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

[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.
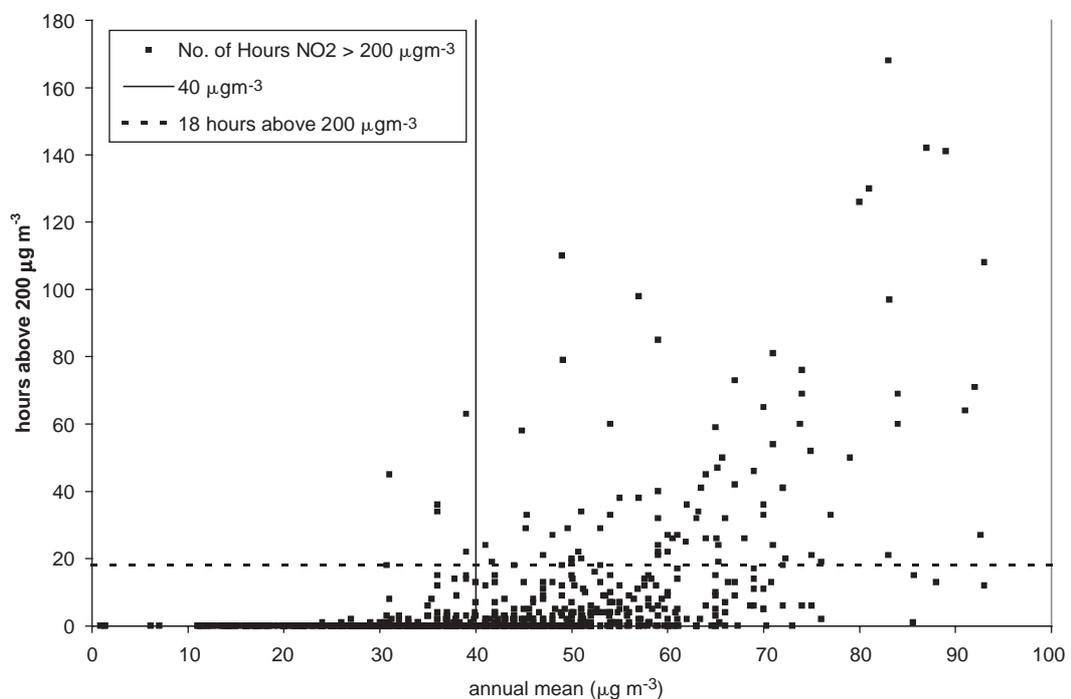
618. There was one measured exceedence of the EU alert threshold for NO$_2$ of a 3-hour mean concentration greater than 400 μg m^{-3} during 2001. This was at the Stockport Bredbury site. Since this site is not part of the UK national networks designated for reporting to the European commission, this 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 ($\mu g\ m^{-3}$).
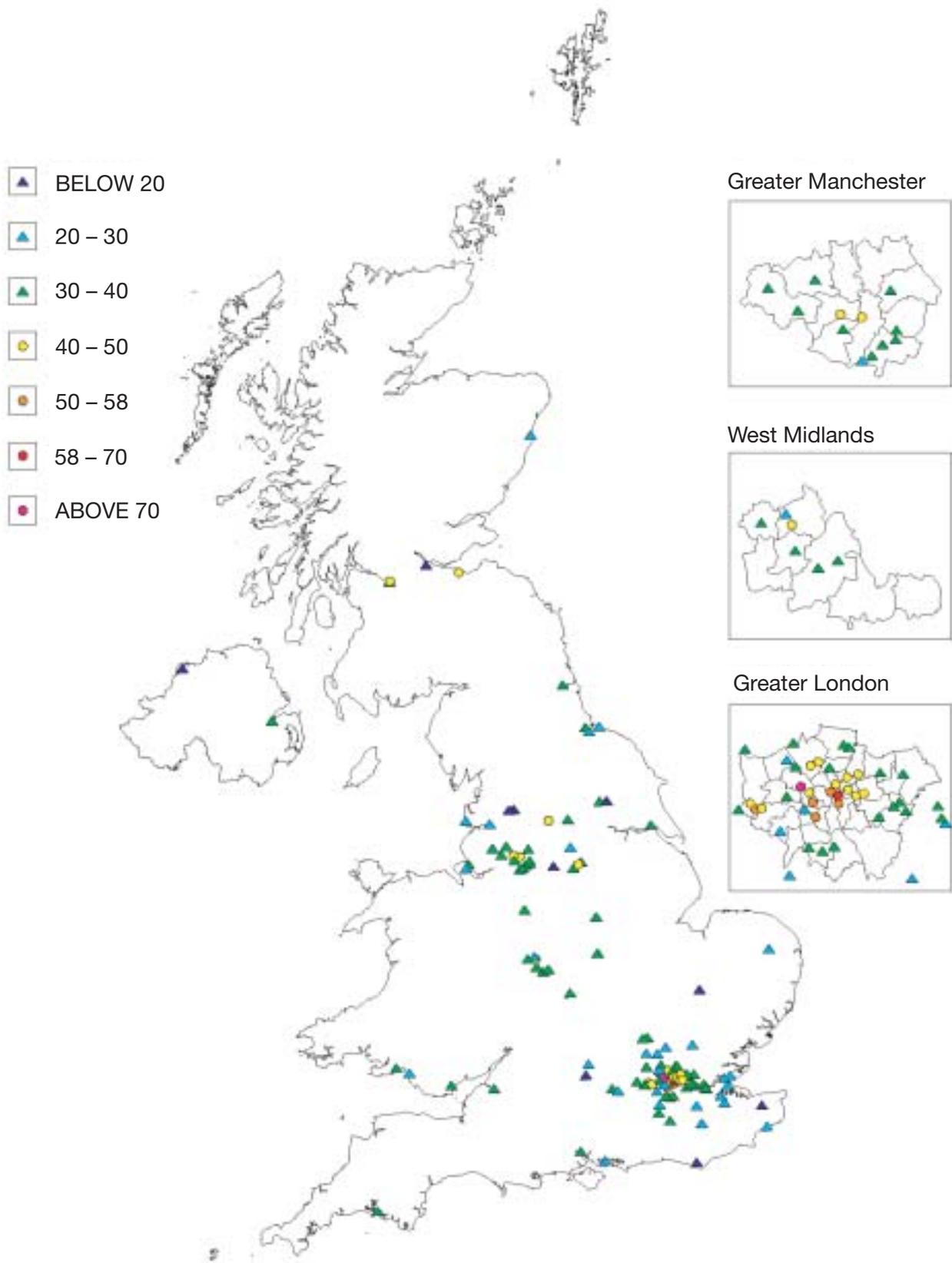
Figure 6.3 Measured annual mean NO$_2$ concentrations in 2001 at roadside and kerbside sites (μg m^{-3}).
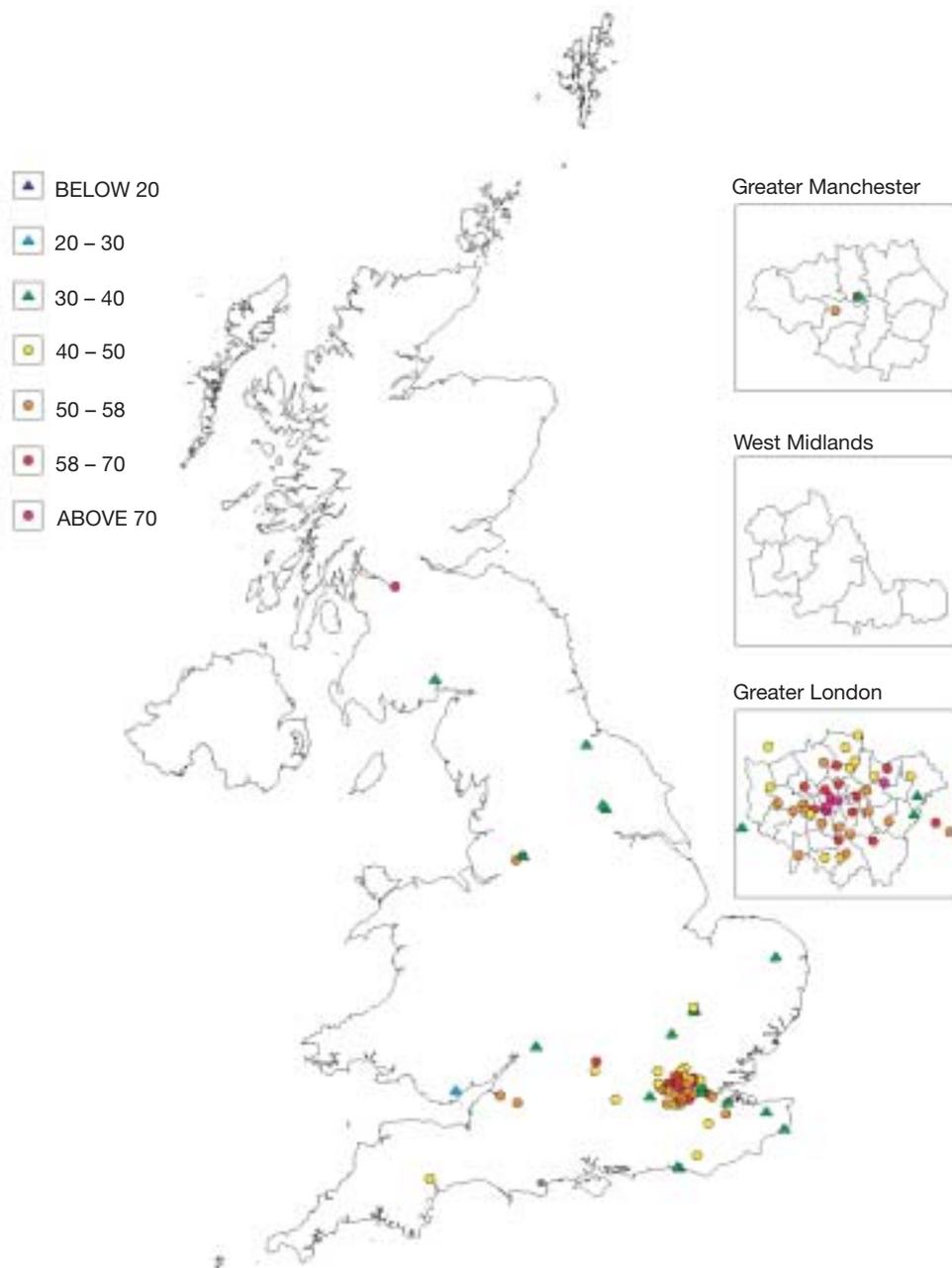
6.2.2 The sites selected for detailed analysis

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.
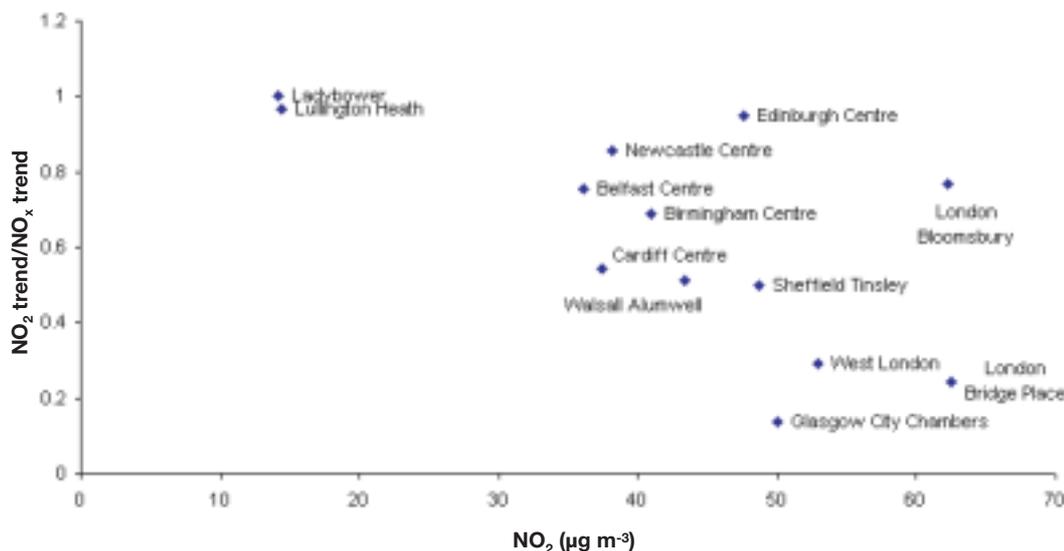
626. Figure 6.4 plots out the ratios of the observed trends in annual mean NO_2 concentrations to those in NO_x against the observed 10-year mean NO_2 concentrations at each of the sites. The trends in NO_2 relative to those in NO_x, decline with increasing average NO_2 level. The rural sites clearly 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.
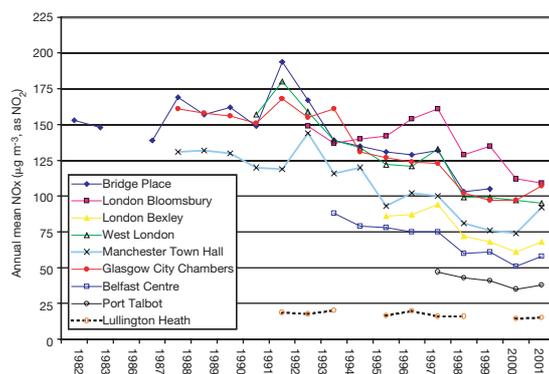
Figure 6.6 Annual mean NO_2 concentrations at selected background sites.
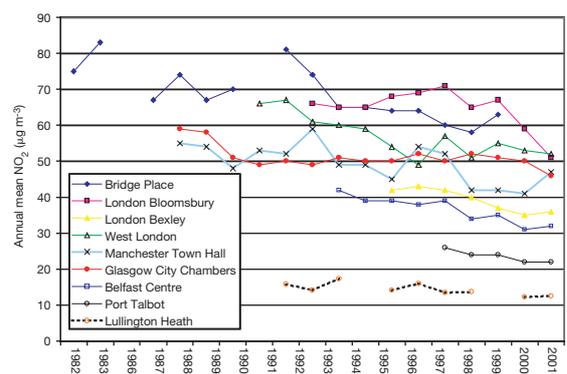
Figure 6.7 Annual mean NO$_x$ concentrations at selected background sites.
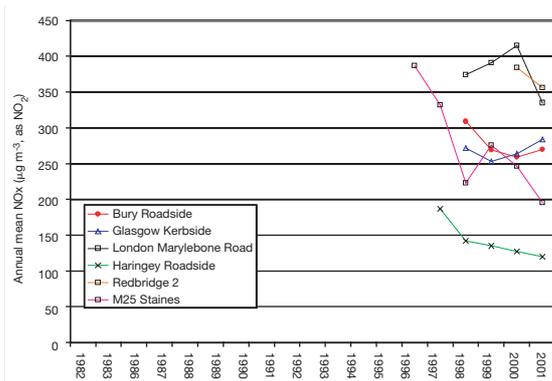
Figure 6.8 Annual mean NO$_2$ concentrations at selected roadside and kerbside sites.
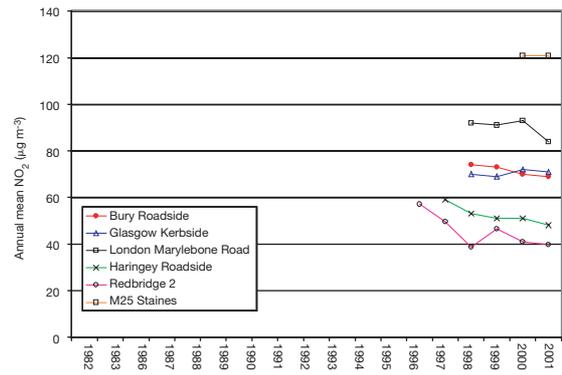
Figure 6.9 Split winter 99.8%ile of hourly average NO$_x$ concentration at selected background sites.
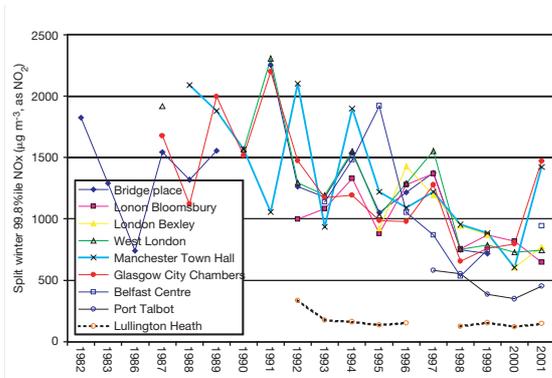
Figure 6.10 Split winter 99.8%ile of hourly average NO$_2$ concentration at selected background sites.
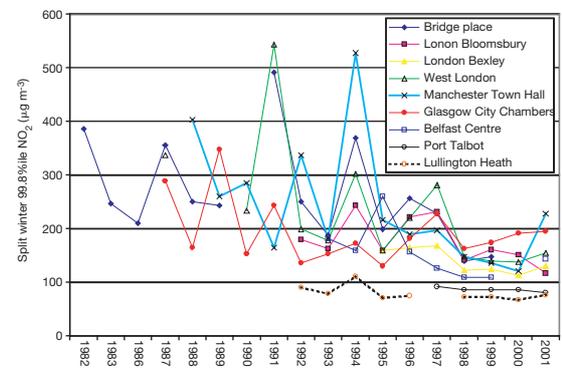
Figure 6.11 Summer 99.8%ile of hourly average NO$_x$ concentration at selected background sites.
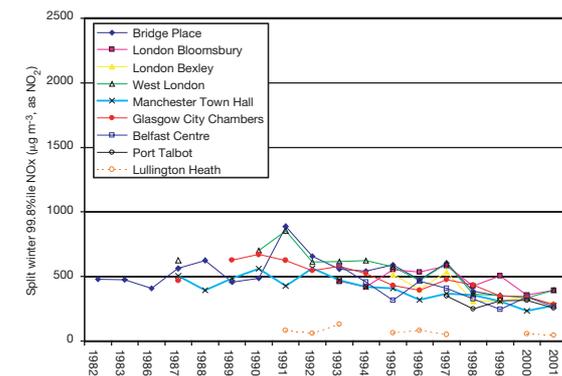
Figure 6.12 Summer 99.8%ile of hourly average NO$_2$ concentration at selected background sites.
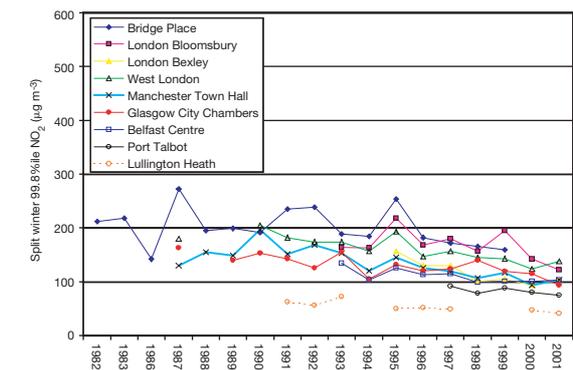
Figure 6.13 Measured annual mean NO$_x$ and NO$_2$ concentrations at selected background sites 1983 – 2001.
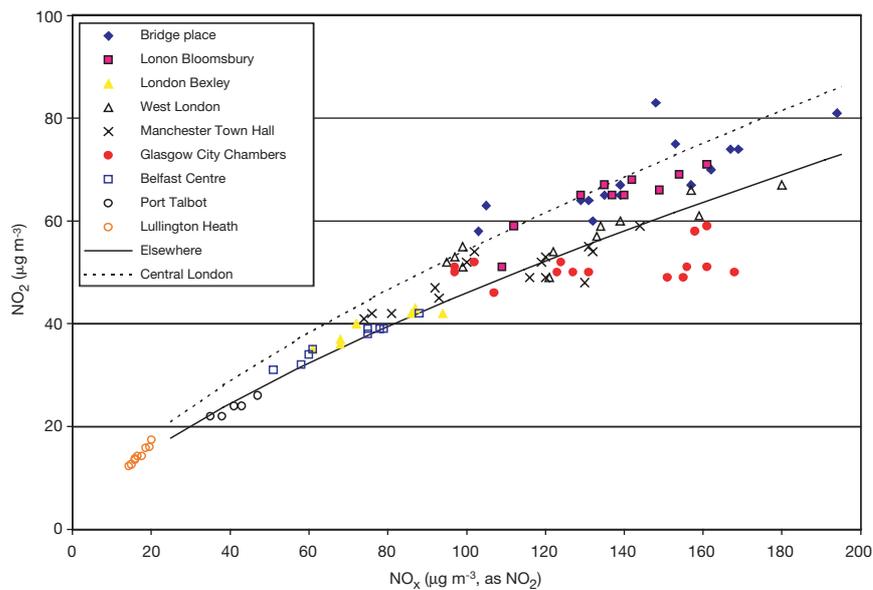
6.2.4 Analysis of hourly NO$_x$ and NO$_2$ meteorological and traffic count data in 2001 at selected roadside and kerbside monitoring sites in 2001

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.
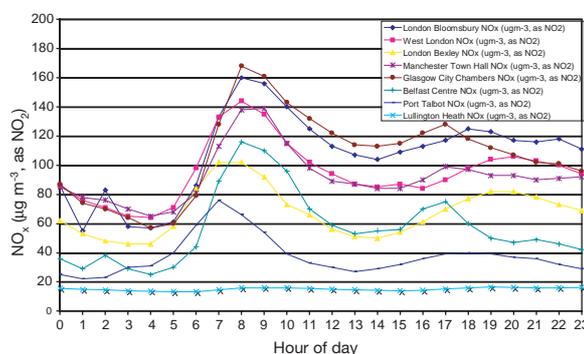
Figure 6.15 Diurnal variation of NO_2 concentrations at selected background sites in 2001.
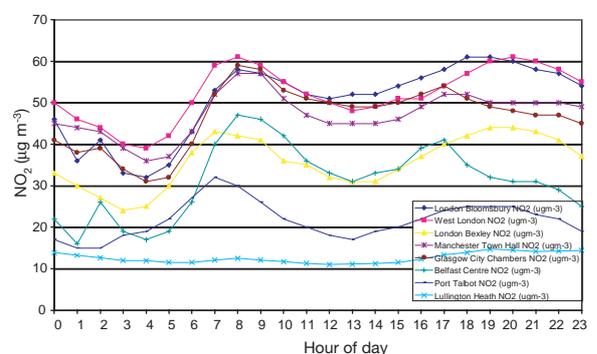
Figure 6.16 Diurnal variation of NO$_x$ concentrations at Marylebone Road and London Bloomsbury in 2001.
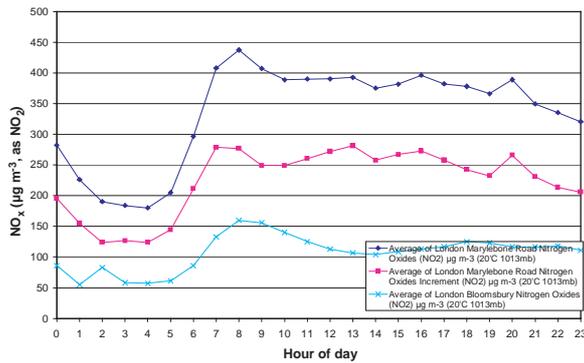
Figure 6.17 Diurnal variation of NO$_2$ concentrations at Marylebone Road and London Bloomsbury in 2001.
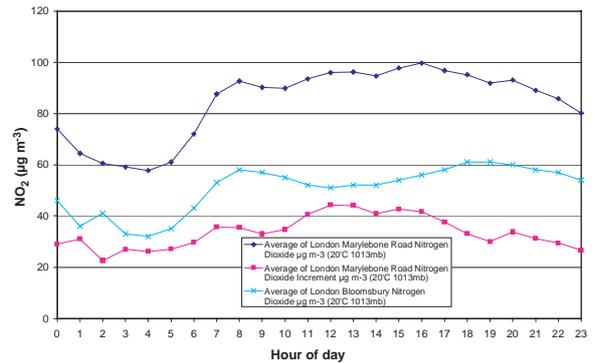
Figure 6.18 Diurnal variation of traffic flow at Marylebone Road in 2001.
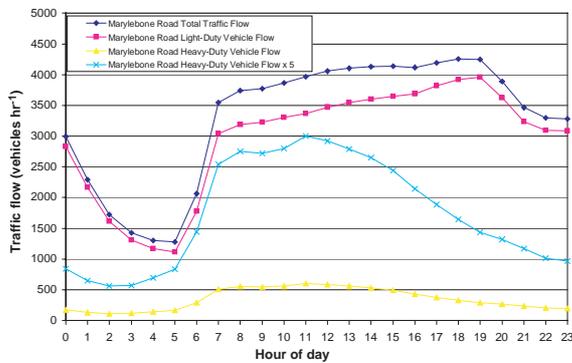
Figure 6.19 Diurnal variation of wind speed at Barking and Dagenham 1 in 2001.
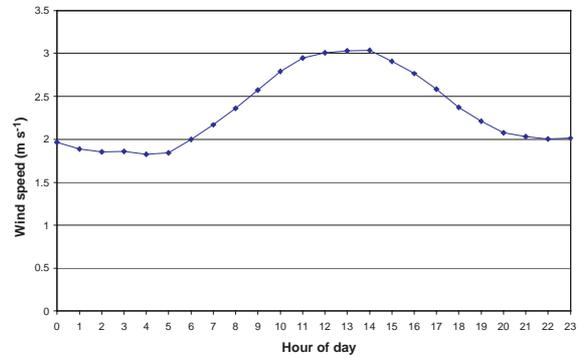
Figure 6.20 Comparison of roadside increment of NO$_x$ concentrations with traffic flow at three sites in 2001.
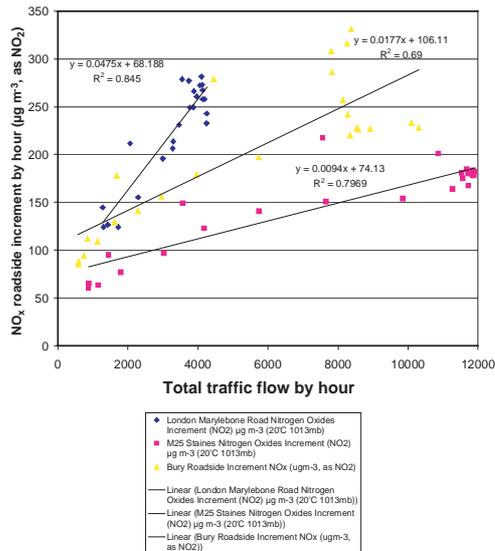
Figure 6.21 Comparison of roadside increment of NO$_2$ concentrations with traffic flow at three sites in 2001.
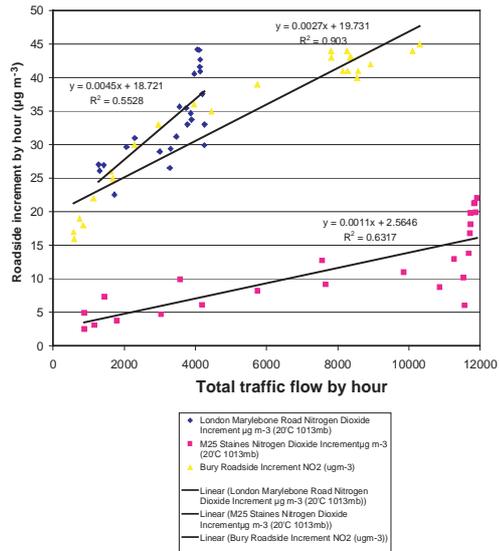
641. Annual average roadside increments in 2001 are listed in Table 6.8 along with the calculated NO$_2$/NO$_x$ ratio for these increments. The total roadside NO$_x$ concentration is dominated by local emissions from the road but the total roadside NO$_2$ concentration includes a large contribution from 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.
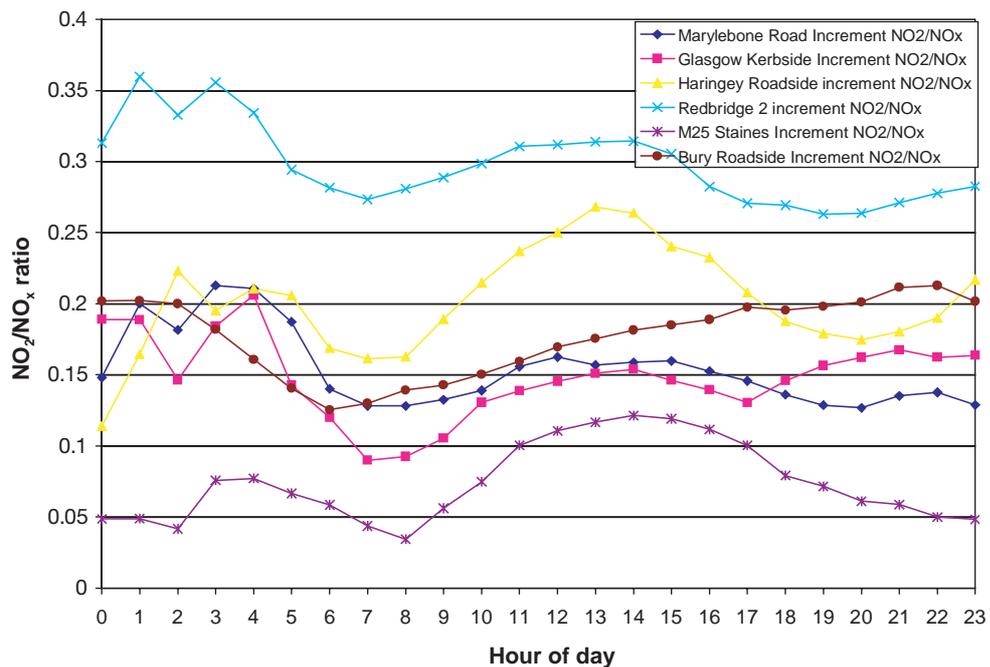
642. The traffic increment NO$_2$ is less than 10% of NO$_x$ at the M25 Staines site, peaking during the afternoon. This is a much lower percentage than at the other sites studied and may, at least partly, reflect the choice of background site (London Teddington), which is located in a park on the outskirts 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.
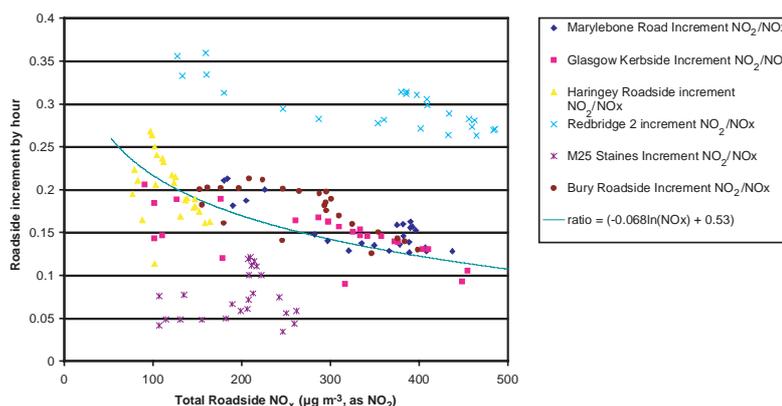
Figure 6.24 Comparison of NO$_2$/NO$_x$ ratio of roadside increment and total roadside NO$_2$.
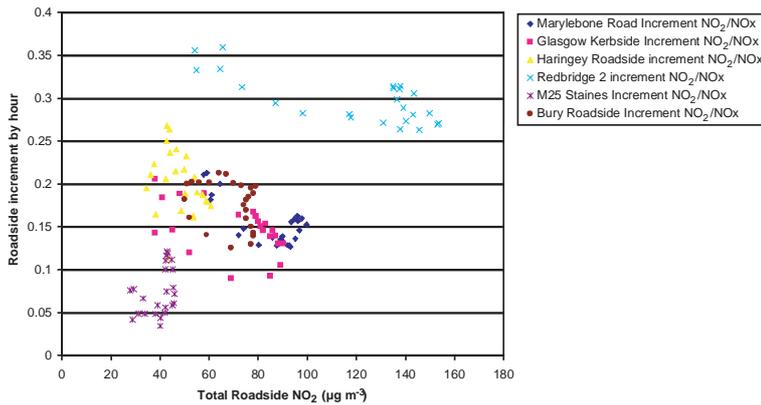
645. The analyses carried out at the six roadside sites considered here suggests that the highest primary NO$_2$ concentrations are found close to congested traffic or where a high proportion of the traffic emissions are from vehicles with diesel engines, such as buses. The lowest primary NO$_2$ concentrations 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].
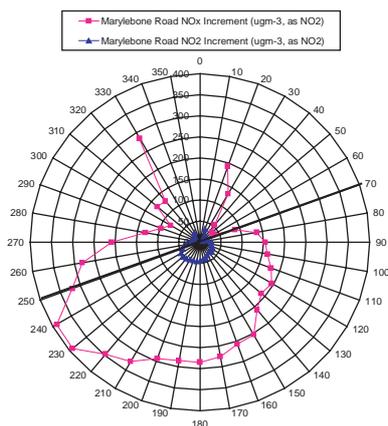
Figure 6.26 NO$_x$ roadside increment at Marylebone Road and wind speed in 2001 (wind direction between 150 and 250).
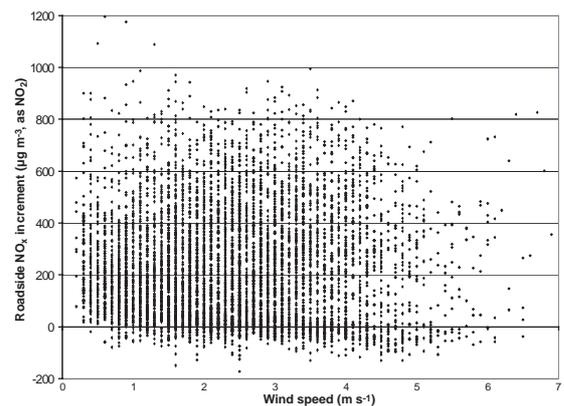
647. Street canyon effects are not evident at any of the other sites selected for detailed analysis. This is illustrated in Figures 6.27 and 6.28, which show the pollution rose and winds speed plots for the roadside increment of hourly NO$_x$ concentrations at Haringey Roadside. The highest NO$_x$ increments 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 NO₂).[5]
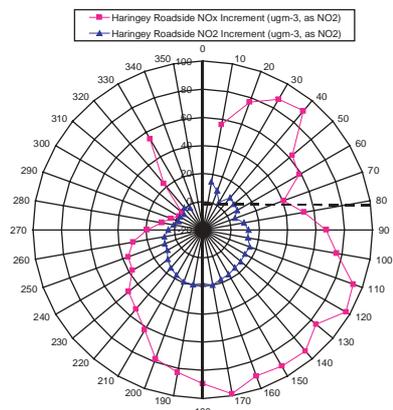
Figure 6.28 NOₓ roadside increment at Haringey Roadside and wind speed in 2001 (all wind directions).
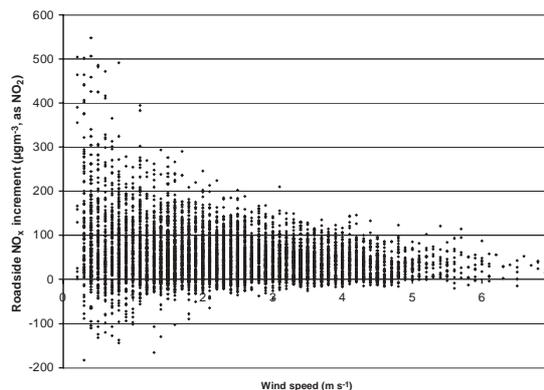
648. The roadside and kerbside sites selected for detailed analysis have been chosen to reflect a range of different concentrations and environments. It is, however, important to consider the relevance of the sites in terms of exposure when comparing the measured concentrations with objectives and limit 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 NO₂ 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 NO₂. 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 NO₂ indicate that under these conditions these instrument types might result in higher NO₂ 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 NO₂ 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 NO₂). 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.
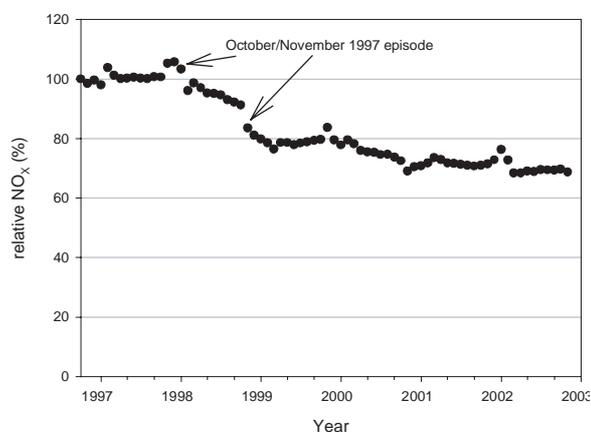
Figure 6.30 Relative monthly annual mean normalised NO_2 in London.

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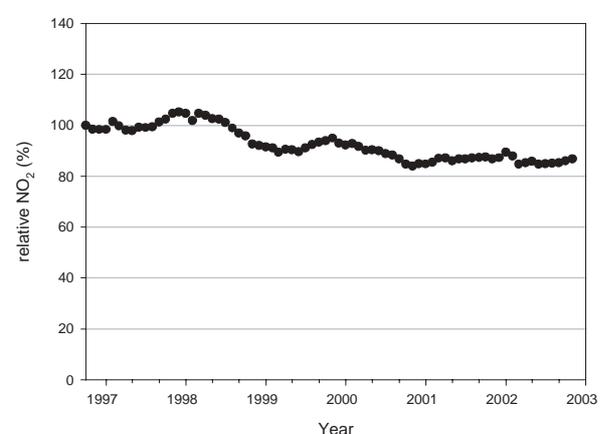
[6] Some of the data for 2002 are unratified. See section 4.9.6.

Figure 6.31 Running annual mean NO$_2$, O$_3$ and OX (NO$_2$ + O$_3$) averaged over five background sites in the LAQN.
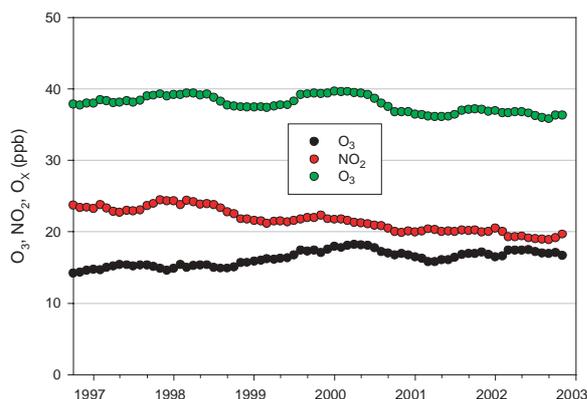
6.2.6 Source apportionment and analysis of recent trends in annual mean NO$_x$ concentrations using site-specific empirical models

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	Commer-cial	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)	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 from 1996 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.
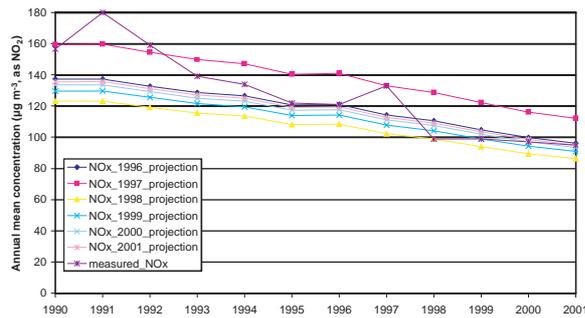
658. Figure 6.33 summarises the results of this type of analysis at the background monitoring sites
selected for detailed analysis. The measured concentrations in each year have been normalised
by dividing by the concentration projected from the 2001 base year at each site. This should
remove the trend 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.
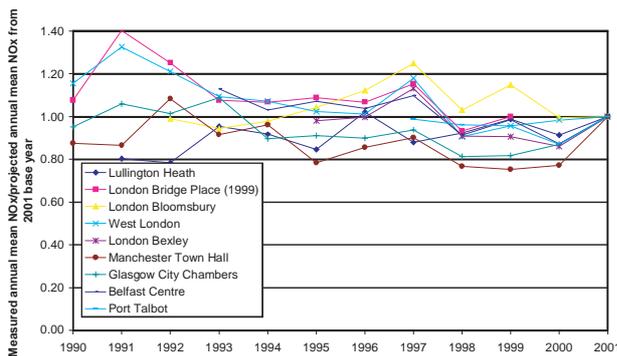
Figure 6.34 Comparison of measured and
site-specific estimates of annual mean NO$_x$
concentrations at roadside and kerbside sites.
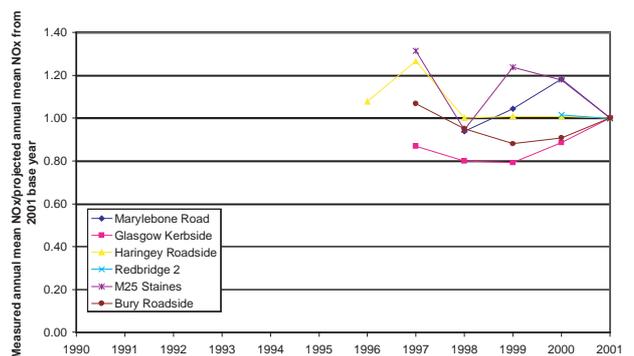
Figure 6.35 Site-specific projections of annual mean NO$_x$ concentrations at Central London/Bridge Place.
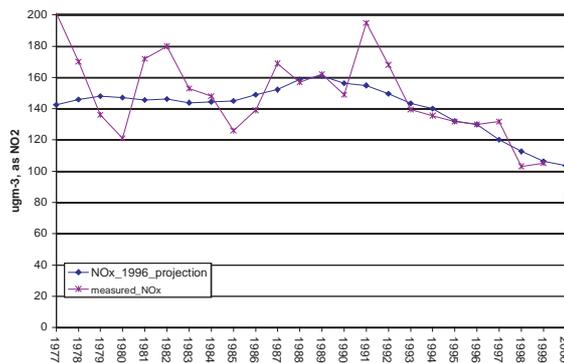
Figure 6.36 Site-specific projections of annual mean NO$_x$ concentrations at Cromwell Road/Cromwell Road 2.
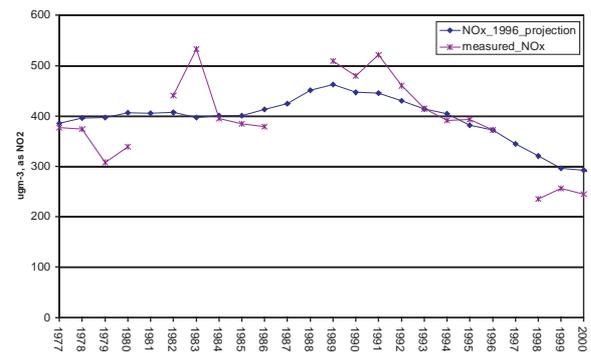
659. NO$_x$ emissions from traffic sources peaked in about 1990 and thus the analyses described above can only be used to illustrate the decline in emissions since then. Figures 6.35 and 6.36 show site-specific projections of annual mean NO$_x$ concentrations at Central London/Bridge Place and Cromwell 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.

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.
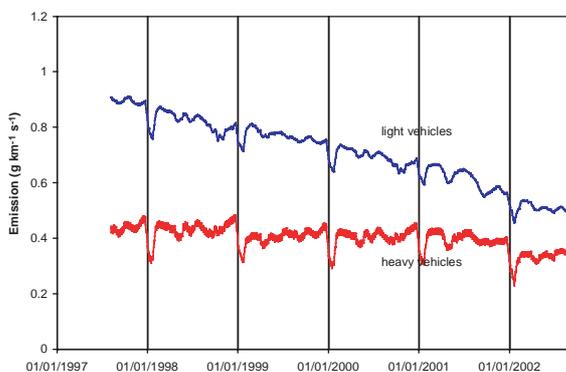
Figure 6.38 Relative NO_x emissions and concentration by day of the week.
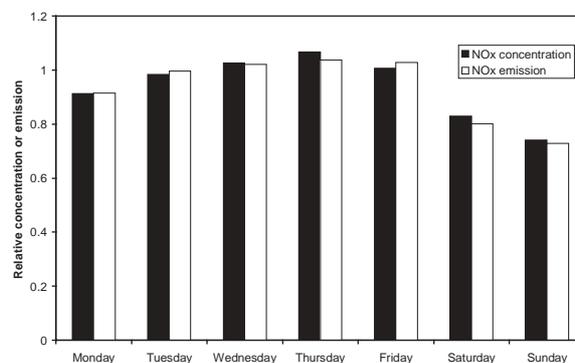
664. Table 6.10 shows that there was a relatively small change in NO_x concentrations at Marylebone Road until 2001, despite a significant downward trend in total emissions. The variation in NO_x concentrations from year to year must therefore be explained by variations in meteorology from year to year (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 ≥ 70º and ≤90º (along the road) or ≥160º and ≤250º 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.
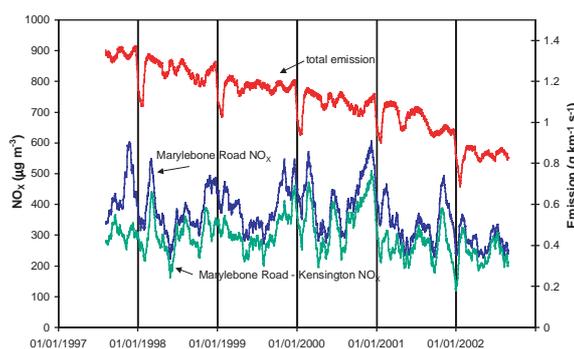
Figure 6.40 Proportion of the time where the wind direction is from a 'polluted direction'.
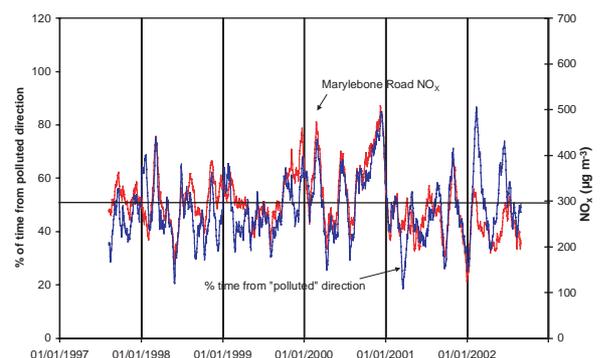
667. Observations of NO$_x$ and NO$_2$ at Marylebone Road show that there is significant variation in the concentrations of NO$_x$ and NO$_2$ year to year, with concentrations only declining in 2001 and 2002 (see Table 6.10 and Figure 6.7 and 6.8). Variations in meteorology from year to year, along with 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.
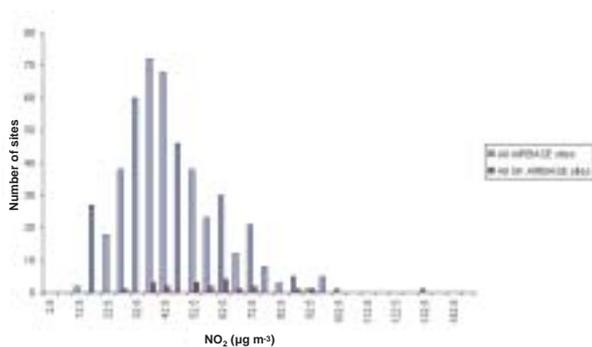
Figure 6.42 The frequency distribution of the annual mean NO_2 concentrations reported for urban background sites in the AIRBASE database for the year 2000.
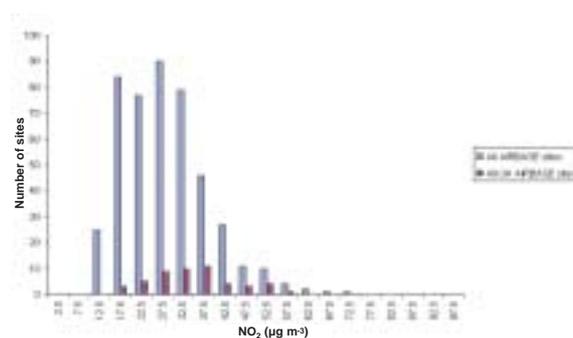
Figure 6.43 The frequency distribution of the annual mean NO$_2$ concentrations reported for rural background sites in the AIRBASE database for the year 2000.
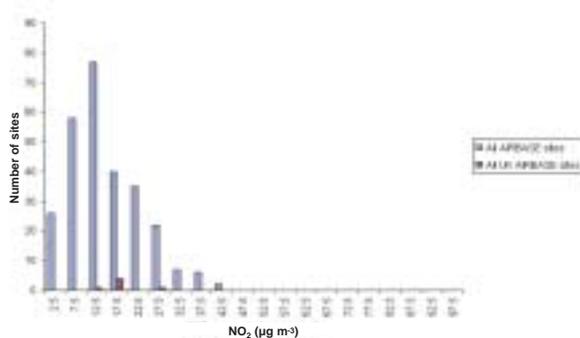
Figure 6.44 The frequency distribution of the annual mean NO$_2$ concentrations reported for rural background sites in the EMEP database for the year 2000.
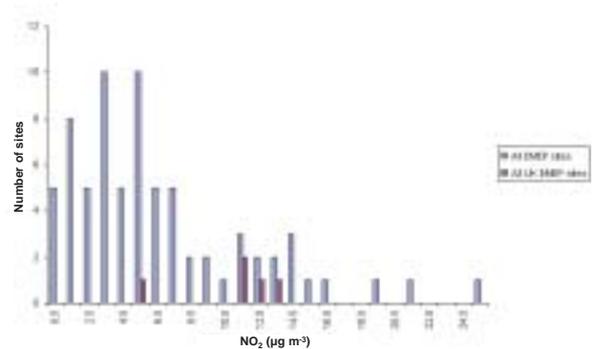
672. Urban sites in AIRBASE have a further subdivision into sites which are industry-influenced. This category contains 146 sites with an average annual mean NO$_2$ concentration of 25.2 µg m^{-3} for the year 2000. This value is slightly lower than that for the urban background category but the difference 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.
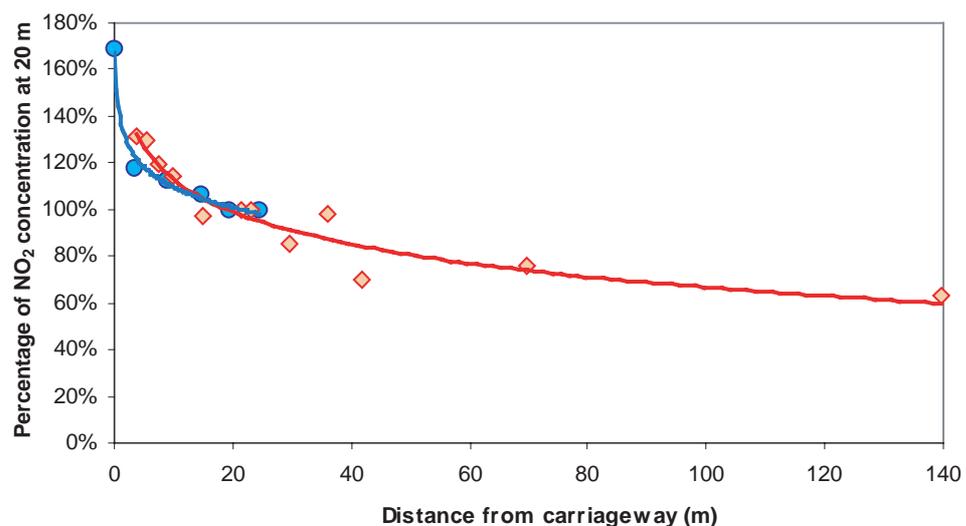
683. A recent study alongside the M25 west of London, also using diffusion tubes, has demonstrated a similar rapid decline over the first 20 m (Hickman *et al.*, 2002). This is also illustrated in Figure 6.45, in which annual average concentrations for both sides of the motorway are shown normalised 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)	ΔNO_2/ ΔNO_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_2 downwind 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 NO₂ 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 (µg 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.
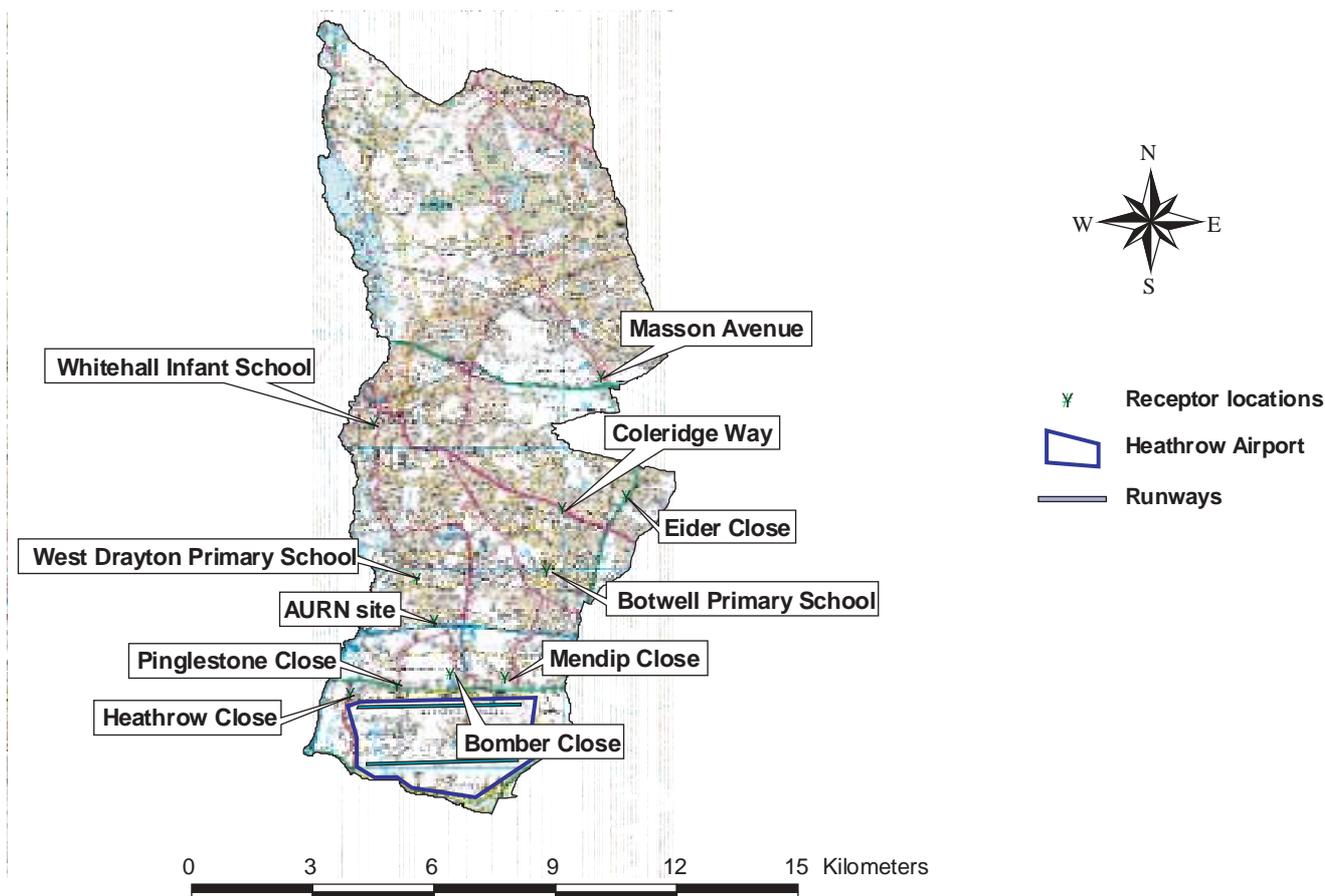
695. Figure 6.47(a) shows a breakdown by general source type of emissions in Hillingdon. The emissions from the airport, which include all aircraft emissions up to 1000 m and all other emissions within the airport perimeter, are dominant. The component parts of the Heathrow emissions are shown in Figure 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) NOₓ emissions from within Hillingdon (2005).
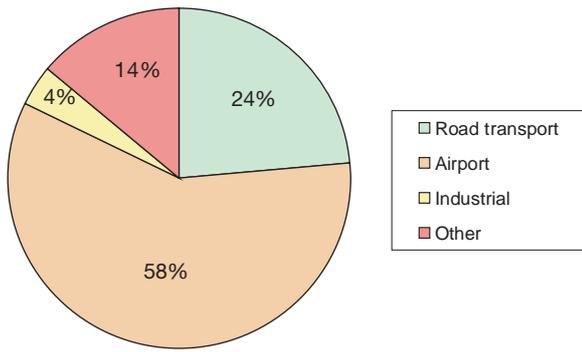
Figure 6.47(b) NOₓ emissions from different Heathrow source groups in Hillingdon (2005).

Road transport 24%
Airport 58%
Industrial 4%
Other 14%

Road vehicles 12%
Airborne aircraft 61%
Taxiing & aircraft holding 13%
Heating 6%
Other 8%
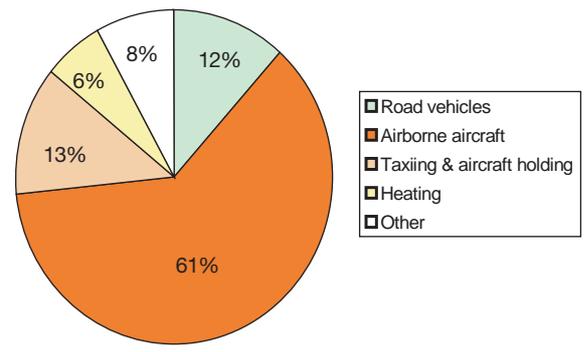
Figure 6.48 Contribution of major source groups to annual average NOₓ concentrations (2005).
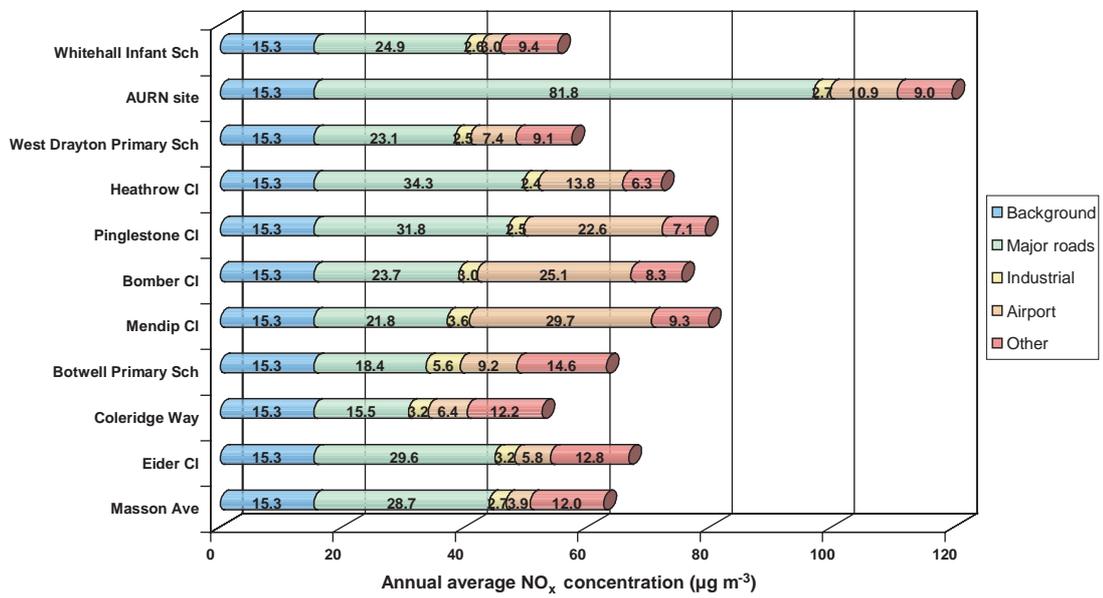
Figure 6.49 Contribution of Heathrow sources to annual average NOₓ concentrations (2005).
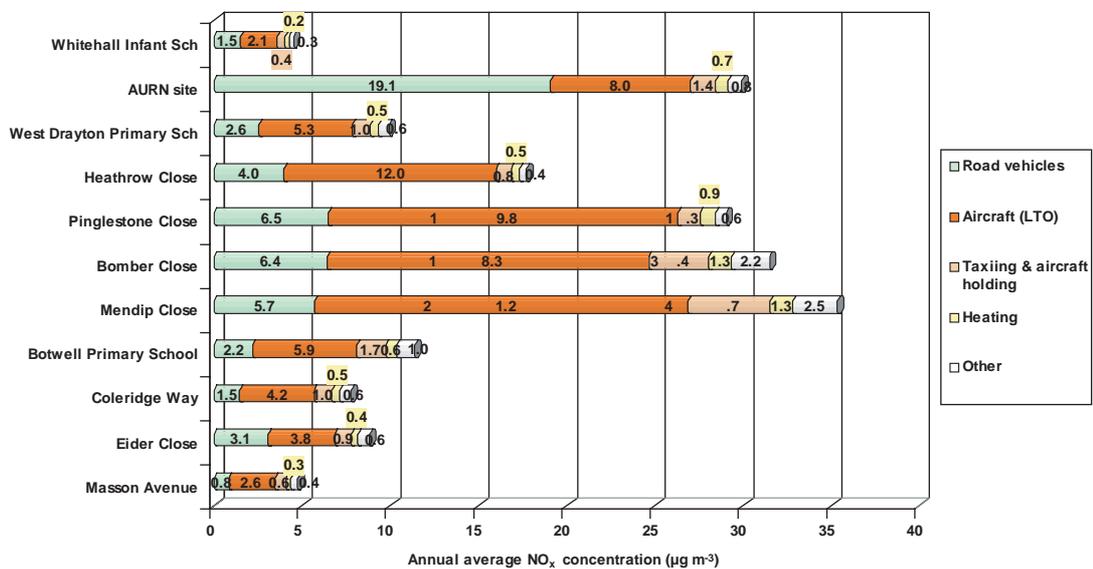
Figure 6.50 Contribution of aircraft at different heights to annual average NO_x concentrations during take-off, climb-out, approach and landing (2005).
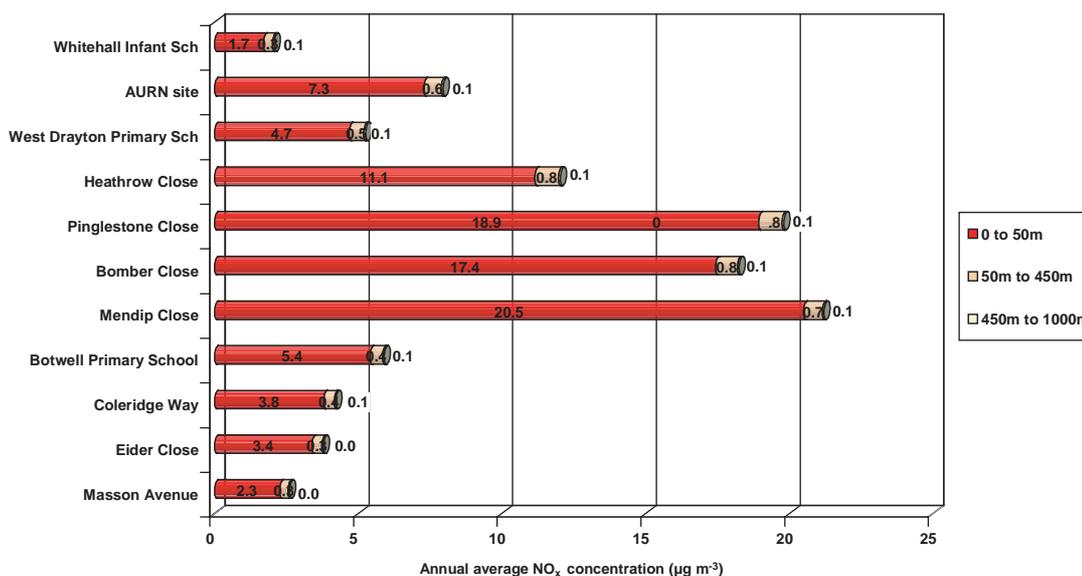
6.4   Modelling results for 2001

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 µg m^{-3} (km^2)	Number of road links > 40 µg m^{-3}	Length > 40 µg 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 µg 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 ($\mu g\ m^{-3}$, as NO_2).
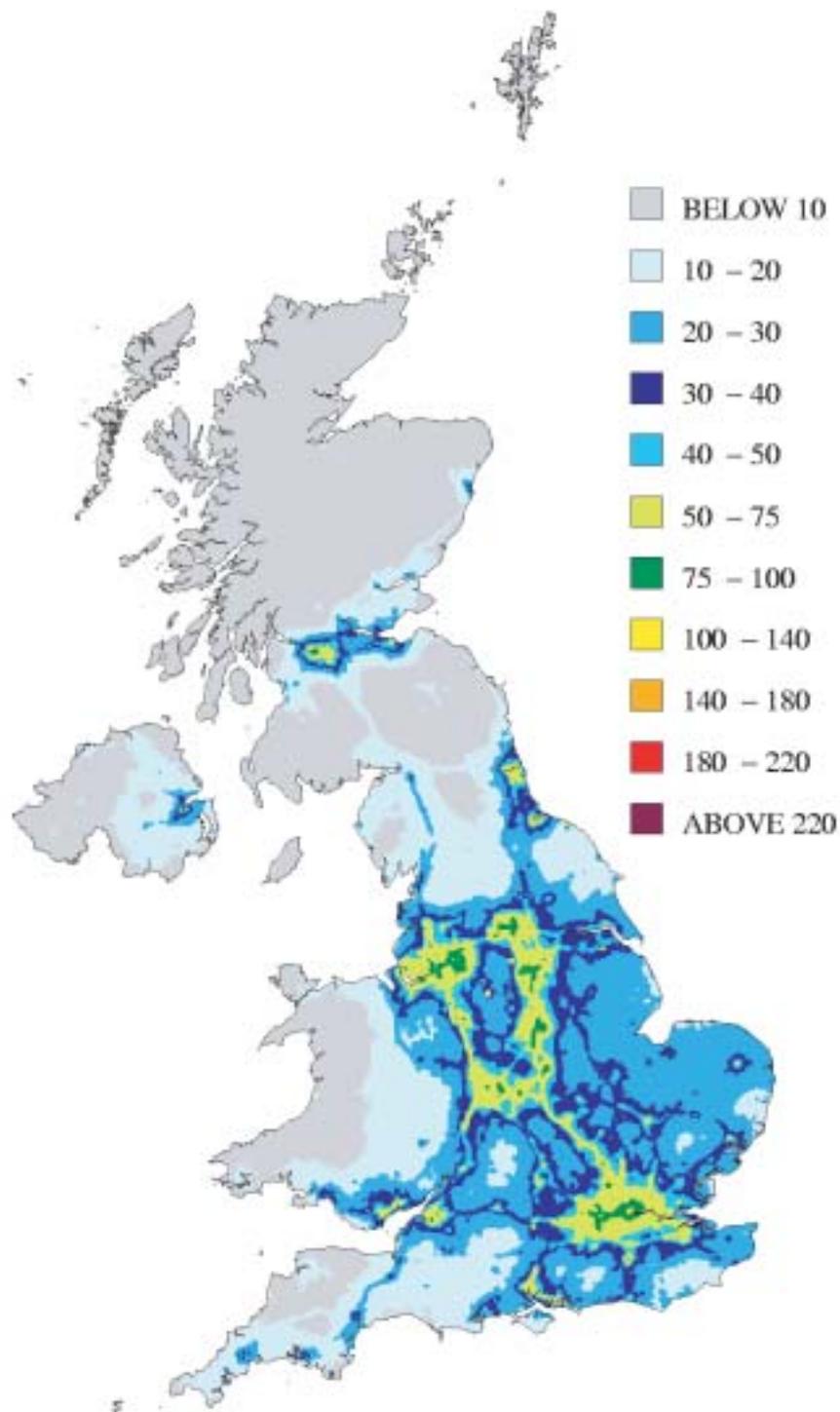
Figure 6.52 Estimated annual mean background NO_2 concentration, 2001 (µg m⁻³).
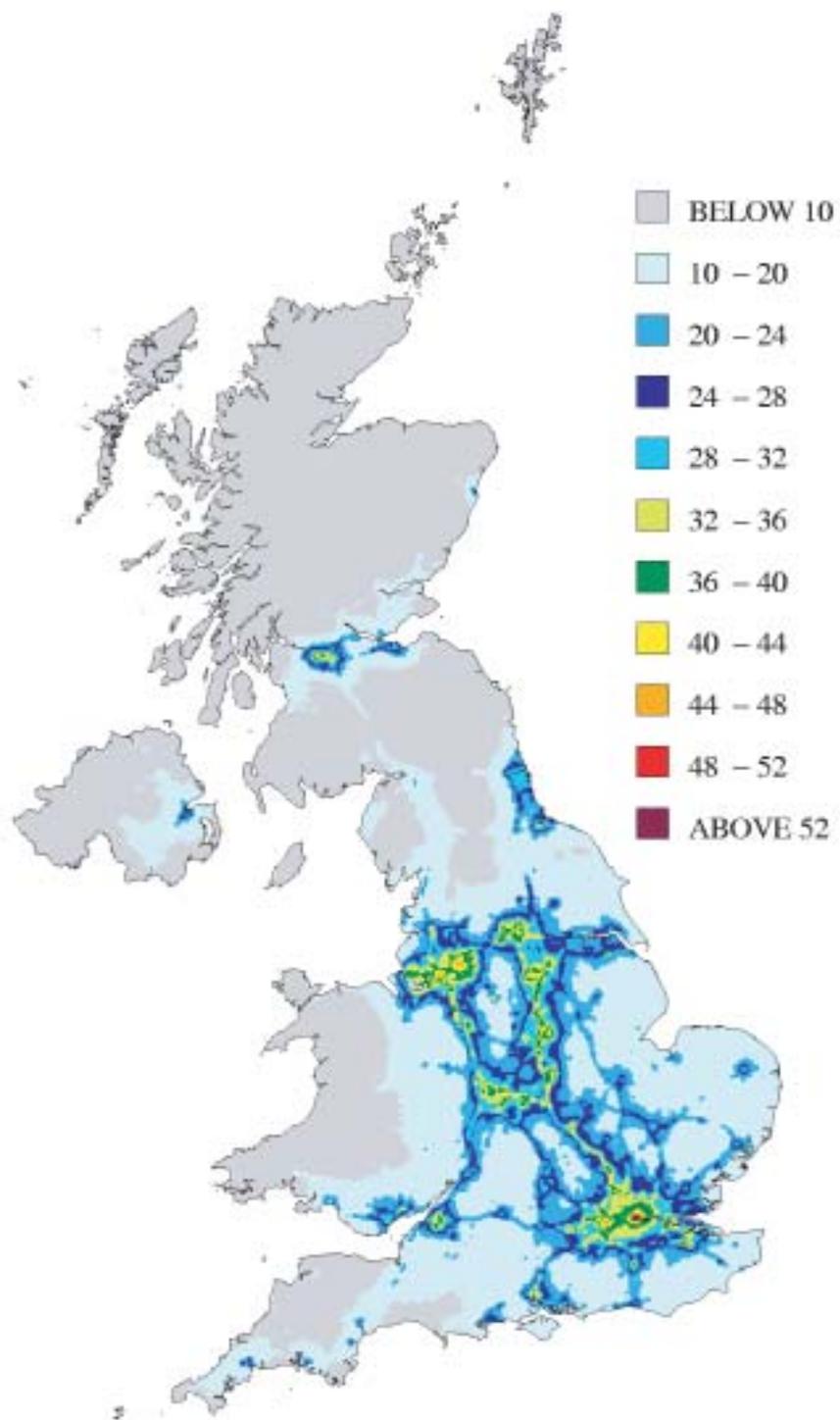
Figure 6.53 Estimated annual mean roadside NO$_x$ concentration, 2001 (µg m^{-3}, as NO$_2$).
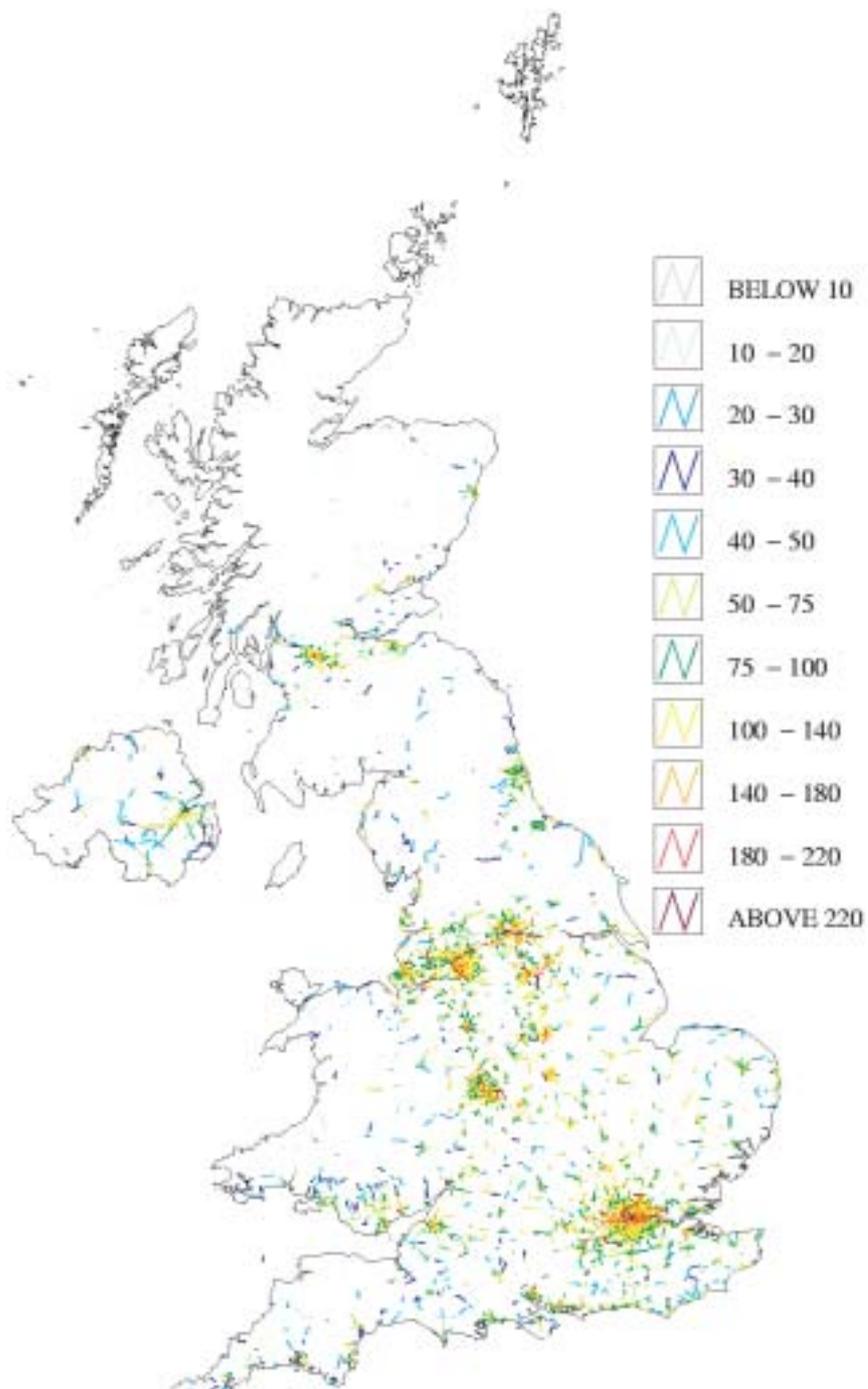
Figure 6.54 Estimated annual mean roadside NO$_2$ concentration, 2001 (µg m^{-3}).
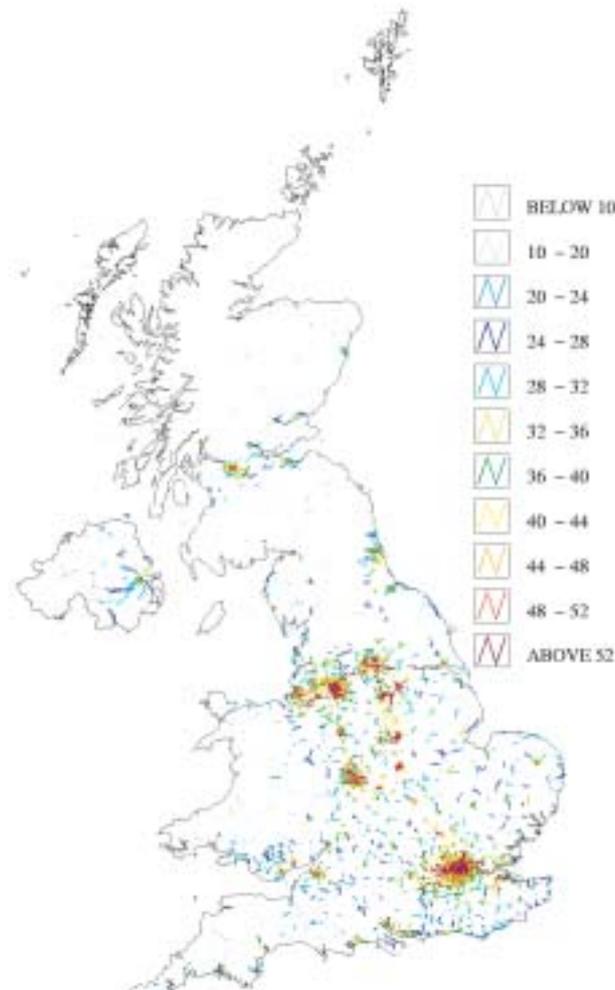
6.4.3 Detailed modelling results for London

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 µg 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 µg 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 µg m^{-3}, whereas the national modelling relates only to background areas exceeding 40 µg m^{-3}.

Figure 6.55 Annual mean NO$_2$ concentrations for 1999 in London estimated using the ERG model (µg m^{-3}).
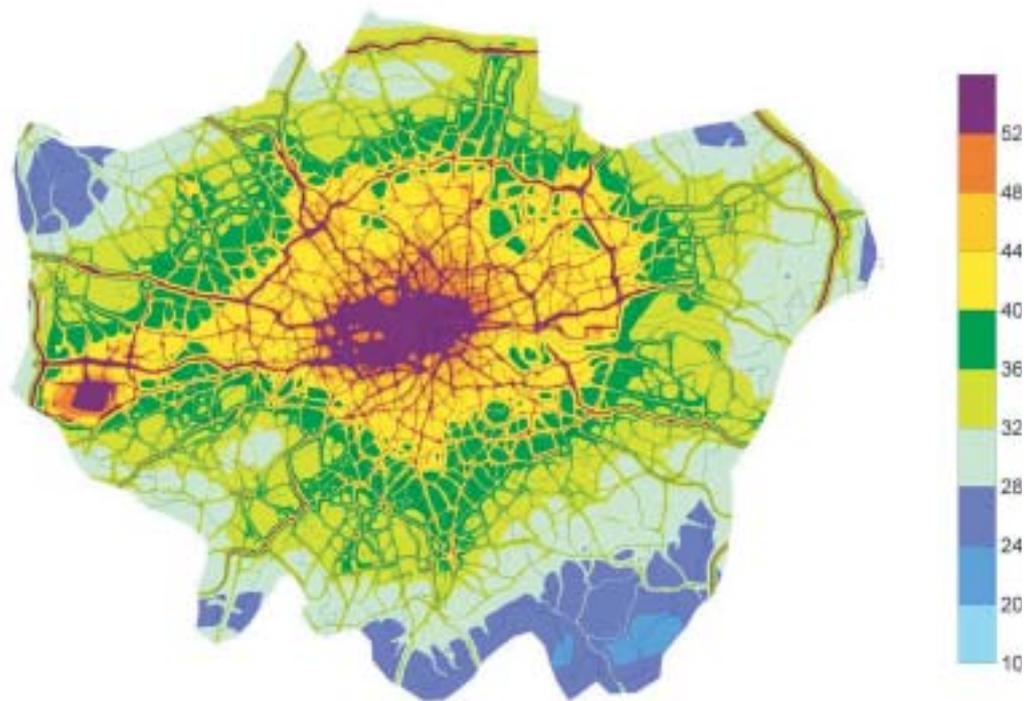
Figure 6.56 Annual mean NO$_2$ concentrations for 2001 in London estimated using the ERG model (µg m^{-3}).

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 (µg m^{-3}).
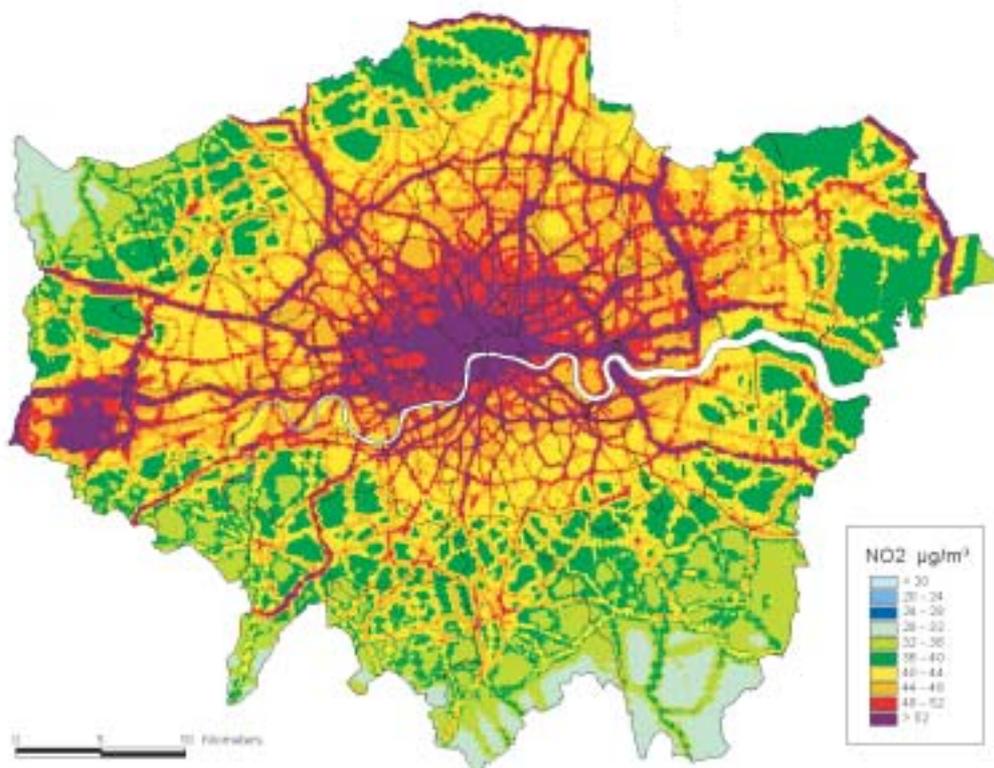
706. The LRCTM (see section 5.4) has also been used to estimate background NO$_2$ concentrations in London using the 1999 LAEI inventory and the resulting map is shown in Figure 6.58, covering over 1500 km^2 of north and central London and its outskirts. The annual mean NO$_2$ concentration for this selected area was found to be 39.5 µg 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 µg m^{-3}. The maximum annual mean NO$_2$ concentration in central London was found to be 57.4 µg 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 (µg m^{-3}).
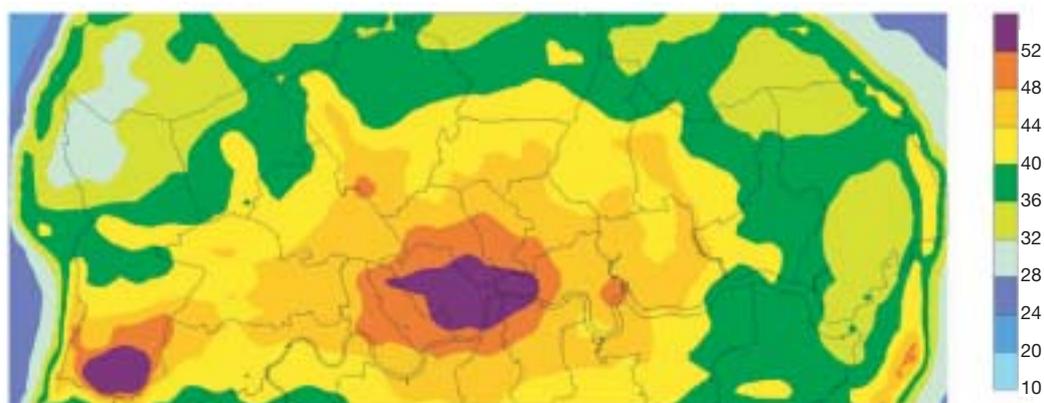
707. The estimates of background concentrations from the CERC and LRCTM modelling are higher than those from the netcen and ERG modelling. This highlights the uncertainties associated with modelled assessments of air quality. An examination of maps of annual mean NO_x derived from the CERC and netcen 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.
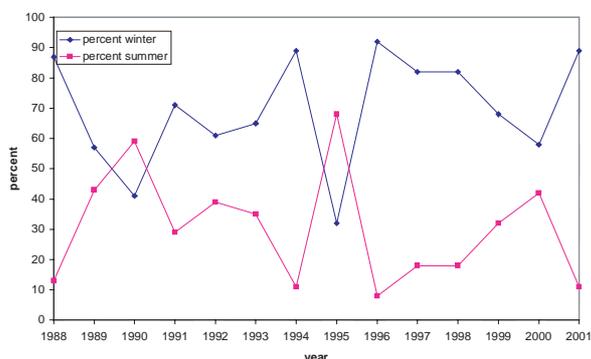
Figure 6.60 The number of sites in the national network and the number of hours per site per year with NO$_2$ concentrations greater than 200 µg m^{-3}.
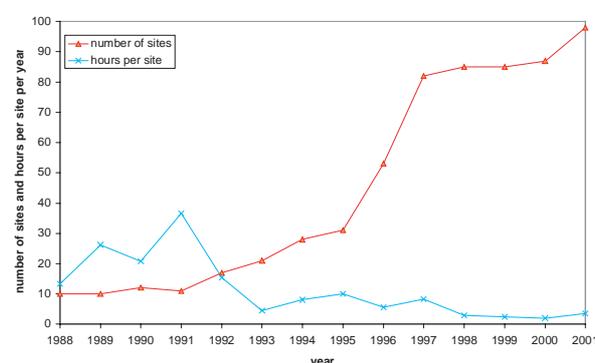
711. The number of monitoring sites has increased approximately 10-fold since 1988 and the number of episodes per site has decreased as emissions have declined (and the number of sites has increased including a greater proportion of sites outside the major conurbations). The majority of hours above the 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.
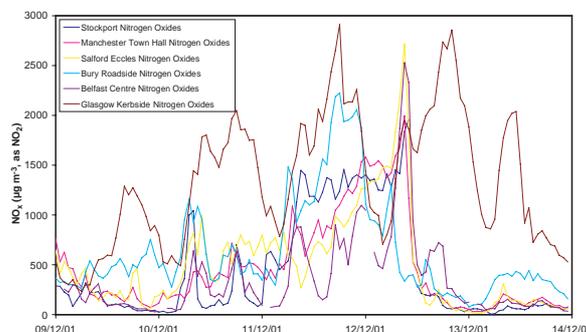
Figure 6.62 Hourly NO$_2$ concentrations during the episode in December 2001 at selected sites.
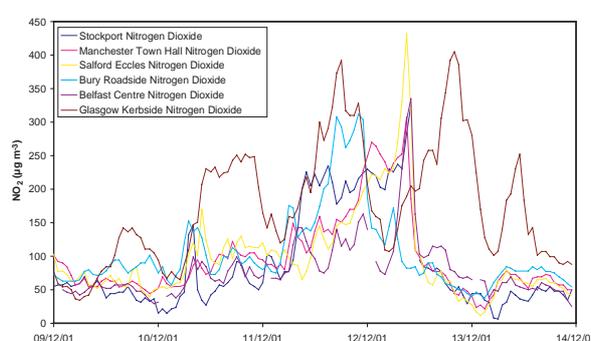
713. Figure 6.63 shows a comparison of the measured hourly concentrations of NO_x and NO_2 at two sites (Marylebone Road and Glasgow Kerbside) for the 4th quarter of 2001. Data for the 4th quarter of 1991 at Cromwell Road are also shown for comparison. This graph illustrates that the behaviour of the 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.
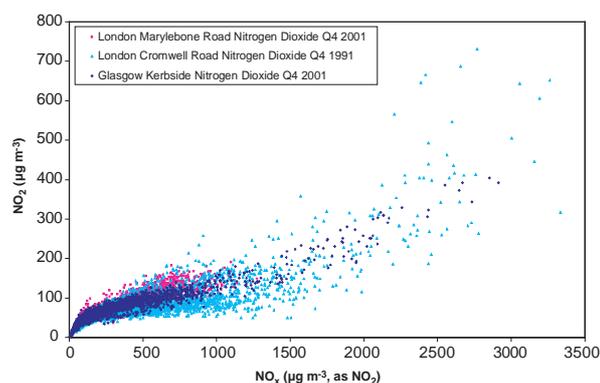
Figure 6.64 Hourly NO_x and NO_2 at Salford Eccles 2001 and Manchester Town Hall 2001 and 1992.
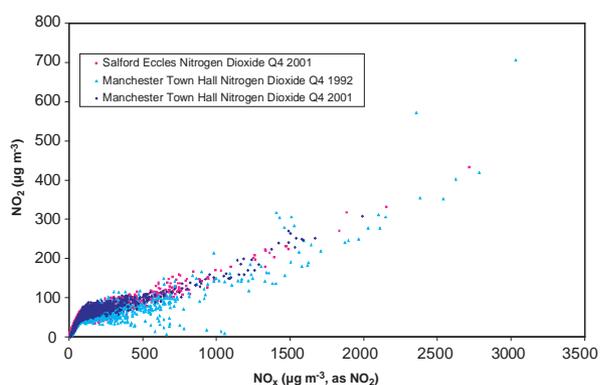
716. The interpolation of hourly NO_2 measurement data from this episode in the Greater Manchester area has been discussed in section 5.6.3.1. Hourly NO_x concentrations during winter episodes exhibit extensive spatial variation due to meteorological and topographic variation and particularly the small-scale 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.
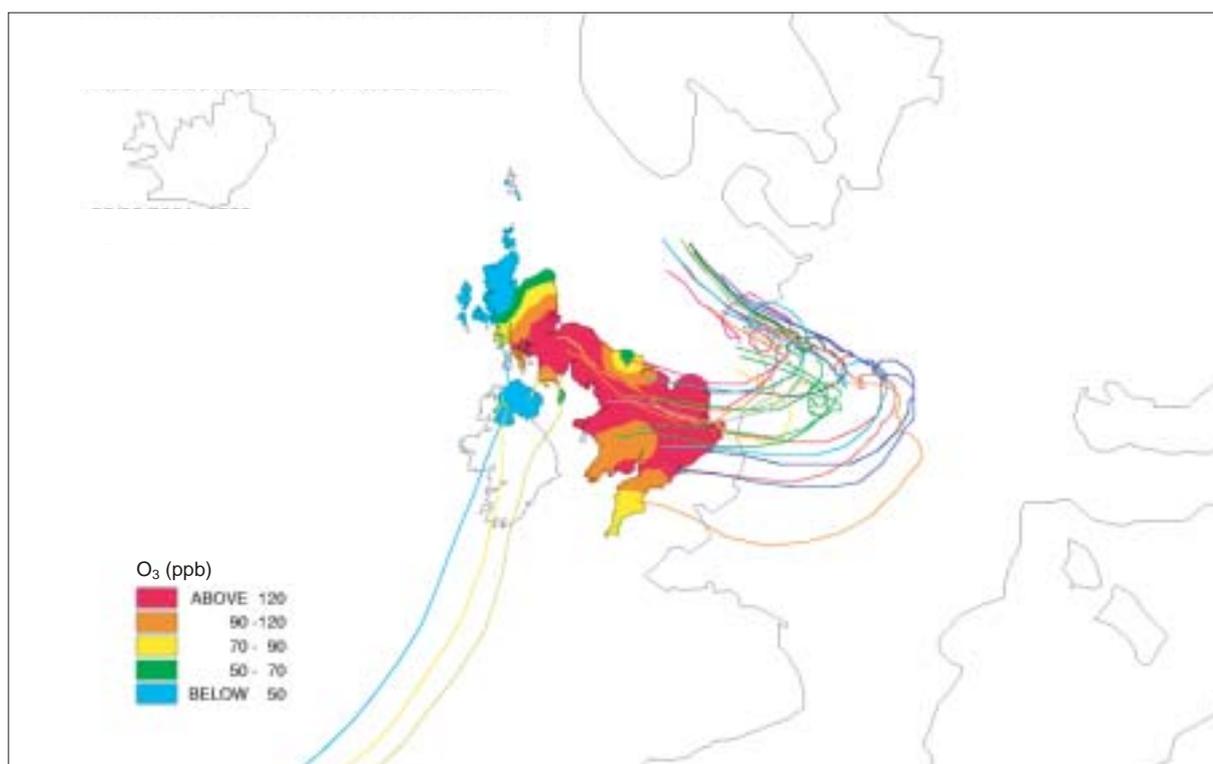
719. Hourly concentrations of NO_x, NO_2, O_3 and total oxidant ($NO_2 + O_3$) during the episode are shown in Figures 6.66 to 6.69. The highest O_3 concentrations were recorded at the rural site of Lullington Heath peaking at 198 µg m⁻³ on 26th June 2001. Oxidant concentrations at this site were dominated 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.
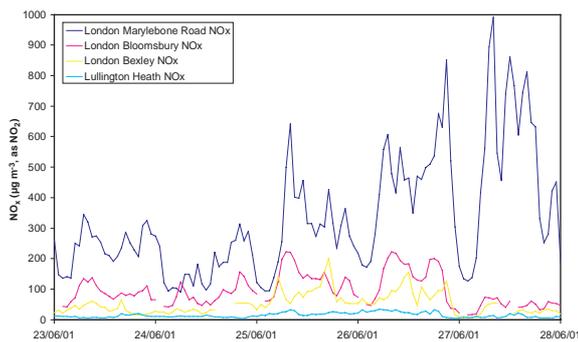
Figure 6.67 Hourly NO_2 concentrations during the episode in June 2001 at selected sites.
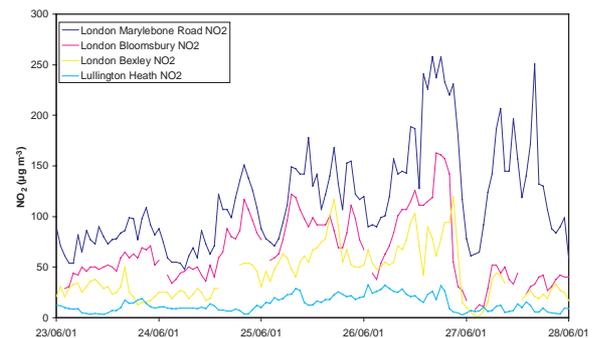
Figure 6.68 Hourly O_3 concentrations during the episode in June 2001 at selected sites.
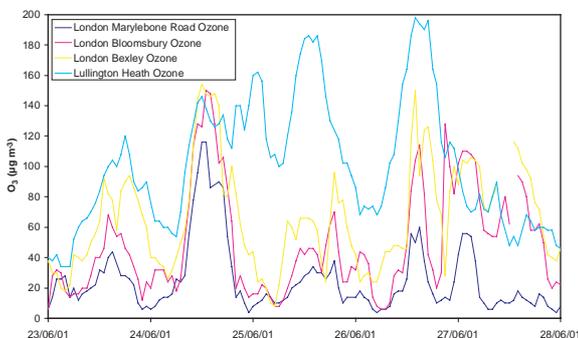
Figure 6.69 Hourly total oxidant ($NO_2 + O_3$) concentrations during the episode in June 2001 at selected sites.
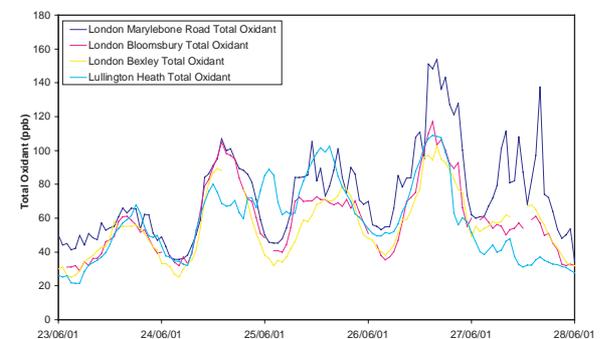
6.6 Influence of changes in mean O_3 concentrations on annual mean NO_2 concentrations

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.
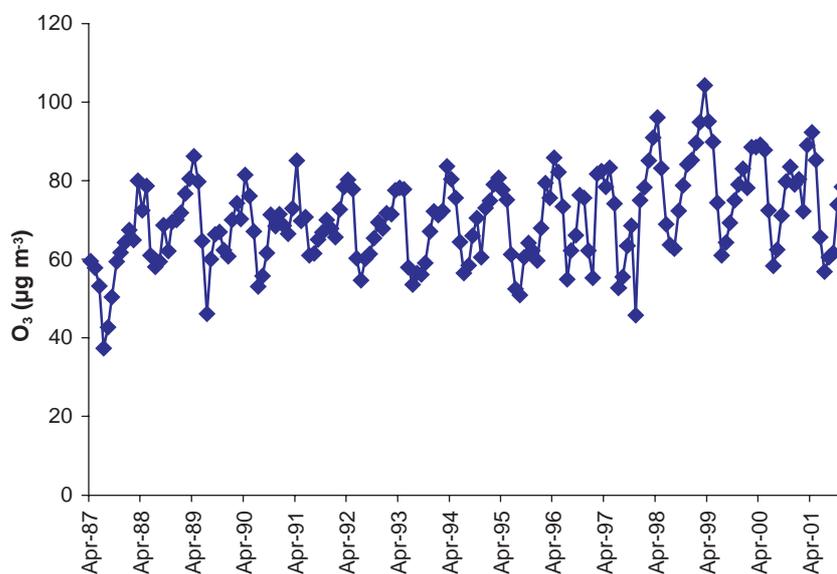
726. This analysis indicates that there has therefore been a considerable upwards trend in the northern hemisphere O$_3$ baseline concentrations in air masses entering the UK which is particularly apparent in the winter and spring. This should have resulted in greater extents of oxidation of NO$_x$ emissions 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 µg 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.
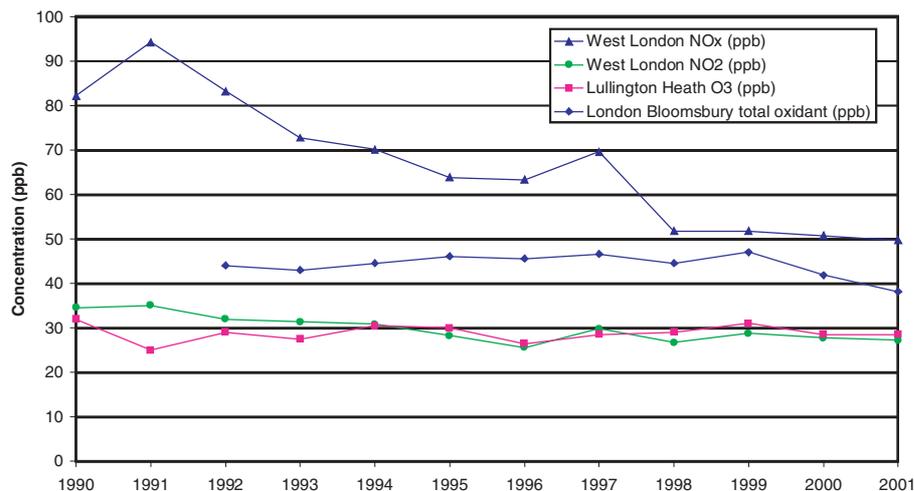
Figure 6.72 Trends in annual mean NO_x, NO_2, O_3 and total oxidant in Glasgow.
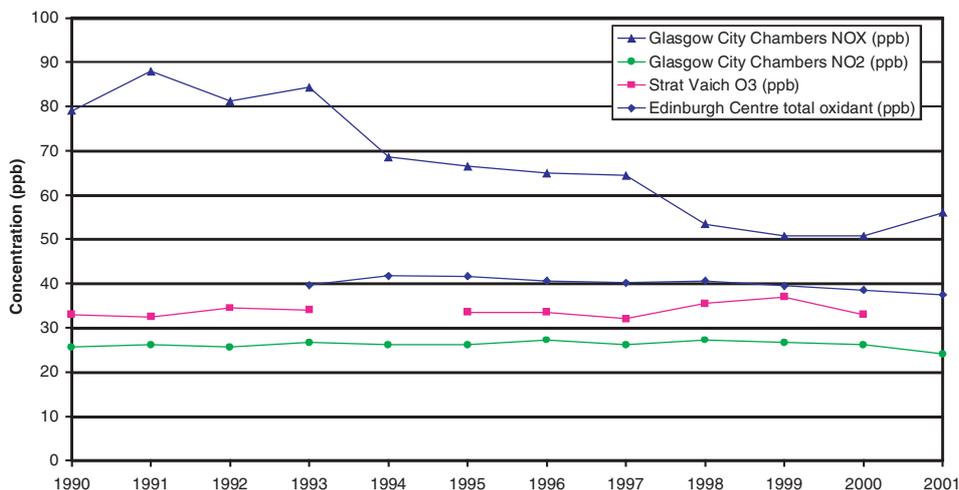
733. Summer episode O_3 concentrations have, however, also declined over this period. Such episodes have been and remain more frequent in south-east England than in Scotland. The contribution of summer episode oxidant to annual mean NO_2 concentrations in London is therefore likely to have declined over 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.