also produce large volumes of output data that can be difficult to interpret. Computer capacities currently limit the smallest horizontal grid sizes that can be utilised to about 1 km x 1 km, whilst integration periods are generally limited to a few days so that is not practical to calculate long term averages. The advantage of these models is their ability to include time varying meteorology in a detailed manner that allows analysis of pollutant episodes. This approach is also compatible with sophisticated chemistry routines.

480. CFD models also solve the fluid dynamic equation, however their focus is on the detailed modelling of complex structure at the small scale (for example, street canyons). They are thus able to aid in understanding of local processes but are not appropriate for calculating concentrations of pollutant right across an urban area.

481. The following sections of this chapter consider both empirical and deterministic models used in studies in the UK. These include widely used methodologies (for example, the National empirical model and ADMS-Urban) through to sophisticated models which have been used for particular case studies (for example, LRTCM trajectory models in London and UAM in Birmingham). A detailed discussion on NO_x:NO_2 relationships is also presented.

482. Table 5.1 describes some of the modelling techniques used in the UK by practitioners, policy makers and researchers. The table does not give model sub classes or version numbers but lists basic names only. Details of model version/developers/suppliers etc. may be found from the reference and/or from the web sites. Most European models are listed on the Model Documentation System: http://air-climate.eionet.eu.int/databases/mds_search.html. US and some other models are also presented on the US-EPA web site: http://www.EPA.gov/scram001/tt22.htm.

Table 5.1 Description of the main models and modelling techniques used in the UK for modelling NO_2 air quality[1].

Model	Basic description	Meteorology	Transport and dispersion	Chemistry	Spatial scale
1. Empirical models, long term (annual) averages only					
DMRB (2003) Highways Agency (2001)	Screening model for road traffic emissions and air quality	Fixed, at 2 m s^{-1} equally distributed from all directions	Incorporates a fixed empirically adjusted Pasquill stability category	NO_x:NO_2 relationships	Local
ERG Carslaw *et al.* (2001)	All emission types; set-up for London	Different statistical analyses for different years	Regression analysis based on ADMS 3 calculations and monitoring data	NO_x:NO_2 relationships	Urban down to local scale including spatial variation at local street scale
NETCEN Stedman *et al.* (2002)	All emission types	Different statistical analyses for different years	Regression analysis based on ADMS 3 calculation and monitoring data for urban background; simple formulation for representative calculation on road segments	NO_x:NO_2 relationships	National to suburban scale representative concentrations for road segments

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[1] This table is intended only to indicate some of the different modelling techniques and models available. It makes no attempt to be exhaustive, and some commercial models have not been included. Omission of a model should not be inferred as conveying any opinion by the Air Quality Expert Group and, equally, inclusion does not constitute an endorsement by the Group.

Model	Basic description	Meteorology	Transport and dispersion	Chemistry	Spatial scale
2. Models employing analytic expressions for concentration (for example, Gaussian models); long and short term average concentrations					
ADMS McHugh *et al.* (1997)	Models for stationary source emissions and/or road traffic emissions	Hourly sequential data from one site	Quasi Gaussian type model using h/M_{MO} parameterisation of boundary layer. Also includes trajectory model and canyon model based on OSPM approach	Generic reaction set	Urban down to local, including spatial variation at local street scale
AERMOD Lee *et al*. (1996) US-EPA www.epa.gov/scram001	Stationary source emissions	Hourly sequential data from one site	Quasi Gaussian type model. h/L_{MO} parameterisation of boundary layer	O_3 limiting method	About 50 km down to local
AIRVIRO	All emission types	Hourly sequential data or fields	Gaussian plume model and Eulerian grid model for pollutant advection and dispersion	Limited	Urban to local
CALINE Benson (1979) US-EPA www.epa.gov/scram001	Road traffic emissions	Hourly sequential data	Gaussian model. Pasquill parameterisation of boundary layer	Discreet parcel method for NO_2 for some versions	Local
CAR	Road traffic emissions				Street scale
OSPM Berkowicz *et al*. (1997)	Road sources in street canyons	Hourly sequential data	Two component model – direct plume component and recirculation component	Various	Street canyon scale

Model	Basic description	Meteorology	Transport and dispersion	Chemistry	Spatial scale
3. Numerical models.					
MM5-CMAQ/ Models 3	Advanced nesting air quality model – US community model for air quality with focus on O_3	MM5	CMAQ	SMVGEAR model based on CBM-IV	Minimum 1 km resolution
EURAD	Nested mesoscale transport model	Mesoscale MM5	Bott Advection scheme	RADM2, CBM4.	Grid size 2 to 80 km
LRTCM Derwent (1999)	Lagrangian column trajectory model	Hourly sequential for 1995	Trajectory model	NO + O_3	1 x 1 km resolution
MEMO	Simulation of mesoscale air motion and passive pollution dispersion at local to regional scale	Non hydrostatic model	TVD scheme	None	Grid resolution 500-10,000 m domain size 10-500 km
NAME	Mesoscale model for pollutant dispersion	Mesoscale model	Lagrangian particle model	NO_x:NO_2 relationships	15 km x 15 km
UAM	Urban air quality model	Generally requires mesoscale model as driver	Eulerian scheme	CBMIV	Minimum 1 x 1 km
4. Computational fluid dynamics					
For example, CFX FLUENT STAR-CD Galpin *et al*. (1985)	General purpose CFD	Meteorology represented as ‘upstream’ profiles, finite volume techniques, range of turbulence parameterisation	Eulerian transport model	Reaction schemes available	Local scale

5.2. Empirical models of NO_x and NO_2 concentrations across the UK

5.2.1 The UK National Scale Empirical Model of NO_2 and NO_x

5.2.1.1 Introduction to the National Scale Empirical Model

483. Empirical models have been used by netcen to estimate both background and roadside annual mean NO_x and NO_2 concentrations across the UK for the purposes of policy development within the UK. They are therefore relevant only to the conditions of the UK and to the time periods appropriate to the observations employed. Maps for 2001 have been prepared for the assessment of ambient concentrations in the UK required under the first Air Quality Daughter Directive (AQDD1) and can be projected to years such as 2005 and 2010 for baseline analysis and scenario testing (see section 7.3). Detailed descriptions of the modelling methods are available from earlier publications (Stedman *et al.,* 1997; Stedman *et al.,* 2001a; b; Stedman and Bush, 2000; Stedman *et al*., 2002). A summary of the approach is provided here along with details of key aspects of the method. The methods used to calculate site-specific projections at monitoring site locations are also described.

484. A map of annual mean background NO_x concentrations in 2001 has been built-up from contributions from:

- distant sources (characterised by the rural background concentration);
- large point sources;
- local area sources.

485. Hence, at locations away from busy roads NO_x concentrations has been estimated as:

Estimated background NO_x concentration (µg m^{-3}, as NO_2) = corrected rural NO_x concentration (µg m^{-3}, as NO_2) + contributions from major point sources (µg m^{-3}, as NO_2) + contributions from area sources (µg m^{-3}, as NO_2)

486. At locations close to busy roads an additional roadside contribution is added to account for contributions to total NO_x from road traffic sources.

487. Empirically derived conversion factors are then used to estimate the NO_2 concentrations from modelled NO_x concentrations.

5.2.1.2 Empirical relationships between NO_x and NO_2 at background locations

488. The following relationships have been derived from monitoring data from 1998 to 2001 inclusive and are presented in Figures 5.1 and 5.2.

Rural Background NO_2 [µg m^{-3}] = 0.7835.NO_x [µg m^{-3}, as NO_2]

(Elsewhere) Background NO_2 [µg m^{-3}] = 1.9301.(NO_x [µg m^{-3}, as NO_2])$^{0.6887}$

Central London Background NO_2 [µg m^{-3}] = 2.28.(NO_x [µg m^{-3}, as NO_2])$^{0.6887}$

489. At rural background locations, the majority of NO_x is present as NO_2, because such areas are generally distant from sources, and oxidant (O_3) is available in excess. In urban areas however, annual mean NO_2 concentrations are more limited by the availability of oxidant. From Figure 5.2 it is also notable that measured NO_2 concentrations in Central London are higher than in other urban areas. This is thought to be due to the large size of the urban area, which enables relatively aged NO_x from other parts of London to contribute to NO_2 in Central London.

Figure 5.1 Relationship between annual mean NO_x and NO_2 concentrations measured at rural background sites in the AURN 1998-2001.

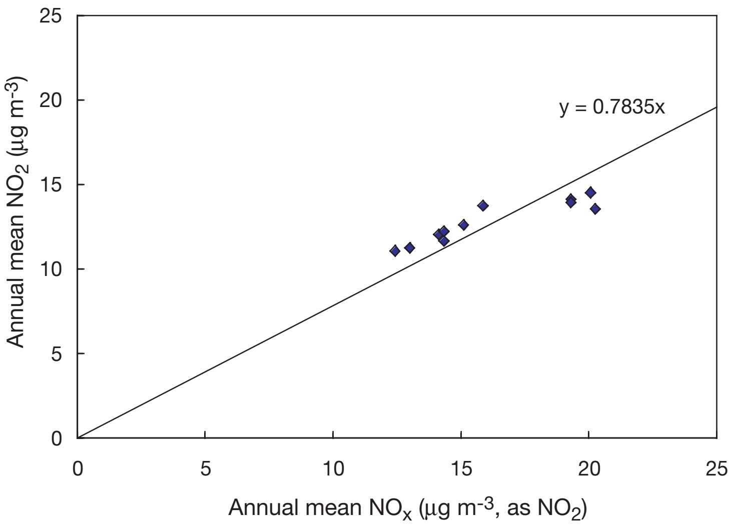
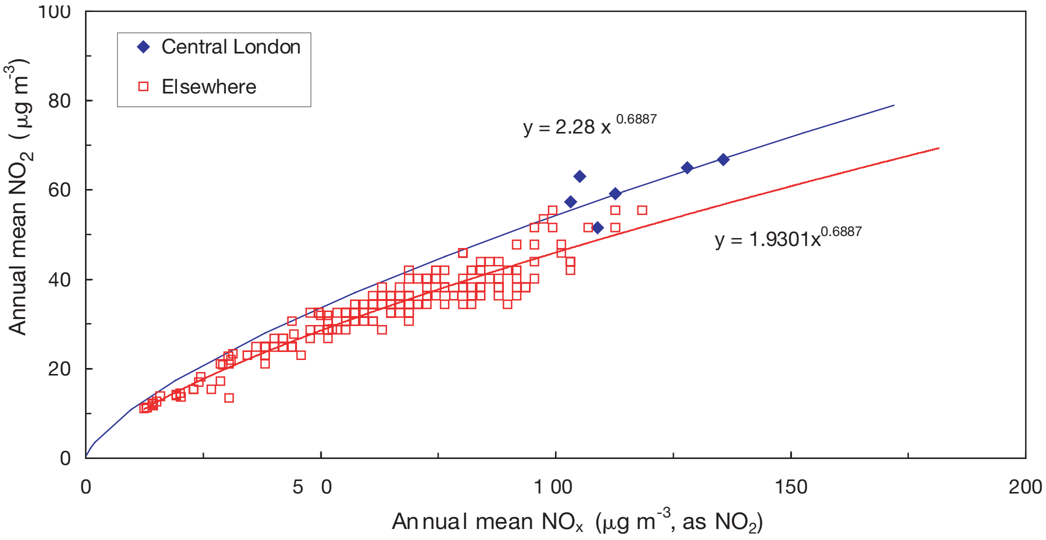


Figure 5.2 Relationship between annual mean NO_x and NO_2 concentrations measured at background sites in the AURN 1998-2001 with at least three years data.



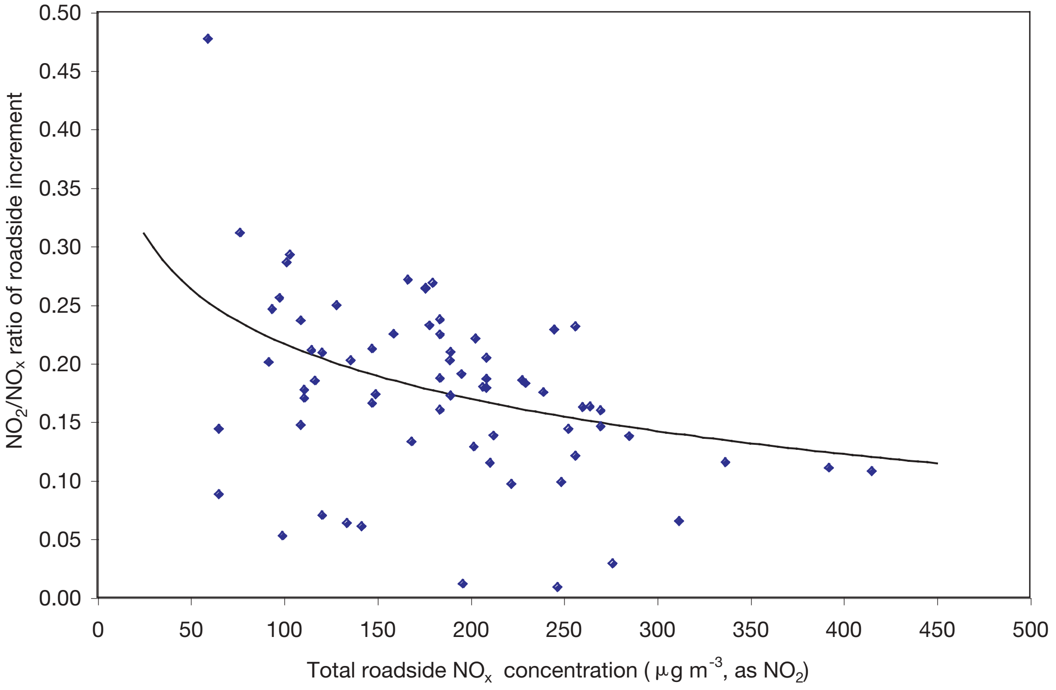
5.2.1.3 NO_x to NO_2 relationships at roadside locations

490. Estimates of annual mean roadside NO_2 concentration have been calculated as the sum of the modelled background NO_2 concentration and an NO_2 roadside increment derived from a modelled NO_x roadside increment using the following equation:

NO_2 Roadside increment (µg m^{-3}) = NO_x Roadside increment (µg m^{-3}, as NO_2). (0.53 – 0.068.ln(total roadside NO_x concentration (µg m^{-3}, as NO_2))

491. This method (developed by Laxen and Wilson (2002b) from a method proposed by Stedman *et al*. (2001a)) takes into account that the NO_2/NO_x ratio at high NO_x locations (close to major roads) will be lower than the same ratio at lower NO_x areas as a result of oxidant limiting. The NO_x to NO_2 relationship for the roadside increment is based on a natural logarithmic best-fit curve as shown in Figure 5.3. Thus the percentage of the roadside increment of NO_x represented by NO_2 falls from about 25% at low total NO_x concentrations to about 10% at the highest concentrations close to busy roads in large urban areas.

Figure 5.3 Relationship between the NO_x/NO_2 ratio of the roadside increment and total roadside NO_x concentration at AURN and TRL road and kerbside sites 1999-2001.



5.2.1.4 *Detailed descriptions of the individual contributions from point sources*

492. Contributions to ground level annual mean NO_x concentrations from point sources have been estimated by modelling each source explicitly using an atmospheric dispersion model (see Section 5.3). The contribution to annual mean NO_x concentrations from NO_x point sources with >250 tonnes per annum emission, was modelled using ADMS 3.1. Contributions from NO_x point sources with <250 tonnes per annum release were modelled using a dispersion matrix approach, at a 1 km receptor resolution. Each source was assumed to emit into a nominal 1 km x 1 km x 50 m volume.

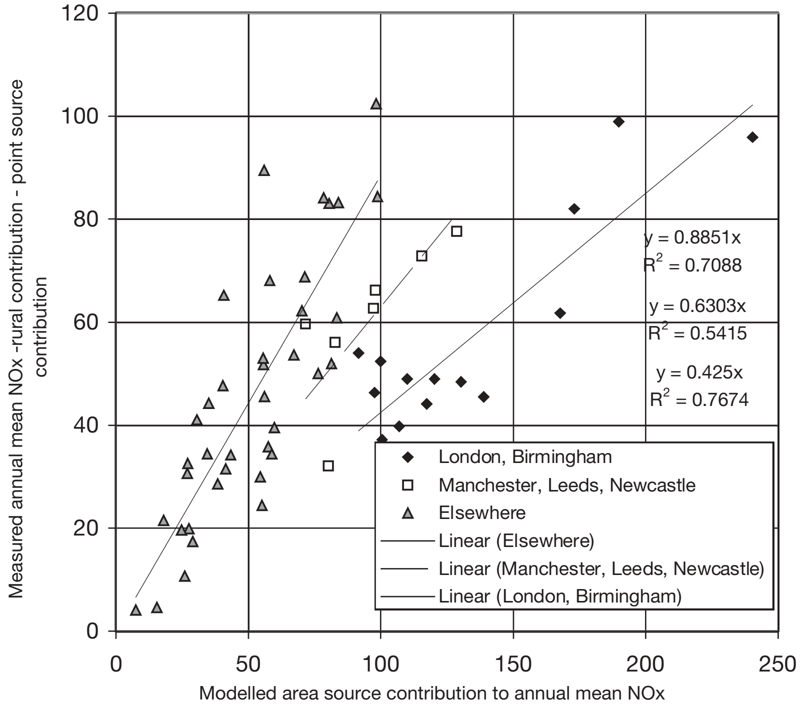
5.2.1.5 *Rural concentrations*

493. Diffusion tube measurement data from the Acid Deposition Monitoring Network (Hayman *et al.*, 2001) were used to estimate rural annual mean background NO_2 concentrations. In a number of instances, measurements from this network are influenced by contributions from nearby point and area sources. Care has been taken to avoid double counting these contributions. The contributions from point sources (as modelled explicitly) and area sources (estimated using the area source modelling methods described below but coefficients derived from earlier work) were subtracted prior to interpolation to obtain estimates of corrected regional background concentrations throughout the UK (section 5.6.4).

5.2.1.6 Area sources

494. An empirical model based method has been adopted to estimate the contribution to ambient concentrations from area sources. An ADMS-derived dispersion matrix has been applied to weight emissions from area sources within a 35 x 35 km square surrounding each monitoring site according to distance and direction of the emissions from the central receptor. 10-year average meteorological data from Heathrow has been used to construct this dispersion matrix and represents the contribution from unit emission in each 1 km x 1 km square to concentrations at the receptor. An example calibration plot for this area source model for 2001 is shown in Figure 5.4. The modelled point source and corrected rural NO_x concentrations have been subtracted from the measured annual mean NO_x concentration at background sites. This is compared with the modelled area source contribution to annual mean NO_x concentration. Emissions from traffic sources have been dispersed from a volume source of height 10 m and stationary area sources have been dispersed from a volume source of height 30 m. Estimates of NO_x emissions for 2000 have been adjusted to values for 2001 using UK sector total emission estimates for 2000 and 2001 available from the NAEI. The modelled area source contribution is therefore directly comparable with the measured concentrations in 2001.

Figure 5.4 Calibration of NO_x area source model for 2001 (µg m^{-3}, as NO_2).



495. The study utilised the air quality model ADMS-Urban. Non airport sources were treated in a standard way as discussed in Chapter 5. Airport sources, including take-off and climb-out, landing, loading bays and taxi areas are represented as a series of volume sources. The calculated concentrations are dependent on the number of individual sources specified and the height and width of these sources. In particular there is considerable model sensitivity to the specified height of the ground level volume sources representing take-off roll and landing. A height of 50 m is used in this study.

496. Examination of figure 5.4 shows that the monitoring sites fall into three groups related to the size of urban area:

- London and Birmingham;
- Manchester, Leeds and Newcastle;
- elsewhere.

497. The elsewhere slope is close to unity, showing that in these locations the un-calibrated model is quite successful in predicting the concentration. The other slopes are lower, reflecting the different meteorological and dispersion conditions in larger cites. Meteorological data from Heathrow is not expected to be representative of central London, for example. The effective roughness in large urban areas is also greater than in rural or smaller urban areas, leading to more efficient dispersion of pollutants.

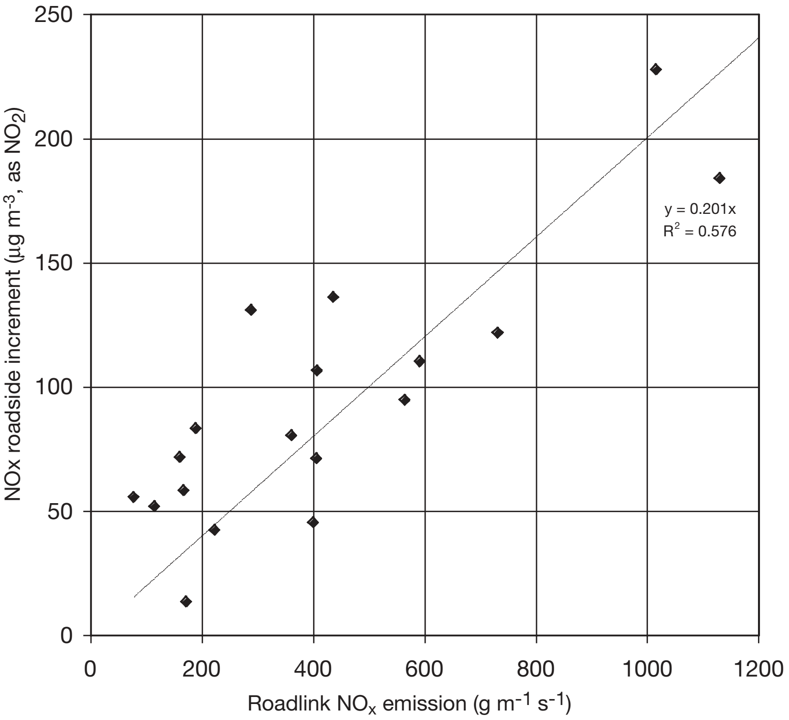
498. The modelled area source contribution can be multiplied by the relevant empirical coefficient to calculate the calibrated area source contribution for each grid square in the country. The corrected rural and point source contributions are then added, resulting in a map of background NO_x concentrations. A map of background NO_2 concentrations can then be calculated from this NO_x map using the NO_x to NO_2 relationships listed above.

5.2.1.7 Roadside concentrations

499. The annual mean concentration of NO_x at a roadside location can be made up of two parts: the background concentration (as described above) and a roadside increment:
roadside NO_x concentration = background NO_x concentration + NO_x roadside increment

500. The NAEI provides estimates of NO_x emissions for major road links in the UK for 2000 derived from traffic counts carried out at a ‘census point’ alongside each link (Goodwin *et al.,* 2001). The estimates for 2000 have been adjusted to provide estimates of emissions in 2001. Figure 5.5 shows a comparison of the roadside increment of annual mean NO_x concentrations at roadside or kerbside national automatic monitoring sites with NO_x emission estimates for the individual road links alongside which these sites are located. The background NO_x component at these roadside monitoring sites was derived from the map described above. This comparison provides an empirical dispersion coefficient, which can be used to calculate estimates of the roadside increment of annual mean NO_x concentrations from road link-specific emission estimates. This empirical model is calibrated using data from roadside or kerbside national automatic monitoring sites, so the estimated roadside concentration will be representative of the places in which the monitoring sites are located (typically 5 m from the kerb, see section 4.7). Thus a single estimate of the roadside concentration is calculated for each major road link in the UK. This is in contrast to more detailed local air quality modelling studies, which typically provide estimates of concentrations at a range of receptor points adjacent to an individual road link.

Figure 5.5 Calibration of NO_x roadside increment model for 2001 (µg m^{-3}, as NO_2).



501. The roadside increments of annual mean NO_x concentrations adjacent to motorways and non-built-up dual carriageways are considerably smaller per unit emissions than for built-up roads in urban areas. This is due to a combination of the wider traffic lanes, faster speeds and the more open aspects typically found on this type of road. An empirically derived factor of 0.25 was therefore applied to emissions from motorways and non-built-up dual carriageways prior to inclusion in the calibration. No attempt has been made to calculate roadside concentrations for non-built-up single carriageway roads.

502. The roadside increment of NO_2 can be calculated from the roadside increment of NO_x using the relationships described above. This is then added to the mapped background NO_2 concentration:

roadside NO_2 concentration = background NO_2 concentration + NO_2 roadside increment

5.2.1.8 Verification of mapped values

503. Figures 5.6 and 5.7 show comparisons of modelled and measured annual mean NO_x and NO_2 concentration in 2001 at background monitoring site locations and Figures 5.8 and 5.9 show similar comparisons for roadside sites. Both the national network sites used to calibrate the models and the Calibration Club sites (and other sites quality assured by netcen) which provide an independent verification of the modelled concentrations are shown. Lines representing y = x – 30 % and y = x + 30% are also shown.

Figure 5.6 Verification of background NO_x model for 2001.

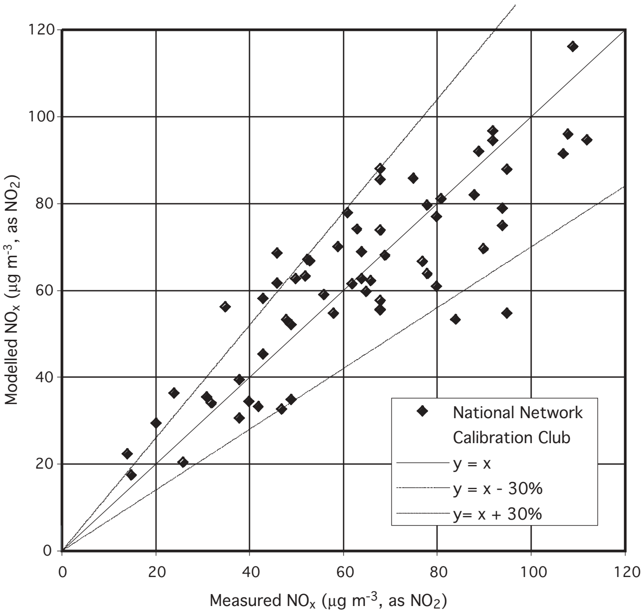


Figure 5.7 Verification of background NO_2 model for 2001.

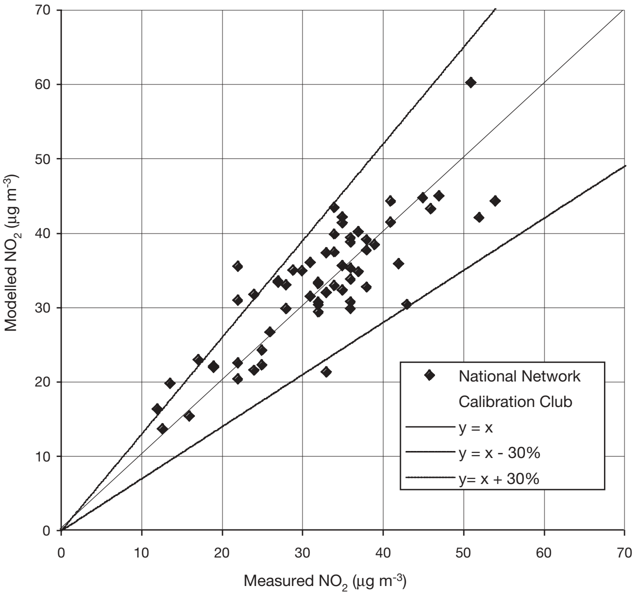


Figure 5.8 Verification of roadside NO_x model for 2001.

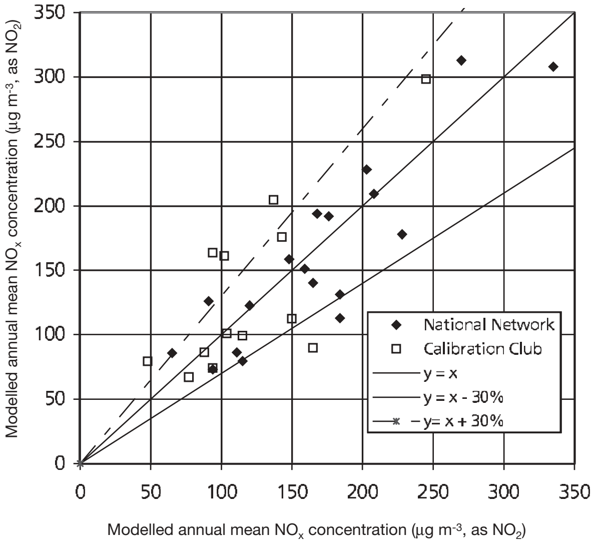
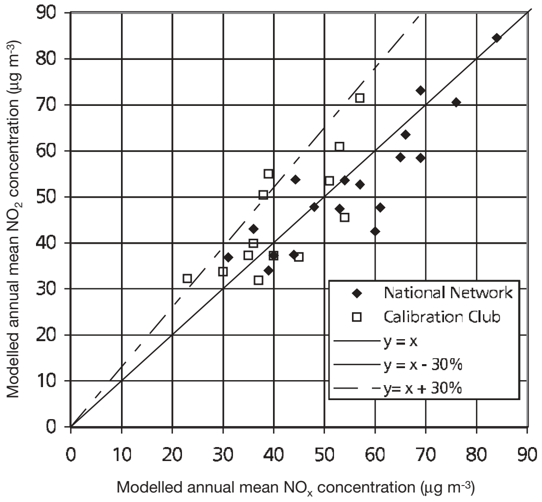


Figure 5.9 Verification of roadside NO_2 model for 2001.



504. Summary statistics for the comparison between modelled and measured NO_x and NO_2 concentrations are listed in Tables 5.2 and 5.3. Comparisons of modelled and measured NO_x concentrations provide an additional check on the reliability of modelled estimates of NO_2. This is because the non-linear relationships between NO_x and NO_2 tend to cause modelled NO_2 concentrations to be relatively insensitive to errors in the dispersion modelling of NO_x.

Table 5.2 Summary statistics for comparison between modelled and measured NO_x and NO_2 concentrations at background sites (µg m^{-3}, as NO_2).

		Mean of measurements	Mean of model estimates	r^2	Number of sites
National Network	NO_x	63.3	62.6	0.72	62
Calibration Club	NO_x	51.3	61.0	0.77	25
National Network	NO_2	32.1	33.0	0.70	62
Calibration Club	NO_2	27.1	32.2	0.55	25

Table 5.3 Summary statistics for comparison between modelled and measured NO_x and NO_2 concentrations at roadside sites (µg m^{-3}, as NO_2).

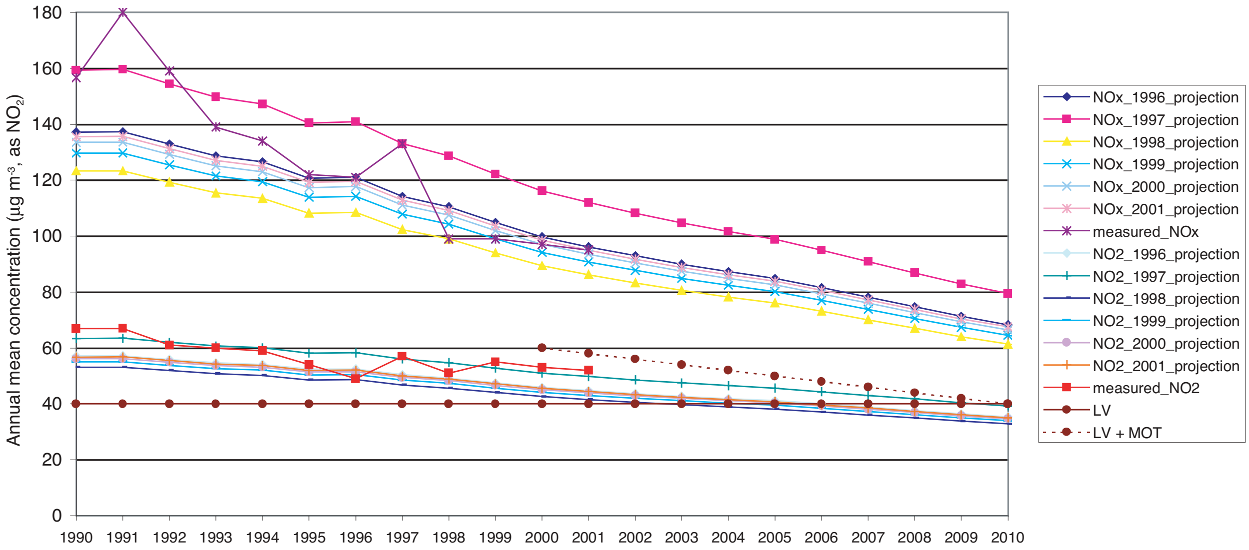
		Mean of measurements	Mean of model estimates	r^2	Number of sites
National Network	NO_x	168.0	160.5	0.79	18
Calibration Club	NO_x	120.2	131.7	0.55	13
National Network	NO_2	55.3	42.4	0.79	18
Calibration Club	NO_2	41.4	45.1	0.57	13

5.2.1.9 Site-specific projections of NO_2 concentrations

505. Site-specific projections of annual mean NO_x and NO_2 concentrations have also been calculated using empirical models derived from the national mapping models. These projections provide a valuable tool for the rapid assessment of the impact of a range of possible policy options on ambient concentrations. Concentrations can also be projected backwards and the comparison with historical measured concentrations can provide useful insights into the reasons for the observed trends in addition to some verification of the model predictions. The site-specific projections also have the advantage of reducing the uncertainties associated with the empirical or dispersion modelling of concentrations.

506. Figure 5.10 shows an example of site-specific projections for the West London monitoring site. The empirical models described above have been used to assign the measured annual mean NO_x concentration in a range of base years to different emission sectors (rural background, traffic, industry, domestic, etc). The contribution for each sector in different years is then calculated using emission statistics and projections from the NAEI. These are then summed to calculate the predicted NO_x concentration in each year. Annual mean NO_2 concentrations are then calculated from the NO_x estimates using the empirical relationships described above.

Figure 5.10 Site-specific projections of annual mean NO_x and NO_2 concentrations at West London



5.2.1.10 *Advantages and disadvantages of the National Empirical Model*

507. The disadvantages of the empirical modelling methods include the following. The models do not attempt a full physical description of the processes taking place in the atmosphere and there is a risk that the empirical calibration of model results could hide errors that tend to cancel out. It is also assumed that any empirical relationships or coefficients will remain valid for other locations or at other times, such as projections to 2010. It is not always straightforward to incorporate adjustments to empirical models to take into account factors such as changes in oxidant concentrations, or primary NO_2 emissions. The spatial resolution of the model outputs is also limited and is restricted to estimates for background and roadside locations. No treatment of concentrations at locations intermediate between roadside and background or at complex locations such as junctions is attempted.

508. The first attempts to map NO_2 concentrations across the UK used a combination of diffusion tube data and population density as a surrogate for NO_x emission estimates at a high spatial resolution (Campbell *et al.,* 1994; QUARG, 1993). The UK scale empirical models have since been extensively developed in a number of important areas and these highlight some of the advantages of empirical modelling methods. NAEI maps of UK emissions at a 1 km x 1 km resolution are now used along with emissions estimates on an individual road link basis for the major road network. This avoids the uncertainties that surround using surrogate statistics other than emission estimates for the empirical modelling. The models are calibrated using automatic monitoring data, which are now available from extensive networks in the UK. A dispersion matrix approach has been adopted to model the contribution from area sources to ambient concentrations. This enables the model to be calibrated directly in ‘concentration units’, improving the transparency of the calibration compared with calibrating the relationships between emissions and concentrations directly. This has the additional advantage that the models derived for a base year for which measured concentrations and emissions inventory estimates are available can be applied to calculate estimates of concentrations for alternative scenarios or projections by revising the emission inventory inputs. Another advantage of empirical modelling methods is that they are generally very computationally efficient in comparison with more complex modelling methods.

5.2.2 Screening models

5.2.2.1 Introduction to the screening models

509. Screening models are designed to provide an initial estimate of pollutant concentrations arising from specific sources. The main characteristic of all screening models is that they require relatively simple inputs, with the results being generated rapidly and with a minimum of computer processing. The predictions are usually used to determine the need for a more detailed assessment. In general screening models are often simplified versions of more sophisticated approaches, which omit or simplify various input parameters such as meteorology. Although screening models are designed to be quick and simple to use, the assumption that they are inaccurate is not necessarily valid but they do take a generally precautionary approach.

510. Screening models are available from the United States Environmental Protection Agency support centre for regulatory air models web site (www.epa.gov/scram001/tt22.htm). All of these models incorporate Gaussian dispersion functions, and are applicable for the determination of relatively conserved species such as CO. In general, NO_2 concentrations are derived from predicted NO_x concentrations, the latter quantity remaining relatively stable in the immediate vicinity of a source. The functions used to convert NO_x to NO_2 are based on empirical relationships derived from simultaneous ambient measurements of the two pollutants.

5.2.2.2 The DMRB Screening Method

511. With respect to UK road transport, one of the most widely used screening approaches is that contained within volume 11 of the Design Manual for Roads and Bridges (DMRB) (Highways Agency, 2003). The DMRB provides guidance on the Environmental assessment requirements for highway schemes in the UK, and has also become an important tool within the local authority review and assessment process for air quality, which is a requirement under the Environment Act 1995. This method was originally developed in the early 1980s, and is broadly based upon the TRL PREDCO Gaussian dispersion model. The approach, originally restricted to the estimation of CO concentrations, has subsequently been revised and extended in 2002 to include NO_2, PM_{10}, benzene and 1,3-butadiene (Boulter *et al*., 2002a). The recent revision incorporates the latest UK road transport emission data (Barlow *et al.,* 2001), the vehicle fleet composition model from the 2000 National Atmospheric Emission Inventory (NAEI), a re-evaluation of roadside pollutant dispersion curves (Hickman *et al.,* 2002), the relationships between annual mean concentrations and other metrics relevant to the air quality objectives, and revised empirical relationships to estimate NO_2 from NO_x concentrations.

512. Prior to the revision, the official DMRB method had taken the form of a series of paper-based tables and graphs, and incorporated a step-by-step procedure for calculating the contribution of road traffic to both area-wide emissions and concentrations of air pollutants at a specified location. The method now takes the form of an Excel spreadsheet (DMRB, 2003).

513. In previous versions of the DMRB, the method for deriving NO_2 concentrations near to a road involved the estimation of the background concentration of NO_x, adding this to a NO_x contribution from the road, then converting to NO_2 using a best-fit relationship between measured NO_2 and NO_x based on data from a wide variety of national monitoring sites. The method now employed to derive NO_2 from NO_x is that proposed by Laxen and Wilson (2002b). This approach assumes that concentrations near to roads are made up of two components: (i) NO_2 from the road traffic and (ii) NO_2 from the background air. The relationship between the atmospheric NO_2 and NO_x concentrations due to road traffic was examined by subtracting the corresponding background values of NO_2 and NO_x from the measured values at monitoring sites. In the simplest terms, the road traffic contribution to NO_2 can be treated as being a constant proportion (approximately 16 %) of road traffic NO_x. However, the actual proportion is dependent on the total amount of NO_x. Hence, a variable factor is preferred because it provides a more logical fit to the measured data. The equation used to estimate total annual mean NO_2 is therefore (see section 5.2.1.3 and Figure 5.3):

$$\mathbf{NO_2\ total = NO_2\ background + NO_2\ road}$$

Where NO_2 road is given by:

$$\mathbf{NO_2\ road = NO_x\ road\ x\ [-0.068ln(NO_x\ total) + 0.53]}$$

This approach has been tested and refined using data for a large number of sites throughout the UK for 1999, 2000 and 2001.

514. An important element of the DMRB revision has been the calibration and adjustment of the method by comparison of predicted annual mean NO_2 concentration with measured concentration using data from 27 roadside monitoring sites between 1998 and 2001. Although such comparisons have been conducted in the past (for example, Mann *et al.*, 2001), the 2002 revision represents the first consideration and incorporation of such results in the development of the method itself.

515. Pollutant concentrations tended to be overestimated by the model. There was found to be a good relationship between the annual average daily traffic flow (AADT), or AADT weighted by distance if more than one road was present, and the overestimation of the contribution of road traffic emissions to pollutant concentrations. The differences between the predicted and measured concentrations were then integrated into the spreadsheet model itself as a series of adjustments. However, although the application of the adjustment factors substantially improved the prediction accuracy of the method at the sites that were considered, a thorough verification of the model at other sites remains a task for the future (Boulter *et al*., 2002a).

5.2.3 Empirical models of NO_x and NO_2 concentrations in London

5.2.3.1 Introduction to the empirical models for London

516. Empirical techniques have been used to predict annual mean concentrations of NO_x and NO_2 in London by the Environmental Research Group at King's College London (ERG). These predictions have been used by the GLA in their development of the Mayor's Air Quality Strategy (GLA, 2002) and for many local authorities in London as part of their responsibilities to review and assess air pollution under the National Air Quality Strategy.

517. The most significant issue affecting predictions of air pollutants in London is the size and complexity of the London conurbation. For this reason, it is not possible to model each source explicitly taking account of hourly to seasonal changes in emissions: it is necessary to make simplifying assumptions. In the ERG model a distinction is made between emissions from the local road network and other sources. The approach uses a dispersion model together with empirical relationships derived from air pollution measurements. The approach aims to address the issue of the partitioning of different source types through a multiple regression using measurements from the LAQN and the AURN. A dispersion model is used to make predictions; the multiple regression with measurements is simply a refinement of the technique. The annual mean concentration of NO_x at a monitoring site is described by a linear regression:

$$NO_x = A.NO_x[roads] + B.NO_x[other] + C$$

Where A, B and C are constants to be derived from the multiple regression, NO_x[roads] is the contribution from the local road network (defined and modelled within 500 m of a monitoring site) and NO_x[other] is the contribution from more distant sources of NO_x i.e. beyond 500 m of a monitoring site.

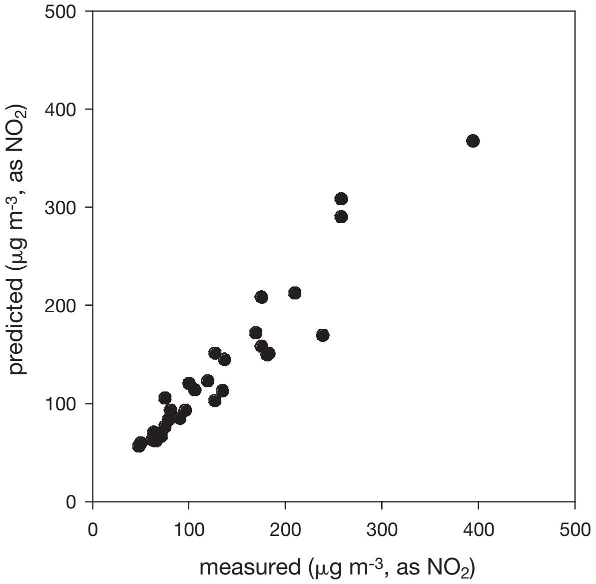
518. Emissions from the road network within 500 m of each monitoring site are modelled in a detailed way. The road network is split up into 10 m sections to take account of the geographic variation in the actual road network. Careful treatment of the road network is necessary because of the steep gradient in concentrations of NO_x close to roads.

519. All other sources of NO_x (except Part A industrial processes, which are modelled explicitly as point sources) and road network emissions greater than 500 m from a monitoring site are represented as shallow volume sources of varying dimension depending on their source characteristics.

520. Emissions from gas combustion (domestic and commercial; the second most important emissions category for NO_x after road transport) are released from a wide range of source heights. Furthermore, the type of building can also influence the release of emissions. This feature of these sources makes it difficult to model them in a consistent way. For example, emissions from these sources are greatest in central London, but they are generally released from a greater height, thus reducing their ground-level impact. At present, there is insufficient information to warrant a more detailed treatment of these sources. However, it is important to recognise that emissions from these sources will be diluted in a very different way to road transport emissions. The regression technique described above aims to use the additional information provided by measurements to assist in the partitioning of ground-level NO_x from these different source types.

521. The effect of the multiple regression is that more emphasis is placed on local road network emissions than would have been the case if each source type were modelled directly. Concentrations of NO_x in London therefore respond very well to changes in the emissions from road transport. A comparison between measured and modelled annual mean NO_x concentrations is shown in Figure 5.11.

Figure 5.11 Comparison between measured and modelled annual mean NO_x concentration in 1999 in London (r_2=0.89).

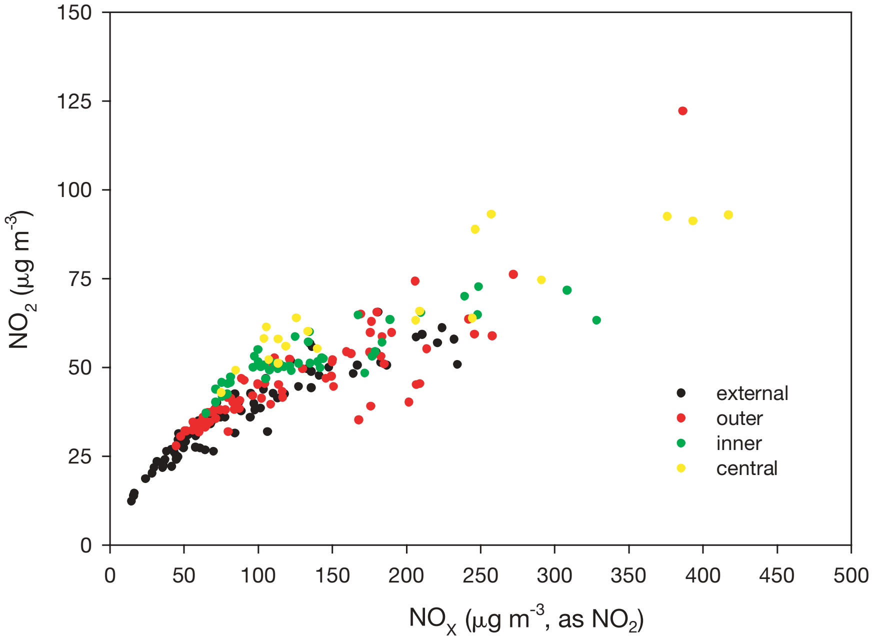


5.2.3.2 *Empirical relationships between NO_2 and NO_x in London*

522. One potential approach to developing a relationship between annual mean NO_x and NO_2 is to plot the annual mean NO_x against the annual mean NO_2, as has been done for most empirical approaches, for example, as used in the national modelling described in section 5.2.1. In the past, it has been difficult to distinguish between different NO_2 environments because of a lack of data. However, in London there is now sufficient monitoring to consider one urban area in isolation. Figure 5.12 shows the annual mean NO_x-NO_2 relationship for sites in and close to London for measurements between 1996-2000. For convenience central is defined as within 5 km of Bloomsbury, inner between 5-10 km, outer 10-20 km and external > 20 km.

523. Figure 5.12 shows higher NO_2 concentrations in central and inner London for a particular value of NO_x. Central London locations receive emissions from all wind directions, thus allowing time for chemistry to occur and emissions to be mixed into incoming trajectories. The other principal difference is between background and roadside/kerbside locations, where the latter always shows a lower NO_2/NO_x ratio for a particular NO_x concentration compared with background locations. Again, this observation reflects the time available for chemistry to occur and the relatively high background NO_2 concentrations that are observed. Figure 5.12 also encompasses the localised effects of higher direct emissions of NO_2, which will affect some locations more than others.

Figure 5.12 Annual mean NO_x vs. NO_2 by location in London (1996-2000).



524. One difficulty with approaches that rely on the relationships between annual mean NO_x and NO_2 is that some assumptions must be made to describe how NO_2 concentrations respond to decreasing concentrations of NO_x. For example, there are no central London sites at low NO_2 concentrations: these have to be interpolated. This interpolation must either include sites in other locations, for example, outer London, or some other guess as to how the curve reaches the origin. The risk is that there is no direct information on which to base future predictions of NO_2 in central London. Specifically, this approach does not provide the concentration of NO_x that must be reached in order that the NO_2 objective is met at a particular site.

525. An alternative approach described below aims to provide some information on how NO_2 might be expected to decline at different sites in London. More information can be found in Carslaw *et al*. (2001).

526. The relationship between hourly NO_x and NO_2 concentrations can be investigated by plotting each hourly value of NO_x against the corresponding NO_2 concentration in a scatter plot, as shown for the Bloomsbury site in Figure 5.13. Derwent and Middleton (1996) found that the relationship between NO_x and NO_2 could be usefully summarised by plotting NO_2 against NO_x in different NO_x 'bins'. The mean NO_2 concentration is averaged according to different ranges of NO_x concentration. Figure 5.14a shows the effect of summarising data in this way for the Bloomsbury site in 1998. However, the precise relationship is always both year- and site-dependent.

527. The NO_x-NO_2 relationship and the frequency distribution shown in Figure 5.14 can be multiplied together and divided by the number of measurements in one year to derive the annual mean NO_2 concentration. The NO_2 concentration derived in this way is identical to the measured annual mean NO_2 concentration. Of more use in a predictive sense, is the calculation of the annual mean NO_2 based on a new concentration of NO_x, for example, the expected annual mean NO_2 concentration for a 50 % reduction in NO_x.

Figure 5.13 The relationship between hourly NO_x and NO_2 concentrations for the Bloomsbury site (1998).

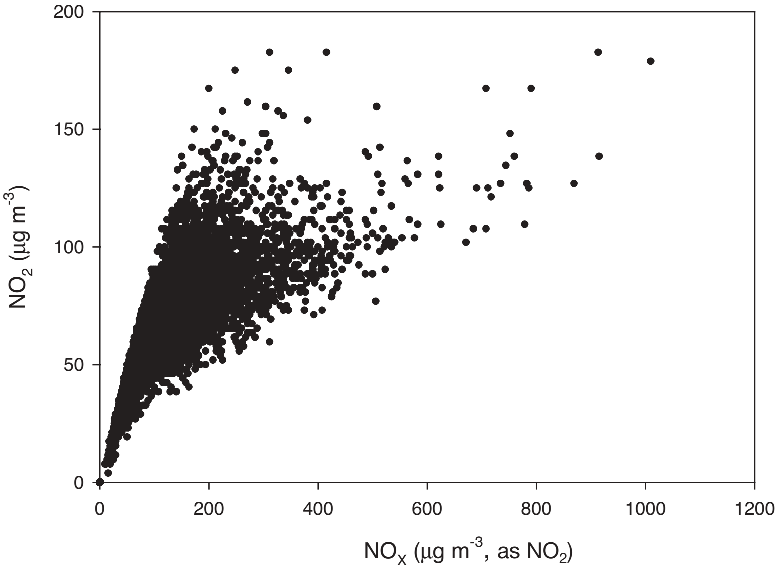
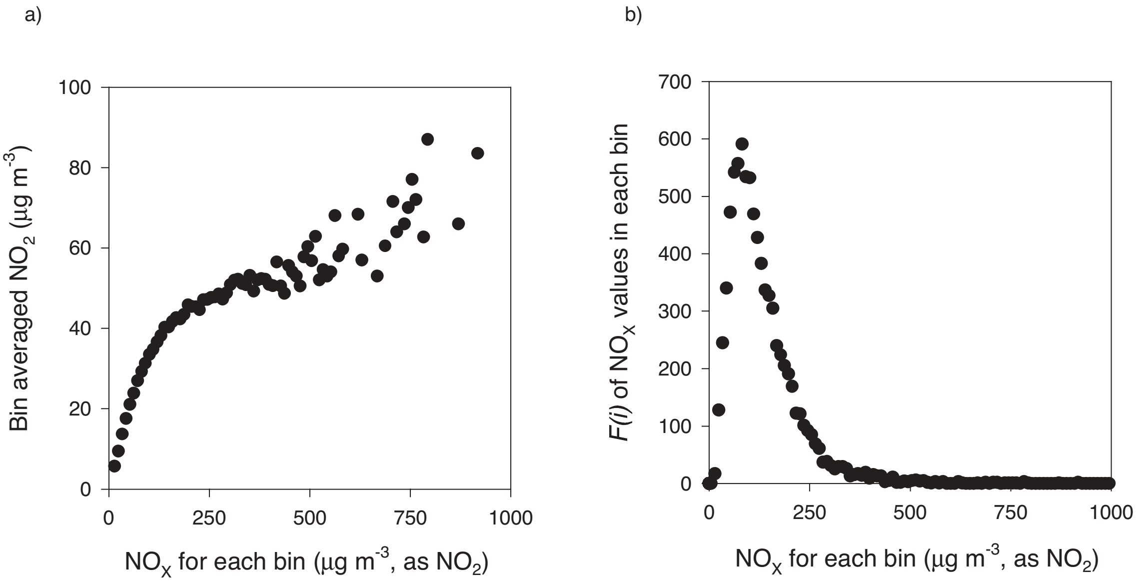
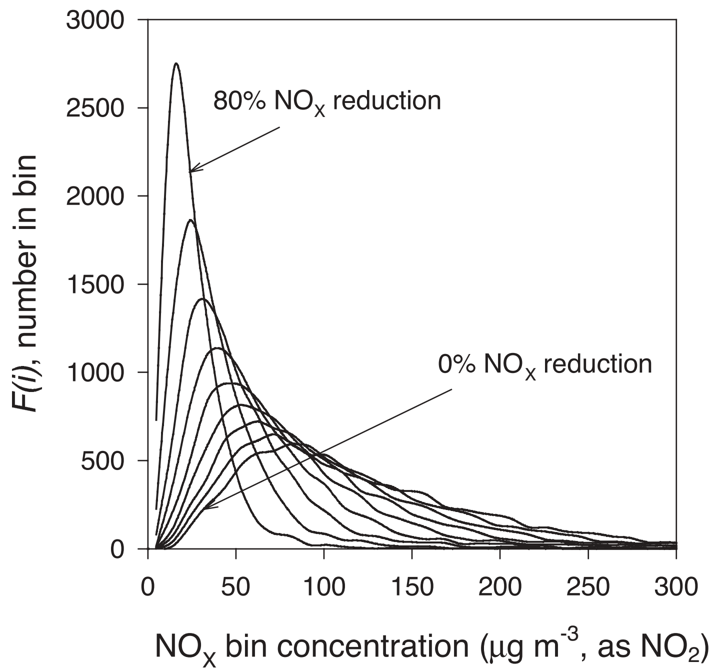


Figure 5.14 (a) The hourly NO_x-NO_2 relationship at Bloomsbury for 1998 showing the bin averaged NO_2 concentrations after sorting into 10 µg m^{-3} NO_x bins and **(b)** the corresponding NO_x frequency distribution F(i) showing the number of points averaged in each bin.



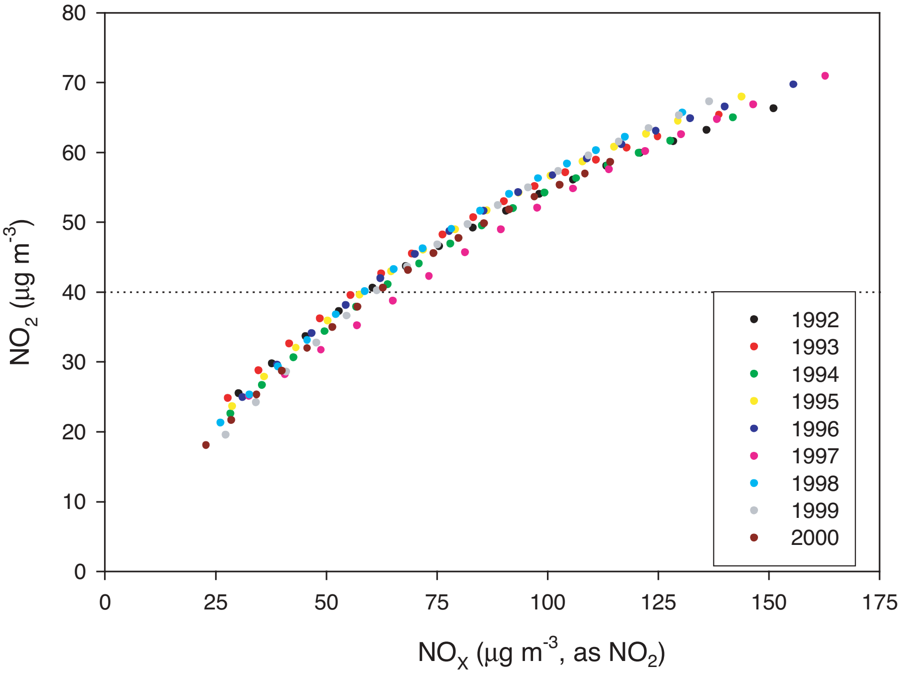
528. Different NO_x reductions can been considered by re-calculating the hourly values of NO_x for different percentage reductions in NO_x from 0 to 80% in 5% intervals, reducing the annual mean NO_x accordingly, and then sorting to find the new frequency distribution. The revised frequency distributions for NO_x are multiplied by the NO_2 vs. NO_x relationship to provide a new estimate of the annual mean NO_2 concentration. Figure 5.15 shows how the frequency distribution changes with reductions in NO_x. The shape of the relationship becomes increasingly narrow and tall as NO_x values migrate to lower concentrations, with increased reductions in NO_x. Second, as the NO_x concentration is reduced an increased number of points move to the NO_x-limited regime.

Figure 5.15 The effect on the NO_x frequency distribution, F(i), at the Bloomsbury site as NO_x concentrations are reduced in 10% intervals from 0 to 80% reduction (1998).



529. By multiplying successive NO_x frequency distributions shown in Figure 5.15 with the NO_x-NO_2 relationship shown in Figure 5.14a and dividing by the number of observations, it is possible to derive an *annual mean* NO_x-NO_2 relationship at a site. This approach indicates how NO_2 concentrations might change as NO_x is reduced at a *single site*. Furthermore, the technique is based on hourly measurements of NO_x and NO_2, which is essential if the change in future NO_2 concentrations is to be predicted.

Figure 5.16 Annual mean NO_x vs. NO_2 (1992-2000) for the Bloomsbury site derived using the ERG approach. The different points for each year show the estimated effect of different % reductions in NO_x.



530. Figure 5.16 shows the results of applying the technique to the Bloomsbury site over 9 different years covering a range of annual mean NO_x and NO_2 concentrations. In general, these derived curves show consistency between the years and indicate that to achieve a NO_2 concentration of 40 µg m^{-3}, NO_x must reduce to approximately 60-70 µg m^{-3} at this site. 1997 stands out as one year where the relationship is slightly different, although the reasons for this are not properly understood (there were a large number of wintertime pollution episodes during 1997). The advantage of the approach is that it provides predicted concentrations of NO_2 given a concentration of NO_x, say from a dispersion model.

5.2.3.3 Advantages and disadvantages of the empirical model for London

531. Many of the advantages and disadvantages of the modelling approach described are the same as those described for the national modelling. The aim of the modelling has been to describe ‘typical’ concentrations expected at different locations. However, it is recognised that actual concentrations depend upon many factors that cannot adequately be described using the techniques described above. These factors include:

- Factors affecting local dispersion that cannot be adequately described using existing dispersion models, for example, locations in a ‘mixed’ setting surrounded by buildings of various heights. Most urban locations do not simply fit into ‘open’ or ‘street canyon’ descriptions. The empirical approach only recognises that concentrations tend to be enhanced in urban locations where many buildings are present compared with open locations.

- Local variation in emissions of NO_x. No account is taken of the variation in emissions along each road link. It would generally be expected that emissions (and therefore concentrations) would be higher close to locations with queuing traffic, for example, near junctions. Again, the techniques provide predictions that represent average or typical conditions.
- Local variation in direct emissions of NO_2. The approach inherently accounts for the local contribution to direct emissions of NO_2. This is both an advantage and a disadvantage. The advantage is that some account is taken of direct emissions, which is useful since emission inventories are not currently detailed enough to account for direct emissions of NO_2. The disadvantage is that the approach does not currently account separately for direct emissions. It is not possible therefore to consider how these emissions might change in future.
- The calculation of future concentrations of NO_2 is dependent on several assumptions, most significantly how future concentrations of NO_2 respond to changes in the concentration of NO_x. The only way that these assumptions can be tested is to compare them with approaches that attempt to account for all or most of the processes controlling concentrations of NO_2, for example, a Lagrangian model (section 5.4). The approach also does not account for changes in future concentrations of tropospheric O_3. The empirical oxidant approach described in section 5.2.4 indicates how O_3 concentrations can be included in empirical models.

5.2.4 Prediction and interpretation of annual mean NO_2 vs. NO_x curves by consideration of oxidant sources and chemistry

532. As described in detail in section 3.2.1, there is a strong chemical coupling between the NO_x species and O_3. Because of this, the consideration of NO, NO_2 and O_3 as a set of species, rather than NO and NO_2 alone, can provide additional information to assist the prediction and interpretation of how the concentration of NO_2 varies with NO_x. Consistent with the definition explained in section 3.2.1, NO_2 and O_3 are collectively considered as oxidant (OX). The method presented here (based on that reported by Clapp and Jenkin, 2001), involves defining (i) how the concentration of OX varies with NO_x and (ii) how the fractional contribution of NO_2 to OX (i.e., NO_2/OX) varies with NO_x. The product of these two quantities yields the dependence of NO_2 on NO_x.

533. Figure 5.17 shows how the annual mean concentrations of OX depend on NO_x for the majority of London AURN sites where the required measurements are available, and for Reading and Harwell (as in section 3.3, graphs presenting data for O_3 and/or OX, exceptionally, make use of the ppb units). The general trend of increasing OX with NO_x, superimposed on a background level, may be interpreted in terms of the 'regional' and 'local' contributions to OX, as described in section 3.3.1. Clearly, however, there are some variations in the magnitudes of these contributions from one site to another. This is further illustrated in Figure 5.18, which shows only the data for five selected sites. In each case an associated regression line is presented, based on an intercept (i.e. regional contribution) fixed at a value of 35.7 ppb, derived from analysis of the entire data set in Figure 5.17. Corresponding regression parameters are presented in Table 5.4, for all the sites in Figure 5.17. The assumption here is that variations in levels of OX from one site to another arise primarily from differences in local input (for example, the fraction of NO_x emitted as NO_2), but that the regional contribution (resulting from the background O_3 level) is the same at all sites in a given region.

Figure 5.17 Variation of annual mean OX mixing ratio with NO_x for London sites where O_3, NO and NO_2 are measured, Reading and Harwell. The number of years of available data vary from one site to another, but all displayed data were obtained during 1992-2000. (adapted from Clapp and Jenkin, 2001).

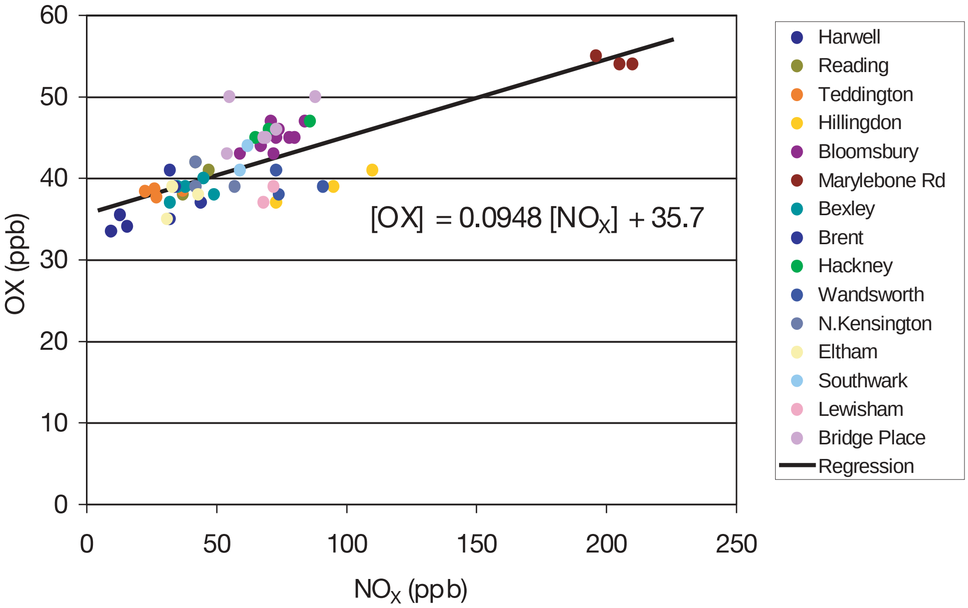


Table 5.4 Summary of [OX] vs [NO_x] and [NO_2]/[OX] vs [NO_x] relationships, and NO_x thresholds derived from the analysis of annual mean data presented in Figures 5.18 and 5.20.

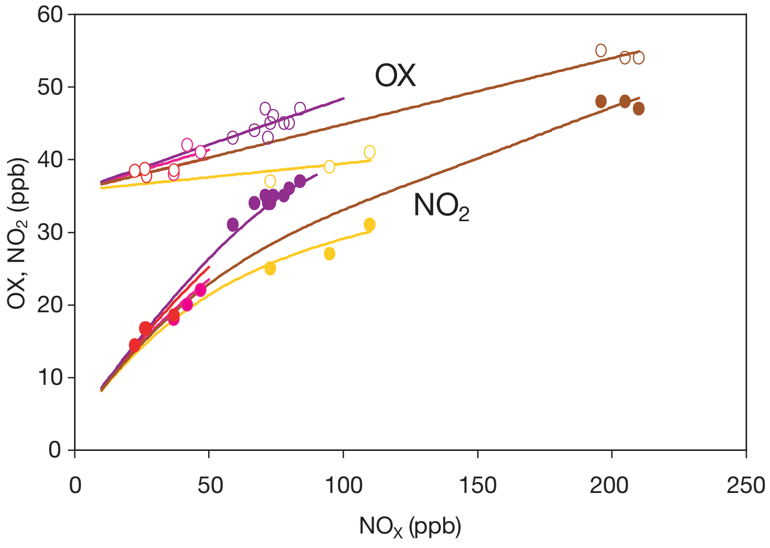
Site	Type	A[1]	[NO_2]/[OX][2]	NO_x threshold[3], μg m^{-3} as NO_2 (ppb)	
				Current method	Carslaw *et al.* (2001)
Bexley	Suburban	0.0771	Fit 1	75.4 (39.4)	–
Bloomsbury	Urban Centre	0.1272	Fit 1	70.6 (36.9)	60.6 (31.7)
Brent	Urban BG	0.0616	Fit 1	77.1 (40.3)	–
Bridge Place	Urban BG	0.1572	Fit 1	68.1 (35.6)	65.6 (34.3)
Eltham	Suburban	0.0477	Fit 1	78.8 (41.2)	–
Hackney	Urban Centre	0.1391	Fit 2	77.3 (40.4)	66.8 (34.9)
Hillingdon	Suburban	0.0375	Fit 2	93.0 (48.6)	100.0 (52.3)
Lewisham	Urban Centre	0.0332	Fit 1	80.7 (42.2)	76.7 (40.1)
Marylebone Rd	Kerbside	0.0914	Fit 2	83.2 (43.5)	71.9 (37.6)
N Kensington	Urban BG	0.0873	Fit 1	74.2 (38.8)	70.6 (36.9)
Reading	Urban BG	0.1121	Fit 2	80.4 (42.0)	–
Southwark	Urban Centre	0.1129	Fit 1	71.8 (37.5)	68.7 (35.9)
Teddington	Urban BG	0.0902	Fit 1	74.1 (38.7)	–
Wandsworth	Urban Centre	0.0449	Fit 2	91.5 (47.8)	89.0 (46.5)

1 Linear [OX] vs [NO_x] relationships given by [OX] = A.[NO_x] + B, derived from data for each site presented in Figure 5.17. The regional OX contribution, B = 35.7 ppb, is assumed site-independent and equivalent to that derived from the composite analysis of all sites.

2 [NO_2]/[OX] vs [NO_x] expressions are polynomial fits to data presented in Figure 5.19:
Fit 1: [NO_2]/[OX] = (1.015×10^{-1}) + (1.367×10^{-2} [NO_x]) – (6.127×10-5 [NO_x]^2) – (4.464×10^{-8} [NO_x]^3): applicable range, 19 – 172 μg m^{-3} NO_x as NO_2 (10 – 90 ppb)
Fit 2: [NO_2]/[OX] = (8.962×10^{-2}) + (1.474×10^{-2} [NO_x]) – (1.290×10^{-4} [NO_x]^2) + (5.527×10^{-7} [NO_x]^3) – (8.906×10^{-10} [NO_x]^4): applicable range, 19 – 402 μg m^{-3} NO_x as NO_2 (10 – 210 ppb).

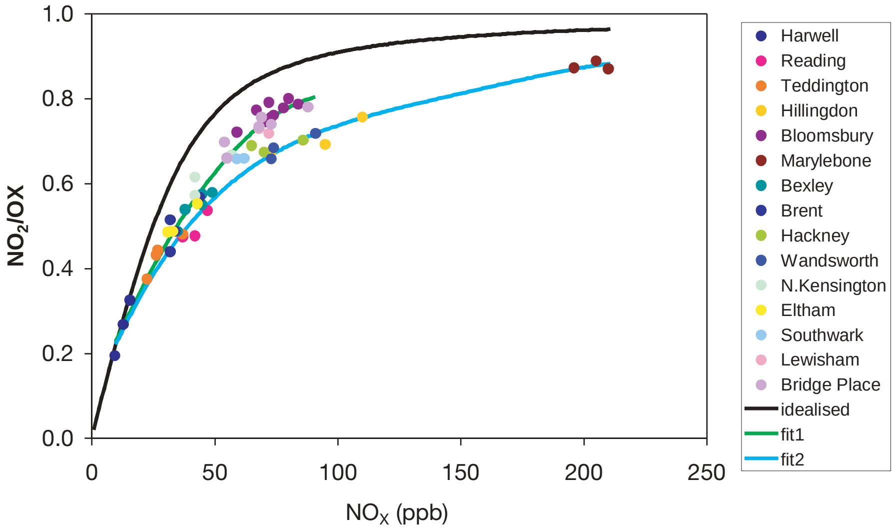
3 Predicted NO_x threshold corresponding to an annual mean NO_2 of 40 μg m^{-3} (21 ppb).

Figure 5.18 Annual mean OX and NO_2 as a function of NO_x at Marylebone Rd (●), Bloomsbury (●), Hillingdon (●), Teddington (●) and Reading (●). Open circles refer to OX, full circles to NO_2. Lines are calculated from the expressions in Table 5.4. (adapted from Clapp and Jenkin, 2001).



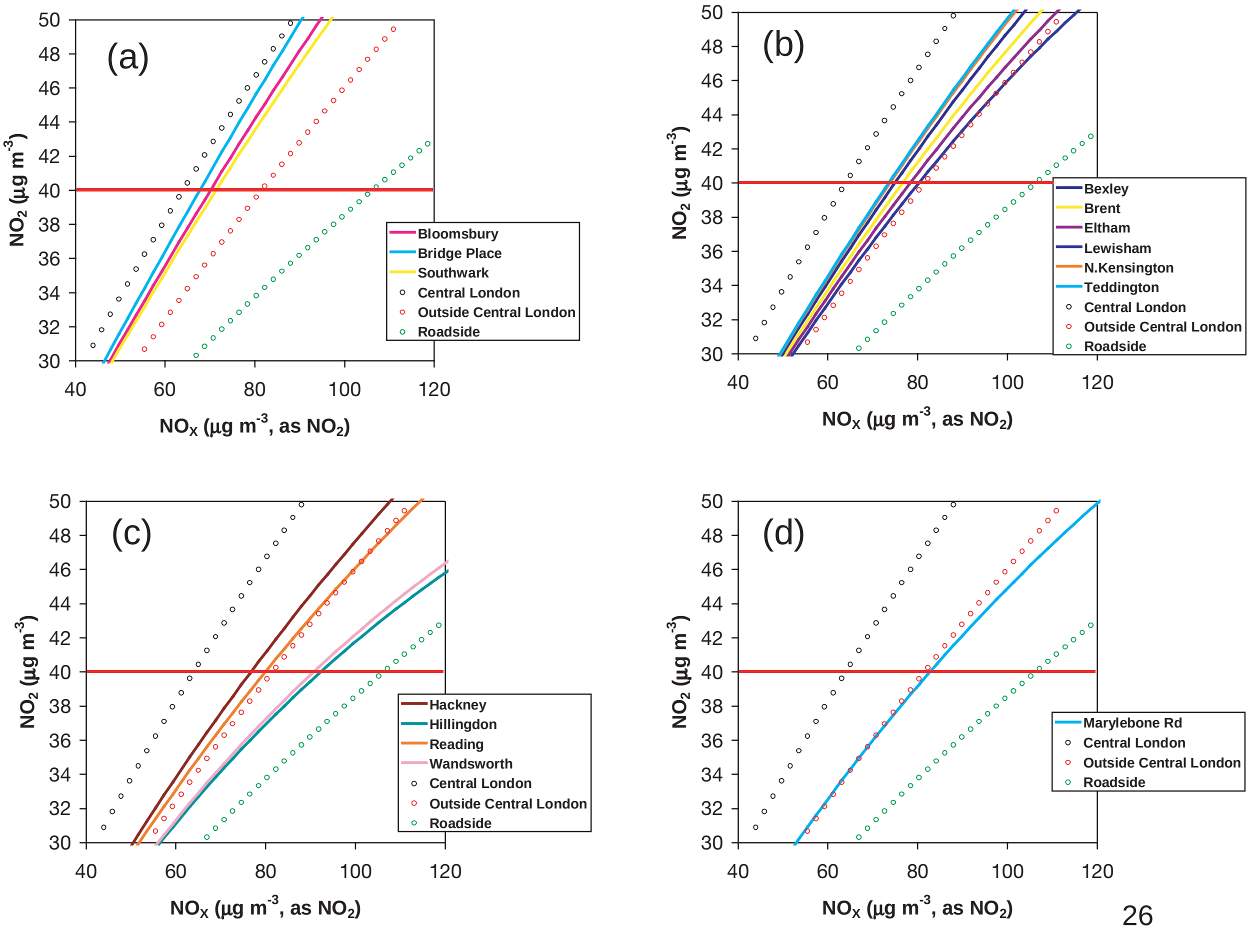
534. Figure 5.19 shows the fraction of OX which is in the form of NO_2, based on the annual average data for the same set of sites. The data appear to fall into two reasonably distinct groups, the first containing Hillingdon, Hackney, Wandsworth, Reading and Marylebone Rd, and the second containing the remaining sites, which generally show higher NO_2/OX ratios. Such differences in the partitioning of NO_2 and O_3 are likely to relate (at least partially) to the time available for reaction of NO with O_3 to occur, following emission of NO. A short time lag between emission and measurement most likely accounts for the lower ratios in the first group identified above. Marylebone Rd is a kerbside site, and the other four sites are located comparatively close to local traffic sources and therefore have partial roadside character.

Figure 5.19 Variation of annual mean $[NO_2]/[OX]$ as a function of NO_x for London sites where O_3, NO and NO_2 are measured, Reading and Harwell. The black line is the calculated idealised variation based on the assumption of photostationary state. (adapted from Clapp and Jenkin, 2001).



535. An idealised variation of NO_2/OX with level of NO_x can be inferred on the basis of the photostationary state relationship (discussed in section 3.2.1). This defines the levels of O_3, NO and NO_2 which co-exist under steady state conditions in a sunlit atmosphere. The variation, calculated with an annual average NO_2 photolysis rate, is also displayed in Figure 5.19. This qualitatively shows the same trend as the observed points, indicating an increasing proportion of OX in the form of NO_2 as NO_x increases.

Figure 5.20 Inferred variation of annual mean NO_2 with annual mean NO_x at (a) central London sites, (b) sites outside central London, (c) intermediate sites and (d) roadside sites, based on the parameters given in Table 5.4. In each panel, the generic dependences of Stedman *et al*. (2002) for urban centre and background sites in central London and outside central London are also shown. A generic dependence for roadside sites used previously (Stedman *et al*., 2001a) is shown for comparison.



536. It cannot, however, provide a quantitative description for annual mean data, for two main reasons. First, the idealised dependence describes how the partitioning of the OX components varies for a range of unique levels of NO_x, whereas each observed data point represents the mean of a large number of measurements at many NO_x levels. If a number of discrete measurements describe a curve of the form shown in Figure 5.19, then the mean of the measurements must lie below the curve. Secondly, the photostationary state relationship assumes there is sufficient time for steady state to be established. As indicated above, this is less likely at sites closer to source and cannot generally be achieved at night. As a result, the idealised curve overpredicts the proportion of OX in the form of NO_2 on an annual mean basis.

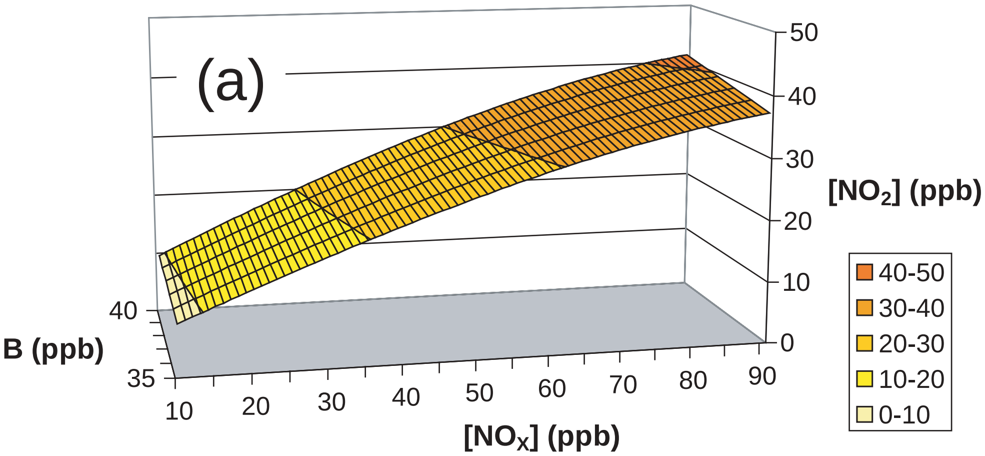
537. The NO_2/OX variation is therefore defined using fitted functions for groups of sites with particular characteristics. The data for the two groups shown in Figure 5.19 yield the two polynomial expressions given in Table 5.4, and illustrated in the figure. In conjunction with the OX vs NO_x regression parameters, these expressions allow NO_2 vs NO_x curves to be inferred for all 14 urban and suburban sites. These are presented with the observed annual mean data in Figure 5.18 for five selected sites.

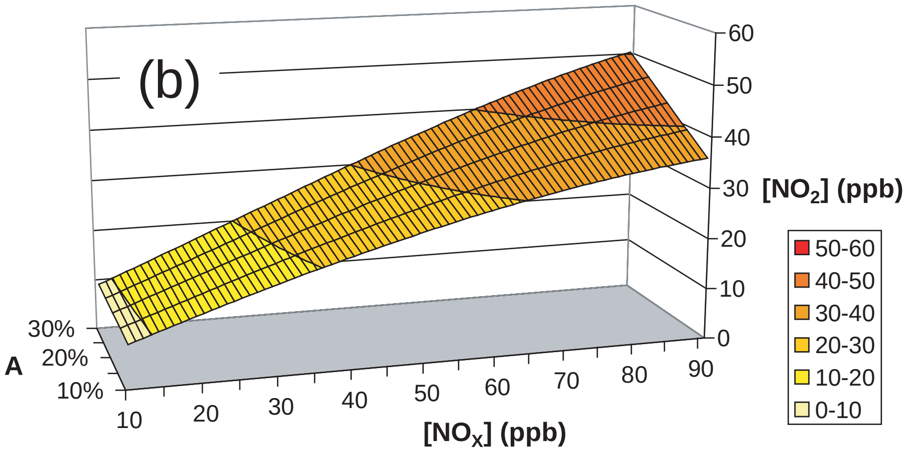
538. The calculated variation of annual mean NO_2 with NO_x is also shown for all 14 sites in Figure 5.20, and the inferred NO_x thresholds which correspond to an annual mean NO_2 concentration of 40 µg m^{-3} (21 ppb) are also presented in Table 5.4. The NO_x thresholds for the sites considered show significant site-to-site variation, covering the range 35.6 – 48.6 ppb (i.e., 68.1 – 93.0 µg m^{-3} as NO_2). The results are thus broadly consistent with threshold values derived from the NO_2 vs. NO_x relationships discussed in the previous subsections (Carslaw *et al*., 2001; Stedman *et al*., 2002). Those reported by Carslaw *et al*. (2001) for specific sites generally lie slightly below the present values, but in reasonable agreement, as also shown in Table 5.4. Stedman *et al*. (2001a) and Stedman *et al*. (2002) derived three generic expressions to describe the NO_2 vs. NO_x relationship for 'central London sites', for 'sites outside central London', and for 'roadside sites', which are compared with the present expressions in Figure 5.20. The present data for urban centre and background central London sites and sites outside central London are consistent with the expressions of Stedman *et al*. (2002).

539. As discussed above, a number of sites can be classified as 'intermediate', as they have partial roadside character, due to the influence of nearby roads. Logically, therefore, the present data for some of these lie at higher NO_x than the generic expressions for the urban centre and background sites, but lower than the generic expression for roadside sites. The present data for Marylebone Rd (the only roadside site available for consideration by the analysis method) lie at significantly lower NO_x than the generic expression of Stedman *et al*. (2001a). Although the NO_2-NO_x dependence provides a good description of the variation of the currently available narrow range, some systematic error may result from the necessary inclusion of Marylebone Rd in the intermediate grouping in the present analysis.

540. This analysis suggests that consideration of NO, NO_2 and O_3 as a set of chemically-coupled species allows variation of (oxidant in the form of) NO_2 as a function of NO_x to be explained on the basis of sources of oxidant and well-understood chemical processes. This allows new insights into the origins of site-to-site variations of NO_2 vs. NO_x curves, by decoupling differences which are due to variations in the time available for chemical processes (for example, the reaction of NO with O_3) from those which are due to variations in the sources of oxidant (OX), for example, the fractional contribution of primary NO_2. The methodology potentially enables predictions that take account of future changes in regional oxidant (i.e. the background O_3 level), or local oxidant inputs which might arise, for example, from modifications in vehicle emissions control technologies or fleet composition.

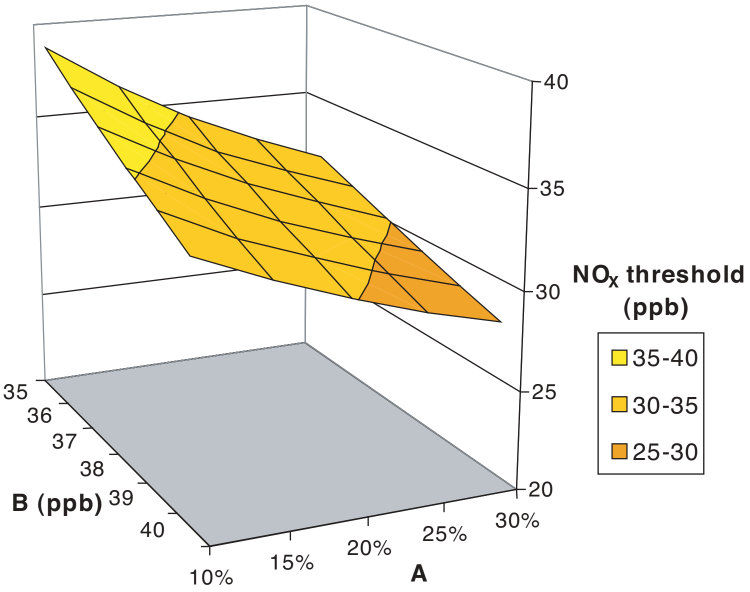
Figure 5.21 The calculated influence of changing the regional background oxidant level (B) and the local oxidant contribution (A) on the variation of NO_2 with NO_x at London Bloomsbury. Consistent with analysis in Table 5.4, variation with changing B carried out with A = 12.72%; variation of A carried out with B = 35.7 ppb.





541. This is illustrated in Figure 5.21, using the London Bloomsbury site as an example. The figure shows the influence of varying the regional background oxidant level (B), and the local oxidant contribution (A) on the calculated NO_2 vs. NO_x curves. In particular, these graphs show that increasing either A or B leads to greater NO_2 concentrations for a given level of NO_x, such that the NO_x threshold corresponding to an annual mean NO_2 concentration of 40 µg m^{-3} (21 ppb) decreases. This is illustrated further in Figure 5.22, for combinations of B (in the range 35-40 ppb) and A (in the range 10-30% of NO_x). The present day reference conditions for Bloomsbury (A = 12.7%; B = 35.7 ppb) correspond to a point close to the high corner of the NO_x surface. The lowering of the NO_x threshold if and when either A or B (or both) are increased is indicative of the additional NO_x reduction that would be required to achieve the annual mean NO_2 concentration of 40 µg m^{-3} (21 ppb).

Figure 5.22 The calculated influence of changing the regional background oxidant level (B) and the local oxidant contribution (A) on the NO_x threshold corresponding to 21 ppb (40 µg m^{-3}) NO_2 at London Bloomsbury.



542. Although this methodology allows the influence of these factors to be considered, it should be noted that it is limited to sites where measurements of O_3 and NO_x are co-located. However, it is possible that the NO_2 vs. NO_x dependence at additional sites possessing similar characteristics (for example, local traffic flows and vehicle fleet composition) may be adequately represented by inference.

5.3 Dispersion and small-scale models

5.3.1 Introduction

543. The class of 'Gaussian-type' models covers a range of model types which are in widespread use across the UK and elsewhere. The main advantage of this general model type is that computer codes can be designed to be relatively straightforward to use by practitioners and they can also be constructed in a way that allows the impact of different physical/chemical factors to be readily understood.

5.3.2 Dispersion processes

544. The first such models of this type were constructed to calculate concentrations of a pollutant emitted from a single point source but the general approach is easily extendable to line, area and volume sources and also to multiple sources. The basic Gaussian formulation for concentration *C* due to a pollutant emission rate *Q* in windspeed *U* is derived as a solution of the diffusion equation as

$$C(x,y,z)=\frac{Q}{2\pi U\sigma_y\sigma_z}\exp\left\{-\frac{1}{2}\left(\frac{(y-y_s)^2}{\sigma_y^2}+\frac{(z-z_s)^2}{\sigma_z^2}\right)\right\} \quad (1)$$

α_y and σ_z are the lateral and vertical standard deviations of the concentration distribution and are therefore a measure of its spread or cross section of the plume.

545. Strictly the expression (1) for the concentration distribution is only valid in a turbulent flow if it is assumed that turbulent diffusion can be represented by a constant turbulent diffusivity. However, it has been applied successfully in the atmospheric boundary layer where there are rapid changes in the diffusivity especially with height. The expression is also valid only if *C* represents the mean concentration, that is the concentration averaged over times greater than the turbulent time scales but less than that associated with changing meteorology; typically it would be an hourly average concentration. For shorter averaging times the concentration will vary significantly from one averaging period to the next because of the random movement of the plume by turbulence and because of in-plume structure which occurs as the plume mixes with ambient air. Expressions similar to (1) have formed the basis both for discussions of dispersion and of many dispersion models. Referring to (2) we see that it describes the mean concentration in terms of the vertical and horizontal plume spreads, the mean wind speed, and a distribution about a plume centreline (y_s, z_s) at downstream distance x. Since the plume spread and the location of the plume centreline depend on the mean and turbulent wind field the expression provides a mechanism by which concentrations can be approximately quantified in terms of the atmospheric boundary layer variables.

546. In early dispersion models (for example, Pasquill and Smith, 1983; R91, ISC) the boundary layer structure was parameterised in terms of a single parameter, the Pasquill stability category, which typically varied from Category A (very unstable) through to D (neutral) to F or G (very stable). The plume spread parameters (z and y) are then determined analytically as functions of stability category and distance downstream. More recent research has shown that one parameter is not sufficient to describe turbulence mixing satisfactorily since both mean flow and turbulence and hence dispersion vary significantly with plume height in the boundary layer. Thus more recent and advanced models (for example, OML, ADMS, AERMOD) parameterise the boundary layer structure in terms of two length scales, the boundary layer height (*h*) and the Monin Obukhov length (L_{MO}) a length scale dependent on mechanical mixing and the surface heat flux. Some models also employ more complex non-Gaussian formulae than (1) to describe the concentration profile normal to the ground. The use of these more physically based parameters (h, L_{MO}) rather than Pasquill categories, also has the advantage that other effects on dispersion such as those of complex terrain can be treated within the model in a consistent manner since these effects can also be described in terms of the boundary layer variables.

547. Many validation studies have been performed on dispersion models for point sources (for example, Hanna et al., 2001). These generally show the better performance of the models based on *h* and L_{MO}.

5.3.3 Practical dispersion models for calculating NO_2 concentrations

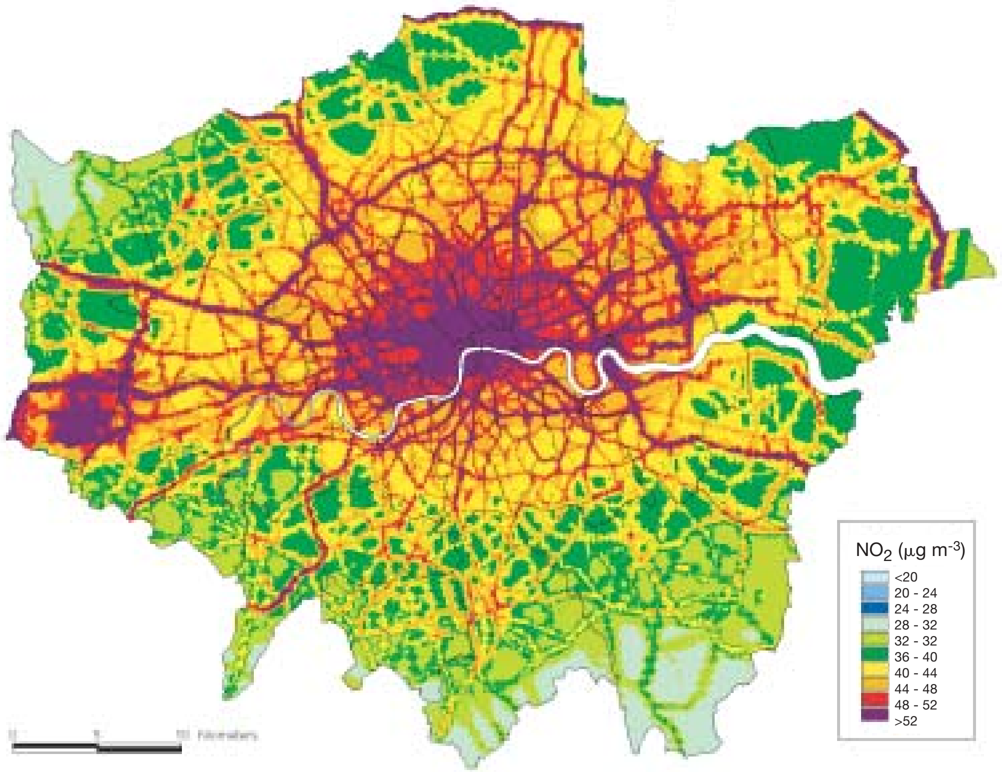
548. Most practical dispersion models have features additional to those required for the dispersion of pollutants emitted from point sources. These include extensions to line source models (analytical or numerical integration of point source models) and sometimes area or volume sources, allowance for deposition and also inclusion of chemical routines. As with the basic formulation for α_z, σ_y, levels of sophistication employed for additional source types vary greatly with, for example, some models employing simple ‘virtual’ upstream point sources to represent line or volume sources (for example, ISC, AERMOD) whilst others describe dispersion from elements within the source (for example, CALINE, ADMS). Models for dispersion from roads include different formulations for the impacts of street canyons or vehicle-induced turbulence.

549. Models also include chemical schemes of very different levels of sophistication dependent on the intended model application. These range from no scheme, to empirical formulae, to the simple chemical reaction schemes, for example the Generic Reaction Set (GRS) used in ADMS, through to more advanced schemes such as CBM IV used in UAM.

550. An analysis, using ADMS 3, of dispersion from point sources (Carruthers *et al.*, 2002) has shown that the chemical scheme employed can have a large influence on both peak and annual NO_2 concentrations. For instance, the O_3 limiting method which assumes all available O_3 reacts with NO_x may result in much higher concentrations of NO_2 than the GRS scheme which allows for reaction times and photodissociation of NO_2. In this study concentrations were also sensitive to the background concentration and the year of the meteorological data.

551. The most advanced models, ADMS-Urban and AIRVIRO, can be used to calculate dispersion at high resolution over large cities. These systems include allowance for a large number of sources of each type, chemical reaction schemes and other advanced options (for example, ADMS-Urban nests the Gaussian-type model with a trajectory model, AIRVIRO offers an Eulerian flow and diffusion scheme as an alternative to a Gaussian model). These models also include emissions database and analysis facilities for manipulating emissions data and calculating emissions from raw activity data. They also usually include GIS functionality for input and display of model output. An example of the annual average NO_2 concentration across London calculated using ADMS-Urban is shown in Figure 5.23.

Figure 5.23 Annual average NO_2 for 1999 across London calculated using ADMS-Urban.



5.3.4 Applications and limitations of dispersion models in urban areas

552. The basic Gaussian plume formula (1) assumes that the turbulent flow is stationary, that is, the statistics of the boundary layer flow are assumed to remain constant over time. This means that the formulation is less effective when there are rapid changes in local weather conditions; it also limits the spatial range from sources for which the models might be expected to perform well, with the upper limit typically set at 30-50 km. Puff models or the nesting of Gaussian-type models within trajectory or Eulerian models are methods for overcoming these limitations.

553. As discussed in Chapter 3, in urban areas the local mean flow and turbulence may change over small spatial scales due to the impact of building structure and local heating effects etc. In practical models such effects can be included, for example with variable surface roughness and/or street canyon models, although such parameterisations cannot describe the precise details of flow and dispersion and there is inevitably a degree of approximation at small spatial scales. The most commonly used practical street canyon model (OSPM, Berkowicz *et al*., 1997) is widely used as a basis for many dispersion models of road traffic emissions in built up areas. This OSPM approach is based on the idealised model of a street as a long road enclosed along its length on both sides by buildings of equal height. The wind passing across the street induces a vortex or recirculating region in the street (Figure 5.24). As the canyon increases in width the vortex is confined to the leeward side of the canyon. The concentration consists of two components, the first resulting from direct emissions that contribute across the whole canyon and represented by a line source formulation, the second of pollutant recirculating in the vortex that of course only contributes in the vortex. OSPM requires only simple data from the user on the street dimensions, and wind strength and direction. The model parameterisation has been optimised and tested against experimental data collected in wind tunnel studies and in field campaigns in long uniform streets. Under these circumstances these models perform quite well.

Figure 5.24 Example of the capability of computational fluid dynamics to model pollutant concentrations from vehicle exhaust – two-dimensional simulation of street canyon circulation in steady, horizontal incident flow.

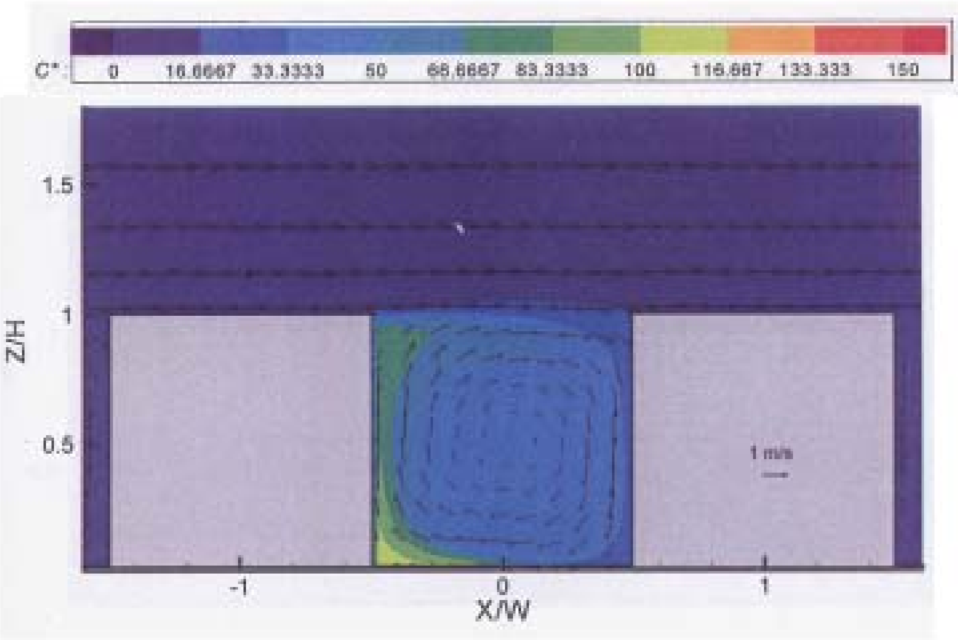
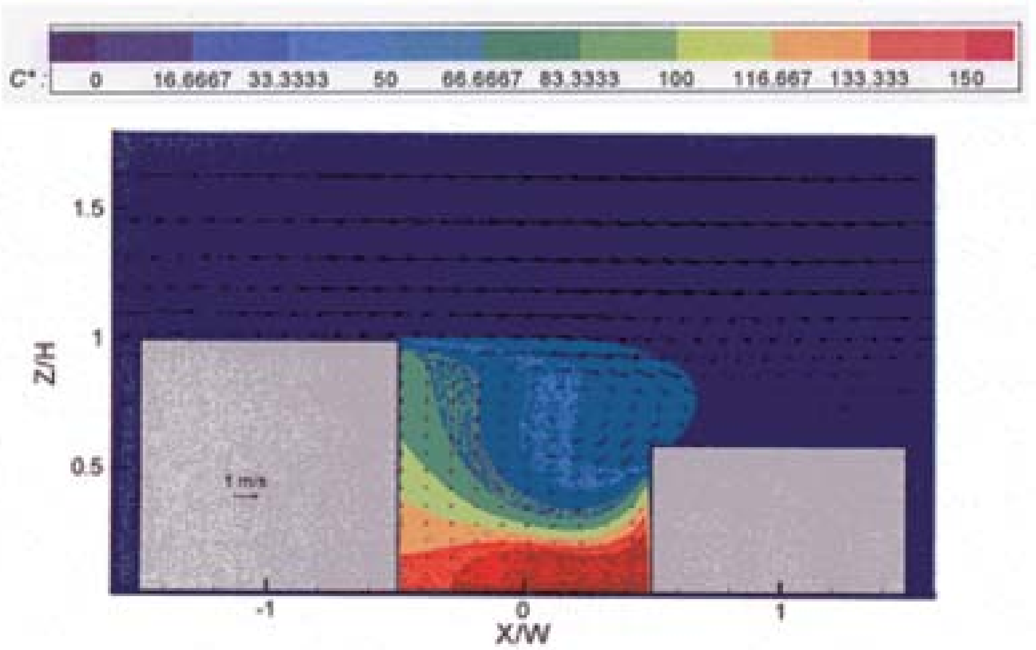


Figure 5.25 Example of the capability of computational fluid dynamics to model pollutant concentrations from vehicle exhaust – asymmetrical canyon, showing marked influence of asymmetry when compared with Figure 5.24.



554. However, most streets do not conform to these simple idealised forms, with buildings of uniform height along them. They also have intersections and cross-roads with air moving between streets as well as exchanging with air above the urban canopy. The effect of introducing asymmetry in the building heights of a street canyon is illustrated in Figure 5.25, where the vortex has been displaced and distorted, changing the ventilation potential of the street, and in this case enhancing. The 3-dimensional dispersion at, for example, a cross-roads, is even more complex- as illustrated in the work of Scaperdas *et al*. (2000). Here an analysis of the intersection between Marylebone Road and Gloucester Place explained a lot of the observed variability observed at the local monitoring station, but wind tunnel experiments and CFD studies illustrated the sensitivity to slight asymmetries and building configurations.

555. The above illustrations are based on more complex computational fluid dynamics (CFD) modelling solving the detailed equations describing the physical flow and turbulence. Several codes of this type are available, and can treat three dimensional flows round quite complex building and street geometries, but they are computationally very demanding, and require very detailed input. They are also more difficult to interpret. For these reasons and because the evidence of validation (for example, Carruthers *et al*., 2003) is that approximations used in practical models can be sufficiently accurate for air quality assessments and policy analysis, practical models are routinely used for complex urban environments.

5.3.5 Validation and verification of dispersion models and intermodel comparison

556. It is important to distinguish between model validation and verification. Model validation generally refers to detailed peer-reviewed studies that have been carried out by the model developer or regulatory agency (for example, Environment Agency or USEPA). Models in general use in the UK have been subject to these detailed and documented validation trials.

557. Model verification generally refers to checks on the model performance that are carried out at a local level or application to a study. Such verification studies may result in adjusting input parameters to improve fit with measured data (for example, surface roughness, MO length etc), by improvements to the emissions data, or by selection of the meteorological data.

558. Comprehensive validation and verification is a prerequisite for using models to predict air pollutant concentrations. This section describes validation of ADMS-Urban, a Gaussian-type model which has been used for and on behalf of more than 80 UK local authorities and is currently being used for modelling studies of the major UK conurbations. This process has included validation in London and also comparisons with two empirical models, the national empirical model (Section 5.2.1) and the ERG model (Section 5.2.3).

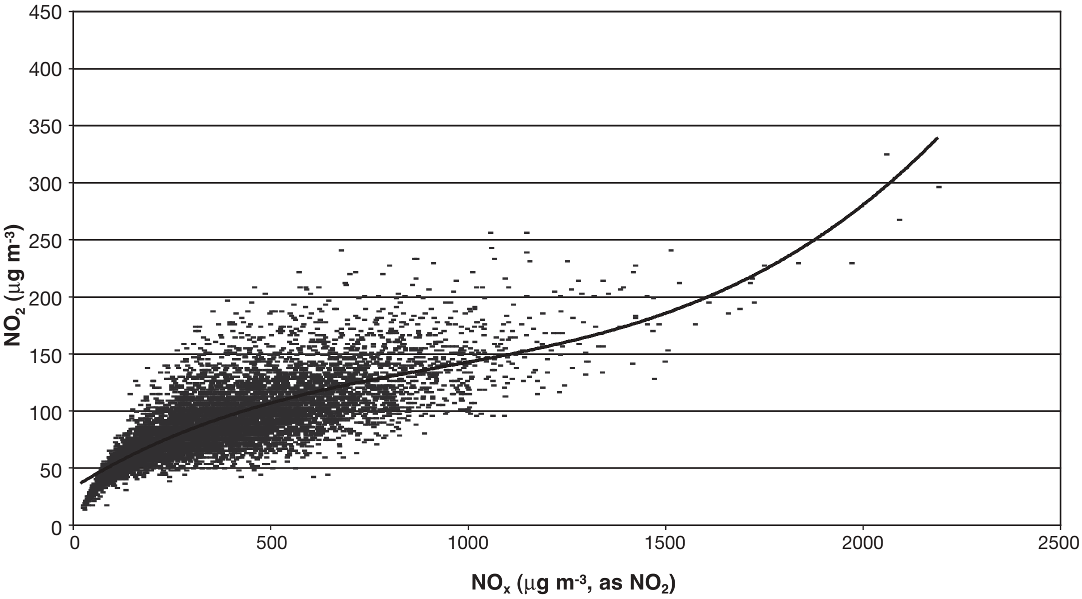
559. The validation study utilised the latest emission inventory for London which is for 1999, meteorological data recorded at Heathrow airport, and rural background pollution for NO_x and O_3 from sites at Harwell, Rochester, Wicken Fen and Lullington Heath. A detailed validation study including model performance statistics derived from the predicted hour by hour concentrations was conducted using hourly data from the 24 AURN sites across London. Table 5.5 summarises the validation of annual mean concentrations for NO_x and NO_2 calculated from NO_x using the GRS scheme. These show the generally good agreement between model and data with all comparisons showing appreciably less than 30% divergence as specified in the Air Quality Daughter Directive. The overall fractional bias showed a 2% underprediction. The model was also able to broadly reproduce the dependence of NO_2 on the NO_x concentration at the monitoring sites; see for example Figure 5.26.

Table 5.5 Monitored and calculated (ADMS-Urban) average NO_2 and NO_x concentrations at London sites (1999).

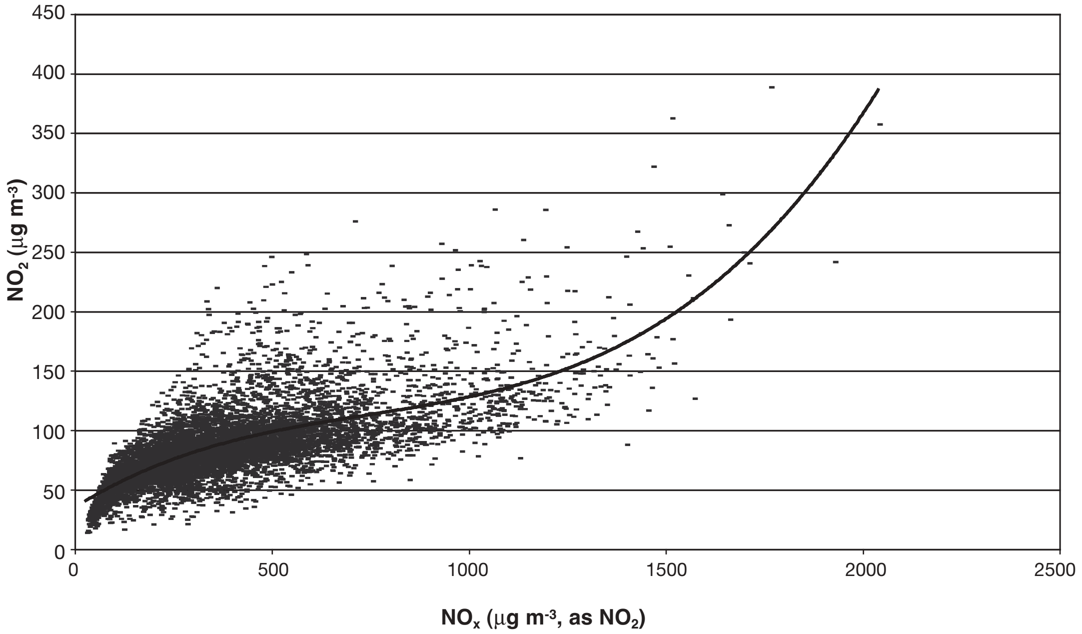
	Annual average NO_x concentrations (μg m^{-3})		Annual average NO_2 concentrations (μg m^{-3})	
	Monitored	Calculated	Monitored	Calculated
A3	257	230	58	67
Camden	211	205	66	71
Cromwell Road	257	260	93	76
Haringey	136	115	51	55
Hounslow	192	132	60	53
Marylebone Road	391	385	91	88
Southwark roadside	228	186	75	67
Sutton roadside	117	77	44	42
Tower Hamlets	241	193	70	71
Roadside mean	**220**	**189**	**68**	**66**
Bexley	69	79	37	40
Bloomsbury	136	119	67	57
Brent	67	77	37	44
Bridge Place	105	109	63	53
Eltham suburban	65	86	36	44
Hackney	136	113	60	55
Hillingdon	167	207	50	63
Lewisham	140	117	54	55
North Kensington	82	101	46	52
Southwark urban centre	119	100	56	50
Sutton suburban	65	67	35	38
Teddington	52	61	32	34
Wandsworth	142	128	52	59
West London	100	92	55	50
Background mean	**103**	**103**	**49**	**50**
Overall mean	**149**	**138**	**56**	**56**

Figure 5.26 Scatter plots of (a) monitored and (b) calculated hourly average NO_x and NO_2 concentrations for Marylebone Road (1999).

(a) Monitored



(b) ADMS-Urban



560. The comparisons show generally good performance of models. However there are significant differences, in particular ADMS shows a greater ratio of NO_2 to NO_x especially for 2005 projections; this results in a greater number of roads showing exceedence of 40 µg m^{-3} in that year, see Table 5.6.

Table 5.6 Comparisons of annual average NO_2 concentrations for 1999 ($\mu g\ m^{-3}$) calculated using ADMS-Urban and the NETCEN and ERG methodologies.

		Measured Value	ADMS-Urban	NETCEN NAEI Background	NETCEN NAEI Roadside	NETCEN Site Specific	ERG
Roadside monitoring sites	A3 roadside	58	67	41	–	–	61
	Camden roadside	66	71	46	69	66	63
	Cromwell roadside	93	76	54	70	92	73
	Haringey roadside	51	55	45	49	48	50
	Hounslow roadside	60	53	44	60	–	–
	Marylebone roadside	91	88	59	83	92	80
	Southwark roadside	75	67	51	66	–	–
	Sutton roadside	44	42	39	–	44	–
	Tower Hamlets roadside	70	71	48	58	67	61
	All roadside sites mean	**68**	**66**	**–**	**–**	**–**	**–**
	ERG roadside sites mean	**72**	**71**	**–**	**–**	**–**	**65**
Background monitoring sites	Bexley suburban	37	40	37	–	–	40
	Bloomsbury urban centre	67	57	63	–	67	65
	Brent urban background	37	44	42	–	–	36
	Bridge Place urban background	63	53	57	–	–	57
	Eltham suburban	36	44	41	–	–	40
	Hackney urban centre	60	55	46	–	–	53
	Hillingdon suburban	50	63	45	–	–	55
	Lewisham urban centre	54	55	43	–	–	–
	N Kensington urban background	46	52	48	–	–	48
	Southwark urban centre	56	50	47	–	–	61
	Sutton suburban	35	38	39	–	–	–
	Teddington urban background	32	34	40	–	–	36
	Wandsworth urban centre	52	59	45	–	–	–
	West London urban background	55	50	47	–	55	57
	All background sites mean	**49**	**50**	**46**	**–**	**–**	**–**
	ERG background sites mean	**49**	**49**	**47**	**–**	**–**	**50**
Roadside and backgroundM monitoring sites	**All AURN sites mean**	**56**	**56**	**46**	**–**	**–**	**–**
	Combined NAEI roadside and background sites mean	**56**	**56**	**52**			
	All ERG AURN sites mean	**57**	**57**	**–**	**–**	**–**	**55**

561. A sensitivity analysis of model performance to input parameters showed relatively little sensitivity to model set-up parameters (minimum Monin Obukhov length, gridded emission source height, surface roughness) but greater sensitivity to changes in the emission inventory, the met data site and the meteorological year – the latter mainly impacted on concentration through significant changes in the pollutant background concentrations.

562. Comparisons between ADMS-Urban, National Empirical Model and ERG methodology have also been conducted at sites across London for both current (1999) emissions and for future projections. Table 5.6 and Figure 5.27 give such a comparison of annual mean NO_2 for 1999 emissions at both roadside and urban background sites. Table 5.7 presents percentage of road segments exceeding a range of defined concentration thresholds of NO_x and NO_2, and finally Figure 5.28 presents scatter plots of comparisons of NO_2 concentrations on road segments calculated by ADMS and ERG methodologies. As detailed in sections 5.2.3 and 5.3, the ERG and ADMS methodologies are able to calculate concentrations at defined receptor points whereas the NETCEN methodology, section 5.2.1, calculates a simple representative value for concentrations close to the road. This is used for both receptor point concentration and concentrations representative of the road segment. Thus care is required in the interpretation of the comparisons.

Figure 5.27 Comparison of measured 1999 annual average pollutant concentrations with values predicted at AURN Sites by ADMS-Urban, NETCEN and ERG for (a) NO_x, (b) NO_2.

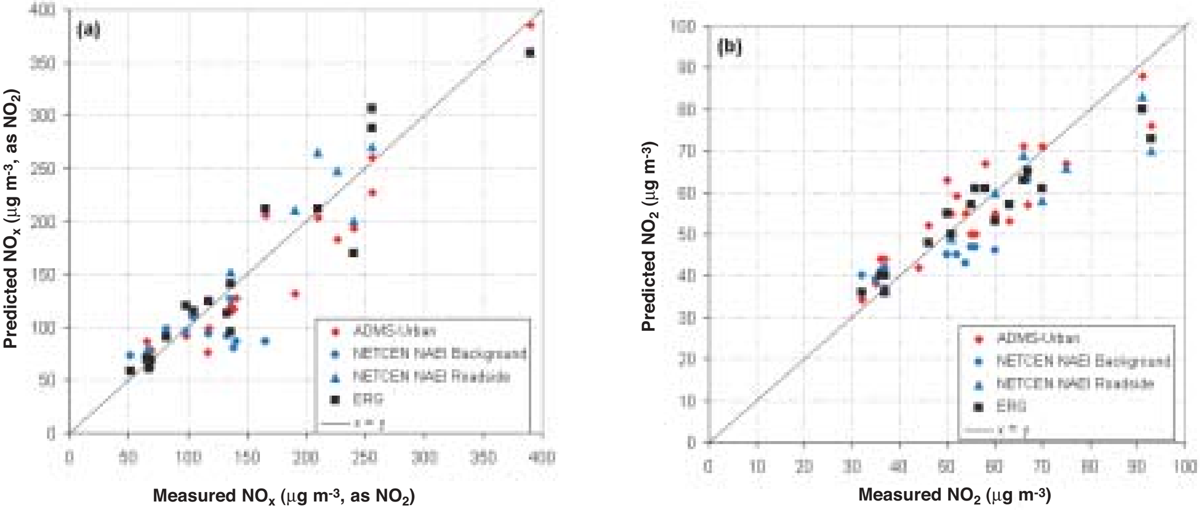
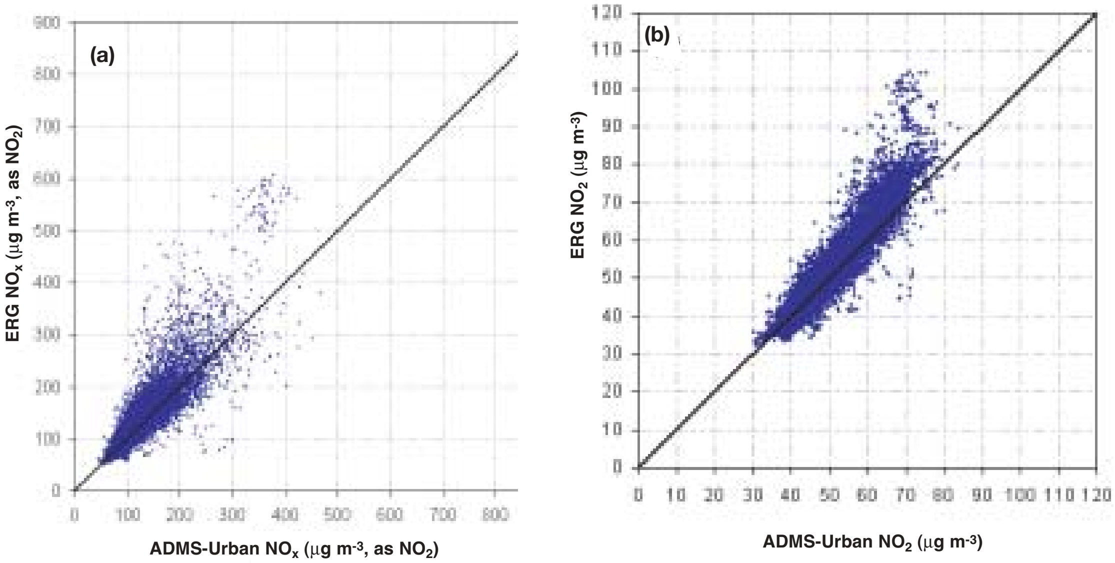


Figure 5.28 Comparison of annual average NO_x and (b) NO_2 concentrations predicted on road links between ADMS – Urban and ERG for 1999.



563. The comparisons between models and data show generally good performance of models. However there are differences, in particular ADMS shows a greater ratio of NO_2 to NO_x and somewhat higher NO_2 concentrations at roadside; this results in a greater number of roads showing exceedence of 40 µg m^{-3} in that year, see Table 5.7. Further comparisons of the three methodologies are presented in Chapter 7. These show divergence of model predictions for future years and significant differences in the predicted areas of exceedence of the NO_2 annual limit value.

Table 5.7 Percentage road length exceeding specified annual mean values.

	ADMS-Urban		ERG
	1999	2004/2005	1999
NO_x >30 µg m^{-3}	100	100	100
NO_x >40 µg m^{-3}	100	98	100
NO_x >50 µg m^{-3}	99	83	100
NO_x >60 µg m^{-3}	90	56	98
NO_2 >20 µg m^{-3}	100	100	100
NO_2 >30 µg m^{-3}	100	97	100
NO_2 >40 µg m^{-3}	87	59	84
NO_2 >50 µg m^{-3}	39	16	42

5.4 Applications of Lagrangian trajectory models

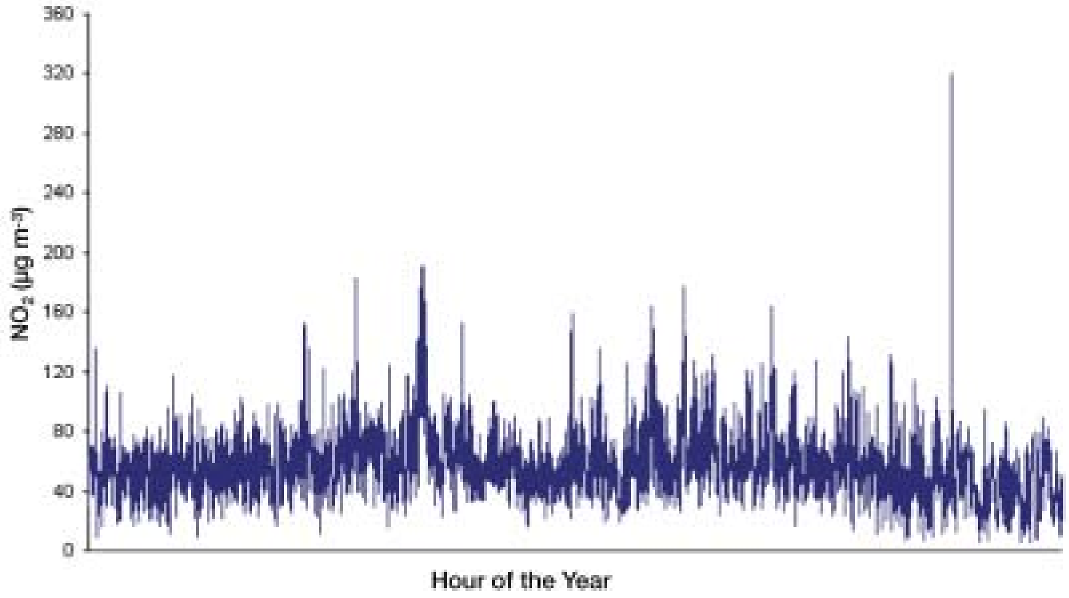
5.4.1 The LRCTM Model for NO_2 in London

564. The London Routine Column Trajectory Model LRCTM (Derwent, 1999) adopts a Lagrangian trajectory approach in which a column of air is advected into an arrival point within the London conurbation. The column of air is divided into 21 layers in the vertical and is advected along a 6-hour trajectory using sequential meteorological data for Heathrow Airport for 1995. Trajectories are initialised using hourly O_3 data for Harwell, Lullington Heath and Sibton for each day of 1995. The model includes dry deposition and chemistry and describes the height and spatial distributions of NO, NO_2 and O_3 at a spatial scale of 1 km x 1 km across London using either the NAEI or ERG-GLA LAEI NO_x emission inventories for 1999, 2005 and 2010.

565. The LRCTM model identifies and describes in some detail the following major sources of NO_2 in London:

- direct emission of NO_2 from NO_x sources, assumed to be 5% by volume in the base case model;
- oxidation of NO by O_3 advected into the model from the northern hemisphere baseline or from regional-scale photochemical pollution events;
- oxidation of NO in the NO + NO + O2 reaction in local-scale wintertime stagnation events.

Figure 5.29 Annual time series of model calculated NO_2 concentrations for an urban background location in central London using 1999 LAEI NO_x emissions.

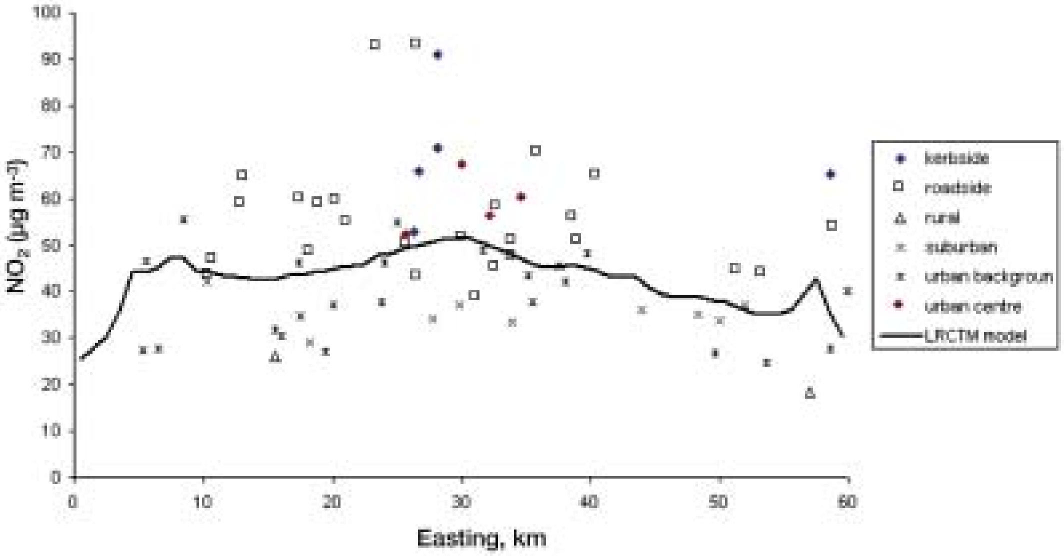


566. Figure 5.29 presents the annual time series of model hourly NO_2 concentrations for a typical urban background location in central London in the base case model using the 1999 LAEI NO_x emission inventory. The plot illustrates the presence of both a local-scale wintertime stagnation event, which occur particularly during November and December and the summertime regional-scale O_3 transport episodes, particularly during the period May to August.

5.4.2 Model validation

567. The LRCTM results on an annual average basis have been carefully compared with observations of NO_2 in Figure 5.30 for a selection of sites which form a west-east transect across London. The comparison has been made for the model surface level by averaging the results across a 10 km width, centred on Westminster. The model is well able to account for the observed variations in NO_2 concentrations along the transect, showing the step up from the rural values on the fringe of the London conurbation. The influence of the M25 corridor is clear at the easterly and westerly ends of the transect, with the peak at the westerly end somewhat higher; reflecting the added influence of Heathrow Airport. The model curve rises towards the centre of London and shows a significant portion of the transect above the 40 µg m^{-3} annual NO_2 air quality standard.

Figure 5.30 Observed annual mean NO_2 concentrations for 1999 at various sites and the model results for a 10 km wide transect from west to east across London, centred on Westminster using the 1999 LAEI NO_x emission inventory.



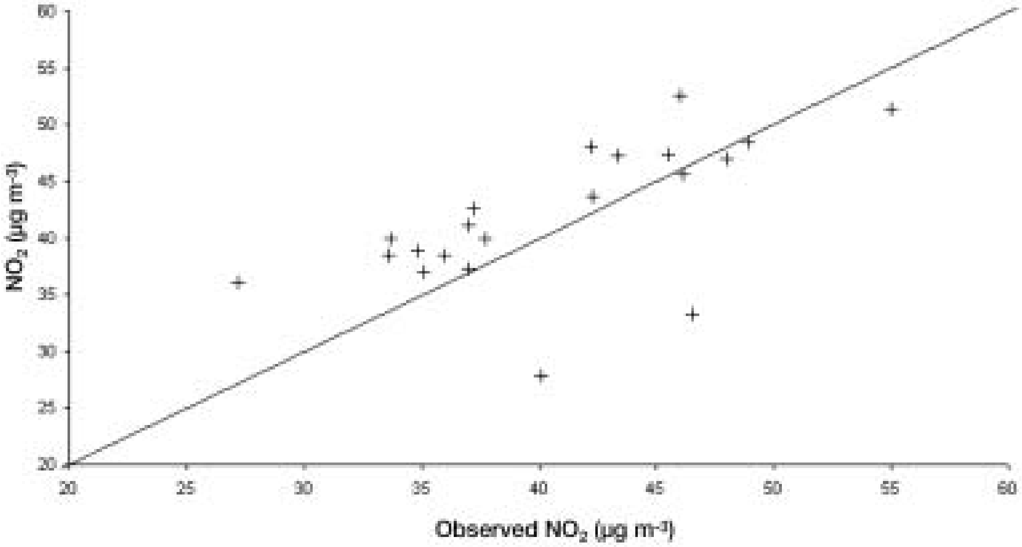
568. The model results appear to overestimate considerably the majority of the observed NO_2 concentrations for the sites marked as suburban and rural, and to underestimate the concentrations for the sites marked as roadside and kerbside sites. Model performance appears much better against the urban background sites. These comparisons are entirely consistent with the model's use of a 1 km x 1 km NO_x emission inventory. The model appears to be able to describe many of the major features of the observed distribution of NO_2 at urban background locations across London.

569. Table 5.8 describes a more detailed evaluation of the model predictions for suburban and urban background sites against the available observations for 1999. The mean model NO_2 concentrations was 42.9 μg m^{-3} compared with the observed concentrations of 41.4 μg m^{-3}. The mean bias was therefore about 1.5 μg m^{-3}, representing a mean percentage bias of 3.7%. The normalised mean square error was 0.02 and all of the points fell within a factor of two of the observations. The scatter plot of model vs. observations is illustrated in Figure 5.31 and was characterised by a correlation coefficient, r_2 of 0.51, indicating a good correlation between model and observation. Altogether these comparisons indicate a satisfactory level of model performance and indicate that the LRCTM model is able to describe the major features influencing the distribution of NO_2 across London.

Table 5.8 Comparison of observed and model calculated annual mean NO_2 concentrations for a range of suburban and urban background sites for 1999.

Site Name	Category	Observed (µg m^{-3})	Model (µg m^{-3})
Bexley 2 – Belvedere	Suburban	33.7	40.0
Bexley 5 – Bedonwell	Suburban	35.1	37.0
Enfield 1 – Bushhill Park	Suburban	33.6	38.5
Haringey 2 – Priory Park	Suburban	37.2	42.6
Hounslow 2 – Cranford	Suburban	42.2	48.0
London Bexley	Suburban	37.0	37.3
London Eltham	Suburban	36.0	38.5
Brentwood – Town Hall	Urban background	40.1	27.8
Ealing 1 – Ealing Town Hall	Urban background	46.0	45.7
Enfield 3 – Salisbury School	Urban background	37.7	39.9
Harrow – Stanmore	Urban background	34.8	38.9
Heathrow Airport	Urban background	55.4	60.7
Hertsmere Background (Borehamwood)	Urban background	27.2	36.1
Islington – Upper Street	Urban background	48.9	48.4
London Brent	Urban background	37.0	41.1
London N. Kensington	Urban background	46.0	52.6
Three Rivers Background	Urban background	46.5	33.4
Tower Hamlets – Bethnal Green	Urban background	43.4	47.3
Tower Hamlets 1 – Poplar	Urban background	45.5	47.4
Waltham Forest – Dawlish Road	Urban background	42.3	43.7
West London	Urban background	55.0	51.4
Newham Tant Avenue	Urban background	48.0	46.9

Figure 5.31 Scatter plot of model predicted annual mean NO_2 concentrations and the observed concentrations at suburban and urban background sites during 1999.

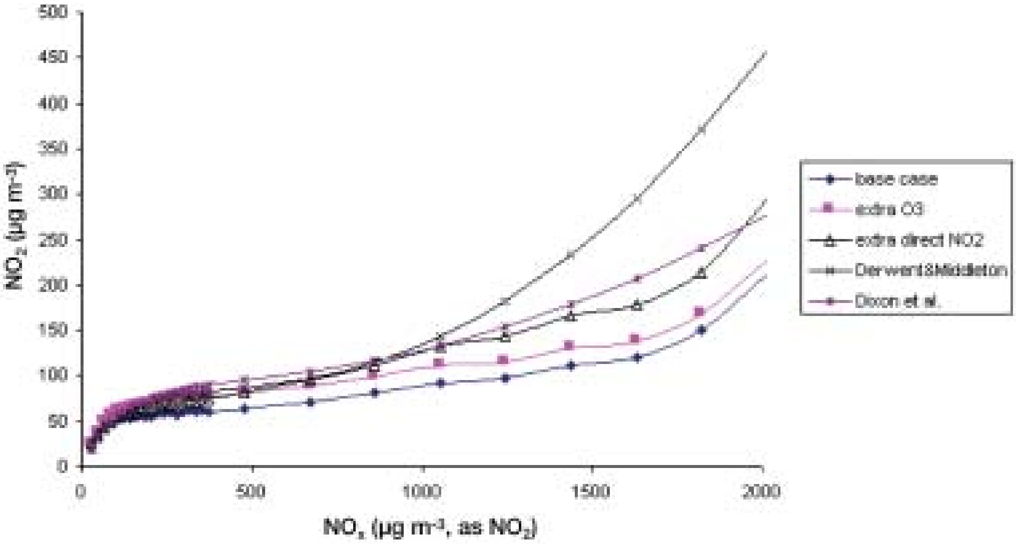


5.4.3 Comparison with empirical models

570. An important application of the LRCTM model has been to provide some underpinning to the empirical models used in policy formulation. An important feature of the observations of NO_2 and NO_x has been that there appears to be a simple relationship between the amount of NO_2 that is present in a polluted air mass and the NO_x concentration. Empirical curves have been derived of the 'so-called' NO_2 vs. NO_x relationship (Derwent and Middleton, 1996; Dixon *et al*., 2001). These empirical curves have been derived by sorting the hourly NO_x concentrations in ascending order and determining the mean NO_2 concentrations for each NO_x concentration. Plots of the resulting NO_2 vs. NO_x relationship from the data are shown in Figures 5.32 and 5.33. Here the observed NO_2 vs. NO_x relationship for Exhibition Road, London during the annual period which contained the 1991 pollution episode, is presented together with the relationship obtained by combining all the sites and years between 1991 and 1997 in London and the West Midlands.

571. Also shown in Figure 5.32 is the NO_2 vs. NO_x relationship obtained by sorting the model results in exactly the same manner as the observations. There is a clear similarity in shape and form between the model and observed NO_2 vs. NO_x relationships.

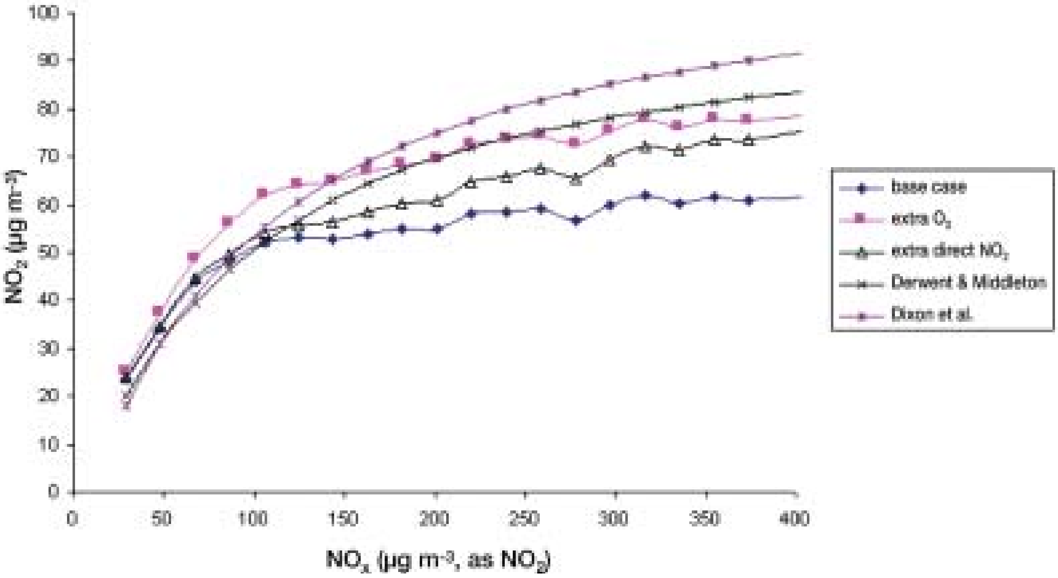
Figure 5.32 Observed and model derived NO_2 vs. NO_x relationships across the entire range of hourly NO_x concentrations.



572. To illustrate the main source of NO_2 in London, Figure 5.32 and 5.33 show two additional model sensitivity cases in addition to the base case model results. The sensitivity case marked 'extra O_3' addresses the difficulties associated with initialising the O_3 concentrations in the air parcels before they enter the London conurbation. Although it is straightforward to initialise the surface O_3 concentrations using observed rural concentrations, it is difficult to know how to initialise the O_3 concentrations throughout the vertical profile. Undoubtedly there is more O_3 aloft than close to the surface but there are no observed O_3 vertical profiles with which to initialise the model accurately. In the sensitivity case, an extra 20 µg m^{-3} of O_3 was added across the vertical profile and this produced a dramatic shift in the NO_2 vs. NO_x relationship derived from the model results. Figures 5.32 and 5.33 show the results from the base case and the 'extra O_3' sensitivity cases across the entire range of NO_x concentrations.

573. The second sensitivity case in Figures 5.32 and 5.33 shows the impact of doubling the fraction by volume of the NO_x emitted as NO_2 from 5 to 10% by volume. Again this sensitivity case generates a different shape NO_2 vs. NO_x relationship to the base case, indicating the importance of the direct emission of NO_2 in determining the model-calculated NO_2 concentrations. Clearly the model fit is improved at the highest NO_x concentrations with the higher direct emissions of NO_2. However, there is little information with which to fix this important model input parameter.

Figure 5.33 Observed and model derived NO_2 vs. NO_x relationships across the lowest range of hourly NO_x concentrations.



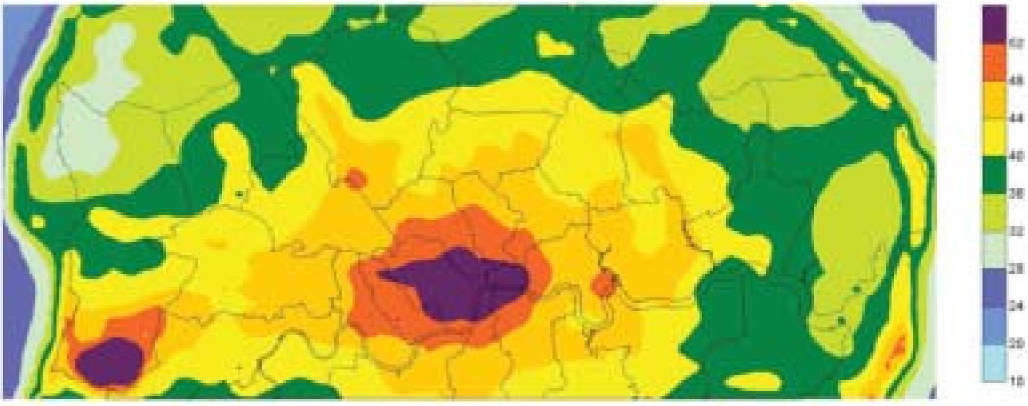
574. The close similarity between the NO_2 vs. NO_x relationships derived from the model and from observations provides strong confirmation that the model is able to describe quantitatively the magnitudes of the sources of NO_2 in London. Sensitivity studies point to the importance of direct emission of NO_2 and oxidation by O_3 as the two most important sources of NO_2 in London. A third source becomes active under wintertime stagnation conditions through the NO + NO + O_2 reaction.

5.4.4 Assessment of current NO_2 air quality across London

575. The LRCTM model can be used to calculate the spatial distribution of annual mean NO_2 concentrations at a spatial resolution of 1 km x 1 km across London as is shown in Figure 5.34. The mapped area contains 1560 grid squares of which 686 (44%) exceed the 40 µg m^{-3} annual mean air quality target. Exceedences are found in the vicinity of Heathrow Airport and across much of the centre of London. Peak model annual mean NO_2 concentrations of

67 µg m^{-3} are found in the vicinity of Heathrow Airport and 57 µg m^{-3} in central London using the LAEI 1999 NO_x emission inventory.

Figure 5.34 Distribution of annual mean NO_2 concentrations in µg m^{-3} calculated with the LRCTM model using 1999 NO_x emissions from the LAEI inventory.



5.5 Eulerian grid modelling

5.5.1 Modelling NO_x and O_3 using the urban airshed model

576. The Urban Airshed Model (UAM-IV) is an Eulerian air quality model developed in the United States for the prediction of O_3 within polluted urban regions. The University of Birmingham has applied the model to predict primary and secondary pollutant concentrations across the West Midlands conurbation.

577. The model requires detailed hourly three-dimensional meteorological fields which were provided through running the Colorado State University RAMS (Regional Atmospheric Modelling System) model using two nested domains, the outer one covering much of England, whilst the inner one covering the West Midlands conurbation with a horizontal spatial resolution of 2 km and 31 vertical levels. For each 2 x 2 km grid square and each vertical level, the RAMS model predicts variables such as air temperature, pressure, wind speed, wind direction and turbulent kinetic energy (TKE). The calculation requires the meteorological information on the boundaries of the outer domain together with spatially disaggregated information on topography and land use types within the domain of the model. The RAMS model was validated in detail against measured data and provides an excellent simulation of hourly meteorological data. The depth of the atmospheric boundary layer was diagnosed from the vertical profile of TKE. The UAM model uses the meteorological fields of the inner domain.

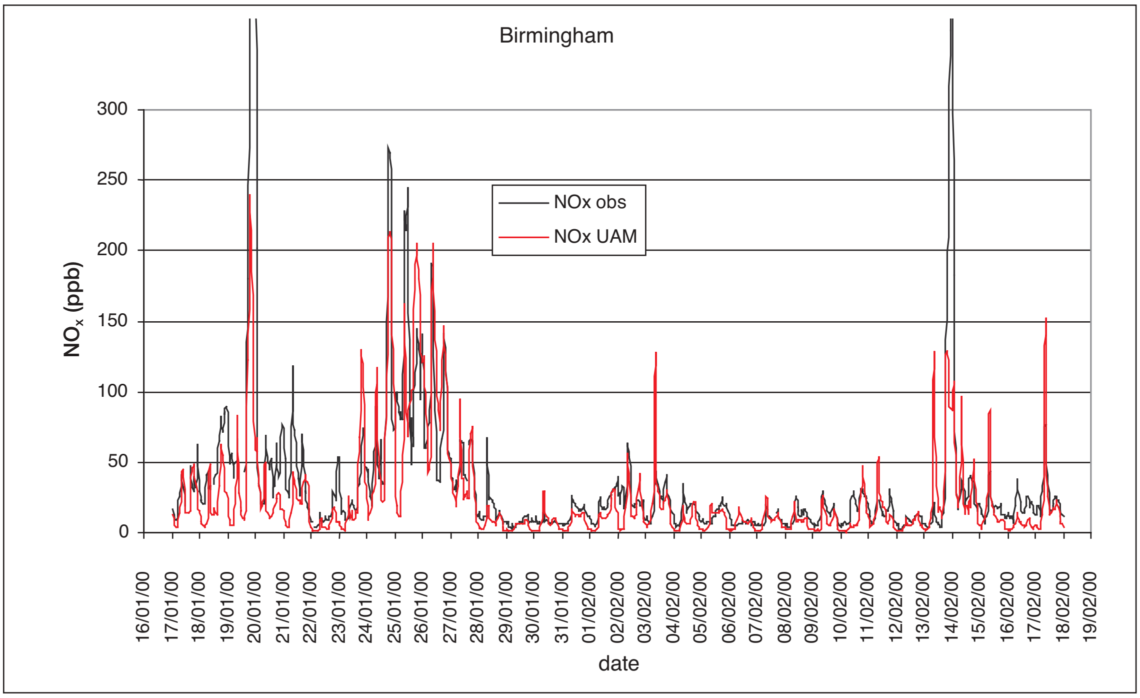
578. In addition to meteorological drivers, the UAM model requires data on primary emissions of pollutants and the 1998 1 x 1 km inventory from the National Atmospheric Emissions Inventory was used across the modelled domain of the West Midlands. Diurnal variations in emissions from major sources such as road traffic were imposed on the inventory. Since the model relies upon an Eulerian formulation, for each time step, pollutants which are emitted and those already within the atmosphere are dispersed according to the level of turbulence and transferred between grid boxes in the model according to the three dimensional wind vectors predicted by the RAMS meteorological model.

579. In addition to the advection and diffusion of primary pollutants, the model needs to account for atmospheric chemical change and accordingly it contains the CBM IV chemical scheme which is a highly condensed scheme using surrogate species to describe carbon bond types and molecules of similar behaviour. It was run with a horizontal resolution of 2 x 2 km and six vertical layers, four of which were in the boundary layer. For each time step in the model, the chemical scheme computes pollutant degradation and formation within each grid box, and the pollutants are then transferred between grid boxes at the end of the time step, as for unreactive primary pollutants. The model computes the atmospheric chemical changes in each 2 x 2 km grid box and for each of the six vertical levels as well as transferring materials between the boxes in response to the results of the meteorological model. Such simulations are highly computationally intensive.

580. It was found particularly important to provide the model with hourly upwind boundary conditions representing pollutant concentrations on the upwind edge of the model domain. These were taken from campaign measurements where available and elsewhere from the nearest available National Network rural monitoring station.

581. The model was run in order to simulate the results of monitoring campaigns which took place over periods of one-month duration in the summer of 1999 and the winter of 1999/2000. The predictions of NO_x were generally good, with a slight under prediction in the summer and a very good reflection of true concentration magnitudes in the winter. The model had difficulty in reproducing exactly the hour-to-hour variation in NO_x, but produced excellent simulations of average diurnal behaviour and reasonable representations of specific episodic conditions. The results for the winter campaign appear in Figure 5.35. In both summer and winter the model predicts rather lower NO_2/NO_x ratios than are actually observed, despite simulations of O_3 being generally good.

Figure 5.35 Model and observed NO_x concentrations at the Birmingham East site during January 2000.



582. Further examples of model output appear in Figures 5.36 and 5.37. In Figure 5.36, predictions of concentrations NO of at the Pritchatts Road site in south Birmingham over the period 24-28 January 2000 are compared with measurements. Also shown are measurements of atmospheric boundary layer depth (Zi) which clearly show the build-up of high NO concentrations when the boundary layer depth was low (200 m), with much reduced concentrations when the boundary layer deepened, especially towards the end of the monitoring period. Figure 5.37 shows the spatial distribution of NO, NO_2 and O_3 across the model domain at 16.00 on 24 June 1999, when the wind was in an easterly sector. Clear O_3 decrements and corresponding NO_2 formation are seen across central Birmingham and the Black Country corresponding to O_3 removal by reaction with traffic-generated NO_x. Also clearly visible are O_3 decrements in the plumes downwind of major point sources to the east of the conurbation, together with NO_2 enhancements.

Figure 5.36 Measured and modelled nitric oxide and boundary layer depths in Birmingham, 24-28 January 2000 from UAM.

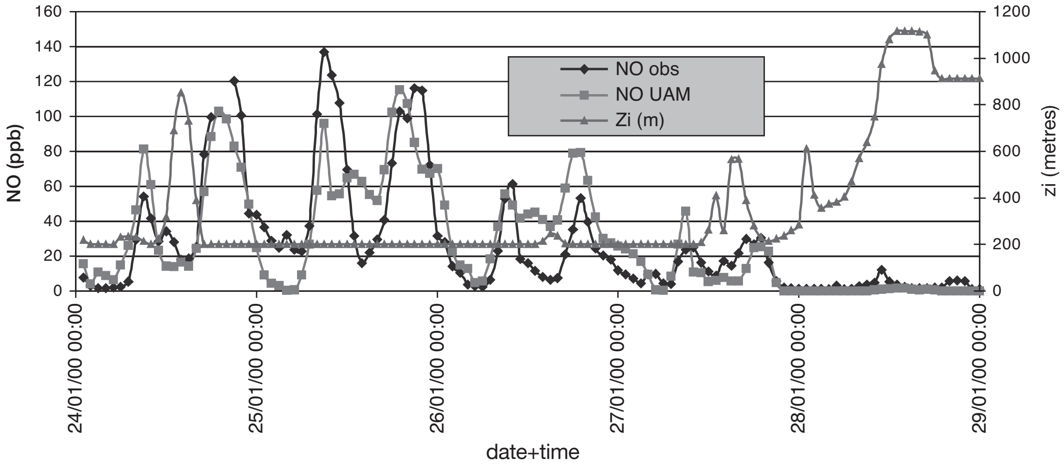
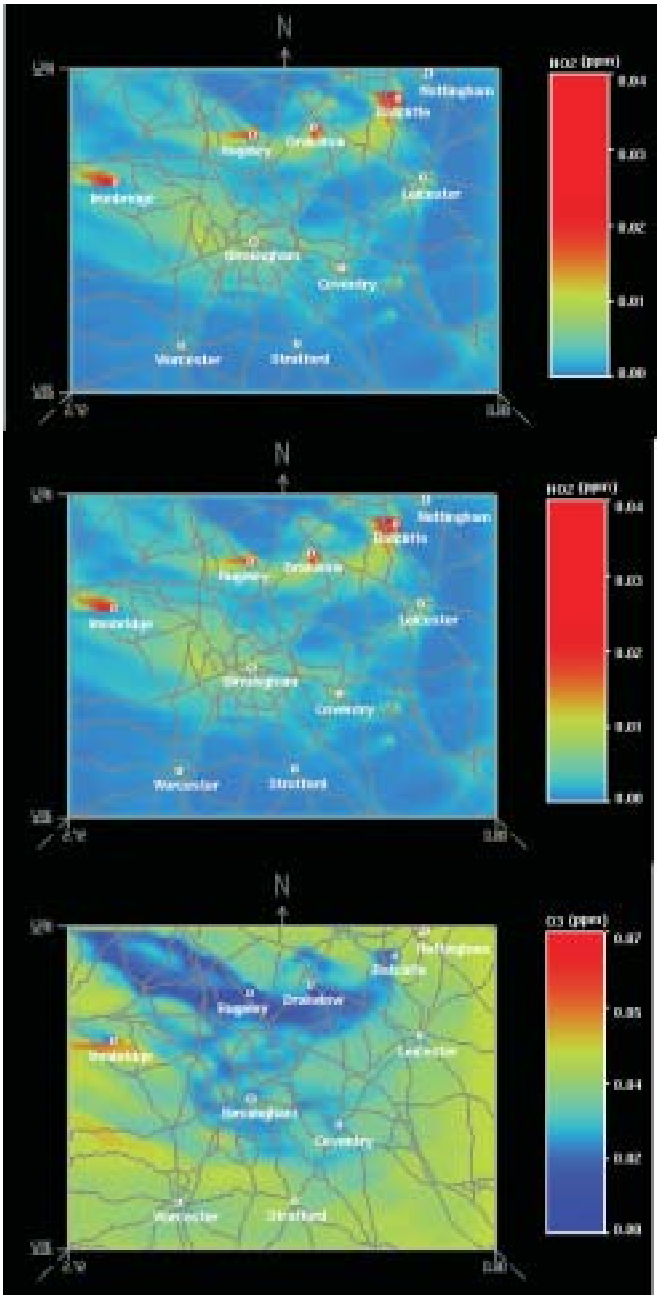


Figure 5.37 Surface concentrations of NO, NO_2 and O_3 on 24th June 1999.



583. The work demonstrates that Eulerian modelling approach has considerable utility in predicting background concentrations of NO_x within urban and rural areas. The model resolution is however insufficient to allow calculations for specific streets and urban hotspots.

5.6 Surface concentration mapping

5.6.1 Introduction

584. Producing a spatial representation of NO_2 concentrations usually requires application of an interpolation procedure, i.e. the estimation of the concentration at an unsampled location using values at sampled locations (such as monitoring stations or dispersion model receptor points).

585. A benefit of surface concentration modelling is the assessment of potential patterns of exposure to specific events, which can otherwise be difficult to estimate. Some techniques also enable generation of error and probability surfaces which can be used to assess the uncertainty in spatial estimates and/or the probability of concentrations exceeding air quality standards. Error estimations associated with an interpolated surface can help inform the location of new monitoring stations, which can in turn help provide a more accurate interpolated surface for future air quality assessments.

586. Disadvantages to interpolation techniques include the difficulty of determining the 'best' interpolation procedure to use for a particular dataset and careful consideration of sample points to ensure that a valid air quality surface is produced. The main policy-related drawback is the lack of any direct future predication capability.

5.6.2 Techniques and characteristics

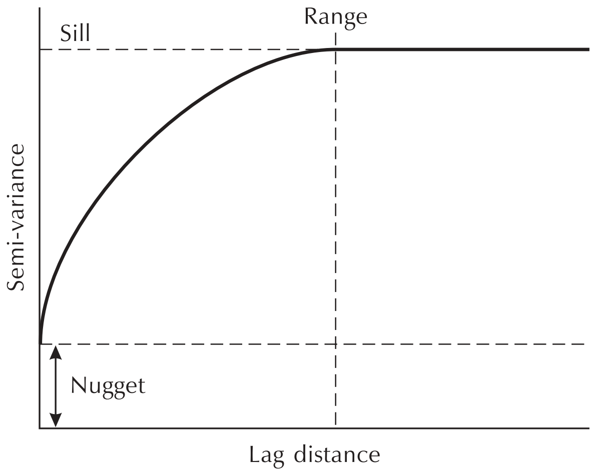
587. Surface concentration modelling of NO_2 using interpolation assumes that the value of NO_2 at a particular point is likely to be more similar to the values at nearby sites than to the values at sites further away. The application of basic interpolation procedures only yields meaningful results if sample points have similar siting characteristics, and are not strongly influenced by specific local sources. It is therefore usual to use data relating to background sites as the basis of such an interpolation exercise. Resultant concentration surfaces indicate a 'broad brush' estimated pattern of background concentrations and do not estimate the influence of particular sources at individual locations, such as around major roads. The national maps produced using the methodology described in section 5.2 of this chapter provide greater spatial detail through the combination of interpolation techniques with other modelling procedures and additional datasets.

588. This restriction does not necessarily apply to interpolating the output of dispersion models where surfaces are estimated from user-defined or automatically selected receptor locations. A regular sampling framework of an appropriate density or one which is developed to be sensitive to known patterns of change (for example, through specifying a higher density around major road links) enables estimates to be generated between points with reasonable confidence.

589. There are numerous techniques available to interpolate monitoring data, each with different characteristics. The underlying interpolation algorithm selected always has an influence on the patterns of concentrations generated from the same input dataset (reference literature on GIS and geostatistics includes: Burrough and McDonnell, 1998; Isaaks and Srivastava, 1989). Although there is no universally accepted mechanism, there are a number of techniques that are commonly employed to map the spatial distributions of air quality data. Interpolation by kriging has been identified by a number of authors as a suitable technique (for example, Lefohn *et al.*, 1987; Phillips *et al.,* 1997).

590. Kriging is a smoothing interpolator which uses regionalised variable theory. In its basic form it requires the spatial structure of the phenomenon under consideration to have the same patterns over the entire area being examined (stationarity). The nature of this pattern is determined through analysis of a semi-variogram (Figure 5.38) which plots the difference in the magnitude of measured values between pairs of sample points as a function of the spatial separation of the points (the lag distance). If the data are spatially auto-correlated, the differences in values for sample pairs is less where the samples are closer together and higher where the sample pairs are further apart. Using the pattern of auto-correlation as a guide, a function is then fitted which best describes the observed relationship. This function will also help determine the range and the sill of the dataset, after which point differences are taken as random spatially uncorrelated noise. The data may also include a nugget value that represents uniform noise in the dataset, for example, as a result of measurement error.

Figure 5.38 Characteristics of the semi-variogram



591. Alternative standard interpolation methods include use of splines, which produce concentration surfaces where surface curvature is minimised between sample points, and inverse distance weighting in which values are estimated using samples in a specified neighbourhood, the nearer samples are the more weight they are assigned.

592. Given that data patterns can exhibit large variability, as a result of source proximity, meteorology and chemical reactions, standard procedures may be inadequate due to an inability to account for different spatial structures within the same dataset. Consequently, variations of standard approaches have been developed (for example, Fuentes, 2001; Ionescu *et al.,* 2000)

593. Exploratory data analysis is an important first stage of any interpolation exercise which helps to identify the statistical characteristics of the sample dataset and the presence of any outliers prior to surface generation. The results of interpolation procedures can be assessed in various ways, for example through a cross comparison of surfaces and their error statistics, or through a comparison of the relative influence of individual sample points. The former can help to identify the optimum surface from a set of possibilities, both in terms of different procedures and also in terms of other specific parameters such as the size and orientation of search neighbourhoods. The latter includes techniques such as cross validation of sample points using a leave-one-out procedure where predictions for known sample sites can be made through the application of the procedure with the remaining samples. This is useful to identify the particular influence of individual sites on the final surface and can be a further basis for the rejection of samples as outliers. Local knowledge can also be an important factor in the assessment of estimated surfaces.

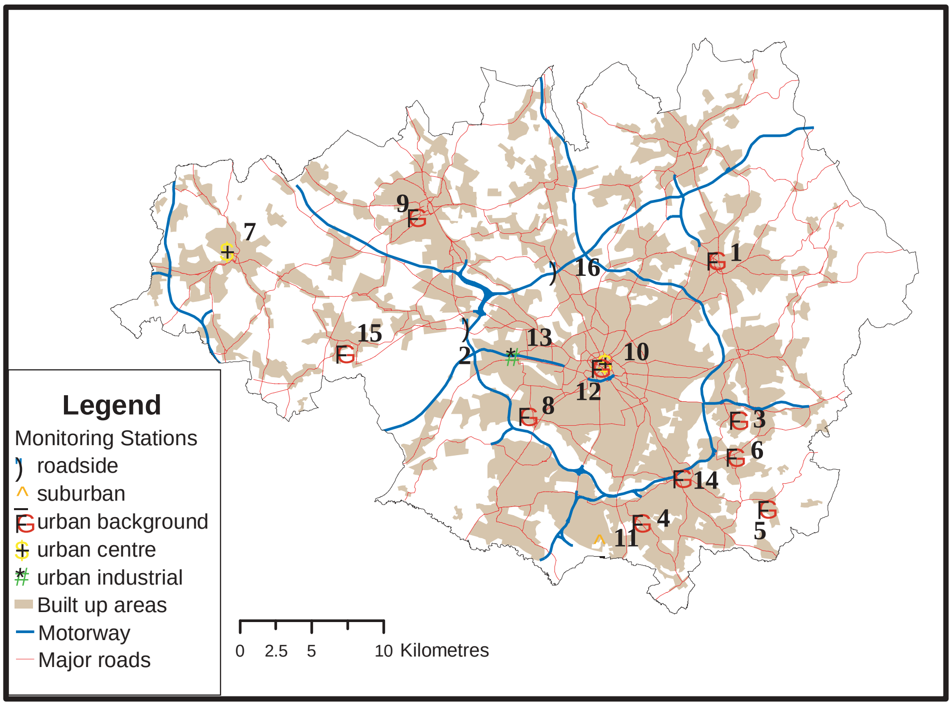
5.6.3. Urban scale interpolation

594. To illustrate some of the techniques and issues outlined above, concentration surfaces have been applied to the estimation of patterns of NO_2 concentrations in the Greater Manchester conurbation, first during a winter-time episode in December 2001 and secondly for annual mean concentrations for 2001.

5.6.3.1 Concentration modelling of episode conditions

595. As is discussed in section 6.5.3, elevated concentrations of hourly NO_2 were experienced in a number of northern UK cities during the period of Tuesday 11th and Wednesday 12th of December 2001. In the conurbation of Greater Manchester, concentrations greater than 200 µg m^{-3} were seen at many sites. A map of the location of sites is given in Figure 5.39. Peak hourly values and the time they were recorded for each site are shown in Table 5.9 and the full time series are shown in Figure 5.40. Although the peak value was experienced at the largest number of sites on the morning of the 12th December, the episode was characterised by a large range of values over relatively small spatial scales and short time periods. This makes the interpolation of concentration values particularly problematic.

Figure 5.39 Location of automatic monitoring stations measuring hourly NO_2 in Greater Manchester, 2001.



596. The sites to form the sample dataset, shown in Table 5.9, were selected primarily on the basis of their siting characteristics and the temporal patterns in concentrations over the period of interest. The urban industrial site at Salford Eccles is near to a relatively large point source of NO_x (which emitted 300 tonnes of NO_x during 2001) but since emissions from elevated sources are not likely to exert an influence during this type of winter-time, and the temporal pattern of concentrations was similar to neighbouring urban background sites (Figure 5.40(a) and (c)), this site was retained. Exploratory data analysis of specific hourly values suggested that the urban background sites of Stockport Bredbury (site no. 6) represents an extreme outlier for some of the hours during the time period and the temporal distribution of values was also found to be atypical compared to the remaining stations in the area. Although there is a strong case for exclusion of this site from the analysis it was included in the interest of completeness.

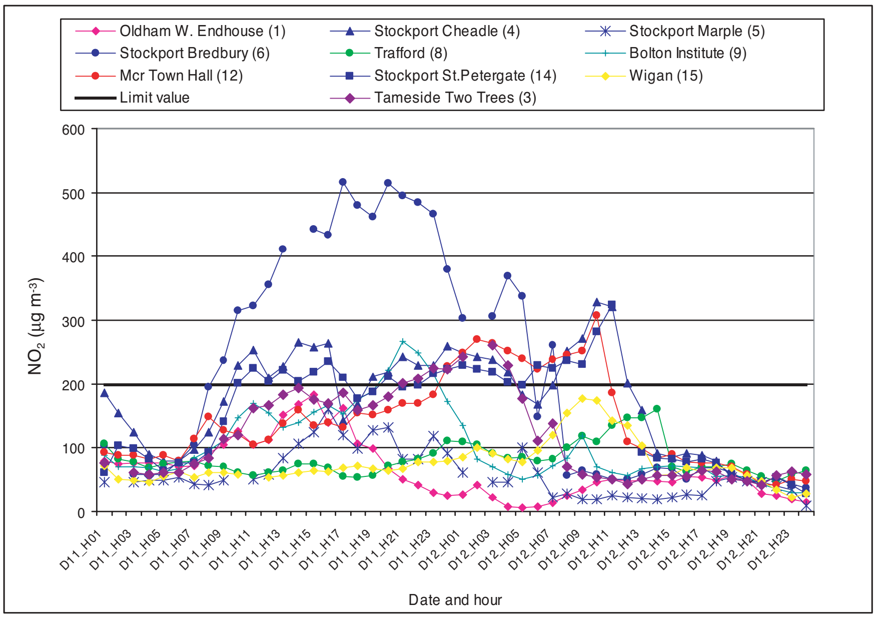
597. Three basic interpolation functions were used to assess the optimum function for surface generation: inverse distance weighting, splines and kriging. The very small number of sample data points in this case restricted the possibilities of using a more sophisticated approach. Since ordinary kriging requires normally distributed data for the generation of probability measures (but not for error and prediction surfaces), it was necessary to identify hours which showed a normal distribution (or which could be transformed to a normal distribution) for the creation of probability surfaces.

Table 5.9 Peak hourly NO_2 concentrations 11/12/01 to 12/12/01 (µg m^{-3}) monitored at AURN and Calclub+ sites in Greater Manchester.

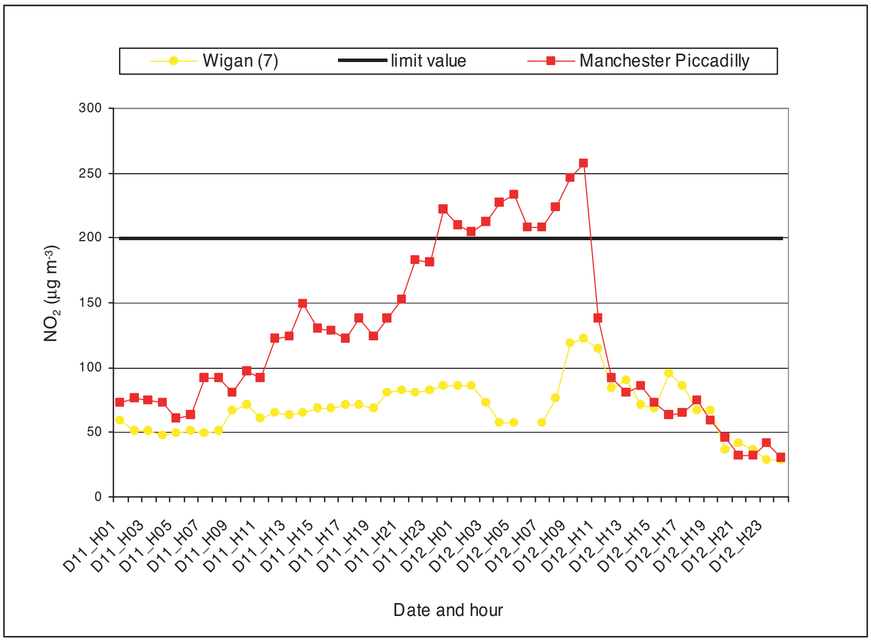
Station	Siting	District (Site name)	Peak value	Peak hour	No. hrs > 200 µg m^{-3}	Modelled
1	Urban Background	Oldham (W. Endhouse) +	183	11/12 15:00	0	Y
2	Roadside	Salford (M60) +	275	12/12 11:00	9	N
3	Urban Background	Tameside (Two Trees) +	262	12/12 15:00	7	Y
4	Urban Background	Stockport (Cheadle) +	329	12/12 10:00	22	Y
5	Urban Background	Stockport (Marple) +	160	11/12 16:00	0	Y
6	Urban Background	Stockport (Bredbury) +	516	11/12 17:00	20	Y
7	Urban central	Wigan (Centre) +	122	12/12 10:00	0	Y
8	Urban Background	Trafford (Background) +	160	12/12 14:00	0	Y
9	Urban Background	Bolton (Institute)	267	11/12 21:00	4	Y
10	Urban central	Manchester (Piccadilly)	258	12/12 10:00	11	Y
11	Suburban	Manchester (South)	159	12/12 10:00	0	N
12	Urban Background	Manchester (Town Hall)	308	12/12 10:00	11	Y
13	Urban industrial	Salford (Eccles)	432	12/12 10:00	11	Y
14	Urban Background	Stockport (St. Petergate)	325	12/12 11:00	21	Y
15	Urban Background	Wigan (Leigh)	178	12/12 09:00	0	Y
16	Roadside	Bury (M60)	311	11/12 23:00	10	N

Figure 5.40 Time series of NO_2 concentrations across Greater Manchester 11th to 12th December 2001 at (a) urban background; (b) urban centre; and (c) other sites.

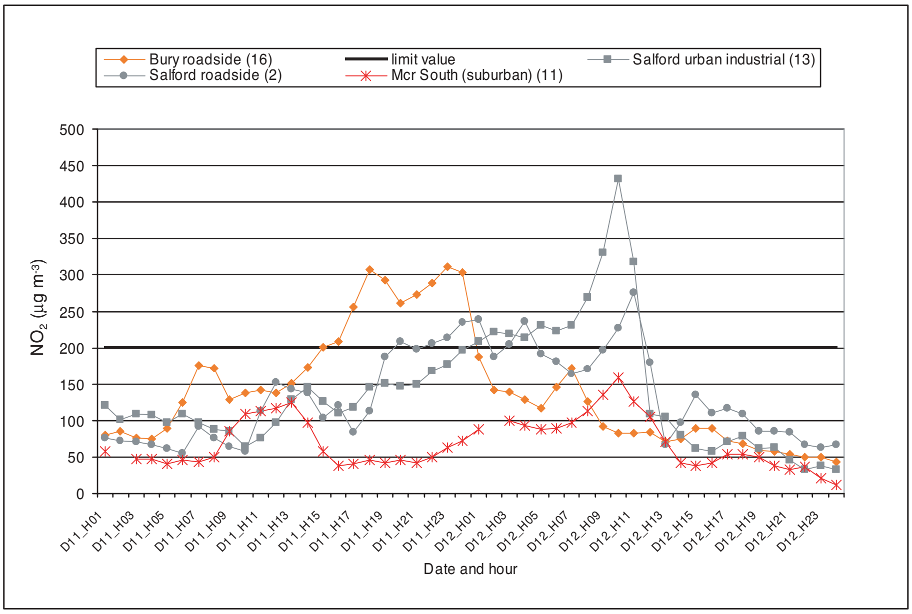
(a) urban background sites



(b) urban centre sites



(c) Other sites

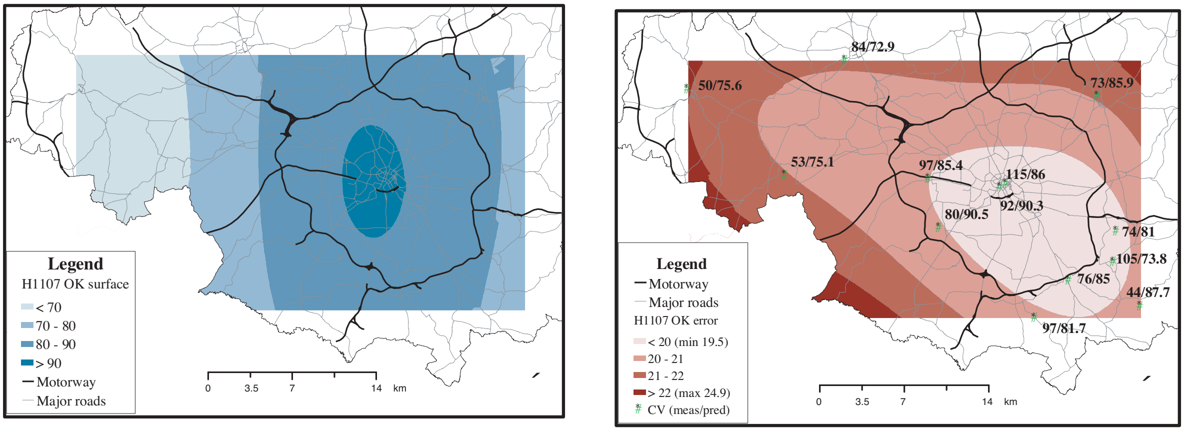


598. The results of a cross comparison of surfaces suggested that splines and ordinary kriging produced least overall error in the final surface estimates although the specific error and parameters in each case did vary according to the hour selected. In the interests of consistency, a time series of surfaces have been produced using ordinary kriging throughout, as this produced the optimum results for most of the hours tested. Examples of estimated NO_2 concentration surfaces for 7 am, 2 pm, and 7 pm on 11th December and 7 am and 9 am on 12th December are shown in Figure 5.41. The morning of the 11th shows that Manchester city centre has the highest NO_2 concentrations with a gradual fall-off towards the periphery of the conurbation, consistent with the annual average pattern of NO_2 concentrations measured at the automatic sites in the area. By 2 pm concentrations of NO_2 have increased considerably and begin to show a distinct pattern of high concentrations towards the south west. This pattern continues into the next day with very high concentrations by 9 am of the 12th. In the morning of the 12th there are also high concentrations in central Manchester which were less pronounced during the afternoon and early evening on the 11th. These surfaces only represent background concentrations. Concentrations of NO_2 within 20-50 m of motorways and major roads (see section 6.3) are likely to be in excess of estimated background values whereas locations with a suburban characteristic are likely to exhibit lower concentrations.

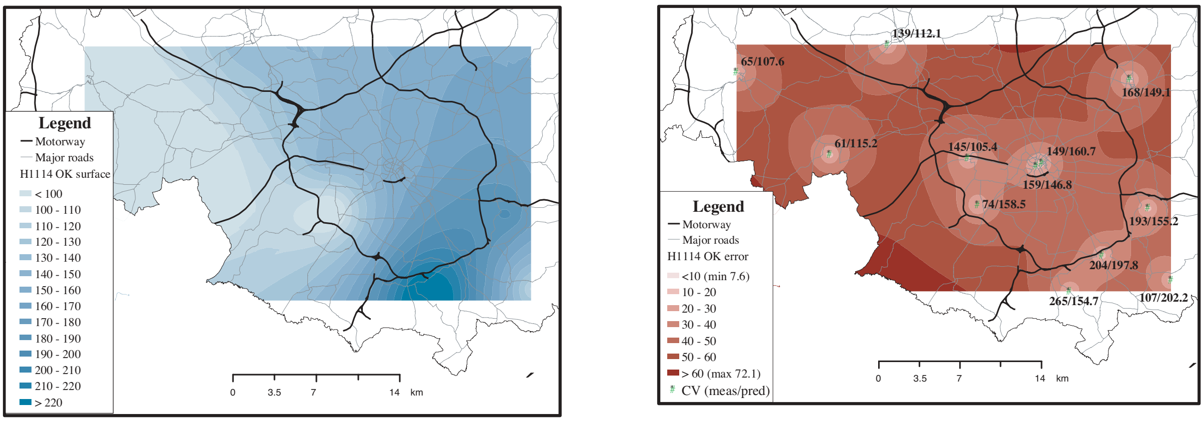
599. Figure 5.41 also provides error surfaces for each of the modelled hours. The most significant errors are associated with time periods with the highest concentrations. Cross-validation indicated large errors showing which particular stations have the most impact on the final surface generated.

Figure 5.41 Selected hourly predicted NO_2 concentration (left-hand maps) and error surface concentration (right-hand maps). Cross validation values refer to measured/predicted values based on leave-one-out procedure.

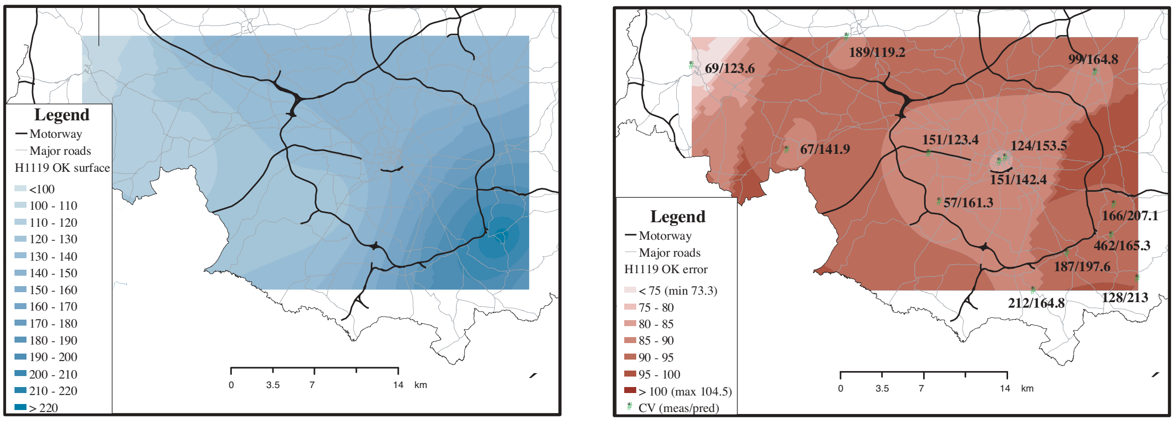
(a) 11th Dec 7am (monitored min 44, monitored max 115) – concentrations are in µg m^{-3}.



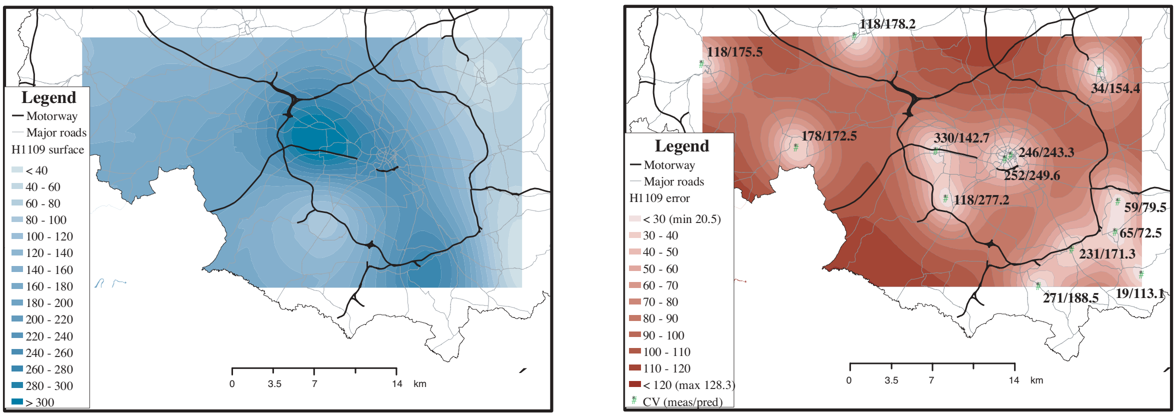
(b) 11th Dec 2pm (monitored min 61, monitored max 265). – concentrations are in µg m^{-3}.



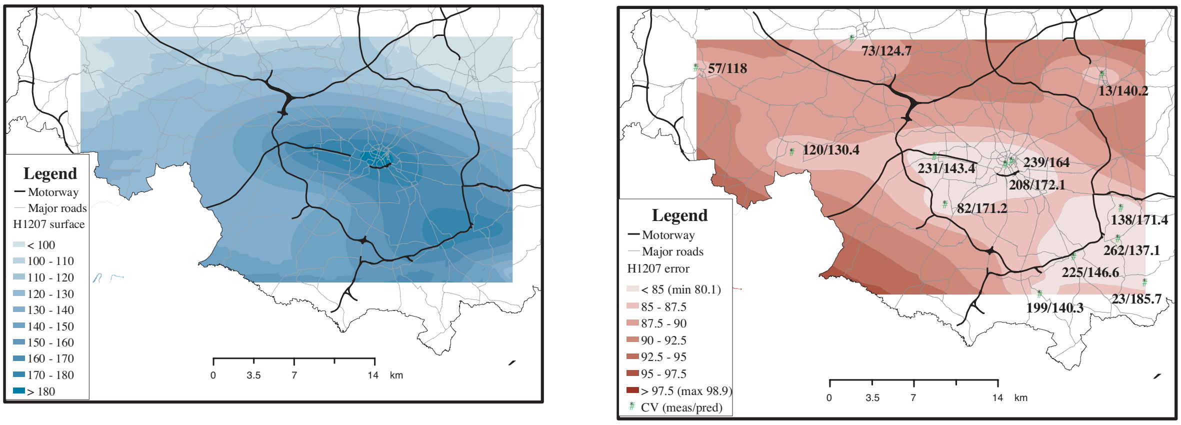
(c) 11th Dec 7 pm (monitored min 55, monitored max 210) – concentrations are in µg m^{-3}.



(d) 12th Dec 7 am (monitored min 13, monitored max 262) – concentrations are in µg m^{-3}.



(e) 12th Dec 9 am (monitored min 19, monitored max 330) – concentrations are in µg m^{-3}.

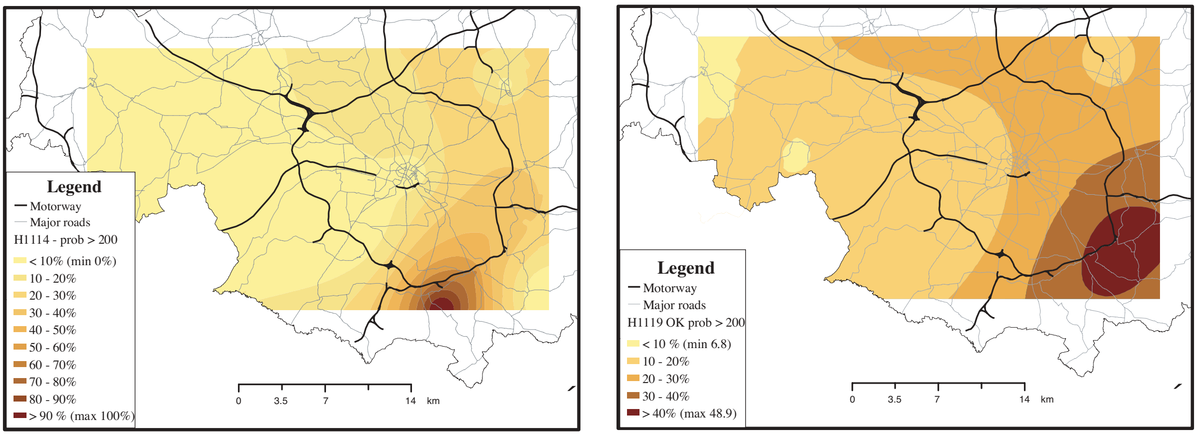


600. Illustrative probability maps for selected hours are shown in Figure 5.42, and indicate the estimated probability that NO_2 concentrations exceeded 200 µg m^{-3} at urban background locations over the conurbation at the times specified. As with the concentration maps, these probabilities do not relate to locations near to large local sources such as major roads and motorways.

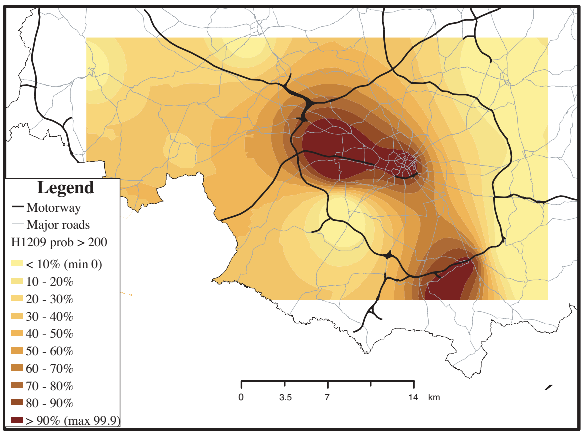
Figure 5.42 Selected hourly probability surfaces for exceedence of 200 µg m^{-3} during 11th and 12th December 2001 (%).

(a) 11th December 2pm

(b) 11th December 7pm



(c) 12th December 9am



601. This urban scale episode work has illustrated a number of issues raised in section 5.6.2 concerning the application of interpolation techniques to estimating surface concentrations of NO_2. The lack of sample points in this example restricts the level of sophistication with which procedures can be applied as well as the robustness of the data produced. The lack of points meant that it was not always possible to include anisotropy (directional) effects which can sometimes help to improve the robustness of predictions.

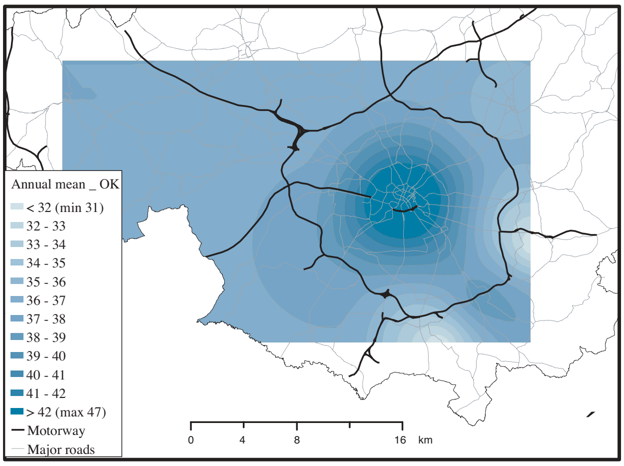
602. The very high variability in concentrations monitored at urban background locations in relatively close proximity (especially in Stockport in the south-west) suggests the strong influence of localised site characteristics (for example, proximity to source or topographic setting). The impact of these characteristics may be particularly important during winter-time episode conditions. Consequently the assumption of stationarity may not then hold for all the hours shown in this example.

5.6.3.2 Concentration modelling of annual means

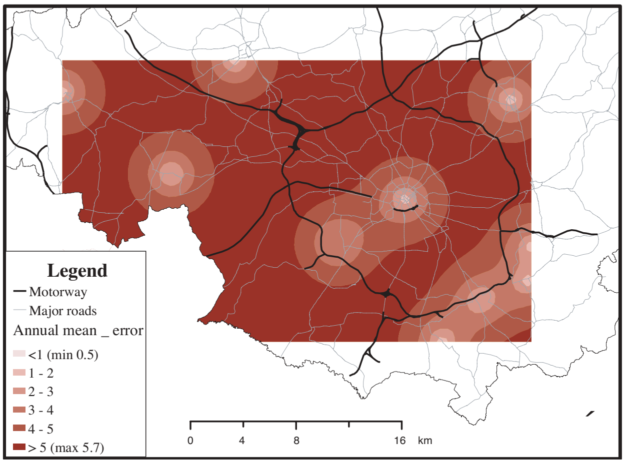
603. Spatial maps of annual average NO_2 concentrations for the Greater Manchester have also been estimated for the sites indicated in Table 5.9 for 2001 using the same basic methodology described in the previous section. Prediction, error and probability surfaces are shown in Figure 5.43. These suggest that there was a very high probability that annual average concentrations of NO_2 exceeded 40 µg m^{-3} in many central Manchester locations in 2001. There is a higher overall confidence with annual mean data as can be seen from the error plots in Figure 5.43.

Figure 5.43 Prediction, error and probability surfaces (exceedence of 40 µg m^{-3}) for annual average NO_2 concentrations in Greater Manchester for the calendar year 2001.

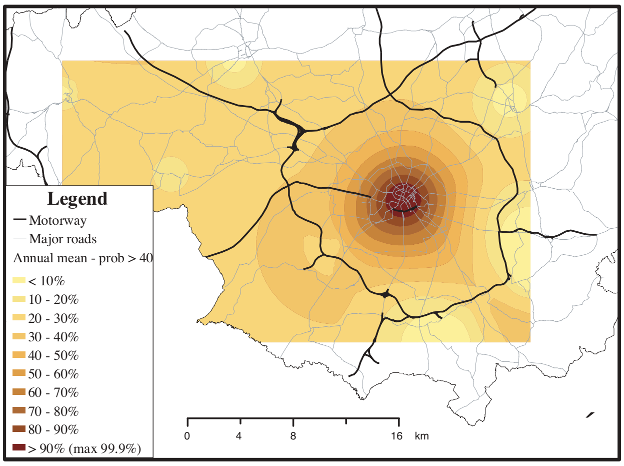
(a) Predictions – concentrations are in µg m^{-3}



(b) error surface – concentrations are in µg m^{-3}



(c) probability surface for exceedence of 40 µg m^{-3} (%)



5.6.4. National scale interpolation

604. At the national scale, fine scale spatial variations are currently best represented with the dispersion model based semi-empirical techniques discussed earlier in this chapter.

605. Measured annual mean concentrations of NO_2 at intermediate and urban background locations in the UK NO_2 Diffusion Tube Network have been routinely used by NETCEN to produce interpolated maps of urban background NO_2 concentrations produced throughout the UK, using a bilinear interpolation model (Loader *et al.*, 2002). The bilinear interpolation method creates a smoothed two-dimensional dataset from bilinear and quadratic functions on x, y, and z datasets. This interpolation method approximates existing points onto a grid (in this case 10 x 10 km), interpolates by linear interpolation, and then improves the estimation by computing gradients at the points using quadratic interpolations. It then refines the values using-distance weighting methods. The new surface smoothes the original data points, it will not necessarily pass though them.

606. The same interpolation algorithm has been used since the network was commissioned in 1993, to ensure consistency for comparative purposes. As with the example of spatial mapping in the Manchester area, these maps only represent NO_2 concentrations at urban background locations and not at roadside or rural locations. Roadside measurements are not interpolated, as concentrations measured at such locations are likely to be representative of only very limited spatial areas.

5.6.5. Summary and conclusions

607. This section has provided an overview of interpolation procedures and their usage in terms of modelling surfaces of NO_2. There are a variety of techniques available and no universally agreed methodology for application to air quality datasets. The examples have shown some of the specific considerations in applying interpolation to NO_2 values at the urban scale, many of which are directly applicable to surfaces produced for the national scale. These include a consideration of monitoring station siting characteristics as well as the extent, density and distribution of points in terms of monitored data or dispersion model receptor points. Although there are drawbacks and difficulties in the application of interpolation procedures, these are essential building blocks to many of the modelling procedures discussed in this chapter.

Chapter 6

NO_2 concentrations and recent trends

Key points

- Measured NO_2 concentrations during 2001 have been assessed for a total of 212 automatic monitoring sites. Data from 15 of these sites have been considered for detailed analysis. Annual mean NO_2 concentrations above 40 µg m^{-3} (the annual mean objective for 2005 and limit value for 2010) were measured at the majority of roadside and kerbside sites in London and at about half of the background sites. Exceedences outside London were largely confined to roadside and kerbside monitoring sites. Sites with more than 18 hours with measured hourly mean concentrations greater than 200 µg m^{-3} (the 1-hour objective for 2005 and limit value for 2010) were less common than exceedences of an annual mean concentration of 40 µg m^{-3}. The annual mean is thus the key assessment level for nitrogen dioxide.
- At the 13 monitoring sites for which there are long-running time series of observations, statistically significant downwards trends have been observed in NO_2 and NO_x concentrations over the 1993-2002 period. Annual mean NO_x concentrations have shown a -5.1 ± 1.6% per year downwards trend compared with -4.4% per year in the UK NO_x emission inventories. Downwards trends in annual mean NO_2 concentrations at urban background sites are also statistically significant and average -3.1 ± 1.7% per year, about two-thirds of the simultaneous NO_x trends. An assessment of rolling annual mean concentrations at 11 background and roadside sites in London since 1995 shows a decrease in NO_x concentrations to 70% and of NO_2 concentrations to 88% of the 1996 values.
- The trends in ambient NO_x and NO_2 concentrations over the last 10–20 years can be reconciled with the changes in emissions by a combination of the source apportionment of current measured concentrations and the trends in emissions in published emission inventories.
- An analysis of the roadside increment of NO_2 concentrations has shown the key role of the emissions of primary NO_2 from traffic sources in determining annual mean NO_2 concentrations at the roadside. The NO_2/NO_x ratios of this increment ranged between 8% and 29% at the six sites studied in detail. The higher ratio is associated with a greater proportion of the emissions being from diesel vehicles in slow moving congested traffic, while the lower ratio is associated with fast moving motorway traffic.
- Road traffic is the dominant source of NO_x at busy roadside locations, but, because of the non-linear relationship between NO_x and NO_2, the roadside NO_2 concentrations are also strongly influenced by the local background NO_2 concentration.
- The contribution to annual mean NO_x concentrations in 2001 from domestic and commercial emissions (primarily natural gas space heating) at background locations in central London is estimated to be about a quarter of the total. This is important because emissions from these sectors are expected to increase somewhat over the next 10 years or so, in contrast to emissions from traffic, which are expected to decline.
- The range in annual mean NO_2 concentrations monitored at urban background sites in the UK are broadly comparable with those monitored across Europe and reported in the AIRBASE database. Proximity to traffic appears to be the major determining influence on the annual mean NO_2 concentrations monitored at over 1600 sites across Europe and reported in the AIRBASE database. Annual mean exceedences of 40 µg m^{-3} NO_2 appear to be widespread in urban centres across Europe.

- Annual mean NO_2 concentrations across Europe have declined during the 1990s at between -2 and -3% per year, trends closely similar to those in national NO_x emissions over the same period. Annual mean NO_2 concentrations at most of the rural and remote rural sites in the EMEP database have also declined during the 1990s, with trends of about -2% per year.
- The few studies of the way in which NO_2 concentrations decline away from the road suggest that concentrations are generally only significantly elevated above background levels within about 20 m of the edge of the road.
- Elevated sources such as power stations have a relatively minor impact on annual mean NO_2 concentrations at ground level, typically in the range 1 to 4 µg m^{-3}.
- Aircraft emissions are the dominant source of NO_x in airports. A source apportionment study at Heathrow shows that emissions from aircraft below 50 m dominate the aircraft contribution to concentrations. This contribution is important relative to road traffic emissions up to about 1 km from runways and away from the immediate vicinity of major roads.
- The use of models enables an extension of analysis based on results of monitoring data to the whole country. The results of dispersion modelling studies for 2001 and 1999 are generally consistent with measurements in terms of exceedences 40 µg m^{-3}. Exceedences are currently widespread throughout the London conurbation in the immediate vicinity of heavily-trafficked roads. These areas of exceedence spread further into urban background locations and fill much of the space between the major road links in the centre of London although the estimated extent of exceedence in background locations varies considerably between the different modelling studies.
- Detailed dispersion modelling studies indicate that between 34-68% of the area of London and alongside (at about 5–10 m from the edge of the road) between 95-98% of major roads in London annual mean NO_2 concentrations exceeded 40 µg m^{-3} in 1999. The national models suggest that concentrations were above this level alongside 90% of major roads in London, 33% in the rest of England, 17% in Scotland and 5% in Wales and Northern Ireland in 2001.
- Episodes when hourly mean NO_2 concentrations exceed 200 µg m^{-3} can be classified into two types. Winter episodes are associated with poor dispersion of primary NO_x emissions, with elevated NO_2 concentrations derived from a combination of NO_x emitted as primary NO_2 and emitted as NO followed by reaction with O_2. Summer episodes are characterised by elevated secondary NO_2 concentrations associated with regional scale photochemical oxidant episodes.
- An analysis of the occurrence of NO_2 episodes since 1988 shows that such episodes have become less common and that the majority are winter episodes in most years. Years with major photochemical episodes such as 1990 and 1995 are dominated by summer episodes. There is some evidence that summer episodes are becoming relatively less important than winter episodes in terms of their contribution to the total number of hours with concentrations in excess of 200 µg m^{-3}.
- A comparison of measured hourly NO_x and NO_2 concentrations during an episode in December 2001 and episodes in the early 1990s shows that the reaction of NO with O_2 can still lead to significant exceedences of 200 µg m^{-3} during extreme weather conditions, even with the decline in NO_x emissions from traffic since early 1990s.
- Northern hemisphere mean O_3 baseline concentrations have shown an upward trend at the Mace Head site on the West Coast of Ireland of 1 µg m^{-3} (0.5 ppb) per year over the period from 1987–2001. An increase in the oxidising capacity of rural air entering urban areas of this magnitude would be expected to increase urban NO_2 levels.

- The impact of the increasing northern hemisphere O_3 baseline concentrations on O_3 and NO_2 levels in urban areas of the United Kingdom may have been confounded by the decreasing intensity of summertime photochemical O_3 episodes and by decreasing NO_x depletion reactions during wintertime.

6.1 Introduction

608. This chapter presents an analysis of current (2001) NO_2 concentrations in the UK and trends in concentrations over the last 10 to 20 years from a combination of information from both monitoring and modelling studies.

609. Automatic NO_x and NO_2 monitoring data for a total of 212 monitoring sites in 2001 have been compiled for this report and the measured concentrations have been compared with the 40 µg m^{-3} concentration threshold set in the AQS objective for 2005 and limit value (LV) for 2010, as well as the associated 1-hour criterion of no more than 18 exceedences of 200 µg m^{-3}. Fifteen sites have been selected for additional detailed analysis of recent trends and of hourly data during 2001. Meteorological and traffic flow information is available for a number of roadside and kerbside sites on an hourly basis and an analysis can provide information on the emissions and processes that influence roadside NO_2 concentrations.

610. The trends in annual mean NO_x and NO_2 concentrations averaged over London have been analysed. Site-specific projection models can be used to reconcile the trends in ambient NO_x concentrations over the last 10–20 years with the changes in emissions, by a combination of the source apportionment of current measured concentrations, and published emission inventories. A detailed examination of the measured trends at the Marylebone Road monitoring site, however, shows that the trends at an individual site can be highly dependent on local influences.

611. A combination of monitoring and modelling methods have been used to assess the impact of NO_x emissions from road traffic, elevated sources such as power stations, and airports, on local NO_x and NO_2 concentrations.

612. Maps of ambient NO_2 concentrations in the UK in 2001 have been calculated for policy analysis and for reporting to the European Commission. These maps provide an assessment of the locations in the UK where concentrations are likely to have exceeded an annual mean concentration of 40 µg m^{-3}. More detailed modelling studies have been carried out for London and the results of these studies are compared. London is of particular interest because it is the area with the highest annual mean NO_2 concentrations in the UK and the modelling results for London also aid the interpretation of the national modelling results in other areas.

613. Episodes when hourly mean NO_2 concentrations exceed 200 µg m^{-3} can be classified into two types. Winter episodes are associated with poor dispersion of primary NO_x emissions and elevated NO_2 concentrations are derived, at least in part, from the reaction of NO with O_2. Summer episodes are characterised by elevated secondary NO_2 concentrations associated with regional scale photochemical oxidant episodes. Examples of both types of episodes during 2001 are presented in order to illustrate the causes of these events along with an analysis of the relative frequency of summer and winter episodes in the UK since 1988.

614. The chapter concludes with a discussion of the evidence for an increase in regional mean O_3 concentrations over the last 10 years or so and the possible impact of this on recent or predicted NO_2 concentrations.

6.2 Measured NO_2 concentrations during 2001 and recent trends

6.2.1 Comparison of measured concentrations in 2001 with threshold concentrations

615. Automatic NO_x and NO_2 monitoring data for 2001 from a total of 212 sites have been collated for this report. Summary statistics have also been collected for earlier years and are available from: www.defra.gov.uk/environment/airquality/aqeg/index.htm[1]. Measurements are available from a considerably greater number of sites than for previous reviews such as the Photochemical Oxidants Review Group's 1990 report on oxides of nitrogen (PORG, 1990), which identified a total of only 21 monitoring sites operating as recently as mid 1989.

616. Table 6.1 shows the number of the NO_2 monitoring sites exceeding various threshold concentrations in 2001. The sites exceeding each threshold are listed in Appendix 4. Annual mean NO_2 concentrations above 40 µg m^{-3} were measured in 2001 at the majority of roadside and kerbside sites in London and at about half of the background sites. Exceedences outside London were largely confined to roadside and kerbside monitoring sites. The annual mean limit value + margin of tolerance[2] defined within the First Daughter Directive for 2001 is 58 µg m^{-3}. The majority of the exceedences of this threshold are at roadside and kerbside sites in London.

Table 6.1 Status of NO_2 monitoring sites in 2001 (R and K = Roadside and Kerbside sites, Other = all other classifications[a]).

		Total number of sites	**No. sites annual average > 40 µg m^{-3} (%)[b]**	**No. sites annual average > 58 µg m^{-3} (%)[c]**	**No. sites where no. of hours NO_2 >200 µg m^{-3} > 18 (%)[d]**	**No. sites where running 3-hrs NO_2 >400 µg m^{-3} (%)[e]**
Scotland	R and K	2	1 (50)	1 (50)	1 (50)	0
	Other	5	2 (40)	0	0	0
Wales	R and K	1	0	0	0	0
	Other	3	0	0	0	0
Northern Ireland	R and K	0	0	0	0	0
	Other	2	0	0	0	0
London	R and K	41	39 (95)	16 (39)	5 (12)	0
	Other	36	17 (47)	2 (6)	1 (3)	0
Rest of England	R and K	33	18 (54)	3 (9)	2 (6)	0
	Other	89	6 (7)	0	4 (4)	1 (1)
UK	R and K	77	58 (75)	20 (26)	8 (10)	0
	Other	135	25 (71)	2 (1)	5 (4)	1 (1)

[a] definitions of the different site types are listed in Appendix 3
[b] annual mean objective for 2005 and annual mean limit value for 2010
[c] annual mean limit value + margin of tolerance for 2001
[d] 1-hour mean objective for 2005 and annual mean limit value for 2010
[e] EU Daughter Directive Alert Threshold

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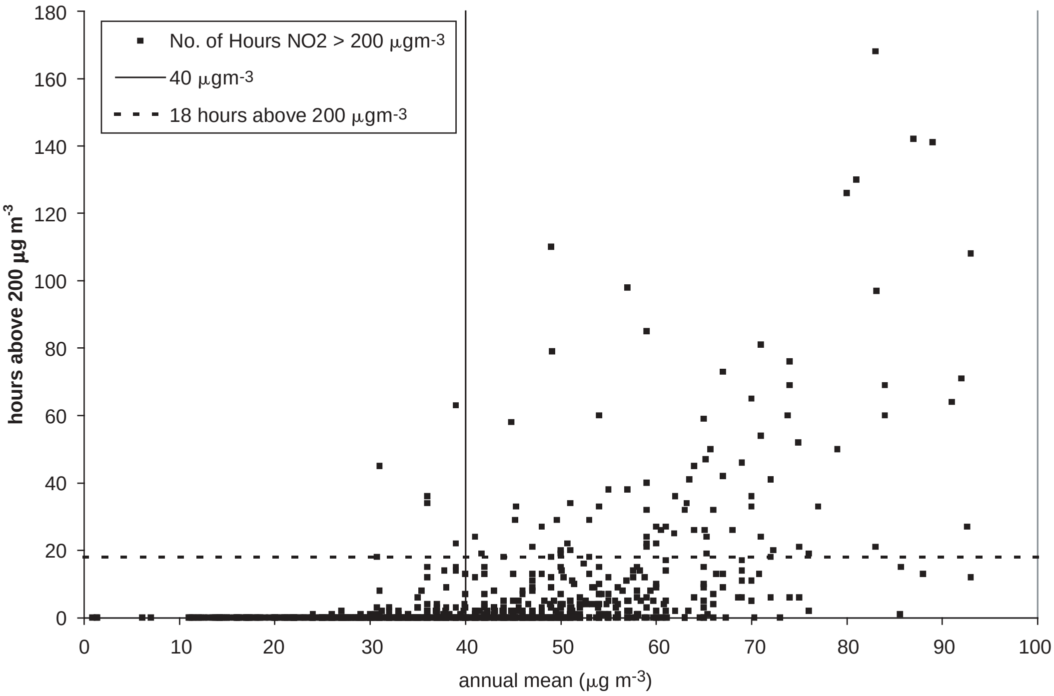
[1] It should also be noted that only data from the monitoring networks described in Chapter 4 have been collated. Although additional monitoring activity is undertaken in the UK, primarily by local authorities and commercial organisations, our analysis has focussed on data from fixed long-term monitoring sites from networks with known quality assurance procedures.

[2] This threshold was chosen to identify locations at risk of an exceedence of 40 µg m^{-3} remaining at a monitoring site in 2010.

617. Sites with more than 18 hours with measured hourly mean concentrations greater than 200 µg m^{-3} were less common than exceedences of an annual mean concentration of 40 µg m^{-3} in 2001. That this is generally the case is confirmed by Figure 6.1, in which the number of hours with concentrations greater than 200 µg m^{-3} has been plotted against the annual mean NO_2 concentration for all of the data collected for this report[3]. Out of the 933 site years of data shown on this figure, 477 (51%) had an annual mean concentration greater than 40 µg m^{-3} and 86 (9%) had more than 18 hours above 200 µg m^{-3}. 81 out of the 86 (94%) of the site years with more than 18 hours above 200 µg m^{-3} also had annual mean concentrations greater than 40 µg m^{-3}. Of the five site years with more than 18 hours above 200 µg m^{-3} but annual mean of 40 µg m^{-3} or lower, all were in Greater Manchester, four in 2001 and one in 1999. These exceedences were due to unusual episode conditions as discussed in Section 6.5. The annual mean is thus the key assessment level for nitrogen dioxide.

Figure 6.1 Comparison of annual mean NO_2 concentrations and the number of hours with concentration greater than 200 µg m^{-3} 1978-2001.



618. There was one measured exceedence of the EU alert threshold for NO_2 of a 3-hour mean concentration greater than 400 µg m^{-3} during 2001. This was at the Stockport Bredbury site. Since this site is not part of the UK national networks designated for reporting to the European commission, this did not trigger an alert.

619. Maps showing measured annual mean NO_2 concentrations at background and roadside/kerbside locations in 2001 in the UK are presented in Figures 6.2 and 6.3.

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[3] The data points for the Redbridge 2 site have been excluded in order to show the remaining data on an expanded scale. Annual means of 120 and 121 µg m^{-3} and 826 and 849 hours above 200 µg m^{-3} were recorded at this site in 2000 and 2001 respectively.

Figure 6.2 Measured annual mean NO_2 concentrations in 2001 at background sites ($\mu g\ m^{-3}$).

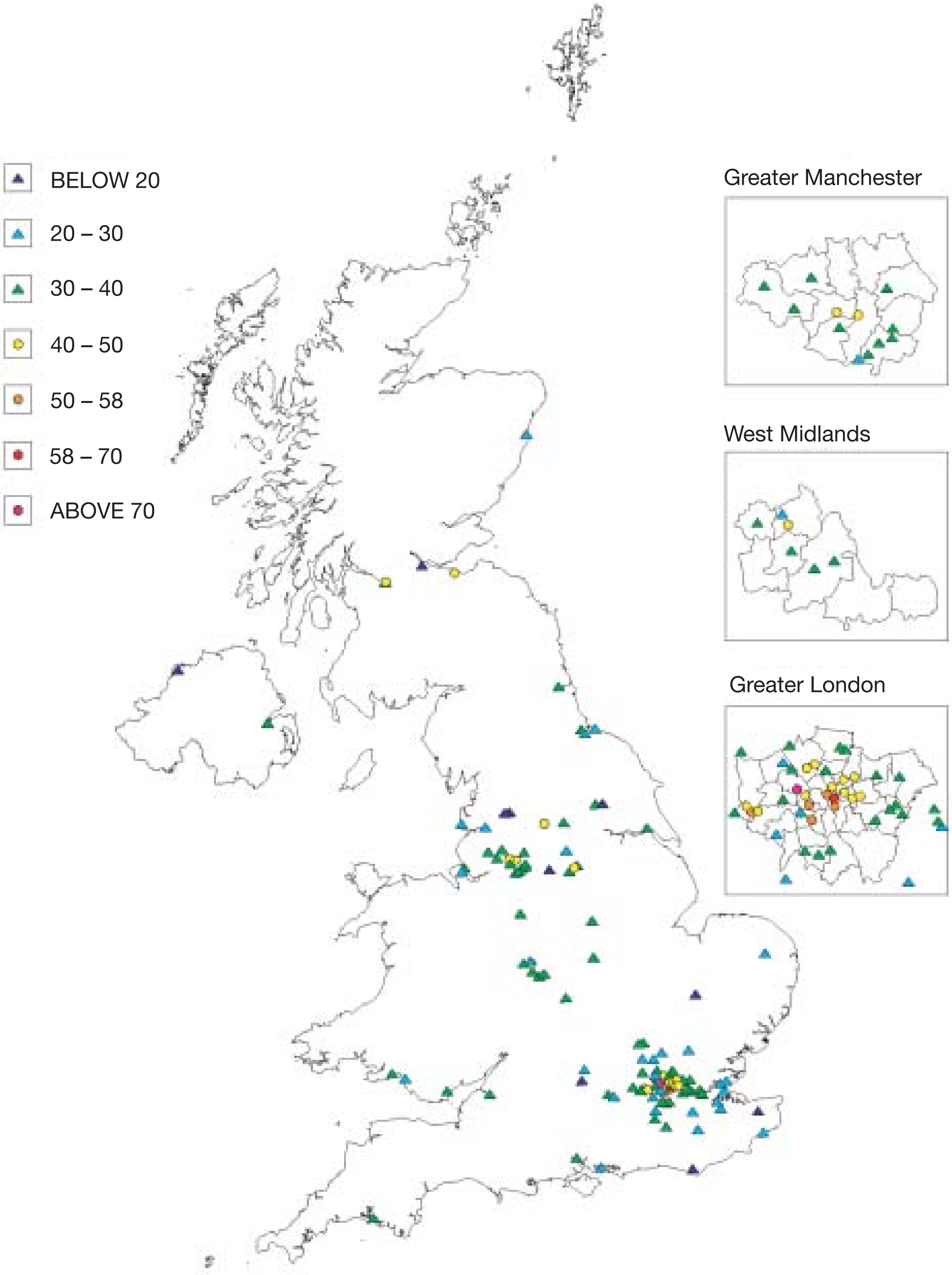
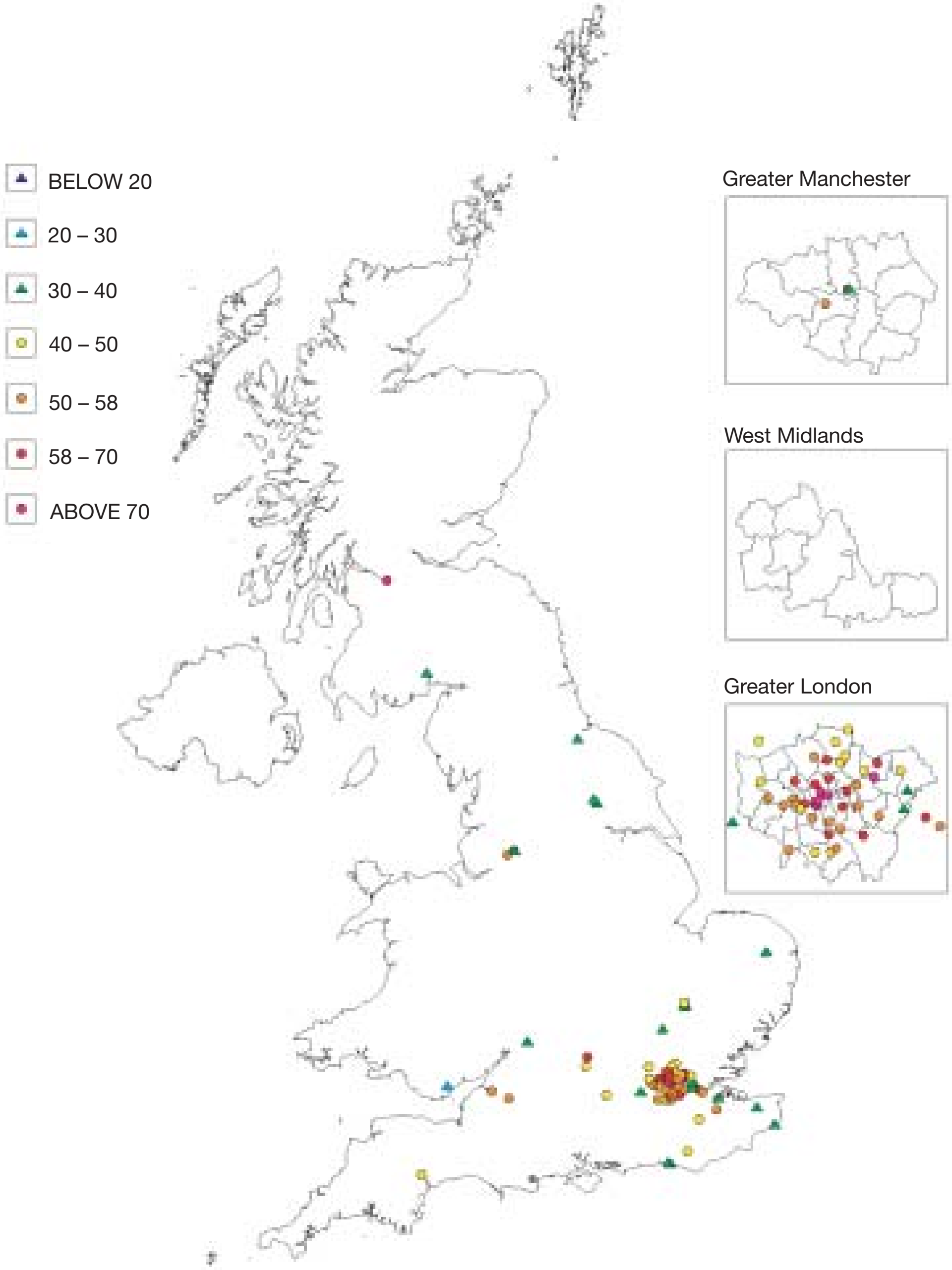


Figure 6.3 Measured annual mean NO_2 concentrations in 2001 at roadside and kerbside sites (µg m^{-3}).



6.2.2 The sites selected for detailed analysis

620. The sites selected for detailed analysis are listed in Table 6.2. These sites have been chosen in order to illustrate a number of important features of measured NO_2 concentrations in the UK. London Bloomsbury and Marylebone Road sites have been chosen to represent urban background and concentrations close to a very busy urban road in central London. The remaining background sites have been chosen to represent the gradient in concentrations from central London through the suburbs to the rural site at Lullington Health and to represent concentrations in different parts of the UK over the last 10 years or so. The Bury Roadside and M25 Staines sites have been chosen to illustrate the impact of emission from motorways on ambient NO_x and NO_2 concentrations. The Haringey Roadside and Redbridge 2 sites provide a contrast between the concentrations measured alongside a relatively busy A-road at Haringey

Roadside and on a traffic island on a very busy and congested one way system at Redbridge 2. It is important to recognise that every site, and particularly roadside and kerbside sites, are subject to local features (see section 6.2.6). While it is not always possible to generalise from the results of measurements at a single location, it is clear that high quality measurement data are key to an understanding of NO_2 concentrations in the UK.

621. Annual mean concentrations and the annual NO_2/NO_x ratio for 2001 for these sites are also listed in Table 6.2. This ratio varies from 0.20 at M25 Staines to 0.84 at Lullington Heath.

Table 6.2 Annual mean concentrations of NO_x and NO_2 in 2001.

Site	Type	NO_x (µg m^{-3}, as NO_2)	NO_2 (µg m^{-3})	NO_2/NO_x	Data capture (%)
Marylebone Road	Kerbside	335	84	0.25	93.8
Glasgow Kerbside	Kerbside	284	71	0.25	98.8
Haringey Roadside	Roadside	120	48	0.40	97.9
Redbridge 2	Roadside	355	120	0.34	87
M25 Staines	Kerbside	196	40	0.20	98
Bury Roadside	Roadside	270	69	0.26	97.7
Lullington Heath	Rural	15	12.6	0.84	93.5
London Bridge Place (1999)	Urban background	105	63	0.60	86.1
London Bloomsbury	Urban centre	109	51	0.47	86.9
West London	Urban background	95	52	0.55	95.4
London Bexley	Urban background	68	36	0.53	93.8
Manchester Town Hall	Urban background	92	47	0.51	98.5
Glasgow City Chambers	Urban background	107	46	0.43	98.7
Belfast Centre	Urban centre	58	32	0.55	87.9
Port Talbot[a]	Urban background	38	22	0.58	95.5

[a] Near a major steelworks

6.2.3 Recent trends in measured NO_x and NO_2 concentrations at selected sites

622. At the 13 monitoring sites for which there are long-running time series of data, clear downwards trends have been observed in NO_2 and NO_x concentrations during the 1990s. Table 6.3 presents the trends in annual mean NO_x and NO_2 concentrations during the 1993-2002 period, determined using a Mann-Kendall test with linear slopes estimated using Sen's method. Averaged over all 13 sites, annual mean NO_x concentrations show a -5.1% per year decline. The individual trends are all statistically significant at the 10% level of significance or better, except for the Edinburgh Centre site. Whereas twelve sites show trends in the range – 3.9 to -8.3% per year, Edinburgh Centre shows a trend of -1.9% per year. There appears to be little difference between the trends in annual mean NO_x concentrations recorded at the two rural sites, Lullington Heath and Ladybower, and the remaining 9 urban background sites.

Table 6.3 Trends in annual mean NO_x and NO_2 concentrations at 13 selected long-running sites over the period 1993-2002.

Site	Trend in annual mean NO_x µg m^{-3} per year or % /yr		Trend in annual mean NO_2 µg m^{-3} per year or % /yr	
Lullington Heath	-0.9	-5.6	-0.8	-5.4
Ladybower	-0.9	-5.1	-0.7	-5.1
Belfast Centre	-3.4	-4.9	-1.3	-3.7
Cardiff Centre	-4.2	-6.1	-1.2	-3.3
Newcastle Centre	-6.9	-8.3	-2.7	-7.1
Birmingham Centre	-4.8	-6.4	-1.8	-4.4
Manchester Town Hall	-2.7	-2.9	-1.6	-3.4
Walsall Alumwell	-5.1	-4.9	-1.1	-2.5
Edinburgh Centre	-1.9	-1.9	-0.8	-1.8
Sheffield Tinsley	-7.3	-5.8	-1.4	-2.9
Glasgow City Chambers	-5.1	-4.4	-0.3	-0.6
West London	-6.1	-5.5	-0.8	-1.6
London Bloomsbury	-5.1	-3.9	-1.9	-3.0

Using a Mann-Kendall test with linear slopes estimated using Sen's method.
The shaded areas show those trends that are statistically significant at the 10% level of significance or better.

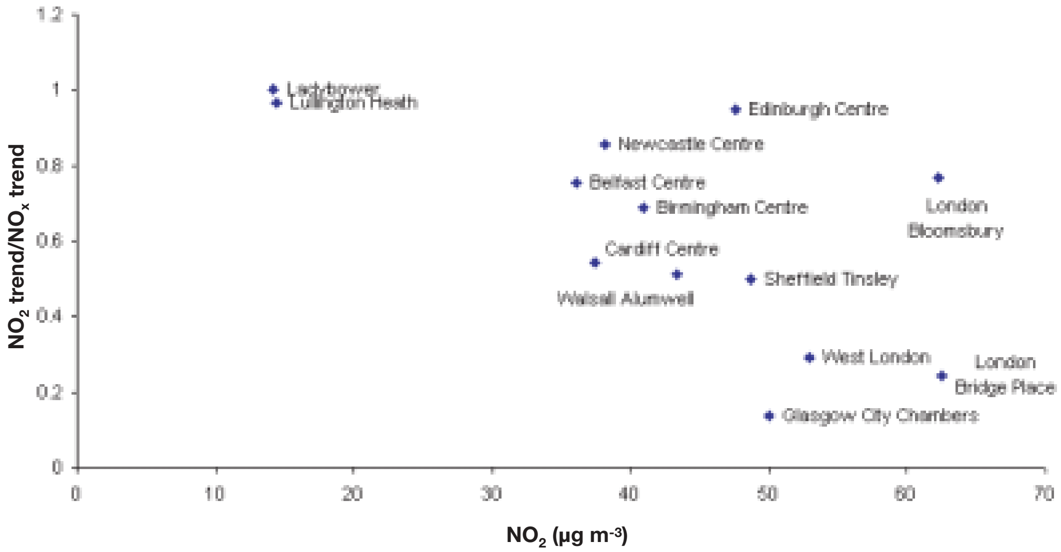
623. The network-average observed NO_x trend of -5.1 ± 1.6 % per year over the period 1993-2002 period appears to be slightly higher than the trend in the UK NO_x emissions, either of total NO_x emissions, -4.4 % per year, or of road transport NO_x emissions, -4.3 % per year, over the 1993-2000 period, see Chapter 2. Better agreement between the observed network mean NO_x trend and the decline in UK NO_x emissions might have been anticipated in view of the stated accuracy of the latter. However, without an explanation of why the observed trends at the Edinburgh Centre and Manchester Town Hall sites, for example, are so different from the network average, it is difficult to take further any comparisons between the observed and emission inventory trends. The discrepancies in trends may point to a slight underestimation of motor vehicle NO_x emissions in the UK inventories.

624. The observed trends in NO_2 and NO_x at the rural sites, Lullington Heath and Ladybower, are highly statistically significant and are similar, lying in the range -5.1 to -5.6% per year. The similarity in the NO_x and NO_2 trends is consistent with the view that in rural locations there is generally enough oxidant to convert all the available NO to NO_2, particularly at nighttime and during the summertime. Under these conditions, rural NO_2 concentrations are generally determined by the availability of NO_x. The trends in NO_x and NO_2 should therefore be closely related on both a % per year and µg m^{-3} basis, and this is what is observed.

625. At urban background sites, trends in annual mean NO_2 concentrations are also statistically significant at most sites, with the exception of the Glasgow City Chambers, Manchester and London Bloomsbury sites. However, Table 6.3 shows that the trends in NO_2 are distinctly smaller than those observed in annual mean NO_x concentrations at the same sites. Averaged over the urban background and kerbside sites, annual mean NO_2 concentrations showed a downwards trend of -3.1 ± 1.7% per year, about two-thirds of the simultaneous NO_x trend.

The discrepancy between these paired NO_2 and NO_x trends is least for the Edinburgh Centre site and greatest for the West London site.

Figure 6.4 Plot of the ratios of the trends in annual mean NO_2 concentrations to those in NO_x on a % per year basis against the observed 10-year mean NO_2 concentrations.



626. Figure 6.4 plots out the ratios of the observed trends in annual mean NO_2 concentrations to those in NO_x against the observed 10-year mean NO_2 concentrations at each of the sites. The trends in NO_2 relative to those in NO_x, decline with increasing average NO_2 level. The rural sites clearly show up with identical slopes as discussed above. There are then a group of seven urban background sites which show ratios in their NO_2 to NO_x trends of about 0.7 on a % per year basis. Such ratios strongly support the empirical relationships between NO_2 and NO_x discussed in Chapter 5. At these urban background sites, availability of NO_x and oxidant, jointly control the observed NO_2 concentrations. They also confirm that in the past, such sites have moved along the same NO_x to NO_2 relationship between years as that defined for one year between the sites. This is an important verification of the assumptions inherent in the empirical models of NO_2 and NO_x described in Chapter 5.

627. There are two of the sites in Figure 6.4 whose NO_2 trends are significantly lower compared to their NO_x trends and these are the West London and Glasgow City Chambers sites. Also added is an additional point for the London Bridge Place site estimated using the 1993-1999 data record. At these three sites, NO_2 trends are small on both a µg m^{-3} or % per year basis. The different relative trends in NO_2 and NO_x observed at these three sites points to there being different site-specific NO_2 trends in the future and to different site-specific expectations from future NO_x emission reductions. At present, it is not clear what the detailed factors are which have led to this different behaviour at these three sites. Evidently, these sites may not fit in with the simplified relationships implied in the empirical models described in Chapter 5.

Table 6.4 Estimates of the trends in monthly mean NO_x concentrations in µg m^{-3} per year over the period 1993-2002 for 13 selected long-running sites.

	LaB	LH	BeC	CC	NC	BiC	MT	WA	EC	ST	GC	WL	LoB
Jan	-0.6	-0.3	-1.8	-3.4	-2.8	-1.9	-2.7	-3.3	+3.6	-2.5	+0.5	-2.5	-3.4
Feb	-2.1	-2.7	-3.0	-6.7	-6.5	-7.0	-6.6	-5.3	-0.4	-8.8	-8.4	-8.0	-7.7
Mar	-0.3	-1.1	-1.5	-3.9	-4.8	-3.5	-1.6	-4.0	-1.6	-8.3	+1.0	-5.0	-4.2
Apr	-0.6	-1.0	-2.0	-3.3	-7.5	-3.7	-3.2	-3.5	-3.8	-6.3	-4.5	-4.0	-1.6
May	-0.6	-0.9	-2.5	-2.7	-8.0	-4.3	-2.5	-3.6	-0.7	-4.8	-5.3	-3.2	-5.0
Jun	-0.6	-0.6	-1.9	-2.8	-6.5	-3.3	-3.0	-4.2	-2.2	-5.5	-5.2	-4.0	-3.1
Jul	-0.9	-0.3	-1.5	-2.9	-4.5	-2.0	-2.7	-4.7	-3.5	-8.0	-3.3	-3.3	-2.3
Aug	-1.3	-0.4	-1.5	-3.3	-4.8	-3.0	-3.9	-4.0	-3.5	-7.0	-4.5	-4.7	-2.9
Sep	-1.1	-1.0	-4.8	-6.0	-5.4	-4.3	-5.3	-6.0	-3.8	-6.5	-7.0	-7.5	-5.3
Oct	-1.0	-0.8	-7.7	-5.0	-10.	-8.3	-11.	-10.	-4.3	-13.	-11.	-9.0	-7.0
Nov	-2.2	-0.8	-8.0	-8.5	-8.7	-10.	-9.3	-10.	-1.8	-12.	-9.9	-12.	-12.
Dec	-0.2	-0.9	-4.3	-4.6	-7.1	-3.4	+10.	-12.	+1.5	-10.	-0.3	-4.5	-7.5

LaB, Ladybower; LH, Lullington Heath; BeC, Belfast Centre; CC, Cardiff Centre; NC, Newcastle Centre; BiC, Birmingham Centre; MT, Manchester Town Hall; Walsall Alumwell; EC, Edinburgh Centre; ST, Sheffield Tinsley; GC, Glasgow City Chambers; WL, West London; LoB, London Bloomsbury.
The shading shows those trend estimates that are statistically significant at better than the 10% level of significance.

628. Monthly mean NO_x concentrations show similar downwards trends at urban background sites compared with those shown by the annual mean NO_x concentrations. In Table 6.4, the trends in the January monthly mean NO_x concentrations, February mean and so on, are shown for the 1993-2002 period. The columns for Cardiff Centre and Newcastle Centre, for example, show that downwards trends in most months are statistically significant and compare closely in magnitudes with those of the annual mean concentrations in Table 6.3.

629. At the rural sites, Ladybower and Lullington Heath, monthly mean NO_x trends show evidence of somewhat stronger downwards trends in wintertime compared with summertime. This seasonal variation is also clearly apparent at most urban sites. There are however some exceptions, with the Edinburgh Centre and Glasgow City Chambers sites showing upwards trends in the wintertime and downwards trends during the remainder of the year. The origins of the observed seasonal variations in monthly mean NO_x trends are not clear.

630. The monthly mean NO_2 trends are presented in Table 6.5 for the 1993-2002 period. The columns for Belfast Centre and Birmingham Centre, for example, show that the downwards trends in most months are statistically significant and compare closely in magnitude to those observed in the annual mean NO_2 concentrations in Table 6.3.

Table 6.5 Estimates of the trends in monthly mean NO_2 concentrations in µg m^{-3} per year over the period 1993-2002 for 13 selected long-running sites.

	LaB	LH	BeC	CC	NC	BiC	MT	WA	EC	ST	GC	WL	LoB
Jan	-0.3	0.6	-0.8	-0.7	-0.3	-0.5	+0.4	0	+1.4	-0.2	+1.6	+1.4	-1.8
Feb	-1.7	-0.5	-1.3	-1.8	-1.8	-2.2	-1.0	-0.6	0	-0.5	-0.9	+1.0	-2.9
Mar	-0.3	-0.4	-1.0	-1.7	-1.4	-1.3	-0.5	-0.4	-0.8	-0.5	+0.3	+0.3	-2.8
Apr	-0.6	-0.5	-1.3	-1.7	-1.7	-1.8	-1.0	-1.1	-1.8	-0.8	-1.0	-1.0	-1.6
May	-0.5	-0.4	-1.4	-1.7	-2.0	-2.9	-1.0	-1.7	-1.0	-0.1	-0.6	0	-2.6
Jun	-0.5	-0.3	-1.0	-1.4	-2.0	-2.1	-1.5	-0.6	-1.2	-0.5	-0.5	-0.4	-2.0
Jul	-0.8	-0.2	-0.8	-1.1	-1.3	-1.8	-1.0	-0.7	-1.7	-2.0	-0.5	0.5	-1.8
Aug	-1.0	-0.4	-0.5	-1.7	-1.2	-1.5	-0.7	+0.2	-2.2	-1.7	-0.4	-0.8	-2.0
Sep	-0.9	-0.3	-1.3	-1.9	-0.7	-2.3	-0.3	-0.4	-1.7	-1.3	-1.2	-1.3	-1.1
Oct	-0.9	0.5	-2.3	-1.9	-1.8	-2.7	-0.7	-1.7	-1.3	-1.5	-0.8	-1.0	-1.9
Nov	-1.7	0	-2.1	-1.7	-0.3	-2.5	-1.1	-1.0	+0.7	-1.0	0	0	-2.2
Dec	-0.2	-0.6	-1.4	-0.6	-1.5	-1.3	0	-1.5	+0.8	-1.3	+0.2	-0.5	-2.0

LaB, Ladybower; LH, Lullington Heath; BeC, Belfast Centre; CC, Cardiff Centre; NC, Newcastle Centre; BiC, Birmingham Centre; Walsall Alumwell; EC, Edinburgh Centre; ST, Sheffield Tinsley; GC, Glasgow City Chambers; WL, West London; LoB, London Bloomsbury.
The shading shows those trend estimates that are statistically significant at better than the 10% level of significance.

631. However, at the more polluted urban background sites, towards the right hand side of Table 6.5, the monthly trends become much less pronounced and deviate further from the annual trends shown in Table 6.3. The deviations tend to be preferentially found during the winter months. At the Glasgow City Chambers and West London sites, the monthly mean NO_2 concentrations during winter months show increasing trends rather than the decreasing trends shown at the other urban background sites. Furthermore, these sites also show much reduced declines during summer months.

632. The lessened downwards trends and increasing trends in winter monthly mean NO_2 concentrations observed at some sites, deserve further comment. At the Glasgow City Chambers and Edinburgh Centre sites, the increasing winter NO_2 trends may have been driven by the observed increasing trends in winter NO_x concentrations observed simultaneously. At the West London site, the winter trends are increasing whilst the corresponding NO_x trends are strongly decreasing, as expected. At the Sheffield Tinsley, Cardiff Centre, Newcastle Centre, Birmingham Centre and Walsall Alumwell sites, winter NO_2 concentration trends are barely downwards, being close to level whilst simultaneous NO_x trends are strongly downwards. Plausible explanations may be the increasing trend in northern hemisphere baseline O_3 concentrations or increasing direct emissions of NO_2. However, without an understanding of the upwards trends in winter NO_x concentrations observed at the Glasgow City Chambers and Edinburgh sites, it is difficult to take the examination of wintertime NO_2 trends further.

633. Figures 6.5 and 6.6 show that annual mean concentrations of NO_x at urban background and urban centre sites have declined over the past decade with NO_2 showing a far more modest fall at most sites with some sites showing no obvious temporal trend. Figures 6.7 and 6.8 show the trends at kerbside and roadside monitoring sites, for which data are only available for a much shorter period. Most of the kerbside and roadside sites show a substantial reduction in mean NO_x between 1996/7 and 2001, but little change in NO_2.

634. Figure 6.10 shows that peak (99.8%ile) hourly winter concentrations of NO_2 are very variable from year to year, with London showing particularly high concentrations in 1991 and Manchester in 1994 (18 exceedences of 200 μg m^{-3} is equivalent to a 99.8%ile of 200 μg m^{-3}). Figure 6.9 shows that 99.8%ile concentrations of NO_x show less inter-annual variability. This is presumably due to the NO + NO + O_2 reaction making a significant contribution to peak winter NO_2 only during rare winter episode conditions (see section 6.5.3). Peak (99.8%ile) hour winter concentrations of both NO_2 and NO_x show a steady decline at background sites between 1990 and 2001. Peak (99.8%ile) hourly concentrations of NO_2 and NO_x are substantially lower in summer than in the winter months (Figures 6.11 and 6.12).

635. Figure 6.13 shows annual mean NO_2 concentrations plotted against annual mean NO_x at the selected urban background and urban centre sites since 1983 along with the empirical relationships for background sites derived from monitoring data from 1998 to 2001 for a much larger number of UK network sites (discussed in section 5.2.1.2). This comparison provides some confirmation of the validity of these relationships because the changes in concentrations over time at a single location are seen to be in reasonable agreement with those predicted by comparing concentrations measured at different locations over a much shorter time span. Close examination of Figure 6.13 does however suggests that the reductions in NO_2 concentrations at individual sites show some tendency to be slightly less than suggested by the curves. The only site that clearly deviates from this pattern is Glasgow City Chambers where annual mean NO_x concentrations have declined from about 160 μg m^{-3}, as NO_2 in the early 1990s to about 100 μg m^{-3}, as NO_2 in 2000, while annual mean NO_2 concentrations have remained roughly constant at about 50 μg m^{-3}. This is discussed further in section 6.6.3.

Figure 6.5 Annual mean NO_x concentrations at selected background sites.

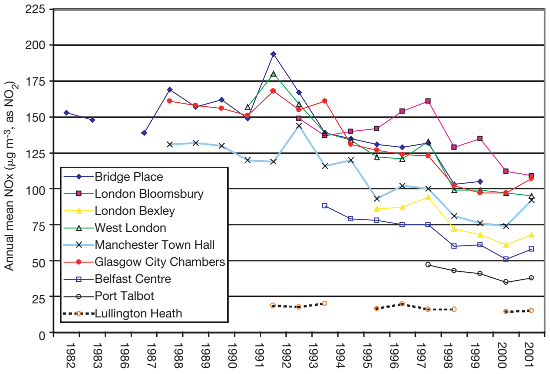


Figure 6.6 Annual mean NO_2 concentrations at selected background sites.

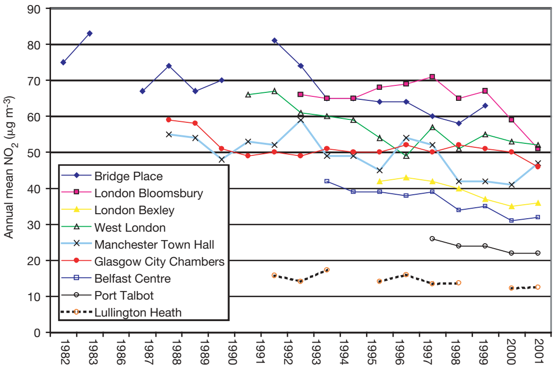


Figure 6.7 Annual mean NO_x concentrations at selected background sites.

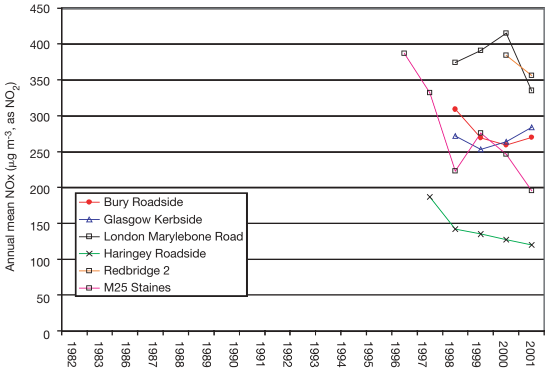


Figure 6.8 Annual mean NO_2 concentrations at selected roadside and kerbside sites.

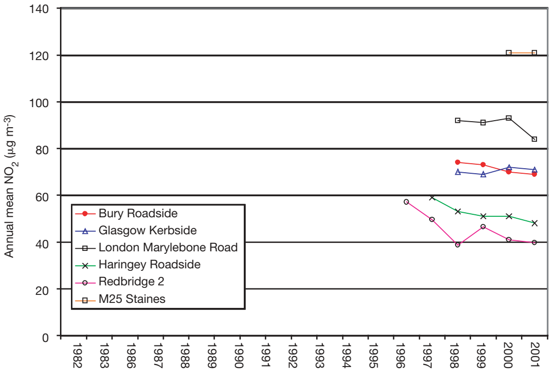


Figure 6.9 Split winter 99.8%ile of hourly average NO_x concentration at selected background sites.

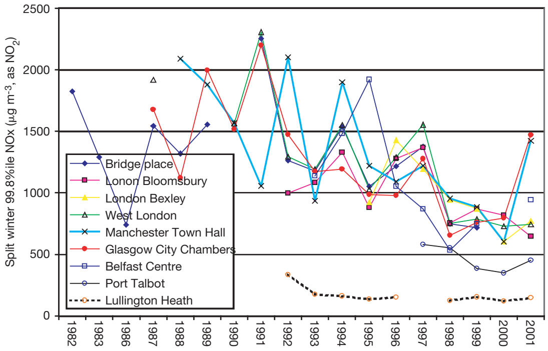


Figure 6.10 Split winter 99.8%ile of hourly average NO_2 concentration at selected background sites.

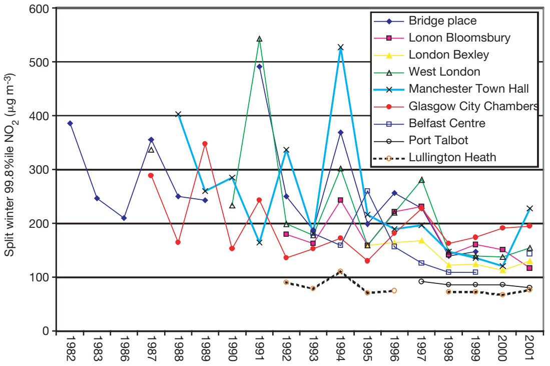


Figure 6.11 Summer 99.8%ile of hourly average NO_x concentration at selected background sites.

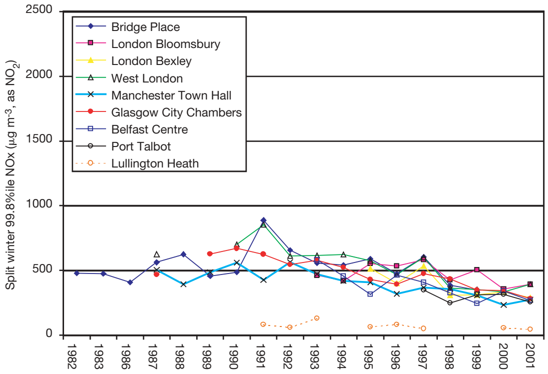


Figure 6.12 Summer 99.8%ile of hourly average NO_2 concentration at selected background sites.

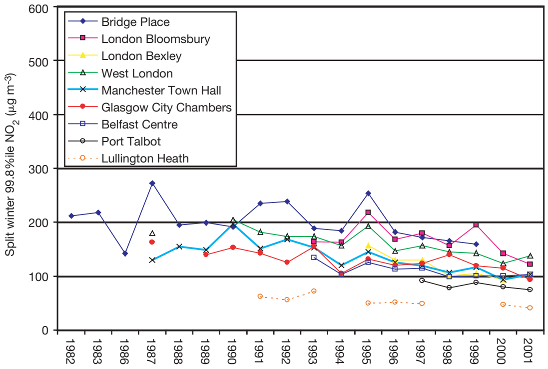
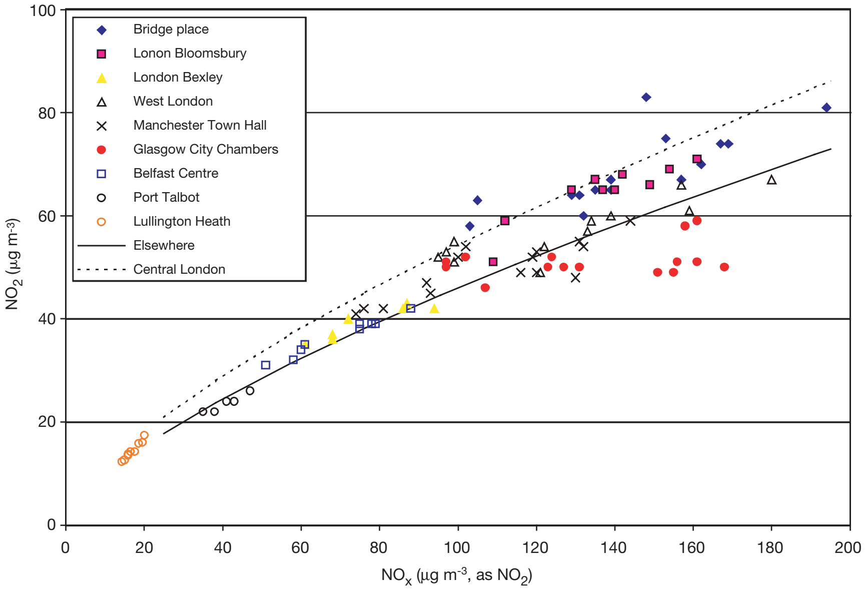


Figure 6.13 Measured annual mean NO_x and NO_2 concentrations at selected background sites 1983 – 2001.



6.2.4 Analysis of hourly NO_x and NO_2 meteorological and traffic count data in 2001 at selected roadside and kerbside monitoring sites in 2001

636. Meteorological data and traffic flow information have been included in the analyses presented here, where available. Meteorological data from Barking and Dagenham 1 has been used for the Marylebone Road, Haringey Roadside and Redbridge 2 sites (this site was chosen to represent wind speeds and directions in the London area. Wind speeds and directions are, of course, very different within the urban canopy, as discussed in Chapter 3). Hourly traffic count data are available for the Marylebone Road and M25 Staines sites; data from the M60 Kirkhams site has been used for the Bury Roadside site.

637. The roadside increment of hourly NO_x and NO_2 concentrations has been calculated at roadside and kerbside monitoring sites by subtracting hourly concentrations measured at nearby background sites from the concentration measured at the roadside or kerbside site. This enables an assessment of the contribution to ambient concentrations from the NO_x sources adjacent to the monitoring sites (that is, the traffic on the road). This approach has been used previously to provide useful insights into traffic increment of roadside PM_{10} concentrations (APEG, 1999). This approach is clearly not perfect since the background sites used are also network monitoring sites and were not established in order to provide the best possible estimate of background concentrations for this type of paired site analysis. The combination of both primary and secondary contributions to ambient NO_2 concentrations also complicates the interpretation of the results. The background sites chosen to calculate the roadside increments at the roadside and kerbside sites selected for analysis are listed in Table 6.6, along with manual and automatic traffic count information, where available. The inlets at all of the roadside and kerbside sites studied are 3 m above the ground except at M25 Staines and Redbridge 2 where the inlet is at 1.5 m. The height of the sampling point is important because the NO_x emissions from cars are very close to the surface of the road but the emissions from some HGVs are at a height of several metres.

Table 6.6 Details of 'paired' roadside and kerbside sites.

Site	Pairs with	Road number	Site location	Manual count 2000[a]	Automatic count 2001[a]
Marylebone Road	London Bloomsbury	A501	1 m from kerb, south of the road, which runs roughly east – west	71952	69700
Glasgow Kerbside	Glasgow City Chambers	–	1 m from kerb, east of the road, which runs roughly north – south	–	–
Haringey Roadside	Haringey 2	A1010	5 m from kerb, west of the road, which runs roughly north – south	13968	–
Redbridge 2	Redbridge 1	A123, A118, A118	1 m from kerb, on an island on a 1–way system. Roads go roughly north, south and west	16596, 25613, 24636	–
M25 Staines	London Teddington	M25	5 m from edge of carriageway, west of the motorway, which runs roughly north – south	194186	177800
Bury Roadside	Bolton	M60, A56	17m from edge of carriageway, north of the motorway, which runs roughly east – west and inside the junction roundabout.	15988 37568	130000

[a] annual average daily traffic flow

638. Table 6.7 shows the percentage contributions to road-link NO_x emissions from different vehicle types from the NAEI emission inventory for 2000. None of the sites analysed are dominated by emissions from cars, for which the contributions to emissions vary between 28% and 37%. Buses and HGVs make large contributions at some sites.

Table 6.7 Percentage of road link NO_x emissions by vehicle type (NAEI 2000).

	%bus	%car	%HGVa[a]	%HGVr[b]	%LGV	%mc[c]
Marylebone Road	17%	37%	9%	25%	12%	0%
Glasgow Kerbside[d]	–	–	–	–	–	–
Haringey Roadside	40%	32%	3%	17%	8%	0%
Redbridge 2	43%	35%	2%	13%	7%	0%
M25 Staines	2%	36%	43%	12%	7%	0%
Bury Roadside[e]	1%	28%	52%	14%	5%	0%

[a] articulated HGV

[b] rigid HGV

[c] motorcycles

[d] Traffic count and hence link emissions estimates are not available for Glasgow Kerbside

[e] motorway traffic only

639. Figures 6.14 and 6.15 illustrate that the hourly concentrations of NO_2 and NO_x at urban sites show the typical diurnal pattern of a traffic-generated pollutant with a substantial morning rush-hour peak (less marked for NO_2 than NO_x and an early evening peak. Figures 6.16 and 6.17 show the diurnal variation in NO_x and NO_2 concentrations at the Marylebone Road and London Bloomsbury sites along with the diurnal variation in the roadside increment of both NO_x and NO_2 concentrations (calculated as the difference between the concentrations measured at these two sites). The roadside increment of NO_x is greater than the background NO_x concentration in all hours of the day, while the roadside increment of NO_2 concentrations is always lower than the background NO_2 concentration. This illustrates that the production of secondary NO_2 close to the road is limited by the availability of oxidant.

640. Figure 6.18 shows the diurnal variation of traffic flow at the London Marylebone Road site in 2001, split by light-duty and heavy-duty vehicles. The heavy-duty vehicle flow is also shown multiplied by 5 in order to expand the scale. There is relatively less heavy-duty traffic in the afternoon and evening. Figure 6.19 shows the diurnal variation of wind speed at Barking and Dagenham 1. Figures 6.20 and 6.21 show comparisons of NO_x and NO_2 roadside increments at Marylebone Road, M25 Staines and Bury Roadside by hour against total traffic flow. Both the NO_x and NO_2 increments increase with traffic flow, as expected. The gradients are steepest at Marylebone Road, showing the less efficient dispersion of the emissions per vehicle at this location than at the two motorway sites (consistent with the street canyon location of the monitoring site). The gradient is lowest at the M25 Staines site, which is consistent with the fact that this site is adjacent to the clockwise carriageway of the motorway and upwind of the road in prevailing south-westerly winds. The Bury Roadside site is further from the edge of the motorway but is downwind of the carriageways in the prevailing wind direction. The results of this analysis of diurnal trends show that the increment of both NO_x and NO_2 concentrations is related to traffic flow but that the relationships is not necessarily simple. Another point to remember is that this diurnal analysis will also be influenced by variation in average meteorological conditions with time of day and that the lowest traffic flows and concentrations usually are in the middle of the night, when dispersion is less efficient due to lower windspeeds and a more stable atmosphere (see Figure 6.19). This may account for the apparent intercept in the relationship, which should logically go through zero.

Figure 6.14 Diurnal variation of NO_x concentrations at selected background sites in 2001.

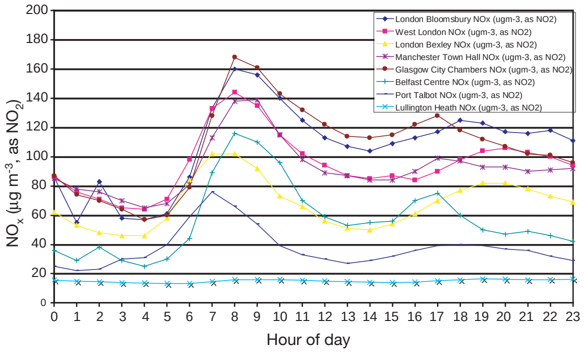


Figure 6.15 Diurnal variation of NO_2 concentrations at selected background sites in 2001.

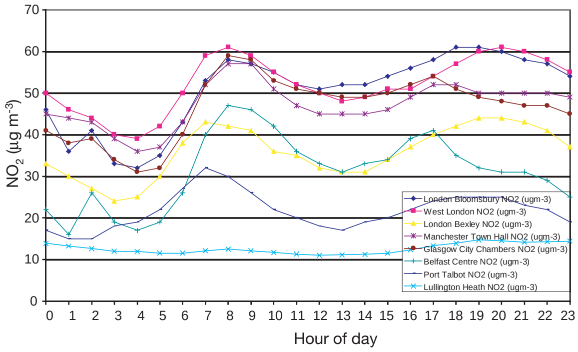


Figure 6.16 Diurnal variation of NO_x concentrations at Marylebone Road and London Bloomsbury in 2001.

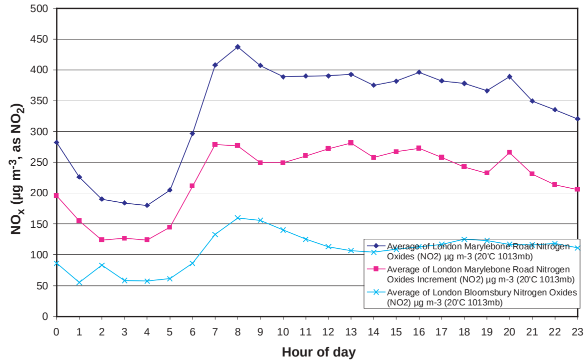


Figure 6.17 Diurnal variation of NO_2 concentrations at Marylebone Road and London Bloomsbury in 2001.

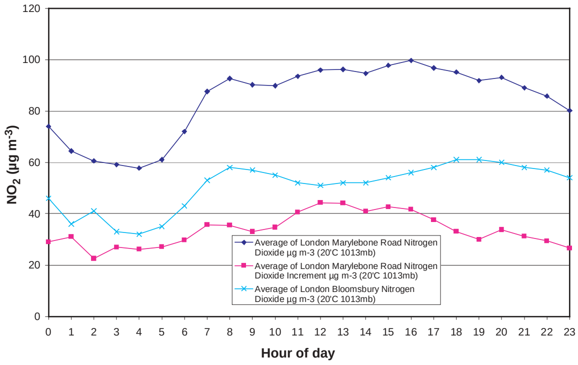


Figure 6.18 Diurnal variation of traffic flow at Marylebone Road in 2001.

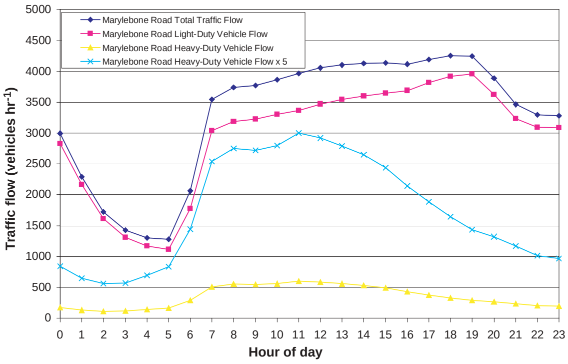


Figure 6.19 Diurnal variation of wind speed at Barking and Dagenham 1 in 2001.

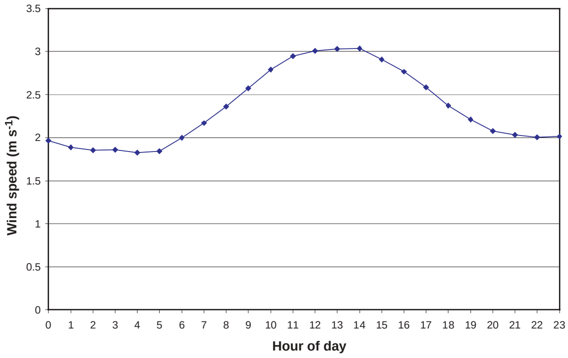


Figure 6.20 Comparison of roadside increment of NO_x concentrations with traffic flow at three sites in 2001.

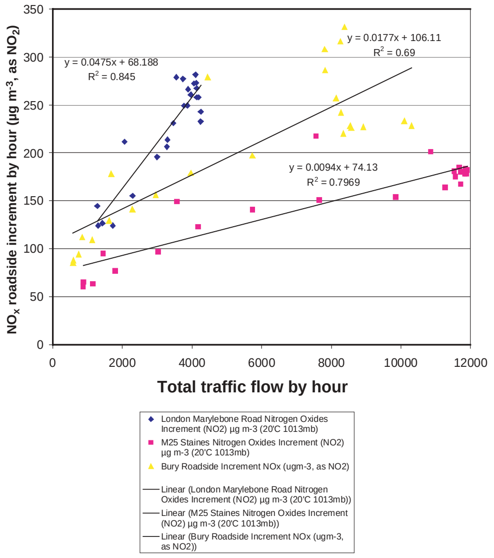
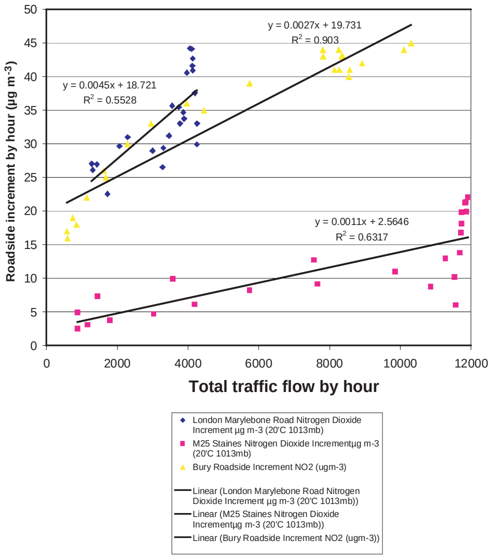
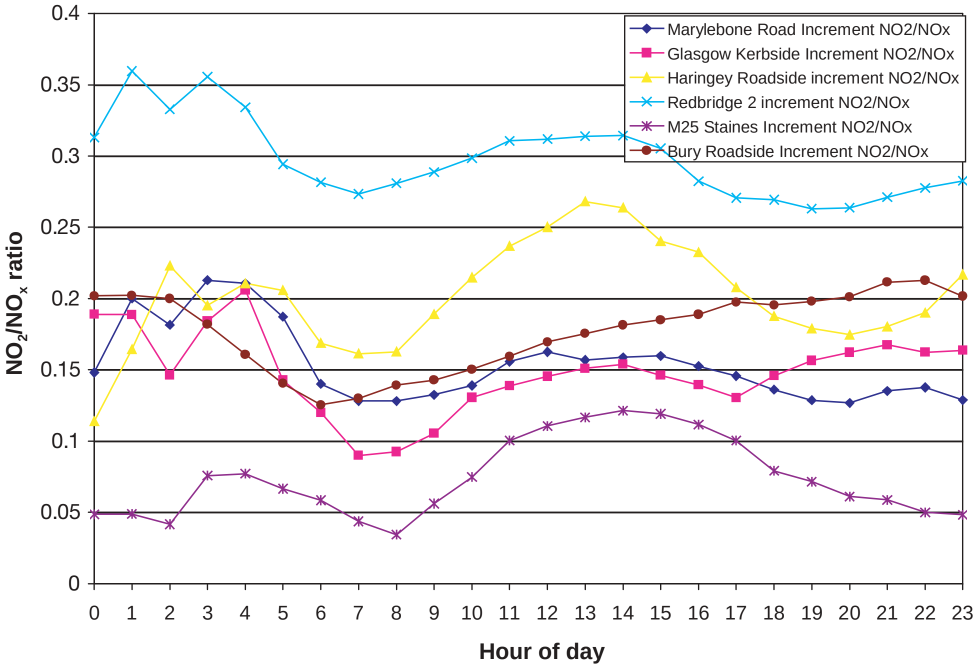


Figure 6.21 Comparison of roadside increment of NO_2 concentrations with traffic flow at three sites in 2001.



641. Annual average roadside increments in 2001 are listed in Table 6.8 along with the calculated NO_2/NO_x ratio for these increments. The total roadside NO_x concentration is dominated by local emissions from the road but the total roadside NO_2 concentration includes a large contribution from urban background NO_2 concentrations. The diurnal variations of the roadside increment NO_2/NO_x ratios are illustrated in Figure 6.22. The ratio is about 15% at both Marylebone Road and Glasgow Kerbside and rather higher at 20% at Haringey Roadside. This is consistent with the lower NO_x increment at this site (a similar amount of oxidant is available to oxidise less NO_x and a greater proportion is therefore converted to secondary NO_2). At the Redbridge 2 traffic island site, the traffic increment in NO_x has a high proportion (about 30%) of NO_2 which shows little diurnal variation. This site is in the middle of a busy junction with frequently congested traffic with a large proportion of bus traffic. This site is discussed in more detail below.

Figure 6.22 Diurnal variation of the NO_2/NO_x ratio of roadside increments in 2001.



642. The traffic increment NO_2 is less than 10% of NO_x at the M25 Staines site, peaking during the afternoon. This is a much lower percentage than at the other sites studied and may, at least partly, reflect the choice of background site (London Teddington), which is located in a park on the outskirts of London. The background NO_2 concentrations subtracted to calculate the roadside increment may therefore have overestimated the background NO_2 concentrations at the M25 Staines site. Another important influence may be the higher traffic speeds at this motorway site. The percentage of the roadside NO_x increment that is NO_2 is greater at the Bury Roadside site at 17%. In general higher NO_2/NO_x ratios seem to be associated with a greater proportion of the emissions being from diesel vehicles in slow moving congested traffic, while lower ratios are associated with fast moving motorway traffic.

Table 6.8 Measured annual mean concentrations and roadside increments at selected roadside and kerbside sites in 2001.

Site	Measured total concentrations			Calculated roadside increments		
	NO_x (µg m^{-3}, as NO_2)	NO_2 (µg m^{-3})	NO_2/ NO_x	NO_x (µg m^{-3}, as NO_2 and (%))	NO_2 (µg m^{-3} and (%))	NO_2/ NO_x
Marylebone Road	335	84	0.25	225 (67)	34 (40)	0.15
Glasgow Kerbside	284	71	0.25	177 (62)	25 (35)	0.14
Haringey Roadside	120	48	0.40	51 (42)	10 (21)	0.20
Redbridge 2	355	120	0.34	277 (78)	80 (67)	0.29
M25 Staines	196	40	0.20	148 (76)	11 (28)	0.08
Bury Roadside	270	69	0.26	202 (75)	35 (51)	0.17

643. Overall the observed NO_2/NO_x ratios of the roadside increments illustrate the importance of primary NO_2 emissions from traffic sources, since these ratios are considerably higher than would be expected if all of the roadside increment of NO_2 were derived by oxidation of emitted NO. The processes leading to the emission of primary NO_2 have been described in Chapter 2. The percentage of NO_x emitted directly as primary NO_2 from most combustion sources, such as petrol vehicles, is generally assumed to be about 5%. The percentage of NO_x emitted as NO_2 from diesel vehicles is thought to be higher than this, particularly for idling engines under very light loads, where the percentage could be up to 30%. The factors influencing the observed NO_2/NO_x ratio of the roadside increment are explored further below.

644. Figures 6.23 and 6.24 show the NO_2/NO_x ratio of the roadside increment by hour of the day, plotted against total roadside NO_x and total roadside NO_2 concentrations for all of the roadside sites analysed. The empirical relationship between this ratio and total NO_x derived from an analysis of annual means at a range of sites for the period 1998- 2001 and discussed in (section 5.2.1.3) is also shown. This shows that the NO_2/NO_x ratio is generally greatest at lower total roadside NO_x concentrations when more oxidant will be available and declines at higher NO_x. This suggests that the proportion of the roadside NO_2 increment represented by primary rather than secondary NO_2 will be greater at higher NO_x concentrations. The agreement between the empirical relationship and data by hour is good at all of the sites except Redbridge 2, with a higher ratio and M25 Staines, with a lower ratio. Figure 6.24 suggests that the NO_2/NO_x ratio also declines as total NO_2 concentration increases.

Figure 6.23 Comparison of NO_2/NO_x ratio of roadside increment and total roadside NO_x.

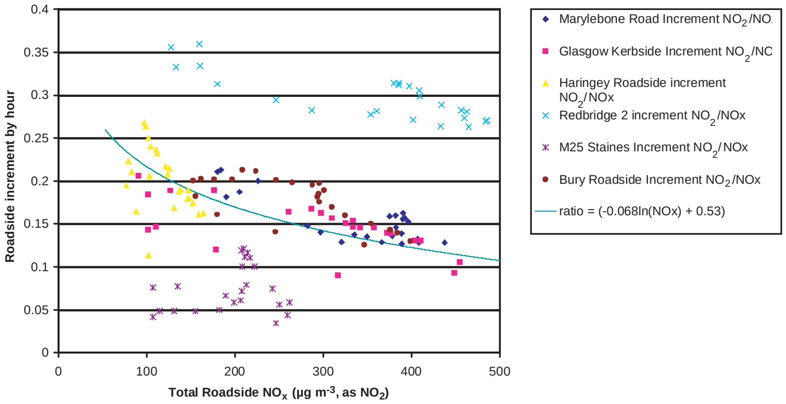
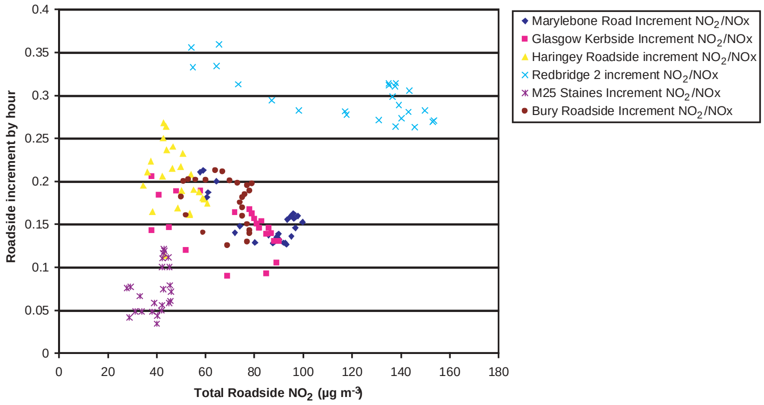


Figure 6.24 Comparison of NO_2/NO_x ratio of roadside increment and total roadside NO_2.



645. The analyses carried out at the six roadside sites considered here suggests that the highest primary NO_2 concentrations are found close to congested traffic or where a high proportion of the traffic emissions are from vehicles with diesel engines, such as buses. The lowest primary NO_2 concentrations are found for fast moving motorway traffic.

646. It was noted earlier that the magnitude of the roadside increment of NO_x concentrations is not only directly related to traffic flow or emissions but is also influenced by dispersion conditions. Figure 6.25 shows a pollution rose for the roadside increments at Marylebone Road site in 2001. The highest NO_x increments at Marylebone Road occur when the wind is from the south west. The monitoring site is situated on the south side of Marylebone Road showing that street canyon effects are evident at this site, leading to relatively poor dispersion of pollutants from traffic sources on the road. This is confirmed by Figure 6.26, which shows that the roadside increment is relatively insensitive to wind speed at this site when the wind is from the south- west.

Figure 6.25 Pollution rose for the roadside increment at Marylebone Road in 2001[4].

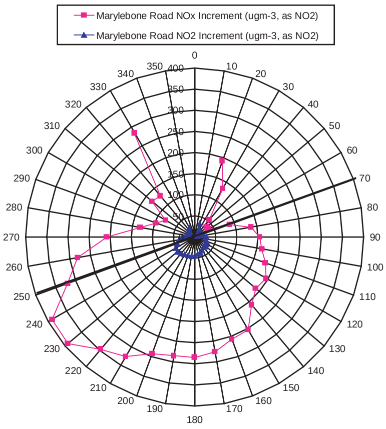
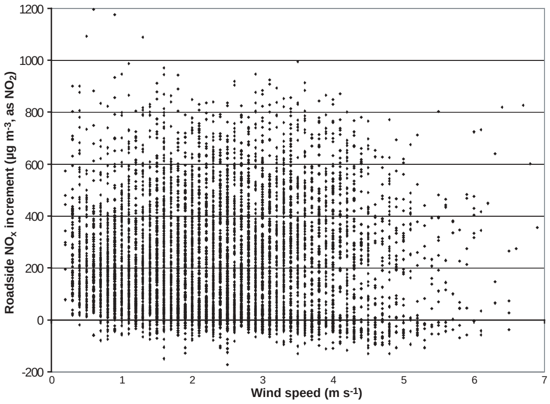


Figure 6.26 NO_x roadside increment at Marylebone Road and wind speed in 2001 (wind direction between 150 and 250).



647. Street canyon effects are not evident at any of the other sites selected for detailed analysis. This is illustrated in Figures 6.27 and 6.28, which show the pollution rose and winds speed plots for the roadside increment of hourly NO_x concentrations at Haringey Roadside. The highest NO_x increments at Haringey Roadside are seen when the wind is from the south-east (from the road towards the monitoring site, which is on the west of the road). In contrast to Marylebone Road, the roadside increment is always low at higher wind speeds.

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[4] The thick line on this plot represents the orientation of the road at Marylebone Road, which is roughly east to west.

Figure 6.27 Pollution rose for the roadside increment at Haringey Roadside in 2001 (µg m^{-3}, as NO_2).[5]

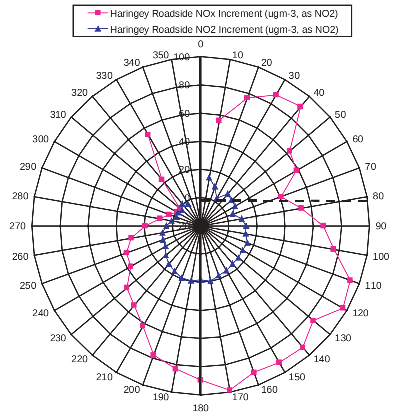
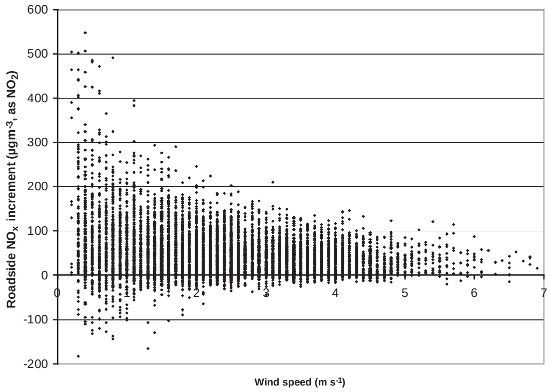


Figure 6.28 NO_x roadside increment at Haringey Roadside and wind speed in 2001 (all wind directions).



648. The roadside and kerbside sites selected for detailed analysis have been chosen to reflect a range of different concentrations and environments. It is, however, important to consider the relevance of the sites in terms of exposure when comparing the measured concentrations with objectives and limit values (see section Chapter 1). Neither the Bury Roadside nor the Redbridge 2 sites can be considered as directly relevant in terms of exposure. The Bury Roadside site is located within the roundabout of a motorway junction. There are houses, and therefore the possibility of relevant exposure, close to the roundabout but not within it.

649. The Redbridge 2 site is of particular interest since it has been identified as having the highest NO_2 concentrations in the UK, despite the fact that it is located in outer London. Furthermore, comparisons with data from other countries suggests that the site is amongst the most polluted in Europe in terms of annual mean NO_2. There are several points that should be noted when considering concentrations of pollutants measured at this site.

- The site is located on a traffic island surrounded by three very busy roads. The traffic along these roads is frequently queuing or moving at crawling pace. There is also a high proportion of buses (up to 6% of the traffic flow on the three road links at this site; in comparison with an average of about 1.3% for London as a whole).
- The limited work that has been carried out on single-chamber instruments (as used at this site, see section 4.2.1) at high-fluctuating concentrations of NO and NO_2 indicate that under these conditions these instrument types might result in higher NO_2 concentrations being recorded. However, it should be noted that whether an instrument is 'single-chamber' or 'dual-chamber' type is only one of many influences that can affect the NO_2 measured (others include instrument design, filtering etc.).
- Carbon monoxide (CO) is also measured at the site. CO is a very good indicator of road traffic and particularly of congested traffic. The CO instrument is not affected by difficulties in measuring rapidly fluctuation high concentrations (at least not in the same way as NO_2). CO concentrations are the highest in London. For example, the annual mean CO concentration in 2000 was 3.2 mg m^{-3} at Redbridge 2, higher than the 2.3 mg m^{-3} at Marylebone Road, and at least double that of any other roadside/kerbside site in London. The measured CO concentration therefore strongly supports the idea that the site is located in an extreme location that is very close to high flows of congested road traffic.

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[5] The thick line on this plot represents the orientation of the road at Haringey Roadside, which is roughly south to north, with a pedestrian road joining from the east.

650. The analyses presented here show that the NO_2 concentrations measured close to complex junctions can exhibit unusual behaviour including the high NO_2 concentrations recorded at Redbridge 2. It is important to understand the complex influences on ambient NO_2 concentrations at these locations, since there may be locations in the UK that show similar behaviour that are of relevance to exposure, but at which concentrations have not been measured.

6.2.5 Recent trends of NO_x, NO_2 and O_3 concentrations in London

651. Concentrations of NO_x have declined in recent years in London, approximately in line with the calculated decline in NO_x emissions (see section 6.2.4). Indicative trends for NO_x and NO_2 are shown in Figures 6.29 and 6.30. These trends have been calculated for 11 LAQN sites (a mixture of background, roadside etc.) starting in October 1995. The rolling annual mean has been calculated on a monthly basis for each site (hence the Figures start in October 1996). For each site, these trends have been normalised such that the value for October 1996 is 100. All sites are then averaged. Considering annual mean rolling concentrations should remove seasonal effects, but still reveals the important influence of meteorology. These plots show that at the end of 2002[6], the NO_x concentration was approximately 70 % of the concentration in 1996. For NO_2 over the same period, the mean value at the end of 2002 was 88 % of that recorded at the beginning of 1996.

652. The plots also highlight some important pollution episodes. The October/November 1997 episode is clearly shown in Figures 6.29 and 6.30. In Figure 6.29, NO_x can be seen to increase during October and November of that year. However, one year later there is a reduction in NO_x between October and November 1998 when the episode is no longer included in the averaging. These effects are also shown in the plot of NO_2 and highlight that major air pollution episodes can affect annual mean concentrations.

653. Consideration has also been given to the trends in O_3 and total oxidant ($NO_2 + O_3$) in London. Only five background sites have been used in the analysis since co-located NO_2 and O_3 monitoring is required. Figure 6.31 shows that concentrations of O_3 have increased about 18% over the period 1996-2002 compared with a reduction of 17% for NO_2. Absolute concentrations of total oxidant have declined gradually to 96% over the same period. This will be due to a decline in primary NO_2 emissions, possibly offset by an increase in background O_3.

Figure 6.29 Relative monthly annual mean normalised NO_x in London.

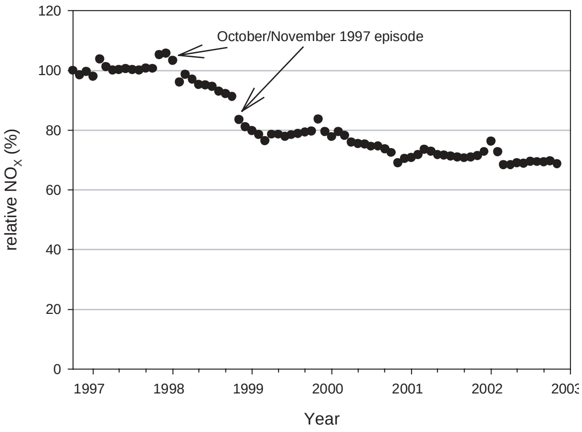
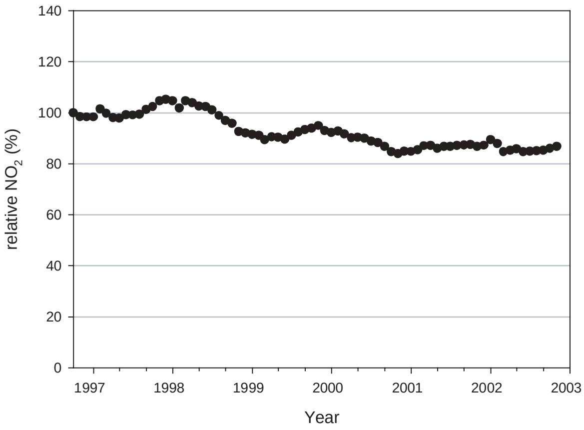


Figure 6.30 Relative monthly annual mean normalised NO_2 in London.



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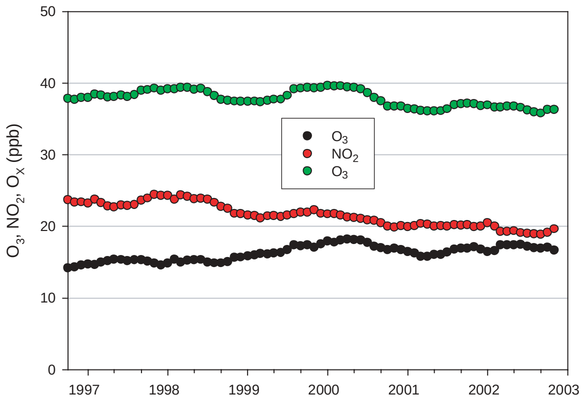
[6] Some of the data for 2002 are unratified. See section 4.9.6.

Figure 6.31 Running annual mean NO_2, O_3 and OX (NO_2 + O_3) averaged over five background sites in the LAQN.



6.2.6 Source apportionment and analysis of recent trends in annual mean NO_x concentrations using site-specific empirical models

654. The trends in ambient NO_x and NO_2 concentrations over the last 10–20 years can be reconciled with the changes in emissions by a combination of the source apportionment of current measured concentrations and published emission inventories.

655. The site-specific empirical modelling approach described in section 5.2.1.9 provides a source apportionment of the measured annual mean NO_x concentration for a particular base year into sectors including traffic, industrial, domestic, major point sources and regional rural contributions. Examples of this source apportionment are listed in Table 6.9 for the sites selected for detailed analysis. The total traffic contribution has been split into the contributions from different vehicle types at a selection of these sites. As expected, traffic sources dominate at the roadside sites with contributions ranging from 70% at Haringey Roadside to 92% at Redbridge 2. The percentage contribution from traffic sources is lower at background sites, ranging from 50% at London Bloomsbury to 71% at Glasgow City Chambers and Belfast Centre. The contribution from emissions from cars is about 30% at all of the sites where this statistic has been calculated. This is because the contribution from cars to the total traffic emissions is lower at the roadside sites than at background locations because the roadside sites studies are generally on roads with a greater proportion of emissions from buses and HGVs than for traffic emissions in general.

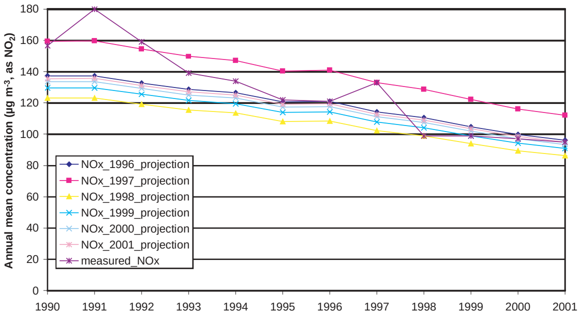
Table 6.9 Source apportionment of annual mean NO_x concentration in 2001 (µg m^{-3}, as NO_2, and percentages in brackets).

Site	Total 2001	Rural	Industry	Commercial	Domestic	Other area	Points	Total traffic	Cars	LGV	HGVr	HGVa	Buses
Marylebone Road	335	12 (4)	7 (2)	7 (2)	19 (6)	7 (2)	0 (0)	281 (84)	114 (34)	32 (10)	65 (19)	23 (7)	47 (14)
Glasgow Kerbside	284	4 (1)	6 (2)	6 (2)	7 (2)	5 (2)	0 (0)	256 (90)					
Haringey Roadside	120	13 (11)	6 (5)	6 (5)	9 (8)	2 (2)	1 (1)	84 (70)	37 (31)	7 (6)	12 (10)	6 (5)	21 (23)
Redbridge 2	355	12 (3)	3 (1)	8 (2)	4 (1)	2 (1)	1 (0)	325 (92)	114 (32)	23 (6)	42 (12)	7 (2)	140 (39)
M25 Staines	196	12 (6)	0 (0)	2 (1)	4 (2)	16 (8)	0 (0)	162 (83)	58 (30)	11 (6)	19 (10)	70 (36)	3 (2)
Bury Roadside	270	12 (4)	3 (1)	3 (1)	5 (2)	4 (1)	2 (1)	242 (90)	80 (30)	15 (6)	32 (12)	111 (41)	5 (2)
London Bloomsbury	109	12 (11)	8 (7)	8 (7)	18 (17)	6 (6)	1 (1)	55 (50)	29 (27)	5 (5)	8 (7)	3 (3)	9 (8)
West London	95	12 (13)	4 (4)	4 (4)	17 (18)	4 (4)	1 (1)	52 (55)	28 (29)	5 (5)	8 (8)	4 (4)	7 (7)
London Bexley	68	12 (18)	3 (4)	3 (4)	5 (7)	5 (7)	2 (3)	38 (56)					
Manchester Town Hall	92	13 (14)	5 (5)	5 (5)	6 (7)	6 (7)	1 (1)	56 (61)	27 (29)	5 (5)	7 (8)	13 (14)	5 (5)
Glasgow City Chambers		107 (4)	4 (7)	7 (7)	7 (7)	7 (6)	6 (0)	0 (71)	76				
Belfast Centre	58	6 (10)	1 (2)	1 (2)	5 (9)	4 (7)	0 (0)	41 (71)					
Port Talbot	38	6 (16)	1 (3)	1 (3)	1 (3)	2 (5)	1 (3)	25 (66)					

656. The contribution from domestic and commercial emissions (primarily natural gas space heating) varies from 5% at Port Talbot to 10% at Belfast Centre and 22% at West London and 23% at London Bloomsbury. The percentage contribution from domestic and commercial emissions at roadside sites is much lower. These are important sources of NO_x, particularly in London, because emissions from these sectors are expected to increase somewhat over the next 10 years or so, in contrast to emission from traffic, which are expected to decline (see section 2.12). Projections of the expected source apportionment of annual mean NO_x concentrations for the future are presented in Chapter 7.

657. Published trends in emissions for each sector (such as those in section 2.8) can be applied to the contributions from each source sector to calculate site-specific estimates of concentrations in earlier years from the measurements in 2001. Figure 6.32 is an example of this type of analysis and shows site-specific estimates of annual mean NO_x at West London back to 1990 from the base years from1996 to 2001. There is reasonably good agreement between the projected and measured trend. This plot also shows the influence of meteorology on the measured concentrations, with the contributions to the annual mean from poor dispersion episodes of 1991 and 1997 clearly visible.

Figure 6.32 Site-specific estimates of annual mean NO_x concentrations at West London.



658. Figure 6.33 summarises the results of this type of analysis at the background monitoring sites selected for detailed analysis. The measured concentrations in each year have been normalised by dividing by the concentration projected from the 2001 base year at each site. This should remove the trend in concentrations due to the year on year changes in emissions, leaving just the year to year variations in concentrations due to dispersion conditions. The majority of the ratios are in the range 0.8 to 1.2 indicating that the trends in measured concentrations can be explained by a combination of the source apportionment and trends in emissions estimates. The remaining year to year variation in annual mean NO_x concentrations (about ± 20%) will be due to a combination of meteorological variations and uncertainties in the source apportionment and emission trends. This is consistent with the modelled indications of likely year to year variations in annual mean NO_x concentrations due to variation in meteorology discussed in chapter 5. The analysis presented here is of NO_x concentrations only, the trends in annual mean NO_2 are also influenced by the availability of oxidant. Figure 6.34 shows a similar analysis for the roadside and kerbside monitoring sites, for which there are considerably less data.

Figure 6.33 Comparison of measured and site-specific estimates of annual mean NO_x concentrations at background sites.

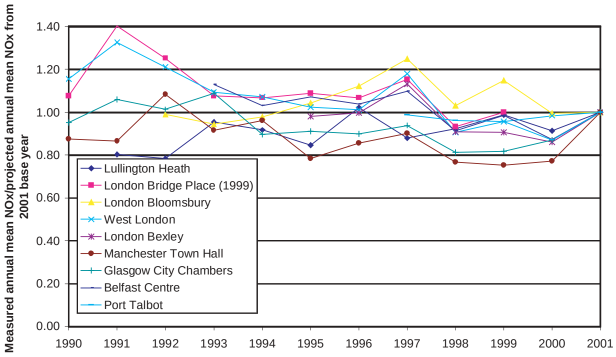


Figure 6.34 Comparison of measured and site-specific estimates of annual mean NO_x concentrations at roadside and kerbside sites.

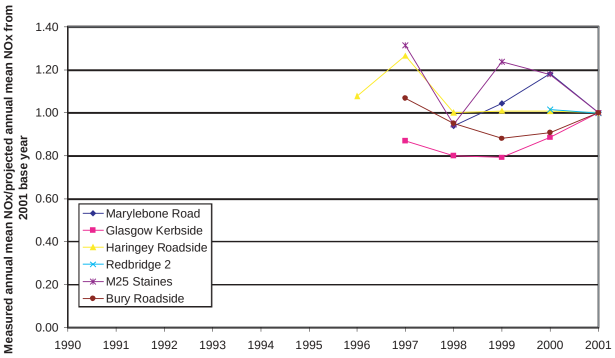


Figure 6.35 Site-specific projections of annual mean NO_x concentrations at Central London/Bridge Place.

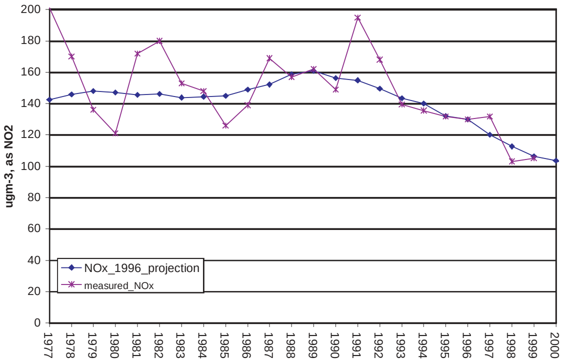
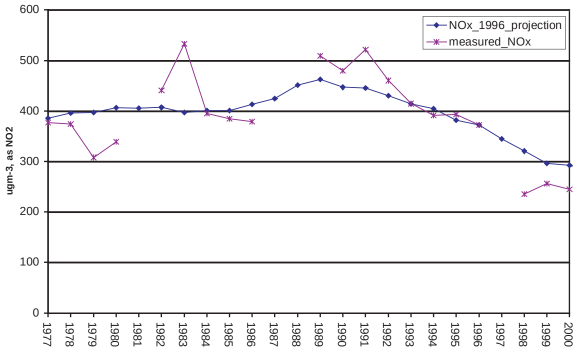


Figure 6.36 Site-specific projections of annual mean NO_x concentrations at Cromwell Road/Cromwell Road 2.



659. NO_x emissions from traffic sources peaked in about 1990 and thus the analyses described above can only be used to illustrate the decline in emissions since then. Figures 6.35 and 6.36 show site-specific projections of annual mean NO_x concentrations at Central London/Bridge Place and Cromwell Road/Cromwell Road 2 back to 1977 from the base year 1996. These plots show considerable year to year variation in measured concentrations but the overall trends are followed by the projections derived from the emissions inventory information[7].

6.2.7 Recent trends in NO_x and NO_2 concentrations at Marylebone Road

660. The analysis presented above shows that the broad pattern of trends in measured NO_x concentrations over recent years is consistent with the expected changes in emissions over the same period. The local conditions at an individual monitoring site can, however, have a significant influence on the measured trends. The trends in measured concentrations and emissions at Marylebone Road have therefore been investigated in some detail. Measurements of NO_x and NO_2 have been made at Marylebone Road since July 1997. The annual means listed in Table 6.10 indicate that the decrease in NO_x and NO_2 was negligible up until 2001. In fact, concentrations measured in 2000 were significantly higher than 1998 and almost as high as 1997. Furthermore, data for 1997 only reflect a proportion of the year and this proportion was significantly influenced by a large pollution episode.

Table 6.10 Annual mean NO_x and NO_2 concentrations at Marylebone Road (µg m^{-3}, as NO_2) and the percentage of each year when the wind direction at Marylebone Road was from a 'polluted direction'.

Year	NO_x	NO_2	Percentage of wind directions from a 'polluted direction'
1997[1]	425	93	45
1998	374	92	46
1999	391	91	48
2000	415	93	55
2001	335	84	44
2002[2]	290	76	–

[1] Part of a year [2] Includes some provisional data.

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[7] There are additional uncertainties in this analysis of trends since the 1970s because the Central London/Bridge Place site has been relocated twice over the period studied and the Cromwell Road site moved to Cromwell Road 2 site.

661. Estimates have been made of the likely change in emissions from 1997 to 2001 using continuous traffic count data and manual counts of traffic at Marylebone Road. The continuous traffic counter records flows of different categories of vehicle for each of the six lanes and also provides an estimate of the vehicle speed on a 15-minute basis. These traffic data have been processed on an hourly basis over the entire period. Manual count data has been used to split these broad vehicle classes into motorcycles, cars, taxis, LGVs, buses, rigid HGVs and articulated HGVs.

662. Emissions have been calculated using the national stock of vehicle ages for all vehicle types except taxis and buses, which make use of London-specific data. Figure 6.37 shows the trend in calculated emissions from light-duty and heavy-duty vehicles. A rolling monthly average has been applied to these data to highlight the overall trend. The downward trend in emissions is more marked for light-duty vehicles, mostly as a result of the increasing proportion of vehicles fitted with catalytic converters. There is a significant dip in vehicle emissions each year around Christmas and New Year, mostly caused by a reduction in the flow of heavy-duty vehicles. Towards the end of 2001, there was an accelerated decrease in the emissions of both light-duty and heavy-duty vehicles, caused by the introduction of a bus lane, causing an overall reduction in traffic volume.

663. There is also a clear day of the week dependence for emissions and concentrations of NO_x at Marylebone Road, as shown in Figure 6.38, which shows that total NO_x emissions increase from Monday to Thursday and then decrease through to Sunday. This pattern is also reflected in the NO_x concentration. The significant change in HGV flows on Sunday is reflected well in both the emissions and concentration measurements. These results therefore provide some confidence that the relative emission rates between light and heavy vehicles are well represented by current emission factors.

Figure 6.37 Emissions from light and heavy-duty vehicles at Marylebone Road.

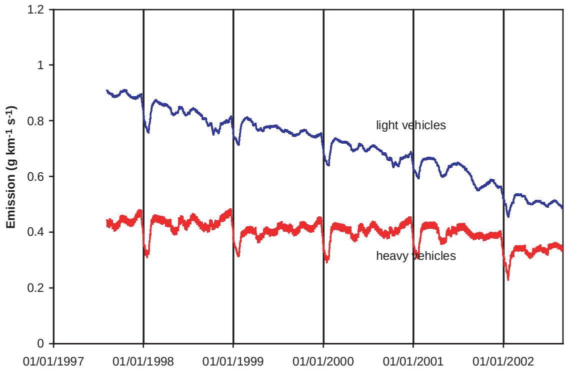
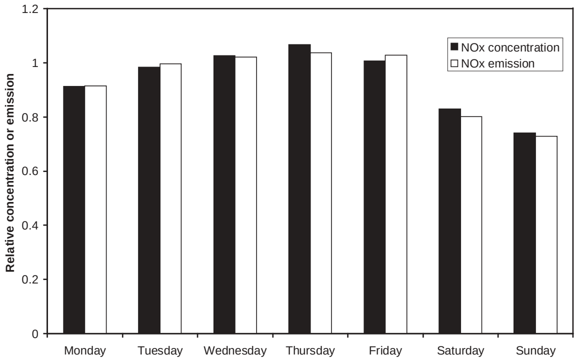


Figure 6.38 Relative NO_x emissions and concentration by day of the week.



664. Table 6.10 shows that there was a relatively small change in NO_x concentrations at Marylebone Road until 2001, despite a significant downward trend in total emissions. The variation in NO_x concentrations from year to year must therefore be explained by variations in meteorology from year to year (if a decrease in emissions is accepted). There are two significant influences on measured concentrations at Marylebone Road: London-wide air pollution concentrations and the local dispersion of pollutants within the street. The roadside increment of concentrations at this site is generally greatest when the wind is from the south-west (see section 6.2.3).

665. Figure 6.39 shows the trends in total emissions of NO_x, NO_x concentration and an indicator of the NO_x roadside increment (Marylebone NO_x minus London Kensington NO_x). The NO_x roadside increment (green line) should reflect more the influence of Marylebone Road itself and not the effect of London-wide pollution episodes that affect all sites. It is clear, for example, that the elevated NO_x concentrations at the end of October/beginning of November 1997 were associated with a London-wide episode of high concentration. Periods where the NO_x roadside increment and the total NO_x concentration are similar are periods where concentrations at Marylebone Road are dominated by local street canyon effects. This is most clearly shown during 2000. The decrease in emissions around Christmas/New Year can be seen in the NO_x concentration observations. The decline in NO_x concentrations in 2001 was significantly influenced by the introduction of a bus lane during the autumn of that year. The bus lane has resulted in a decline in emissions from both heavy and light vehicles (shown in the plots), as well as moving the 'centre of gravity' of the emissions further away from the kerb.

666. In order to show that there are significant periods where NO_x is influenced by the street canyon, the proportion of the time during each year when the wind direction was from a 'polluted direction' is listed in Table 6.10 and plotted as a running monthly mean in Figure 6.40. A 'polluted direction' is defined as a direction ≥ 70° and ≤90° (along the road) or ≥160° and ≤250° street canyon. It is clear that there was a significantly higher proportion of polluted wind directions during 2000 than any other year. Indeed, this explains the relatively high concentrations of NO_x and NO_2 during that year. A closer inspection of 1999 also shows that this was a year influenced by London-wide air pollution episodes and a relatively high proportion of polluted wind directions.

Figure 6.39 Trends in total NO_x emission and concentration at Marylebone Road.

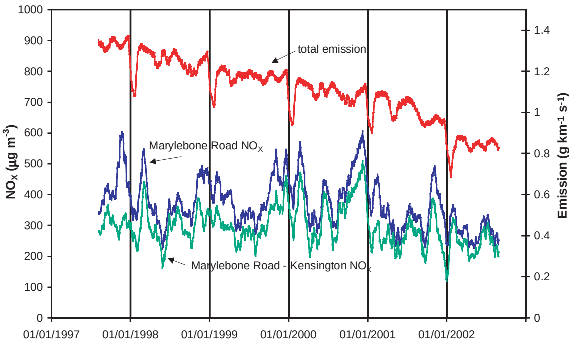
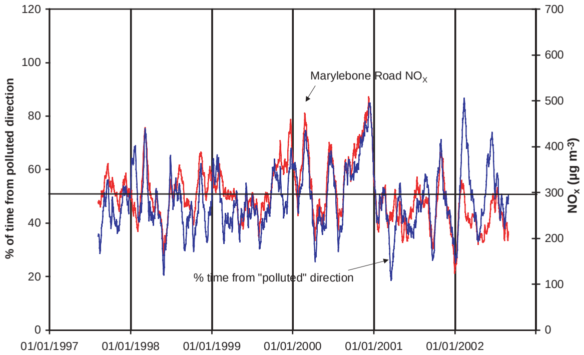


Figure 6.40 Proportion of the time where the wind direction is from a 'polluted direction'.



667. Observations of NO_x and NO_2 at Marylebone Road show that there is significant variation in the concentrations of NO_x and NO_2 year to year, with concentrations only declining in 2001 and 2002 (see Table 6.10 and Figure 6.7 and 6.8). Variations in meteorology from year to year, along with the street canyon effects at this site, provide at least a partial explanation of the unexpectedly high concentrations measured during 1999 and 2000, at a time when emission were expected to have declined. The introduction of a bus lane in 2001 is also thought to have contributed to the reduction in measured concentrations observed in 2001 relative to earlier years. It is clear therefore that local influences at monitoring sites can profoundly affect observed concentrations and these should be considered when assessing trends in air pollution, particularly at a site such as Marylebone Road where data are available for only six years.

6.2.8 NO_2 levels in the United Kingdom in the context of European levels

6.2.8.1 Analysis of the AIRBASE database for the Year 2000

668. NO_2 data for a large number, over 1600, of sites across Europe have been collected by the European Environment Agency into the AIRBASE database which is accessible at: http://etc-acc.eionet.eu.int/databases/airbase.html. A web-based public-access visualisation tool, AirView, which is accessible from this site allows the database to be queried and selected data to be presented as maps, graphs or tables.

669. EU member states have been obliged to present NO_2 air quality data to the European Commission through its directive on the exchange of information on air pollutants covered by health protection directives. Annual mean NO_2 data are stored by site type and by year. Clearly, it is difficult to harmonise site classifications between each country and these difficulties can influence the analysis of the database. Despite these reservations, the airborne site classification is broadly consistent with that used in the UK, although the terminology is different. Although there is already a large body of information in AIRBASE, it is apparent that the database is incomplete. This situation has arisen because submissions to AIRBASE are organized at the national level and hence there is a variable amount of local authority, municipality and devolved administrative regional monitoring data.

670. The AIRBASE classification with the most polluted sites is the so-called 'traffic-influenced' category. A total of 479 sites have valid data entries for the year 2000, with an average annual mean NO_2 concentration of 43.8 µg m^{-3} recorded. Of these 479 sites, 262 sites reported annual mean concentrations in excess of 40 µg m^{-3}. The 'traffic-influenced' category contains 20 UK sites with an average annual mean concentration of 56.5 µg m^{-3}, significantly higher than the AIRBASE average. Of these 20 UK sites, 16 sites reported concentrations in excess of 40 µg m^{-3}. The frequency distributions of annual mean NO_2 concentrations for all sites and for the UK sites in this category are illustrated in Figure 6.41. In general terms, UK sites appear to be distributed towards higher concentrations compared with the rest of the AIRBASE sites.

671. Urban background sites in the AIRBASE classification appear to be associated with significantly lower annual mean NO_2 concentrations in the year 2000 compared with the traffic-influenced sites. This confirms the view found from the UK monitoring that proximity to traffic is the most important determinant in urban NO_2 levels. A total of 457 urban background sites have valid data entries for the year 2000, with an average annual mean concentration of 27.9 µg m^{-3}. Of these sites, 56 sites reported annual mean concentrations in excess of 40 µg m^{-3}. The urban background category contains 50 UK sites with an average annual mean concentration of 34.4 µg m^{-3}, significantly higher than the AIRBASE average for this category. Of these 50 UK sites, 12 sites reported concentrations in excess of 40 µg m^{-3}, a larger fraction than for AIRBASE as a whole. The frequency distributions of annual mean NO_2 concentrations for all sites and for the UK sites are illustrated in Figure 6.42. Again, UK sites appear to be preferentially distributed towards higher concentrations compared with the rest of the sites in the AIRBASE database.

Figure 6.41 The frequency distribution of the annual mean NO_2 concentrations reported for traffic-influenced sites in the AIRBASE database for the year 2000.

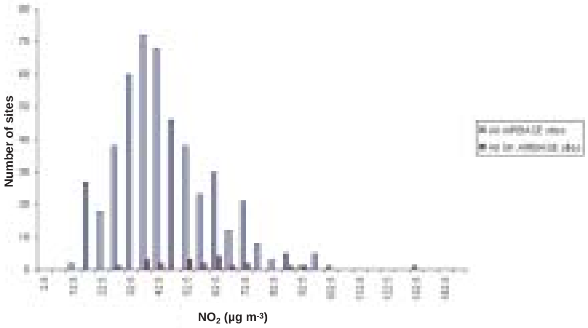


Figure 6.42 The frequency distribution of the annual mean NO_2 concentrations reported for urban background sites in the AIRBASE database for the year 2000.

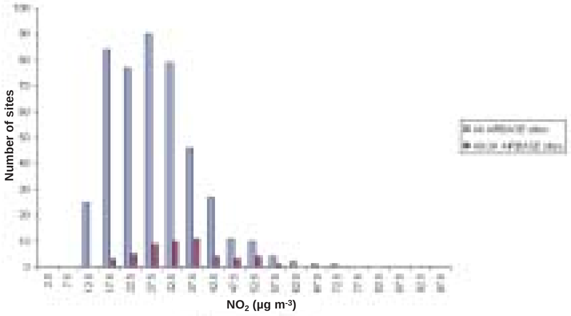


Figure 6.43 The frequency distribution of the annual mean NO_2 concentrations reported for rural background sites in the AIRBASE database for the year 2000.

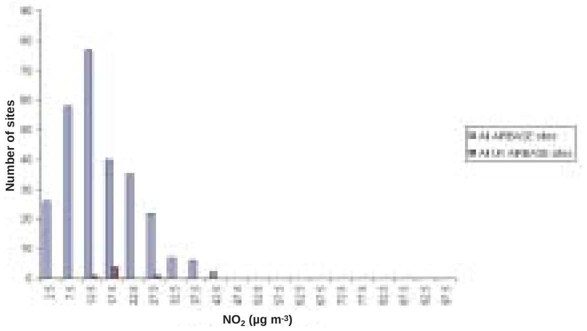
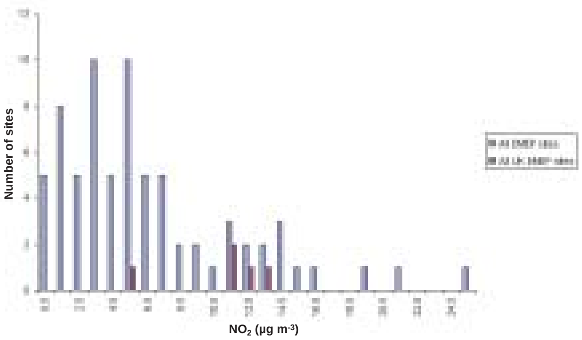


Figure 6.44 The frequency distribution of the annual mean NO_2 concentrations reported for rural background sites in the EMEP database for the year 2000.



672. Urban sites in AIRBASE have a further subdivision into sites which are industry-influenced. This category contains 146 sites with an average annual mean NO_2 concentration of 25.2 μg m^{-3} for the year 2000. This value is slightly lower than that for the urban background category but the difference is probably not significant. This lack of difference confirms the view found from UK monitoring (see section 6.3.3), that industrial sources are not important in determining urban NO_2 concentrations across Europe as a whole. The AIRBASE database has a suburban category which contains 221 sites with an average annual mean NO_2 concentration of 27.5 μg m^{-3}. This value is not significantly different from the average value for the urban background category, suggesting that the subdivisions within the AIRBASE database have not necessarily identified new exposure regimes for urban NO_2.

673. The rural background category in AIRBASE contains entries for 272 sites with valid data entries for the year 2000. The average annual mean NO_2 concentration for these sites was 14.5 μg m^{-3}, about half that of the urban background sites. Of these sites, only two (one each in France and Italy) reported exceedences of 40 μg m^{-3}. This category contains 6 UK sites with an average annual mean concentration of 12.7 μg m^{-3}, not significantly different from the AIRBASE average for this category. Figure 6.43 shows the frequency distributions for the annual mean concentrations for all AIRBASE and the UK AIRBASE sites. The AIRBASE distribution appears to show a larger fraction of cleaner sites compared with the situation in the UK as indicated by the AIRBASE database.

674. For all these site categories above, UK levels appear to be broadly comparable with those found across Europe. However, without a more detailed assessment of the comparabilities of the sites and their classifications, it is difficult to make a more detailed assessment of the UK levels in the context of those found across Europe.

6.2.8.2 Trends in annual mean NO_2 concentrations during the 1990s in the AIRBASE database

675. Across the entire AIRBASE database, there are 33 sites for which valid data entries are provided for the annual mean NO_2 concentrations for all years between 1990 and 2000, inclusive. A non-parametric method (Sens' method) has been used to determine whether there are any statistically significant trends within these annual mean quantities for any of the 33 sites. The selected sites are not restricted to any particular AIRBASE site category. Over the 1990-2000 period the 33-site average annual mean NO_2 concentration showed a statistically significant (at the 1% level) downwards trend of -0.7 μg m^{-3} per year, equivalent to -2% per year. In terms of individual sites, 12 sites showed statistically significant downwards trends at the 1% level of significance, and 6 more at the 5% level of significance. These trends were generally in the range -0.7 to -1.3 µg m^{-3} per year level, that is about –2 to –3% per year. Such trends are closely similar to but slightly smaller than the trends in national NO_x emissions which have been close to –3% per year over the EU during the 1990s.

6.2.8.3 Analysis of NO_2 concentration trends at rural sites in the EMEP Network

676. From the 1990s onwards, EMEP has coordinated a network of rural and remote rural NO_2 monitoring sites generating daily mean NO_2 data. The summaries of the data are available in the EMEP database which is accessible at www.nilu.no/projects/ccc/emepdata.html.

677. In the year 2000, annual mean NO_2 concentrations at EMEP sites were found to lie in the range 10–24 µg m^{-3} across much of Europe, from the UK through to Poland and from Italy to Spain. Concentrations fall off to 2–4 µg m^{-3} into southern Scandinavia and to 1–2 µg m^{-3} in the Arctic regions. The rapid fall off with distance away from the centre of Europe reflects the relatively short lifetime for the oxidation of NO_2 through to nitric acid and nitrate aerosol.

678. Annual network mean NO_2 concentrations have been in the range from 6.3–7.9 µg m^{-3} during the 1990s, with the value for the year 2000 at 6.6 µg m^{-3}. This is significantly smaller than the average for the year 2000 found for the rural background category in the AIRBASE database. This reflects the stricter siting criteria adopted for EMEP sites which ensures their siting in remote and sometimes mountainous regions, well away from pollution sources. Figure 6.44 shows the frequency distribution of annual mean concentrations for the year 2000 in the EMEP database. The distribution of annual mean values for the UK EMEP sites is clearly shifted towards higher concentrations, reflecting the UK's general proximity to the large NO_x sources in the industrial and population centres of north west Europe.

679. The annual EMEP network mean shows a highly statistically significant downwards trend of -0.12 µg m^{-3} per year or -2% per year over the period of the 1990s. This is again somewhat less than the trend in NO_x emissions of -3% per year for the EU countries over the same period. An analysis was also performed on the annual mean NO_2 concentrations over the period 1990-2000 using the Sen's method for all EMEP sites with complete records. At almost all of the sites, the annual mean NO_2 concentrations showed downward trends. At 15 of the sites, the trend estimates obtained using the Sen's method were significant at the 10% probability level or better.

6.3 Impacts of emissions from specific sources on local NO_x and NO_2 concentrations

6.3.1 Introduction

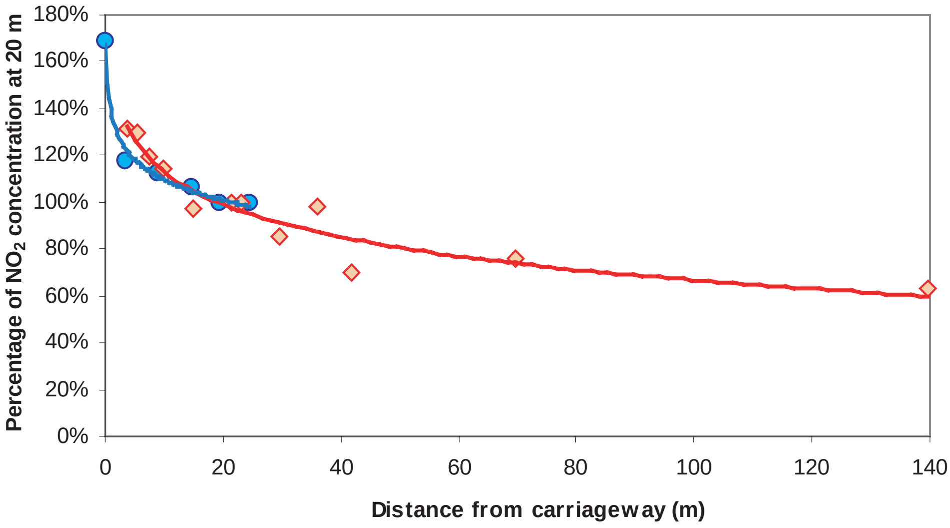
680. Measured NO_x and NO_2 concentrations during 2001 and recent years have been analysed in some detail in this report. The modelling methods that can be used to calculate maps of and projections for future years have also been described in the Chapter 5. Before presenting the results of these modelling studies for 2001 it is useful to review the typical impacts of NO_x emissions from some key source sectors. The impacts of road traffic, elevated sources such as power stations and airports on local NO_x and NO_2 concentrations from a combination of monitoring and modelling methods are considered.

681. Road traffic is clearly an important source of NO_x and contributed almost 50% of UK emissions in 2000 (see section Chapter 2). Concentrations are generally highest close to major roads but the profile of the decline in concentrations away from the edge of the road is not very well understood, particularly in urban areas. It is important to consider the emissions from power stations, because they contributed over 20% of total UK NO_x emission in 2000. The impact of these emission on ground level concentrations is however relatively small due to the tall chimneys from which these emissions are released. Aircraft emission only contributed about 1% of the UK total NO_x emissions in 2000 but it is important to assess the contribution of aircraft and other airport related emissions to local air quality. This is because airports are one of the few sectors for which emissions are expected to show a significant increase over the next 10 to 20 years. Emissions from aircraft are expected to increase to approximately 140% of the 2000 total by 2010 and to approximately 175% by 2020.

6.3.2 Road traffic

682. There have been few studies to show the way in which concentrations decline away from roads because such studies require the simultaneous measurement of concentrations at different distances from a road. Early work in London using diffusion tubes showed a sharp decline with distance from the road, with concentrations close to the local background beyond about 20 m from the edge of the road (Laxen & Noordally, 1987; Laxen *et al.*, 1988). This is illustrated in the results for a transect away from York Road (Figure 6.45), based on the average of two one-week exposures, normalised to 100% at about 20 m from the kerb.

Figure 6.45 NO_2 concentrations measured on a transect away from a busy central London road (circles), and the M25 motorway (diamonds) normalised to 100% at about 20 m from the edge of the carriageway The data points have been fitted using a logarithmic relationship, which accounts for 99% and 91% of the variance respectively.



683. A recent study alongside the M25 west of London, also using diffusion tubes, has demonstrated a similar rapid decline over the first 20 m (Hickman *et al.*, 2002). This is also illustrated in Figure 6.45, in which annual average concentrations for both sides of the motorway are shown normalised to 100% at about 20 m from the edge of the carriageway. The reduction does not appear to be quite as rapid at the motorway, as in central London. This may be due to the more open nature of the site and the greater width of the road.

684. Recent studies using a continuous monitor located at different distances from the A1(M) motorway in North Yorkshire provide results that are consistent with this pattern, although in this case the findings were based on a series of non-simultaneous 15-minute measurements during periods when the sites were downwind of the motorway (Lansley and Seakins, 2003).

685. Monitoring has been carried out in London at two distances from the kerb of Marylebone Road, using automatic monitors (Green & Fuller, 2003). This is a national monitoring site described at www.erg.kcl.ac.uk. Sampling was carried out at 1.4 and 5.5 m from the kerb, at a height of 3 m, for a six week period in January/February 2003. Average NO_x concentrations were 351 and 277 µg m^{-3} at 1.4 and 5.5 m respectively, representing a 21% reduction over this distance, while NO_2 concentrations were 100 and 91 µg m^{-3} respectively, representing a 9% reduction. O_3 concentrations were also measured, allowing an analysis of total oxidant concentrations. This showed a daily average NO_2:NO_x ratio of 0.14 for the road traffic component (see also section 5.2.4), with no evidence of any chemical reactions taking place over this distance.

686. There are two consequences arising from the apparently rapid decline away from the road:

- concentrations beyond about 20-50 m from the edge of the road will be essentially indistinguishable from the local background, taking account of measurement uncertainty and the normally high background contribution to measured roadside concentrations;
- elevated concentrations are generally confined to a zone extending out to about 20 m from the edge of the road although local conditions vary, especially in street canyons. This is evident in the results from both the national monitoring network and the monitoring carried out by local authorities for review and assessment purposes, which shows that exceedences of the 40 µg m^{-3} are likely to be confined to locations within 10 m of the kerb of all roads outside of major conurbations (Laxen *et al.*, 2002). The distance from the road centre line will, of course, depend on the width of the road.

687. The exact shape of the falloff with distance from the road is clearly crucial to our understanding of distances over which 40 µg m^{-3} may be exceeded. It is also important that models are able to reproduce this. There is thus an important need for further measurement studies, especially in more built up areas, together with comparisons with model results. The relatively rapid decline in concentrations away from the roadside does however provide some support for the simplified treatment of roadside concentrations within the national modelling studies in which concentrations are predicted for roadside and background locations only.

6.3.3 Elevated sources such as power stations

688. The areas with the highest NO_2 concentrations are generally associated with urban areas and roads. However, a number of rural measurements in the vicinity of major stationary sources of NO_x have been analysed by Futte *et al*. (2002). These sites form part of a number of monitoring networks run by power generators around power stations in Yorkshire and the Midlands designed to detect the influence of the power stations emissions on ground level concentrations in the region. The authors investigated a number of empirical approaches to estimating the long-term NO_x and NO_2 contributions to ground-level concentrations arising from major stationary sources. Table 6.11 summarises the estimated annual mean NO_2 at monitoring sites from major stationary sources, derived from those methods relying only on measurements at the rural monitoring sites listed and no modelling. The total concentrations at these sites are generally slightly higher than those at other rural sites, for example, Harwell, Lullington Heath and Wicken Fen.

Table 6.11 Annual mean NO_x and NO_2 concentrations and estimated contribution to annual means from power station sources.

Site	County	Year	NO_x (µg m^{-3}, as NO_2)	NO_2 (µg m^{-3})	ΔNO_2* (µg m^{-3})	$\Delta NO_2/\Delta NO_x$*
Carr Lane	North Yorkshire	1998	28.5	21.2	1.3-2.7	0.73-0.80
Cliffe	North Yorkshire	1998	26.9	16.8	1.0-2.5	0.29-0.74
Featherstone	West Yorkshire	1998	39.9	25.4	1.1-11.5	0.54-0.74
Sherburn	North Yorkshire	1998	33.8	20.6	1.0-5.7	0.40-0.77
Smethalls	North Yorkshire	1998	41.3	26.0	1.5-10.7	0.50-0.73
Gateforth	North Yorkshire	1998	32.3	20.4	1.0-3.8	0.47-0.81
Temple Hirst	North Yorkshire	1998	33.4	21.6	1.7-5.2	0.49-0.72
Cliffe	North Yorkshire	1999	25.8	18.0	0.8-3.2	0.51-0.74
Gateforth	North Yorkshire	1999	29.6	21.2	0.8-4.4	0.58-0.76
Featherstone	West Yorkshire	1999	34.6	23.9	1.3-9.2	0.56-0.74
Howden	North Yorkshire	1999	28.5	19.9	0.6-5.0	0.52-0.74
Sherburn	North Yorkshire	1999	30.0	21.0	0.8-4.8	0.50-0.78
Smeathalls	North Yorkshire	1999	35.5	24.1	1.7-9.0	0.52-0.72
Temple Hirst	North Yorkshire	1999	28.7	19.9	1.7-4.6	0.50-0.70
Womersley	North Yorkshire	1999	26.7	19.9	0.6-4.6	0.65-0.77
Bottesford	Leicestershire	1998	19.9	13.6	1.0-2.3	0.59-0.80
Jenny Hurn	Lincolnshire	1998	19.1	13.2	1.1-2.3	0.50-0.84
Thorney	Nottinghamshire	1998	23.9	16.6	1.7-4.2	0.53-0.78
Weston	Nottinghamshire	1998	32.9	22.9	1.1-9.4	0.64-0.89
Grove	Nottinghamshire	1998	22.3	15.7	1.3-2.9	0.56-0.74
Bottesford	Leicestershire	1999	16.4	13.0	0.4-2.5	0.60-0.81
Jenny Hurn	Lincolnshire	1999	17.0	12.2	1.3-2.9	0.49-0.79
Thorney	Nottinghamshire	1999	19.5	14.9	1.3-3.6	0.56-0.80
Weston	Nottinghamshire	1999	23.1	16.4	0.8-6.9	0.56-0.78
Grove	Nottinghamshire	1999	18.0	14.5	1.0-2.5	0.65-0.80
Harwell	Oxfordshire	1998	24.4	18.1		
Lullington Heath	East Sussex	1998	15.9	13.7		
Wicken Fen	Cambridgeshire	1998	20.1	14.6		
Wicken Fen	Cambridgeshire	1999	19.2	14.1		

*$\Delta NO_2/\Delta NO_x$ is the ratio of the estimated incremental NO_2 (ΔNO_2) to the incremental NO_x (ΔNO_x) from the local sources. The range shown represents the spread in values associated with the various empirical methods of interpretation used.

689. The contributions of the major power stations to the measured concentrations at these sites can be derived by a variety of approaches:

- *background corrected method*. This uses the measured hourly mean NO_2 and NO_x concentrations at each site corrected for background concentrations by averaging measured concentrations at sites in the network, judged not to be downwind of any local power station. The method is not suitable for rural monitoring sites in the Thames Estuary as local power stations make a much smaller relative contribution to NO_x concentrations in this area;
- *proportional method*. This assumes that the NO_2downwind of local power station is given by ΔNO_x ((total measured NO_2/total measured NO_x);
- *SO_2 ratio method*. This uses the typical ratio of SO_2 to NO_x in power station emissions, taken to be 3 in ppm by volume, from which ΔNO_x from a local power station can be estimated from the measured SO_2. In this method ΔNO_2 is set equal to $\Delta NO_x \times$ (total measured NO_2/total measured NO_x);
- *SO_2 ratio, background corrected method*. This is a refinement in which the SO_2 concentration is first corrected for background levels of SO_2. In this method ΔNO_2 is set equal to ΔNO_x (total measured NO_2/total measured NO_x);
- *dispersion modelling method*. ΔNO_x concentrations can be predicted using the ADMS 3 dispersion model as the basis of another method of deriving $\Delta NO_2/\Delta NO_x$ ratios, but these are not shown in Table 6.11.

690. Futter *et al*. (2002) list the estimated ΔNO_x and ΔNO_2 over 25 site years in the period 1998 and 1999 (as summarised in Table 6.11). It turns out that there are substantial differences in the amount of NO_x estimated to arise from local power station emissions depending on the method used. Generally *background corrected* ΔNO_x > *SO_2 ratio* ΔNO_x > *SO_2 ratio background corrected* ΔNO_x > *dispersion modelling* ΔNO_x. The actual annual average concentrations of ΔNO_2 are small, at most 10 μg m^{-3} and generally 2 to 4 μg m^{-3}. These small contributions to total annual mean concentrations are part of the reason that inaccuracies are introduced in all the methods. The average ratio of annual average NO_2 to annual average NO_x at the sites was 0.65. The ratio for the estimated power station component varied broadly between 0.4 and 0.8, depending on the method.

691. In an earlier analysis, also based on rural netweork monitoring data, Webb and Hunter (1998) considered the influence of power station emissions on the occurrence of peak concentrations of NO_2 at rural monitoring sites. The interpretation was somewhat easier as it is usually straightforward to identify short-term plumes at the ground from their NO_x to SO_2 ratios. The authors showed that exceedences of hourly average concentrations above 200 μg m^{-3} are extremely unlikely. The reason is the same as that arising in connection with long-term averages: the limitation on the oxidation of NO by oxidant.

6.3.4 Airports

692. Air traffic numbers continue to increase significantly whilst emissions from individual aircraft are reducing at rather modest rates. For this reason the emissions of NO_x from the aircraft sector is increasing both in absolute terms and relative to other sectors such as road transport. As an illustration of the contribution to NO_x by emissions related to airports some examples are taken from a source apportionment study conducted for the London Boroughs of Hillingdon and Hounslow and the Borough of Spelthorne for receptor points in the neighbourhood of Heathrow Airport for the year 2005.

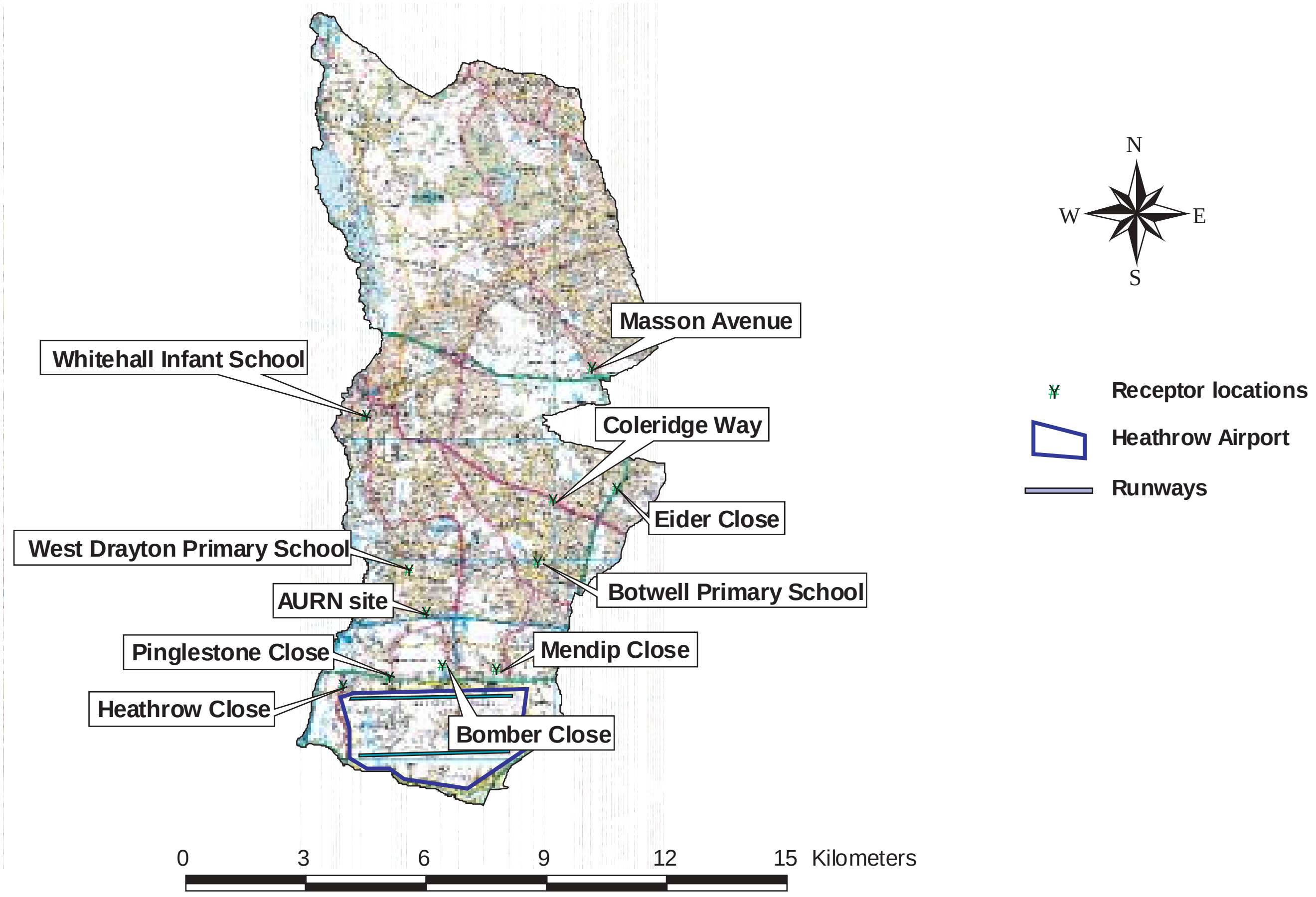
693. Emissions data were taken from four different sources: the emissions inventory for Heathrow, 1998; the London Atmospheric Emissions Inventory (LAEI), supplied by the Greater London Authority (GLA), February 2002; the Surrey Traffic Model; and the February 2002 emissions inventory for Slough. It is recognised that the revised inventory (Underwood and Walker, 2003) exhibits lower aircraft emissions (for example, a reduction of 19% for ground based aircraft) and these emissions would result in lower calculated concentrations, however, the general conclusions of this illustrative study are not changed. Meteorological data from Heathrow for the year 1999 were used in the modelling. Background concentration data were obtained from rural monitoring sites and adjusted to be appropriate for the year 2005. Emission data were projected forward to 2005 taking account of traffic and airport growth. Terminal 5 comes on-stream in 2008/9 and will add further to the airport growth.

694. Details of the receptor locations within Hillingdon for which the source apportionment exercise was carried out using ADMS-Urban are given in Table 6.12 together with the total annual average NO_2 concentrations. The locations are shown in Figure 6.46. The site closest to the airport, Heathrow Close, is only 300 m from the northern runway, whilst Masson Avenue, the most distance site, is 8.4 km from the runway.

Table 6.12 Source apportionment receptor point locations within Hillingdon.

ID	Description	Location	Annual average NO_2 concentration 2005 ($\mu g\ m^{-3}$)
1	Masson Avenue	511019, 184714	39.3
2	Eider Close	511642, 181709	40.4
3	Coleridge Way	510073, 181410	35.4
4	Botwell Primary School	509681, 179870	40.5
5	Mendip Close	508640, 177199	47.0
6	Bomber Close	507307, 177301	45.0
7	Pinglestone Close	505996, 177006	45.6
8	Heathrow Close	504842, 176789	42.1
9	West Drayton Primary School	506473, 179674	37.7
10	AURN site	506900, 178620	51.9
11	Whitehall Infant School	505432, 183532	36.8

Figure 6.46 Location of source apportionment receptor points.



695. Figure 6.47(a) shows a breakdown by general source type of emissions in Hillingdon. The emissions from the airport, which include all aircraft emissions up to 1000 m and all other emissions within the airport perimeter, are dominant. The component parts of the Heathrow emissions are shown in Figure 6.47(b). In this figure, road vehicles includes airside vehicles and Heathrow-generated traffic on major roads. This shows that most emissions are from aircraft in the Approach, Landing, Take-off, Climb-out (LTO) cycle. Figure 6.48 shows source apportionment of NO_x at the 11 receptor points, again broken down by the general source types. Close to the airport (Mendip Close, Bomber Close) the impact of the airport emissions is comparable with the impact of traffic emissions. However other sites are dominated by traffic emissions. Figure 6.49 shows source apportionment corresponding to the airport emissions Figure 6.47(b). This shows the dominant impact of the LTO aircraft emissions – except for the AURN site where road traffic associated with the airport gives a large contribution. Figure 6.50 shows the impact of LTO aircraft emissions from different height bands. Emissions in the lowest 50 m dominate concentrations, with higher level emissions contributing very little to ground level concentrations of NO_x.

Figure 6.47(a) NO_x emissions from within Hillingdon (2005).

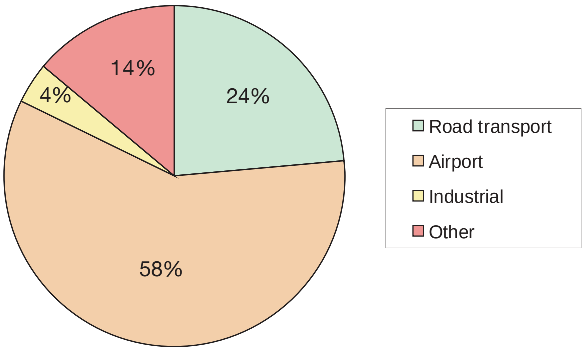


Figure 6.47(b) NO_x emissions from different Heathrow source groups in Hillingdon (2005).

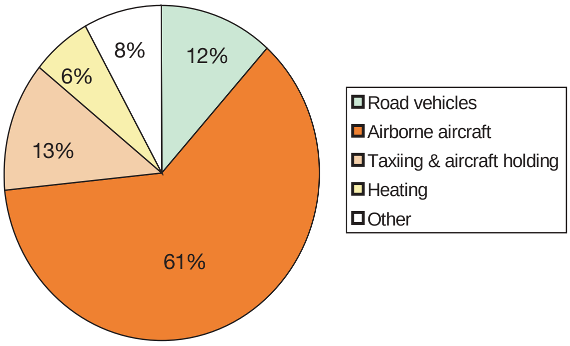


Figure 6.48 Contribution of major source groups to annual average NO_x concentrations (2005).

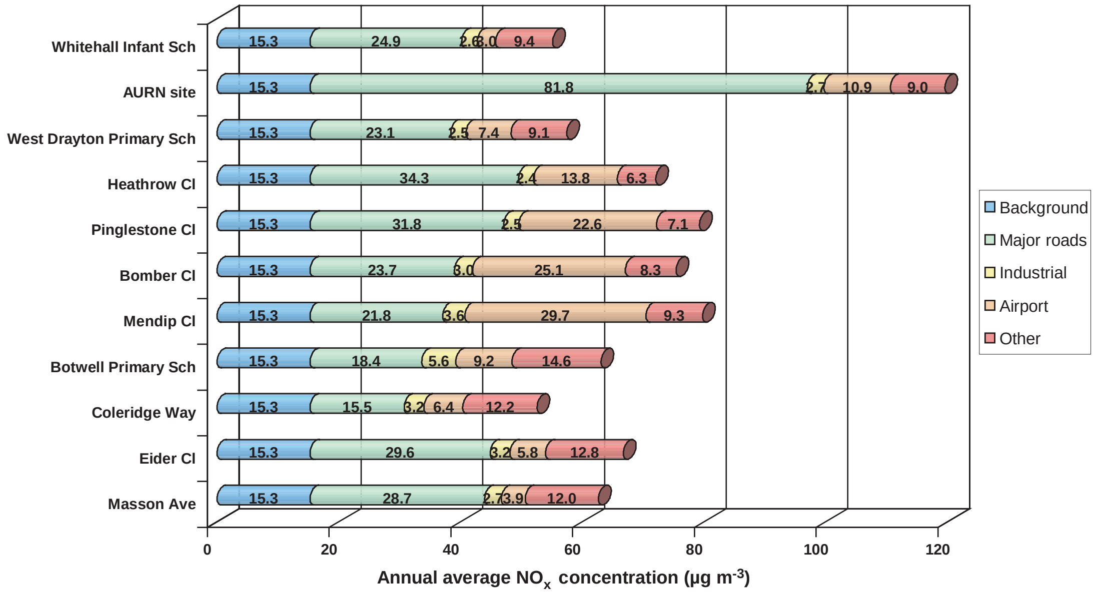


Figure 6.49 Contribution of Heathrow sources to annual average NO_x concentrations (2005).

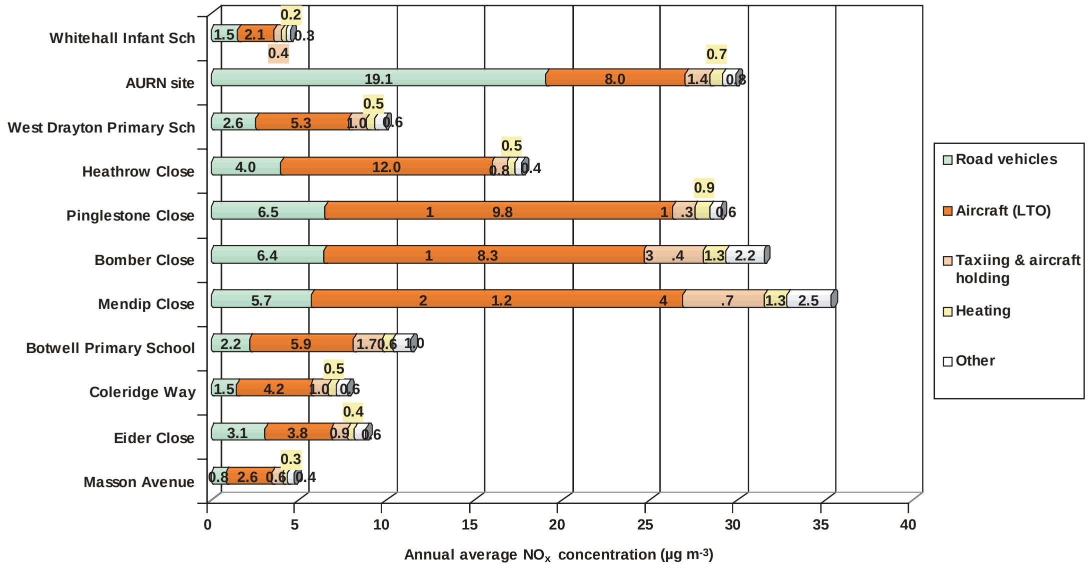
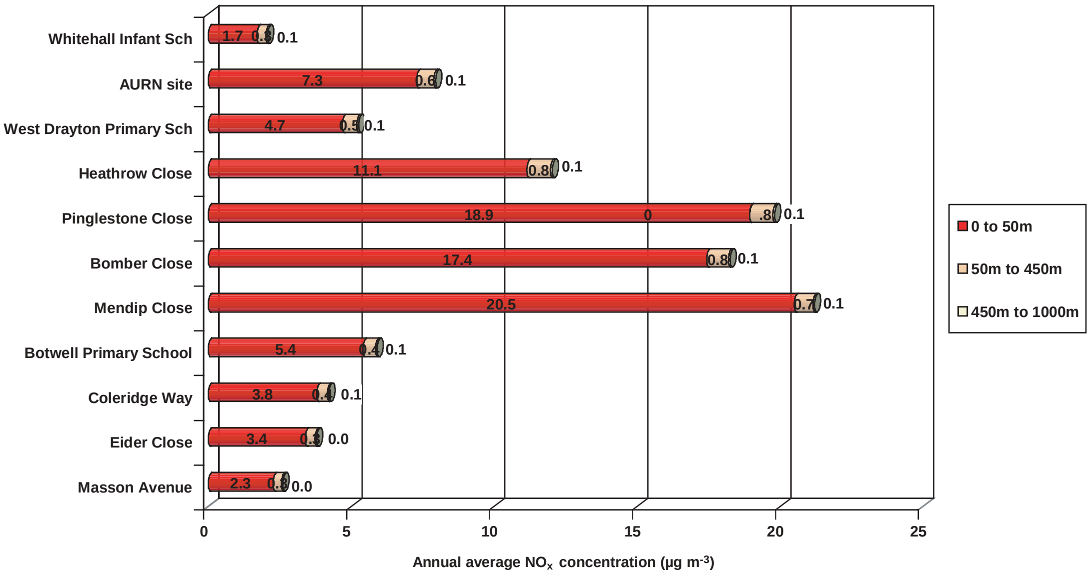


Figure 6.50 Contribution of aircraft at different heights to annual average NO_x concentrations during take-off, climb-out, approach and landing (2005).



6.4 Modelling results for 2001

6.4.1 Introduction

696. Maps of ambient NO_2 concentrations in the UK in 2001 have been calculated for policy analysis and for reporting to the European Commission (Stedman *et al* 2002). These maps provide an assessment of the locations in the UK where concentrations are likely to have exceeded an annual mean concentration of 40 μg m^{-3}. Analysis at the UK scale is clearly limited as to the level of detail of the treatment of emissions, dispersion and chemistry that can be considered. More detailed modelling studies have been carried out for London and the results of these studies are compared. The different modelling studies are in reasonable agreement with each other in terms of the extent of exceedence of 40 μg m^{-3} at roadsides in London but one study predicts a much larger area of exceedence in background locations.

697. Modelled projections of concentrations in 2005 and 2010 are discussed in section Chapter 7.

6.4.2 Mapped results of national empirical models for 2001

698. Maps of annual mean NO_x and NO_2 concentrations in 2001 at both background and roadside locations have been calculated by netcen for the UK using the empirical modelling methods described in section 5.2 and are shown in Figures 6.51 to 6.54. The modelling results in terms of exceedence of 40 μg m^{-3} are summarised in Table 6.13. The modelled extent of exceedence is consistent with the measurements in 2001 discussed in section 6.2.1. Roadside concentrations across most of London are estimated to be above 40 μg m^{-3}, as are background concentrations in central London. Outside London, exceedences are largely but not entirely confined to roadside locations with roughly one third of the total length of built-up major roads in the rest of England with modelled concentrations greater than 40 μg m^{-3}. There are also less extensive roadside exceedences in Scotland, Wales and Northern Ireland.

Table 6.13 Summary of national empirical model results for annual mean NO_2 concentrations in 2001 (areas defined by 1 x 1 km background estimates, roadside concentrations calculated for built-up major road links).

	Area (km^2)	Number of links	Length of links (km)	Area > 40 $\mu g\ m^{-3}$ (km^2)	Number of road links > 40 $\mu g\ m^{-3}$	Length > 40 $\mu g\ m^{-3}$ (km)
Scotland	77769	435	1310.8	18 (0%)	138 (32%)	222.9 (17%)
Wales	20694	572	1022.0	2 (0%)	26 (5%)	49.8 (5%)
N Ireland	13627	124	950.2	0 (0%)	6 (5%)	47 (5%)
London	1591	1888	1766.9	182 (11%)	1759 (93%)	1588.3 (90%)
Rest of England	128519	6069	10922.4	559 (0%)	2709 (45%)	3619.6 (33%)
UK	242200	9088	15972.4	761 (0%)	4638 (51%)	5527.6 (35%)

699. The national roadside empirical modelling method has been calibrated using data from roadside or kerbside national automatic monitoring sites, so the estimated roadside concentration will be representative of the places in which the monitoring sites are located (typically 5–10m from the kerb). Thus a single estimate of the roadside concentration is calculated for each built-up major road link in the UK (A-roads and motorways; a link is defined as the length of road between junctions with other major roads). This is in contrast to more detailed local air quality modelling studies, which typically provide estimates of concentrations at a range of receptor points adjacent to an individual road link providing a more complete picture of the concentrations in all locations.

700. Concentrations at locations intermediate between roadside and background are not estimated in the national modelling. Similarly the national modelling does not account for the elevated concentrations at junctions (but see section 2.6.2.4 on the limitations of emissions estimates and dispersion modelling studies at junctions and Chapter 1 on the relevance of exposure at junctions). It should be noted, however, that no attempt has been made to assess the relevance of each road link modelled within the national modelling in terms of exposure. It is possible that there is no relevant exposure within 10 m of some of the roads modelled to exceed 40 $\mu g\ m^{-3}$. Such an assessment would be impractical on the national scale and is more appropriate to the LAQM review and assessment process discussed in section 7.3.10.

701. The modelling results are also subject to considerable uncertainties, although the overall uncertainty is less than the ±30% required by the EU AQDD1 for assessment with respect to the annual mean NO_2 LV (section 5.2.1.8). An analysis of the frequency distribution of modelled concentrations for both current case and projections for future years is included in Chapter 7. This assessment shows the likely impact which small changes in modelled concentrations would have on the extent of predicted exceedences.

Figure 6.51 Estimated annual mean background NO_x concentration, 2001 (µg m^{-3}, as NO_2).

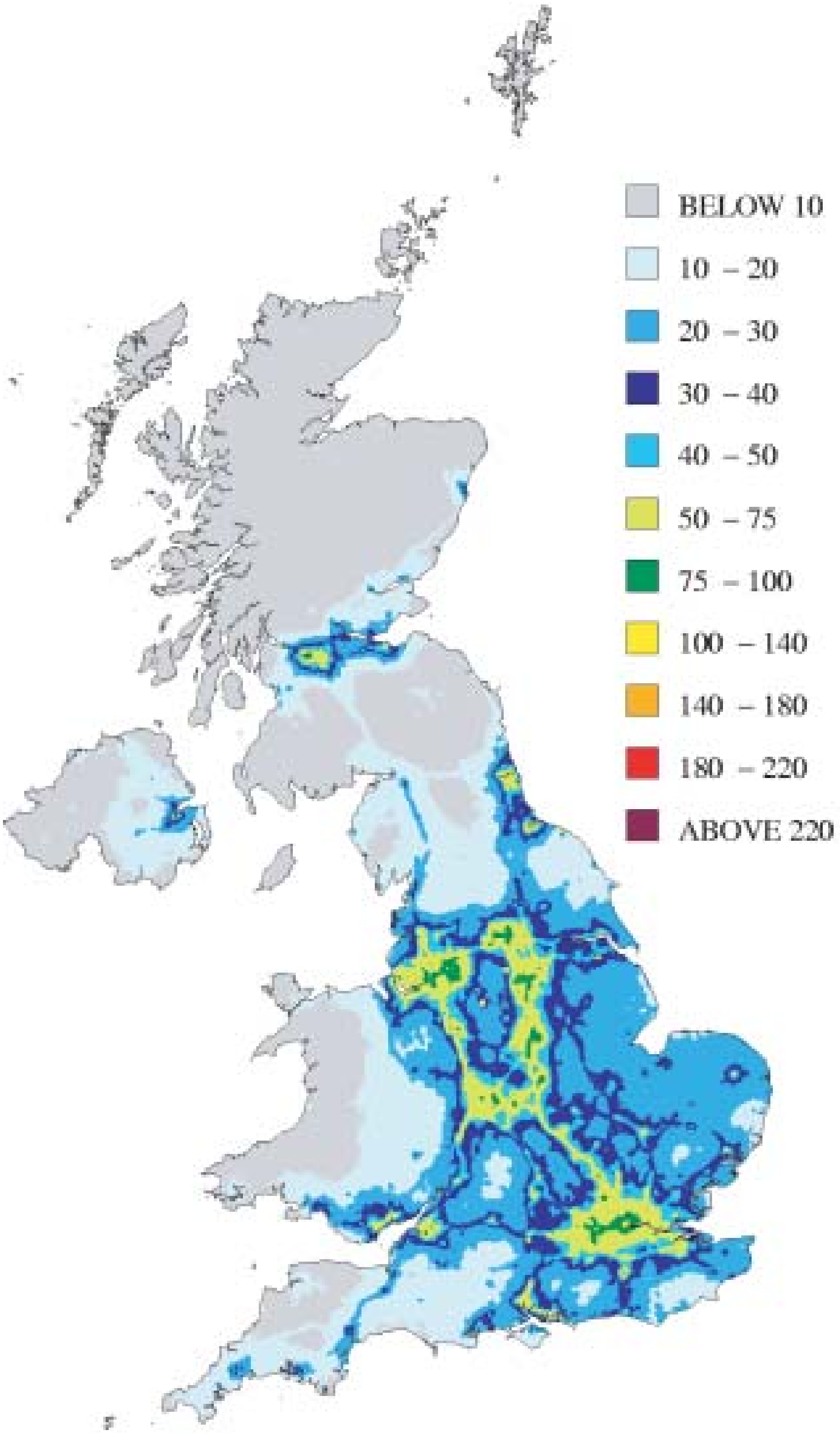


Figure 6.52 Estimated annual mean background NO_2 concentration, 2001 (µg m^{-3}).

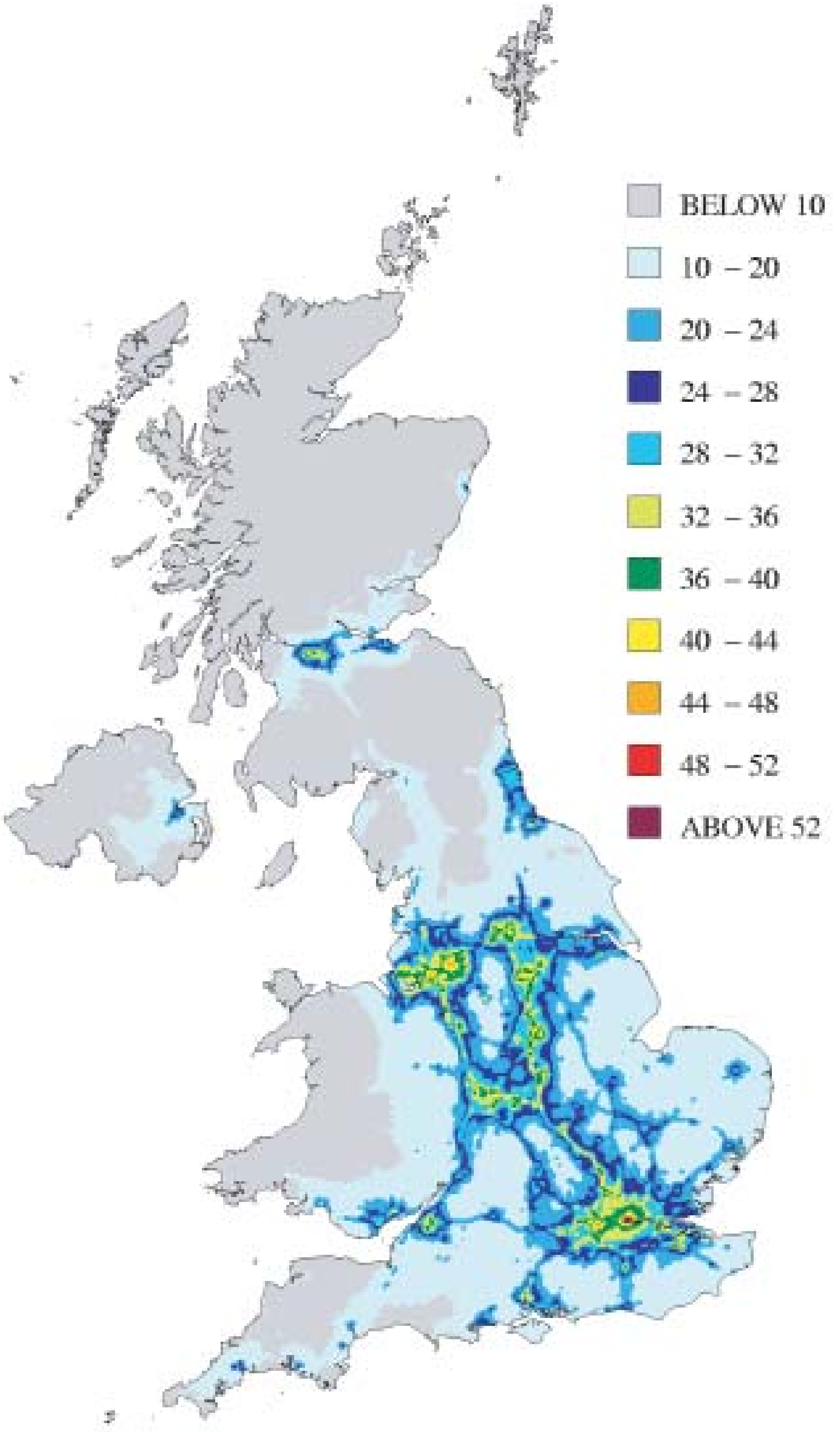


Figure 6.53 Estimated annual mean roadside NO_x concentration, 2001 ($\mu g\ m^{-3}$, as NO_2).

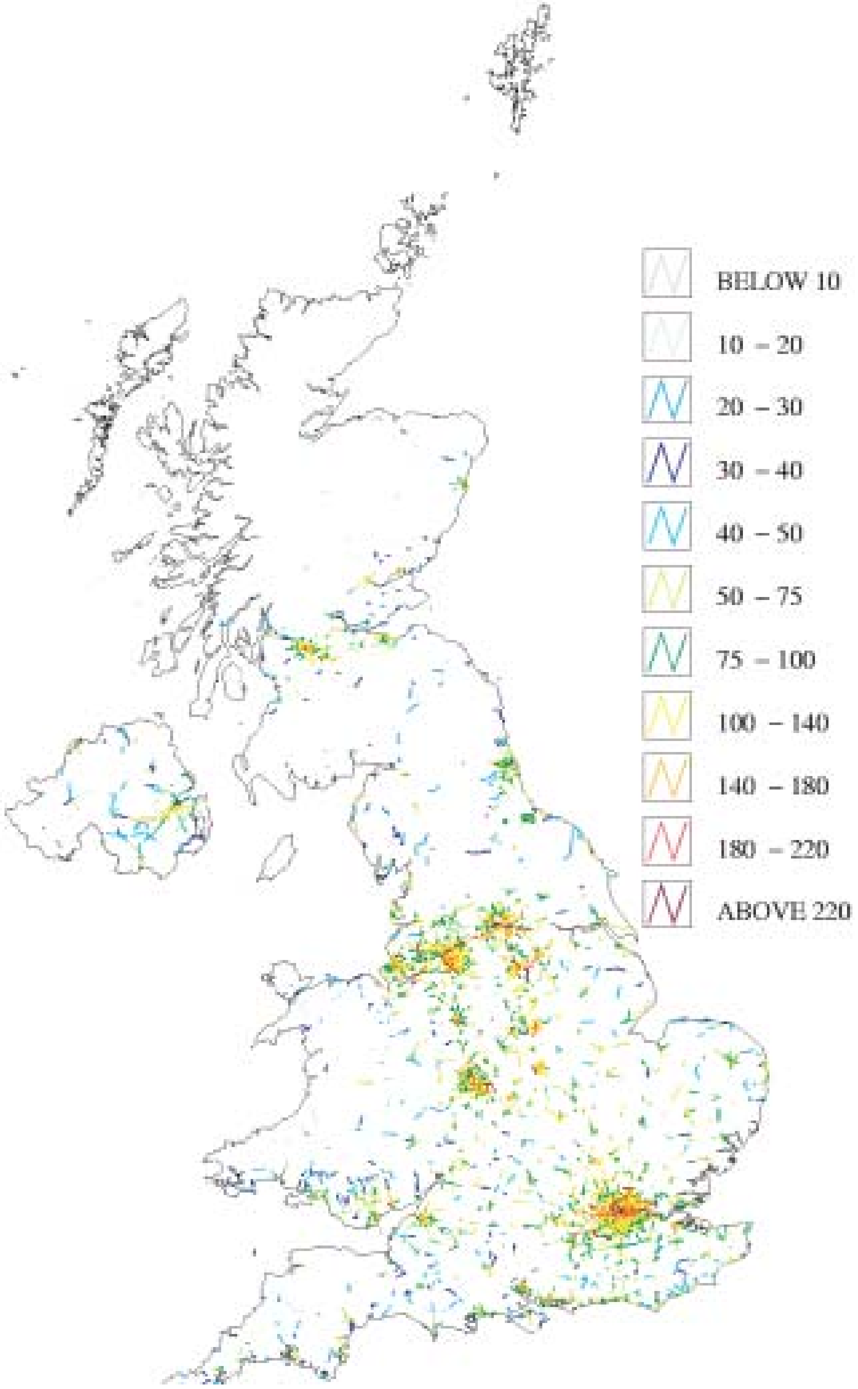
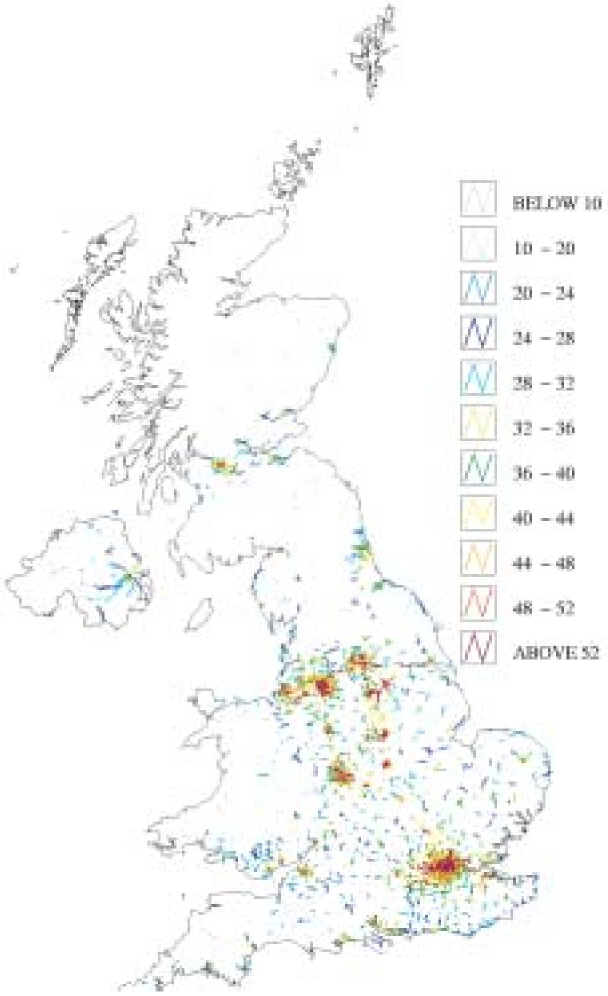


Figure 6.54 Estimated annual mean roadside NO_2 concentration, 2001 ($\mu g\ m^{-3}$).



6.4.3 Detailed modelling results for London

702. Annual mean NO_2 concentrations across London have been modelled by ERG for 1999 and 2001, by CERC for 1999 and by the Met Office using the LRCTM for 1999. These modelling studies are complimentary to the netcen national modelling work. They provide some confirmation of the validity of the results of the national modelling. They provide a much more detailed assessment of the extent of exceedences in London and this also aids the interpretation of the national modelling results in other areas.

703. Figures 6.55 and 6.56 show predictions of annual mean NO_2 concentrations for 1999 and 2001 in London calculated using the ERG approach (see section 5.2.3) The ERG 2001 results are based on projections from the 1999 inventory. An initial comparison with measurements suggests the modelled annual mean NO_2 concentrations compare reasonably with observations during 2001. The figure shows that almost all of central and inner London exceeds 40 $\mu g\ m^{-3}$ as well as locations close to roads in outer London. Exceedences are also shown in the vicinity of Heathrow Airport. The calculated area of Greater London exceeding 40 $\mu g\ m^{-3}$ in 1999 is estimated to be 627 km^2 (34%), declining to 361 km^2 (19%) in 2001. This value for 2001 is somewhat higher than the 11% predicted in the national modelling for 2001 (Table 6.13). A strict comparison is not however possible, as the ERG calculations relate to the total surface area exceeding 40 $\mu g\ m^{-3}$, whereas the national modelling relates only to background areas exceeding 40 $\mu g\ m^{-3}$.

Figure 6.55 Annual mean NO_2 concentrations for 1999 in London estimated using the ERG model (µg m^{-3}).

Figure 6.56 Annual mean NO_2 concentrations for 2001 in London estimated using the ERG model (µg m^{-3}).

704. Estimates of the length of major roads (A-roads and motorways) exceeding an annual mean NO_2 concentration of 40 µg m^{-3} in London have also been made using the ERG model. The length of road statistic is somewhat arbitrary because the statistic depends on the distance from the road at which the statistic is calculated. Calculations have been made at a distance of 10 m from the road centreline based on receptor points placed at 100 m intervals along each side of the major roads in the Greater London area. Table 6.14 summarises the ERG modelling results for 1999 and 2001 in terms of the lengths of road exceeding an annual mean concentration of 40 µg m^{-3}. In 1999 it is estimated that 95% of the major road length exceeded 40 µg m^{-3}, declining to 84 % in 2001. This is in close agreement with the estimate of 90% derived from the national modelling. The percentage of all of the roads in the LAEI (major and minor) exceeding 40 µg m^{-3} is lower than for major roads only. This is as expected because the traffic flows and hence emissions are generally less on the more minor roads.

Table 6.14 Summary of ERG model results for annual mean roadside NO_2 concentrations in 1999 and 2001 in London (roadside concentrations calculated for major roads, minor roads and all roads in the LAEI).

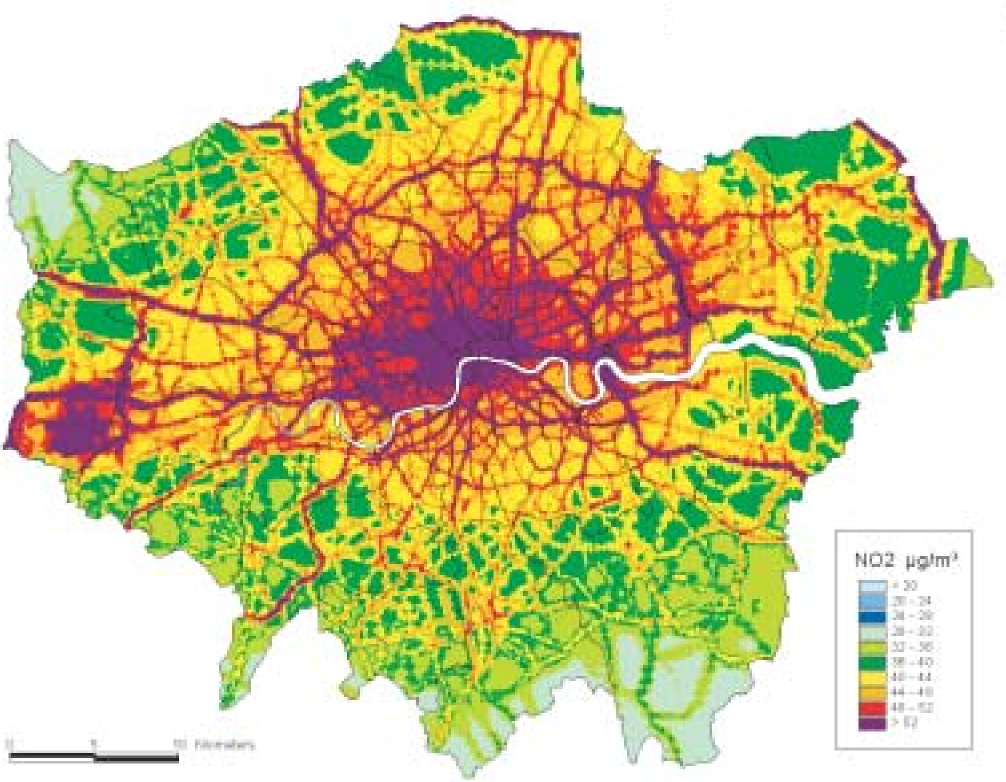
	Length of major road > 40 µg m^{-3} (km and percent)	**Length of minor roads > 40 µg m^{-3} (km and percent)**	**Length of all LAEI roads > 40 µg m^{-3} (km and percent)**
Total length modelled	1937	2076	4013
1999	1880 (95%)	1443 (70%)	3323 (83%)
2001	1627 (84%)	1134 (55%)	2761 (69%)

705. Figure 6.57 shows predictions of annual mean NO_2 concentrations for 1999 in London calculated by CERC using ADMS urban (see section 5.3.3). The figure shows a considerably greater area exceeding 40 µg m^{-3} (68% of Greater London) than predicted in the other modelling studies (Figures 6.54, 6.57 and 6.58). Once again exceedences are also shown in the vicinity of Heathrow Airport. Estimates of the length of roads exceeding 40 µg m^{-3} in 1999 in London have also been made using the CERC model and are listed in Table 6.15. Lines representing the location of the major roads within Greater London have been overlaid onto the modelled concentration maps. For each road the concentration has been calculated as the average of all the modelled concentrations for the 10 x 10 m grid squares intersected by the road. In 1999 it is estimated that 98% of the major road length exceeded 40 µg m^{-3} and 89% of all the roads in the LAEI exceeded this concentration. This is in agreement with the estimates from the other modelling studies. All of the studies suggest that most of the A roads and motorways in London exceeded 40 µg m^{-3} in 1999 and 2001.

Table 6.15 Summary of CERC model results for annual mean roadside NO_2 concentrations in 1999 and 2001 in London (roadside concentrations calculated for major roads, minor roads and all roads in the LAEI).

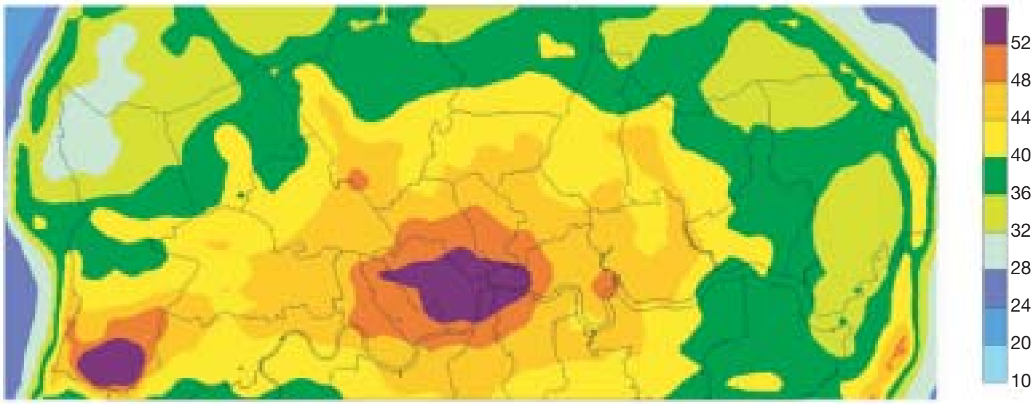
	Length of major road > 40 µg m^{-3} (km and percent)	**Length of minor roads > 40 µg m^{-3} (km and percent)**	**Length of all LAEI roads > 40 µg m^{-3} (km and percent)**
Total length modelled	1746	1910	3656
1999	1659 (98%)	1591 (83%)	3250 (89%)

Figure 6.57 Annual mean NO_2 concentrations for 1999 in London estimated using the CERC model ($\mu g\ m^{-3}$).



706. The LRCTM (see section 5.4) has also been used to estimate background NO_2 concentrations in London using the 1999 LAEI inventory and the resulting map is shown in Figure 6.58, covering over 1500 km² of north and central London and its outskirts. The annual mean NO_2 concentration for this selected area was found to be 39.5 $\mu g\ m^{-3}$. The highest annual mean NO_2 concentrations in the LRCTM model results are found in the area between Heathrow Airport and the M25. The maximum annual mean NO_2 concentration was found to be 67.1 $\mu g\ m^{-3}$. The maximum annual mean NO_2 concentration in central London was found to be 57.4 $\mu g\ m^{-3}$.

Figure 6.58 The spatial distributions of the annual mean NO_2 concentrations predicted with the LRCTM model with 1999 LAEI NO_x emissions ($\mu g\ m^{-3}$).



707. The estimates of background concentrations from the CERC and LRCTM modelling are higher than those from the netcen and ERG modelling. This highlights the uncertainties associated with modelled assessments of air quality. An examination of maps of annual mean NO_x derived from the CERC and netcen studies suggests that different treatments of the conversion of NO_x to NO_2 in the different models is likely to be the most important cause of the difference. The CERC ADMS Urban calculations derived NO_2 from NO_x on an hourly basis using a simplified chemical reaction scheme while both the netcen and ERG approaches applied empirical relationships to estimates of annual mean NO_x concentrations. The estimates of roadside concentrations are more consistent across the different studies. Some of the differences between the model results are also explained by the different base years of the calculations. Measured annual mean NO_2 concentrations in London were lower in 2001 than in 1999 by about 7% (averaged over 19 national network sites; range +4% to -24%). The area of London with concentrations greater than 40 µg m^{-3} is very sensitive to changes in concentrations, as shown by the comparison of the 1999 and 2001 model results in Figures 6.57 and 6.58. The results of the different modelling methods are compared further in section Chapter 7.

6.5 NO_2 episodes

6.5.1 Introduction

708. Episodes when hourly mean NO_2 concentrations exceed 200 µg m^{-3} can be classified into two types. More efficient oxidation of NO than under non-episode conditions plays an important role in both types of episode. Winter episodes are associated with the poor dispersion of primary NO_x emissions and elevated NO_2 concentrations are derived, at least in part, from the reaction of NO with O_2. Summer episodes are characterised by the elevated secondary NO_2 concentrations associated with regional scale photochemical oxidant episodes. Examples of both types of episodes during 2001 are presented here in order to illustrate the causes of these events.

709. A comparison of measured hourly NO_x and NO_2 concentrations during the episode in December 2001 and episodes in the early 1990s shows that the reaction of NO with O_2 can still lead to significant exceedences of 200 µg m^{-3} during extreme weather conditions, even with the decline in NO_x emissions from traffic since the early 1990s.

6.5.2 Analysis of the percentage of hours with NO_2 concentrations above 200 µg m^{-3} in the summer and winter

710. Since NO_2 episodes can generally be classified into two different types, with different causes, it is important to understand the relative importance of the two types in determining the number of exceedences. A detailed analysis of the conditions leading to each hour with a measured NO_2 concentration above 200 µg m^{-3} within the database of measured concentrations would be impractical. Figure 6.59, however, shows an analysis of the exceedences 200 µg m^{-3} at national network sites from 1988 to 2001 classified by the time of year in which the events were recorded. The number of hours with concentrations above 200 µg m^{-3} per site per year and the number of sites in the network in each year are shown in Figure 6.60.

Figure 6.59 The distribution of hours with NO_2 concentrations greater than 200 µg m^{-3} between the winter (January to March and October to December) and the summer (April to September) periods.

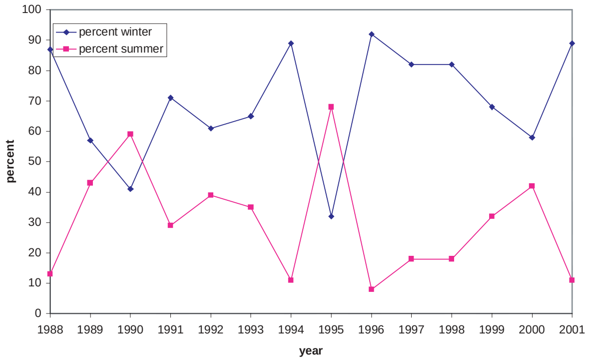
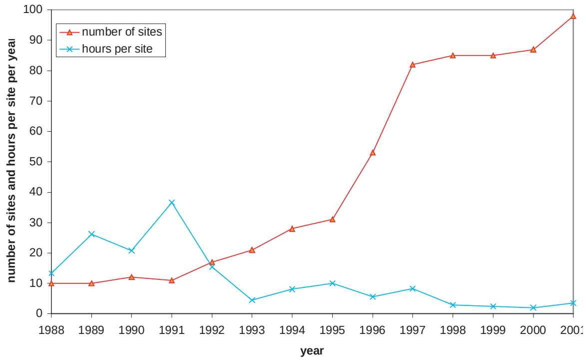


Figure 6.60 The number of sites in the national network and the number of hours per site per year with NO_2 concentrations greater than 200 µg m^{-3}.



711. The number of monitoring sites has increased approximately 10-fold since 1988 and the number of episodes per site has decreased as emissions have declined (and the number of sites has increased including a greater proportion of sites outside the major conurbations). The majority of hours above the 200 µg m^{-3} are in the winter for most years. Years with notable photochemical episodes (1989, 1990, and 1995) are clearly seen. A slight downward trend in the percentage of summer episodes is evident. This could be because the emissions leading to peak oxidant events have declined faster than local NO_x emissions.

6.5.3 An example of a winter NO_2 episode: 10 to 12 December 2001 in the North West of the UK

712. NO_2 concentrations above 200 µg m^{-3} were measured at a number of national network monitoring sites in the north west of the UK in mid December 2001. Concentrations above 200 µg m^{-3} were measured at sites in Greater Manchester, Glasgow, Belfast, Liverpool, Edinburgh and West Yorkshire. The highest concentrations were observed in Greater Manchester and Glasgow. Figures 6.61 and 6.62 show times series of NO_x and NO_2 concentrations at selected sites. The meteorological conditions associated with this type of episode are discussed in section 3.2.5.3.

Figure 6.61 Hourly NO_x concentrations during the episode in December 2001 at selected sites.

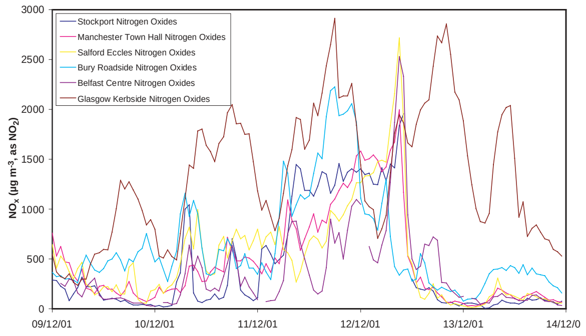
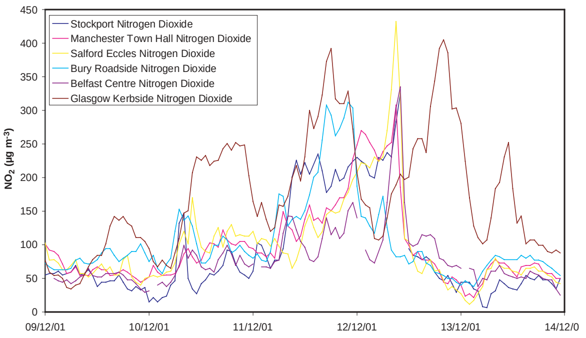


Figure 6.62 Hourly NO_2 concentrations during the episode in December 2001 at selected sites.



713. Figure 6.63 shows a comparison of the measured hourly concentrations of NO_x and NO_2 at two sites (Marylebone Road and Glasgow Kerbside) for the 4th quarter of 2001. Data for the 4th quarter of 1991 at Cromwell Road are also shown for comparison. This graph illustrates that the behaviour of the NO_2 concentrations at Glasgow Kerbside and Marylebone Road is similar up to about 1000 μg m^{-3} of NO_x, with a high NO_2/NO_x ratio at very low NO_x, rapidly flattening off as all available oxidant (O_3) is exhausted. The majority of NO_2 at NO_x concentrations in the range between about 100 and 1000 μg m^{-3}, as NO_2, will be primary NO_2. The gradient then becomes steeper as the NO + NO + O_2 chemistry becomes important, forming more secondary NO_2 (the rate of this reaction depends on the square of the NO concentration, see section 3.31. This behaviour is confined to winter episode conditions, such as were seen in the north west of the UK in December 2001 but not in London at this time. The NO + NO + O_2 chemistry is important because it leads to a greater proportion of the total NO_x concentration being oxidised to NO_2 at higher NO_x concentrations (above about 1000 μg m^{-3}, as NO_2) than at lower concentrations (between about 100 and 1000 μg m^{-3}, as NO_2). An examination of Figure 6.63 would suggest that a measured NO_2 concentration of 400 μg m^{-3} would consist of about 15% of secondary NO_2 oxidised by regional O_3, 25% oxidised by the NO + NO + O_2 reaction and about 60% directly emitted as primary NO_2.

714. The comparison with Cromwell Road during 1991 is interesting. Hourly NO_2 concentrations in London during December 1991 were the highest recorded in UK since monitoring starting in the 1970s (Bower *et al.*, 1994). The NO_x concentrations up to 3000 μg m^{-3} observed in 2001 are of similar magnitude to those observed in December 1991 in London even with the introduction of cleaner vehicles over the intervening period. NO_2 concentrations were generally lower at the same NO_x concentration in 1991 than in 2001, when NO_x was in the range 100-750 μg m^{-3}. This is presumably due to an increase in the ratio of primary NO_2/NO_x in emissions in 2001 caused by changes in vehicle technology, or the greater prevalence of diesel vehicles. The change in slope to higher NO_2/NO_x ratios is at several hundred μg m^{-3}, as NO_2, higher NO_x in 1991 than in 2001, also consistent with an increase in primary NO_2 over this period

715. Figure 6.64 shows a similar comparison of hourly NO_x and NO_2 at Salford Eccles and Manchester Town Hall in the 4th quarter of 2001 and Manchester Town Hall in the 4th quarter of 1992. The two sites show very similar behaviour in 2001, with slightly higher NO_x and NO_2 recorded at Salford Eccles. Manchester Town Hall shows similar behaviour in 1992 at high NO_x concentrations but rather lower NO_2 at the same NO_x at lower, more typical hourly NO_x concentrations. Again this observation is consistent with an increase in primary NO_2 emissions relative to total NO_x emissions between 1992 and 2001.

Figure 6.63 Hourly NO_x and NO_2 at Glasgow Kerbside 2001, London Marylebone Road 2001 and Cromwell Road 1991.

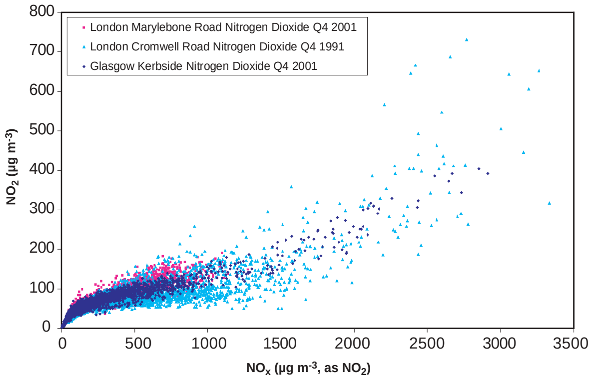
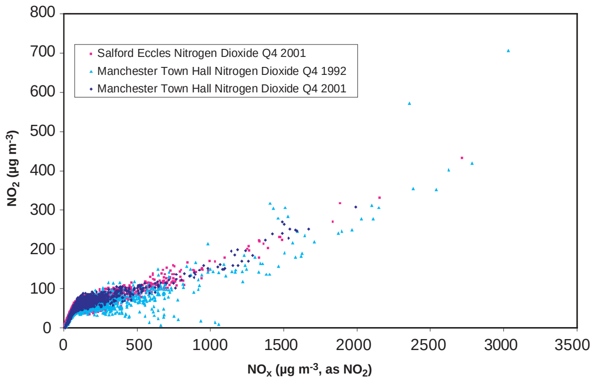


Figure 6.64 Hourly NO_x and NO_2 at Salford Eccles 2001 and Manchester Town Hall 2001 and 1992.



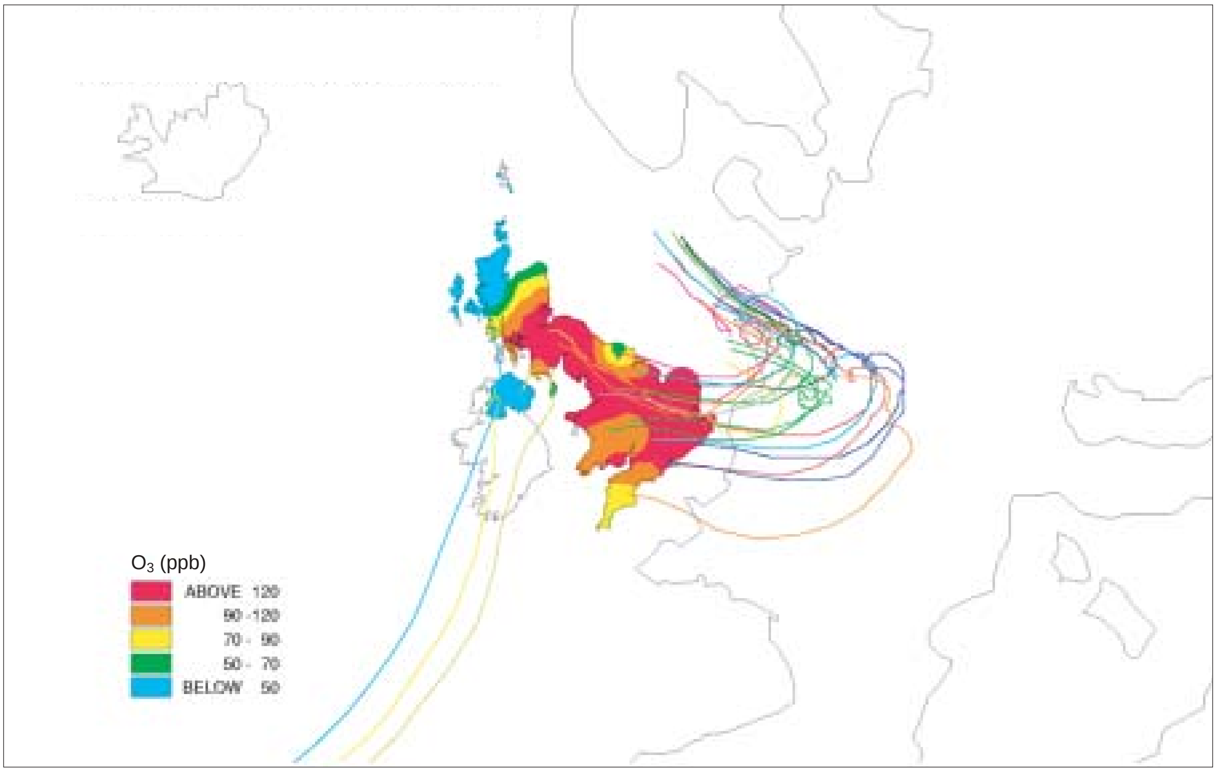
716. The interpolation of hourly NO_2 measurement data from this episode in the Greater Manchester area has been discussed in section 5.6.3.1. Hourly NO_x concentrations during winter episodes exhibit extensive spatial variation due to meteorological and topographic variation and particularly the small-scale spatial variation in emission strength (this was minimised to some extent in the NO_2 analysis presented in section 5.6.3.1 by the exclusion of roadside monitoring sites from the interpolation). The spatial variation in NO_2 concentrations would be expected to be rather less, with the same influences of meteorological and topographic variation and less variation with emissions strength. This is because the observed NO_2 will be a combination of secondary NO_2 formed by oxidation with O_3 to some extent 'filling in the gaps' between the locations close to sources where primary NO_2 will dominate. This is, however, not the case during extreme winter episode conditions when the NO + NO + O_2 reaction becomes important and oxidation becomes more efficient. Surface concentration modelling using standard techniques is likely to be unable to provide a realistic representation of ambient concentrations if the combination of emissions and meteorology mean that this reaction is only having a large effect at some locations.

717. Four out of five of the occasions when a site has recorded more than 18 hours above 200 µg m^{-3} and annual mean concentrations equal to or lower than 40 µg m^{-3} were in Greater Manchester during 2001. It is possible that increases in the proportion of NO_x emitted as NO_2 could lead to more frequent exceedences of an hourly mean concentration of above 200 µg m^{-3} at sites with annual mean concentrations less than 40 µg m^{-3} in the future.

6.5.4 An example of a summer time NO_2 episode: 26 June 2001 in London

718. This is an example of an NO_2 episode related to a summertime photochemical episode. NO_x concentrations are much lower under these conditions than during winter NO_2 episodes. The elevated levels of oxidant available during these episodes lead to a greater conversion of NO to secondary NO_2 than under non-photochemical episode conditions. Summer episodes are generally associated with periods of warm weather and light winds from the east bringing oxidant precursors to the UK (see section 3.2.5.4). O_3 concentrations and the air mass back trajectories during this episode are illustrated in Figure 6.65.

Figure 6.65 Forecast O_3 concentrations and associated back trajectories 26 June 2001 12:00.



719. Hourly concentrations of NO_x, NO_2, O_3 and total oxidant ($NO_2 + O_3$) during the episode are shown in Figures 6.66 to 6.69. The highest O_3 concentrations were recorded at the rural site of Lullington Heath peaking at 198 µg m^{-3} on 26th June 2001. Oxidant concentrations at this site were dominated by O_3. NO_x and NO_2 concentrations were higher at the suburban London Bexley site. O_3 concentrations were generally slightly lower than at Lullington Heath. Oxidant concentration was mostly O_3 on 24th June 2001 but with contributions from both O_3 and NO_2 on 26th June 2001. NO_x and NO_2 concentrations were higher still at the London Bloomsbury urban centre site. The peak NO_2 concentration on 26th June 2001 was 161 µg m^{-3}. Much higher NO_x concentrations were recorded at the Marylebone Road site and oxidant concentrations were dominated by NO_2. O_3 concentrations were low. The peak NO_2 concentration at Marylebone Road on 26th June 2001 was 258 µg m^{-3}.

720. Looking at the four sites together, O_3 concentrations decline as expected with increasing NO_x concentration. Oxidant levels follow a broadly similar temporal pattern at the four sites driven by the diurnal variation in O_3 concentrations. Oxidant concentrations were higher at Marylebone Road on the 26-27/06/2001 than at the other sites, presumably in part due to direct emission of NO_2 from vehicles on the road. The variation in oxidant concentrations is more similar to the variation in NO_2 concentrations at this site.

Figure 6.66 Hourly NO_x concentrations during the episode in June 2001 at selected sites.

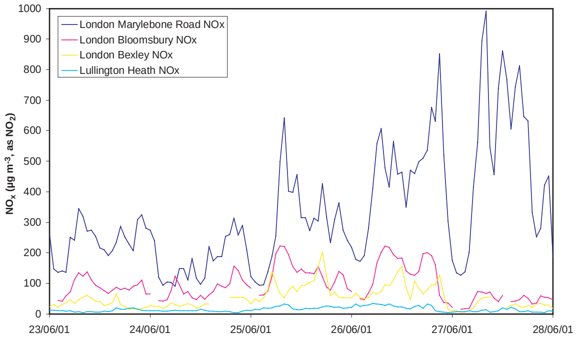


Figure 6.67 Hourly NO_2 concentrations during the episode in June 2001 at selected sites.

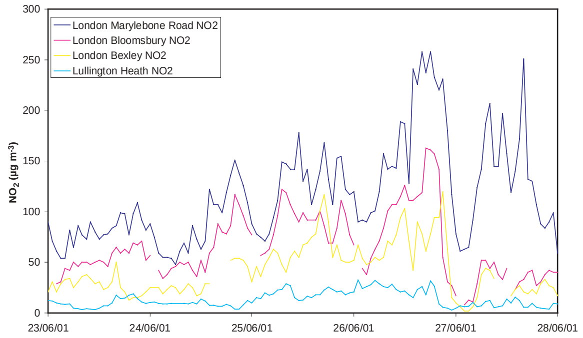


Figure 6.68 Hourly O_3 concentrations during the episode in June 2001 at selected sites.

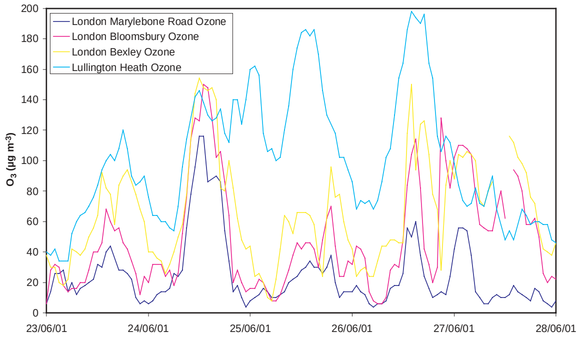
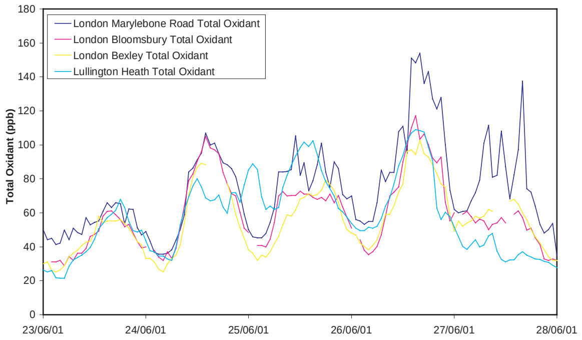


Figure 6.69 Hourly total oxidant ($NO_2 + O_3$) concentrations during the episode in June 2001 at selected sites.



6.6 Influence of changes in mean O_3 concentrations on annual mean NO_2 concentrations

6.6.1 Introduction

721. It is recognised that three factors, amongst many others, may have influenced the long-term trends in the NO_2 concentration distribution:

- the integrated NO_x emissions in the upwind environment;
- the O_3 oxidation capacity which converts NO to NO_2;
- changing direct emissions of NO_2.

722. In urban areas, NO_2 levels are largely determined by primary emissions of NO_2 and by the oxidation capacity of the O_3 in the upwind rural environment. There are generally always enough NO_x emissions to convert the upwind O_3 from the regional background into urban NO_2. In rural areas, the oxidation capacity of the O_3 is usually much larger than that required to convert the majority of the NO_x emissions into NO_2. In urban areas NO_2 levels are controlled by the availability of O_3 in the upwind environment and by direct NO_2 emissions whereas in the rural areas NO_2 levels are controlled by the availability of NO_x emissions.

723. The oxidising capacity of the O_3 in rural areas is in turn influenced by a number of factors. Much of the O_3 in rural areas of the UK is advected in to the British Isles from the north Atlantic region on south-westerly, westerly and north-westerly winds. Under these conditions, rural O_3 levels are close to northern hemisphere baseline concentrations. However, when other meteorological conditions prevail, rural O_3 levels are influenced by O_3 sources and sinks in the UK and in the rest of Europe. This oxidation capacity needs to be quantified and an assessment made of whether or not there have been any changes in its magnitude over the recent past in order to understand the long-term trends in NO_2 levels in the UK.

6.6.2 Measured O_3 concentrations at Mace Head

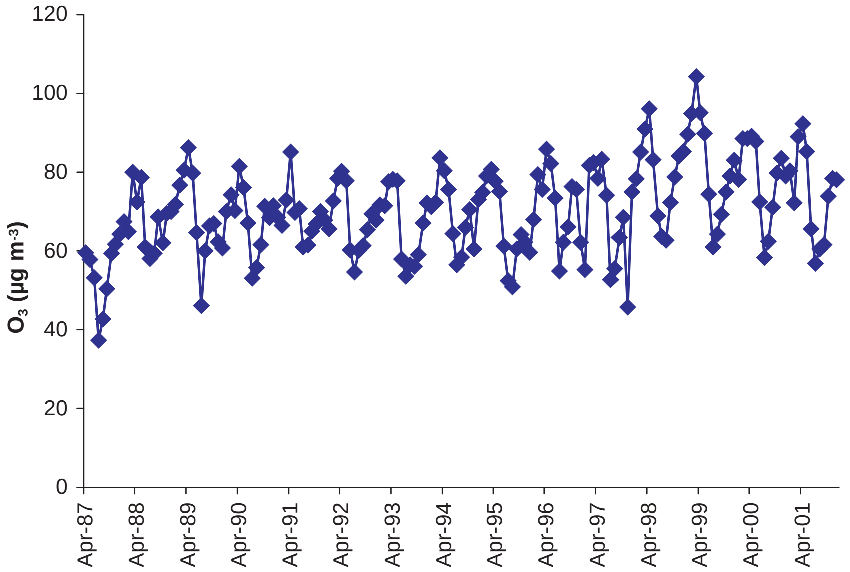
724. The O_3 monitoring records have been examined for the remote atmospheric baseline station at Mace Head in Ireland on the Atlantic Ocean coastline, provided by Professor Peter Simmonds of the University of Bristol. Each hourly O_3 observation has been assigned either to the 'unpolluted' category, having travelled across the Atlantic Ocean, or 'polluted', if it had crossed any part of the British Isles or European mainland. The 'unpolluted' category represents the air masses which enter the UK from the north Atlantic region and characterises the oxidation capacity that underpins much of UK urban NO_2 air quality.

725. Figure 6.70 illustrates the time series of the monthly mean O_3 concentrations in the 'unpolluted' category from 1987 through to 2000. The figure shows a largely level period after an initial rise, followed by the last few years with a rising trend. Monthly mean O_3 concentrations have risen with a statistically-significant upwards trend of +0.96 µg m^{-3} per year over the entire period from 1987 to 2001. Table 6.16 presents the corresponding trends by month in this dataset. All months show a trend towards increasing monthly mean concentrations and for ten months of the year these trends are statistically significant using the Sen's method. Estimated trends are greatest during the months from October to May and smallest from June through to September. The months with the maximum and minimum trends are February +1.82 µg m^{-3} per year and August +0.35 µg m^{-3} per year.

Table 6.16 Trends in the monthly mean O_3 concentrations in 'unpolluted' air masses by month at Mace Head, Ireland over the period from 1987 to 2001 and their level of statistical significance (+ at the 10%, * at the 5% and ** at the 1% probability level).

Month of the year	Statistical significance	Trend in µg m^{-3} per year
Jan	+	0.924
Feb	*	1.822
Mar	**	1.390
Apr	*	1.384
May	**	1.104
Jun	*	0.958
Jul	+	0.682
Aug	+	0.350
Sep		0.422
Oct	*	1.172
Nov		1.182
Dec	*	1.080

Figure 6.70 Mean monthly O_3 concentrations in µg m^{-3} in 'unpolluted' air masses at the Mace Head, Ireland atmospheric baseline station over the period from 1987 to 2000.



726. This analysis indicates that there has therefore been a considerable upwards trend in the northern hemisphere O_3 baseline concentrations in air masses entering the UK which is particularly apparent in the winter and spring. This should have resulted in greater extents of oxidation of NO_x emissions into NO_2 concentrations, all other factors being equal, in urban areas where the supply of oxidants controls the levels of NO_2.

727. To see if there is any influence of this hemispheric baseline increase in O_3 on NO_2 levels in the United Kingdom, the O_3 data for each of the long-running sites in the rural O_3 monitoring network has been examined. The statistic chosen was the monthly mean maximum hourly O_3 concentration recorded in each month from 1990 through to 1999. (The trend in monthly mean O_3 concentrations over this period would be expected to be dominated by decreases in local NO_x emissions.) The trends were determined using the non-parametric Mann-Kendall test and the Sen's slope estimate. The results are presented in Table 6.17. In contrast to the corresponding analysis of the carefully sorted Mace Head data in Table 6.16, few of the estimates in Table 6.17 are statistically significant.

Table 6.17 Estimates of the annual trends in monthly maximum O_3 concentrations in $\mu g\ m^{-3}$ per year over the period 1990-1999 for each site in the UK rural O_3 monitoring network. The shading shows those trend estimates that are negative.

	AH	Bot	Bus	Esk	Gla	Har	HM	Lad	LN	Sib	SV	YW
Jan	0.4	2.2	0.2	0.6	1.0	0.4	1.0	2.0	0.0	1.8	1.0	0.6
Feb	0.8	2.4	1.0	1.2	1.8	0.6	1.2	1.2	1.2	0.8	1.2	1.0
Mar	0.0	1.8	0.6	0.6	1.0	-0.2	1.4	1.2	0.2	0.0	1.0	0.0
Apr	-0.6	2.0	0.6	-0.4	-0.8	-0.6	0.2	0.6	-0.2	-1.0	-0.4	-1.0
Ma	-1.8	0.8	-0.6	-0.2	-1.0	-2.2	0.0	1.4	-1.0	-1.6	-0.6	-1.8
Jun	-0.4	1.2	0.0	-0.1	-0.6	-0.4	-0.6	0.2	0.2	-0.8	0.2	0.0
Jul	-2.6	-1.4	-1.2	-0.8	-1.6	-1.4	-2.2	-2.6	-1.4	-1.6	-0.8	-1.6
Aug	-0.6	1.4	0.4	1.8	1.6	-0.8	1.4	0.8	0.8	-1.2	1.2	-2.4
Sep	-0.8	3.0	0.8	0.8	0.8	0.6	0.8	0.6	-1.6	0.8	1.0	-0.4
Oct	-0.2	2.4	0.8	0.6	0.4	1.0	-0.2	0.2	-0.6	-0.2	1.0	-0.2
Nov	1.2	1.2	0.6	0.0	-0.2	1.2	-0.2	1.0	-0.4	0.8	1.6	1.0
Dec	1.2	2.4	1.0	0.8	0.8	1.6	0.4	1.0	0.8	0.6	1.4	1.2

AH, Aston Hill, Powys; Bot, Bottesford, Leicestershire; Bus, Bush Estate, Midlothian; Esk, Eskdalemuir, Dumfries and Galloway; Gla, Glazebury, Cheshire; Har, Harwell, Oxfordshire; HM, High Muffles, North Yorkshire; Lad, Ladybower, Derbyshire; LN, Lough Navar, Fermanagh; Sib, Sibton, Suffolk; SV, Strath Vaich, Highland; YW, Yarner Wood, Devon.

728. The pattern of O_3 trends revealed in Table 6.17 for the UK, is markedly different to those identified in Table 6.16 for Mace Head. There are a significant number of sites and months with downwards trends as indicated by the shading. This suggests that during the summer months, trends in the mean monthly maximum O_3 concentration have been declining over the UK. This is presumably due to the impact of emission reductions aimed at reducing the intensity of regional scale photochemical episodes. Because of the marked year-on-year variability in the timing of these photochemical episodes, the monthly trends are also highly variable and the trend estimates tend not to be statistically significant. More robust statistics such as the number of episode days per year and the maximum 8-hour mean O_3 concentration show clear downwards trends at all UK rural O_3 monitoring sites. However, as a result of the significant year-on-year variability in the timing of regional scale photochemical episodes and their decreasing magnitude and frequency, it has not been possible to discern whether there has been an influence during the summer months from the northern hemisphere baseline O_3 increase during the 1990s.

729. During the winter months, all the UK rural O_3 monitoring sites show upwards trends in the monthly mean maximum O_3 concentrations. Whilst this may be taken as confirmation that the increasing northern hemisphere O_3 baseline concentrations may well have influenced UK rural O_3 levels, this is not in itself conclusive. There are other influences that may explain the observed O_3 increases at the rural O_3 monitoring sites. For example, reducing traffic emissions of NO_x should have reduced the extent of NO_x depletion reactions and this may have led to increased O_3 levels in rural areas during wintertime.

6.6.3 Comparison of measured NO_x, NO_2, O_3 and total oxidant trends in London and Glasgow

730. The trends in annual mean concentrations of NO_x, NO_2, O_3 and total oxidant in London and Glasgow have been examined to see whether they are consistent with the increase in background O_3 concentrations at Mace Head. Figure 6.71 shows annual mean NO_x and NO_2 trends at West London. Annual mean regional O_3 at Lullington Heath and total oxidant (O_3 + NO_2) at London Bloomsbury (where co-located measurements are made) are also shown. Figure 6.72 shows a similar analysis at Glasgow City Chambers, with regional O_3 at Strath Vaich and total oxidant at Edinburgh Centre.

731. A summary of the gradients determined from these data by linear regression is presented in Table 6.18. The downward slopes in annual mean NO_x are very similar at West London and Glasgow Centre at -7.19 and -7.44 µg m^{-3} as NO_2. This is matched by a decline in annual mean NO_2 of -1.32 µg m^{-3} at West London but not at Glasgow City Chambers where annual mean NO_2 concentrations have remained roughly constant for the last 10 years (see section 6.2.3). Regional O_3 concentrations are roughly constant at Lullington Heath and increasing at Strath Vaich. Total oxidant levels show a small decline at both London Bloomsbury and Edinburgh Centre.

732. The different response of annual mean NO_2 to reductions in annual mean NO_x due to reductions in NO_x emissions in London and Glasgow could be due to the increase in hemispheric O_3 concentrations over this period. This increase may have had a greater effect in the north and west of the UK and less influence in the south and east where the background influence on regional O_3 levels will be much smaller, due to the lower frequency of westerly winds in the south east. Some oxidant may also be lost to dry deposition to the surface as westerly air masses approach the south east across England.

Table 6.18 Gradients determined by linear regression of annual mean concentrations. Available data between 1990 and 2001.

	West London	Glasgow City Chambers
NO_x (µg m^{-3} as NO_2) per year	-7.19 (-3.76 ppb)	-7.44 (-3.89 ppb)
NO_2 (µg m^{-3}) per year	-1.32 (-0.69 ppb)	+0.04 (+0.03 ppb)
O_3 (µg m^{-3}) per year	+0.04 (+0.02 ppb) (Lullington Heath)	+0.30 (+0.15 ppb) (Strath Vaich)
Total oxidant (ppb) per year	-0.31 (London Bloomsbury)	-0.38 (Edinburgh Centre)

Figure 6.71 Trends in annual mean NO_x, NO_2, O_3 and total oxidant in London.

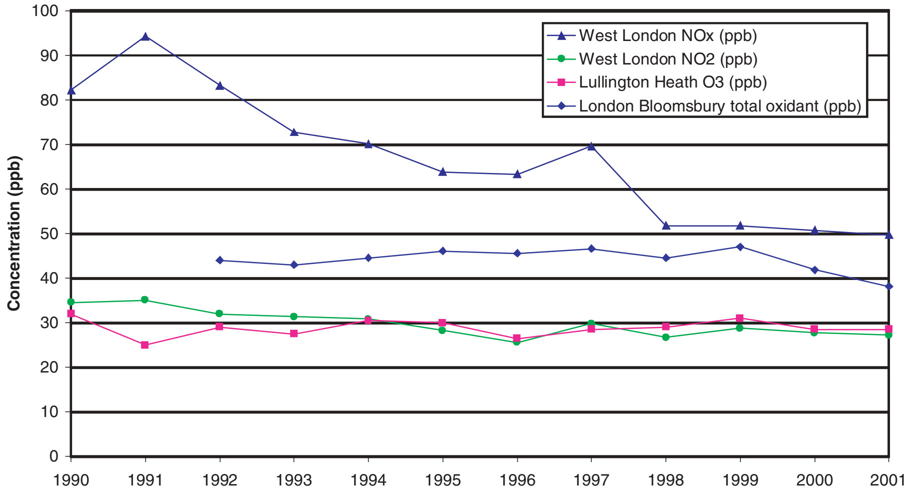
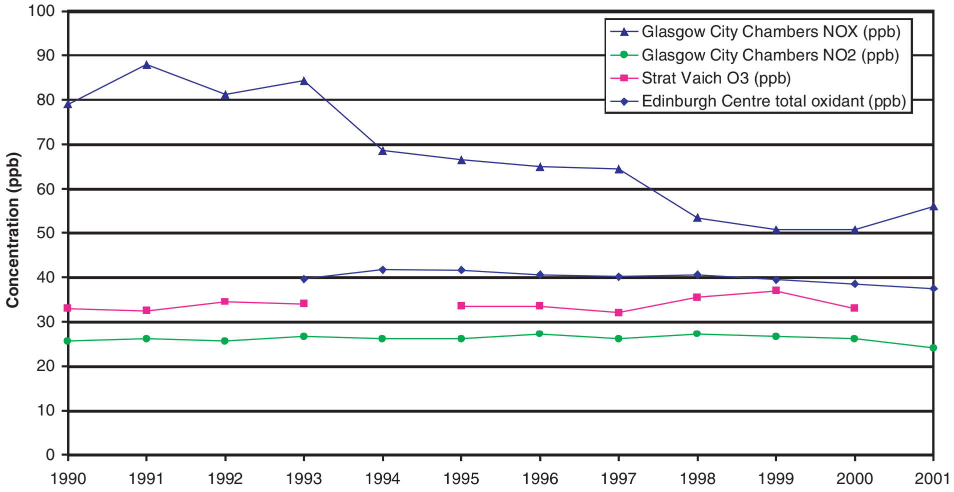


Figure 6.72 Trends in annual mean NO_x, NO_2, O_3 and total oxidant in Glasgow.



733. Summer episode O_3 concentrations have, however, also declined over this period. Such episodes have been and remain more frequent in south-east England than in Scotland. The contribution of summer episode oxidant to annual mean NO_2 concentrations in London is therefore likely to have declined over the last ten years, while the contribution to annual mean NO_2 concentrations in Glasgow will always have been much smaller. This may also have contributed to the difference in relative NO_x and NO_2 trends between these sites. This may have masked the effect of increasing hemispheric O_3 concentrations in London. Annual mean concentrations of both NO_x and NO_2 have declined at Belfast Centre since 1993, in contrast to the relative insensitivity of annual mean NO_2 concentrations at Glasgow City Chambers to NO_x reductions.

734. The impact of the increasing northern hemisphere baseline O_3 concentrations on O_3 and NO_2 levels in urban areas of the UK may well have been confounded by the decreasing intensity of summertime photochemical O_3 episodes and by decreasing NO_x depletion reactions during wintertime as discussed above and in section 6.2.3. It is clear that further research will be required to understand the influences of the changes in oxidant concentrations on NO_2 concentrations in the UK.

Chapter 7

Projected NO_2 concentrations in 2005 and 2010

Key points

- NO_x emissions from road traffic are projected to decrease in the future and the empirical and dispersion model studies indicate that this will lead to a significant reduction in exceedences of 40 µg m^{-3} at the roadside and in background locations, both nationally and in London.
- Projections of annual mean NO_2 concentrations in 2005 and 2010 have been calculated for the 212 monitoring sites for which data are available for 2001. The number of sites with annual mean NO_2 concentration exceeding 40 µg m^{-3} shows little decline between 2001 and 2005 in London but a much greater reduction in the rest of the UK. Concentrations are projected to decline further between 2005 and 2010 with the number of sites with projected concentrations exceeding 40 µg m^{-3} also expect to decline in London by 2010.
- The national modelling projections suggest that in 2005 an annual mean concentration of 40 µg m^{-3} is likely to be exceeded alongside 65% of major roads in London, 18% in the rest of England, 8% in Scotland, 3% in Northern Ireland and 2% in Wales. By 2010 this is expected to reduce to 23% of major roads in London, 5% in the rest of England, 2% in Scotland and zero in Northern Ireland and Wales.
- Additional detailed dispersion modelling studies for London suggest that, despite reductions in projected NO_x emissions between 1999 and 2010, projected exceedences of 40 µg m^{-3} are likely alongside 53-62% of the length of major roads in London in 2005, falling to 26-35% in 2010. Projections of the extent of exceedence in background locations are less consistent between different models, which predict exceedence areas in Greater London of between 5-32% for 2005 and 3-13% for 2010. The predicted areas of exceedences are, however, very sensitive to small changes in modelled concentrations.
- The Local Air Quality Management (LAQM) process has identified the need for approximately one third of the local authorities in Great Britain to declare air quality management areas. The majority of these are for NO_2. While national studies are inevitably focused on London and other conurbations, as well as on the most heavily trafficked roads, the LAQM studies have shown that exceedences can also occur in smaller towns, where narrow streets coincide with congested traffic. An analysis of automatic monitoring data collected as part of the LAQM process suggests that exceedences of 40 µg m^{-3} as an annual mean NO_2 concentrations will be more likely in major conurbations than outside. Outside conurbation, exceedences are more likely alongside single rather than dual carriageway roads. Exceedences will therefore be unlikely more than 9 m from the kerb of single carriageway roads and more than 5 m from the kerb of dual carriageway roads outside conurbations.
- Traffic sources and the total of domestic and commercial emissions are predicted to each contribute roughly one third of the total annual mean NO_x concentration by 2010 in central London. The remaining third includes contributions from regional rural concentrations, industry, other area and point sources.

- Analysis using the current policies NO_x emission projections in the national scale empirical model suggest that reductions in NO_x emissions well beyond those anticipated will be required if a concentration of 40 μg m^{-3} is to be met at all locations by 2005 or 2010. Site-specific modelling for the kerbside location monitoring site at Marylebone Road in London indicate that emission reductions of 90% relative to the 2005 base case traffic emissions or of 65% of the 2005 base case emissions for all sectors would be required to reduce the roadside annual mean NO_2 concentration to 40 μg m^{-3}. Slightly smaller percentage reductions relative to the base case emissions would be required in 2010 due to the expected decline in base case emissions between 2005 and 2010 as a result of current policies.
- Projections of the 99.8 percentile of 1-hour NO_2 concentrations have been calculated for London using a dispersion model. The results suggest that 1-hour exceedences of 200 μg m^{-3} will be less likely in 2005 and 2010 compared with 1999. Peak hourly NO_2 concentrations are, however, highly variable from year to year and dependent on the occurrence of particular summer or winter episode weather conditions.
- The empirical relationships between NO_x and NO_2 developed for the current atmosphere may not hold into the future if baseline O_3 concentrations increase and if the mass fraction of NO_x emitted directly as NO_2 also increases, leading in either case to increased difficulties with achieving an annual mean concentration of 40 μg m^{-3}. Modelling studies suggest that a 12 μg m^{-3} increase in baseline O_3 concentration could lead to an increase in annual mean NO_2 concentration in London of between about 4 and 7 μg m^{-3}. These issues highlight the complexities of predicting future NO_2 concentrations, involving the interactions of influences at global, regional and local scales.

7.1 Introduction

735. Projections of NO_2 concentrations in 2005 and 2010 are key to an assessment of NO_2 concentrations in the UK because the annual and 1-hour AQS objectives have been set for 2005 and subsequent years and the EU LVs apply from 2010 (see Chapter 1). Models are the only means of assessment for future concentrations. This is in contrast to the assessment of current and recent NO_2 concentrations presented in Chapter 6 where a combination of measurements and modelled estimates are available. There are a number of elements of a modelling assessment of future NO_2 concentrations that need to be reasonably well understood before a policy assessment can be made. These include the source apportionment of current concentrations, the expected trends in emissions and the non-linear responses of ambient concentrations to changes in emissions, such as the influences of background concentrations and atmospheric chemistry.

736. Projections of annual mean NO_2 concentrations have been estimated for individual monitoring site locations across the UK using the national empirical model. Projections of both annual mean and hourly NO_2 concentrations (for comparison with the 1-hour objective and LV) have been calculated for sites in London using a more detailed dispersion model. The site-specific projections have the advantage of enabling a direct comparison with current measured concentrations. The national model has also been used to calculate estimates of the emissions reductions that would be needed to reduce concentration to an annual mean concentration of 40 μg m^{-3} at selected monitoring site locations.

737. Mapped projections of annual mean NO_2 concentrations in 2005 and 2010 have been calculated for background and roadside locations across the UK. These results have been compared with the results of more detailed dispersion modelling studies for Greater London in terms of the expected extent of exceedence (areas and lengths of roads) of 40 μg m^{-3}. The frequency distributions of the modelled concentrations have been examined in order to assess the likely impact of small changes in model estimates on the extent of exceedence.

738. The projections presented here have been derived from a combination of base case emission inventory projections and the best estimates of the response of ambient concentrations to changes in emissions. The base case emission projections are based on an analysis of current national and international policies and their likely effect on future NO_x emissions. This chapter concludes with a discussion of some key issues in relation to predicting future NO_2 concentrations. These are the possible impacts of an increase in hemispheric background O_3 concentrations and an increase in the proportion of NO_x emitted as primary NO_2. Neither of these issues are sufficiently well understood at present to be fully incorporated into the base case projections. The sensitivity analyses presented here, however, indicate that these changes have some potential to offset the predicted decreases in NO_x emissions over the next ten years or so, resulting in smaller reductions in ambient NO_2 concentrations than suggested by the base case projections.

7.2 Site-specific projections of NO_2 concentrations in 2005 and 2010

7.2.1 Projections of annual mean concentrations derived from empirical site-specific models

739. Projections of annual mean NO_2 concentrations in 2005 and 2010 have been calculated by netcen from measured concentrations in 2001 using the site-specific projection models described in section 5.2.1.9 and applied in section 6.2.4 to the interpretation of recent trends in concentrations. These analyses and projections are based on the NAEI. Site-specific projections for selected sites are illustrated in Figures 7.1 to 7.4. These projections are listed in Table 7.1 for the sites selected for detailed analysis in Chapter 6. Projections have also been calculated using a simpler 'year factor' approach and the table shows the range of the projections calculated using these two different methods. Generic background and roadside year factors have been used to scale measured concentrations at all sites in this simpler method. This has the advantage of making the calculation much simpler but any spatial variation in emission trends is not accounted for. These 'year factors' have been calculated by averaging the results of site-specific projections at 11 background and 8 roadside national network sites. These year factors have also been published in the LAQM Technical Guidance TG(03) (DEFRA, *et al*, 2003).

Figure 7.1 Site specific projections of NO_x and NO_2 concentrations at West London (µg m^{-3}, as NO_2).

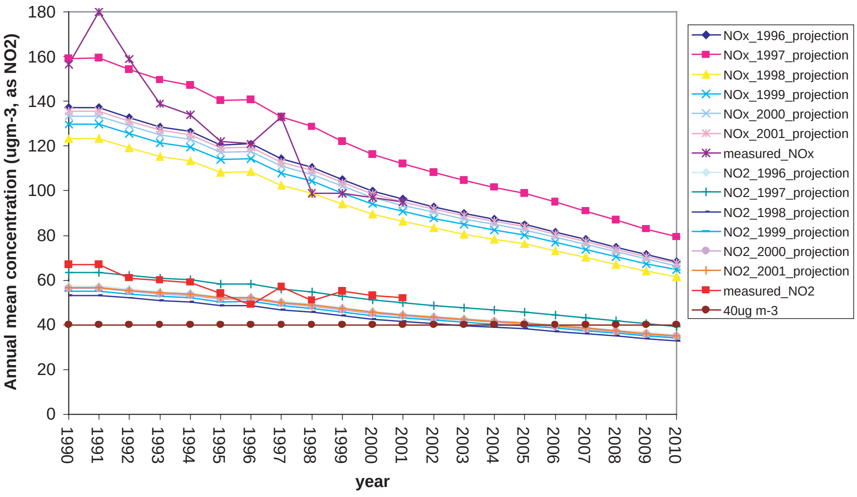


Figure 7.2 Site specific projections of NO_x and NO_2 concentrations at Manchester Town Hall ($\mu g\ m^{-3}$, as NO_2).

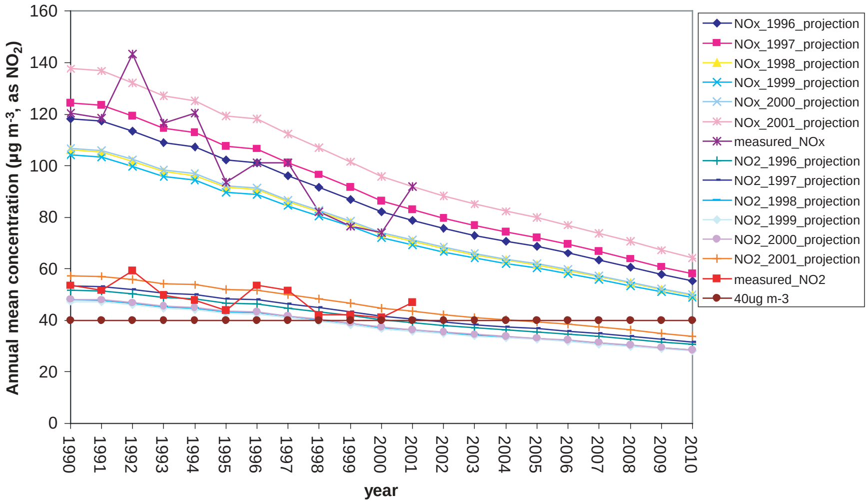


Figure 7.3 Site specific projections of NO_x and NO_2 concentrations at Belfast Centre ($\mu g\ m^{-3}$, as NO_2).

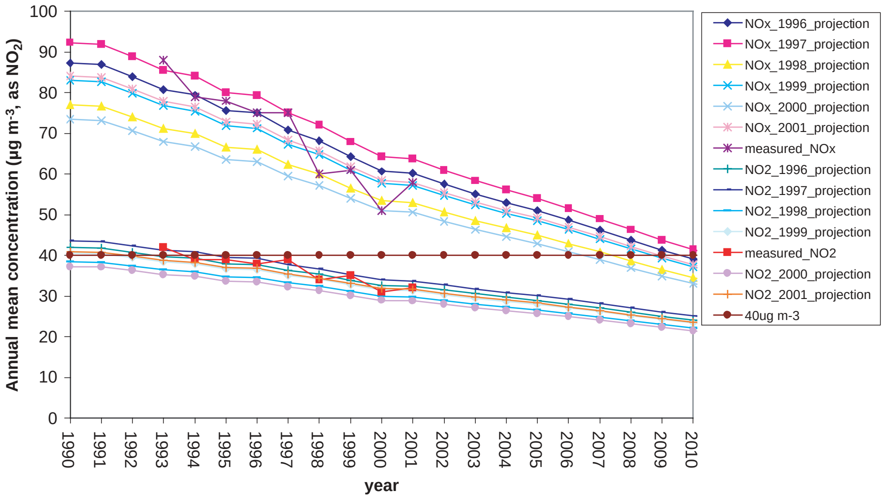


Figure 7.4 Site specific projections of NO_x and NO_2 concentrations at Glasgow Kerbside ($\mu g\ m^{-3}$, as NO_2).

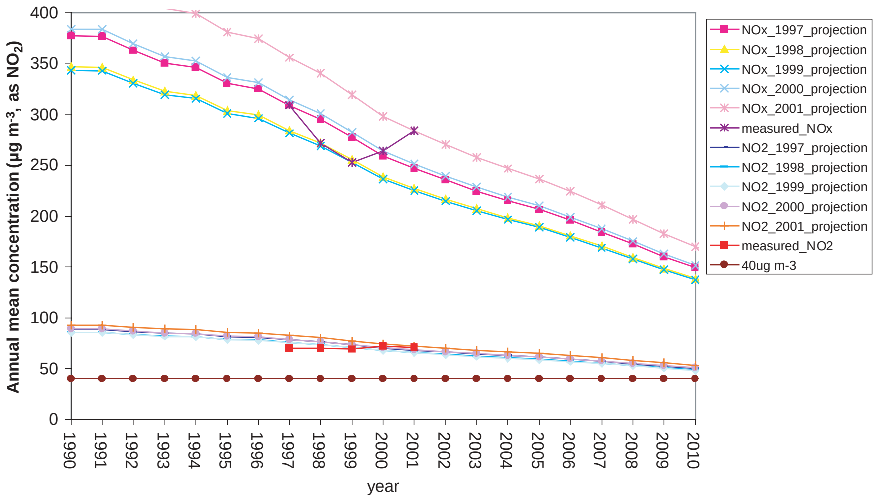


Table 7.1 Site-specific and year factor projections of annual mean NO_x and NO_2 concentrations from 2001 base year (µg m^{-3}, as NO_2). The range of the concentrations predicted by these two methods is listed. Values greater than 40 µg m^{-3} are in bold.

		Measured 2001		**Projections 2005**		**Projections 2010**	
Site	Type	NO_x	NO_2	NO_x	NO_2	NO_x	NO_2
Marylebone Road	Kerbside	335	**84**	278 – 282	**75 – 80**	195 – 201	**62 – 66**
Glasgow Kerbside	Kerbside	284	**71**	236 – 237	**63 – 65**	170	**52 – 53**
Haringey Roadside	Roadside	120	**48**	100 – 102	**43**	72 – 80	35 – 36
Redbridge 2	Roadside	355	**120**	288 –294	**66 – 107**	204 – 213	**55 – 88**
M25 Staines	Kerbside	196	**40**	157 –163	36 – **54**	117 – 119	29 – **45**
Bury Roadside	Roadside	270	**69**	219- 224	**61 – 62**	151 – 162	**50 – 51**
Lullington Heath	Rural	15	12.6	13	10 – 11	10 – 11	8 – 10
London Bridge Place (1999)	Urban background	105	**63**	83 – 84	**48 – 54**	67	**41 – 46**
London Bloomsbury	Urban centre	109	**51**	95 – 98	**46 – 54**	76 – 81	40 – **47**
West London	Urban background	95	**52**	83 – 84	**41 – 47**	66 – 67	35 – 40
London Bexley	Urban background	68	36	59	32 – 33	47 – 48	28
Manchester Town Hall	Urban background	92	**47**	80	39 – **43**	64	34 – 37
Glasgow City Chambers	Urban background	107	**46**	93	**42 – 44**	73 – 74	36 - 37
Belfast Centre	Urban centre	58	32	49 – 50	28 – 29	38 – 40	24 – 25
Port Talbot	Urban background	38	22	31 – 33	20 – 21	24 – 26	17
Roadside > 40 µg m^{-3}			**5**		**5 – 6**		**4 – 5**
Background > 40 µg m^{-3}			**5**		**4 – 5**		**1 – 2**
Total > 40 µg m^{-3}			**10**		**10**		**5 – 7**

740. The NO_x projections calculated using the two methods are very similar. The projected NO_2 concentrations are at times quite different because NO_x projections are used to derive projections of NO_2 concentrations using non-linear NO_x/NO_2 relationships within the site-specific model. NO_2 concentrations are projected directly from measured NO_2 concentrations in the year factor approach. The largest difference in predicted NO_2 concentrations are at those sites such as Redbridge 2 and M25 Staines, where the current measured concentrations are not consistent with the empirically derived relationships between NO_x and NO_2 concentrations (see Chapters 5 and 6).

741. Measured annual mean NO_2 concentrations in 2001 were greater than 40 µg m^{-3} at 10 out of 15 of the sites select for detailed analysis. Projected exceedences remained at 10 of the sites in 2005 for both the site-specific and year factor projections. This is reduced to between 5 and 7 sites in 2010 for the two different approaches.

742. The year factor approach has also been applied to calculate projected annual mean NO_2 concentrations in 2005 and 2010 from measured concentrations in 2001 at all of the sites for which data has been collated for this report. The results of this analysis are summarised in Table 7.2. The reduction in the number of sites with concentrations in excess of 40 µg m^{-3} is broadly in line with the mapped results of the national empirical models presented later in this chapter. This is as expected since the two analyses are based on the same emission inventory projections and similar models. The projected concentrations for each site are presented in Appendix 3. The number of sites with annual mean NO_2 concentration exceeding 40 µg m^{-3} shows little decline between 2001 and 2005 in London but a much greater reduction in the rest of the UK. Concentrations are projected to decline further between 2005 and 2010 with the number of sites with projected concentrations exceeding 40 µg m^{-3} also expected to decline in London by 2010.

Table 7.2 Status of NO_2 monitoring sites in 2001 and projections to 2005 and 2010 (R and K = Roadside and Kerbside sites, Other = all other classifications)

		Total number of sites	**No. sites annual average > 40 µg m^{-3} measured 2001 (%)**	**No. sites annual average > 40 µg m^{-3} projected 2005 (%)**	**No. sites annual average > 40 µg m^{-3} projected 2010 (%)**
Scotland	R and K	2	1 (50)	1 (50)	1 (50)
	Other	5	2 (40)	1 (20)	0
Wales	R and K	1	0	0	0
	Other	3	0	0	0
Northern Ireland	R and K	0	0	0	0
	Other	2	0	0	0
London	R and K	41	39 (95)	35 (85)	21 (51)
	Other	36	17 (47)	12 (33)	6 (17)
Rest of England	R and K	33	18 (55)	15 (45)	6 (18)
	Other	89	6 (7)	3 (3)	0
UK	R and K	77	58 (75)	51 (66)	28 (36)
	Other	135	25 (12)	16 (12)	6 (4)

7.2.2 Receptor point projections from dispersion modelling studies

743. Projected NO_x and NO_2 concentrations in 2005 and 2010 have been calculated for monitoring sites in London by CERC using ADMS-Urban from a 1999 base year and are shown in Table 7.3. These analyses and projections are based on the LAEI. The total number of sites with modelled annual mean NO_2 concentrations greater than 40 µg m^{-3} reduces from 21 out of 23 in 1999 (when measured concentrations were above 40 µg m^{-3} at 18 sites) to 17 in 2005 and 15 in 2010. Table 7.4 shows a comparison of predicted NO_x and NO_2 concentrations in 2010 at six sites in London for which predictions are available from both the netcen and CERC models. There is generally good agreement between the model results although there is some tendency for the CERC results to be slightly higher, for example for NO_2 at Haringey Roadside.

Table 7.3 Annual average NO_x and NO_2 concentration predicted by dispersion modelling ($\mu g\ m^{-3}$, as NO_2). Base year is 1999. Values greater than 40 $\mu g\ m^{-3}$ are in bold.

	Measured 1999		Modelled 1999		Modelled 2005		Modelled 2010	
	NO_x	NO_2	NO_x	NO_2	NO_x	NO_2	NO_x	NO_2
Roadside monitoring sites								
A3	256	**58**	227	**67**	138	**55**	105	**48**
Camden	210	**66**	204	**71**	143	**61**	117	**55**
Cromwell Road	256	**93**	260	**76**	193	**69**	141	**59**
Haringey	136	**51**	115	**55**	84	**48**	71	**42**
Hounslow	191	**60**	132	**53**	88	**44**	69	**40**
Marylebone Road	390	**91**	386	**88**	290	**78**	220	**71**
Southwark Roadside	227	**75**	183	**67**	132	**59**	107	**52**
Sutton Roadside	117	**44**	76	**42**	57	34	46	29
Tower Hamlets	241	**70**	193	**71**	138	**61**	113	**55**
Background monitoring sites								
Bexley	69	37	78	**40**	61	34	52	31
Bloomsbury	136	**67**	120	**57**	96	**52**	78	**46**
Brent	67	37	76	**44**	57	38	50	32
Bridge Place	105	**63**	111	**53**	90	**48**	71	**42**
Eltham	65	36	86	**44**	67	36	55	32
Hackney	134	**60**	113	**55**	84	**48**	71	**42**
Hillingdon	166	**50**	206	**63**	132	**55**	101	**48**
Lewisham	139	**54**	117	**55**	86	**46**	71	**42**
North Kensington	82	**46**	99	**52**	76	**44**	65	**40**
Southwark Urban Centre	118	**56**	99	**50**	80	**44**	67	38
Sutton Suburban	65	35	67	38	52	32	42	27
Teddington	52	32	59	34	48	29	40	25
Wandsworth	141	**52**	128	**59**	94	**50**	76	**44**
West London	99	**55**	92	**50**	71	**42**	61	38
Roadside sites > 40 $\mu g\ m^{-3}$		9		9		8		8
Background sites > 40 $\mu g\ m^{-3}$		9		12		9		7
Total sites > 40 $\mu g\ m^{-3}$		18		21		17		15

Table 7.4 Comparison of projected annual mean NOx and NO_2 concentrations in 2010 for selected monitoring sites in London (µg m^{-3}, as NO_2).

	netcen		CERC	
	NOx	NO_2	NOx	NO_2
Marylebone Road	195 – 201	62 – 66	220	71
Haringey Roadside	72 – 79	35 – 36	71	42
London Bridge Place	67	41 – 46	71	42
London Bloomsbury	76 – 81	40 – 47	78	46
West London	66 – 67	35 – 40	61	38
London Bexley	47 – 48	28	52	31

744. Mapped projections derived from the ERG modelling are presented later in this chapter. Site-specific projections have also been made by ERG at fewer sites than listed in Table 7.3 and only for 1999 and 2005. Further details relating to these predictions can be found in Carruthers *et al* (2002). Predictions have been made London-wide for 1999, 2005 and 2010 and these are considered later in this chapter.

745. Overall the modelled base case predictions of annual mean NO_2 suggest that concentrations below 40 µg m^{-3} will not be achieved across the whole of the UK by 2010. Outside London the exceedences are likely to be confined to the roadsides of busy roads while many roadside locations and some background locations are expected to exceed in London.

7.2.3 Source apportionment of concentrations in 2005 and 2010

746. The netcen site-specific model works by projecting the contributions to annual mean NO_x from different source sectors into the future and then adding the contributions together to get the total annual mean NO_x concentration. The source apportionment of the measured annual mean NO_x concentrations at selected monitoring sites in 2001 was discussed in section 6.2.4. Figures 7.5 and 7.6 show how the contributions from individual emission sectors are estimated to vary between 1996 and 2020 at London Bloomsbury and Marylebone Road. It is clear that the contributions from the domestic, commercial and regional rural contributions are much more important in 2010, relative to emissions from road traffic sources, than in 2001 as the contributions from traffic sources decline and the contributions from some other sectors show small rises.

Figure 7.5 Source apportionment of annual mean NO_x concentrations at London Bloomsbury (2001 base year).

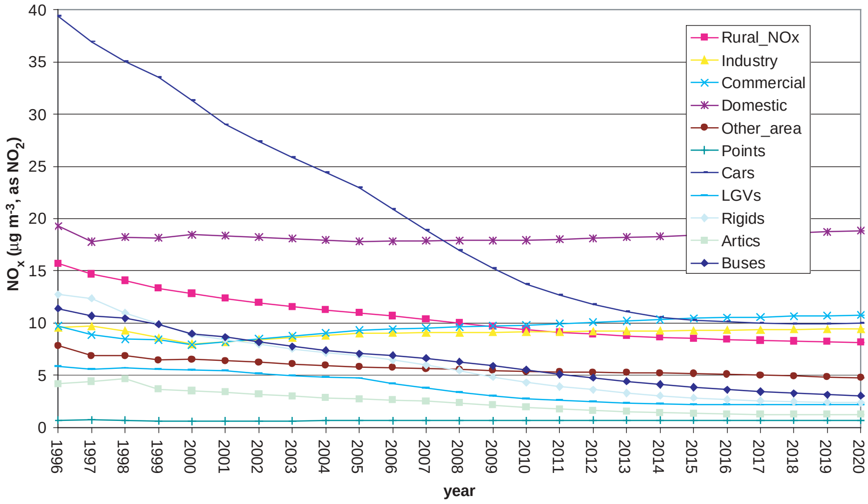
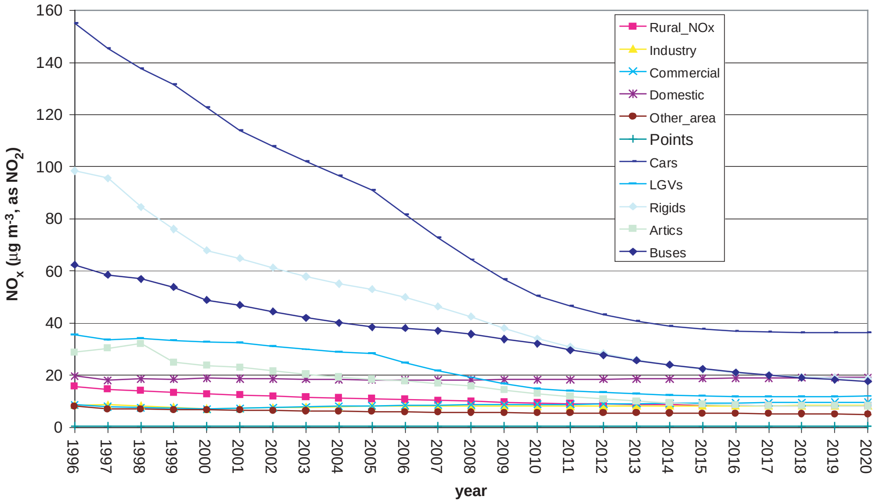


Figure 7.6 Source apportionment of annual mean NO_x concentrations at Marylebone Road (2001 base year).



747. The source apportionment of the projected annual mean NO_x concentrations in 2010 at the sites selected for detailed analysis is listed in Table 7.5 and can be compared with the analysis for 2001 in Table 6.9. Traffic sources are expected to continue to dominate at the roadside sites with contributions ranging from 57% at Haringey Roadside to 86% at Redbridge 2 (down from 70% and 92% in 2001). The percentage contribution from traffic sources is lower at background sites, ranging from 35% at London Bloomsbury to 59% at Belfast Centre (down from 50% and 71% in 2001). The percentage contribution from domestic and commercial emissions increases relative to 2001 to vary from 9% at Port Talbot to 16% at Belfast Centre, 32% at West London and 34% at London Bloomsbury. At London Bloomsbury, traffic sources and the total of domestic and commercial emissions are expected to each contribute roughly one third of the total annual mean NO_x in 2010. The final third of the total annual mean NO_x concentration includes 12% from regional rural concentrations, 11% from industry, 7% from other area sources and 1% from other point sources. The percentage contributions from domestic and commercial emissions at roadside sites are much lower at between 5% and 20% at the sites studied.

Table 7.5 Source apportionment of annual mean NO_x concentration in 2010 ($\mu g\ m^{-3}$, as NO_2), and percentages in brackets) derived from the netcen site-specific models.

Site	Total 2001	Rural	Industry	Commercial	Domestic	Other area	Points	Total traffic	Cars	LGV	HGVr	HGVa	Buses
Marylebone Road	195	9 (5)	8 (4)	9 (5)	18 (9)	6 (3)	1 (0)	145 (74)	50 (26)	15 (8)	34 (17)	13 (7)	32 (16)
Glasgow Kerbside	170	3 (2)	7 (4)	7 (4)	7 (4)	4 (2)	0 (0)	142 (84)					
Haringey Roadside	80	10 (12)	6 (8)	7 (8)	9 (11)	2 (2)	1 (1)	46 (57)	19 (24)	4 (5)	6 (8)	3 (4)	13 (16)
Redbridge 2	204	9 (5)	4 (2)	10 (5)	3 (2)	2 (1)	1 (0)	175 (86)					
M25 Staines	119	9 (8)	0 (0)	2 (2)	4 (3)	19 (16)	0 (0)	85 (71)					
Bury Roadside	151	9 (6)	3 (2)	3 (2)	5 (3)	3 (2)	2 (2)	41 (27)	41 (27)	8 (6)	16 (11)	57 (38)	3 (2)
London Bloomsbury	81	9 (12)	9 (11)	10 (12)	18 (22)	5 (7)	1 (1)	28 (35)	14 (17)	3 (3)	4 (5)	2 (2)	6 (7)
West London	67	9 (14)	5 (7)	5 (8)	16 (24)	4 (6)	1 (1)	27 (40)	14 (20)	3 (4)	4 (6)	2 (3)	5 (7)
London Bexley	48	9 (19)	3 (6)	3 (7)	5 (11)	5 (10)	2 (5)	20 (43)					
Manchester Town Hall	64	10 (15)	6 (9)	6 (9)	5 (8)	6 (9)	2 (3)	30 (47)	13 (20)	3 (4)	4 (6)	8 (12)	3 (5)
Glasgow City Chambers	73	3 (4)	8 (11)	9 (12)	6 (9)	4 (96)	0 (0)	42 (58)					
Belfast Centre	38	5 (12)	1 (3)	1 (3)	5 (13)	4 (10)	0 (0)	22 (59)					
Port Talbot	24	5 (20)	1 (4)	1 (4)	1 (5)	2 (7)	1 (4)	13 (55)					

748. The contributions from each vehicle type at each site can be further sub-divided by 'Euro class' (the new vehicle emission standards to which they belong, see section 2.6.5). The split of emissions between the different Euro classes has been assumed to be the same within each vehicle type across the whole of the UK because the same fleet composition information has been used everywhere. Figures 7.7-7.11 show the source apportionment of projected annual mean NO_x between Euro class for each vehicle type at London Bloomsbury. Curves for other sites would have exactly the same shape and relative contributions from different Euro classes but with a different vertical scale for each vehicle type. The contribution from Pre-Euro vehicles is relatively unimportant for all vehicle types after about 2005 and the contribution from Euro I is similarly very small by about 2010. Bus fleets have the slowest turnover and thus a relatively larger contribution in each year from vehicles meeting the older emissions standards. Figure 7.11 is, however, representative of the UK fleet of buses and it is expected that all London Buses will meet at least Euro II standards by 2005.

Figure 7.7 Source apportionment of annual mean NO_x concentrations at London Bloomsbury: Cars by Euro class (2001 base year, P = Petrol, D = Diesel) (μg m^{-3}, as NO_2).

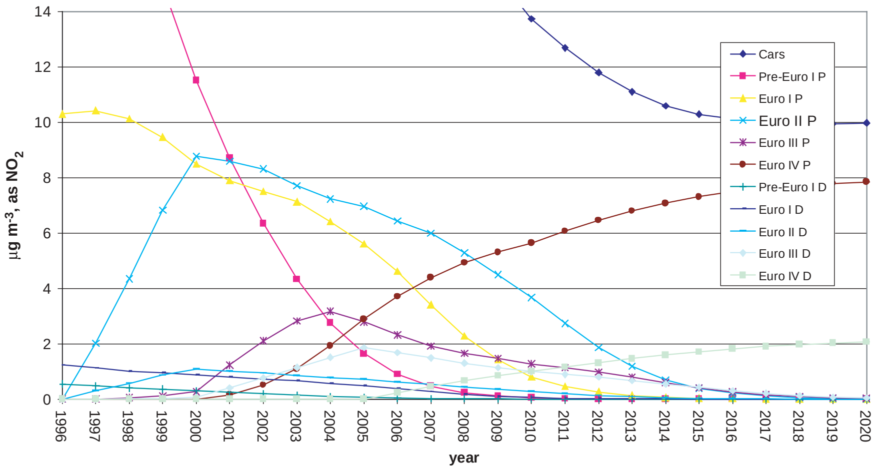


Figure 7.8 Source apportionment of annual mean NO_x concentrations at London Bloomsbury: Light Goods Vehicles by Euro class (2001 base year, P = Petrol, D = Diesel) (μg m^{-3}, as NO_2).

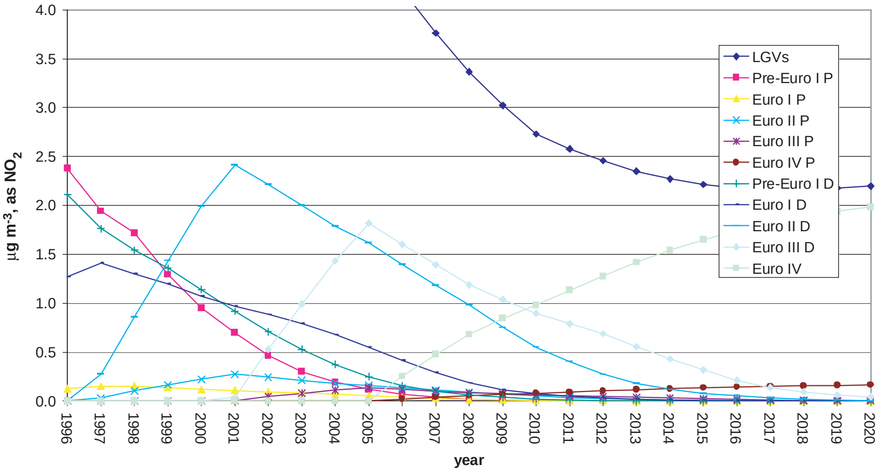


Figure 7.9 Source apportionment of annual mean NO_x concentrations at London Bloomsbury: Rigid Heavy Goods Vehicles by Euro class (2001 base year) (μg m^{-3}, as NO_2).

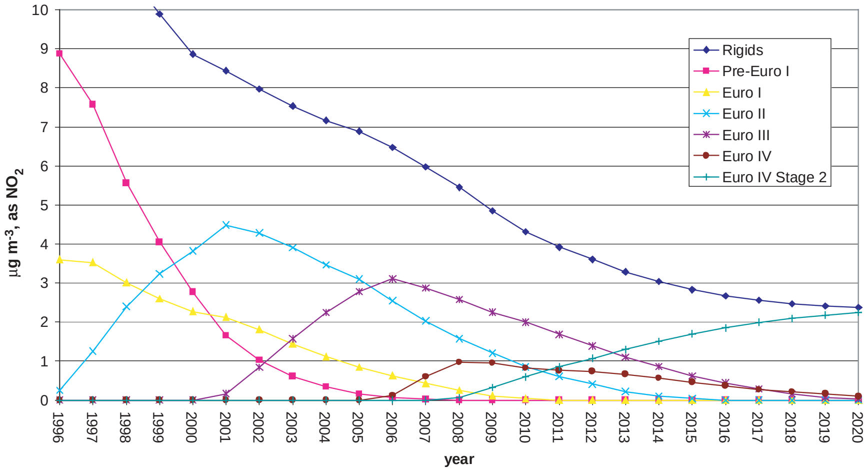


Figure 7.10 Source apportionment of annual mean NO_x concentrations at London Bloomsbury: Articulated Heavy Goods Vehicles by Euro class (2001 base year) (µg m^{-3}, as NO_2).

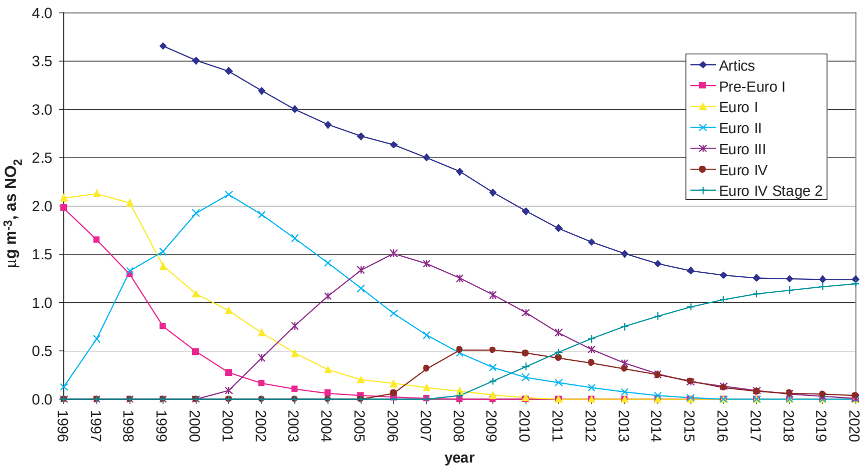
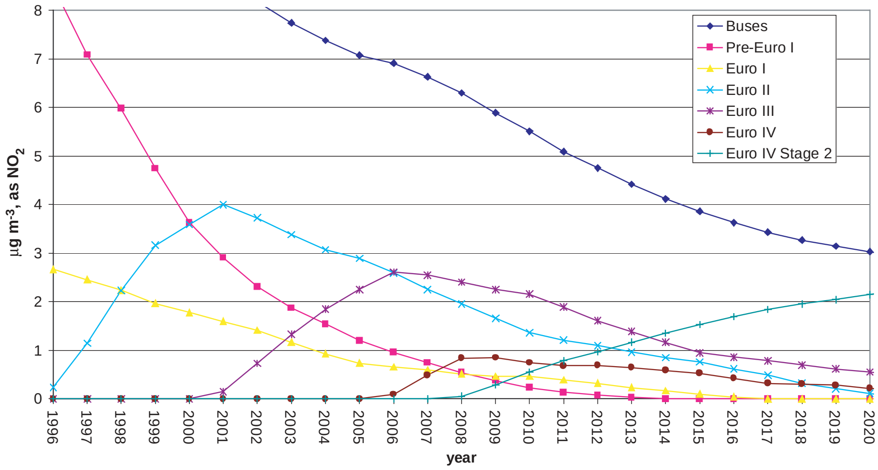


Figure 7.11 Source apportionment of annual mean NO_x concentrations at London Bloomsbury: Buses by Euro class (2001 base year) (µg m^{-3}, as NO_2).



7.2.4 Estimates of emission reductions required to reduce annual mean NO_2 concentrations to 40 µg m^{-3}

749. The netcen site-specific model has also been used to estimate the additional reductions in emissions, beyond those in the base case emission projections, required to reduce annual mean NO_2 concentrations to 40 µg m^{-3}. The results of this analysis are listed in Table 7.6 for both the reductions in traffic emissions required if the emissions from all other sectors remain at base case values and for reductions in emissions from all sectors. The required reductions in 2005 range from a reduction of 90% of base case traffic emissions at Marylebone Road (of 65% of base case emissions from all sectors) to 5% (3% for all sectors) at West London. No reductions beyond those assumed in the base case are required at Manchester Town Hall and Belfast Centre. The reductions relative to the base case are lower in 2010 due to the reduction in base case emissions from 2005. A reduction of 87% of base case traffic emissions (of 53% of all sector emissions) would be required at Marylebone Road to reduce annual mean NO_2 to 40 µg m^{-3}.

Table 7.6 Percentage emission reductions required to reduce annual mean NO_2 concentrations to 40 µg m^{-3} in 2005 and 2010 at selected monitoring sites. Relative to the base case emission projections, calculated using the netcen site-specific model.

	2005		2010	
Reductions applied to	Traffic emissions	All emissions	Traffic emissions	All emissions
Marylebone Road	90	65	83	53
Glasgow Kerbside	63	52	45	35
Haringey Roadside	15	10	0	0
Bury Roadside	57	48	37	29
London Bloomsbury	70	35	53	21
West London	5	3	0	0
Manchester Town Hall	0	0	0	0
Belfast Centre	0	0	0	0

7.2.5 Projections of hourly mean concentrations

750. The CERC dispersion modelling study also provides predictions of hourly mean NO_2 concentration in 2005 and 2010 from a base year of 1999. The 99.8th percentiles of modelled hourly mean concentrations in 2005 and 2010 have been calculated for the 23 national network sites in London listed in Table 7.3. A 99.8th percentile of 200 µg m^{-3} is equivalent to no more than 18 hours with concentrations greater than 200 µg m^{-3} for a full year of data. The model over-predicts 99.8th percentile concentrations in 1999 with 13 sites over 200 µg m^{-3} in comparison with the one measured exceedence of 200 µg m^{-3} (at Marylebone Road). The poor agreement between modelled and measured exceedences is as expected, since it is considerably more difficult to correctly predict concentrations for a single hour than for an annual mean (see Chapter 5). The 99.8th percentile concentrations are predicted to decline by an average of 18% from the 1999 level by 2005 at roadside and 16% at background sites and by 25% and 22% by 2010. Peak hourly NO_2 concentrations are, however, highly variable from year to year and dependent on the occurrence of particular summer or winter episode weather conditions, as discussed in section 6.5. Measured 99.8th percentile NO_2 concentrations were above 200 µg m^{-3} at 5 out of a total of 97 national network monitoring sites during 2001.

7.3 Mapped projections of NO_2 concentrations in 2005 and 2010

7.3.1 Introduction

751. Modelled projections of NO_2 concentrations are subject to considerable uncertainty. This uncertainty can arise from the treatment of dispersion and chemical processes within the models, the meteorological data used, the emissions inventories and projections included and the boundary conditions for the models, such as the regional concentrations of pollutants. A full analysis of the uncertainty associated with the modelled projections from a particular modelling study has not been attempted for this report. Such an analysis would not be practicable, in any case, given current understanding of the factors that influence ambient NO_2 concentrations and the fact that emission projections are necessarily influenced by policy, economic and political factors that are beyond the scope of this report. An indication of the uncertainty associated with the modelled predictions can, however, be provided by an examination of the range of the results obtained from the different studies.

7.3.2 Mapped results of national empirical models

752. Maps of annual mean NO_2 concentrations in 2005 and 2010 at both background and roadside locations have been calculated by netcen for the UK using the empirical modelling methods described in section 5.2.1 and are shown in Figures 7.12 to 7.15. These maps can be compared with the maps for 2001 presented in section 6.4.2. These projections have been derived using the empirical calibration coefficients derived for 2001 and NAEI projections of emissions for 2005 and 2010. These maps have therefore effectively been calculated using '2001 meteorology'. The modelling results in terms of exceedence of 40 µg m^{-3} as an annual mean are summarised in Table 7.7 and can be compared with the analysis for 2001 presented in Chapter 6. The modelled extent of exceedence is broadly consistent with the projection for individual monitoring site locations presented in Table 7.2 above. The greatest decline in the length of major road exceeding 40 µg m^{-3} is between 2005 and 2010. 65% of the length of major roads in London are projected to remain above this concentration in 2005, reducing to 23% by 2010. Background concentrations in central London are projected to remain above 40 µg m^{-3} for a small number of 1 km squares. The number of exceedences at roadside locations outside London is projected to be much reduced by 2010 relative to 2001. There are projected to be no roadside exceedences in Wales or Northern Ireland by 2010.

Table 7.7 Summary of national empirical model results for annual mean NO_2 concentrations in 2005 and 2010 (areas defined by 1 x 1 km background estimates, roadside concentrations calculated for built-up major road links).

				2005			2010		
	Area (km^2)	Number of links	Length of links (km)	Area > 40 µg m^{-3} (km^2)	Number of road links > 40 µg m^{-3}	Length > 40 µg m^{-3}	Area > 40 µg m^{-3} (km^2)	Number of road links > 40 µg m^{-3}	Length > 40 µg m^{-3}
Scotland	77769	435	1310.8 (0%)	0 (16%)	70 (8%)	108 (0%)	0 (3%)	11 (2%)	22
Wales	20694	572	1022.0 (0%)	2 (2%)	11 (2%)	21 (0%)	1 (0%)	0 (0%)	0
N Ireland	13627	124	950.2 (0%)	0 (3%)	4 (3%)	26 (0%)	0 (1%)	1 (0%)	4
London	1591	1888	1767 (5%)	78 (75%)	1407 (65%)	1152 (3%)	48 (35%)	670 (23%)	410.6
Rest of England	128519	6069	10922 (0%)	66 (25%)	1538 (18%)	1941 (0%)	33 (7%)	405 (5%)	5441
UK	242200	9088	15972 (0%)	146 (20%)	3030 (33%)	3250 (0%)	82 (12%)	1087 (6%)	981

753. Figure 7.16 shows the frequency distributions of projected concentrations at the roadside of major roads in 2001, 2005 and 2010 in the UK and in London. The mode of the UK frequency distribution is above 40 µg m^{-3} in 2001 but below in 2005 and well below in 2010. The mode for roads in London is rather higher and is still just above 40 µg m^{-3} in 2005. This analysis indicates that small changes in modelled concentrations would have a large impact on the estimated percentage of road length expected to exceed 40 µg m^{-3} in 2001 and 2005. The percentage of road length projected to exceed 40 µg m^{-3} in 2010 will be less sensitive to small changes in estimated roadside concentrations, particularly outside London, where the expected percentage exceedence is small.

Figure 7.12 Estimated annual mean background NO_2 concentration, 2005 (µg m^{-3}).

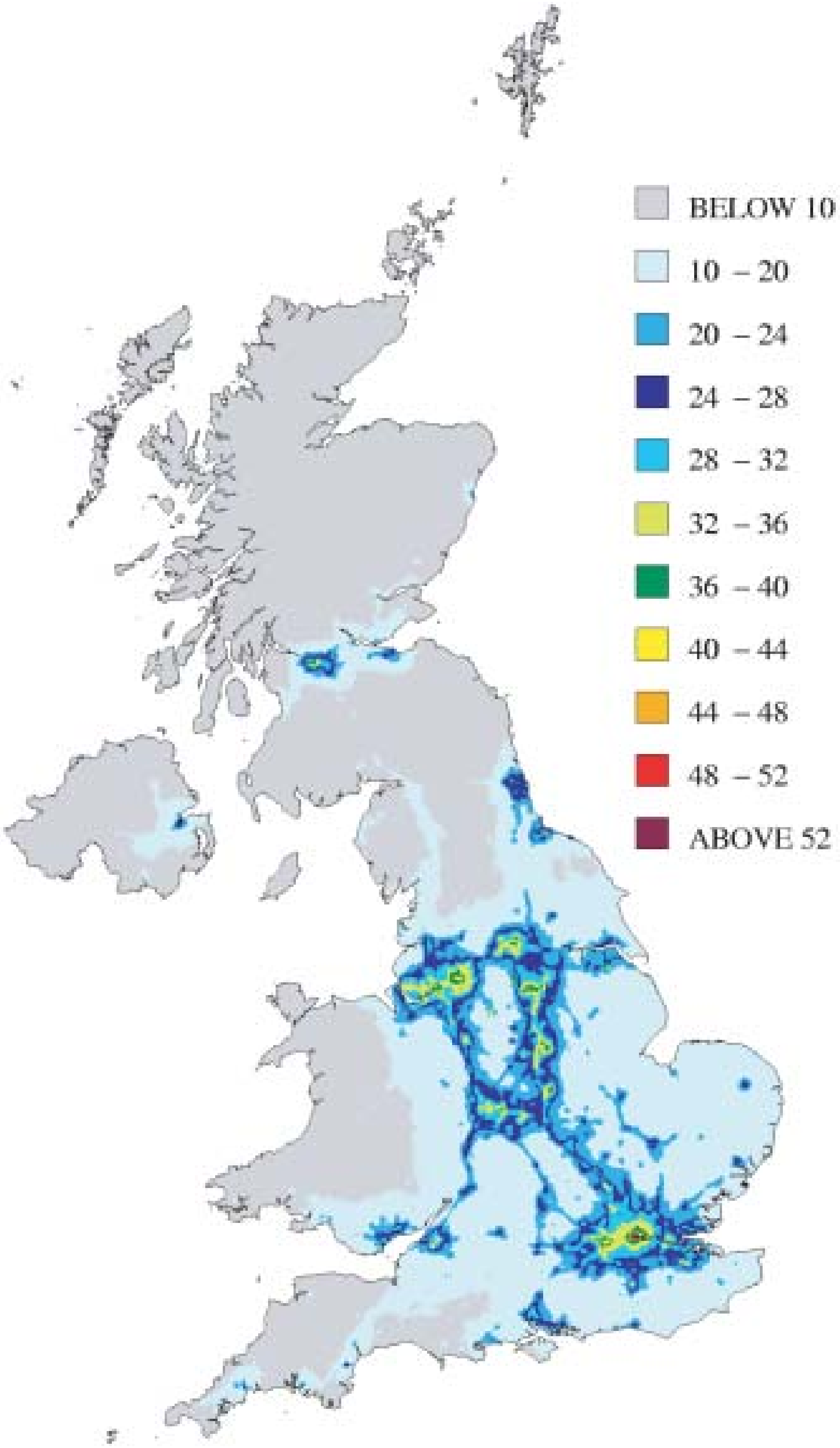


Figure 7.13 Estimated annual mean background NO_2 concentration, 2010 (µg m^{-3}).

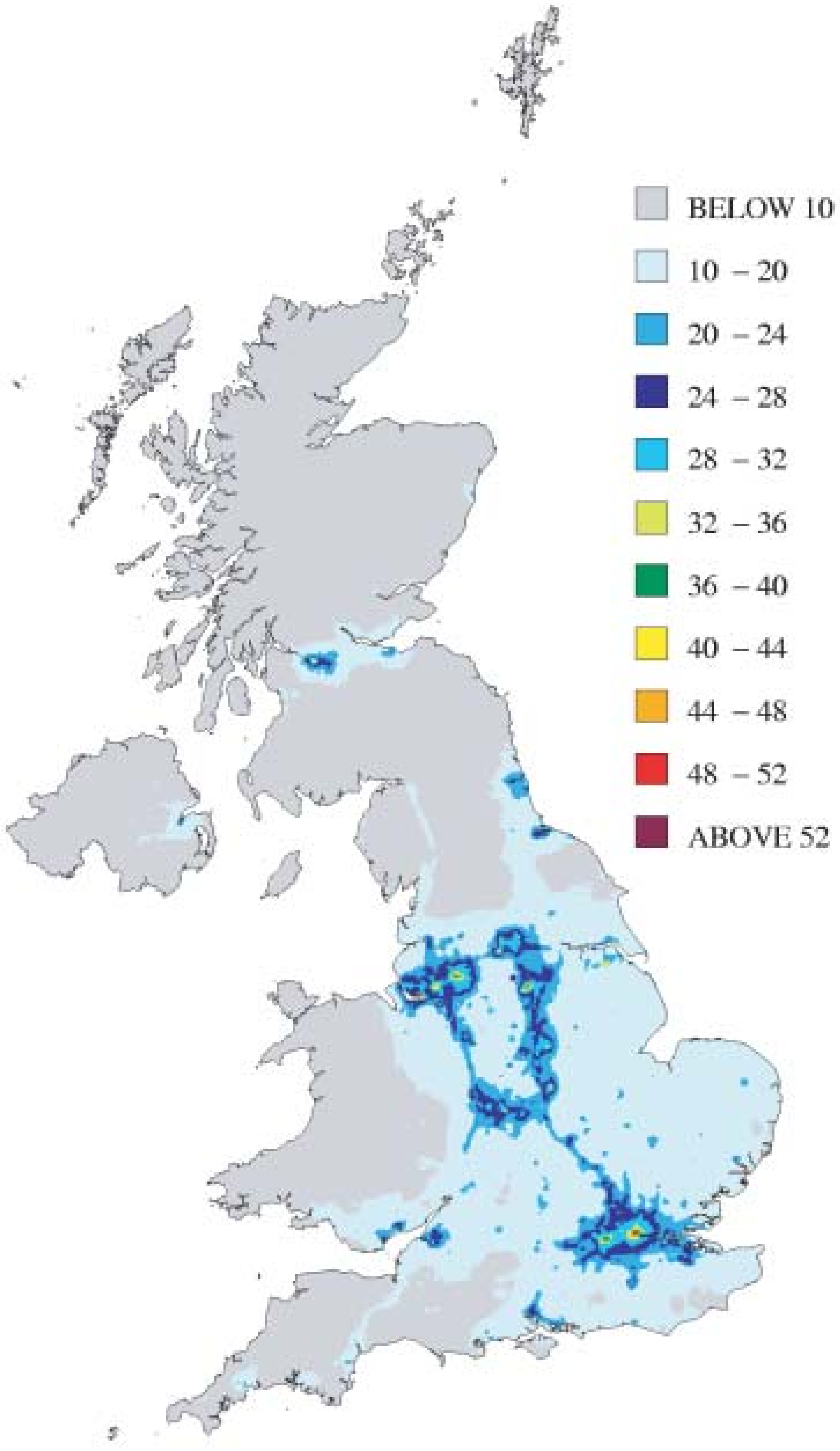


Figure 7.14 Estimated annual mean roadside NO_2 concentration, 2005 (µg m^{-3}).

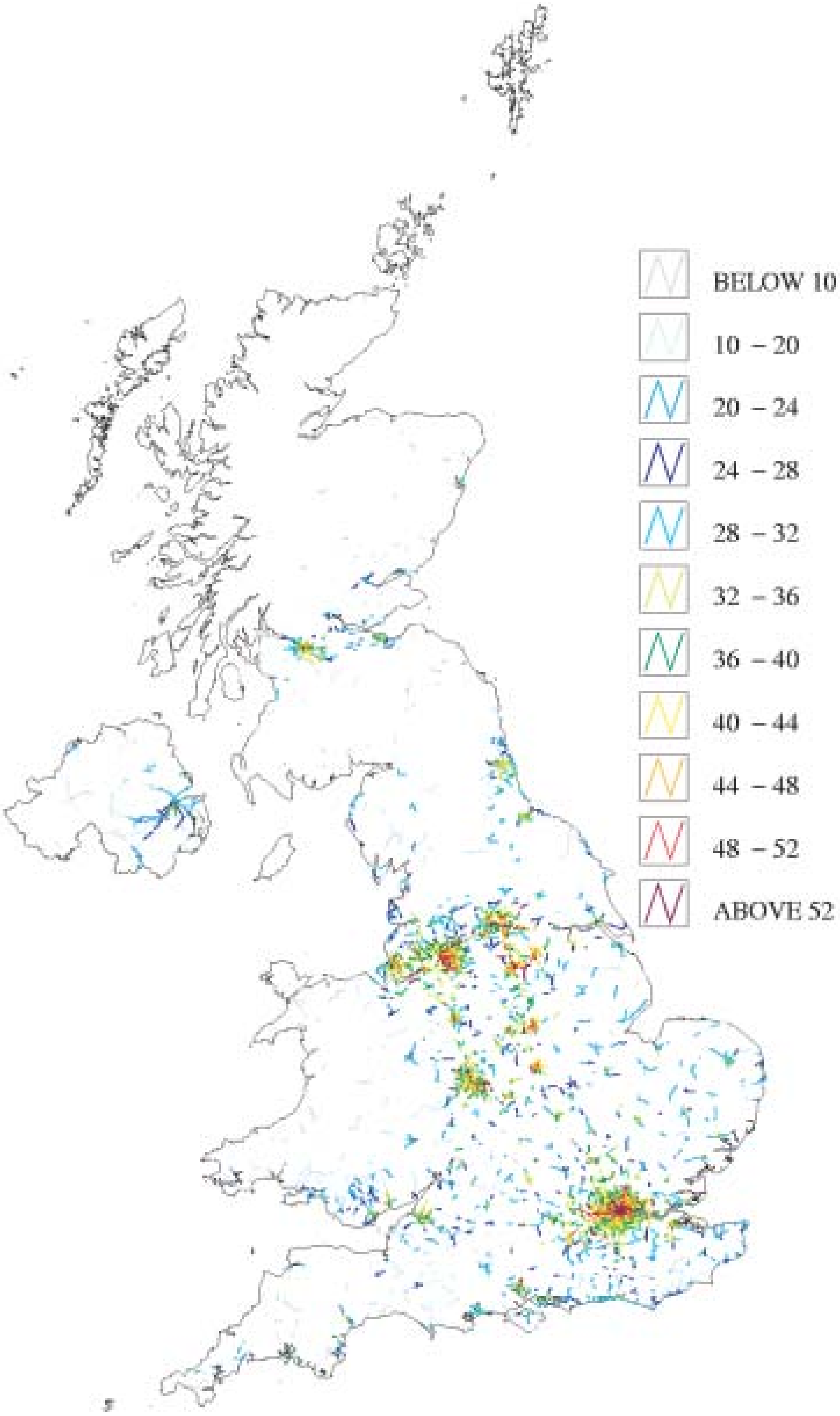


Figure 7.15 Estimated annual mean roadside NO_2 concentration, 2010 ($\mu g\ m^{-3}$).

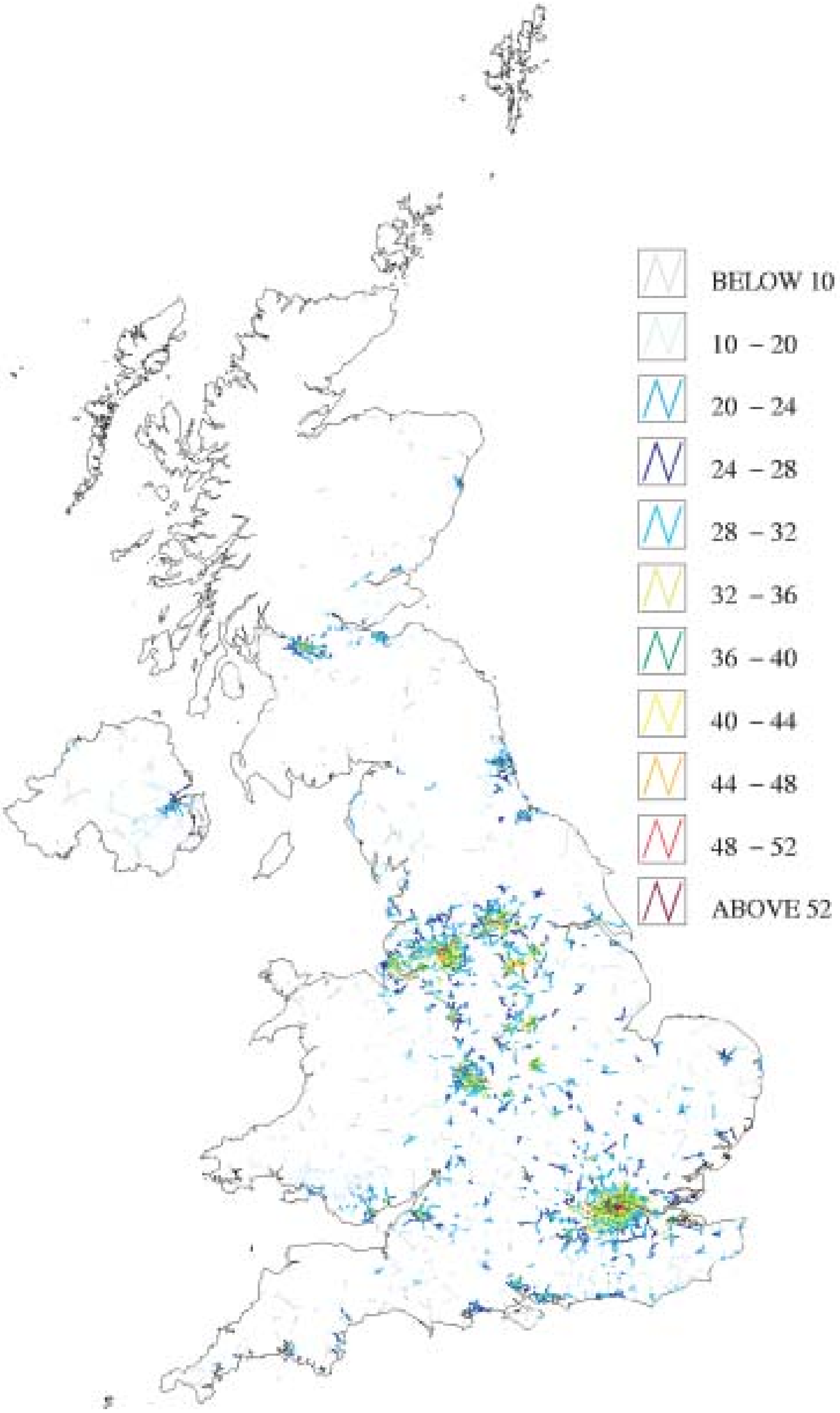
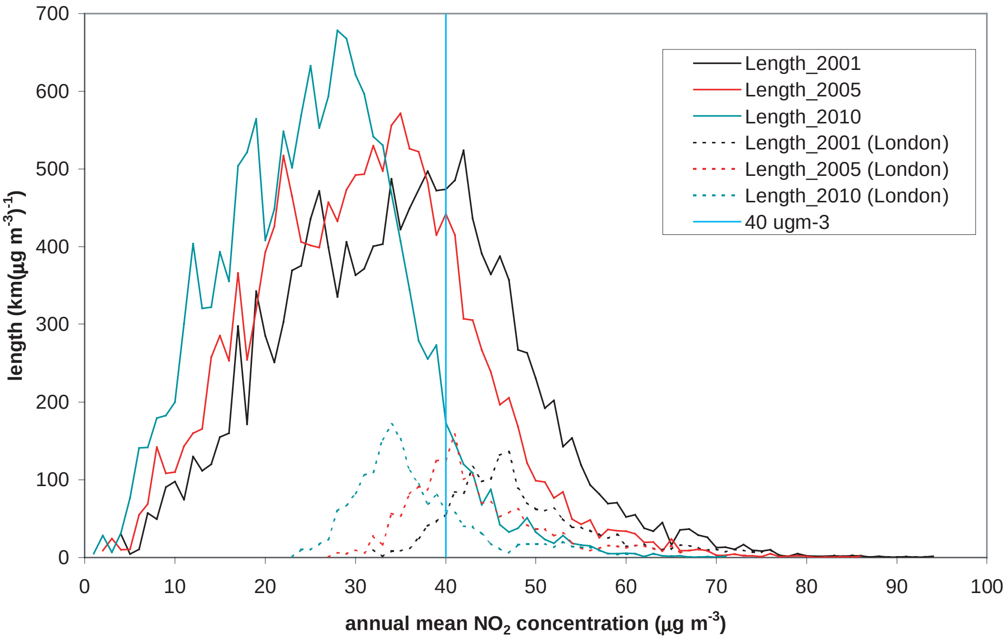


Figure 7.16 Frequency distribution of modelled roadside annual mean NO_2 concentrations from the netcen model.



7.3.3 Detailed modelling results for London

754. Projections of annual mean NO_2 concentrations in London for 2005 and 2010 have been calculated by ERG and by CERC from a 1999 base year using the methods described in the Chapter 5 and LAEI emissions projections. Maps for 1999 and 2001 were presented in the section 5.4.3. The maps for 2005 and 2010 are presented here along with frequency distributions of results and comparisons with the national scale analysis and the modelled concentrations for 2001. Projections of background concentrations from the LRCTM for 2005 and 2010 are also presented and discussed.

755. Figures 7.17 and 7.18 show the ERG modelling results for the London Area. Table 7.8 lists the calculated area of London exceeding the 40 μg m^{-3} for 1999, 2001, 2005 and 2010. These results are somewhat higher than those predicted in the national modelling. The most significant difference is that the ERG calculations relate to the total surface area exceeding 40 μg m^{-3} whereas the national modelling relates only to background areas exceeding this value.

Figure 7.17 Annual mean NO_2 in London for 2005 using ERG model (μg m^{-3}).

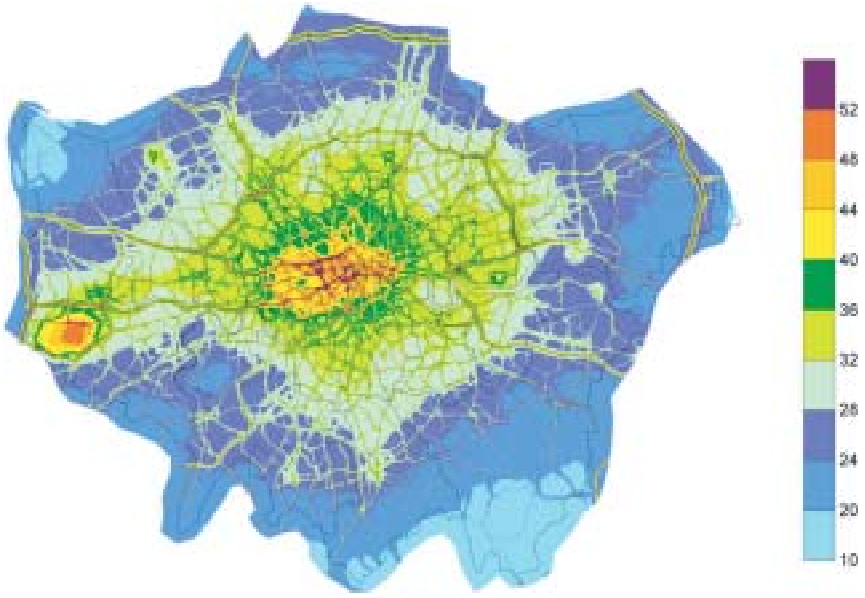


Figure 7.18 Annual mean NO_2 in London for 2010 using ERG model (µg m^{-3}).

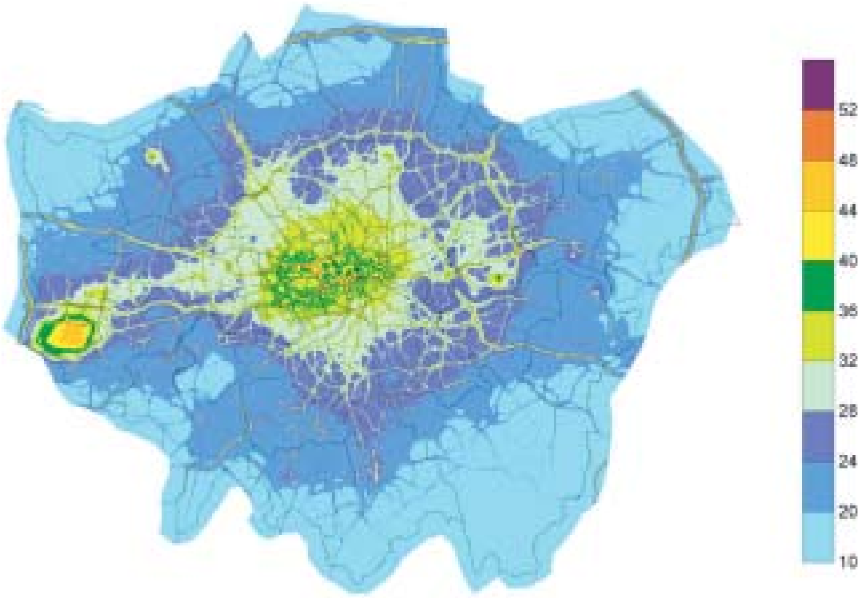
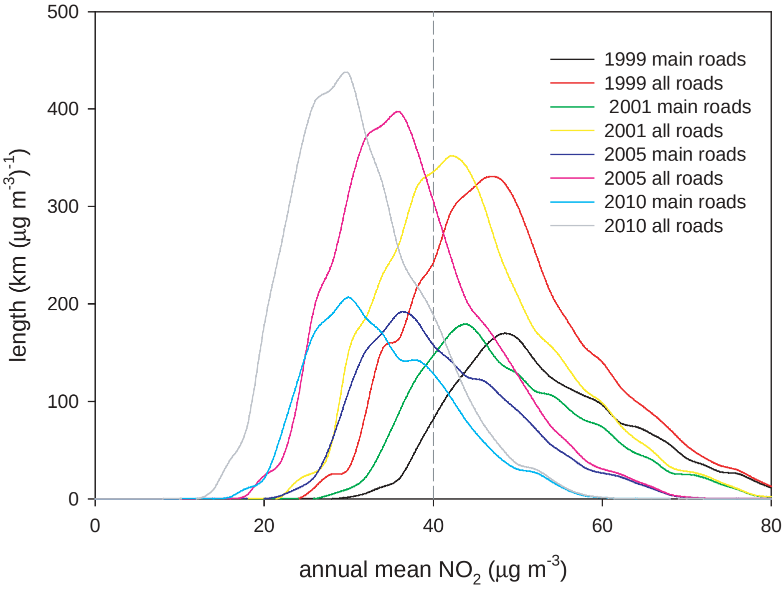


Table 7.8 Area and length of road exceeding an annual mean NO_2 concentration of 40 µg m^{-3} in London using the ERG approach.

	km^2 > 40 µg m^{-3}	Length of major road > 40 µg m^{-3} (km and percent)	Length of minor roads > 40 µg m^{-3} (km and percent)	Length of all LAEI roads > 40 µg m^{-3} (km and percent)
Total area/length modelled	1854	1937	2076	4013
1999	627 (34%)	1880 (95%)	1443 (70%)	3323 (83%)
2001	361 (19%)	1627 (84%)	1134 (55%)	2761 (69%)
2005	134 (7%)	1027 (53%)	553 (27%)	1580 (39%)
2010	36 (2%)	500 (26%)	173 (8%)	673 (17%)

756. Estimates of the length of major road (A-roads and motorways) exceeding an annual mean NO_2 concentration of 40 µg m^{-3} have also been made using the ERG model for London. The length of road statistic is somewhat arbitrary because the statistic depends on the distance from the road at which the statistic is calculated. Calculations have been made at a distance of 10 m from the road centreline based on receptor points placed at 100 m intervals along each side of the major roads in the London area. A total major road length of 1937 km has been considered. In 1999 it is estimated that 95% (1880 km) of the road length exceeds 40 µg m^{-3} (84%, 1627 in 2001). By 2005 this decreases to 53% (1027 km, compared with 1152 km from the national model), decreasing to 26% (500 km compared with 410 km from the national model) in 2010. Figure 7.19 shows the frequency distributions for each of the years and highlights 40 µg m^{-3}.

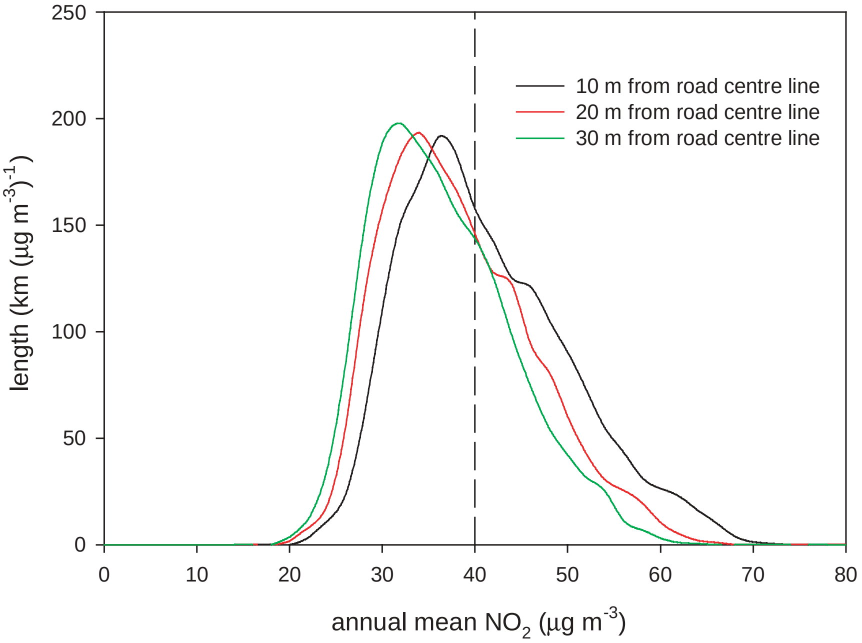
Figure 7.19 Frequency distribution of annual mean NO_2 calculated at 10 m from the road centreline for major and all LAEI roads in London using the ERG methodology.



757. The LAEI considers approximately 4000 smaller road links than the NAEI and it is therefore possible to calculate exceedence statistics for all roads in the LAEI. The total road length in London considered by the LAEI is 4013 km. Figure 7.19 shows the frequency distributions for all roads in the LAEI in addition to major roads only. The mode of each distribution remains similar in each case but the proportion of roads exceeding 40 µg m^{-3} is, as expected, less. In 2005 39% of LAEI roads are predicted to exceed at 10 m, decreasing to 17% in 2010.

758. The ERG estimates of the extent of roadside exceedence depend on the assumptions concerning the distance from the road centreline. Estimates have been made for 2005 at three distances: 10, 20 and 30 m. Figure 7.20 shows the effect of these assumptions. The results are very sensitive to the distance from the road assumed because of the fall-off in concentrations away from roads. At 10 m, 53% of the road length exceeds 40 µg m^{-3}, but this falls to 42% and 34% for 20 m and 30 m from the road centreline respectively. It is likely that the national empirical modelling will typically relate to the 10 m distance from the road centreline on this basis. However, because the national modelling is based on data from roadside and kerbside measurements, which are varying distances from actual road centrelines, there will be some uncertainty in the actual distance.

Figure 7.20 Frequency distribution of annual mean NO_2 calculated at 10, 20 and 30 m from the road centreline for major roads in London using the ERG methodology for 2005.



759. Figures 7.21 and 7.22 show the CERC modelling results for the Greater London Area for 2005 and 2010. The area exceeding 40 µg m^{-3} is calculated to decrease from 68% in 1999 to 32% in 2005 and 13% in 2010. These results are higher than those predicted in the ERG and netcen modelling. Figure 7.23 shows frequency distributions for the percentage of the total area of Greater London derived from the CERC model results. A frequency distribution for roadside concentrations is shown in Figure 7.24 all roads in LAEI and in Figure 7.25 for major roads only. The mode of the all LAEI roadside concentrations is at approximately 40 µg m^{-3} in 2005. The CERC modelling results in terms of the area and length of road exceeding 40 µg m^{-3} are listed in Table 7.9.

Table 7.9 Area and length of road exceeding an annual mean NO_2 concentration of 40 µg m^{-3} in London using the CERC approach.

	km^2 > 40 µg m^{-3}	**Length of major roads > 40 µg m^{-3} (km and percent)**	**Length of minor roads > 40 µg m^{-3} (km and percent)**	**Length of all LAEI roads > 40 µg m^{-3} (km and percent)**
Total area/length modelled	1574	1746	1910	3656
1999	1074 (68%)	1719 (98%)	1591 (83%)	3250 (89%)
2005	504 (32%)	1378 (79%)	893 (47%)	2271(62%)
2010	205 (13%)	857 (49%)	410 (21%)	1267 (35%)

Figure 7.21 Annual mean NO_2 in London for 2005 using CERC model (µg m^{-3}).

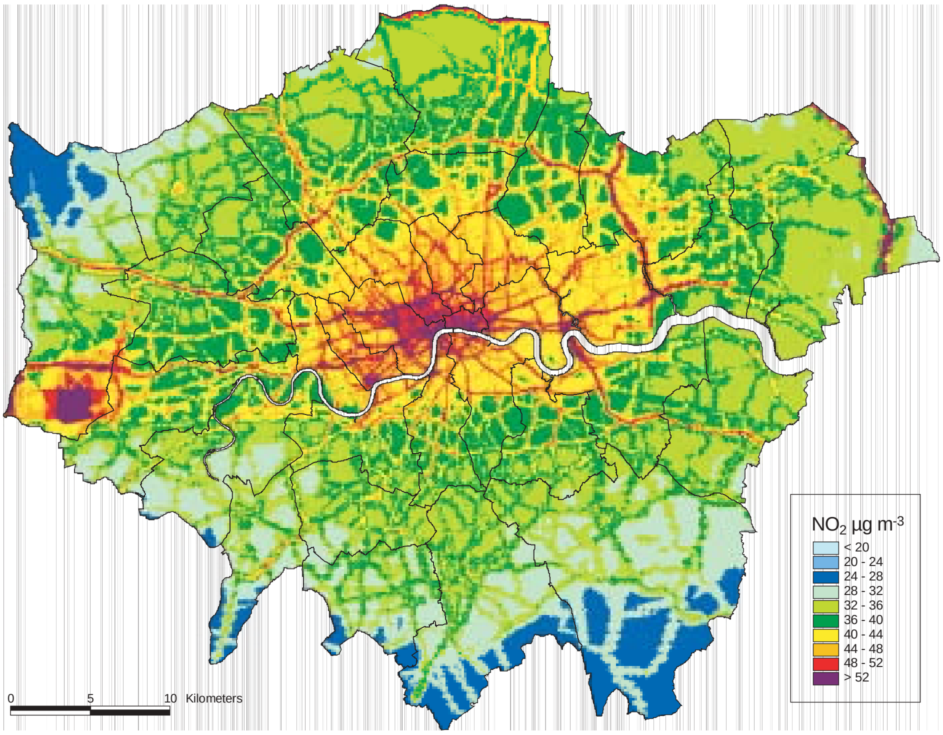


Figure 7.22 Annual mean NO_2 in London for 2010 using CERC model (µg m^{-3}).

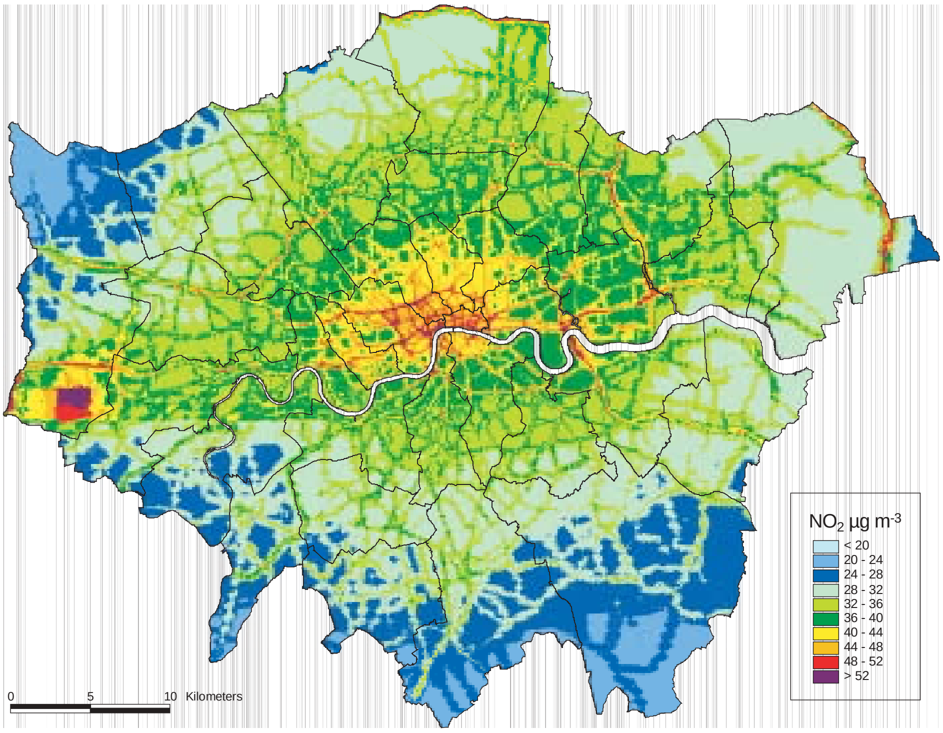


Figure 7.23 Frequency distribution of annual mean NO_2 concentration across the area of Greater London derived from CERC model results.

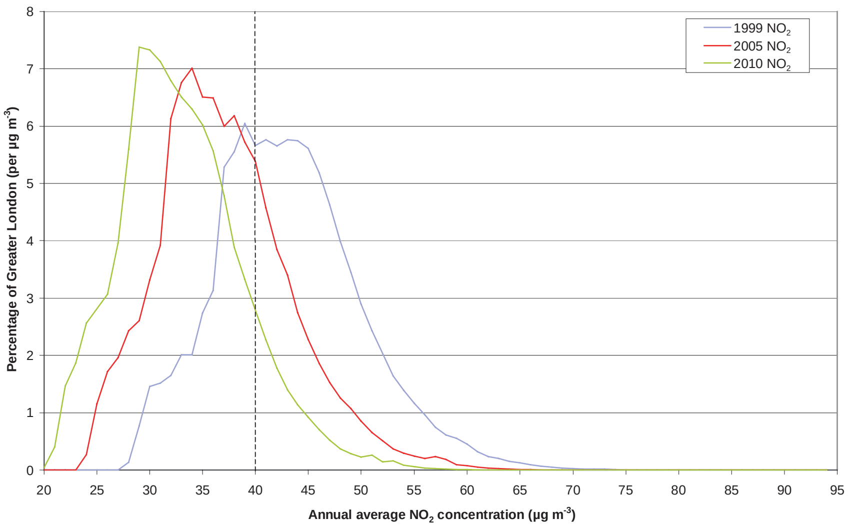


Figure 7.24 Frequency distribution of annual mean NO_2 concentration for all LAEI roads in London using the CERC model.

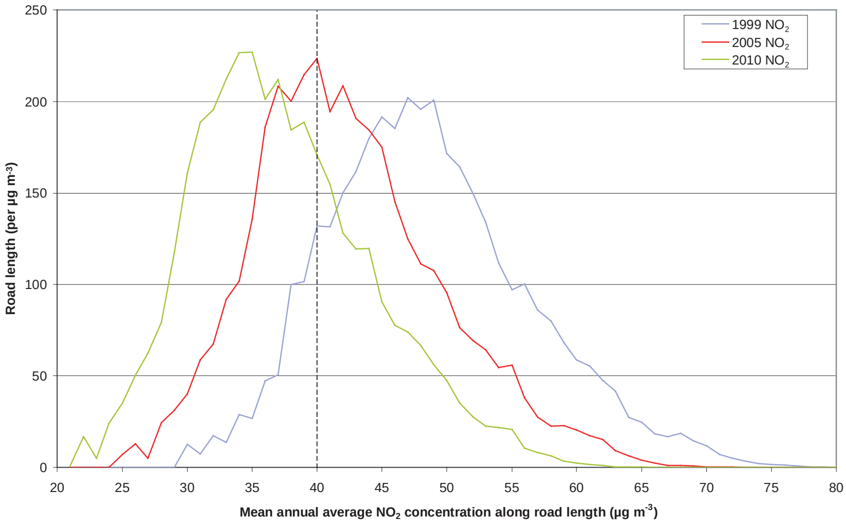
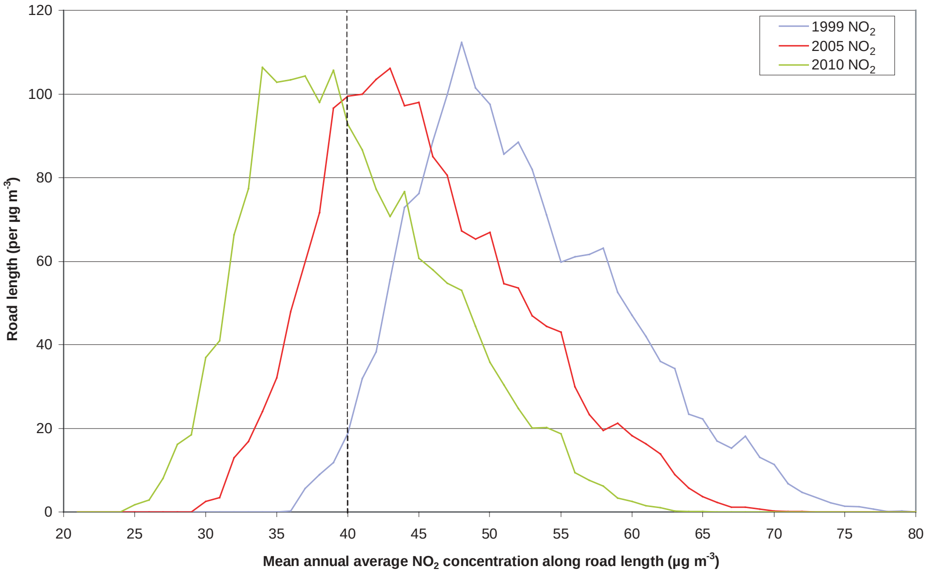


Figure 7.25 Frequency distribution of annual mean NO_2 concentration for major roads in London using the CERC model.



760. The LRCTM (see section 5.4) has also been used to predict background NO_2 concentrations in London. The LRCTM model has been used to test the assumptions inherent in projecting NO_2 concentrations into the future and to examine the impact of policy measures on NO_2 concentrations in London up to the years 2005 and 2010. The concern here has been about the extent to which the empirical models are able to address quantitatively the future NO_2 air quality situation because they necessarily have been built using contemporary NO_2 and NO_x air quality data.

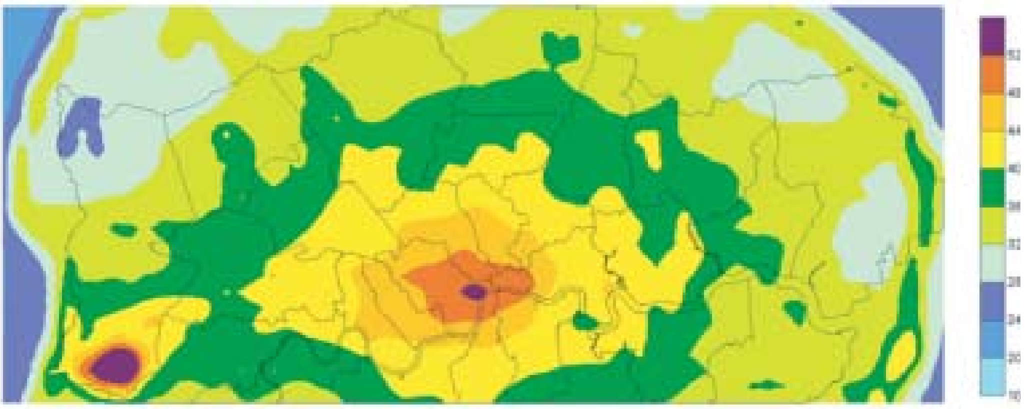
761. The spatial distributions of the annual mean NO_2 concentrations across London have been determined using the LAEI NO_x emission inventories for 2005 and 2010 for comparison with the map for 1999 presented in section 6.4.3. These distributions are plotted out in Figure 7.26 for a substantial area of the London conurbation covering over 1500 km_2 of north and central London and its outskirts. The mean annual mean NO_2 concentration for this selected area was found to be 39.5 µg m^{-3} with 1999 NO_x emissions, 35.8 µg m^{-3} with 2005 emissions and 33.7 µg m^{-3} with 2010 emissions. The reduction in annual mean NO_2 concentrations amounted to 9% for 2005 and 15% for 2010 whilst the reduction in annual mean NO_x concentrations were a factor of two higher at 21% and 31%, respectively. NO_x concentrations fell at a slightly lower rate compared with NO_x emissions because of the contribution to emissions from the south-east region generally and from long range transport from Europe. Both these terms had been held constant in the LRCTM between 1999 and 2010. Mean NO_2 concentrations fell at a significantly lower rate compared with NO_x concentrations because the NO_2 vs. NO_x split changes with NO_x concentration. As NO_x concentrations fall, the fraction of the NO_x that is present as NO_2 increases so that NO_2 concentrations always fall less quickly compared to NO_x concentrations and NO concentrations fall more quickly. This behaviour found in the LRCTM model results explains the origins of the non-linear relationship found between NO_2 and NO_x concentrations which is the basis of the empirical modelling approaches described Chapter 5.

762. The highest annual mean NO_2 concentrations in the LRCTM model results are found in the area between Heathrow Airport and the M25 as is shown in Figure 7.26. With 1999 NO_x emissions, the maximum annual mean NO_2 concentration was found to be 67.1 µg m^{-3} and this decreased by 7% to 62.2 µg m^{-3} with 2005 emissions and by 14% to 58.0 µg m^{-3} with 2010 emissions. Over central London, the maximum annual mean NO_2 concentrations was found to be 57.4 µg m^{-3} and this decreased by 8% to 52.7 µg m^{-3} with 2005 emissions and by 13% to 49.8 µg m^{-3} with 2010 emissions.

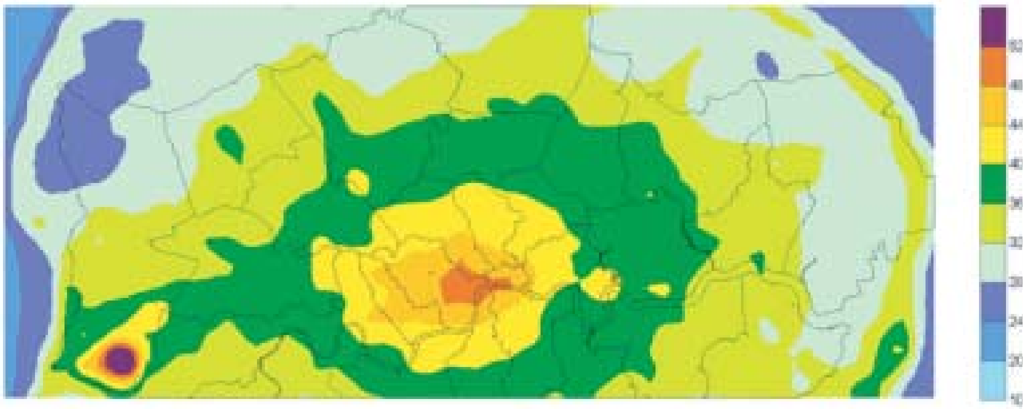
763. The area of exceedence of 40 µg m^{-3} shrinks dramatically from 44% of the area modelled in 1999 to 20% in 2005 and 10% in 2010. A significantly greater reduction in NO_x emissions would be required if exceedences are to be eliminated altogether in central London. In viewing Figure 7.26 account should be taken of the coarse resolution of the LRCTM model. This means that the plotted annual mean concentrations give a reasonable reflection only of likely annual mean NO_2 concentrations in urban background and suburban locations. The model is not able to describe the NO_2 concentrations likely to be experienced in roadside and kerbside locations in 2010. This consideration reinforces the conclusion that significantly greater reductions in NO_x emissions will be required compared to those currently contemplated, if exceedences of 40 µg m^{-3} are to be eliminated by 2010.

Figure 7.26 The spatial distributions of the annual mean NO_2 concentrations (µg m^{-3}) predicted with the LRCTM model with:

a) 2005 LAEI NO_x emissions



b) 2010 LAEI NO_x emissions



7.3.4 Comparison of the different model predictions for London

764. The areas modelled in each of the studies are different and thus the percentages of the total areas modelled exceeding 40 µg m^{-3} presented in the previous section are not directly comparable. Table 7.10, however, shows a comparison of the model results for London for concentrations in background locations for the three studies for which the model domains include the whole of Greater London. The area modelled using the LRCTM does not cover the whole of Greater London and Table 7.11 therefore provides a comparison for the area that is common to all four studies. Figure 7.27 shows the location of this common model domain in relation to that of the CERC, Met Office (LRCTM) and netcen studies. The ERG model domain is similar to that of CERC but extends slightly outside Greater London.

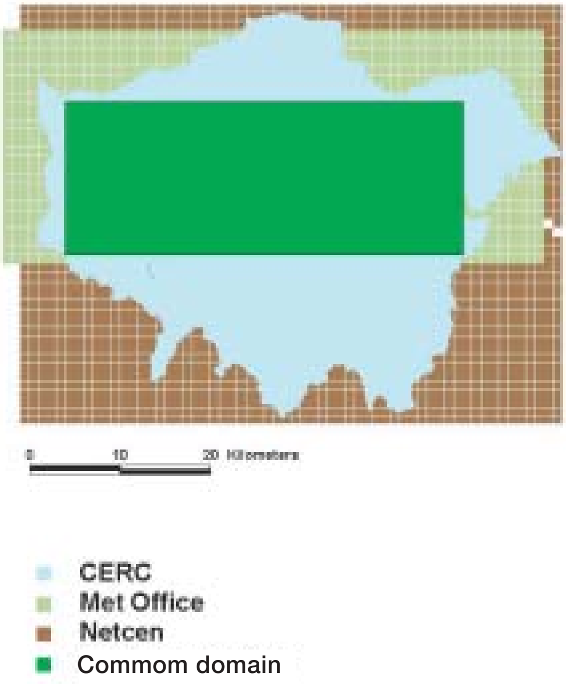
Table 7.10 Comparison of the percentage of the area of Greater London predicted to exceed an annual mean NO$_2$ concentration of 40 µg m^{-3} (total area modelled = 1574 km^2).

Model	Locations modelled	Base year	1999	2001	2005	2010
Netcen	background	2001	–	12	5	3
ERG	all	1999	38	21	10	3
CERC	all	1999	68	–	32	13

Table 7.11 Comparison of the percentage of the area of the common model domain predicted to exceed an annual mean NO$_2$ concentration of 40 µg m^{-3} (total area modelled = 748 km^2).

Model	Locations modelled	Base year	1999	2001	2005	2010
Netcen	background	2001	–	22	10	6
ERG	all	1999	65	39	18	5
CERC	all	1999	88	–	55	24
LRCTM	background	1999	75	–	38	20

Figure 7.27 The areas of London modelled in the different studies.



765. The largest percentages of exceedence are predicted by the CERC and LRCTM models. All of the model results show a reduction in the area above 40 µg m^{-3} between 1999/2001 and 2010 but with exceedences remaining in central London and in the vicinity of Heathrow. The area of exceedence at Heathrow predicted in the netcen model is, however, much smaller than predicted in the other three models.

766. Table 7.12 shows a comparison of the lengths of roads in London predicted to exceed an annual mean NO_2 concentration of 40 µg m^{-3} for the different model results (but note that the exact length of road modelled varies between the different studies). The netcen and ERG results for major roads are very similar to each other in both 2001 and 2010, although the netcen results are somewhat higher for 2005. The percentage of minor road length predicted to exceed the 40 µg m^{-3} is considerably less than for major roads in both the ERG and CERC model results. This suggests that the majority of possible roadside exceedences in 2010 are likely to be confined to major rather than minor roads. This is an important result because it suggests that the national scale modelling of concentrations for major roads only should provide a reasonable approximation of the likely exceedences in 2010 for all roads. The CERC results, however, suggest that a greater percentage of roads in London are at risk of exceeding 40 µg m^{-3} in 2005 and 2010 than indicated by the other two models.

Table 7.12 Comparison of the percentage of the length of road in London predicted to exceed an annual mean NO_2 concentration of 40 µg m^{-3} at the roadside.

Model	Road network	Road length modelled (km)	Base year	1999	2001	2005	2010
Netcen	major roads	1767	2001	-	90	65	23
ERG	major roads	1937	1999		84	53	26
ERG	minor roads	2076	1999		55	27	8
ERG	all LAEI roads	4013	1999		69	39	17
CERC	major roads	1746	1999	98	-	79	49
CERC	minor roads	1910	1999	60	-	47	21
CERC	all LAEI roads	3656	1999	89	-	62	35

767. There are clearly important differences between the modelled predictions of annual mean NO_2 concentrations in 2005 and 2010 from the different models and these differences highlight the uncertainties associated with modelled predictions of future air quality. All of the model results show a significant reduction in the areas and lengths of roads above 40 µg m^{-3} between 1999/2001 and 2010. The estimates of the extent of background exceedence in 2010 range from 3% of Greater London for the netcen and ERG modelling to 13% for the CERC modelling (between 5-6% of the common model domain for the netcen and ERG modelling to 20-24% for the CERC and LRCTM modelling). The predicted area exceeding 40 µg m^{-3} statistic is very sensitive to small changes in concentration. A sensitivity analysis of the calculated area above 40 µg m^{-3} for the ERG predictions for 2005 shows that the area changes by a factor of about 2 for a 10% change in predicted concentrations. The predictions of the extent of roadside exceedence in 2010 range from 23-26% of major roads for the netcen and ERG modelling to 49% for the CERC modelling.

768. It is important to attempt to understand the reasons for these differences between the model results. The factors that may influence the different model results include:

- the source apportionment of 'current concentrations';
- the emission inventory;
- the emission projections;
- dispersion modelling methods;
- the treatment of sources outside London;
- the treatment of the conversion of NO_x to NO_2.

7.3.5 Source apportionment and emissions inventories

769. The netcen modelling is based on the NAEI inventory and projections, while the remaining studies all used the LAEI inventory and projections. A comparison of the source apportionment of annual mean NO_x concentrations between the netcen analysis presented in this report and a similar study by CERC (Blair *et al.*, 2003) shows that the results of the two studies are broadly consistent. This suggests that both the emission inventories and the source apportionment of current NO_x concentrations, and thus the general treatment of dispersion, are reasonably consistent in these two studies. Maps of annual mean NO_x derived from the CERC and netcen studies for both 1999/2001 and 2005 and 2010 are also reasonably consistent, which suggests that the relative declines in emission in the two inventories over the period from 1999/2001 to 2010 are consistent.

770. The NO_x concentrations predicted for 2005 and 2010 using the ERG approach, however, show much greater declines from the 1999 concentrations than suggested by the other studies (Carruthers *et al.*, 2002). The source apportionment of current concentrations between local road traffic and other sources within the ERG model is derived from a multiple regression with measurements (see section 5.2.3.2). Non-road traffic sources therefore make a much smaller relative contribution to current concentrations of NO_x in the ERG model compared with road transport. The modelled decline in concentrations of NO_x in the ERG model is therefore steeper than in the other studies. This is in contrast to the other models, in which the relative contribution from the non-traffic sources is greater. This affects the projections of NO_x concentrations because the non-traffic emissions are not expected to show the same decline as the traffic emissions between 1999/2001 and 2010 (see Chapter 2).

7.3.6 The treatment of dispersion

771. The LRCTM predicts the largest area of exceedence and highest concentrations in the Heathrow area and the netcen model predicts the lowest concentrations in this area. This is likely to be due to a combination of differences in the treatment of the dispersion from aircraft sources and differences between the emission projections within the NAEI and LAEI. The NAEI was used for the netcen projections and the LAEI was used for the other three projections. Aircraft sources have been modelled in the same way as all other sources within the LRCTM, with rapid mixing into the boundary layer. The height of the aircraft sources has been taken into account in the other studies. The greater assumed height of the majority of the emissions from aircraft in the other three studies leads to lower modelled ground level concentrations per tonne emitted, than the modelled concentrations for sources close to the ground such as traffic.

7.3.7 Sources outside London

772. The treatment of sources outside the model area is also important and can have an important influence on the area or length of road predicted to exceed 40 µg m^{-3}. The influence of regional O_3 concentrations is discussed below. All of the models also include contributions to NO_x concentrations in London from regional concentrations in the south east of England and these range between 13 to 19 µg m^{-3}, as NO_2 in 1999. The regional NO_x concentrations have been held constant between 1999 and 2010 in the LRCM modelling. Regional NO_x concentrations have been assumed to decline in line with changes in UK total NO_x emissions in the netcen and CERC studies, leading to a reduction of approximately 30% between 1999 and 2010. Regional NO_x concentrations have been assumed to decline in line with changes in NO_x emissions in London in the ERG study, leading to a reduction of almost 50%.

7.3.8 Chemistry

773. The different treatments of the conversion of NO_x to NO_2 in the different models is likely to be an important cause of the differences in predicted NO_2 concentrations. The CERC ADMS Urban and LRCTM calculations derived NO_2 from NO_x on an hourly basis using a simplified chemical reaction scheme while both the netcen and ERG approaches applied empirical relationships to estimates of annual mean NO_x concentrations. The more deterministically based models might be expected to provide a more accurate description of the chemistry of NO_2 formation, however both these models and the more empirically based models are far from being complete descriptions of all the physical and chemical processes taking place in the ambient air.

7.3.9 Treatment of direct NO_2 emissions

774. The different modelling approaches also take account of direct NO_2 emissions in different ways. The empirical models implicitly include the contribution made by direct NO_2 emissions for the current day. There will be uncertainty in the use of these methods when making forward projections if the proportion of direct NO_2 emissions changes; in addition it is difficult to take account of the detailed spatial variation in the proportion of the direct emissions that is NO_2. Conversely, although the deterministic models can specify the proportion of primary NO_2 in NO_x sources they are limited by uncertainties concerning direct emissions of NO_2. They have assumed a uniform NO_2 emission from traffic of 5% of NO_x by volume. Material presented in this report suggests that direct emissions could be higher in locations with a high proportion of slow-moving diesel vehicles for example, central London. For this reason, more of the NO_2 in the deterministic approaches may be chemically derived than is implicit in the empirical models. It is possible that these approaches will respond more slowly to reductions in NO_x compared with the empirical approaches.

7.3.10 Findings from the LAQM process

775. All local authorities in the UK have carried out a review and assessment of air quality in their area. This represents the first round of what are to be regular reviews and assessments to be completed every three years (Defra, 2003). The first round was a three-stage process, examining the likelihood of an exceedence of one or more of the objectives for the 7 regulated pollutants. The third, more detailed, stage was required most commonly for nitrogen dioxide, against the annual mean objective. In many cases the detailed assessment included automatic monitoring for NO_x using a chemiluminescence analyser, for periods ranging from 1 to 12 months. The results from these studies, all of which were focussed on road traffic sources, have been collated and analysed (Laxen *et al.*, 2002). The focus of the analysis was on the influence of distance from the edge of the road, with the data examined in three groupings: single carriageway roads outside of major conurbations; dual carriageway roads outside of major conurbations; and all roads in major conurbations. Data from national network sites were included, to provide a more comprehensive data set. The findings are summarised in Table 7.13 in terms of distances within which likely exceedences of the annual mean NO_2 objective might occur. An exceedence was judged likely if the 1999/2000 concentration was above 46 μg m^3. It is clear that exceedences are more likely:

- in major conurbations, rather than outside; and
- alongside single carriageway roads, rather than dual carriageways and motorways, outside major conurbations.

Conversely, exceedences are unlikely for locations:

- more than 9 m from the kerb of single carriageway roads outside major conurbations; and
- more than 5 m from dual carriageways and motorways outside major conurbations.

The finding that exceedences outside of major conurbations are likely to be confined to a narrow strip close to the road is consistent with the rapid fall-off in concentrations with distance from the kerb as seen in section 6.3.2.

Table 7.13 Likely exceedences of annual mean NO_2 objective in 2005 identified from monitoring near to roads (from Laxen *et al.*, 2002).

Location	Distances from kerb with exceedences[a]	Exceedences as proportion of number of near road sites[b]	Exceedences as proportion of all sites[c]	Total number of sites in analysis
Outside Major Conurbations – Single Carriageway Roads	0–9 m	36%	24%	59
Outside Major Conurbations – Dual Carriageways and Motorways	0–5 m	40%	12%	17
In Major Conurbations – All Roads[d]	0–35 m	81%	72%	18

a Outside this distance range no likely exceedences were identified.
b Proportion of sites within the distance range in previous column.
c Proportion of all sites (these were all within 100 m of the road, mostly within 50 m).
d The M25 was counted as being in a major conurbation.

7.4 Key issues for projections

7.4.1 Introduction

776. The projections presented here have been derived from a combination of base case emission inventory projections and our best estimates of the response of ambient concentrations to changes in emissions. The base case emission projections are based on an analysis of current national and international policies and their likely effect on future NO_x emissions. A full analysis of the uncertainties in the modelled projections of concentrations in 2005 and 2010 has not been carried out. The range in the results for London from the different models shows that the uncertainties are quite significant. It is clear, however, that exceedences of 40 μg m^{-3} in 2005 and 2010 are likely, but the exact extent of likely exceedence is uncertain. There are two issues that have not been incorporated into the detail of the base case projections. These are the possible impacts of an increase in hemispheric background O_3 concentrations and an increase in the proportion of NO_x emitted as primary NO_2. They have some potential to offset the predicted decreases in NO_x emissions over the next ten years or so, resulting in smaller reductions in ambient NO_2 concentrations than suggested by the base case projections.

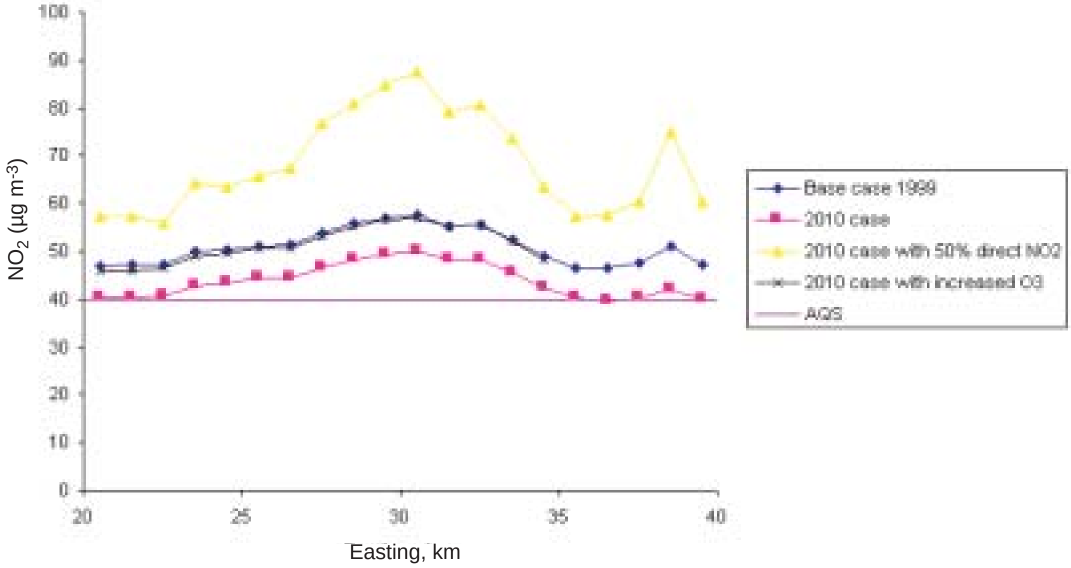
7.4.2 Policy impacts of other influences on future NO_2 air quality

777. Empirical models can only address those changes in future NO_2 air quality that are driven by NO_x emission changes because they are built from contemporary NO_x and NO_2 air quality data. The LRCTM model has been used to test whether there are other potential factors over and above changes in NO_x emissions that could potentially influence future NO_2 air quality and the exceedences of 40 µg m^{-3}.

778. Figure 7.28 presents the annual mean NO_2 concentrations calculated for an east-west transect through central London using the 1999 LAEI NO_x emissions inventory as a base case and with the corresponding 2010 inventory. The transect covers the region of annual mean NO_2 above 40 µg m^{-3} and exceedences in central London which is apparent in Figure 7.26. Annual mean NO_2 concentrations decrease at all points along the transect with 2010 compared with 1999 NO_x emissions.

779. Figure 7.28 also shows the results of two sensitivity cases for 2010 that cannot be easily investigated with empirical models. The first sensitivity case addresses the issue of any increase in the direct emissions of NO_2 in the future. The second sensitivity case addresses what happens if baseline O_3 concentrations continue to rise in the British Isles and increase the capacity of urban atmospheres to convert NO into NO_2.

Figure 7.28 Annual mean LRCTM model NO_2 concentrations along a transect through central London with 1999 and 2010 NO_x emissions, showing the impact of increased O_3 baseline concentrations and of increasing direct emissions of NO_2 .



7.4.3 Increasing direct emissions of NO_2

780. The first sensitivity case assumed that the fraction of NO_x emitted directly as NO_2 changed from 5% by volume in the base case to 50% by volume in the 2010 case for all sources in the inventory. Under these conditions, NO_2 concentrations along the transect then rise significantly above those in the 1999 case and a significant gap opens up between the future NO_2 concentrations in 2010 and 40 µg m^{-3}, see Figure 7.28.

781. The results of this model sensitivity case suggest that it is important to quantify the direct emissions of NO_2 from diesel-engined cars, buses and goods vehicles in the contemporary fleet and in the future. It is also particularly important to determine the influence of catalytically-regenerative diesel particle traps on the direct emissions of NO_2 from the buses to which they have, and will be, fitted in the near future. It should be noted however, that the 50% of all NO_x emissions emitted directly as NO_2 should be considered as an extreme worst case sensitivity test rather than a realistic scenario.

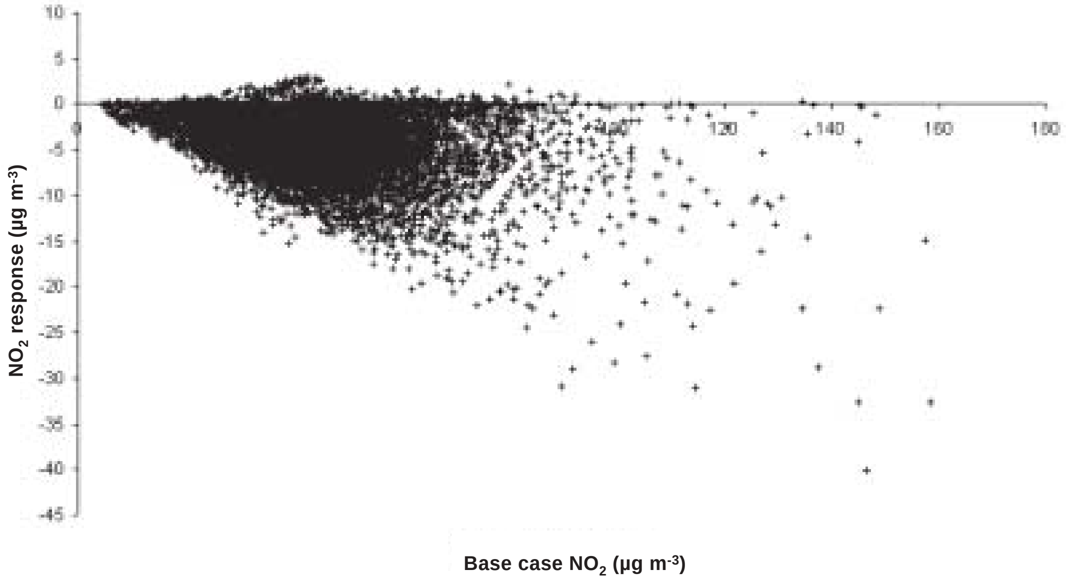
7.4.4 Increasing hemispheric O_3 baseline concentrations

782. The second sensitivity case assumed that baseline O_3 concentrations may rise significantly in the future and that they continue to rise at the same annual rate of increase as that observed historically for background air masses at Mace Head, Ireland during the 1990s. The assumed increase varies by month of the year and is highest in the winter and lowest in the summer, as described in section 6.6.2. The average increase is approximately +1 µg m^{-3} (+0.5 ppb) O_3 per year. Under these assumptions, an additional 2010 case was modelled with both decreased future NO_x emissions and increased O_3 baseline concentrations. Under these conditions, future NO_2 concentrations remained close to 1999 levels, despite the significant reduction in NO_x emissions, see Figure 7.28. It appears that increasing O_3 baseline levels could increase the oxidising capacity of London's atmosphere, enough to offset any improvements anticipated from the reduction of NO_x emissions by the year 2010.

783. In attempting to summarise these results for the 2010 cases compared with the base case, it becomes apparent that there is a less than linear relationship between annual mean NO_2 concentrations and London-wide total NO_x emissions. That is to say, if London-wide total NO_x emissions decrease by x% between 1999 and 2010, then annual mean NO_2 concentrations tend to decline by less than x%. This less-than-linear relationship between NO_2 and NO_x was investigated using the LRCTM model.

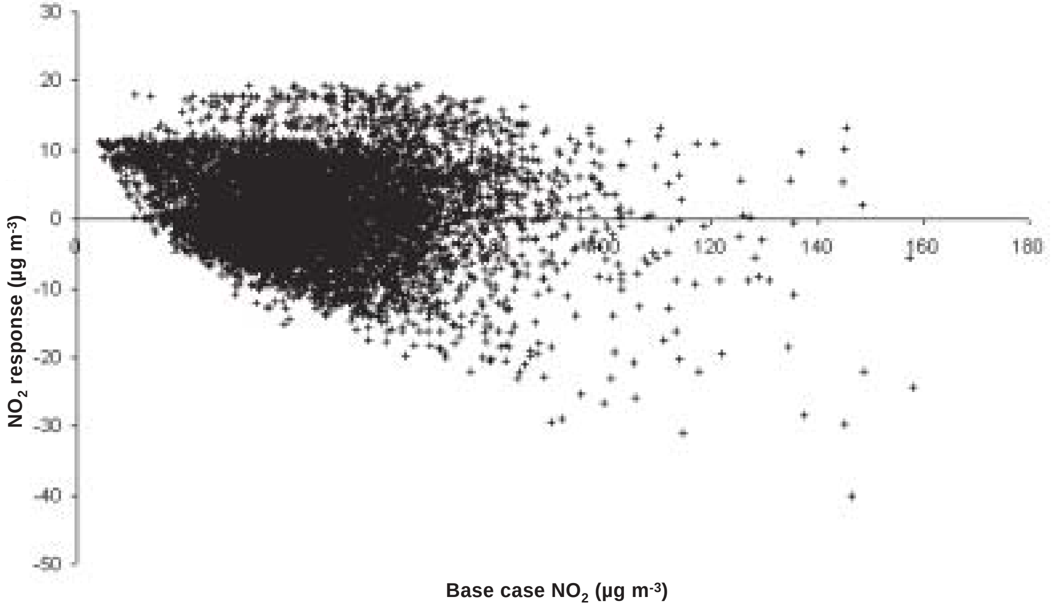
784. Figure 7.29 presents the NO_2 responses for each hour at a location in central London, expressed as 2010 case minus 1999 case so that a NO_2 decrease shows up as a negative value in the plot. Each NO_2 response was plotted out as a scatter plot against its respective base case NO_2 concentration. At each NO_2 concentration level in the base case, there was a large variation in NO_2 response which varied over the range from zero up to about 20%, despite there being about a 38% reduction in NO_x emissions overall. For some hours, there was no apparent NO_2 response to the reduction in NO_x emissions. These are the hours in which NO_2 formation is entirely O_3-limited. Decreasing NO_x emissions then makes no difference to NO_2 formation. For those hours close to the upper-limit surface, NO_2 concentration responses are about 20% for a 38% reduction in emissions, representing an efficiency of about one half to two-thirds. Here NO_2 formation is entirely NO_x limited. Decreasing NO_x emissions then makes a direct difference to NO_2 formation but the efficiency is not unity because for every 2 µg m^{-3} decrease in NO_x as NO_2 (1 ppb), NO_2 decreases by about 1.2 µg m^{-3} (0.6 ppb) and NO decreases by about 0.5 µg m^{-3} (0.4 ppb) because of the photostationary state relationship.

Figure 7.29 Scatter plot of the hourly LRCTM model NO_2 concentration responses against the base case NO_2 concentration for changing NO_x emissions between 1999 and 2010.



785. Figure 7.30 presents the corresponding NO_2 responses to decreasing emissions between 1999 and 2010 and increasing baseline O_3 in 2010. The influence of the increased oxidation capacity has been dramatic and NO_2 concentrations in some hours have increased despite the reductions in NO_x emissions. Overall the decreases in NO_2 concentrations in the NO_x-limited hours have been compensated by the increases in NO_2 concentrations during the O_3-limited hours. Annual mean NO_2 concentrations are thus left more or less unchanged from 1999 to 2010.

Figure 7.30 Responses to NO_x emission reduction and increased baseline O_3 of each hourly mean LRCTM model NO_2 concentration plotted against the base case NO_2 concentration.



786. On this basis, it is concluded that urban NO_2 concentrations in London are surprisingly sensitive to increases in the hemispheric O_3 baseline concentrations in the air masses as they impinge on the London region. This conclusion is entirely derived from model calculations and as yet is unsupported by observations. It is important then to examine the available NO_x-NO_2-O_3 monitoring records from long running sites to establish whether there has been any influence from increasing O_3 baseline levels in NO_2 levels. If such a relationship between hemispheric O_3 and urban NO_2 can be discerned from the observations then it may be necessary to take into account hemispheric O_3 trends in planning future NO_x emission reductions in urban areas. The limited analysis of measured NO_x, NO_2 and O_3 concentrations at West London and Glasgow City Chambers presented in section 6.6.3 suggests that the impact of changes in hemispheric baseline O_3 levels may be variable across the UK, with a greater impact in Glasgow than in London, although the analysis is confounded by changes in summer episode O_3 concentrations.

7.4.5 ADMS-Urban calculations of the impact of increased O_3 concentrations

787. The possible impacts of increasing concentrations of hemispheric O_3 on NO_2 concentrations in London in 2010 have also been estimated using ADMS-Urban. The estimated monthly average increases in O_3 concentrations listed in Table 7.12 in Chapter 6 were added to each hourly background O_3 concentration input into the model for the relevant month. This increased the annual mean regional O_3 concentration in 2010 from 61.52 µg m^{-3} to 73.72 µg m^{-3} (30.76 ppb to 36.86 ppb)

788. The increase in regional O_3 concentration results in an increase in annual mean NO_2 concentration of 4.4 µg m^{-3} (2.3 ppb) averaged across the 23 sites modelled and 3.6 µg m^{-3} (1.9 ppb) at the 14 background sites. The corresponding increases in modelled O_3 concentrations are 7.2 µg m^{-3} (3.6 ppb) averaged over all site sites and 8.3 µg m^{-3} (4.1 ppb) at background sites. While the increase in total input oxidant concentration (NO_2 + O_3) is conserved in both the LRCTM and ADMS-Urban calculations, the partitioning of the increase in oxidant between NO_2 and O_3 is different. For the same 12 µg m^{-3} (6 ppb) increase in input O_3 concentrations the resulting average increase in annual mean NO_2 ranges from about 3.8 µg m^{-3} (2 ppb), for the ADMS-Urban calculations for background sites, to about 6.7 µg m^{-3} (3.5 ppb) for the LRCTM calculations. This difference is likely to be due to the different treatment of dispersion and mixing within the two models since the chemical schemes adopted are similar. The results of these two studies confirms that an increase in hemispheric O_3 concentrations could have an important impact on projected annual mean NO_2 concentrations in 2010 and that this is clearly an important area for further research.

Chapter 8

Conclusions

789. The Expert Group has examined monitoring data for 212 sites across the UK, which show that the annual mean NO_2 concentrations have exceeded 40 µg m^{-3} by a wide margin in recent years. This value of 40 µg m^{-3} is both the UK objective and the EU limit value, although the former is to be achieved by 2005, while the latter is to be achieved by 2010. Many of these observed exceedences are found in London. They have also been reported in other conurbations, but because the density of monitoring sites is much lower than in London, the full extent of the exceedences is difficult to gauge from observations. The Local Air Quality Management process operated by local authorities has also identified current exceedences in smaller towns, especially for locations alongside narrow congested town centre roads, where houses can be close to the kerb. Examination of the European Environment Agency air quality database has shown that current NO_2 concentrations exceed 40 µg m^{-3} at many locations throughout Europe, and thus the situation in the UK is not unique. Direct comparisons with other European cities cannot, however, be made because of differences in approaches to siting of monitoring stations in the various countries. Whilst the hourly mean objective value of 200 µg m^{-3} per year (with 18 allowed exceedences) is occasionally breached, it represents a much less challenging target for air quality management than the annual mean objective.

790. The Expert Group has examined a hierarchy of modelling tools to quantify the spatial distribution of the annual mean NO_2 concentrations from NO_x emission inventories. The general level of agreement found between models and observations is good and confirms that the available NO_x emission inventories give an accurate picture of actual NO_x emissions and that two main processes control the annual mean distribution of NO_2 in urban areas. These two processes are the oxidation of emitted NO to NO_2 by O_3 transported in from the surrounding rural areas, and direct emissions of nitrogen dioxide by vehicular traffic. The general level of agreement found between model and observation at roadside, kerbside and urban background sites away from the immediate influence of roads, argues strongly that models are able to provide a reasonable description of the fall-off in annual mean concentration with increasing distance from roads. On this basis, the Expert Group was able to confirm that exceedences of 40 µg m^{-3} annual mean NO_2 are widespread throughout the UK. Exceedences are unlikely to be limited to the London conurbation and may also be found in all major conurbations, in all the devolved administrative regions of the UK and along all the heavily-trafficked arterial roads.

791. The widespread exceedences of 40 µg m^{-3} NO_2 as an annual mean remain, despite a decade of decreasing NO_x emissions. Running mean NO_x concentrations have declined by up to 3% per annum at urban background sites during the late 1990s. However, simultaneous running mean NO_2 concentrations in London show significantly lesser downward trends or no trend at all. The Expert Group has explained this important difference in the behaviour of NO_x and NO_2 with time through oxidant limitations on chemical production of nitrogen dioxide and increased direct emissions of NO_2. Despite the reduction in NO_x emissions, concentrations of NO in pollution episodes still reach levels at which the thermal oxidation of NO by O_2 plays an important role in producing NO_2, as it did in the December 1991 London episode.

792. The assumption that the relationships in the empirical models between NO_x and NO_2 imply cause and effect relationships has been examined. Using physically-based process models, the Expert Group has been able to show that the empirical relationships between NO_x and NO_2 are exactly those expected if oxidation of NO by O_3 and direct emission of NO_2 are indeed the two major sources of urban NO_2. However, empirical models are only valid for the range of conditions experienced during their derivation, and changes in, for example, background O_3 concentrations will require model reformulation, and imply that the current models will probably not be reliable in 2010.

793. Despite continuing decreases in urban NO_x emissions projected through to the years 2005 and 2010, modelling studies reviewed by the Expert Group still predict continuing exceedences of the annual mean objective and limit value for NO_2 beyond the relevant dates. Inevitably, these models contain a number of simplifications and inadequacies. They are driven with input data which themselves contain approximations and are subject to uncertainties. Much reliance has been placed on dispersion models and empirical relationships between NO_x and NO_2. Where possible they have been verified and validated against observations but this has been difficult outside of the London conurbation because of the more limited availability of high quality monitoring data with long enough time series.

794. The Expert Group remains convinced that, on the basis of current projections of traffic volumes and vehicle sales and predictions of the reduction of vehicle NO_x emissions on a kilometre travelled basis as new control technologies are introduced, some exceedences of the annual mean objectives and limit values for NO_2 will remain in the years 2005 and 2010 respectively. These will be less widespread than at present, but nonetheless still appreciable. Crucial to these predictions are the assumptions about traffic growth, rate of turnover in the vehicle fleet and NO_x emissions from new vehicles in the future. If future traffic flows exceed expectations, or emission control technologies fail to deliver anticipated reductions, continued exceedences will be more widespread. On the basis of current evidence, heavy duty diesel vehicles emitted 43% of NO_x emissions from UK road transport in 2000, but this conclusion is based on poorly characterized emissions. Levels of NO_x and of direct NO_2 emissions from these vehicles need to be determined through the continuation of robust emission research programmes focusing on both current and future vehicle technologies.

795. NOx emissions are primarily in the form of NO, but there is evidence for significant amounts of NO_2 emitted directly from the tailpipe of diesel vehicles, with levels possibly as high as 25% of total NO_x emissions. Current evidence suggests that increasing the diesel car penetration rate in the UK fleet would lead to a small increase in NO_x emissions from road traffic in urban areas, but a more significant increase in primary NO_2 emissions. An increase in diesel car sales in 2010 from 22% of new car sales (close to the current rate) to 30% would increase urban UK road transport emissions of NO_x in 2010 by 0.7% and NO_2 emissions by 3%. In an extreme case, where diesel car sales increased to 50% of new car sales, urban UK road transport emissions of NO_2 in 2010 would be increased by 6%.

796. Progress in the Government's Ten Year Plan for transport and other transport policies will need to be monitored regularly if the UK is going to reduce NO_x emissions sufficiently to meet the National Emissions Ceiling Directive target for 2010. The Government's recently proposed National Emission Reduction Plan for implementing the Large Combustion Plant Directive will help ensure emission reductions for stationary combustion sources are realised, but levels of activity in the power generating and industry sectors and effectiveness of abatement measures adopted will need to be monitored to ensure the reductions do occur. Uncertainties in the emission projections, and the sensitive drivers that are used to derive them, will need to be considered when progress towards meeting the NECD targets is reviewed.

797. The Expert Group is aware that the local air quality management process being undertaken by local authorities is leading to the development of local air quality action plans to deal with hot-spots. These measures are welcomed by the Expert Group and should help reduce NO_2 concentrations at critical locations. However, because of the devolved nature of this local authority work, the Expert Group has been unable to form a view on the extent to which these measures will contribute to meeting the objectives and limit values.

798. There are reasons to believe that the current projections for future urban NO_2 concentrations may be optimistic. If northern hemisphere baseline O_3 concentrations continue to rise and influence rural O_3 concentrations in the UK, then the relationships between urban NO_2 and NO_x concentrations will alter, resulting in higher than expected future annual mean NO_2 concentrations. The Expert Group were unable to establish clear evidence for or against this hypothesis based on historic monitoring data. There is a clear need for developing new predictive models able to allow for changes in background O_3. Furthermore, if catalytically-regenerative particulate traps that are being retrofitted to diesel powered vehicles significantly increase direct emissions of NO_2, as indicated by studies carried out in the USA, there will be further breaches of the air quality objective and limit value. The Expert Group was unable to find any measurements of direct emissions of NO_2 from vehicles retro-fitted with catalytically-regenerative traps in the UK and were hence unable to form a considered view on whether this is likely to become a problem. However, given the limited number of diesel vehicles in the national fleet which are likely to be retrofitted with these types of regenerative traps, the problem is unlikely to be widespread. It may be more acute in certain areas where local fleets operate which have been targeted with the retrofitting of CRTs in order to control local particulate emissions. Central London, where a significant number of buses have been retrofitted, may be an example. In the future manufacturers of new vehicles and engines will be required to meet increasingly stringent limits on total NO_x as well as on particulate emissions for Euro IV type-approval. Already, particulate trap technology exists which includes downstream NO_2 emission control.

799. The Expert Group has given careful consideration in this report to the processes involved in the emission, formation, dispersion and removal of NO_2, that ultimately determine exposures of the general population to NO_2. However, the Expert Group is only too aware that in this analysis, NO_2 should not be considered in isolation. Traffic sources emit a number of pollutants in addition to NO_x and of particular concern is the emission of particulate matter. Measures taken to reduce particulate matter emissions may increase public exposure to NO_2, particularly if they focus on the important role of the particle emissions from diesel-engined vehicles in contributing to the exceedence of the air quality objectives for PM_{10}. Increasing direct emissions of NO_2 from diesel vehicles retro-fitted with particle traps would lead to increased airborne concentrations of NO_2.

800. NO_2 should not be considered in isolation from other pollutants. In developing air pollution controls, there may therefore be trade-offs between different pollutants, and it is a matter of concern that much of local air quality management is currently driven by exceedences of one air quality objective: the annual mean of 40 $\mu g\ m^{-3}$ for NO_2. The Expert Group feels that a more flexible and holistic approach to air quality management might deliver more effective control strategies.

Chapter 9

Research recommendations

801. Research requirements have been identified in the following areas:

802. A better understanding of vehicle behaviour and of instantaneous emissions from traffic at specific locations like road junctions and areas, where modelling emissions by the more simplistic average speed approach is inadequate. Refined estimates of the effects of instantaneous traffic activity (speed/load/rates of acceleration).

803. Provision of robust NO_x emission factors and the fraction of NO_x emitted as NO_2 for current and future road vehicles that can be used in inventories and dispersion models. Emphasis should be placed on measurements of emissions of NO_x and primary NO_2 from heavy duty diesel vehicles meeting current and future emission standards.

804. Measurement of primary NO_2 emissions from other ground-level combustion sources, particularly other transport and machinery types using diesel engines (for example, rail locomotives, construction plant , non-road mobile machinery) and domestic burner and combustion appliances.

805. Investigation of the effects of aircraft emissions at major airports on ground level NO_2, including measurements of emissions and dispersion modelling.

806. Assessment of the effects of domestic emissions on urban background concentrations of NO_2, including forward projections.

807. Development of a better understanding of the effects of meteorology on NO_2 concentrations, including: (i) investigation of the effect of urban environments on meteorological fields; (ii) measurements of meteorological parameters above and below the urban canopy; (iii) assessment of the significance of the choice in models of input meteorological data (station and year), via modelling sensitivity studies; (iv) Coordinated and co-located measurements of air quality, meteorology and detailed traffic activity and better quality control of those weather data which are already collected at urban air quality monitoring stations; (v) evaluation of remote sensing techniques to allow local monitoring of meteorological parameters to be extrapolated to other locations within the urban boundary layer.

808. Research into the use of other modelling approaches, that have not been used extensively in this report for the assessment of NO_2 concentrations, is required. These approaches include computational fluid dynamics (CFD) for the consideration of small spatial and temporal scales, for example, close to roads where the meteorology can be influenced by buildings, and Eulerian approaches for the urban to regional scale. One important application of such models is in helping to build the understanding required to carry out a scientific assessment of the empirical and deterministic models used extensively in this report.

809. Investigation of the determinants of NO_2 concentrations at roadside hotspots, including meteorology, mixing of O_3 from the urban canopy, and chemical kinetics.

810. Further assessment of the contributions of free radical-driven oxidation processes to NO-to-NO_2 conversion in polluted urban environments.

811. Further comparisons of the national empirical model and dispersion models in cities other than London, to confirm the applicability of the conclusions from London elsewhere. Implementation of this recommendation may be difficult, however, because of the more limited extent of hourly continuous monitoring in other major conurbations.

812. Support for national empirical modelling via improved provision of automatic NO_2 measurements, of known high quality, outside London and especially in Scotland, Wales and Northern Ireland. Specific requirements include roadside monitoring data in the West Midlands and greater co-location of rural NO_x and O_3 measurements.

813. Further investigation of the interactions between trends in O_3 and NO_2

814. Further modelling of the relationship between the northern hemisphere scale increase in O_3 and urban scale NO_2 problems. Remote background O_3 is rising as shown by the monitoring data for the Mace Head site in Ireland and the Strath Vaich site in the Highlands of Scotland. There is uncertainty regarding the influence of these rising levels once they have reached urban areas.

815. Modelling studies on the effects of possible increases in the proportion of primary NO_2 emissions on the exceedence of annual mean NO_2 targets.

816. Investigation of various aspects relating to the spatial characteristics of NO_2 concentrations, including: (i) the spatial characteristics of air quality episodes; (ii) the influence of interpolation errors in modelling procedures; and (iii) the influence of particular siting and sampling characteristics on interpolated surfaces. Developments are needed in methodologies for the wide application of interpolation procedures to policy work.

817. Better collection and availability of detailed information on site characteristics, including grid references to 1 m resolution and distance from kerb to enable more extensive GIS and other data analysis and verification of model results.

818. More studies on the decline in NO_x and NO_2 concentrations away from the kerb of major roads in both rural and urban areas, including: (i) long-term studies using chemiluminescent automatic monitors, and; (ii) measurement of both NO_x and NO_2 concentrations for at least one month in the summer and one month in the winter, to aid understanding of seasonal influences.

819. Research on the effectiveness of local air quality management measures on the abatement of NO_2 exceedences.

820. Development of a more holistic approach to air quality management that reflects the trade-offs between abatement strategies for NO_2 and for other pollutants.

Abbreviations and glossary

Abbreviations

AADT	Annual Average Daily Traffic (vehicles per day)
ADMS	Atmospheric Dispersion Modelling System
AQMA	Air Quality Management Area
Aric	Atmospheric Research Information Centre
ARTEMIS	Assessment and Reliability of Transport Emission Models and Inventory Systems
ATC	Automatic Traffic Counts
AURN	Automatic Urban and Rural (air quality monitoring) Network
BAM	Beta Attenuation Monitor
BAT	Best Available Techniques
CCGT	Combine cycle gas turbines
CCTV	Closed Circuit Television
CEN	European Committee for Standardisation
CLRTAP	Convention on Long-Range Transboundary Air Pollution
CNG	Compressed Natural Gas
CO	carbon monoxide
CO_2	carbon dioxide
COMEAP	Committee on the Medical Effects of Air Pollutants
COPERT III	Computer Programme to Calculate Emissions from Road Transport (version III)
CORINAIR	COoRdination d'INformation Environmentale
COST	European Cooperation in the field of Scientific and Technical Research
CRTs	Continuously Regenerating Particulate Traps
CSTM3	Central Scotland Transport Model
CVS	Constant volume sampler
Defra	Department for Environment, Food & Rural Affairs
DfT	Department for Transport
DMRB	Design Manual for Roads and Bridges
EA	Environment Agency
EFD (or EFDB)	Emissions Factor Database
EfW	Energy from Waste

EGR	Exhaust Gas Recirculation
EIA	Environmental Impact Assessment
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe
EP68	Dti's Energy Paper 68
EPEFE	European Programme on Emissions, Fuels and Engine technologies
EPER	European Pollutant Emission Regsiter
ERG	Environmental Research Group at King's College London, developers of the empirical modelling approach most widely used in London
EU	European Union
EU15	The 15 Member State countries of the European Union
FIGE	Forschungsinstitut Geräusche und Erschütterungen
FTIR	Fourier Transform Infra-red Spectroscopy
GDP	Gross Domestic Product
GIS	Geographical Information System
GLA	Greater London Authority
GSS	Guidance for estimating the air quality impact of Stationary Sources
HC	Hydrocarbons
HDV	Heavy Duty Vehicles – Road vehicles greater than 3.5 tonnes weight (GVW).
HECA	Home Energy Conservation Act 1995
HGV	Heavy Goods Vehicle – Road vehicle greater 7.5 tonnes (GVW). Where GVW is the gross vehicle weight i.e the combined weight of the vehicle and the goods.
HGVa	Heavy Goods Vehicle – articulated
HGVr	Heavy Goods Vehicle – rigid
HONO	Nitrous acid
HNO_3	Nitric acid
HRTM	Heathrow Road Traffic Model
ICAO	International Civil Aviation Organisation
IIASA	International Institute for Applied Systems Analysis
IPC	Integrated Pollution Control
IPCC	Intergovernmental Panel on Climate Change
IPPC	Integrated Pollution Prevention and Control
ISO	International Standards Organisation
LAEI	London Atmospheric Emissions Inventory

LGV	Light Goods Vehicles – goods vehicles less than 3.5 tonnes in weight
LAQN	London Air Quality Network
LEZ	Low emission zone. A specific type of 'Clear Zone'.
LRC	London Research Centre
LTO	Landing and take-off
LTS	London Transport Study
mg m^{-3}	milligrams per cubic metre of air
µg m^{-3}	micrograms per cubic metre of air
NAEI	National Atmospheric Emissions Inventory
NAMAS	National Accreditation of Measurement and Sampling
NECD	National Emissions Ceilings Directive
netcen	National environmental technology centre, part of AEA Technology plc
NMVOC	Non-methane volatile organic compound
NO	Nitrogen monoxide, also termed nitric oxide
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides (NO + NO_2)
NRTF	National Road Traffic Forecasts
NWS	National Waste Strategy (Scotland)
O_2	Oxygen
O_3	Ozone
OS	Ordnance Survey
OX	Oxidant potential. A sum of O_3 and NO_2
PAN	Peroxyacetyl nitrate
PI	Pollution Inventory
PM_{10}	Particulate matter with an (equivalent aerodynamic) diameter of ten microns (10 µm) or less
$PM_{2.5}$	Particulate matter with an (equivalent aerodynamic) diameter of 2.5 microns (2.5 µm) or less
ppb	parts per billion (1,000,000,000)
ppm	parts per million
QA/QC	Quality Assurance and Quality Control
RO_2	organic peroxy radicals
SCR	Selective Catalytic Reduction
SEPA	Scottish Environment Protection Agency
SO_2	sulphur dioxide

TEMPRO	Trip End Model PROjections of growth in travel demand, and the underlying car ownership and planning data projections
TEOM	Tapered Element Oscillating Microbalance
TRAMAQ	DfT funded, Traffic management and air quality research programme. http://www.roads.dft.gov.uk/roadnetwork/tramaq/
TRL	Transport Research Laboratory
TÜV	German Accreditation Organisation
UKAS	United Kingdom Accreditation Service
ULSD	Ultra-Low Sulphur Diesel
ULSP	Ultra-Low Sulphur Petrol
UNECE	United Nations Economic Commission for Europe
USA	Updating and Screening Assessment
USEPA	United States Environmental Protection Agency
VOC	Volatile organic compound
WASP	Workplace Analysis Scheme for Proficiency
WDF	Waste Derived Fuel (or Refuse Derived Fuel)

Glossary

Accuracy	A measure of the closeness of the agreement between the result of a measurement and the true value (see also Uncertainty and Precision).
Air quality objective	Policy targets generally expressed as a maximum ambient concentration to be achieved, either without exception or with a permitted number of exceedences within a specified timescale (see also air quality standard).
Air quality standard	The concentration of a pollutant, and associated averaging period, which is without significant effect on human health at a population level.
Ambient air	Outdoor air in the troposphere, excluding workplace air
Annual mean	The average of the concentrations measured for each pollutant for one year. In the case of the air quality objectives this is for a calendar year.
AQMA	Air Quality Management Area, an area which a local authority has designated for action, based upon predicted exceedences of Air Quality Objectives.
Area source	Emission sources that are too diffuse to identify individually (for example residential properties)
Atmospheric boundary layer	The lowest 100 to 3000 m of the atmosphere. That part of the troposphere that is directly influenced by the presence of the earth's surface, and responds to surface forcing with a time scale of about an hour or less. The forcing includes frictional drag, evaporation and transpiration, heat transfer, pollutant emission, and terrain induced flow modification.
Atmospheric dispersion model	A mathematical, often computer-based method for calculating pollutant concentrations from emissions data and specified meteorological conditions. Models vary from screening models to detailed, 'new-generation' types.
AURN	Automatic Urban and Rural Network of air pollution measurement sites, managed by contractors on behalf of Defra and the devolved administrations.
Baseline	Concentration of a pollutant (usually used referring to ozone) at a location unaffected by local or regional sources.
Blending height	Height over the ground at which the ground inhomogeneity is not perceived, over which the various internal boundary layers merge into a layer having a horizontally homogeneous structure.
Calibration (modelling)	The process of multiplying the output of a model by a fixed correction factor to give, on average, a 1:1 relationship with measured data.
Calibration (monitoring)	The process of reducing the uncertainty of monitoring data by controlled tests on the analyser, normally traceable to internationally accepted measurements standards.

Concentration	The amount of a (polluting) substance in a volume (of air), typically expressed as a mass of pollutant per unit volume of air at standard conditions of temperature and pressure (eg micrograms per cubic metre, µg m^{-3}) or as the ratio of the number of molecules of the pollutant to the total number of molecules in the volume of air (for example, parts per billion, ppb).
Conservative	Tending to over-predict the impact rather than under-predict.
Convective boundary layer	The atmospheric boundary layer under strong insolation, which causes positive buoyancy and the generation of turbulence (as opposed to the stable boundary layer).
Correlation coefficient	The fraction of the variability in one set of data that is proportional to the value of some other set of data.
Diffusion equation	Mathematical expression relating the rate of change of concentration in time to the rate of change of concentration over distance.
Data capture	The percentage of all the possible measurements for a given period that were validly measured.
Emission	The amount of a (polluting) substance emitted in a certain amount of time, typically expressed as a mass of pollutant per unit time (for example, grams per second, or tonnes per year for a single source). May also be expressed per unit length of a road (for example, g s^{-1} m^{-1}), or per unit area of an urban area (for example, t a^{-1} km^{-2}).
Emissions inventory	A quantification and compilation of emission sources by geography and time, usually including data covering one or several years.
Euro I	Europe-wide vehicle standard that required vehicles manufactured after 1992 to achieve set emissions limits. For petrol cars this was achieved by the fitting of three way catalysts.
Euro II, III, IV & V	Europe-wide vehicle standards that are progressively stricter, for years 1996, 2000, 2006 and 2008 respectively.
Exceedence	A period of time where the concentration of a pollutant is greater than the appropriate air quality objective.
Free radical	A molecule with an unpaired electron. Most free radicals are highly reactive. NO and NO_2 are examples of free radicals that are comparatively stable.
Inertial sublayer	The remaining part of the urban surface layer above the roughness sublayer where surface layer scaling (Monin-Obukhov similarity) applies.
Internal boundary layer	The atmospheric layer that develops after a change in ground roughness or surface temperature. Its height increases with the distance from the change.
Line source	A long, narrow source of emissions such as a road, railway line or shipping lane.
Maximum hourly average	The highest hourly reading of air pollution obtained during the time period under study.

Metadata	Information about a dataset which provides the data analyst with an essential description of the data capture process. For example, the instrument used, temporal resolution, monitoring site details, quality control process applied, etc.
mg m^{-3}	Milligrams per cubic metre of air. A unit for describing the concentration of air pollutants in the atmosphere, as a mass of pollutant per unit volume of air. This unit is one thousand times larger than the µg m^{-3} unit listed below. Valid comparisons of concentrations given in mg m^{-3} usually require the volume of air to be corrected to a common temperature and pressure.
Microgram (µg)	One millionth of a gram.
µg m^{-3}	Micrograms per cubic metre of air. A unit for describing the concentration of air pollutants in the atmosphere, as a mass of pollutant per unit volume of air. A concentration of 1 µg m^{-3} means that one cubic metre of air contains one microgram of pollutant. Valid comparisons of concentrations given in µg m^{-3} usually require the volume of air to be corrected to a common temperature and pressure.
Micrometre (µm)	One millionth of a metre, also referred to as a micron.
Minor roads	Non A roads or motorways.
Mixed layer	The major part of the convective boundary layer. Within the mixing layer the vertical profiles of most meteorological variables are roughly constant with height due to the intensive vertical mixing. The mixing height is the height of the layer next to the ground over which pollutants or any constituents emitted within this layer or entrained into it become vertically dispersed by convection or mechanical turbulence within a time scale of approximately 30 minutes.
Model adjustment	Following model verification, the process by which modelled results are amended. This corrects for systematic error.
Multiple regression	A method of statistical analysis that identifies empirically how a variable of interest (for example, pollutant concentration) depends on the value of several determinants (for example, emissions, wind speed, altitude).
nm	Nanometre 10^{-9} metres.
Oxidation	The increase in the proportion of oxygen in a molecule.
Percentile	A value that is the rank at a particular point in a collection of data. For instance, a 98th percentile of values for a year is the value that 98% of all the data in the year fall below, or equal.
Photochemical processes	Chemical reactions which occur following the absorption of light by a molecule.
Photolysis	The decomposition of molecules following the absorption of light.
PM_{10}	Airborne particulate matter passing a sampling inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter and which transmits particles of below this size.

$PM_{2.5}$	Airborne particulate matter passing a sampling inlet with a 50% efficiency cut-off at 2.5 μm aerodynamic diameter and which transmits particles of below this size.
Point source	A stationary location or fixed facility from which pollutants are discharged or emitted, this may include a number of stacks or a large plant.
ppb	Parts per billion. The concentration of a pollutant in air in terms of molar ratio. A concentration of 1 ppb means that for every billion (10^9) molecules in a volume of air, there is one molecule of the specified pollutant present. For practical purposes in ambient air, the molar ratio and volume ratio (the volume occupied by the pollutant gas within a given volume of air) are identical.
ppm	Parts per million. The concentration of a pollutant in air in terms of molar ratio. A concentration of 1 ppm means that for every million (10^6) molecules in a volume of air, there is one molecule of the specified pollutant present. For practical purposes in ambient air, the molar ratio and volume ratio (the volume occupied by the pollutant gas within a given volume of air) are identical.
Precautionary approach	In air quality modelling, erring on the side of predicting concentrations higher than those that actually occur or will occur, when there are known to be significant uncertainties in the model.
Precision	A measure of the closeness of the agreement between the results of successive measurements where the true value remains constant (see also Accuracy and Uncertainty).
Ratification (monitoring)	Involves a critical review of all information relating to a data set, in order to amend or reject the data. When the data have been ratified they represent the final data to be used (see also Validation).
Regression	*See multiple regression.*
Road link	A length of road, usually in a traffic model or emissions inventory, which has the same flow of traffic along its length. Usually, a link is the road from one junction to the next.
Roughness	A length-scale describing how a bumpy surface influences the development of turbulence in a flow of air above that surface.
Roughness sublayer	The lowest part of the urban surface layer, where the strong influence of the buildings and roughness elements and turbulence is three-dimensional.
Running mean	A mean – or series of means – calculated for overlapping time periods, and is used in the calculation of several of the national air quality objectives. For instance, an 8-hour running mean is calculated every hour, and averages the values for eight hours. The period of averaging is stepped forward by one hour for each value, so running mean values are given for the periods 00:00 – 07:59, 01:00 – 08:59 etc. By contrast a non-overlapping mean is calculated for consecutive time periods, giving values for the periods 00:00 – 07:59, 08:00 – 15:59 and so on. There are, therefore, 24 possible 8-hour means in a day (calculated from hourly data) and 3 non-overlapping means.

Surface layer	The lowest 10% of the atmospheric boundary layer. Frictional drag, heat conduction and evaporation from the surface causes substantial changes with height of the mean wind speed, temperature and humidity. However, the turbulent fluxes are relatively constant with height in the surface layer; hence the surface layer is also known as the constant flux layer.
TEOM	Tapered element oscillating microbalance. Equipment used for measuring fine particulate matter such as PM_{10}.
True value	The value of a concentration, for example, which is entirely consistent with the definition of the units in which it is given. This is the value that would be obtained by a perfect measurement.
Uncertainty	A measure, associated with the result of a measurement, that characterizes the range of values within which the true value is expected to lie. Uncertainty is usually expressed as the range within which the true value is expected to lie with a 95% probability, where standard statistical and other procedures have been used to evaluate this figure. Uncertainty is more clearly defined than the closely related parameter *accuracy*, and has replaced it on recent European legislation.
Urban boundary layer	The atmospheric boundary layer over an urban area. Its structure, at least in its lowest layers, is modified by the high drag and sensible heat fluxes of the urban area, especially their strong in homogeneity.
Validation (modelling)	Refers to the general comparison of modelled results against monitoring data carried out by the model developers.
Validation (monitoring)	Screening monitoring data by visual examination to check for spurious and unusual measurements (see also Ratification).
Verification (modelling)	Comparison of modelled results versus any local monitoring data at relevant locations.

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Appendix 1

Technical annex to Chapter 2

Emission controls and abatement technologies

1. Section 2.5 gave a brief description of three of the main techniques used to control NO_x emissions from stationary and mobile combustion sources. These were low-NO_x burners for stationary combustion sources and Exhaust Gas Recirculation and Three-Way Catalyst technology for motor vehicles. However, there are a number of other technologies already available or under development that are showing promise for NO_x emission abatement.

2. In this Annexe, more fundamental and design characteristics of the three technologies discussed in Section 2.5 are provided and a description is made of several other important methods for controlling NO_x emissions, at present not widely used in the UK. This is not meant to be an exhaustive list of all methods either available now or under development.

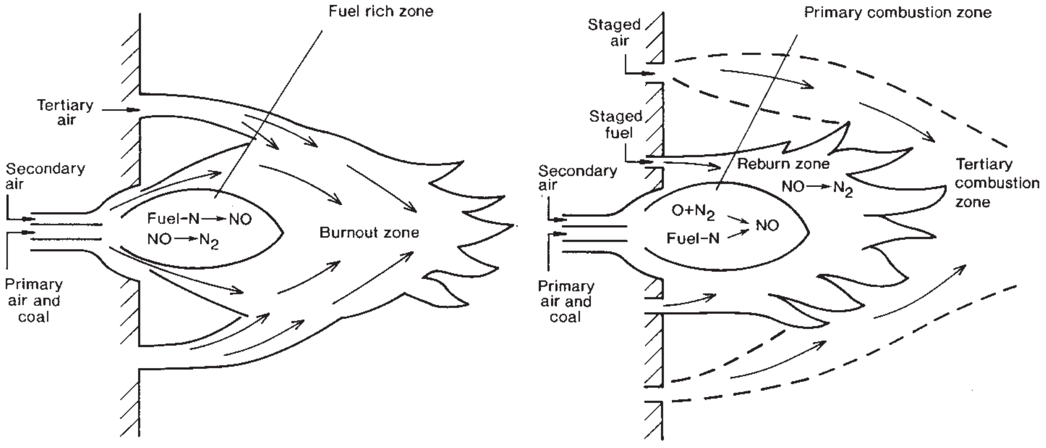
Emission abatement for stationary combustion sources

Low-NO_x burners

3. Low-NO_x burners work on the principle of staged combustion in which the fuel and air are mixed in the burner in stages to create regions of different temperature and fuel/air ratio in the flame. Fuel-rich regions allow the conversion of NO and its precursors to N_2

4. For pulverised coal combustion, there are two main types of low-NO_x burner designs which both involve the use of staged combustion to achieve the desired effect. These are air-staged and fuel-staged burners, shown schematically in Figure A1, or variants of these.

Figure A1.1 Low-NO_x burner concepts for pulverised coal flames.



Air – Staged Burner

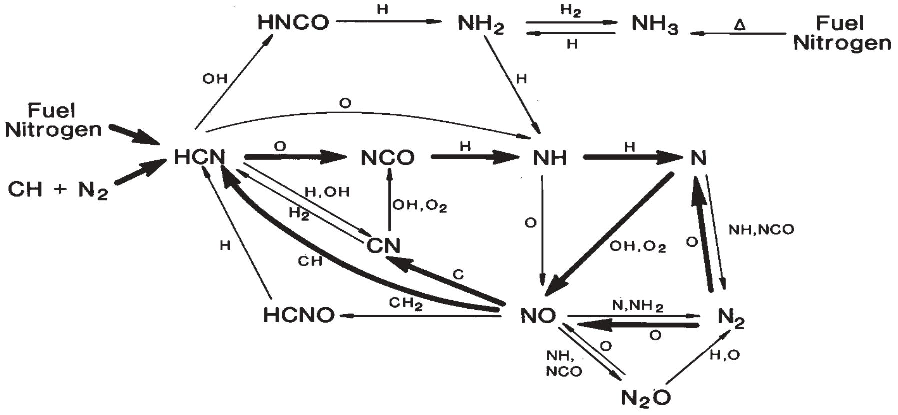
Fuel – Staged Burner

5. In air-staged burners, the first stage of combustion occurs in a slightly fuel-rich zone, at the optimum fuel/air stoichiometric ratio (1.1–1.3) for conversion of fuel-nitrogen to N_2. A second stage of combustion is operated fuel-lean by addition of secondary air in such a way as to complete fuel burnout, with careful temperature control to minimise the formation of thermal-NO in this zone. There are various designs of air-staged burners which differ in the way the two combustion zones are created. The aerodynamics of the fuel-air mixing are key design factors controlling the effectiveness of air-staged burners, ensuring the optimum residence time and temperatures in each stage.

6. In fuel-staged burners, NO is allowed to be formed in the primary combustion zone, but fuel is injected downstream to create a secondary fuel-rich or 'reburn' zone where the NO is reduced to N_2. Further air is added downstream to complete combustion of the excess fuel in a tertiary combustion zone, again with careful control of temperature to minimise thermal-NO formation.

7. Understanding how NO is converted to N_2 in low-NO_x burners requires an understanding of the full cycle of nitrogen chemistry in combustion systems, from formation of NO from N_2 and fuel-bound nitrogen in the first instance. The main steps in the chemical mechanism involved in the formation and removal of NO in combustion is shown in Figure A2.

Figure A1.2 Chemical mechanism for formation and removal of NO in combustion.



8. The key reactions responsible for conversion of NO to N_2 in the fuel-rich zones of both types of low-NO_x burners are those between NO and small hydrocarbon and nitrogen-containing radicals (CH, CH_2, NH, NH_2 and NCO) which are present under these conditions. The reaction sequence is complex, but the main steps are shown in Figure A2. Among the key reactions are:

$$CH_2 + NO \rightleftharpoons HCNO + H$$

$$NH_2 + NO \rightleftharpoons N_2 + H_2O$$

$$NH_2 + NO \rightleftharpoons N_2 + H + OH$$

9. A chemical cycle exists (Figure A2) interconverting NO with species like HCN and other reduced nitrogen species. While these compounds have the potential to continuously regenerate NO, the key point is that each time the chemistry passes through one loop, some of the nitrogen is taken out of the loop as N_2.

10. The effectiveness of these methods for reducing NO_x emissions depends on a number of factors including burner design, operation and coal type. NO_x reductions of between 30 and 60% have been achieved.

Selective catalytic reduction

11. Selective catalytic reduction (SCR) is a well developed and widely used aftertreatment methods for reducing NO_x emissions in the flue gas of a stationary combustor. The process involves the reduction of NO (and NO_2) to N_2 with approximately stoichiometric amounts of ammonia (NH_3) over a catalyst bed. The overall reaction is:

$$4NO + 4NH_3 + O_2 \rightleftharpoons 4N_2 + 6H_2O$$

12. The catalyst effectively reduces the temperature at which this reaction occurs to 350–400°C, but this is still sufficiently high for it to be necessary to install the SCR reactor near the boiler if the flue gas is not to be reheated. This can cause problems if the flue gas is laden with SO_2 and particulate; SO_2 can poison the catalyst and particulates cause clogging.

13. A range of different catalyst materials can be used. Vanadium pentoxide (V_2O_5) supported on titanium dioxide (TiO_3) are one of the most effective and widely used materials as they have a high resistance to SO_2. Other substances exhibiting catalytic activity are oxides of iron and copper and the metals platinum, palladium, rhodium and ruthenium.

14. SCR can achieve 80–90% efficiency in NO_x reduction.

Selective non-catalytic reduction

15. The selective non-catalytic reduction process (SNCR), also known as the thermal-deNO_x process, reduces NO by addition of NH_3 in the flue gas without the use of a catalyst. NH_3 is injected in the high temperature region of the boiler to reduce NO to N_2 thermally by the same overall reaction as occurs in SCR, shown above. The presence of excess oxygen is required and a residence time of 0.1 to 0.4 seconds.

16. A feature of this process is that it is only effective over a narrow temperature window between 1050–1350°C. This can be a problem if the flue gas temperature changes with load. However, the position and width of the temperature window is affected by additives, for example, hydrogen and hydrogen peroxide shift the temperature window down to 900–1220°C.

17. The reaction mechanism is a homogeneous gas-phase one, the key step being one of the reactions important in low-NO_x burner mechanisms:

$$NH_2 + NO \rightleftharpoons N_2 + H_2O$$

$$NH_2 + NO \rightleftharpoons N_2 + H + OH$$

18. The supply of sufficient NH_2 radicals to sustain this reaction and competition with reactions of NH_2 with oxidising radicals leading to the formation of NO explains the existence of the temperature window.

19. The efficiency of NO_x removal by SNCR is ~60%.

20. A variation of the SNCR thermal deNO_x process involves injecting solid cyanuric acid $(HOCN)_3$ into the flue gas. Upon heating, this compound decomposes to gaseous isocyanic acid (HNCO) which itself thermally decomposes to radicals engaging in a sequence of reactions removing NO similar to those in the thermal-deNO_x mechanism. This method is effective over a broader temperature window than the thermal deNO_x process involving NH_3, typically 430–1130°C. Very high efficiencies of NO_x reduction can be achieved (>90%) and this method has been applied to smaller-scale boilers. Urea can also be used as an SNCR reagent.

A disadvantage of these techniques is that fairly significant concentrations of N_2O can be formed as a by-product.

Emission abatement for mobile combustion sources

Exhaust gas recirculation

21. Exhaust Gas Recirculation (EGR) controls NO_x formed in internal combustion engines by recycling a fraction of the exhaust gases through a control valve from the exhaust to the engine where it acts as a diluent, thereby reducing the peak flame temperature and hence the rate of NO formation in the burned gases through the thermal-NO mechanism. Increasing the amount of gas recirculated reduces the rate of NO formation. However, it also reduces the combustion rate making stable combustion more difficult to achieve. Nevertheless, substantial reductions in NO_x emissions (60–80%) can be achieved with 15–25% EGR, about the maximum amount of EGR the engine will tolerate under normal conditions.

22. Reductions in NO_x emissions from EGR can be accompanied by modest increases in CO and hydrocarbon emissions, depending on engine conditions. In diesel engines, EGR can, under certain conditions increase particulate (PM) emissions, but this problem can be resolved by careful engine design.

Three-way catalysts for petrol vehicles

23. As discussed in Section 2.5.2, three-way catalysts have been fitted on all new petrol cars and vans sold in Europe since 1992. A single catalyst bed can reduce NO and oxidise CO and HC if the fuel/air ratio is maintained close to stoichiometric (i.e. neither fuel or oxygen are in excess).

24. There is a narrow range of fuel/air ratios close to the stoichiometric condition in which high conversion efficiencies of all three pollutants are achieved, so an oxygen sensor in the exhaust is used to detect whether the engine is operating on the fuel rich or lean side of stoichiometric and to adjust the fuel system accordingly. In reality, the fuel/air ratio fluctuates around the optimum level, but all three pollutants can still be removed with high efficiency if the appropriate catalyst material is used. Commercial three-way catalyst units use various combinations of the metals platinum (Pt), palladium (Pd) and rhodium (Rd) as the active catalyst materials on various substrates. The reactions on the catalyst surface removing NO are:

$$2NO + 2CO \rightleftharpoons N_2 + 2CO_2$$

$$2NO + 2H_2 \rightleftharpoons N_2 + 2H_2O$$

25. Catalysts are only effective when the temperature is above 250–300°C. This is known as the catalyst light-off temperature. When the vehicle is started with the engine cold, it takes a certain time for the catalyst to warm up to its optimum temperature by the exhaust gases. During this time, emissions are higher than they are when the catalyst is at its optimum temperature. This contributes to the excess emissions during the cold start period of a trip. Recent developments in three-way catalyst technology have concentrated on reducing the time it takes for catalysts to reach the light-off temperature, thus reducing the excess cold start emissions. This has been achieved by modifying the composition of the active catalyst material and changing the thermal capacity of the substrate. For example, higher palladium content reduces the light-off temperature. It has also been achieved by physically moving the catalyst unit closer to the engine or using supplementary heating.

26. Three-way catalysts are susceptible to poisoning by sulphur present in the fuel, significantly reducing its efficiency and durability. This has been the motivation for reducing the sulphur content of fuels from levels of above 300 ppm in petrol in the early 1990s to below 50 ppm today in the UK, levels that meet the European Fuel Directive limits for 2005. This is ultra-low sulphur petrol grade. However, even at this level, the performance of the catalyst can be compromised and consideration is currently being given to implementing directives and policies which set a ceiling of 10 ppm on the sulphur content of fuels – so-called sulphur-free fuels.

27. Reduced catalyst light-off time together with improved catalyst materials, engine design and cleaner fuels are together likely to be the means by which petrol vehicles will meet the stringent Euro IV standards.

DeNO$_x$ systems for diesel vehicles using selective catalytic reduction

28. Selective catalytic reduction systems have potential for reducing emissions of NO$_x$ in exhausts of heavy duty diesel vehicles. These work in much the same way as SCR systems for stationary combustion except they use urea instead of NH$_3$ as the active reagent. A 32% solution of urea in water is stored on the vehicle and injected into the exhaust upstream of the SCR catalyst. Urea is a safe and relatively inexpensive reagent which when heated liberates NH$_3$, initiating the SCR deNO$_x$ process on the catalyst surface at 250–500°C:

$$(NH_2)_2CO + H_2O \rightleftharpoons 2NH_3 + CO_2$$

$$4NO + 4NH_3 + O_2 \rightleftharpoons 4N_2 + 6H_2O$$

29. On-board diagnostics with NO$_x$ or NH$_3$ sensors linked to a dynamic urea injection control system are required to optimise the performance of the SCR system. An oxidation catalyst downstream of the SCR catalyst may be used to eliminate the possibility of NH$_3$ slip.

30. This is still an emerging technology for mobile applications, but tests show it can achieve 80% reduction in NO$_x$ emissions from heavy duty diesel vehicles, allowing them to meet the 2008 Euro IV standards. It requires ultra-low sulphur diesel (<50 ppm) to be used, but the technology is reasonably sulphur-tolerant. An infrastructure for the supply of urea-water solution is a prerequisite for widespread application.

31. SCR systems based on urea combined with Continuously Regenerating Trap (CRT) technology have been developed to simultaneously reduce both PM and NO$_x$ emissions from heavy duty diesel exhausts.

DeNO$_x$ systems using NO$_x$ traps

32. NO$_x$ traps are another exhaust aftertreatment technology which have potential for reducing NO$_x$ emissions from diesel and other lean-burn vehicles. However, they are believed to be more appropriate for light duty diesel vehicles, rather than heavy duty vehicles. The system works by adsorbing NO$_x$ on a catalyst surface in the exhaust during lean cycles at temperatures between 250–450°C. Desorption occurs during periodic, short rich 'spikes' in the exhaust when the NO$_x$ is reduced to N$_2$ in the rich environment and the catalyst trap regenerated. Using sophisticated engine management systems, engine cycles must be adjusted so that periodic rich excursions will regenerate the trap, burning off NO$_x$ at 650°C without producing significant amounts of particulates.

33. The adsorbing material is highly susceptible to sulphur poisoning. To be effective NO_x traps require very low levels of sulphur in the fuel, even below 15 ppm. This is still an emerging technology for mobile applications, but tests show that NO_x emission reductions as high as 80% can be achieved with this technology and with the prospect of very low sulphur fuels coming on the market, NO_x traps will have potential for meeting Euro IV standards.

Alternative fuels

34. A number of alternative fuels for road vehicles have been developed for addressing a range of different environmental and energy supply issues. Vehicles running on compressed natural gas (CNG) and liquefied petroleum gas (LPG) show potential benefits in reducing emissions compared with conventional fuels. However, while many laboratory tests show improvements in terms of NO_x emissions when vehicles are run on LPG or CNG fuels, tests carried out on vehicles while 'on-road' show the benefits are often less obvious. This may in part be due to the limited amount of 'on-road' data available for vehicles running on these fuels, making it difficult to allow thorough comparisons with conventional fuels. Much of the available data are associated with relatively small pilot studies using 'specially maintained' vehicles. More thorough testing is clearly required.

35. Other advanced vehicle technologies will undoubtedly show NO_x emission benefits. Hybrid electric and fuel cell powered vehicles will substantially reduce or eliminate NO_x emissions from vehicles at point of use.

Off-road mobile machinery

36. As well as road vehicles, emissions have recently become regulated for off-road vehicles and mobile machinery which run on diesel engines between 18 and 560 kW power. These cover a range of different machinery which may run on gas oil or road diesel used in industry (for example, forklifts, compressors), construction (for example, cranes, bulldozers), agriculture (for example, tractors, combines) and airport support vehicles. Directives 97/68/EC and 2000/25/EC set limits of NO_x and other pollutant emissions according to engine rating introduced in two stages between 1998 and 2003.

37. Many of the abatement methods discussed above for road vehicles will be applicable to off-road machinery, provided the quality of the fuel used is adequate, for example, ultra-low sulphur grade diesel may be necessary for some technologies.

UK NO_x emission projections for road transport

38. Section 2.12.1 discussed the NAEI emission projections for road transport. Tables A1 and A2 in this Annexe provide figures for UK and urban UK road transport emissions of NO_x by vehicle type. These are based on the new vehicle emission factors discussed in Section 2.7.2.1 and the figures are projections from 2000 and consistent with the plots in Figure 2.13.

Table A1.1 UK road transport emissions of NO_x.

ktonnes		1990	1995	2000	2005	2010	2015	2020	2025
Cars	Petrol	752.5	579.6	383.1	223.9	146.1	114.1	114.3	118.3
	DERV	7.1	19.0	31.1	42.8	37.5	35.7	35.3	36.5
	All Cars	759.6	598.7	414.2	266.7	183.7	149.8	149.6	154.8
LGV	Petrol	52.8	36.7	16.3	4.7	2.1	1.7	1.8	2.0
	DERV	20.4	36.0	51.0	53.4	37.7	31.5	32.1	35.2
	All LGV	73.2	72.7	67.3	58.1	39.8	33.3	33.9	37.2
HGV	Artic	194.9	188.9	205.9	160.6	108.1	76.2	74.3	81.7
	Rigid	187.8	138.9	107.7	83.0	54.8	36.0	30.4	31.1
	ALL HGV	382.7	327.7	313.6	243.6	162.9	112.2	104.6	112.8
Buses		69.6	63.0	48.3	40.7	30.7	21.9	17.4	15.8
Motorcycles		0.8	0.7	0.9	1.0	1.1	1.2	1.2	1.3
All DERV		479.8	445.8	444.0	380.5	268.8	201.3	189.5	200.3
All Petrol		806.2	617.1	400.3	229.6	149.3	117.0	117.4	121.6
All Vehicles		**1286.0**	**1062.8**	**844.3**	**610.0**	**418.1**	**318.3**	**306.8**	**321.9**

Table A1.2 Urban UK road transport emissions of NO_x.

ktonnes		1990	1995	2000	2005	2010	2015	2020	2025
Cars	Petrol	270.0	222.6	165.5	116.3	74.6	53.8	52.9	54.7
	DERV	3.3	9.0	13.3	18.1	15.2	14.3	14.1	14.5
	All Cars	273.3	231.6	178.8	134.4	89.8	68.1	67.0	69.2
LGV	Petrol	20.1	13.8	6.4	2.4	1.2	1.0	1.0	1.1
	DERV	9.1	15.7	20.7	21.3	14.9	12.2	12.2	13.2
	All LGV	29.2	29.5	27.0	23.7	16.1	13.2	13.3	14.3
HGV	Artic	24.2	21.4	18.3	14.4	10.7	7.4	7.0	7.5
	Rigid	58.5	45.4	30.3	27.2	17.9	11.9	10.1	10.4
	ALL HGV	82.7	66.8	48.6	41.6	28.6	19.3	17.1	18.0
Buses		41.6	45.3	34.6	30.8	22.3	15.9	12.6	11.5
Motorcycles		0.3	0.3	0.3	0.3	0.4	0.4	0.4	0.5
All DERV		136.7	136.8	117.2	111.9	80.9	61.7	56.1	57.2
All Petrol		290.4	236.6	172.2	119.0	76.2	55.2	54.4	56.3
All Vehicles		**427.2**	**373.4**	**289.4**	**230.9**	**157.1**	**116.9**	**110.5**	**113.5**

Appendix 2

Factors affecting diffusion tube performance

1. This appendix presents a review of the possible factors affecting the performance of nitrogen dioxide diffusion tubes. Although diffusion tubes are, and are likely to remain, important for general assessment of NO_2 air quality, this AQEG report utilises data obtained almost exclusively from chemiluminescence analysers, rather than diffusion tubes, for its analyses of NO_2 in the UK. Therefore this report is not the appropriate place for an exhaustive review of the literature.

2. The following factors are reviewed together with potential explanatory mechanisms for their effects on diffusion tube performance:

- the laboratory preparing and analysing the tubes;
- the exposure interval – weekly, 2-weekly or monthly;
- the time of year;
- the exposure setting – sheltered or exposed;
- the exposure location – roadside or background;
- the tube preparation method; and
- the exposure concentration and NO_2/NO_x ratio.

It is important to recognise that the effect of one factor on a diffusion tube measurement may act to obscure or offset the effects of other factors acting on the same measurement. This complicates attempts at comparing data from different studies, and of interpretation of results.

Role of laboratory

3. Early studies involving monthly exposure at a number of sites around the UK of tubes prepared with 50% TEA-in-acetone found an average diffusion tube bias of +26%, although this varied from site to site (Campbell *et al.*, 1994). Bush *et al.* (2001a) carried out a more extensive validation study over a full year in 1996/97 using tubes also prepared and analysed by Harwell Laboratory using 50% TEA-in-acetone. This study identified an overall positive diffusion tube bias of +8% for monthly exposure. Heal *et al.* (1999), using tubes prepared and analysed in their own laboratory with 50% TEA-in-acetone, found an average over-estimation of +1% for tubes exposed monthly over a 4 month summer period in Edinburgh. However it is not a universal picture that diffusion tubes over-estimate concentrations. For a number of years AEA Technology carried out an annual one-month intercomparison exercise using tubes prepared and analysed by a wide range of UK laboratories. Tubes were exposed at an urban background site (Walsall) alongside an automatic monitor. These exercises have shown that diffusion tubes exposed monthly can both under- and over-estimate concentrations by up to ±30% or more. The indications were of a consistency in the performance of a particular laboratory from year to year. A more recent study by Laxen & Wilson (2002) has examined data from collocation studies carried out by 23 local authorities, giving 44 site-years worth of data. This has confirmed that diffusion tubes both over- and under-estimate concentrations and that the most significant factor affecting tube performance is the laboratory preparing and analysing the tubes. The reasons for the differences between the laboratories are currently not understood. No criticism of the laboratories should be implied as there is no carefully defined procedure to follow. In practical terms, what matters is that laboratories are consistent in their performance. This may allow a diffusion tube bias to be determined and applied to the results from that laboratory.

Role of exposure interval

4. Bush *et al.* (2001a) cite unpublished evidence from the 1990s that tubes exposed for four week periods produced lower concentrations than those exposed for one week, especially during summer months. The study by Heal *et al.* (1999) during a 4 month summer period in Edinburgh also found tubes exposed for four-weeks to give much lower concentrations than those exposed for one-week intervals in parallel (ratios to a chemiluminescence analyser of 1.01 and 1.26, for four and one week exposures, respectively). In a separate study over 11 months, Heal *et al.* (2000) found average ratios between diffusion tube and chemiluminescence analyser of 1.06, 1.15 and 1.24 for exposure intervals of 4 weeks, 2 weeks and 1 week, respectively. On the other hand, the national collocation study carried out by Bush *et al.* (2001a) over a full year found only a very small enhancement for 2-week exposures compared with 4-week exposures. The recent examination of collocation studies by Laxen & Wilson (2002) found indicative evidence of a significant effect of exposure interval, but this was not based on results for tubes supplied and analysed by the same laboratory, so must be treated with some caution. Overall, the evidence points to the exposure interval affecting performance, with concentrations being lower for longer exposure intervals. Heal *et al.* (2000) postulated some, as yet unidentified, exposure-interval dependent loss of nitrite at the adsorbent to explain the decrease in effective uptake of NO_2 with exposure interval. They argued that this loss factor, acting in the opposite direction to a positive bias on diffusion tube performance from within-tube chemistry (see section on Mechanisms), explained the effective closer agreement between their diffusion tube measurements and continuous analyser as exposure interval was increased.

Role of time of year

5. Heal *et al.* (1999) have argued on a theoretical basis, that chemistry taking place within the diffusion tubes (see section on Mechanisms), means that they will over-estimate by a greater amount in the summer at a polluted site, but that there should be little or no overestimation, and consequently no seasonal effect in such a bias, at a low pollution site. A similar theoretical study by Bush *et al.* (2001b) also predicted that tubes would over-estimate by a greater amount in summer than winter, although they concluded this would occur at all types of site, not just polluted sites. Kirby *et al.* (2001) report results of a 12-month survey using 10% TEA-in-water tubes at a site in Cambridge that showed greater over-estimate in summer than winter, +18% *cf.* +13%. In a separate study, also in Cambridge, they reported a summer (March-August) over-estimate of +7% and a winter (December-March) under-estimate of -6%. Tubes were exposed for 2-week intervals in both cases. Bush *et al.* (2001a) on the other hand reported no seasonal difference for 50% TEA-in-acetone tubes exposed monthly at 17 sites across the UK. Laxen & Wilson (2002) have analysed data for tubes prepared by a single laboratory using two different methods, 50% TEA-in-acetone and 50% TEA-in-water, and exposed alongside a chemiluminescence monitor for 1-month at a time over an annual period, at 5 and 6 different locations respectively. The results show no evidence of a seasonal pattern for tubes prepared with 50% TEA-in-acetone (Figure A1). In the case of tubes prepared by the same laboratory using 50% TEA-in-water there is evidence of higher diffusion tube/chemiluminesence ratios during the four months August, September, October and November (Figure A2). A similar pattern was found in the results for 10% TEA-in-water tubes prepared and analysed by a different laboratory, based on 12 site-years of data. In this case the diffusion tube/chemiluminesence ratios were higher in August, September and October (Laxen & Wilson, 2002). Heal *et al.* (2000) present diffusion tube and collocated chemiluminesence data for an 11-month period for tubes prepared with 10% TEA-in-water exposed at a city centre site in Cambridge. The diffusion tube/chemiluminesence ratios were significantly higher in the period mid-September to mid-November (no data were available for August and December). There is thus evidence for a dependency upon time of year (but not a simple summer/winter split) for tubes prepared with TEA-in-water, but not for tubes prepared with 50% TEA-in-acetone. There is currently no explanation for this pattern associated with preparation method.

Figure A2.1 Ratio of diffusion tube to chemiluminescence for tubes prepared and analysed by one laboratory using 50% TEA-in-acetone. Box and whisker plots for each month for 5 site-years of data. (Laxen & Wilson, 2002).

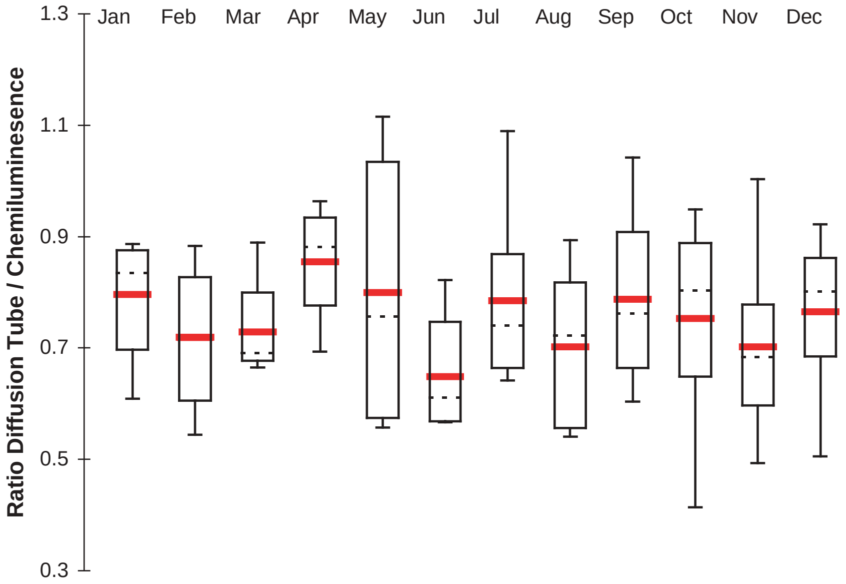
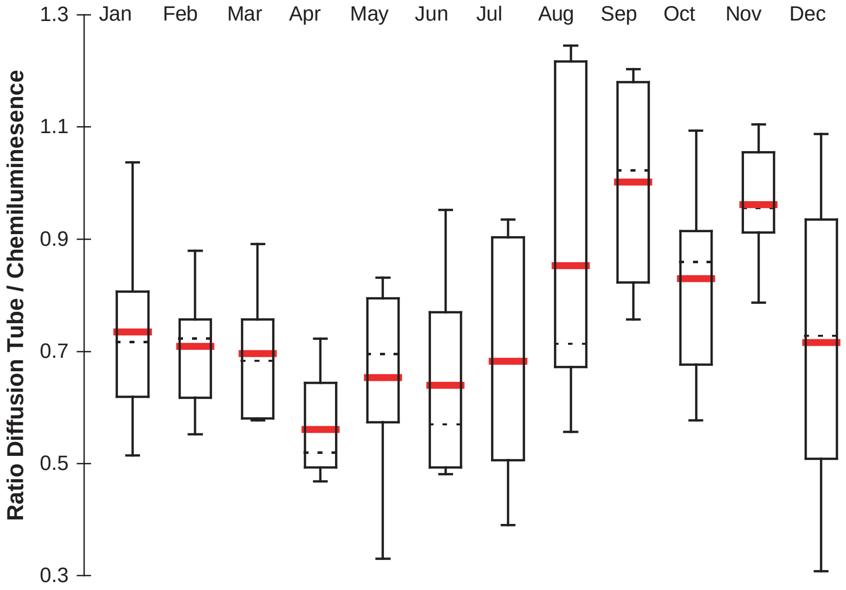


Figure A2.2 Ratio of diffusion tube to chemiluminesence for tubes prepared and analysed by one laboratory using 50% TEA-in-water. Box and whisker plots for each month for 6 site-years of data. (Laxen & Wilson, 2002).



Role of exposure setting

6. Bush *et al.* (2001a) found unsheltered tubes, exposed monthly for a full year at 17 locations around the UK, over-estimated concentrations by +10%, while tubes sheltered beneath an inverted bucket under-estimated concentrations by -10%. Kirby *et al.* (2001) found a similar behaviour, with unsheltered tubes at a location in Cambridge over-estimating by +16%, while the sheltered tube beneath an inverted bucket over-estimated by +3%. This is an artificial situation, as tubes are not routinely exposed in this way, but Campbell *et al.* (1994) report a study that suggests tubes in an exposed location on a roof top will over-estimate by up to +40% due to wind induced turbulence in the mouth of the diffusion tube, compared with tubes in a more sheltered location near to a wall (see below under Mechanisms).

Role of exposure location – roadside/background

7. Heal *et al.* (1999), in a study using 50% TEA-in-acetone tubes exposed for one-week in Edinburgh, found that 2 roadside sites over-estimated by +50 and +37%, while an urban background site over-estimated by +13%. Kirby *et al.* (2001) report a +14% over-estimate for tubes exposed for 2-week intervals at an urban background (Norwich) site, but a lower (+4%) over-estimate at a roadside (Marylebone Road, London) site. The difference between the two studies may be associated with differences in concentrations of NO_x, NO_2 and O_3 at the various sites, with a resultant impact on the photochemical equilibrium within the tubes (see below under Mechanisms). The examination by Laxen & Wilson (2002) of results obtained by a number of local authorities found that the performance of tubes exposed for a month at a time at roadside sites fell within the range of the performance at background sites (comparison based on tubes supplied and analysed by the same laboratory and prepared in the same way). The evidence for a consistent difference between roadside and background monitoring location is therefore equivocal.

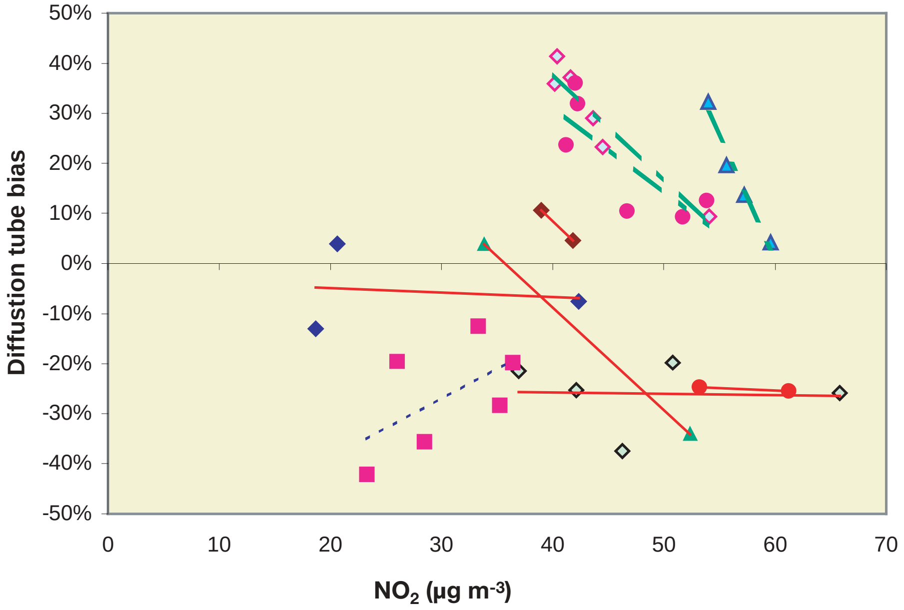
Role of tube preparation method

8. The comparative study by Kirby *et al.* (2000) using 2-week exposure intervals over 5 months showed similar performances for tubes prepared using 50% TEA-in-acetone, 10% and 20% TEA-in-water, but tubes prepared with 50% TEA-in-water gave lower concentrations. AEA Technology (2002), in a study over one month (October), compared tubes prepared by three different laboratories using 50% TEA-in-acetone, 20% and 50% TEA-in-water. As with Kirby *et al.* (2000) 50% TEA-in-acetone tubes gave the highest concentrations, with 50% TEA-in-water tubes giving the lowest concentrations. However, unlike the Kirby *et al.* (2000) study, 20% TEA-in-water tubes gave intermediate values. In contrast, the data compiled by Laxen & Wilson (2002) for monthly exposure of tubes prepared by one laboratory using 50% TEA-in-water (six sites) and 50% TEA-in-acetone (5 sites), over a full year, showed no difference in the performance. The method of tube preparation does not therefore affect performance in a consistent way.

Role of exposure concentration and NO_2/NO_x ratio

9. Atkins *et al.* (1986) found no dependency of the performance of 50% TEA-in-acetone tubes on NO_2 concentration when exposed weekly to concentrations over the range 10–110 µg m^{-3}. In contrast, Kirby *et al.* (2001) found that the performance of 10% TEA-in-water tubes was strongly related to the total NO_2 concentration. Tubes exposed to 1-week or 2-week NO_2 concentrations (chemiluminescence) of around 55 µg m^{-3} under-estimated by around 0 to -10%, while tubes exposed to concentrations of around 25-30 µg m^{-3} over-estimated by around +30 to +45%. Heal *et al.* (1999) reported similar behaviour for 50% TEA-in-acetone tubes exposed weekly to NO_2 concentrations in the range 30–65 µg m^{-3}. Laxen & Wilson (2002) found a more confusing picture for annual mean performance of tubes prepared in different ways and exposed for 1-week, 2-week or monthly time intervals (Figure A3). Some of the variability may follow from the relationships also observed between 1-week tube performance and NO_2:NO_x, NO:NO_2 and O_3:NO_2 ratios and explained by a within-tube chemistry mechanism (Heal *et al.*, 1999, Heal *et al.*, 2000, Kirby *et al.*, 2001). If ambient concentrations of NO_x, NO_2 and O_3 are affecting tube performance, then it will clearly be in a complex way.

Figure A2.3 Diffusion tube bias vs annual mean concentration (automatic monitor). Data grouped by laboratory and tube preparation method. Lines are linear best-fit relationships: solid lines for 50% TEA in water, monthly exposure; small dashed line for 50% TEA in water, monthly exposure; and small/large dashed lines for 10% TEA in water, 1-week and 2-week exposure. (Laxen & Wilson, 2002).



Mechanisms

10. The two principal mechanisms proposed to account for overestimation by diffusion tubes are (i) a shortening of the diffusion length in the tube due to turbulence in the mouth of the tube brought about by higher windspeeds, and (ii) reaction within the tube between NO and O_3 that are not at photostationary equilibrium in the ambient air. Even when the ambient NO and O_3 are at photostationary equilibrium, additional NO_2 can be generated within the tube since the photolytic component of the NO-NO_2-O_3 equilibrium is switched off within the tube. To date no convincing mechanism has been put forward to account for under-estimation of concentrations, although (as mentioned above in Role of Exposure Interval) Heal *et al.* (1999, 2000) postulate some form of light-dependent loss of the nitrite from the adsorbent.

11. The issue of a wind induced shortening of the diffusion length was first examined by Atkins *et al.* (1986). They postulated that there should be greater turbulence in the mouth of the tube at higher windspeeds, giving rise to a shorter diffusion length and hence higher 'apparent' concentrations. However they found no relationship between windspeed and tube performance, suggesting this mechanism is not significant. Campbell *et al.* (1994), on the other hand, concluded that over-estimation by diffusion tubes at some, but not all, sites in a UK-wide study in 1990/91, was likely to be due to wind-driven shortening of the diffusion length. They supported this with a study using tubes of different length in an exposed roof-top location and in a more sheltered location by a building wall. This showed that the sheltered tubes experienced no reduction in diffusion length, while the exposed tubes showed a 2 cm reduction in diffusion length. For the standard 7.1 cm tube this would translate to a 40% increase in concentration. A similar study by Gair & Penkett (1995) also showed a constant shortening of the diffusion length, in their case 2.3 cm, for tubes of 4 to 14 cm length exposed on a roof. However, they found only a weak relationship between average windspeed over the exposure period and the shortening of the diffusion length. They concluded that for the standard diffusion tube the over-estimation would be 25% for their short period study at a rooftop location. Subsequent studies by Kirby *et al.* (2001) confirmed the findings of Atkins *et al.* (1986), finding no correlation between performance and windspeed. They did though find a reduced uptake for sheltered tubes, as did Bush *et al.* (2001a). The picture is thus somewhat confusing. There does seem to be evidence to support the view that turbulence in the mouth of the tube shortens the diffusion length, thus increasing the apparent concentration, but this effect is clearly not related to average windspeed over the sample period.

12. The potential for photolytic enhancement of concentrations was addressed when diffusion tubes were first proposed. Atkins *et al.*, (1986) used a chemical model to show that the reduced photolysis within the tube could lead to over-estimation of 11% during daylight hours in a low pollution area and 6% over-estimation in a polluted area. Taking account of no over-estimation during night-time the overall over-estimation would be substantially less. The mechanism postulated involves the photolytic breakdown of NO_2 being switched off within the tubes, as the acrylic walls do not transmit the necessary UV energy. This allows the NO within the tube to react with O_3 to create additional NO_2, without the photolytic back reaction occurring. There is sufficient time for this reaction, as the transport time for the diffusing species within the diffusion tube is of the order of 2 minutes, which is similar to the NO + O_3 reaction time (Heal & Cape, 1997). Clearly such an effect will be dependent on the ambient concentrations of NO and O_3. Heal & Cape (1997) have carried out detailed modelling of the chemistry within tubes, and concluded that this would produce an overall over-estimate of NO_2 concentrations by 14% in winter and 28% in summer at an urban location, and 8-9% at a rural location in both summer and winter. The over-estimation is greater than suggested by Atkins *et al.* (1986) and the low pollution – high pollution pattern is different. Bush *et al.* (2001b) have carried out further modelling, which shows the greatest over-estimation will occur when the annual mean NO_x lies between 60-100 µg m^{-3}. Their modelling suggests that the over-estimation would range from a maximum of around 7% in winter to 25% in summer. At high concentration, such as might be found close to busy roads (>200 µg m^{-3} NO_x), the over-estimation is predicted to be negligible, at <5%. Measurements at different times of year and at different types of location have been used by these authors to support the theoretical behaviour, but the full set of data analysed above do not support the chemistry mechanism fully. For instance, the apparent lack of seasonal pattern in the performance of tubes made up with 50% TEA-in-acetone is not consistent with the predictions of the chemistry model, neither is the odd behaviour of tubes made up with TEA-in-water, which produce higher concentrations in the months August to November, while the peak photolytic season, runs from May to August.

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Appendix 3

Definition of site classes

Kerbside (U1)

A site sampling within 1 m of the edge of a busy road.
Source influences: Local traffic
Examples of objectives:
- Identifying vehicle pollution blackspots
- Assessing worst case scenarios
- Evaluating impacts of vehicle emission control technologies
- Determining impacts of traffic planning/calming schemes.

Roadside (U2)

A site sampling between 1 m of the kerbside of a busy road and the back of the pavement. Typically this will be within 5 m of the road, but could be up to 15 m.
Source influences: Local traffic
Examples of objectives:
- Assessing worst case population exposure
- Evaluating impacts of vehicle emission controls
- Determining impacts of traffic planning/calming schemes.

Urban centre (U3)

A non-kerbside site, located in an area representative of typical population exposure in town or city centres (for example, pedestrian precincts and shopping areas). This is likely to be strongly influenced by vehicle emissions, as well as other general urban sources of pollution. Sampling at or near breathing-zone heights will be applicable.
Source influences: Vehicle, commercial, space heating
Examples of objectives:
- Identification of long-term urban trends.

Urban background (U4)

An urban location distanced from sources and therefore broadly representative of city-wide background conditions, for example, elevated locations, parks and urban residential areas.
Source influences: Vehicle, commercial, space heating
Examples of objectives:
- Trend analysis
- Urban planning
- Traffic and land-use planning.

Urban industrial (U5)

An area where industrial sources make an important contribution to the total pollution burden.

Source influences:	Industrial, motor vehicles

Examples of objectives:

Assessing local impacts on health and amenity
Process optimisation
Source attribution/identification
Providing model input data
Model development/validation
Local planning and plant authorisation.

Suburban (SU)

A location type situated in a residential area on the outskirts of a town or city.

Source influences:	Traffic, commercial, space heating, regional transport, urban plume downwind of a city

Examples of objectives

Traffic and land-use planning
Investigating urban plumes.

Rural (R1)

An open country location, in an area of low population density, distanced as far as possible from roads, populated and industrial areas.

Source influences:	Regional long-range transport, urban plume

Examples of objectives

Ecosystem impact studies
Assessing compliance with critical loads and levels for crops and vegetation
Investigating regional and long-range transport
Identification of O_3 ‘hot spots’.

Remote (R2)

A site in open country, located in an isolated rural area, experiencing regional background pollutant concentrations for much of the time.

Source influences:	Regional/hemispheric background

Examples of objectives

Assessing ‘unpolluted’ global or hemispheric background conditions
Long-range transport studies
Long-term baseline trend analysis.

Special (Sp)

A special source-oriented category covering monitoring studies undertaken in relation to specific emission sources such as power stations, petrol stations, car parks or airports.

Appendix 4

Lists of sites exceeding threshold concentrations

Table A4.1 Roadside and kerbside sites with measured annual mean NO_2 concentration in 2001 exceeding 40 µg m^{-3} and 'year factor' projection of concentration for 2005 and 2010 (µg m^{-3})

Site name	Site type	2001	2005	2010
Redbridge 2 – Ilford Broadway	Roadside	120	107	88
Kens and Chelsea – King's Rd	Roadside	86	76	63
London Marylebone Road	Kerbside	84	75	62
Kens and Chelsea – Knightsbridge	Kerbside	83	74	61
London Cromwell Road 2	Roadside	76	68	56
Camden – Shaftesbury Avenue	Roadside	75	67	55
Glasgow Kerbside	Kerbside	71	63	52
Hammersmith Broadway	Roadside	70	62	51
Bury Roadside	Roadside	69	62	51
Tower Hamlets Roadside	Roadside	69	62	51
Croydon 5 – Norbury	Roadside	69	61	50
Islington – Holloway Road	Roadside	67	60	49
Camden Kerbside	Kerbside	66	59	48
Brent – Ikea	Roadside	65	58	48
Redbridge 3 – Fullwell Cross	Roadside	65	58	48
Southwark Roadside	Roadside	65	58	48
Dartford Roadside – St Clements	Kerbside	62	55	45
London Bromley	Roadside	61	54	45
Haringey 3 – Bounds Green	Roadside	61	54	44
Oxford Centre	Roadside	60	54	44
Salford M60	Roadside	58	52	43
Lambeth – Christchurch Road	Roadside	58	52	42
Barnet – Tally Ho Corner	Kerbside	58	51	42
Bath Roadside	Roadside	57	51	42
Bristol Old Market	Roadside	57	51	42
Croydon 4 – George Street	Roadside	56	50	41
Hounslow 4 – Chiswick High Rd	Roadside	56	50	41

Site name	Site type	2001	2005	2010
Greenwich 5 – Trafalgar Road	Roadside	55	49	40
Ealing 2 – Acton Town Hall	Roadside	54	48	40
Hounslow Roadside	Roadside	54	48	40
London A3 Roadside	Roadside	54	48	40
Newham Cam Road	Roadside	54	48	40
Wandsworth 4 – High Street	Roadside	53	47	39
Richmond Mobile 6	Roadside	51	46	38
Gravesham Roadside – A2	Roadside	51	46	38
Maidstone Roadside – Fairmeadow	Kerbside	51	46	38
Crystal Palace	Roadside	51	45	37
Ealing Mobile – Southall	Roadside	51	45	37
A2 Falconwood	Roadside	51	45	37
Enfield 4 – Derby Road, Upper Edmonton	Roadside	50	45	37
Broxbourne Roadside	Roadside	50	44	37
Tonbridge Roadside – Town Centre	Roadside	49	44	36
Haringey Roadside	Roadside	48	43	35
Redbridge 4 – Gardner Close	Roadside	48	43	35
Cambridge Parker Street	Roadside	47	42	34
Cambridge Silver Street	Roadside	46	41	34
Watford Roadside	Roadside	46	41	33
Richmond Mobile 5	Roadside	45	40	33
Croydon 2 – Purley Way, 5 Ways	Roadside	45	40	33
Hillingdon – South Ruislip	Roadside	45	40	33
Stert St (Abingdon)	Roadside	45	40	33
Richmond – Castlenau	Roadside	44	39	32
M4	Kerbside	44	39	32
E. Herts Roadside (Sawbridgeworth)	Roadside	44	39	32
Sutton Roadside	Roadside	44	39	32
Enfield 2 – Church Street	Roadside	42	38	31
Havering 3 – Romford	Roadside	42	37	31
Exeter Roadside	Roadside	41	37	30

Table A4.2 Background sites with measured annual mean NO_2 concentration in 2001 exceeding 40 µg m^{-3} and 'year factor' projection of concentration for 2005 and 2010 (µg m^{-3})

Site name	Site type	2001	2005	2010
Brent – Harlesden	Urban background	76	69	59
Corp. of London – Senator House	Urban background	60	54	46
London Southwark	Urban centre	55	50	43
Heathrow LHR2	Airport	54	49	42
London Wandsworth	Urban centre	53	48	41
West London	Urban background	52	47	40
London Bloomsbury	Urban centre	51	46	40
Islington – Upper Street	Urban background	50	45	39
London Hackney	Urban centre	48	44	37
Manchester Town Hall	Urban background	47	43	37
Tower Hamlets – Bethnal Green	Urban background	47	42	36
London Hillingdon	Suburban	46	42	36
Glasgow City Chambers	Urban background	46	42	36
Newham Tant Avenue	Urban background	46	42	36
Sheffield Tinsley	Urban industrial	45	41	35
Tower Hamlets 1 – Poplar	Urban background	45	41	35
Bradford Centre	Urban centre	44	40	34
Waltham Forest – Dawlish Road	Urban background	43	39	34
Edinburgh Centre	Urban centre	43	39	33
London N. Kensington	Urban background	42	38	33
Walsall Alumwell	Urban background	42	38	33
Salford Eccles	Urban industrial	42	38	33
Hounslow 2 – Cranford	Suburban	41	37	32
Barnet – Finchley	Urban background	40	37	31
Barnet – Strawberry Vale	Urban background	40	36	31

Table A4.3 Sites with more than 18 exceedences of 200 µg m^{-3} hourly mean NO_2 concentration in 2001 (number of hours above 200 µg m^{-3})

Site name	Site type	
Redbridge 2 – Ilford Broadway	Roadside	826
Bristol Old Market	Roadside	98
Kens and Chelsea – Knightsbridge	Kerbside	97
Trafford	Urban background	63
London Marylebone Road	Kerbside	60
Glasgow Kerbside	Kerbside	54
Stockport Cheadle	Urban background	45
Stockport Bredbury	Urban background	34
Dartford Roadside – St Clements	Kerbside	25
Brent – Ikea	Roadside	24
Stockport	Urban background	22
Redbridge 3 – Fullwell Cross	Roadside	19
Brent – Harlesden	Urban	19

Table A4.4 Sites with 3-hour mean NO_2 concentration greater than 400 µg m^{-3} in 2001

Site name	Site type
Stockport Bredbury	Urban background

Appendix 5

Respondents to the draft Nitrogen Dioxide in the United Kingdom Report

Comments were gratefully received from the following organisations and/or individuals on the draft of this report, which was published in May 2003:

Air Monitors Ltd

Mr Robert Appleby

BAA Heathrow

British Airways

Dr Simon Watts

Energy Saving Trust

Environment Agency

Greater London Authority

Highways Agency

Professor Stephen Holgate

Innogy plc

Johnson Matthey

Dr David Muir

Natural Environment Research Council (NERC)

nsca

Kirby Associates Limited

Scottish Environment Protection Agency

Welsh Air Quality Forum

Nitrogen Dioxide in the United Kingdom

PB 9025A

Correction

Within the Air Quality Expert Group report *Nitrogen Dioxide in the United Kingdom* Table 7.3, Annual average NO_x and NO_2 concentration predicted by dispersion modelling, was printed with incorrect column headings. Four main column headings above the data should appear as follows: ‘Measured 1999’, ‘Modelled 1999’, ‘Modelled 2005’ and ‘Modelled 2010’. Beneath these main headings subheadings that read ‘NO_x’ and ‘NO_2’ should appear, repeated four times, above the columns of numbers. The column headings to the left – ‘Site’ and ‘Type’ – should not be there. Table 7.3 appears correctly in the web version of this document, which is available at: http://www.defra.gov.uk/environment/airquality/aqeg/nitrogen-dioxide/index.htm