Chapter 4

Sources of particles in the UK

Key points

- The source emissions relevant to atmospheric concentrations of particles are related to a complex multi-pollutant mixture that is present over a range of different particles sizes. These sources include primary, directly emitted, material and secondary material that is formed subsequently in the atmosphere. Particle sources can also be grouped into those originating from anthropogenic activities and those that are natural in origin.

- Total UK emissions of PM_{10} in 2001 were 180 kilotonnes (180 kt). Transport (in particular road transport) and industrial processes are the two most important sources of PM_{10} and each account for 27% of total UK emissions. Emissions from the domestic combustion sector (17% of the total) are also important.

- Considering the road transport sector, tyre and brake wear emissions in 2001 accounted for ~23% of the total road transport emissions. Resuspended road dust also makes an important contribution, one that is not accounted for in national estimates of UK total PM_{10} emissions. Recent work suggests that the resuspended component of PM_{10} can be as large as, and in some cases much larger than, exhaust emissions. Resuspended PM_{10} is, however, variable in magnitude and difficult to quantify with any certainty. In addition, both tyre and brake wear and resuspended PM_{10} appear to be much more important for heavy duty vehicles than for light vehicles such as cars. The type approval test for particulate emissions is only applicable to compression ignition engines. No legislation is currently in place to limit the particulate emissions from spark ignition engines. PM emissions associated with spark ignition engines are not well defined, but although they do not represent significant emissions in terms of mass, they can represent an important source of ultrafine particles.

- Type approval PM measurements for road vehicle emissions testing are based upon a gravimetric filter-based method that is measured at 52°C. No attempt is, therefore, made to include the volatile component.

- For road vehicle emissions testing, the emission sampling strategy can lead to the formation of sampling artefacts. Whereas various PM metrics could be measured at the point of formation or emission, limited work has been undertaken to characterise the changes in PM morphology between the point of emission and the ultimate receptor. The sampling strategy and instrument choice will dictate the measured PM emission value. International programmes across the EU go someway to provide standardised transient test protocols. Measurements undertaken outside these standardised in-service test protocols must be treated with caution.

- The shipping sector makes an important contribution to EU emissions of SO_2, which is an important particle precursor gas. Recent estimates suggest that by 2010 shipping will be equivalent to more than 75% of total land-based emissions of SO_2. Shipping emissions of SO_2 are also of local importance in the vicinity of major UK ports.

- The importance of each emission sector depends markedly on the particle size considered. For total UK emissions of coarse particles (PM_{10} – $PM_{2.5}$) in 2001, the industrial processes sector was most important (34%) followed by domestic combustion (19%). However, for particles smaller than $PM_{2.5}$, the road transport sector was the most important (39%), followed by industrial processes (21%). The road transport sector was even more important when particles smaller than $PM_{0.1}$ are considered: it accounts for 54% of total emissions.

- Between 1990 and 2000, total UK PM_{10} emissions, which have been quantified in the NAEI, have fallen by 42% from 309 to 178 kt. These reductions have been driven by a reduction in coal use in electricity generation and industrial processes. However, the decline in road transport emissions of 37% has also made a significant contribution to the overall decline.

- Total UK emissions of PM_{10} are expected to decline by 28% between 2000 and 2010, primarily due to a decline in power station and road transport emissions. Emissions of precursor gases are also expected to decline over this time period. SO_2 is expected to decline by ~52%, mostly as a result of the introduction of regulations to control the sulphur content of liquid fuels. The decline in emissions of NO_X by 30% is dominated by reductions in the road transport sector. For NMVOCs the estimated 28% reduction is mostly the result of reductions from industrial processes and further reductions from the road transport sector. Decreasing animal numbers and a reduction in fertilizer use is expected to reduce ammonia (NH_3) by 10%. Current projections suggest that total UK PM_{10} emissions will remain virtually unchanged between 2010 and 2020.

- There are important differences between the NAEI and the LAEI. Currently, the NAEI provides estimates for more sources than the LAEI, in particular emissions from vehicular tyre and brake wear and construction. However, increased use of local activity data in the LAEI is also likely to account for some of the differences reported.

- Current Department for Transport (DfT) projections forecast that diesel car sales will reach 42% by 2010, from 14% in 2000. The current NAEI forecast of 30% in 2010 means that emissions of PM_{10} will be higher than currently projected. Estimates suggest that UK urban emissions in 2010 will be 3.8% higher than currently projected. The effect on PM_{10} emissions is greater than that for emissions of NO_X, which are projected to be 1.5% higher on the same basis.

- At a local level, a preliminary analysis of the London Congestion Charging Scheme (CCS) estimates that exhaust emissions of PM_{10} have declined by ~12% in the charging zone as a result of reduced congestion and reduced flows of certain vehicle types. The 12% reduction is likely to underestimate the full benefit since the reduction in vehicle flows would also be expected to result in further reductions in PM_{10} resulting from tyre and brake wear as well as resuspended material, which have not yet been quantified.

- No legislation is in place to limit non-exhaust PM emissions through tyre, brake and clutch wear. Thus, given that tailpipe emissions are reducing, the relative contribution of non-exhaust emissions is increasing.

- Uncertainties in total national emissions of PM_{10} in 2001 range from –19% to +46% at the 95% CI. This range is considerably higher than that for the precursor gases: SO_2 (±3%), NO_X and NMVOCs (±8%) and NH_3 (+19% to –18%). It is also highly likely that the uncertainty estimates for particle size fractions less than PM_{10} will be higher than the uncertainty in PM_{10} emissions, although this uncertainty has not been quantified.

- A recent review of certain particle emissions by the NAEI has revised the estimates of particle emissions from important sources. Emissions from road vehicle tyre and brake wear have almost doubled, therefore increasing the non-exhaust particle emission rates. Emissions of PM_{10} from natural gas have been revised downwards, such that emissions are 25% of previous totals. The revision of the natural gas emission rates could significantly affect road source apportionment of particle emissions in urban areas. These revisions, together with others outlined in this report, will be implemented in the 2003 version of the NAEI.

4.1 Introduction

4.1.1 Scope

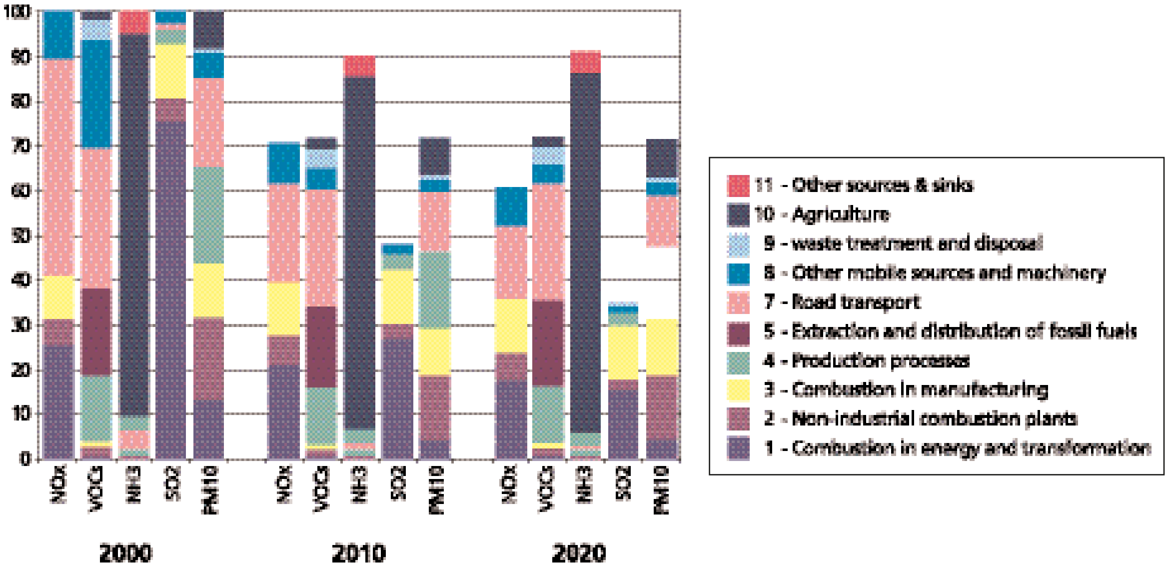
107. This chapter considers the sources of particle emissions in the UK and Europe. It focuses on the sources of emissions for which emission factors exist and is chiefly concerned with the emissions that are considered as part of national and local emissions inventories. Where available, the chapter highlights the characteristics of particle emissions from different source types, such as emissions rates and particle size distributions. Consideration is also given to both the emissions of important particle precursor gases, some of which are of natural origin, and the principal atmospheric chemistry routes that result in secondary particle formation.

108. There are many important natural sources of particles, which contribute significantly to ambient concentrations. Particles from these sources – for example, forest fires and wind blown resuspension – tend to be difficult to predict in terms of their magnitude, emission rate and the description of their temporal emission characteristics. Many sources of natural particles are essentially random in origin and are best described through atmospheric measurements and source apportionment studies. It is generally not possible to identify useful or meaningful emission rates for comparison with manmade sources. Nevertheless, emissions of these particles can, under certain conditions, make a large contribution to observed concentrations of particles and can be significantly more important than manmade primary particles that are generally well characterised in terms of their emission rate. The importance of some natural particle sources is discussed further in Chapter 6.

109. Key data have been considered from both UK and European particle emission inventories and these are presented as both emissions by source type and historical trend data. Where possible, emissions have been summarised by particle size fraction. National emission projections are also highlighted by source sector. Consideration has been given to the effectiveness of technical and non-technical measures to reduce particle emissions by highlighting different abatement technologies, such as vehicle particle filters, as well as the effects of different policies affecting particle emissions, such as the London CCS.

110. This chapter relies extensively on information contained in the NAEI. More detailed information relating to the methodologies for particle emissions inventory compilation and datasets can be found at the NAEI website at http://www.naei.org.uk. An overview of the principal primary and secondary sources of PM_{10} for 2000, 2010 and 2020 is shown in Figure 4.1.

Figure 4.1 UK emissions of PM_{10} and precursor gases relative to 2000 levels, based on a business-as-usual scenario.



4.1.2 Legislation and regulatory framework controlling emissions

4.1.2.1 Control of industrial sources of particles

111. Most industrial sources of particles are subject to control regimes set up by the Pollution Prevention and Control (England & Wales) Regulations 2000, the Pollution Prevention and Control (Scotland) Regulations 2000 and the Pollution Prevention and Control (Northern Ireland) Regulations 2003 which were introduced under the Pollution Prevention and Control Act 1999. Further information on these regulations can be found in the AQEG NO_2 report (AQEG, 2004) and a table listing how the major industrial sources of PM are covered by the different control regimes in England and Wales is available in Annex 1.

112. In general, the introduction of the regimes as a result of the Pollution Prevention and Control Act 1999 will not significantly change the types of conditions placed upon operators with regard to PM emissions from those issued under the older regimes set up by the Environmental Protection Act 1990.

113. For many processes where emissions of PM occur in a manner largely controlled through venting, the most important condition will be an emission limit, expressed in terms of a maximum allowable concentration of PM in the vented gases. For each process, emission limits are set by the regulators based on an assessment of what can be achieved using 'best available techniques not entailing excessive cost' (BATNEEC). In most cases the limits set are likely to correspond to those values suggested in guidance documents published by Defra and regulators. For example, the Environment Agency's guidance for combustion processes suggests that emission limits of 25 mg m^{-3} or less are achievable for coal-fired plants, depending upon the appliance type (Technical Guidance Note S3 1.01). Emission limits for most processes subject to LAPC are set at 50 mg m^{-3}.

114. Certain industrial processes are also subject to additional EC Directives, including waste incinerators and large combustion plants. The Waste Incineration Directive (2000/76/EC) replaces earlier directives and will set an emission limit of 10 mg m^{-3} for dust from nearly all UK incineration processes from 2006 onwards. In contrast, most plants are currently meeting emission limits of 30 mg m^{-3} or more.

115. The Large Combustion Plant Directive (2001/80/EC), which updated the earlier Directive 88/609/EC – introduced controls for combustion plants with an output at or above 50 MWth. New plants (those granted a licence after 1 July 1987) must comply with emission limits for PM, NO_X and SO_2. Existing plants must currently meet emission limits set out in the earlier directive but, from 2008 onwards, these plants will be required either to meet new emission limits or to comply with a 'national emissions reduction plan', which would achieve the same overall reduction in emissions. At the present time the Large Combustion Plant Directive is not transposed into UK law and the UK has still to decide how to achieve compliance. The latest UK Government statement indicates it hopes to adopt a modified approach that combines both these strategies, but whatever it decided in the end, individual plants may opt out and instead accept a limit on their operating hours for the period 2008–2015.

116. The Environmental Protection Act 1990 and the IPPC Directive also deal with emissions of the particulate precursor pollutants NO_X, SO_2 and NMVOC in a similar fashion to the controls placed on emissions of primary PM. As already mentioned, the Large Combustion Plant Directive introduced controls on NO_X and SO_2 from combustion plant with an output at or above 50 MWth. Additional legislation covering NMVOC emissions includes EC Directives on solvent use and petrol distribution.

4.1.2.2 Control of emissions from road vehicles

117. Particulate emissions from new diesel vehicle exhausts have been regulated by EU Directives since the early 1990s. The Directives set limits on emissions of particulates during Type Approval testing over specified vehicle or engine test cycles, some of which have changed in recent years in order to make the tests more representative of the performance of the vehicle on real road conditions. Since the first set of regulations – frequently referred to as Euro I – came into effect in 1992–1993, a succession of more stringent Directives have been brought in to further reduce particulate emissions from both light and heavy duty diesel vehicles, currently extending to tougher limits on new vehicles sold after 2008 (Euro IV vehicle standards (Official Journal, 1999)). Even tighter standards for new vehicles sold after 2010 (Euro V) are currently being considered by the EC.

118. There are currently no regulations on particulate emissions from petrol engines, but for conventional petrol engine technologies, particulate emissions are much lower than they are from diesel engines. However, emissions of NO_X and hydrocarbons, which are precursor pollutants to particulates in the atmosphere, are regulated for both petrol and diesel vehicles (AQEG, 2004). Relatively small amounts of the precursor pollutants NH_3 and SO_2 are emitted from vehicles, but their emissions are not regulated. Emissions of NH_3 can be affected by abatement technologies designed to control NO_X emissions, sometimes increasing their emissions (AQEG, 2004). Emissions of SO_2 are mainly dependent on the sulphur content of the fuels used.

119. The Type Approval limit values on PM, NO_X and hydrocarbons set by the various Directives for the different vehicle types since 1990 are summarised in Annex 2.

120. As well as emission limits for Type Approval testing of new vehicles and engines, the Roadworthiness Testing Directive 96/96/EC and its amendment 1999/52/EC sets limits during in-service testing of vehicles (the MOT). Rather than by mass, particulate emissions are limited according to monitoring by a free acceleration smoke opacity test.

121. The introduction of tighter vehicle emission standards has been accompanied by the introduction of tighter fuel quality standards. These have led to a continuous reduction in the sulphur content of petrol and diesel, which has been necessary to pave the way for the development of emergent exhaust treatment technologies successful in reducing primary particulate emissions and emissions of precursors. For example, most types of particulate trap technologies developed for diesel engine exhausts will only work using fuels with sulphur content below 50 ppm. The presence of sulphur in fuels seriously affects the performance of many abatement technologies. EU Directive 98/70/EC introduced successively tighter limits on the sulphur content of petrol and diesel. The maximum permitted level of 50 ppm (to be achieved by 2005) is already met by virtually all fuels supplied at UK filling stations. Directive 2003/17/EC has set a limit of 10 ppm on the levels of sulphur in petrol and diesel that can be sold from 2009. Even in the absence of particulate emission abatement devices fitted on diesel engine exhausts, reducing the sulphur content of diesel alone reduces particulate emissions by small amounts.

4.1.2.3 Control of emissions from other transport and machinery

122. There are no direct limits or controls on PM emissions from shipping, however there are controls on SO_2 emissions, which will also reduce emissions of PM. The Sulphur in Liquid Fuel Directive (1999/32/EC, see Section 4.1.2.4) does not cover marine heavy fuel oil but another international agreement (MARPOL) sets a limit of 1.5% for heavy fuel oil used in the North Sea, English Channel and Baltic Sea. A revision to the Sulphur in Liquid Fuel Directive has been proposed that would introduce the same or, in some instances, tighter limits into the Directive.

123. Emissions of NO_X, CO, unburned hydrocarbons and smoke from aircraft engines are subject to standards agreed by the International Civil Aviation Organisation (ICAO). These standards have been implemented in the Air Navigation (Environmental Standards) Order 2002.

124. Emissions of particulates, NO_X and hydrocarbons are regulated for 18–560 kW diesel engines used for non-road mobile machinery. These cover many different types of off-road machinery used in construction, mining, quarrying, industry and agriculture. Directive 97/68/EC defined emission limits (in g/kWh) to be implemented in two stages, between 1998 and 2003, depending on the power rating of the engine (Official Journal, 1998). Directive 2000/25/EC covered emission limits for agricultural tractors (Official Journal, 2000). These engines normally run on gas oil, a diesel fuel with higher sulphur content (currently 1500 ppm) than road diesel. As the sulphur content of this fuel is likely to be reduced, the effect this has on primary particulate emissions is not known. The EC has recently proposed an amendment to Directive 97/68/EC to cover diesel engines on railcars, railway locomotives and inland waterway vessels.

4.1.2.4 Control of emissions from other sources

125. The Environmental Protection Act 1990 banned the burning of crop residues in England and Wales with limited exemptions from 1993 onwards. Crop burning is still allowed in Scotland and Northern Ireland but is not common practice.

126. The Clean Air Act of 1956 introduced the concept of Smoke Control Areas, where domestic combustion of coal is prohibited and only designated smokeless solid fuels can be used. This Act has led to a major reduction in levels of PM in urban areas.

127. The Sulphur in Liquid Fuel Directive sets limits on the sulphur content of heavy fuel oil and gas oil burnt by sectors other than shipping.

4.1.2.5 National emissions ceilings

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

129. Within the EU, the National Emissions Ceilings Directive set emissions ceilings for 2010 for each Member State for the same four pollutants listed in the Gothenburg Protocol (Official Journal, 2001). A number of Member States reduced their ceilings somewhat below the levels included in the Protocol. The UK reduced its ceiling for NO_X (as NO_2) emissions from 1181 kt set by the Gothenburg Protocol to 1167 kt set by the National Emissions Ceilings Directive. The UK ceilings for SO_2, NMVOCs and NH_3 set by the National Emissions Ceilings Directive are 585 kt, 1200 kt and 297 kt, respectively.

4.2 Sources of primary emissions

4.2.1 Estimation of emissions

130. Primary particle emissions can be estimated using several approaches, depending on the source type being considered. Often direct measurements of particles from different sources are available as well as information on how emissions vary in relation to the activity being undertaken. One example of this would be the road transport sector. Information is often available on the composition and size distribution of particles from sources within this sector. For other sectors data that characterise particle emissions are also available, but generally less information is available compared with the road transport sector. Often, therefore, emissions from a particular sector are derived using generic fuel-related factors. These factors have been derived for common fuel types such as coke, coal and fuel oil. Emissions from combustion processes are, in principle, straightforward to quantify and characterise, although often there is insufficient information available to do so.

131. Often it can be difficult to quantify primary emissions of particles. Many sources are often transient in nature, for example, emissions from fires or some agricultural emissions. Not only does the transient nature of these emissions make it difficult to quantify quantities of emissions released to the atmosphere, but the characteristics of the sources themselves can be very variable. In the case of agricultural or quarrying emissions, for example, the emissions can be dependent on other factors such as meteorology. This makes it very difficult to be specific about the quantity of material released to the atmosphere or the physical and chemical characteristics of the material. The difficulty in quantifying emissions from these sources also presents difficulties for dispersion modellers, who require emissions input data in the form of a mass release per unit time to derive ambient concentrations. Often, as highlighted in Chapter 6, it is necessary to rely on atmospheric measurements instead of directly inferring the concentrations from these sources.

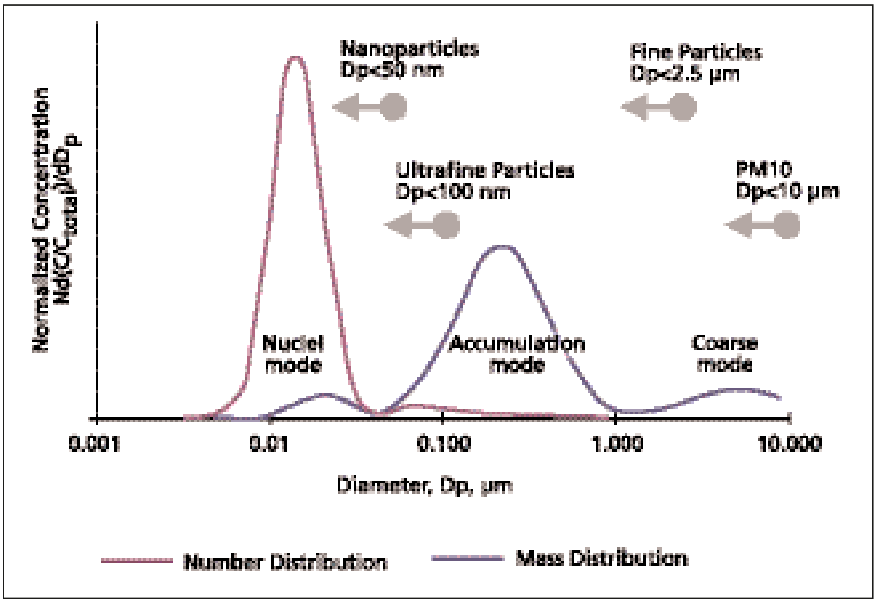
4.2.2 Transport

4.2.2.1 Particle formation mechanisms and definitions

132. An understanding of the mechanisms by which exhaust particles are formed is crucial to the interpretation of measurements and to the understanding of their limitations. The formation mechanisms and the ways in which particles are defined are closely linked.

133. Particles measured in diesel engine exhaust typically exhibit a trimodal distribution. The three distinct size modes are the nucleation mode (sometimes referred to as nuclei or nanoparticles), the accumulation mode and the coarse mode. Typical number-weighted and mass-weighted particle size distributions are shown in Figure 4.2.

Figure 4.2 Typical particle size distributions and definitions as measured in diesel vehicle exhaust (source: Kittleson, 1998).



134. The nucleation mode consists of primary particles that are composed mainly of lower volatility organics and sulphur compounds, but also some solid carbon and metallic particles. The nucleation mode has traditionally been defined as particles with a diameter of 5–50 nm. However, on the basis of the E-43 project in the US, Kittelson *et al*. (2002) have suggested that this range ought to be redefined as 3–30 nm. The accumulation mode ranges in size from ~30–500 nm. As its name suggests, this mode consists of particles formed by agglomeration of the smaller primary particles. The coarse mode consists of particles larger than 1,000 nm, which are formed by re-entrainment of material previously deposited on cylinder and exhaust system surfaces (Kittelson, 1998).

135. The general process of particle formation during combustion has been described in detail by Heywood (1988). Particle formation begins with the creation of carbonaceous material (soot) in the cylinder. Condensation reactions of gas-phase species such as unsaturated hydrocarbons and PAHs lead to the appearance of the first recognisable solid material (soot spherules) at combustion temperatures of between 1000 K and 2800 K. A phase of particle growth then follows inside the cylinder that includes surface growth of spherules by the adsorption of gas-phase components as well as by their coagulation and agglomeration. At temperatures above 500°C the particles are principally clusters of spherules, with individual spherule diameters of 15–30 nm. Once these clusters have left the cylinder they are then subject to a further mass addition process as the exhaust gases cool; as temperatures fall below 500°C, the particles become coated with adsorbed and condensed high-molecular weight organic compounds. Almost all of the particles found in the exhaust pipe before dilution are present as these carbonaceous agglomerates (accumulation mode particles), with a small amount of metallic ash and adsorbed materials.

136. However, the processes leading specifically to the presence of nucleation mode particles during measurement are not well understood. The nucleation mode can be viewed as a rather ephemeral and highly variable phenomenon. Most nucleation mode particles (up to 90%) are thought to originate from the condensation of lower volatility organics in the exhaust gas during exhaust dilution, rather than during combustion. However, nucleation is a non-linear process and the number of particles formed is very sensitive to the nature of the sampling conditions.

137. If a nucleation mode is observed during measurement – and this is not always the case – then the number of particles it contains will greatly exceed the number in the accumulation mode, although the nucleation mode particles contribute little to the total mass. The nuclei mode typically contains 0.1–10% of the particle mass and up to 90% or more of the particle number. Most of the mass is found in the accumulation mode, with the coarse mode typically accounting for 5–20%.

4.2.2.2 The regulatory test for exhaust emissions of PM

138. For exhaust emissions of PM[1], current Type Approval legislation in Europe only applies to heavy duty diesel engines and light duty vehicles powered by diesel engines. The legislation does not take into account the presence of the different particle modes and simply requires gravimetric measurement of the total mass of particles collected on a filter during the Type Approval test. The units used for reporting are g/kWh for heavy duty engines and g/km for light duty vehicles.

139. The legislation prescribes the use of a full-flow dilution system, known as a constant volume sampler (CVS), as the reference procedure for certification. The entire exhaust stream from the engine or vehicle is mixed with ambient air in a dilution tunnel, and a constant total air flow rate is maintained under all running conditions. The use of a constant air flow rate simplifies the calculation of mass-based emission factors. For light duty vehicles a single dilution tunnel is normally sufficient, but it is now customary to use a secondary dilution tunnel for heavy duty applications. This secondary dilution tunnel helps to accommodate the high exhaust flow rates from large-volume engines. Full specifications of this sampling system are given in the associated EU directive and are discussed further elsewhere (Moon and Donald, 1997; Stein, 2001; Kittelson *et al*., 1999).
For the sampling of PM, a portion of air is drawn from the dilution tunnel through a stainless steel probe configured with a 'Chinese hat'-type design facing into the exhaust stream; the gas is then passed through two series-mounted Teflon-coated glass fibre filters[2] that have been pre-weighed and preconditioned. A temperature of 51.7°C must not be exceeded at the sampling point. The exposed filter papers, after post-conditioning under controlled temperature and humidity conditions, are weighed using a microbalance. As with the sampling of PM in ambient air (see Chapter 5), the material collected on the filter includes not only solids but also liquid material, which would condense in the form of droplets at the test temperature, such as H_2SO_4 and high-boiling point hydrocarbons.

140. It is interesting to note that Switzerland has made an official statement saying that not only the mass but also the number of solid particles in the size range 20–300 nm must be measured and curtailed. This was in response to the Swiss legal requirement to minimize carcinogenic air pollutants using BAT, which currently includes particulate traps with efficiencies of >99%.

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[1] It is conventional to use the term 'particle' to refer to the airborne state and to use 'particulate' or 'PM' to refer to the material collected on a filter under the conditions of the Type Approval test.

[2] The second filter acts as a back-up to check for sample breakthrough.

On 1 September 2002, a trap retrofit regulation for diesels >37 kW used in all larger construction operations where traps must be VERT-verified (a verification system for particulate control devices derived from a research programme from 1994–2000, sponsored by Swiss, German and Austrian health authorities) to reach >95% filtration based on solid particle number definition came into force. Although Switzerland is free to regulate retrofit measures in highly polluted areas, Type Approval for new vehicles must be in accordance with EU regulations.

141. The limitations of the regulatory test method, in-service emissions testing and particle research programmes are discussed in Annex 3.

4.2.2.3 Particle size and number emissions by vehicle and fuel type and legislation class

142. Concerns have been raised that modern diesel engines, which emit a low mass of particles, have high emissions of particles in the nanometre size range. For example, in a study of the steady-state emissions of a (then) new technology diesel engine, Baumgard and Johnson (1996) observed a particle concentration of 10^9 cm^{-3}, around three orders of magnitude greater than expected. Bagley *et al*. (1996) found that there was a 15- to 35-fold increase in total particle number from a 1991 diesel engine compared with a 1988 engine, both running on very low-sulphur fuel. This was due to a 30- to 60-fold increase in the number of primary particles and occurred despite a substantial reduction in the mass of PM.

143. Ntziachristos and Samaras (2003) evaluated three diesel passenger cars manufactured under conditions of improving technology and operated on identical fuel. The vehicles selected corresponded to Euro 0 (pre1992), Euro I (1992–1996) and Euro III (2000–2005) emission standards. The cars were driven over a range of operational conditions, including three steady-state speeds (50, 90 and 120 km/h), one cold urban cycle, one hot urban cycle and an extra-urban cycle. A number of observations indicated emerging priorities for exhaust particle characterisation. PM reductions brought by technology improvements were not consistently accompanied by decreases in total particle number concentration and surface area. Hence, the introduction of additional metrics seems justified for more effective particle emission understanding and control. On the other hand, technology improvement was strongly apparent in the solid particle number concentration. This may provide an additional, more sensitive, measurement complementing PM mass emissions. Moreover, the median size of the solid particles was the least sensitive metric to either vehicle technology improvements or vehicle operation condition. This information may potentially be used to distinguish diesel particle emissions from other sources. Finally, all measurements referring to total particle emissions (except mass) showed a higher sensitivity to vehicle operation conditions rather than vehicle technology. This observation is consistent with the formation of nuclei mode particles that are not discerned from the simple PM measurements. Clearly, then, measurements of additional parameters – such as total particle concentration and surface area – measured according to well-defined test protocols are necessary to fully explore emission performance under varied operational conditions.

144. With respect to vehicle type it should be noted that although diesel engines emit higher concentrations of particles than gasoline engines, the total number of vehicle kilometres travelled by gasoline-powered vehicles in the UK is greater than that of diesels. Additionally, vehicles powered by spark ignition (SI) engines typically emit smaller particles than vehicles powered by diesel engines and are an important source of fine particles and nanoparticles. It has been estimated that two-thirds of the total fine particle mass occurs from SI engines (Latham *et al.*, 2001). New gasoline direct injection (GDI) engines emit much higher particle concentrations than conventional engines and may approach diesel levels under some conditions (Kittelson, 2000).

145. The EU Fifth Framework clustered projects ARTEMIS and PARTICULATES were designed to develop new exhaust emission factors for all transport modes. In particular the PARTICULATES project focussed on the development of a measurement protocol for in-service emissions of PM. One of the outputs of these projects was the provision of average speed-based emission factors for a range of particulate metrics – including particle size distributions, the number of particles and their surface area – for Euro I to Euro IV standards and for a range of fuel types and after-treatment devices. Results from this programme are expected in early 2005.

146. The measurement of various particulate metrics, in addition to mass, has highlighted the importance of sampling conditions. Diesel light duty vehicles generally emit $>10^{14}$ particles/km, measured as solid particles using an ELPI. Direct injection spark ignition engines emit 10–20-times fewer particles than the equivalent diesel-engined vehicle (Ntziachristos *et al*. 2004b).

147. Particulate emissions from heavy duty engines are higher than those associated with light duty engines. Total particle emissions associated with conventional Euro I to Euro III heavy duty engines are in the range 10^{14} particles/kWh. When operated with low sulphur fuels, vehicles equipped with PM traps generally produce very low mass emissions, low numbers of carbonaceous particles and low total numbers of particles. This effect of low sulphur fuels is greatest when associated with high speed and temperature operation (Thompson *et al*. 2004).

148. Several initiatives, including the PMP programme, are investigating the possibility of supplementing the PM Type Approval procedure with a standard based upon particle numbers. Whereas the PMP programme is expected to issue a recommendation during 2005, the Swiss BUWAL has proposed a standard of 10^{14} particles/km for light duty diesel vehicles, reducing to 10^{11} particles/km for those vehicles fitted with a diesel particulate trap. No proposed Type Approval standard has been set for heavy duty vehicles.

4.2.2.4 Estimating emissions from road transport

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

150. The methods used for calculating emissions from road transport in an emissions inventory were described in detail in the AQEG report on NO_2. (AQEG, 2004). Only specific points related to the calculation of emissions of PM_{10} from road vehicles in the most recent (2002) version of the NAEI are commented on here.

Table 4.1 Exhaust emission factors for PM (g km^{-1}) for different road types used in the NAEI , based on data from TRL (Barlow *et al*., 2001).

Vehicle type	Euro class	Urban	Rural	Motorway
Petrol cars	ECE 15.04	0.0241	0.0151	0.0182
	Euro I	0.0029	0.0040	0.0089
	Euro II	0.0008	0.0014	0.0054
	Euro III	0.0008	0.0014	0.0054
	Euro IV	0.0008	0.0014	0.0054
Diesel cars	Pre-Euro I	0.154	0.131	0.167
	Euro I	0.057	0.049	0.077
	Euro II	0.051	0.037	0.058
	Euro III	0.032	0.023	0.036
	Euro IV	0.017	0.012	0.019
Petrol LGVs	Pre-Euro I	0.0300	0.0241	0.0696
	Euro I	0.0038	0.0066	0.0142
	Euro II	0.0009	0.0013	0.0048
	Euro III	0.0009	0.0013	0.0048
	Euro IV	0.0009	0.0013	0.0048
Diesel LGV	Pre-Euro I	0.287	0.243	0.358
	Euro I	0.080	0.079	0.195
	Euro II	0.082	0.081	0.200
	Euro III	0.057	0.057	0.141
	Euro IV	0.037	0.037	0.090
Rigid HGVs	Pre-1988	0.890	0.730	0.718
	88/77/EEC	0.426	0.350	0.344
	Euro I	0.220	0.180	0.177
	Euro II	0.153	0.124	0.123
	Euro III	0.111	0.089	0.089
	Euro IV (2006)	0.025	0.019	0.019
	Euro IV (2008)	0.025	0.019	0.019
Artic HGVs	Pre-1988	0.714	0.556	0.488
	88/77/EEC	0.626	0.487	0.428
	Euro I	0.577	0.474	0.466
	Euro II	0.399	0.323	0.321
	Euro III	0.288	0.233	0.231
	Euro IV (2006)	0.064	0.049	0.049
	Euro IV (2008)	0.064	0.049	0.049
Buses	Pre-1988	1.416	0.647	0.551
	88/77/EEC	0.614	0.281	0.239
	Euro I	0.311	0.169	0.167
	Euro II	0.203	0.117	0.116
	Euro III	0.147	0.084	0.084
	Euro IV (2006)	0.044	0.018	0.018
	Euro IV (2008)	0.044	0.018	0.018
Motorcycles (2 stroke)	All	0.04	0.04	0.04
Motorcycles (4 stroke)	All	0.12	0.12	0.12

Key: LGV, light goods vehicle; HGV, heavy goods vehicle.

4.2.2.5 Exhaust emission factors for PM

151. Surveys of road vehicle exhaust emission factors are on-going within the UK and elsewhere. The last revision to the UK database was undertaken by TRL on behalf of the DfT (Barlow *et al.*, 2001). The NAEI incorporates data from the TRL emission factor database for vehicles up to Euro II standards (that is, pre-2000 models). A substantial part of the measurements of Euro I and Euro II emission factors come from several test programmes funded by DfT and Defra and carried out at UK test laboratories between 1999 and 2001 (Barlow *et al.*, 2001). The measurements were made on dynamometer test facilities under various simulated real-road drive cycles.

152. Table 4.1 shows PM emission factors for each class of vehicle calculated from the emission factor equations at typical speeds on urban, rural and motorway roads. These emission factors are currently used in the NAEI. The vehicle types match the traffic activity data that are used in the national emission calculations. The emission factors in this table refer to vehicles running on ultra-low sulphur fuels, with a sulphur content of around 50 ppm, fuels which are currently available at most filling stations around the UK. In cases where the vehicles tested were running on diesel fuels with higher sulphur content, as would have been the norm in the 1990s when the emission measurements were made, the measured emission factors were corrected in deriving the factors shown in Table 4.1. The fuel correction factors were based on various studies and empirical equations relating exhaust emissions to fuel quality taken from the European EPEFE Programme (EPEFE, 1995).

153. Emissions from Euro III and Euro IV classes must be estimated through an examination of the proposed future legislation and the technical ability to meet these new limits. This is because either the emission standard has yet to be introduced at all or because insufficient numbers of vehicles have been tested, the Euro standard having only recently entered the fleet. As discussed in the AQEG report on NO_2 this estimation process can result in significant uncertainties in future emissions factors (AQEG, 2004).

154. Table 4.1 provides emission factors for Euro III and Euro IV vehicles, the latter referring to standards that will come into effect in 2006. These are based on scaling factors that are assumed to apply across all speeds relative to emission factors for Euro II vehicles. For diesel light duty vehicles, the scaling factors were estimated by the NAEI on the basis of current emission levels for Euro II vehicles and the amount emissions will need to be reduced to achieve the Type Approval limit values, taking into account differences in the drive cycles used and the contribution that cold start emissions will make to emission over the new 98/69/EC test cycle. Consideration was given to the emissions durability requirements of vehicles set in the Directive. This states that when using a deterioration factor of 1.2 to the emission factor of the vehicle when new, PM emissions from light duty diesel vehicles must be below the limit value after 80,000 km for Euro III vehicles and 100,000 km for Euro IV vehicles. The emission factors for in-service Euro III and IV vehicles are therefore taken to be a factor of 1.2 lower than the amount derived from the limit value when new and are then allowed to increase by a factor of 1.2 at a linear rate over 80,000 km (Euro III) or 100,000 km (Euro IV). The figures in Table 4.1 represent the value of the Euro III and IV emission factor for new vehicles. As no limit values are set by the Directives on PM emissions from petrol vehicles, emission factors for Euro III and IV vehicles are retained at Euro II levels.

155. For heavy duty vehicles, it was assumed that Euro II vehicles are currently at their emission limits and will be reduced no more than required to achieve the new emission limits for Euro III and IV, using scaling factors from the COPERT III database (EEA, 2000). COPERT III is the methodology and database for calculating road transport emission factors. It was developed by a group of experts for the European Environment Agency (EEA) and is recommended for national emission inventory reporting under CORINAIR.

4.2.2.6 Road transport activity data

156. To calculate UK emissions from road transport for the NAEI, the hot exhaust emission factors are combined with annual UK vehicle kilometre data for different vehicle types on different roads and data on the composition of the vehicle fleet by age, size and fuel mix. Further details are provided in the AQEG report on NO_2. (AQEG, 2004). The NAEI mainly draws on transport statistics published by the DfT and the Transportation Unit, DoE, Northern Ireland.

157. The NAEI makes an assumption that a small proportion of cars fitted with three-way catalysts fail each year. However, it is believed that current car models and future models are more durable than the first generation (Euro I) catalyst cars. This is as a result of on-board diagnostic systems fitted to modern vehicles and the more stringent durability requirements set out in the Directives. Although there are no definitive data available on catalyst failure, on the basis of discussions with experts in industry and at the DfT and evidence, for example, from studies analysing MOT test data, equipment warranty returns and car fleet maintenance records (DfT, 2002), the NAEI has adopted different failure rate assumptions for each Euro class as follows:

- Euro I cars: 5% per annum;
- Euro II cars: 1.5% per annum; and
- Euro III–IV cars: 0.5% per annum.

158. It is assumed that 95% of failed catalysts are repaired after the vehicle is three years old and reaches the age of MOT testing.

4.2.2.7 Cold start emissions

159. The excess emissions that arise when the vehicle is started with its engine cold or below its optimum operating temperature are calculated using a method taken from COPERT III. The procedure uses equations that take account of the effects of ambient temperature on the excess emissions and its effect on the distances travelled with the engine cold. The distances travelled with cold engines also depend on average trip lengths: the shorter the average trip lengths are, the greater the contribution of cold start emissions. The method calculates the ratio of cold to hot exhaust emissions for light duty vehicles that are used to calculate the overall cold start emissions from these vehicles.

160. Further details on the emission calculation methodology used in the NAEI for road transport are provided in the NAEI report (Goodwin *et al*., 2001).

4.2.2.8 Non-exhaust road transport PM

161. The PM generated by road transport activity can be categorised according to its mode of formation. It is generally assumed that fuel combustion is the primary mechanism by which particles are formed, and a considerable body of research on vehicle exhaust particulate emissions has been compiled. However, there are a number of other processes, involving mechanical abrasion and corrosion, that can also result in PM being released directly to the atmosphere. These processes include: tyre wear; brake wear; clutch wear; road surface wear; corrosion of chassis, bodywork and other vehicle components; and corrosion of street furniture and crash barriers.

162. The abrasion and corrosion processes can also lead to the deposition of particles on the road surface. The material that collects on the road surface, often referred to as 'road dust', may also contain exhaust particles, de-icing salt and grit deposited during winter maintenance and matter from a range of sources that are not related to road transport (for example, crustal and vegetative material and material from industrial/commercial/domestic activity). Road dust may subsequently be suspended or resuspended in the atmosphere as a result of tyre shear, vehicle-generated turbulence and the action of the wind. Clearly, the source apportionment and quantification of non-exhaust PM measured in the vicinity of roads is a rather complex task (Luhana *et al*., 2004).

163. Non-exhaust emission sources may contribute significantly to atmospheric particle concentrations. However, the data relating to the emission rate, size and composition of particles arising from such sources are far from comprehensive, and more accurate emission factors are required for emission models and inventories.

4.2.2.9 Road vehicle tyre wear

164. Relatively little data exists that quantifies the emission rates, size distribution and composition of vehicular tyre wear. Different experiments have, however, been carried out to quantify emissions of particles from tyre wear. These experiments have included the collection of samples close to roads and tests undertaken on dynamometers (rolling roads).

165. The NAEI uses g/km emission factors to estimate PM emissions from tyre and brake wear. However, the method used to estimate the emission factors for the 2002 NAEI has been revised from earlier versions of the inventory in light of a recent review of PM emissions from these sources carried out for the UNECE Task Force on Emission Inventories[3] and the EMEP/CORINAIR Emission Inventory Guidebook[4]. The review examined a number of experimental studies on the wear of tyre and brake material and the amount that becomes airborne. Tyre tread wear is a complex physiochemical process that is driven by the frictional energy developed at the interface between the tread and the road pavement (Veith, 1995). The rate of wear varies enormously depending on a combination of factors, including tyre characteristics, vehicle characteristics, road surface characteristics and vehicle operation. Typical wear rates of 10–90 mg km^{-1} per tyre have been reported, with 1–15% by mass of passenger car tyre wear material emitted as PM_{10}. A recent measurement campaign by TRL, involving six mileage accumulation vehicles, derived a typical factor of 74 mg km^{-1} for tyre wear (Luhana *et al*., 2004).

4.2.2.10 Road vehicle brake wear

166. During forced deceleration, vehicle brake linings are subject to large frictional heat generation and associated wear. This mechanical process generates particles from both the brake lining and the brake surface.

167. The wear rate of brake linings for passenger cars has been estimated to be around 10–20 mg vkm^{-1} and up to three- or four-times higher for heavy duty vehicles. Although there have been only limited studies, it appears that the proportion of material emitted as PM_{10} is higher from brake wear than from tyre wear (USEPA 1995; TNO, 1997). Typical PM_{10} emission factors of 1−9 mg vkm^{-1} have been reported for light duty vehicles and 20−40 mg vkm^{-1} for heavy duty vehicles. However, these same studies have concluded that the proportion of brake wear PM with an aerodynamic diameter of <0.1 μm could be as high as 33% of the total brake-wear PM emission.

4.2.2.11 Estimation of tyre and brake wear emissions

168. For both tyre and brake wear, the UNECE method provides emission factors for different vehicle types and provides speed correction factors. These imply higher emissions per km at lower speeds than at high speeds. For heavy duty vehicles, a load correction factor is also provided and tyre wear emissions depend on the number of axles.

169. Based on average speeds on UK roads, Table 4.2 shows emission factors for different vehicle types on different road types. Units are in g of PM_{10} per vehicle km, but in the case of tyre wear emissions from heavy duty vehicles, units are in g PM_{10} per axle km.

170. Use of these new emission factors in the NAEI has almost doubled the estimate of tyre and brake wear emissions in the UK in 2001 compared with previous estimates, mainly due to increased estimates for tyre wear emissions. However, the emission factors must still be regarded with a high degree of uncertainty.

4.2.2.12 Road vehicle resuspension

171. The resuspension of particles by road vehicles occurs through two principal mechanisms. First, particles can be resuspended from the wear and shear of tyres acting on the road surface. Second, particles can be resuspended as a result of the turbulence caused by moving vehicles. Since resuspended material is non-exhaust in origin, there are no direct measurements available that quantify emissions in terms of g km^{-1}. Most estimations of the importance of resuspension have been based on receptor modelling techniques at specific locations. In many cases, however, these studies suggest that vehicular resuspension is as important, if not more important, than exhaust emissions of particles from vehicles. The studies also indicate that HGVs are significantly more important in terms of resuspension compared with smaller vehicles such as passenger cars. Because the significance of resuspension is governed by many factors (for example, vehicle type, road surface condition and meteorological conditions), resuspended material is highly variable in terms of its source emission rate. It is therefore difficult to derive representative emission rates that can be expressed in terms of g km^{-1}.

Table 4.2 Average PM_{10} emission factors for tyre and brake in the UK.

		Tyre wear	**Brake wear**
Cars	Urban	0.00874 g km^{-1}	0.0117 g km^{-1}
	Rural	0.00680 g km^{-1}	0.0055 g km^{-1}
	Motorway	0.00579 g km^{-1}	0.0014 g km^{-1}
LGVs	Urban	0.01380 g km^{-1}	0.0182 g km^{-1}
	Rural	0.01074 g km^{-1}	0.0086 g km^{-1}
	Motorway	0.00915 g km^{-1}	0.0021 g km^{-1}
HGVs	Urban	0.00918 g axle km^{-1}	0.0510 g km^{-1}
	Rural	0.00737 g axle km^{-1}	0.0271 g km^{-1}
	Motorway	0.00608 g axle km^{-1}	0.0084 g km^{-1}
Buses	Urban	0.00937 g axle km^{-1}	0.0536 g km^{-1}
	Rural	0.00737 g axle km^{-1}	0.0271 g km^{-1}
	Motorway	0.00608 g axle km^{-1}	0.0084 g km^{-1}
Motorcycles	Urban	0.00376 g km^{-1}	0.0058 g km^{-1}
	Rural	0.00292 g km^{-1}	0.0028 g km^{-1}
	Motorway	0.00249 g km^{-1}	0.0007 g km^{-1}

There is also a potential difficulty with double counting in inventories because a proportion of particles assumed to be resuspended in origin might have already been accounted for elsewhere. Nicholson (2000) has estimated a mean PM_{10} emission factor of 40 mg km^{-1} for the UK based on the current UK mix of vehicles.

172. A comprehensive set of recent measurements undertaken in Nevada and North Carolina quantified the contributions from tyre wear, brake wear and resuspension for a range of conditions (Abu-Allaban *et al*., 2003). Measurements were made at eight locations and the emissions were quantified for light and heavy vehicles. Typically, exhaust particles were within ±30% at the different sites, whereas the other non-exhaust particles varied greatly between sites. The resuspended component depended on the vehicle speed, the degree of road silting and the proportion of heavy vehicles. Brake wear was only important on exit from a freeway. No evidence was found of tyre wear at any location. Furthermore, no statistically significant difference was found between the locations in Nevada and North Carolina for the non-exhaust particle emission rates calculated. Table 4.3 shows the key results from the study for PM_{10}. Of interest is the comparison between light and heavy vehicles since many of the particle components will depend on the specific conditions in Nevada and North Carolina, which have different climatic conditions compared with the UK. These results show that exhaust emissions from heavy vehicles are a factor of seven higher than those from light vehicles; the resuspended and brake-wear component is a factor of ten higher for heavy vehicles. The mean emission rate factor calculated in the Abu-Allaban study for heavy vehicles – 2.247 g km^{-1} is more than factor of five higher than the total non-exhaust particle emissions that were estimated for heavy vehicles on Marylebone Road (see Chapter 6).

Table 4.3 Emission factors for different sources of road vehicle PM_{10} (mg km^{-1}) (adapted from Abu-Allaban *et al.*, 2003).

Vehicle type	Total emission	Exhaust	Resuspension	Brake wear
Heavy	2570±1297	211±47	2247±617	124±71
Light	265±126	29±7	224±59	12±8

4.2.2.13 Other non-exhaust road vehicular emissions

173 Emissions from vehicles also arise from clutch wear and the corrosion of vehicular materials. Very little information exists on the likely composition and magnitude of these emissions, although these sources could make a non-negligible contribution to non-exhaust emissions.

4.2.2.14 Emission factors for other transport

174 Emissions are estimated from other transport sources including shipping, railway locomotives and aircraft. Emission estimates are based on the use of emission factors combined with either fuel-use statistics or, in the case of aircraft, numbers of take-off and landing cycles. Estimates for rail locomotives attempt to reflect the differences in emissions from different types, but estimates for aircraft and shipping are more simplistic. Further details of the emission estimation methodology for these transport sectors are given in the AQEG report on NO_2 (AQEG, 2004). Emission factors and emissions estimates from the most important non-road transport sources are shown in Table 4.4.

Table 4.4 Emission factors and emission estimates for PM_{10} from non-road transport.

Source	Emission factor	Emission in 2001 (kt)
Aircraft	0.204 kg t^{-1} fuel	0.2
Coastal shipping	1.07 kg t^{-1} gas oil	0.7
Fishing vessels	1.07 kg t^{-1} gas oil	0.1
Naval shipping	1.07 kg t^{-1} gas oil	0.3
Intercity trains	2.23 kg t^{-1} gas oil	0.6
Freight trains	0.816 kg t^{-1} gas oil	0.1
Agricultural vehicles	6.04 kg t^{-1} gas oil	2.0
Industrial off-road vehicles	4.10 kg t^{-1} gas oil	5.1
Airport support vehicles	3.93 kg t^{-1} gas oil	0.1
Garden machinery	8.70 kg t^{-1} DERV	0.1

4.2.2.15 Rail

175. Emissions from railways, that is, diesel locomotives, are estimated from fuel consumption estimates using fuel-related emission factors. Although total UK emissions from railways contribute in a minor way to the total, it is likely that they could be more important locally, for example, close to a rail terminus with a large proportion of non-electric trains.

4.2.2.16 Aircraft

176. Aircraft do not make a large contribution to total UK emissions of PM_{10}. The turbines used by aircraft tend to produce high concentrations of NO_X and low concentrations of PM_{10}. Even for large airports such as Heathrow, the contribution made by aircraft is not significant. Of more significance are the emissions from activities related to aircraft, such as those from airside vehicles. Dispersion modelling described in Chapter 8 also shows that PM_{10} emissions from aircraft do not make a large contribution to local concentrations of primary PM_{10}. There is however a lack of measurements related to aircraft emissions compared with other source sectors.

177. Within the EU ARTEMIS project, a new aircraft emission factor database and model is being developed. While this will largely draw upon existing emissions data, the project includes new emissions measurements from taxi and thrust operations, recorded at London Heathrow, Frankfurt, Vienna, Wroclaw Strachowice and Warsaw Okecie airports during the period 2001–2002.

4.2.2.17 Shipping

178. The shipping sector makes a small contribution to total PM_{10} emissions in the UK. Like rail, it is likely that shipping could be of significance at a local level close to busy ports. Emissions can arise from both the large diesel engines used to power ships and the auxiliary engines. Currently, most ships in UK waters use fuel that has a high sulphur content, which tends to increase emissions of particles. However, as discussed in Section 4.3, the most important impact that ships make towards particle emissions is likely to be their contribution to secondary aerosol through emissions of SO_2 and NO_X. Emissions from ships can be very transient in nature and their patterns of activity complex.

179. The result is that ships are now one of the biggest sources of SO_2 emissions in the EU. Recent research for the EC shows that by 2010, ship emissions of SO_2 are likely to be equivalent to >75% of all land-based emissions, including emissions from all transport modes, combustion plants and heating engines which burn liquid fuels (Entec, 2002).

180. Throughout the EU, ship emissions contribute 20–30% to the concentrations of secondary inorganic particles in most coastal areas (COM, 2002). For the reasons outlined above, the EC is seeking to reduce SO_2 and particle emissions from ships and this is now an important environmental priority.

4.2.2.18 Non-road mobile machinery

181. Emissions from off-road machinery cover a range of portable or mobile equipment powered by diesel or petrol engines. They include agricultural and forestry equipment, construction equipment, industrial machinery and portable generators, aircraft support vehicles and domestic house and garden equipment.

Further details are given in the AQEG report on NO_2 (AQEG, 2004). The emission factors used in the inventory, in combination with machinery population and usage estimates, are given in g PM kWh^{-1} for different types of equipment and engine sizes. Table 4.4 provides averaged emission factors for the UK population of machinery expressed in units of fuel used. These implicit fuel-based emission factors are taken from the 2001 NAEI.

4.2.2.19 Chemical composition of PM emissions from transport sources

182. Particulate emissions from diesel exhausts contain mainly elemental carbon and heavy hydrocarbons derived from the fuel and lubricating oil that may be partially volatile. The particulates contain a large proportion of the toxic PAHs found in diesel exhausts. A small proportion of the emitted particle mass is sulphate derived from the sulphur present in the fuel, although this component can be expected to decline as the sulphur content of fuel is reduced. The EPEFE Auto-Oil Programme (EPEFE, 1995) characterised the particulates emitted from diesel vehicles running on a number of different fuel formulations, but all had a sulphur content of around 400 ppm, about ten-times higher than current levels. The studies found 70–80% of the particulate mass was elemental carbon; 13–23% was heavy hydrocarbons from fuel and lube oil; and 7% was sulphate. The primary contribution to sulphate is, however, negligible compared with secondary sulphate formation.

183. Particulate emissions from tyre wear include elemental carbon; hydrocarbons, including PAHs; several metals, including zinc and iron; and other inorganic compounds containing sulphur, silicon and calcium. Zinc is particularly abundant in tyre material. Chemical markers used to attribute tyre wear debris include benzothiazole, 2-(4-morpholinyl) benzothiazole, n-cyclohexyl-2-benzothiazolamine, styrene butadiene rubber, zinc and extractable organic zinc.

184. A variety of metals and PAHs are present in the material of brake linings. Various organic compounds (including PAHs), elemental carbon and inorganic compounds containing silicon, barium, iron, magnesium, phosphorus and chlorine have been found in brake dust.

4.2.3 Stationary sources

185. This section considers emissions of PM from stationary sources in general and discusses methods for deriving emission estimates of PM_{10}. The emission estimation procedure and emission factors described in the following section relate to the 2001 version of the NAEI produced in 2003, the most up-to-date version used by modellers. Since then the NAEI has paid considerable attention to the estimation of particulate emissions from stationary sources, with particular regard to the emission factors used in the inventory. This review is on-going, but some of the main sources – where changes to the emission factors or estimation methods are recommended – are summarised later in this section.

4.2.3.1 Overview of stationary sources

186. PM is emitted by a wide range of sources, including most major industries as well as homes and commercial premises. Natural sources of particles are also important. The most significant sources are the combustion of fuels in power stations, industrial/commercial-scale plants and particularly small residential combustion units. Residential-scale combustion units tend to emit more PM per unit of fuel consumed compared to larger combustion plants used by industrial and commercial operations. This is due to the lower efficiency of combustion in domestic fires, which produces greater quantities of incompletely oxidised products in the waste gases, and the use of abatement systems on larger industrial plant to remove PM emissions.

187. Industrial processes are a second major source of PM emissions. These emissions generally occur as a result of one of three mechanisms:

(i) the grinding, crushing or abrading of larger materials, for example, cement clinker grinding;

(ii) the evaporation of volatile matter from heated materials, for example, process emissions from glass and non-ferrous metal furnaces; and

(iii) the entrainment of fine PM in gas streams, for example, spray drying of chemicals.

188. An important issue for PM emissions from many industrial processes is that a proportion of these emissions are from ‘fugitive sources’. By this it is meant that emissions occur directly from a process into the workplace or outside atmosphere, without first being collected in by ducting or another container, and then being vented to atmosphere. The significance of fugitive emissions is that because they are not contained, they are difficult both to quantify and to abate.

189. Other, more minor, sources of PM include agricultural processes and waste incineration. Agricultural sources include the suspension of PM from agricultural land and entrainment of fine PM in vented gases from animal housing. Incineration of municipal solid waste, clinical waste, sewage sludge and chemical wastes are all very minor sources of PM, particularly so in recent years due to increasingly lower emission limits for persistent organic pollutants from incineration plants.

4.2.3.2 Derivation of emission estimates

190. Emission estimates for PM_{10} can be derived in one of two ways: with the use of national activity data such as fuel consumption data and emission factors or by the aggregation of emission estimates provided by process operators for individual sites, for example, via the Environment Agency’s Pollution Inventory. The estimates provided by operators may be derived using emission factors and plant-specific activity data or may be based on stack monitoring or other measurements. The methodologies used by process operators in a given sector may not have been consistent and had not been detailed in the sources of these data. Nonetheless they are used in the NAEI, when available, because it is considered that they represent an improvement on estimates made using national activity data and emission factors. Furthermore, consistency should improve in emission estimates under the Pollution Inventory as Guidance Notes are published by the Environment Agency. A more detailed description of the methodology used for the 2001 NAEI for the most important sources of PM_{10} emissions is given in the following sections.

Table 4.5 Emission factors and emission estimates for PM10 emissions from industrial and commercial sources as used in the 2001 version of the NAEI produced in 2003.

Source	Emission factor	Emission in 2001 (kt)
Coal burnt at power stations	319 g t^{-1} coal	15.7
Fuel oil burnt at power stations	426 g t^{-1} fuel oil	0.4
Natural gas burnt at power stations	0.153 g $therm^{-1}$ gas	1.4
Autogenerators:		
burning coal	2500 g t^{-1} coal	4.0
burning gas	0.152 g $therm^{-1}$ gas	0.2
Oil and gas industry/refineries:		
burning fuel oil	516 g t^{-1} fuel oil	1.0
burning gas oil	1070 g t^{-1} gas oil	0.1
burning OPG	0.147 g $therm^{-1}$ OPG	0.2
burning gas	0.326 g $therm^{-1}$	0.3[b]
Iron, steel, chemical and other industry[c]:		
burning coal (other industry)[d]	2500 g t^{-1} coal	5.1
burning coke (other industry)[d]	288 g t^{-1} coke	0.1
burning fuel oil	1032 g t^{-1} fuel oil	0.8
burning gas oil	250 g t^{-1} gas oil	0.6
burning gas	0.326 g $therm^{-1}$	1.7[b]
burning LPG	0.295 g $therm^{-1}$	0.1
burning wood	440 g t^{-1} wood	0.3
burning waste oils	250 g t^{-1} oil	0.1
Commercial/public sector:		
burning coal	2500 g t^{-1} coal	0.7
burning fuel oil	1032 g t^{-1} fuel oil	0.3
burning gas oil	250 g t^{-1} gas oil	0.3
burning gas	0.326 g $therm^{-1}$	1.4[b]
Coke oven processes	43.8 g t^{-1} coke	0.2
Cement clinker production	165 g t^{-1} clinker	1.7
Coal burnt at lime works	4380 g t^{-1} coal	0.5
Natural gas burnt at lime works	0.342 g $therm^{-1}$ gas	0.1[b]
Fugitive emissions from lime works	336 g t^{-1} lime	1.0[b]

Source	Emission factor	Emission in 2001 (kt)
Sinter plant	250 g t^{-1} pig iron	2.5
Blast furnaces	115 g t^{-1} pig iron	1.1
Basic oxygen furnaces	74 g t^{-1} BOF steel	0.8
Electric arc furnaces	138 g t^{-1} EAF steel	0.4
Foundries	625 g t^{-1} castings	0.9
Other iron and steel sources	8.70 g t^{-1} pig iron	0.1
Ore stockpiles	104 g t^{-1} pig iron	1.0
Aluminium smelting	604 g t^{-1} metal	0.2
Anode baking	1740 g t^{-1} anode	0.4
Alumina production	1110 g t^{-1} alumina	0.1
Ammonia-based fertilizer processes	—	0.5
Titanium dioxide processes	—	0.1
Other chemical processes	—	0.7
Fletton brick manufacture	204 g t^{-1} bricks	0.2
Non-Fletton brick manufacture	360 g t^{-1} bricks	2.2
Primary lead/zinc production	474 g t^{-1} metal	0.1
Secondary aluminium processes	369 g t^{-1} metal	0.3
Other non-ferrous metal processes	[a]	0.1
Copper semis and alloys	192 g t^{-1} metal	0.1
Flat glass	428 g t^{-1} glass	0.3
Glass wool	1520 g t^{-1} glass	0.2
Wood products manufacture	433 g t^{-1} product	1.4
Asphalt manufacture	30 g t^{-1} asphalt	0.7
Cement batching	12.8 g t^{-1} cement	0.3
Other part B processes	[a]	4.6

Key: OPG, other petroleum gas; LPG, liquid petroleum gas. [a]Because of the highly varied nature of the processes included, no meaningful emission factor can be derived. [b]These figures are currently being revised. See Section 4.2.4. [c]It should be noted that these are implied, aggregate factors in some cases averaged across several industry sectors. [d]Coal and coke are not burned in the iron and steel industry – these figures refer to other industries.

4.2.3.3 Power stations

191. Estimates of PM emissions from most UK power stations are available via regulators: the Environment Agency publishes emission estimates in their Pollution Inventory for England and Wales; the Scottish Environment Protection Agency (SEPA) publishes emissions data intended for submission to the European Pollutant Emissions Register

(EPER); and the Department of the Environment (Northern Ireland) provides emissions data for power stations, directly for use in the NAEI. There are some gaps in the dataset – for example, for small, regional, generators – but emissions from these processes are estimated either by extrapolation of emissions for other sites or by the use of literature-sourced emission factors, depending upon the fuel being burnt. Emission factors and emission estimates for the most important fuels, included in the 2001 version of the NAEI, are shown in Table 4.5.

192. The emission estimates for power stations are assumed to be among the most reliable in the PM_{10} inventory. This is based on the fact that emission estimates are provided for individual sites by the process operators, who will have access to data on fuel characteristics, fuel consumption, abatement plant efficiency and emissions monitoring data.

4.2.3.4 Industrial and commercial combustion processes

193. The NAEI divides industrial and commercial combustion sources into a number of subsectors, the most important of which (in terms of fossil fuel use) are:

- autogenerators;
- refineries;
- iron and steel;
- oil and gas industry;
- other industry; and
- public services.

194. Emissions of PM for the above and other combustion sources are estimated largely using literature-based emission factors and national fuel consumption statistics. The exception is refineries, where emission estimates are based on estimates provided by process operators for inclusion in the Pollution Inventory and other sources maintained by regulators. Emission factors and emission estimates for the sub-sectors listed above and the most important fuels, included in the 2001 version of the NAEI, are shown in Table 4.5. It should be noted that the factors shown here are averaged across a range of industrial and combustion processes of various scales of operation. Detailed information on emission factors and calculations for specific sectors can be found in the NAEI annual reports. The NAEI provides an opportunity for feedback from industry sectors and has a programme of continuous improvement. The inventory uses data provided by the major iron and steel producers, but there are a number of smaller iron foundries operating smaller Part B processes that are included in the iron and steel emission sector. For these, emissions per tonne of fuel tend to be higher than from the larger plant. Little coal and coke is burned in the iron and steel sector.

195. The emission estimates for industrial and commercial combustion plants are considered more uncertain than those for power stations and have, since the 2001 version of the NAEI, been identified as requiring review and updating. The use of a single or a few emission factors for plants burning a particular fuel is overly simplistic and will be replaced with a more detailed treatment starting with the 2003 version of the NAEI. It is not yet possible to state what impact this will have on UK emission estimates for PM_{10}; however, it is likely that estimates for the generally large plant in sectors such as autogenerators and the iron and steel sector will decrease, whereas estimates for the generally small plants in sectors such as public services will increase.

4.2.3.5 Domestic combustion

196. Emissions from domestic combustion are estimated using literature-based emission factors and national fuel consumption data. Emission factors and emission estimates for the most significant sources, included in the 2001 version of the NAEI, are shown in Table 4.6.

Table 4.6 Emission factors and emission estimates for PM_{10} emissions from domestic combustion.

Fuel burnt	Emission factor	Emission in 2001 (kt)
Coal	10400 g t^{-1} coal	14.9
Anthracite	3594 g t^{-1} anthracite	4.0
Solid smokeless fuels	5600 g t^{-1} SSF	2.5
Wood	7900 g t^{-1} wood	7.1
LPG	0.393 g $therm^{-1}$ LPG	0.1
Natural gas	0.326 g $therm^{-1}$ gas	4.2

Key: SSF, smokeless solid fuel.

197. PM_{10} emission factors for domestic combustion and for gaseous fuels in particular are very uncertain and are currently the subject of review. Emission factors will depend upon the type of combustion device – for example, open fire or enclosed boiler – yet the current approach does not take account of these differences. Changes will, therefore, be made to the methodology and the emission factors used for the 2003 version of the NAEI. Some of the main findings of the recent review are summarised in Section 4.2.4.

4.2.3.6 Coke and SSF manufacture

198. Emissions from the manufacture of coke are reported separately for those emissions derived from fuels burnt and emissions from processes such as oven loading and discharging, product quenching and crushing. Emissions from the manufacture of SSF using low temperature carbonisation processes are reported under a single category covering both fuel-related and process emissions. The estimates for fuel combustion during coke manufacture are based on literature-based emission factors and national energy consumption data and are relatively small compared with emissions from processes. The process emission estimates for coke works and the emission estimates for SSF manufacture are derived from emissions data provided by process operators for inclusion in the Pollution inventory. The emission factors and emission estimates for the only significant source – coke oven processes – included in the 2001 version of the NAEI are summarised in Table 4.5.

199. The emission estimates for combustion of gaseous fuels are very uncertain, but the total estimate for coke ovens (that is, fuel combustion plus process emissions) is based on the emission estimates given in the Pollution Inventory so that the overall uncertainty in emission estimates for this source is equal to the uncertainty in the Pollution Inventory data. These data are provided by process operators and might therefore be expected to be robust, but PM_{10} emissions from coke ovens (and SSF manufacture) will include a fugitive component, which is difficult to quantify. On balance, therefore, it is assumed that emission estimates for both types of processes are more uncertain than the estimates for combustion processes.

4.2.3.7 Cement and lime processes

200. Emissions from the manufacture both of cement clinker and lime will include a contribution from the combustion of fossil fuels and also from 'process' sources such as volatilisation of some components of raw materials and dust generated during crushing and grinding operations. As with coke ovens, fugitive emissions occur and are difficult to quantify. Emission factors and emission estimates for significant sources in cement and lime works, included in the 2001 version of the NAEI, are shown in Table 4.5. Note that the figures for lime production in Table 4.5 are those used in the 2001 NAEI, but these have recently been revised downwards and are likely to be closer to 0.3 kt in 2001. These changes will appear in the 2003 version of the inventory.

201. Emission estimates for cement and lime works are generally expected to be of a quality similar to those for industrial combustion plants and are considered acceptable given the uncertainty of estimates elsewhere in the PM_{10} inventory (with the exception of the estimate for fugitive emissions from lime processes, which is being reviewed). The possibility of under-reporting of emissions from fugitive sources cannot be ruled out.

4.2.3.8 Iron and steel industry

202. Emissions of PM_{10} from iron and steel processes are estimated and reported under the following categories:

- sinter plants;
- iron and steel blast furnaces;
- basic oxygen furnaces;
- electric arc furnaces;
- iron and steel foundries;
- other iron and steel (including iron ore handling, slag processing, continuous casting and miscellaneous secondary processing); and
- stockpiles (emissions from stockpiles of feed materials).

203. These categories relate to emissions from these processes due to any combustion of fuels and due to 'process' emissions, such as from the charging and tapping of furnaces. Emissions from the combustion and process sources are, however, reported separately in the NAEI in the case of blast furnaces. Combustion-related emissions are estimated using literature-based factors, whereas process emission estimates are based mainly on data reported in the Pollution Inventory or provided directly by process operators for use in the NAEI. Emission factors and emission estimates for the most important sources from the iron and steel industry are shown in Table 4.5. As stated earlier, these factors are implied factors that include processes in smaller iron foundries where factors are higher per unit of output than larger plant. The figures in Table 4.5 refer to implied factors in 2000 carried forward to 2001. These are under review and are expected to be superseded in later versions of the inventory.

204. Many of the emission estimates for the iron and steel industry shown in Table 4.5 are considered very uncertain because of the scope for fugitive emissions from these processes, although they may be known with greater accuracy for larger iron and steel plant. The figures for blast furnaces, basic oxygen furnaces and electric arc furnaces are, perhaps, most reliable, whereas those for foundries and stockpiles are probably the least reliable. The uncertainty factors for these processes are under review.

4.2.3.9 Aluminium production

205. In the UK, most aluminium is produced by the pre-baked anode cell process, although one plant operated the older Soderberg Cell process until 2000. Emissions occur both from the smelting process and from the preparation of anodes by baking. Alumina was produced from bauxite ore at one plant in Scotland until 2002 and emissions occurred from the grinding and drying stages of the process. Emission factors and emission estimates are shown in Table 4.5 and emission estimates are assumed to be fairly reliable.

4.2.3.10 Chemical processes

206. Emissions from chemical processes are estimated based on data given in the Pollution Inventory (Environment Agency, 2002a) and are reported in the following categories:

- chemical industry (alkyl lead): emissions from the manufacture of alkyl lead petrol additives;
- chemical industry (ammonia-based fertilizer): emissions from manufacture of ammonium nitrate;
- chemical industry (carbon black): emissions from the non-fuel use of petroleum fractions in the manufacture of carbon black;
- chemical industry (chromium chemicals): emissions from manufacture of chromium-based chemicals;
- chemical industry (magnesia): emissions from the production of magnesia;
- chemical industry (phosphate based fertilizer): emissions from manufacture of superphosphate from phosphate rock;
- chemical industry (titanium dioxide): emissions from manufacture of titanium dioxide; and
- chemical industry: emissions from chemical processes not included elsewhere.

207. In general, activity data are not available for these processes; thus, although the emissions data can be used in the NAEI, it is not possible to derive meaningful emission factors. Emissions from the most important sources from chemical processes are shown in Table 4.5.

208. Emission estimates for chemical processes are likely to be fairly robust, although the possibility of under-reporting of emissions from fugitive sources cannot be ruled out.

4.2.3.11 Bricks, tiles, refractories and ceramics

209. Fletton bricks are manufactured from Lower Oxford Clay, which contains significant levels of sulphur-containing organic material. This organic material is burnt when the bricks are fired and contributes heat to the process, thus less fossil fuel is required. The combustion of this material does lead to emissions of CO_2, CO, SO_2, VOCs and PM, and these emissions have been estimated based on data available in the Pollution Inventory (Environment Agency, 2003a). Since these data will also include emissions from the burning of fossil fuels, an estimate is first made of fossil fuel use by Fletton brick producers and the emissions from these fuels are calculated. These figures are subtracted from the Pollution Inventory data to leave estimates of the emissions from the burning of the organic component of the clay alone.

210. Non-Fletton bricks are manufactured from clays other than Lower Oxford Clay. These clays do not contain significant quantities of organic material and the only emission of significance is likely to be PM, which is generated mainly during the firing of the bricks. Emissions of this pollutant are also likely during the manufacture of tiles, refractories and ceramics. Emissions of PM_{10} have been estimated using emission factors based on USEPA data, as recommended in Passant *et al*. (2002). Emission factors and estimates for significant sources from Fletton and non-Fletton brick manufacture are shown in Table 4.5.

211. Estimates for Fletton bricks are probably more robust than those for non-Fletton bricks, although the quality of both estimates is likely to be acceptable given uncertainties elsewhere.

4.2.3.12 Other processes

212. Although it is likely that almost all manufacturing processes give rise to some emissions of PM, the Pollution Inventory only contains estimates for those where emissions are expected to be significant. Currently, estimates are made for the following fifteen processes:

- primary lead/zinc production (ceased operation in 2003);
- secondary copper production (ceased operation in 2000);
- secondary lead production;
- secondary aluminium production;
- nickel refining;
- copper semis and alloys;

- zinc alloys;
- zinc oxide production;
- lead batteries manufacture;
- other non-ferrous metals;
- glass production (split into flat, container, domestic, lead crystal, continuous filament glass fibre, wool, frits, and special glass);
- asphalt manufacture (roadstone coating);
- cement batching;
- wood product manufacture; and
- other Part B (LAPC) processes.

213. Emission estimates for all processes except the last five are based on Pollution Inventory data (Environment Agency, 2002a), with the full time-series being completed by extrapolation of the Pollution Inventory data based on metal production or consumption data in the first seven cases and based on Department of Trade and Industry (DTI) indices of manufacturing output in the other three cases. Emission factors can be generated for those sectors where the Pollution Inventory data can be compared with published activity data for the sector. Emission factors for glass production are based on data reported in the Pollution Inventory in the case of frits, continuous filament glass fibre and glass wool and from data provided by the process operator for flat glass (Pilkington Glass Ltd, 2003). Information contained in the EMEP/CORINAIR Emission Inventory Guidebook (EMEP/CORINAIR, 1999) and the European IPPC Bureau's BREF note for glass manufacture (EIPPCB, 2000) was used for the other types of glass production. The emission factors for asphalt manufacture, cement batching and wood product manufacture are taken from the USEPA compilation of emission factors (USEPA, 1997). Other Part B processes cover a range of industrial processes including coal handling, metal processing, galvanising and coating. Emission estimates for these Part B processes are based on emission estimates for 'typical' processes – which were originally available from the UK Emission Factor Database at http://rsk.co.uk/ukefd/ – multiplied by estimates of the number of processes in the UK. Emission factors and emission estimates for the most important of these sources are given in Table 4.5. Although these sources may not be important nationally, they could be important local sources.

214. The estimates for other Part B processes in particular are subject to a very high level of uncertainty, although estimates for many of the sources are not as robust as those for combustion processes. The contribution of fugitive emissions could be underestimated.

4.2.3.13 Mining and quarrying

215. Emissions of PM_{10} from open-cast mining and quarrying were estimated using an USEPA (1997) factor of 0.1 g kg^{-1} of material throughput. This gives a very crude estimate of 20.6 kt in 2001, which does not take into account changes in technology and abatement over time. The NAEI methodology for this sector is currently being reviewed (see Section 4.2.4.3).

4.2.3.14 Construction

216. The emission estimates for construction are based on an emission estimate for 1990 presented in the third QUARG report (QUARG, 1996). Estimates for other years are extrapolated from this figure on the basis of the value of construction work done, giving a figure of 4.3 kt for 2001. This very crude estimate is currently being reviewed (see Section 4.2.4.3).

4.2.3.15 Waste incineration

217. The NAEI estimates emissions from the categories municipal solid waste (MSW) incineration, clinical waste incineration, sewage sludge incineration, animal carcass incineration and chemical waste incineration. The emission factors and emission estimates for the most important sources are shown in Table 4.7. The factor for MSW incineration is supported by an estimate of 38g/tonne waste based on site-specific data from waste management activities in a study carried out for Defra by Enviros Consulting and Birmingham University (www.defra.gov.uk/environment/waste). No emission factor is available for animal carcase incineration, since the source of data used for this sector only gives an estimate of total emissions from the sector (AEA Technology, 2002). Emission estimates for offshore flaring are provided by the United Kingdom Offshore Operators Association (UKOOA, 2002).

Table 4.7 Emission factors and emission estimates for PM_{10} from incineration and flaring.

Source	Emission factor	Emission in 2001 (kt)
MSW incineration	24.7 g t^{-1} waste	0.1
Clinical waste incineration	270 g t^{-1} waste	0.1
Chemical waste incineration	320 g t^{-1} waste	0.1
Animal carcase incineration	—	0.3
Offshore flaring	0.604 g m^{-3} gas flared	1.1

218. Emission estimates for incineration processes are likely to be fairly robust, whereas the reliability of the UKOOA figure for offshore flaring is likely to be less so. Dust emissions from flaring in other sectors of industry (chemicals, refineries) are not currently included in the NAEI but are unlikely to be as important as emissions from offshore flaring.

4.2.3.16 Agriculture

219. Emission sources include dust from agricultural activities such as ploughing, dust from animal housing and the use of fuels to provide heating. Straw is burned as a fuel but the *in situ* burning of stubble and crop residues has been prohibited since the mid-1990s, with limited exemptions. Emission factors and emission estimates for the most significant sources are shown in Table 4.8.

220. Emission estimates for agricultural operations must be considered among the most uncertain in the NAEI due to the fugitive nature of most of the emissions.

Table 4.8 Emission factors and emission estimate for PM_{10} from agriculture.

Source	Emission factor	Emission in 2001 (kt)
Agricultural activities	0.105 g t^{-1} cereals	2.0
Straw combustion	11000 g t^{-1}straw	2.2
Housed livestock:		
laying hens	19.5 g per head	0.5
broilers	58.8 g per head	6.6
other poultry	74.1 g per head	2.5
pigs	260 g per head	1.4
dairy cattle	130 g per head	0.4
other cattle	41.9 g per head	0.3

4.2.3.17 Foot and mouth pyres

221. During 2001 the UK suffered from an outbreak of foot and mouth disease. Due to the highly infectious and debilitating nature of the disease, action taken to contain the outbreak included the slaughter of over 6,000,000 farm animals with the subsequent need for disposal of carcasses. The slaughter included 4,200,000 animals for disease-control purposes and 2,300,000 animals for welfare reasons. Approximately one-third of the carcasses were disposed of by burning (including *in situ* burning, mass pyres and incineration). The burning of carcasses on mass pyres occurred between March 2001 and May 2001 and involved the burning of animals together with specified quantities of fuels. The fuels included wooden railway sleepers, kindling wood, straw, coal and diesel oil. Emissions of air pollutants occurred both due to the combustion of these fuels and the burning of the carcasses. Emissions of PM_{10} were estimated for inclusion in the Pollution Inventory. Table 4.9 shows the estimates and emission factors expressed as emissions per animal carcass.

Table 4.9 Emission factors and emission estimates for PM_{10} from foot and mouth pyres.

Source	Emission factor	Emission in 2001 (kt)
Cattle pyres	12.5 kg per carcass	3.0
Sheep pyres	0.682 kg per carcass	0.6
Pig pyres	3.01 kg per carcass	0.2

4.2.3.18 *Chemical composition of PM emissions from stationary sources*

222. Information on particle composition is not currently collected for the NAEI, so no data on the chemical composition of particles emitted can be provided. In general, however, it would be expected that emissions from small and inefficient combustion processes would largely consist of carbon, whereas emissions from larger combustion processes would contain some carbon, but would predominantly consist of inorganic matter deriving from the mineral content of the fuel. This may include metals in the fly ash. PM emissions from most industrial processes (for example, cement manufacture), quarrying and construction would be inorganic matter, often chemically similar to the raw materials or products of the process. These will usually contain a high proportion of alkaline earth metal compounds (for example, calcium).

4.2.4 A recent review of PM emission factors for industrial processes and combustion

223. The NAEI is currently carrying out a review of PM emission factors for a range of industrial processes, combustion, mining and quarrying and construction.

224. Analysis of the most significant industrial emitters of PM_{10} and smaller particle sizes indicates several important sectors:

- coal combustion and natural gas combustion for power generation public utility power stations or other industry subsectors;
- iron and steel sintering plants; and
- manufacture of non-Fletton bricks.

Each of these sectors and the manufacture of cement clinker have been reviewed.

225. For many sources, the emission factor reference documents generally refer to the USEPA publication AP42 for sources of raw data (often via several other references). Although AP42 is a valued reference, it is important to note that – within a number of sections – emission data have not been updated for many years. This is particularly relevant to PM size speciation profiles, some of which – despite advances in abatement technology – have not been updated for over a decade.

226. A brief summary of the main recommendations of the NAEI review and proposed changes to emission factors is given in the following sections.

4.2.4.1 *Review of industry processes*

227. The review of industrial emissions of PM_{10} has highlighted several issues including the fundamental question of what is PM_{10} when emitted from a process. In general, the NAEI includes emission estimates based on a 'filterable' PM_{10} release. However, the emission factors within the NAEI for natural gas combustion assessed in this task also include a 'condensable' fraction.

228. The USEPA defines two fractions to a PM_{10} emission from an industrial or combustion source. The filterable PM_{10} fraction in a stack emission can be determined using USEPA Method 201A and is essentially material that is smaller than nominally 10 µm (separated from larger PM using a miniature cyclone) and that can be retained by a filter at stack temperature. Condensable PM is also considered to be PM_{10} and is material collected downstream of the sampling filter from probe rinses and material collected within a chilled impinger train (USEPA Method 202).

229. The test procedure for condensable PM (Method 202) is an official USEPA test procedure, and emission inventories in the US are required to consider both filterable PM_{10} and the condensable PM_{10} emissions. However, the test method has been the subject of considerable debate over whether it provides data that are representative of condensable emissions. It is intended to replicate secondary formation of particulate by condensation downstream of the stack. However, although the sampling procedure lowers the temperature of the sampled gases, there is no concurrent dilution action. Interfering effects are also possible. Limited evidence from dilution tunnel experiments suggest that Method 202 may overestimate condensable emissions.

230. The NAEI incorporates PM_{10} data reported by process operators to the regulatory authorities. The regulatory authorities require operators of selected processes to report PM_{10} emissions for inclusion in the Pollution Inventory (or EPER) above a threshold of 10 t year ^{1}. This information is then incorporated into the NAEI. The methodologies adopted by process operators for estimating PM_{10} emissions includes a mixture of direct measurements, emission factors or factors applied to a total particulate emission.

231. The protocol for inventory reporting adopted by the Electricity Supply Industry Joint Environment Programme (JEP) is typical in that it applies a PM_{10} factor to the total particulate emission. The JEP protocol states that the Electricity Supply Industry total particulate emission data for coal-fired stations are based on continuous monitoring systems. These systems do not monitor the condensable fraction. Similarly, other operators may base emissions data on short-term measurements of particulate emission, which almost certainly do not include assessment of the condensable fraction. Conversely, other operators may include condensable material in their reports through the use of USEPA default emission factors for their process.

232. The scale of these inconsistencies and their impact on the NAEI is unknown. The consistency of Pollution Inventory/EPER reporting by process operators will be addressed in time by various initiatives, including guidance from the regulatory authorities or operator initiatives such as the JEP protocol.

233. Hence, methodology issues result in inconsistencies in the treatment of PM_{10} within the NAEI. The bulk of existing PM_{10} emission data in the NAEI are for 'filterable' PM_{10} and, hence, to ensure consistency within the NAEI, only filterable PM_{10} data should be considered. Default PM_{10} and other PM_{10} emission factors used in the NAEI for industrial and combustion sources should be reviewed to determine if any include a condensable component.

234. The review has concluded that for the manufacture of cement clinker, combustion of coal and iron and steel sintering processes, no changes to the current PM_{10} emission factors should be made, but further work is required to develop more robust particle size speciation data for these sources. Currently used USEPA speciation data are considered out-of-date and not appropriate for UK plants. For the manufacture of non-Fletton bricks, both a revision to the PM_{10} emission factor and measurements of size speciation data are recommended.

4.2.4.2 Review of emissions from the combustion of natural gas

235. Natural gas is a key fuel for industry and power generation. Combustion of natural gas is among the five highest emitters of particulates for several PM fractions. This is perhaps surprising given the inherently low potential for particulate emission from the combustion of refined gas. The scale of gas use is increasing and the significance of the sector is also increasing as other sources decline. The emission of PM from combustion of natural gas is also significant for the 'other industry' sector.

236. At present, the UK Electricity Supply Industry JEP inventory reporting protocol reports PM_{10} emissions from gas combustion as 0 or operators report emissions below the reporting threshold (10 t $year^{-1}$). However, the NAEI estimates UK emissions of PM_{10} from combustion of natural gas using emission factors and activity statistics. For the electricity supply industry, the NAEI default emission factor is currently 154 kg $Mtherm^{-1}$; however, the source of this factor is unclear. The default factor is about half the USEPA emission factor for total PM (filterable and condensable PM), which is currently used for the other industry and other gas combustion sectors within the NAEI (338 kg $Mtherm^{-1}$).

237. The review indicates that the USEPA emission factor for filterable PM from combustion of natural gas (84 kg $Mtherm^{-1}$) should be used by the NAEI in all sectors. Measurement of filterable PM_{10} at a power station combined cycle gas turbine for Defra and a smaller gas turbine for the Environment Agency as well as data for gas combustion published by EMEP indicate PM_{10} emission factors of a similar order of magnitude to the default USEPA factor. However, it should be noted that these emission measurements are likely to have a high uncertainty.

238. The proposed emission factor is approximately half that of the current 'default' factor used for electricity generation. However, it is traceable and consistent with PM_{10} emissions from combustion in other areas of the NAEI (that is, it is consistent with the definition of PM_{10} from other sources reported in the Pollution Inventory). Application of the filterable emission factor to combustion of gas in the domestic sector represents a 75% reduction in the emission factor currently used. The consequences of this change to estimates of low-level emissions of PM_{10} in urban areas can be expected to be significant and requires evaluation within the inventory.

4.2.4.3 Review of emissions from mining, quarrying and construction

239. A review of PM emissions from the mining, quarrying and construction sectors is still in progress. However, early indications are that the PM emission factors currently used in the NAEI for many of these processes may be too high and need to be reduced.

4.2.5 Fireworks and associated fires

240. Fireworks and the fires associated with bonfire night are important sources of particle emissions in the UK. It is known from ambient concentrations (see Chapter 8) that emissions from these sources can result in concentrations of PM_{10} that are greater than 50 µg m^{-3} for several days each year. These sources of particles can, therefore, be locally important, although no emission rate estimates are available to compare with other primary sources of PM. This source can also represent an important source of metals in the atmosphere, which are either contained in the fireworks themselves or originate from the combustion of materials that contain metals.

4.3 Secondary particle precursors

4.3.1 Origin of secondary particles

241. Secondary particles are generated by gas-to-particle conversion processes, following the formation of products of particularly low volatility or high solubility from the gas phase oxidation of emitted precursor gases. In addition to this, transfer of material from the gaseous to the particulate phase may also occur as a result of the reactions of gaseous species with aerosol or aqueous droplets, when the products of such reactions remain either wholly or partially in the condensed phase. In this section, the sources of secondary particle precursors are identified and a description is given of the chemical processes that convert these precursors into the major components of secondary particles, namely sulphate, nitrate, ammonium and organic.

4.3.2 Sulphate

242. The major precursor to sulphate aerosol formation in the UK is SO_2. As is described further in Section 4.4.3, SO_2 is mainly derived from the combustion of solid fuel and petroleum products; in the UK its major sources are power generation and industrial combustion processes.

243. As shown in Figure 4.3, the oxidation of emitted SO_2 can occur by both gaseous phase and aqueous phase routes. The predominant gas phase reaction for SO_2 is the reaction with the hydroxyl radical (OH):

$$\mathrm{OH + SO_2\ (+M) \rightarrow HOSO_2\ (+M)} \qquad (1)$$

where M is a third body, most commonly nitrogen (N_2). At a typical background OH concentration of 0.04 ppt, SO_2 is oxidized at a rate of about 0.3% h^{-1}, although the conversion rate is correspondingly greater at elevated OH concentrations consistent with a photochemical episode (that is, up to approximately 0.1 ppt). Gaseous H_2SO_4 is subsequently formed by the following sequence of reactions:

$$\mathrm{HOSO_2 + O_2 \rightarrow SO_3 + HO_2} \qquad (2)$$
$$\mathrm{SO_3 + H_2O\ (+M) \rightarrow H_2SO_4\ (+M)} \qquad (3)$$

244. In comparison with reaction (1), these reactions occur essentially instantaneously (typically on the timescales of 1 μs and 10 ms, respectively, in the boundary layer) and are consistent with OH-initiated oxidation, leading to quantitative conversion of SO_2 to gaseous H_2SO_4. Consequently, the fractional conversion of SO_2 to gaseous H_2SO_4 is determined by how efficiently the gas-phase oxidation route, initiated by the reaction with OH radicals, competes with the alternative oxidation or removal routes for SO_2. H_2SO_4 is sufficiently involatile to generate secondary particles by the processes of nucleation and condensation, as described in Section 2.1. The gas phase oxidation of SO_2 may, therefore, result in the formation of new particles or the growth of existing ones.

245. Although the gas phase oxidation of SO_2 can dominate under specific conditions (for example, summertime photochemical episodes), oxidation in the aqueous phase is generally more important. This is largely governed by how frequently polluted air masses encounter clouds and occurs at a rate of about 1% h^{-1}, on average, under UK conditions (Metcalfe *et al.*, 1995). As shown in Figure 4.3, the solvated form of SO_2 ($SO_2.H_2O$) is present in equilibrium with the ionic species HSO_3^- and SO_3^{2-}. In all these species, sulphur is in the +4 oxidation state; they are usually collectively denoted S(IV). Oxidation to the +6 oxidation state (S(VI) or 'sulphate') is promoted by the presence of aqueous phase oxidants and catalysts. As is described in detail elsewhere (Finlayson-Pitts and Pitts, 1999), dissolved hydrogen peroxide (H_2O_2) and ozone (O_3) are believed to be particularly important, although oxidation by O_3 becomes progressively less efficient with increasing acidity of the cloud droplets. The chemical processing of SO_2 by the cloud therefore leads to the oxidation of S(IV) to S(VI) and, upon evaporation, the resultant particles will contain sulphate. Unlike the gas-phase oxidation route, however, aqueous phase oxidation cannot result in new particle formation.

4.3.3 Nitrate

246. Nitrate aerosol is formed from the oxidation of emitted NO_X. As is described in detail elsewhere (AQEG, 2004), and in Section 4.4.3, NO_X is mainly derived from fossil fuel combustion, with its major sources in the UK being road transport and power generation.

247. As shown in Figure 4.4, nitrate aerosol is formed from the oxidation of NO_2, which is itself mainly derived from the oxidation of emitted NO. The predominant daylight oxidation route for NO_2 is initiated by its reaction with OH:

$$OH + NO_2\ (+M) \rightleftarrows HNO3\ (+M) \qquad (4)$$

which leads to the quantitative formation of gaseous HNO_3. At a typical background OH concentration of 0.04 ppt, NO_2 is converted to HNO_3 at about 5% h^{-1} by this reaction, although the conversion rate is correspondingly more rapid at elevated OH concentrations consistent with a photochemical episode (that is, up to ~0.1 ppt). HNO_3 is highly soluble and also shows a strong affinity for surfaces. It is, therefore, scavenged by existing particles and droplets to form nitrate aerosol, in competition with removal by deposition to the ground.

248. Nitrate aerosol is also generated via the formation of the higher oxide, N_2O_5, which occurs as follows:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad (5)$$
$$NO_3 + NO_2\ (+M) \rightleftarrows N_2O_5\ (+M) \qquad (6)$$

249. This reaction sequence is very inefficient during the day and at higher levels of NO because the intermediate NO_3 radical photolyses and reacts with NO rapidly, leading to NO_X regeneration. Because N_2O_5 is in equilibrium with NO_3 (reaction 6), it also cannot persist at significant concentrations below these conditions. During the night, significant NO_2 to N_2O_5 conversion can occur, with the potential for nitrate aerosol generation from the reaction of N_2O_5 with H_2O on existing particle surfaces or aqueous droplets:

$$N_2O_5(g) + H_2O\ (+aerosol) \rightarrow 2HNO_3\ (adsorbed\ or\ dissolved\ (7)$$

Figure 4.3 Major chemical processes involved in sulphate aerosol production.

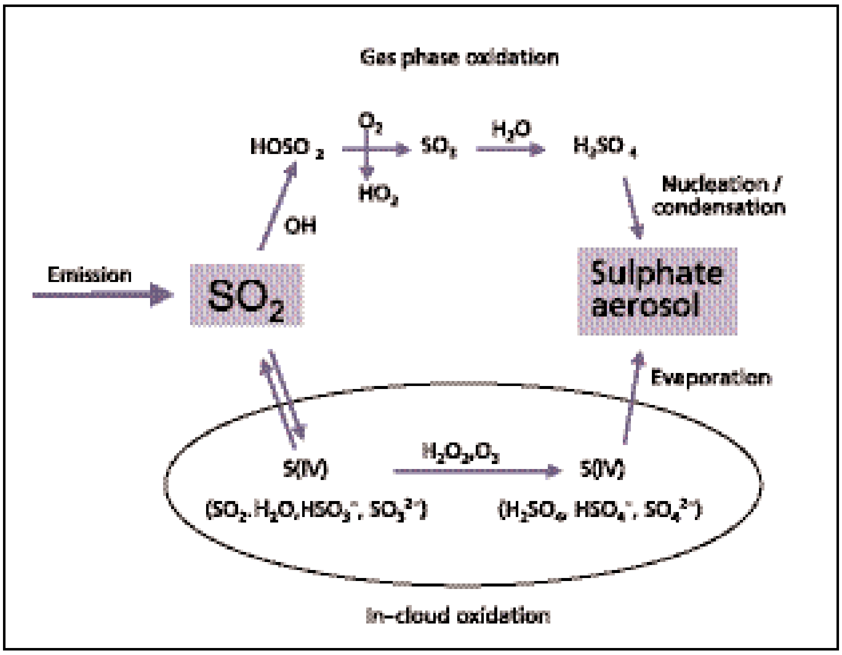
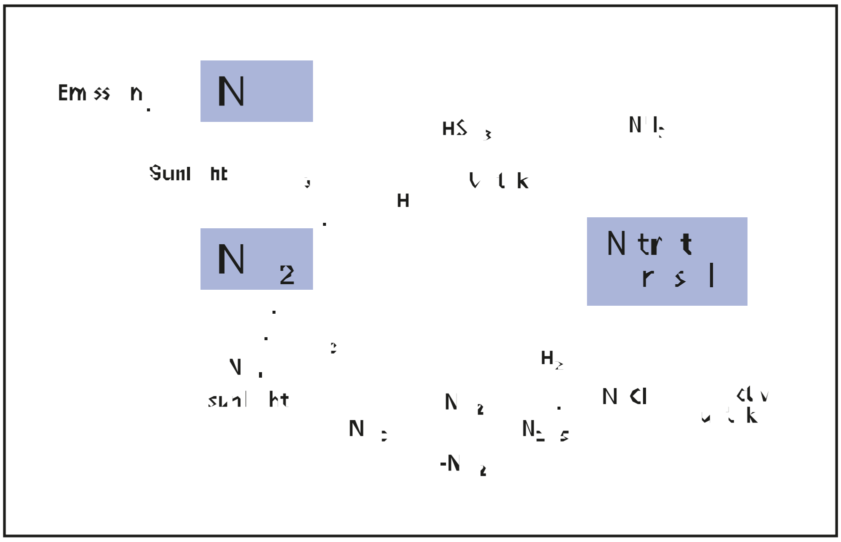


Figure 4.4 Major chemical processes involved in nitrate aerosol production.



250. The reaction of N_2O_5 with sea salt aerosol also leads to the formation of nitrate aerosol, with partial regeneration of NO_2 (and release of chlorine), as follows:

$$N_2O_5(g) + NaCl\ (aerosol) \rightarrow NaNO_3(aerosol) + ClNO_2(g) \quad (8)$$

$$ClNO_2 + sunlight \rightleftarrows Cl + NO_2 \quad (9)$$

251. The conversion of NO_2 to nitrate aerosol, via N_2O_5, potentially occurs at a comparable rate to that from OH-initiated oxidation during daytime. However, its efficiency can be reduced by the reaction of NO_3 with residual night time NO or with some organic compounds.

4.3.4 Ammonium

252. The precursor to ammonium aerosol formation is ammonia (NH_3). As is described further in Section 4.4.3, NH_3 emissions are dominated by agricultural sources, which are mainly due to the decomposition of urea and uric acid in livestock waste. A small, but increasing, contribution is also made by road transport, from cars fitted with three-way catalysts.

253. NH_3 is the most abundant alkaline gas in the atmosphere and, therefore, plays a significant role in neutralizing acids. It is efficiently taken up into acidic sulphate and nitrate aerosols, formed by the processes described above, leading to the formation of ammonium aerosol. As a consequence, NH_3 uptake also serves to regulate the acidity of aerosol and cloud droplets, which, in turn, influences the solubility and oxidation rate of species such as SO_2.

254. As is described in Sections 2.1 and 2.2, NH_3 also plays a role in the ternary nucleation of sulphate aerosols and reacts with gaseous HNO_3 to form particulate NH_4NO_3.

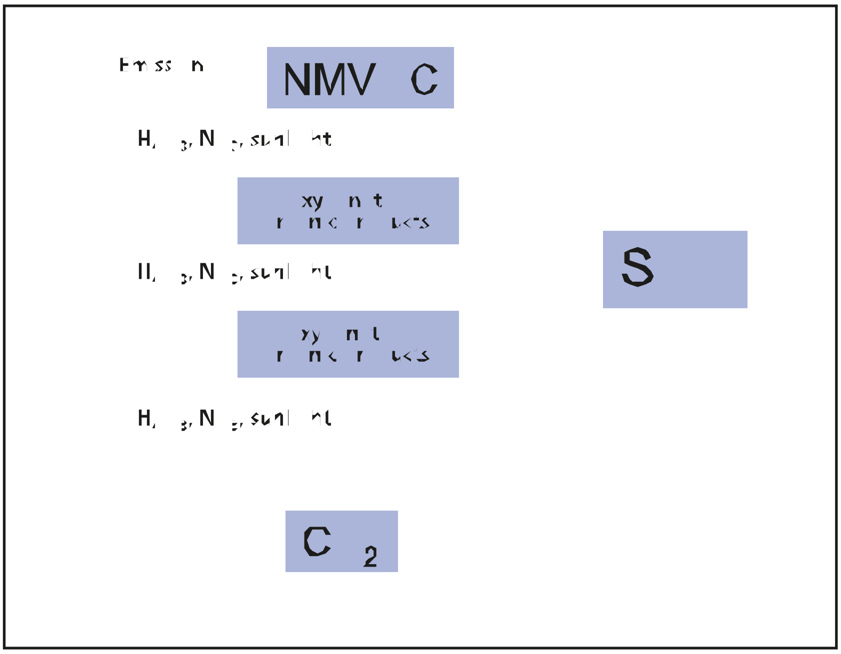
4.3.5 Organics

255. Secondary organic aerosol (SOA) is formed when the oxidation of emitted NMVOCs generates oxygenated products that undergo gas-to-particle transfer. As is described further in Sections 4.4.3 and 4.4.4, important precursor NMVOCs may be emitted from both anthropogenic and biogenic sources.

256. It is well known that hundreds of individual NMVOCs are emitted (Goodwin *et al.*, 2001), with contributions from many different classes (for example, alkanes, and aromatics). The gas phase oxidation of each NMVOC broadly follows the pattern illustrated in the simplified schematic in Figure 4.5. This shows that the oxidation may be initiated by one or more reactions, the relative importance of which depends on the structure of the given NMVOC and on the ambient conditions. The initial oxidation step leads to the production of a set of organic products containing polar oxygenated functional groups, such as aldehyde (–C(=O)H), ketone (–C(=O)–), alcohol (–OH), nitrate ($–ONO_2$) and carboxylic acid (–C(=O)OH) groups, which tend to make the products less volatile and more water soluble. Further oxidation may introduce additional functional groups, such that the ‘second generation’ set of products (and subsequent generations) may be of even lower volatility and higher solubility. However, the oxidation mechanisms also lead to fragmentation of the carbon chains to form lower molecular weight oxygenates (which are more volatile), and the gas phase oxidation ultimately converts all the carbon in the emitted NMVOCs into CO_2. This is in stark contrast, therefore, to the situation for SO_2 and NO_X, for which the ultimate oxidation products (sulphate and nitrate) are the forms that are most associated with the particle phase. SOA is, therefore, formed from the gas-to-particle transfer of partially oxidized organic material, which occurs in competition with further oxidation in the gas phase.

257. As a result of the complexity of the emitted NMVOC mixture and the oxidation chemistry, the atmosphere contains many thousands of structurally different organic oxygenates, which possess a wide range of properties (for example, reactivity, volatility and aqueous solubility) and, therefore, different propensities to undergo gas-to-particle transfer. The importance of emitted NMVOCs as SOA precursors is, therefore, very variable, with most emitted compounds being very inefficient at generating SOA (indeed, the vast majority of atmospheric organic material resides in the gaseous phase, even though the small fraction present in particles makes an important contribution to particle mass). However, certain classes of NMVOCs are more likely to lead to aerosol formation by virtue of their general high reactivity and types of oxidation product formed. Of particular significance are large, cyclic, unsaturated compounds: the oxidation products tend to be of comparatively high molecular weight and contain two or more polar functional groups, these products are, therefore, of lower volatility. Two such classes of emitted NMVOCs, which generate SOAs, are monoterpenes (for example, α and ß-pinene) and aromatic hydrocarbons (for example, toluene and xylenes), although other classes may also contribute. Monoterpenes are naturally emitted NMVOCs that are estimated to account for a substantial fraction (30–80%) of the UK biogenic total, with an important contribution made by coniferous trees (for example, Dore *et al*., 2003; EMEP-CORINAIR, 2004). Aromatic hydrocarbons are primarily of anthropogenic origin, accounting for about 15% of the UK anthropogenic NMVOC total (Goodwin *et al*., 2001). The majority of this derives from road transport emissions.

Figure 4.5 Simplified representation of NMVOC oxidation to CO_2 and formation of SOA as a by-product.



258. Monoterpenes undergo rapid photo-oxidation and reaction with O_3. Particularly significant oxidation products are bi- and multifunctional organic acids, such as pinic acid and pinonic acid. The results of chamber studies demonstrate that these acids play a major role in the SOA formation process (Yu *et al*., 1999a; Glasius *et al*., 2000; Kuckelmann *et al*., 2000); they have also been detected and quantified in ambient aerosols (Kavouras *et al*., 1999; Yu *et al*., 1999b; ODowd *et al*., 2002). Chamber studies of the photo-oxidation of aromatic hydrocarbons have also confirmed that SOA is formed (Odum *et al*., 1997; Kleindienst *et al*., 1999). Although the composition of SOA from aromatic oxidation is less well characterized, highly substituted cyclic products (for example, nitrocresols, catechols and quinones) and ring-opened bi- and multifunctional acids have been detected in the particle phase (Forstner *et al.*, 1997; Jang and Kamens, 2001).

259. The formation of SOA from monoterpene and aromatic hydrocarbon oxidation in chamber experiments has generally been interpreted successfully in terms of the partitioning of semi-volatile oxidation products between the gaseous and particulate phases (Odum *et al.*, 1996; Hoffmann *et al.*, 1997; Seinfeld and Pankow, 2003), as described for PAHs in Section 2.2. However, it is also becoming clear that the partitioning organic species can be further stabilized in the condensed phase by association and polymerization reactions (Tobias and Ziemann, 2000; Kalberer *et al.*, 2004), which may be enhanced in acidic aerosols (Jang *et al.*, 2002), and it is probable that such processes play an important role in atmospheric SOA formation.

4.4 Emission estimates by source type

4.4.1 Directly estimated primary emissions

260. Figure 4.6 shows the contribution made by different source sectors by particle size for 2001 based on an analysis of data from the NAEI. In general the particles derived through combustion processes dominate the smaller size fractions. For example, road transport is responsible for only 18% of total PM_{10} emissions but 35% of PM_1 emissions. The converse is true for other sectors, such as production processes, where their contribution diminishes as the particle size fraction reduces. It is clearly important, therefore, to consider particle size when considering the contributions made by different source sectors. Further consideration is given to PM emissions by size distribution in the next section.

Figure 4.6 Contribution made by different source sectors by particle size.

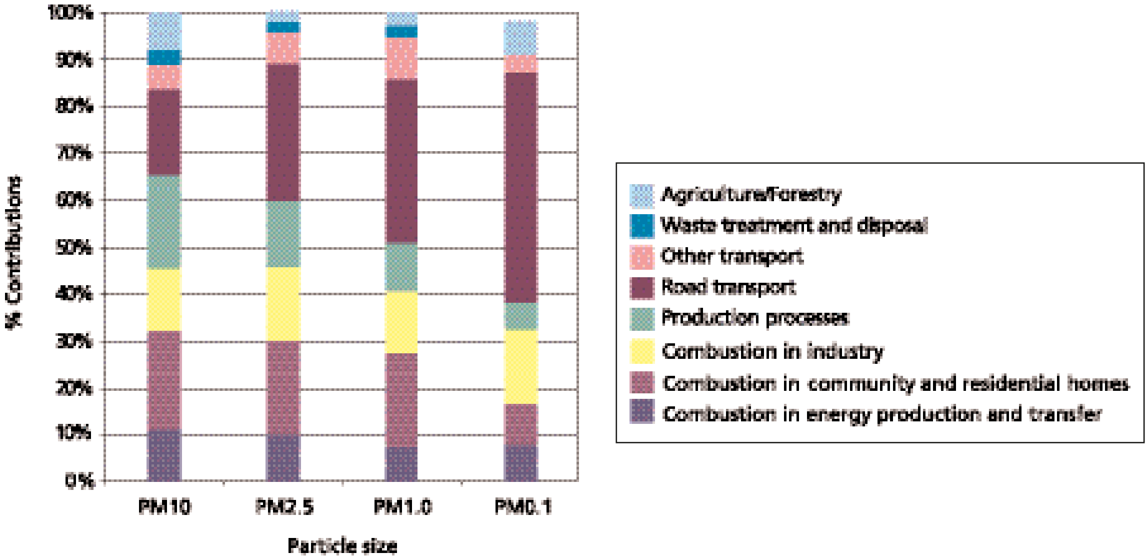


Table 4.10 Emission estimates for PM by size fraction in 2001.

Sector	Emissions in kt	Percentage by size fraction			
	PM_{10}	$PM_{(10\text{-}2.5)}$	$PM_{2.5}$	PM_1	$PM_{0.1}$
Electricity generation:					
Coal	15.7	56.1	43.9	19.1	8.3
Other fuels	2.0	5.0	95.0	90.0	40.0
Industrial/commercial combustion:					
Coal	5.4	35.2	64.8	29.6	7.4
Natural gas	3.7	0.0	100.0	100.0	51.4
Other fuels	6.8	45.6	54.4	32.4	14.7
Domestic combustion:					
Coal	12.6	61.9	38.1	31.7	2.4
Smokeless fuels	6.5	73.8	26.2	4.6	1.5
Wood	7.1	29.6	70.4	29.6	8.5
Natural gas	4.2	0.0	100.0	100.0	9.5
Other fuels	0.7	28.6	71.4	71.4	14.3
Industrial processes:					
Iron and steel	7.3	37.0	63.0	41.1	20.5
Cement and lime	3.3	46.4	53.6	25.0	10.7
Bricks, glass and ceramics	3.8	26.3	73.7	68.4	34.2
Quarrying	20.6	70.9	29.1	8.3	0.0
Construction	4.3	69.8	30.2	11.6	0.0
Other	9.0	40.0	60.0	51.1	18.9
Transport:					
Road transport – petrol	3.0	16.7	83.3	70.0	33.3
Road transport – diesel	27.4	10.2	89.8	85.0	50.0
Road transport – brake /tyre wear	8.9	44.9	55.1	10.1	7.9
Off-road vehicles	7.2	22.2	77.8	69.4	15.3
Other transport	1.9	10.5	89.5	84.2	15.8
Waste disposal:					
Foot and mouth pyres	3.8	60.5	39.5	31.6	2.6
Other	1.5	0.0	100.0	100.0	13.3
Agriculture	13.7	83.9	16.1	15.3	15.3
Total	180	43.3	56.7	41.2	17.2

Table 4.11 Contribution to UK PM emissions by source sector and size fraction in 2001.

Sector	Percentage by size fraction			
	PM_{10}	$PM_{2.5}$	PM_1	$PM_{0.1}$
Electricity generation:				
Coal	8.7	6.7	4.0	4.2
Other fuels	1.1	1.9	2.4	2.6
Industrial/commercial combustion:				
Coal	3.0	3.4	2.2	1.3
Natural gas	2.1	3.6	5.0	6.1
Other fuels	3.8	3.6	3.0	3.2
Domestic combustion:				
Coal	7.0	4.7	5.4	1.0
Smokeless fuels	3.6	1.7	0.4	0.3
Wood	3.9	4.9	2.8	1.9
Natural gas	2.3	4.1	5.7	1.3
Other fuels	0.4	0.5	0.7	0.3
Industrial processes:				
Iron and steel	4.0	4.5	4.0	4.8
Cement and lime	1.8	1.7	1.1	1.1
Glass, bricks and ceramics	2.1	2.7	3.5	4.2
Quarrying	11.4	5.9	2.3	0.0
Construction	2.4	1.3	0.7	0.0
Other	5.0	5.3	6.2	5.5
Transport				
Road transport – petrol	1.7	2.4	2.8	3.2
Road transport – diesel	15.2	24.1	31.3	44.3
Road transport – brake/tyre wear	4.9	4.8	1.2	2.3
Off-road vehicles	4.0	5.5	6.7	3.6
Other transport	1.1	1.7	2.2	1.0
Waste disposal:				
Foot and mouth pyres	2.1	1.5	1.6	0.3
Other	0.8	1.5	2.0	0.6
Agriculture	7.6	2.2	2.8	6.8

4.4.2 Emissions of $PM_{2.5}$ and fine PM

261. The NAEI includes estimates of emissions of $PM_{2.5}$, PM_1 and $PM_{0.1}$. These estimates are derived by applying a set of size distribution profiles to the NAEI estimates of PM_{10} emissions. Generally speaking, PM_{10} emissions from industrial combustion processes and road transport are considered to contain more fine material than, for example, mechanically generated PM_{10} from quarries and construction sites. Table 4.10 summarises the NAEI estimates for fine PM and Table 4.11 shows how the percentage contribution of major source sectors changes for each size fraction. This table shows how emissions from industrial combustion processes and road transport contribute an increasingly large proportion of UK emissions with decreasing size while emissions from quarries and construction become increasingly trivial with decreasing size. The contribution of diesel powered road vehicles increases from 15% for PM_{10} to 45% for $PM_{0.1}$.

262. The size distributions used in the NAEI are mostly taken from the USEPA Compilation of Emission Factors (known as AP-42). These size profiles are given for a specific level of control and it is not clear how appropriate these profiles are for the UK (although a critique of the NAEI size profiles is currently being prepared). In general, the proportion of fine material in PM_{10} emissions will increase as the level of abatement increases because abatement technologies preferentially collect the coarser particles ($<PM_1$). Thus, if the level of control in the UK is greater than that assumed in the USEPA profiles, then it is likely that emissions of PM_1 and $PM_{0.1}$ in particular may be underestimated. This will have significance for the time series of emission estimates for the finer particle sizes (Section 4.5.2), where these were based on particle size fractions that stay constant with time.

263. A quantitative uncertainty assessment of the inventory for fine particles has not been undertaken, but the uncertainties must be considered higher than those for the estimates of PM_{10} emissions. A current review has not revealed any additional information on size distribution profiles for sources other than road transport exhaust emissions. Recent data on modern petrol and diesel car exhaust emissions seem to indicate the proportion of PM_{10} emitted as $PM_{0.1}$ is considerably lower than the current inventory figure of 50% by mass.

264. The NAEI produces national maps of emissions of PM and its precursors in the UK. Although these will not replace detailed local inventories, they provide a valuable basis from which a local inventory can be built and validated and are used in modelling assessments required for national policy-making.

4.4.3 Emissions of particle precursors

265. As explained in Section 4.3, the pollutants NO_X, SO_2, NMVOCs and NH_3 all contribute to the formation of PM in the atmosphere. Emissions of these precursor pollutants arise from a number of anthropogenic sources and the main sectors contributing to their emissions in the UK in 2001 are illustrated in Figure 4.7, based on the 2001 NAEI.

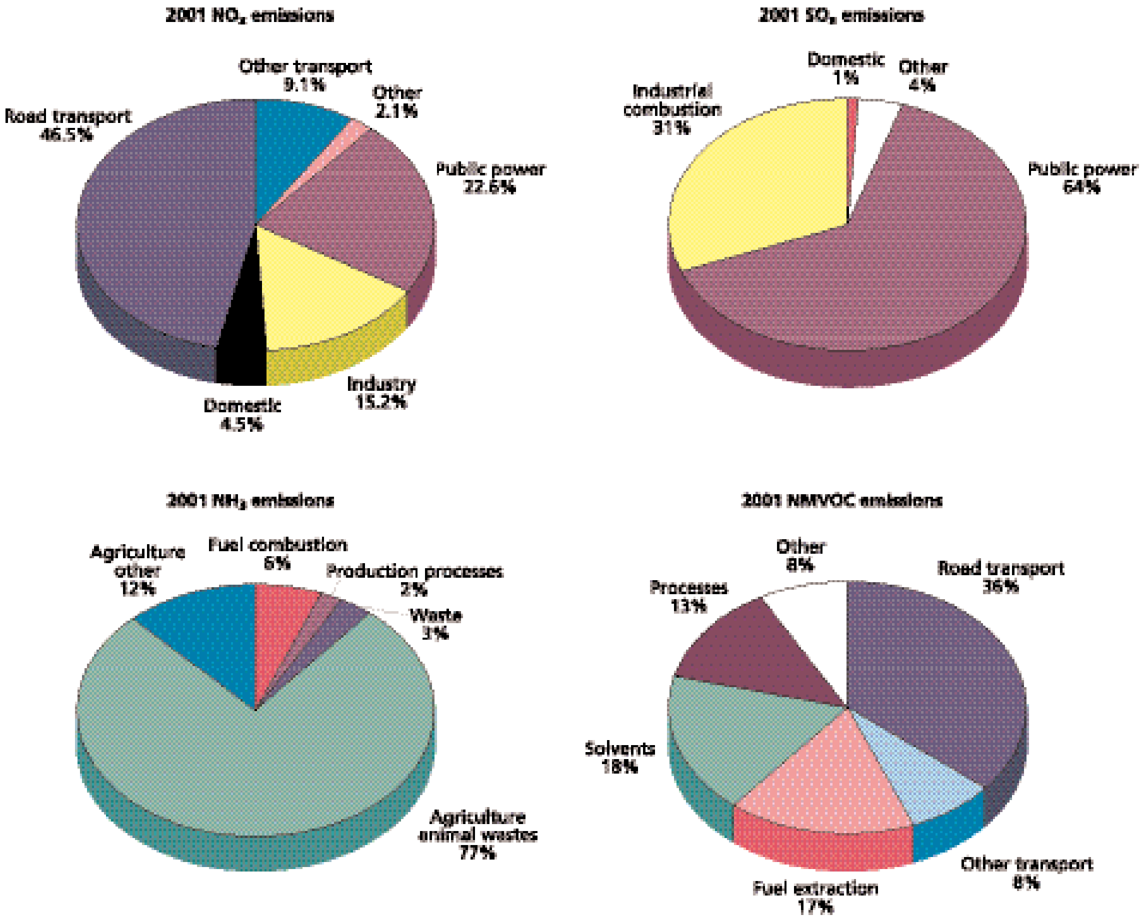
4.4.3.1 NO_X emissions

266. Emissions of NO_X occur mainly from combustion of fossil fuels and have been discussed in detail previously (AQEG, 2004). The most significant sources of NO_X are from road transport and public power generation. Emission occurs as a result of nitrogen present in the air and/or as a trace component in the fuel (for example, in coal) and the high temperatures of combustion. Emission estimates for NO_X from stationary sources are largely based on the use of emission factors combined with national energy statistics. However, emissions data provided by individual process operators for inclusion in the Pollution Inventory are also used, for example, in the cases of power stations and other large combustion plants. For these sources, emission estimates are considered to be of high quality. Area sources – which comprise smaller, but more numerous combustion sources (such as the domestic sector) – cannot be characterised to the same level of detail, and emission estimates are considered less accurate. Emissions from road transport are calculated using emission factors and transport activity data as discussed in Section 4.2.2.4 and in the report on NO_2 by AQEG (AQEG, 2004). Emissions of NO_X from road vehicles have been reasonably well quantified.

4.4.3.2 SO_2 emissions

267. Emissions of SO_2 also occur mainly as a result of the combustion of fossil fuels in which sulphur is naturally present as a trace component in varying amounts. Oxidation to SO_2 occurs during the high temperature combustion process. Sulphur is particularly prevalent in solid fuels such as coal. Consequently, the main consumer of coal – currently the electricity supply industry – is the most important source of SO_2. Total UK coal consumption in 2001 was 63.1 Mt, compared with the 105.9 Mt that were consumed 10 years earlier and the 154.7 Mt consumed 1970. Emissions have also been reduced through the adoption of measures to minimise emissions, including the use of lower sulphur content coals, as well as end-of-pipe abatement technology, such as the use of flue-gas desulphurisation (FGD) at coal-fired power stations. Emission estimates for SO_2 are generally derived by combining emission factors based on fuel sulphur contents with fuel use statistics. Some data provided by process operators for inclusion in the Environment Agencys Pollution Inventory are also used. Consequently, the quality of the inventory for SO_2 emissions is expected to be particularly high.

Figure 4.7 UK emissions of PM precursor pollutants from anthropogenic sources in 2001 by sector (data from the 2001 NAEI).



4.4.3.3 NMVOC emissions

268. Emissions of NMVOCs occur from a wider range of source types than for any other pollutant. The term NMVOC includes many hundreds of chemical species that are emitted to the atmosphere. Each of these has a different propensity to forming PM in the atmosphere based on its structure and chemical reactivity. The NAEI includes information on the species emitted from each source and this information has been published (Passant, 2002); it is also available at www.airquality.co.uk.

269. Major sources include domestic combustion, chemical processes, refineries, whisky production, production and distribution of liquid fossil fuels, industrial solvent use, domestic solvent use, combustion and evaporation of fuels from road and off-road vehicles. Emissions from combustion occur as a result of incomplete combustion of the fuel, reflecting the inefficiencies of the combustion process. Although these are important sources, non-combustion or fugitive sources made up nearly half of all UK emissions of NMVOCs in 2001. Total NMVOC emission estimates are generated via a number of approaches including use of literature-based emission factors, use of estimates provided by industry, estimates based on data held by regulators and emissions data reported in the Pollution Inventory and other inventories compiled by regulators. Fugitive emissions are more difficult to characterise compared to those arising from combustion sources and consequently their emissions estimates are associated with higher degrees of uncertainty.

4.4.3.4 NH_3 emissions

270. NH_3 emissions in the UK are dominated by emissions from agricultural animal wastes from the breakdown of urea and uric acid. Emission estimates for these processes are taken from the official UK NH_3 agricultural emissions inventory, which is compiled annually (Misselbrook *et al*., 2003). This inventory combines official UK agricultural statistics, such as livestock numbers, with emission factors from UK-based measurement programmes. The calculation methodology takes into account parameters such as nitrogen excretion rates and manure management techniques. However, NH_3 emissions are considered to be less well characterised than other pollutants. This is primarily because measurement of NH_3 from agricultural sources is considerably more uncertain than combustion emissions. Emissions from other sources are small, however, the contribution of road transport is growing due to the increasing use of three-way catalysts, which generate some ammonia as a by-product of the catalytic NO_X reduction process in the exhaust. This is discussed in detail in the AQEG NO_2 report (AQEG, 2004).

4.4.4 Biogenic emissions

271. The NAEI also provides estimates of emissions of the precursor pollutants from natural (biogenic) sources. In the UK, these mainly relate to NMVOC and NH_3 emissions, although forest fires and accidental fires can also be sources of NO_X. NMVOCs – in particular isoprene and monoterpenes – are emitted from several natural and agricultural sources such as forests. The emission rate depends on factors such as temperature, light intensity and relative humidity: it, therefore, displays seasonal and interannual variability and may increase substantially during hot summertime weather. Simpson *et al*. (1999) have estimated the total NMVOC emissions in the UK from forests, heathland, pastures and crops to be 178 kt for an average year, but this would be expected to vary with long-term meteorological trends and changes in land use. The NAEI currently adopts this figure, which it is assumed remains unchanged each year and is currently around 13% of anthropogenic UK sources of NMVOCs in 2001. This estimate is currently being reviewed.

272. Emissions of NH_3 occur naturally from humans and domestic and wild animals. The NAEI includes estimates from domestic and wild animals, human breath and sweat and nappies. These amounted to around 22 kt of NH_3 emitted in 2001, around 7.5% of anthropogenic sources including agriculture.

273. Emissions of NO_X occurring from forest and accidental fires are not currently estimated.

Table 4.12 Comparison of 2001 PM_{10} emissions by sector between the NAEI and the LAEI.

Sector	NAEI (t y^{-1})	LAEI (t y^{-1})	LAEI/NAEI (%)
Road transport exhaust	—	—	—
Major roads	—	—	—
Motorcycles	54	71	118
Cars	403	506	122
Taxis	—	97	—
Cars and taxis (combined)	403	603	150
Buses and coaches	116	51	44
LGVs	399	551	138
Rigid HGVs	306	355	116
Articulated HGVs	385	336	87
Minor roads	1104	189	17
Total road transport exhaust	2771	2157	78
Domestic	913	596	65
Other transport	342	204	60
Combustion in domestic, commercial, industry and agriculture	2024	1540	76
Brake and tyre wear	427	0	0
Construction	579	0	0
Total	6911	4517	65

4.4.5 Local emissions inventories

274. The general principles of inventory compilation for local inventories were considered in the AQEG NO_2 report (AQEG, 2004). Local inventories tend to cover specific urban conurbations compiled mostly by the London Research Centre. Particular attention was focussed on the LAEI, in part because London represents the largest urban conurbation in the UK, but also because there is significant dispersion modelling being undertaken for London. It is important, therefore, to consider the differences between the NAEI and LAEI since both have been used for predictions of current and future air quality concentrations and for assessments of source apportionment. Although there were differences highlighted in total NO_X emissions between the NAEI and the LAEI in the AQEG NO_2 report, there was broad agreement by sector. The LAEI typically estimated emissions of NO_X to be less than the NAEI.

275. The situation for particles is, however, more complex for many reasons, including the difficulty in estimating emission rates for some sectors highlighted in Section 4.2. There are currently two releases of the LAEI for 1999 and 2001.

276. The principal difference between the two inventories relates to the activity data used to derive total emissions; identical emission factors have been used for the majority of source sectors. Considering the 1999 LAEI, there are some important differences compared with the NAEI for particles. The most important of these was the assumption in the LAEI that particle emissions from natural gas combustion are negligible. Furthermore, the LAEI did not consider non-exhaust particles from road transport, including those from tyre and brake wear and particle resuspension. These are considered to be significant sources in the NAEI. Nevertheless, non-exhaust particles from road transport were not estimated in the LAEI 2001 version (LAEI, 2003). The Greater London Authority (GLA) is currently reviewing the available information on these sources with a view to incorporating them into the LAEI. Estimates for some sectors, for example, natural gas, were revised in the 2001 release of the LAEI. However, a recent review of PM_{10} emissions from natural gas combustion suggests that emissions should be 25% of current (2001) estimates (see Section 4.2.4.2). Referring to Table 4.12, this revision would reduce the contribution made by natural gas combustion to total London PM_{10} emissions in the LAEI from 20.1% to 5.9%. It is difficult to make a precise comparison between the NAEI and LAEI because of the different source sector definitions used. A summary of the emissions comparison for PM_{10} for 2001 is shown in the Table 4.12.

277. Beyond the differences outlined above, there are other differences that could have a significant bearing on the comparison between the NAEI and LAEI. Some of these differences are outlined below.

- The LAEI and NAEI use the same emissions factors, suggesting that the principal differences between the two inventories are related to emissions activity data.
- The LAEI calculates emissions from the major road network based on the measured hourly distribution of vehicle types.
- The LAEI uses detailed information on vehicle speeds (the 'moving-car-observer' method, where an instrumented car records vehicles speeds on a second-by-second basis on Londons major roads). Different vehicle speed estimates could account for significant differences between the two inventories.
- The LAEI uses London-specific vehicle age and technology information for important vehicle classes such as taxis and TfL buses. In the 2001 LAEI, 60%

of buses were assumed to have particle traps fitted, effectively reducing emissions by 95%. This assumption probably accounts for the differences in PM_{10} emissions between the two inventories shown in Table 4.12.

- Analysis of the vehicle activity and emissions totals shows that the LAEI and NAEI are similar in terms of their estimates of emissions on the major road network, that is, in terms of mass of emissions per unit distance travelled. Furthermore, the total vehicle kilometre estimates within the Greater London area are similar: 29.2 (LAEI) versus 30.9 (NAEI) billion vehicle kilometres. There are, however, differences in the treatment of minor roads. A larger proportion of minor roads are assumed in the NAEI compared to the LAEI since more roads are treated explicitly in the LAEI. Furthermore, the LAEI assumes a different vehicle stock for minor roads and vehicle speed, for example, significantly fewer HGVs and buses. It appears that the latter point accounts for the most important differences between the two inventories for km2 PM_{10} total emissions.

- There are differences in methodology and emission factors used for the calculation of emissions from rail and shipping.

278. Emissions of PM_{10} vary across London because of the patterns of use of different vehicle types in London. There are relatively higher numbers of buses and taxis in central London and this has an important effect on the sources of PM_{10} in this part of the city. Figure 4.8 shows that in 2001, taxis were responsible for an estimated 25% of total road transport emissions in central London, whereas buses were responsible for around 5%. The contribution from LGVs to PM_{10} emissions is important across London. The comparatively low figure for buses is the result of 60% of the bus fleet having particle filters fitted, which were assumed to reduce PM_{10} by 95%. It also should be noted that emissions from the newest buses in Londons fleet are lower still, however, and that this is not yet reflected in the NAEI. This bus fleet in London is newer and lower-emitting than the national bus fleet. This is accounted for in the NAEI, leading to a lower emission rate for buses in London than is apparent for urban areas in the UK as a whole.

279. Figure 4.9 shows the relative contributions of NO_X by area of London. Because particle filters have a very small effect on emissions of NO_X, buses contribute to a relatively higher proportion of NO_X emissions (19%) in central London. These spatial variations in emissions are an important consideration for source apportionment studies.

4.4.6 Changes in the 2002 NAEI

280. The latest version of the inventory (the 2002 NAEI) incorporates some improvements to the methodologies and modifications to activity data or emission factors for certain sectors. These changes have led to revisions in the inventory estimates for PM_{10} emissions produced in the 2001 NAEI, the version used for most of the air quality modelling and assessments. Overall, these have little effect on estimates of total UK emissions of PM_{10}, but the contributions from certain sectors have changed somewhat. The most important changes are summarised as follows.

Figure 4.8 Relative emissions of PM_{10} by area of London (LAEI, 2003).

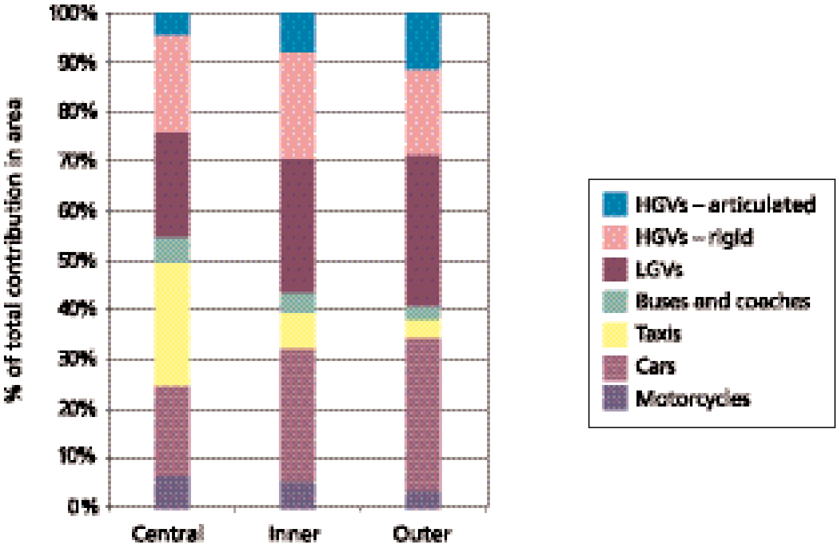
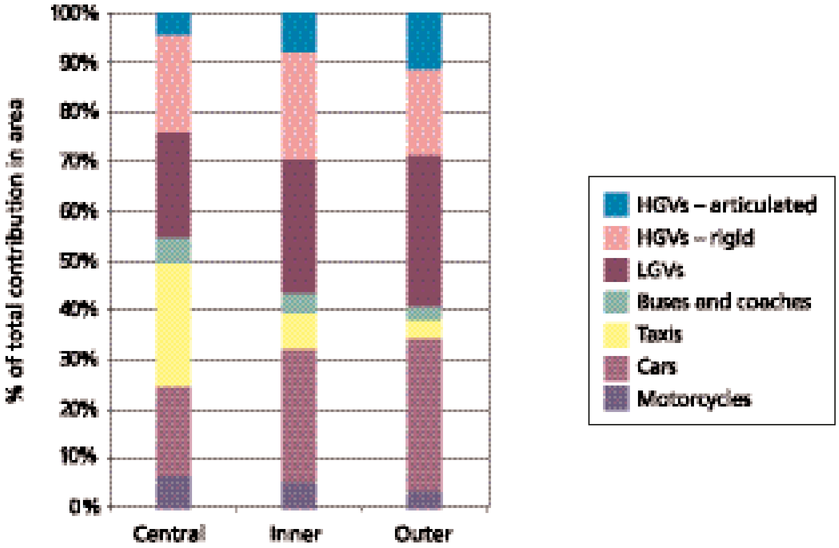


Figure 4.9 Relative emissions of NO_X by area of London (source: LAEI, 2003).



- Revisions to estimation methods for tyre and brake wear emissions. This was discussed in Section 4.2.2.11. The changes led to an increase in the estimate of 2001 emissions from 4.9 kt to 8.9 kt. The change is mainly due to changes in the estimate of tyre wear emissions.

- Revisions to the calculation of emissions from various industrial (mainly coal) combustion sources due to changes in the way emissions data from the Pollution Inventory are used in the NAEI and changes in some of the DTI data on fuel consumption by these sectors. These changes led to an overall reduction in PM emissions.

281. As mentioned in Section 4.2.4, in addition to having made these changes, the NAEI is currently carrying out a review of PM emission factors for a range of industrial processes, combustion, mining and quarrying and construction. The output from this review is expected to feed into the next version of the inventory to be compiled for release in 2005 (2003 NAEI).

4.4.7 Uncertainty in national emissions estimates

4.4.7.1 Quantification of uncertainties in emission estimates for sources covered in the inventory

282. Quantifying the uncertainties in the PM_{10} emission estimates is important if the trends in ambient concentrations and the contributions of different sources are to be understood. This also helps to prioritise future work by pointing to those areas where further information is required to improve the reliability of the inventory. The NAEI has made a detailed and quantified assessment of the uncertainties in the national emission estimates for PM_{10} and other pollutants in 2001 using a direct simulation approach, as described by Passant (2003) and AQEG (2004).

283. The procedure used corresponds to the IPCC Tier 2 approach discussed in the *Good Practice Guidance* (IPCC, 2000), as well as the Tier 2 method proposed in the draft *Good Practice Guidance for CLRTAP Emission Inventories*, produced for inclusion in the *EMEP/CORINAIR Guidebook on Emission Inventories*.

284. Uncertainties in the emission estimates arise from uncertainty in both the emission factor and activity rate used in the calculations. In some cases, emission estimates are supplied directly by process operators or regulators (for example, the Pollution Inventory) and an uncertainty is assessed on this. The main sources of uncertainty in emission inventory estimates are in the emission factors used. Uncertainty estimates for these are largely based on expert judgment, guided by the range in emission factors given by literature sources and elsewhere. Industrial trade associations and industrial process operators provide a final source of expert opinion. The report by Barlow *et al*. (2001) provided an indication of the range of measured emission factors for road vehicles. Table 4.13 gives a summary of the assumptions made for uncertainty for the major PM_{10} sources. The uncertainty factors are under review for some industry sources, e.g. iron and steel, where some industry contacts suggest the uncertainty factors are lower than shown in the table. It should be noted, however, that the uncertainty factors cover all parts of the given industry process.

285. The uncertainty associated with emission estimates for road and off-road transport, small industrial processes and smaller combustion processes is assumed to be greater than the uncertainty for emission estimates for large combustion plants and large industrial processes such as cement production. This is because the emission estimates for the former are all based on literature-based emission factors, whereas the emission estimates for the latter are all taken from estimates for individual plants, provided by process operators and at least some are based on measurements of emissions.

286. The uncertainty analysis gives an overall uncertainty in the annual total for UK PM_{10} emissions in the 2001 version of the NAEI of –19% to +46% at the 95% CI for the mean value emission figure of 191 kt (that is, 155–279 kt). This range is considerably wider than for SO_2 (±3%) and NO_X and NMVOCs (both ±8%). It should be noted that the uncertainty estimates quoted here only refer to uncertainty in the national annual emission totals, not to emissions from particular sources. Neither do they reflect uncertainties in emissions at a particular time or location. The result may appear lower than expected and is a result of a statistical

Table 4.13 Parameters used in the analysis of uncertainty in the PM_{10} inventory for 2001.

Source	Uncertainty expressed as limits relative to the mean value	
	Emission factor	**Activity data**
Combustion:		
Coal and fuel oil-fired power stations	±20%	NA
Other coal combustion	30–300%	±1%
Refinery combustion of fuel oil	±40%	±4%
Domestic wood	50–200%	±30%
Domestic anthracite	30–300%	±1%
Domestic SSF	30–300%	±3%
Natural gas (all sectors)	30–300%	±2%
Agricultural use of straw as fuel	30–300%	±50%
Industrial processes:		
Quarrying	10–1000%	NA
Construction	10–1000%	NA
Non-Fletton bricks	20–500%	±30%
Steel production processes	30–300%[a]	±1%
Chipboard manufacture	20–500%	±30%
Part B processes	20–500%	NA
Cement production	±40%	NA
Road transport:		
Petrol cars/vans	0–100%	±10%
Diesel cars – cold start	±60%	±40%
Diesel vans – cold start	±60%	±60%
Diesel cars/vans – other	±40%	±10%
HGV/buses/coaches	±60%	±5%
Brake wear	10–1000%	±10%
Other transport:		
Off-road vehicles	50 – 200%	±80%
Waste disposal:		
Foot and mouth pyres	50 – 200%	±5%
Agriculture:		
Arable farming	±80%	NA
Livestock housing	±50%	NA

Key: NA, not applicable because emission estimate is taken directly from the Pollution Inventory or from other sources. [a]This covers all parts of the production process. Industry contacts suggest this range is too high: this is under review.

interpretation of roughly similar contributions from several different emission sources each with moderately high uncertainties. It is worth noting that IIASA concluded a very similar level of uncertainty to the UK's figures in their independently derived estimate of UK emissions of NO_X using a similar statistical approach (Suutari *et al.*, 2001).

287. A key source analysis was also undertaken following IPCC Tier 2 methodology to identify the major contributors to inventory uncertainty (Passant *et al*., 2002). Key sources will often, but not always, be those sources that contribute most to national emissions. For PM_{10}, a number of key sources were identified. These were:

- quarry processes;
- domestic combustion of coal, wood and anthracite;
- construction processes;
- coal-fired power stations and autogenerators;
- road vehicle brake wear;
- gas oil use in off-road vehicles and machinery;
- industrial burning of coal;
- domestic gas combustion;
- livestock housing; and
- Part B processes (vehicle respraying, timber processes and so on).

288. The results of the uncertainty analysis apply to estimates for the year 2001 and the 2001 version of the NAEI only. One issue that should be considered is that since emissions are believed to be declining from many sectors as a result of legislation, it is vital that the emission factors and other data used in the NAEI are updated regularly to reflect any reductions in emissions that are occurring. This will require continuous and detailed research to be carried out to provide evidence for any reductions, thereby allowing the emission factors used for the annual NAEI emission estimates to continue being up-to-date. A case in point is the road transport emission factors.

4.4.7.2 Sources omitted in the inventory

289. The statistical method used in the uncertainty analysis is useful for investigating the likely impact of uncertainties in emission estimates. However, it is not helpful for identifying missing sources in the inventory and the impact this has on uncertainties. A study of uncertainty in the NAEI identified a number of potential sources of PM emissions that have been previously omitted from the NAEI, although some of these sources have been included in the current 2002 version of the NAEI (Passant, 2003). Emissions from the omitted sources were not expected to be important, it being considered unlikely that their inclusion would increase the UK estimate of PM_{10} emissions by more than a few per cent. They could be more important at particular times at a local level. The sources still omitted from the NAEI are:

- flaring of waste gases at oil refineries and at chemical works;

- use of chemical process wastes as fuels;
- use of natural gas as a fuel by railway operators and processes for the manufacture of ammonia;
- use of coke by the iron and steel industry but excluding coke use in coke ovens, sinter plant and blast furnaces;
- fuel use by military aircraft;
- natural fires, for example, forest fires, and accidental fires, such as house fires and car fires;
- small-scale burning of waste including garden and other bonfires; and
- burning of wastes produced during well testing on offshore oil and gas installations.

290. In some cases insufficient data are currently available to enable emissions to be estimated. In other cases (including the third, fourth and fifth items in the above list) it will be possible to include emission estimates in future versions of the NAEI. Of the remaining sources, the priorities for estimation of emissions should be natural and accidental fires and small-scale burning of waste. It is recommended that research be done to collect the data necessary for estimating emissions from these sources.

291. Uncertainty assessments were also undertaken for emissions of black smoke, which were more uncertain than PM_{10} emissions (+76 to –50 %). Also of relevance is the uncertainty of important particle precursor emissions.
The uncertainty in the UK totals are:

NO_X ±7%, SO_2 +4 to 3% and NH_3 +19 to –18%.

Overall there is more uncertainty associated with primary particle emissions compared with the particle precursor emissions.

4.4.7.3 Local emissions in critical areas and ratio to NO_X

292. The limit values and objectives place specific emphasis on the characterisation of local emissions at hotspots, in particular busy roads. However, due to the uncertainties in traffic flow characteristics, and the potential contribution from road traffic resuspension, quantification of emissions at critical urban locations such as these can give rise to substantial problems.

293. It is useful to carry out a comparison of PM_{10}:NO_X ratios for both emissions and concentrations, and this has been carried out for the Marylebone Road (kerbside) and London North Kensington (background) sites for 2001. Table 4.14 overleaf summarises the NO_X and PM_{10} concentrations at each site.

Table 4.14 Concentrations of NO_X and PM_{10} at Marylebone Road and Kensington (2001).

	NO_X (μg m^{-3})	PM_{10} (μg m^{-3}, TEOM)
Marylebone Road	337	33
North Kensington	75	20
Increment	262	13

294. The PM_{10}:NO_X ratio of the increment in concentrations is 0.05 (mass basis), which can be used to represents an estimate of the ratio of road traffic emissions from Marylebone Road. Detailed emissions calculations from the LAEI suggest a ratio of 0.063 (mass basis) for 2001. The LAEI emissions estimate includes only exhaust emissions, and does not include emissions from tyre and brake wear or from resuspension.

295. These values can be used in a dispersion model to predict the PM_{10} concentration that is due to vehicle emissions alone, assuming that the model is able to produce accurate estimates of NO_X. On this basis, a dispersion model would predict 262*0.063 = 16.5 μg m^{-3} PM_{10}. This 16.5 μg m^{-3} can then be added to the background concentration of 20 μg m^{-3} (measured at North Kensington) to yield a total of 36.5 μg m^{-3}.

296. The predicted PM_{10} concentration of 36.5 μg m^{-3} is greater than the measured concentration of 33 μg m^{-3} but does not include the additional contributions from tyre and brake wear and resuspension. Inclusion of these other sources would lead to a predicted concentration significantly higher than the measured value. On this basis that there appears to be an important discrepancy between PM_{10}:NO_X ratios based on emissions data and ambient concentrations at this location. Further analysis of other sites is required to determine whether there is a similar conclusion, and if so what is the explanation.

4.5 Time series of emissions

4.5.1 UK emissions

4.5.1.1 Stationary fuel combustion

297. The decline in UK emissions of PM_{10} with time shown in Figure 4.10 is primarily due to the reduction in coal use and the more extensive use of natural gas. In Figure 4.10, the category 'Other' includes emissions from off-road transport, waste treatment and disposal as well as emissions from agriculture. Emission estimates for the resuspension of dust from roads are not typically included in officially reported UK emissions (as the source is not a primary source). However, for completeness, the emission estimates from this source are included. Domestic emissions (a subset of commercial, residential and institutional combustion) have fallen from 222 kt in 1970 (41% of the total emission) to 33 kt in 2001 (18% of the total). Similar reductions were observed from the public power and industrial combustion sectors. In the UK, fuel switching from coal to gas has been driven by the Clean Air Acts and price differentials. However, domestic coal can still be a significant source in some smaller towns and villages, in Northern Ireland and in areas associated with the coal industry.

298. There has also been a drive to control particulate emissions from large industrial sources. So in addition to fuel switching, reductions can also be attributed to the installation of abatement equipment (and to a lesser extent increases in plant efficiencies). There are a number of less well-pronounced features evident in Figure 4.10. Emissions in 1984 were significantly lower than previous years due to the miners' strike. Considerably less coal was used for both electricity generation and domestic heating, which was reflected by a marked decrease in PM_{10} emissions. For a number of other years, elevated emissions from the domestic sector can be attributed to particularly cold winters.

4.5.1.2 Production processes

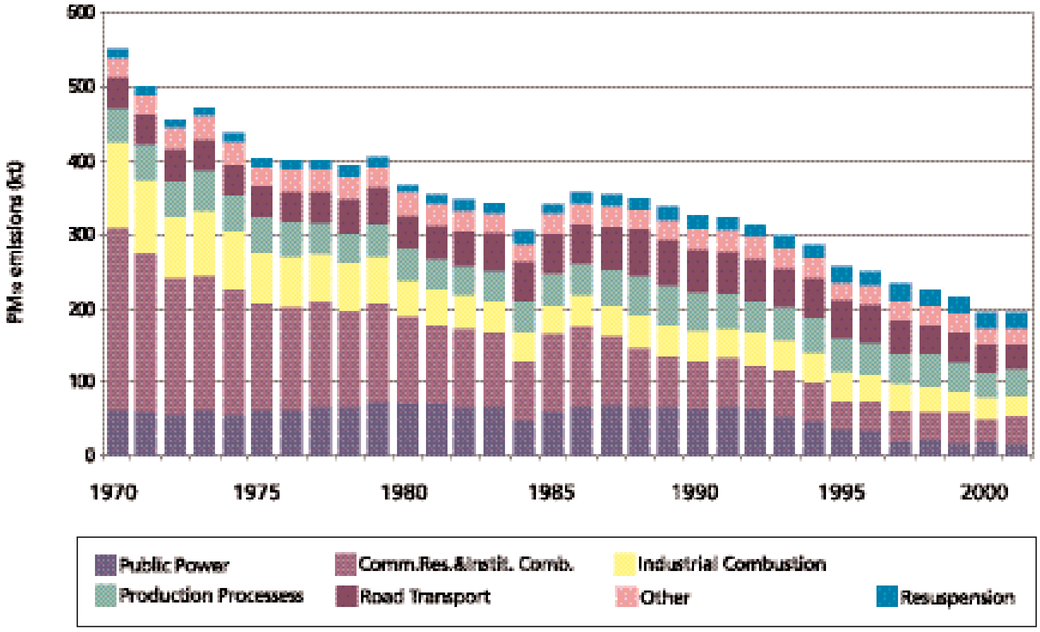
299. These include industrial sources not associated with combustion, that is, the production of metals, cement, lime, coke and chemicals as well as bulk handling of dusty materials, construction, mining and quarrying. Emissions from these sources are difficult to quantify due to the contribution of fugitive emissions. This also makes it inherently difficult to abate emissions and helps to explain why there has been little decrease with time during 1970–2001.

4.5.1.3 Road transport

300. Emissions from road transport (exhaust and tyre and brake wear) decreased by 38% between 1990 and 2001 in spite of a continuous increase in traffic over this period. The decrease is mainly due to the penetration of petrol cars fitted with catalysts in the fleet, the phasing out of leaded petrol and the fleet penetration of newer heavy duty diesel vehicles meeting tighter emission standards. Although emissions of PM from petrol cars are small compared with diesel vehicles, the large number of petrol cars without catalysts in the fleet in the late 1980s meant they made a significant contribution to the PM inventory. Their contribution has declined as lower PM-emitting cars with catalytic converters penetrated the fleet in the 1990s, replacing non-catalyst cars. Furthermore, PM emissions from non-catalyst cars running on leaded petrol are estimated to be around a factor of three times higher than emissions from non-catalyst cars running on unleaded petrol. So the gradual phasing out of leaded petrol during the 1990s helps further explain the trends in emissions from petrol cars. The downward trend in road transport emissions of PM has been tempered by the fact that emissions from diesel vans and cars have been increasing over this period. Although European emission standards have been reducing PM emissions from these vehicles over the 1990s, this has been offset by a large growth in diesel car and van activity between 1990 and 2001. The increase in emissions appears to be reaching a peak now as the impact of tighter Euro standards starts to dominate.

301. The time series of UK PM_{10} emissions is shown in Figure 4.10 split by source sector.

Figure 4.10 Time series of UK PM_{10} emissions.



4.5.2 Emission trends by particle size: the 2001 NAEI time series

302. Typically, particles arising from sources such as quarrying and other processes are the coarsest. The combustion of solid fuels also results in relatively coarse particles, compared to the fine particulate from gas combustion. As a result, these sectors give rise to relatively large emissions for PM_{10}. For the finer particulate emissions, road transport becomes the dominant source. Emissions from gas combustion give rise to fine particulate material; however, the small magnitude of particulate emissions gives rise to relatively lower emissions from these sources.

303. The uncertainties in the emission estimates and the time series of the finer particle size ranges must be considered to be high and more uncertain than the estimates for PM_{10}. As discussed in Section 4.4.2, it is not clear how appropriate the size profiles used in the inventory are for the UK and how they are effected by abatement. Due to the lack of information, the inventory assumes that the size fractions for each source remain unchanged with time, implying that abatement has no effect on the particle size distribution. This may not be the case, as many particulate emission controls for stationary sources may be more effective for the coarser particles than the finer ones ($<PM_1$). This means that applying constant particle size fractions to the inventory for PM_{10} emissions from each source could lead to underestimations of the finer particle size emissions. A critique of the NAEI PM size fractions is currently being prepared. Figure 4.11 indicates the emissions for different particulate size fractions by sector, based on the 2001 NAEI.

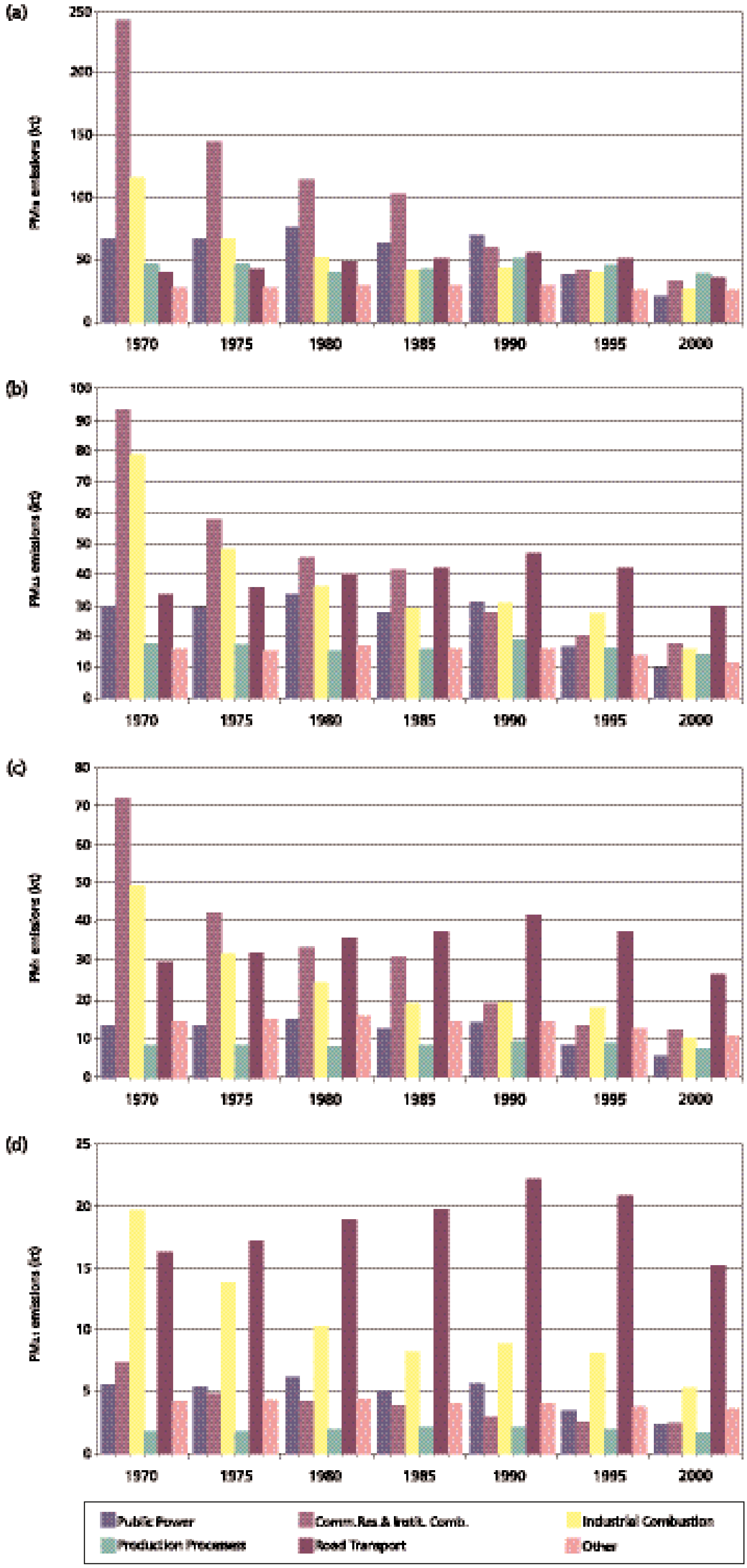
4.5.3 Emissions by fuel type

304. This section highlights the emissions of PM through the combustion of major fuel types and particle size (PM_{10} and $PM_{0.1}$). Two major source sectors are considered: public power and commercial, residential and institutional combustion.

4.5.3.1 Public power

305. Historically, emissions of PM have been dominated by emissions from coal, as shown in Figure 4.12. Despite the large reduction in emissions from coal use,

Figure 4.11 Time trends in PM emissions for (a) PM_{10}, (b) $PM_{2.5}$, (c) PM_1 and (d) $PM_{0.1}$ (source: NAEI, 2001).



these emissions are still large compared to those from other fuels. For the smaller particle sizes there is less impact from coal combustion and an elevated impact from gas combustion.

4.5.3.2 Commercial, residential and institutional combustion

306. Emissions from this sector follow a similar pattern to power generation, that is, the sector is dominated by coal combustion, as shown in Figure 4.13. However, the combustion of gaseous fuels is more important for fine particle emissions compared with the power generation sector. In this sector there is a relatively high contribution from wood, straw and MSW. Emissions from the combustion of wood are more important to total emissions of PM_{10}, whereas the combustion of straw is particularly important at $PM_{0.1}$. Emissions from MSW are small.

4.5.4 Trends in PM_{10}–$PM_{2.5}$ emissions

307. Figure 4.14 shows the trends in PM_{10}–$PM_{2.5}$ from 1990 to 2001 for total UK emissions by source sector. The most significant reductions have been for the energy production and industrial combustion sectors, which have seen reductions of 75% and 46% in emissions between 1990 and 2001, mostly as a result of a reduction in the use of coal in these sectors. Overall the reduction in PM_{10} $PM_{2.5}$ has been 44%.

Figure 4.12 Time series trends in PM emissions from public power for (a) PM_{10} and (b) $PM_{0.1}$.

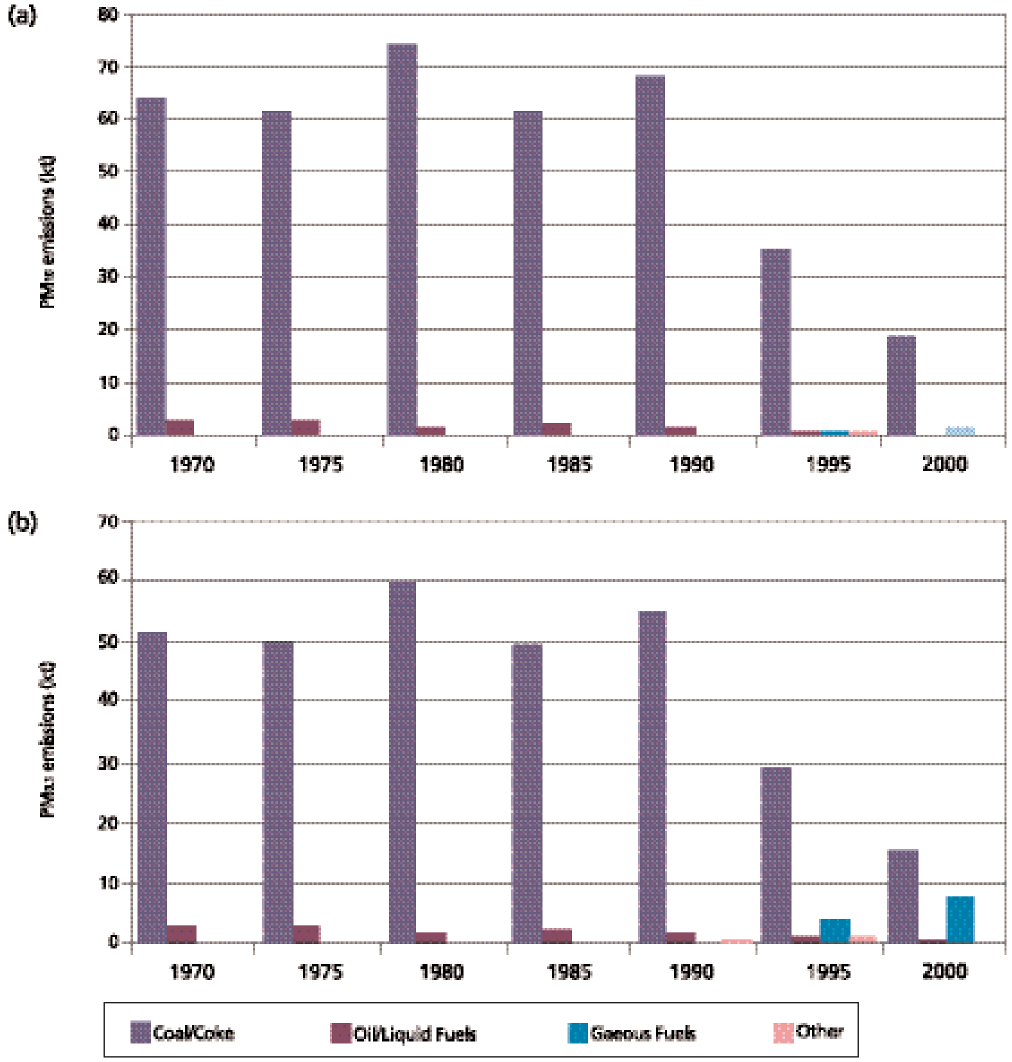


Figure 4.13 Time series trends in PM emissions from commercial, residential and institutional combustion for (a) PM_{10} and (b) $PM_{0.1}$.

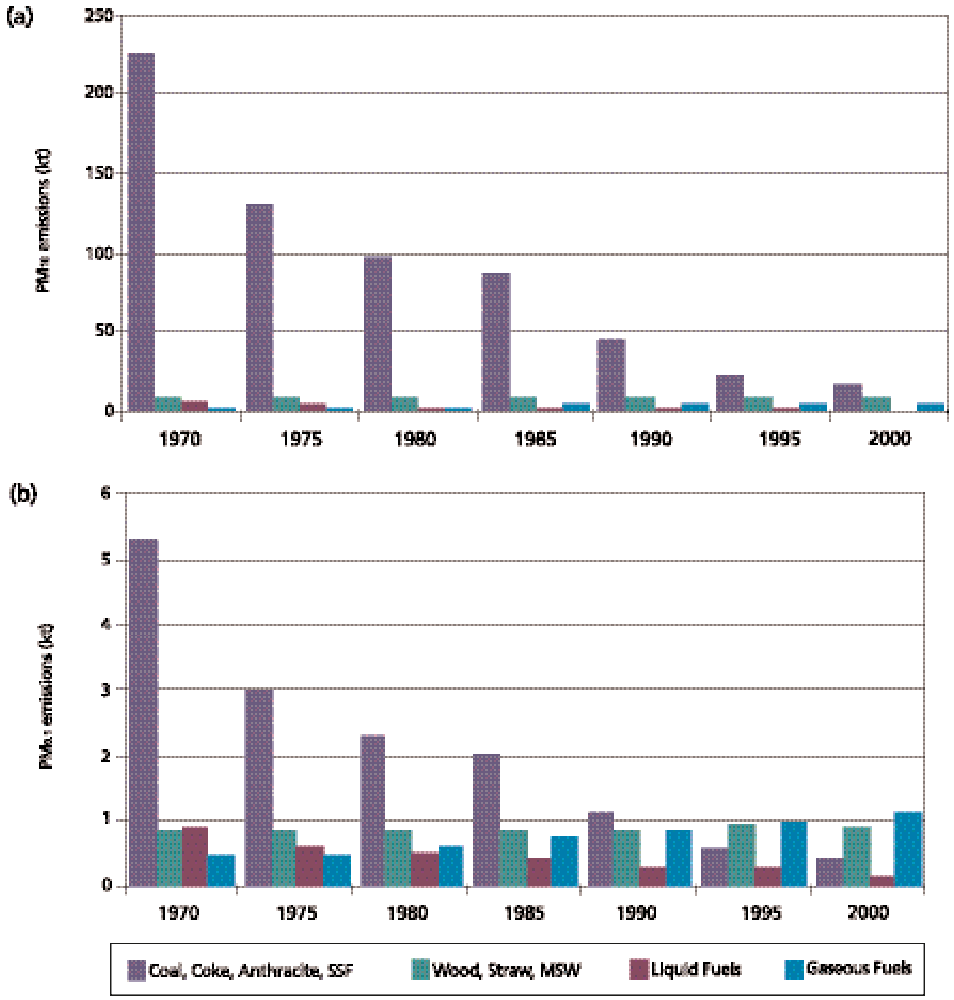
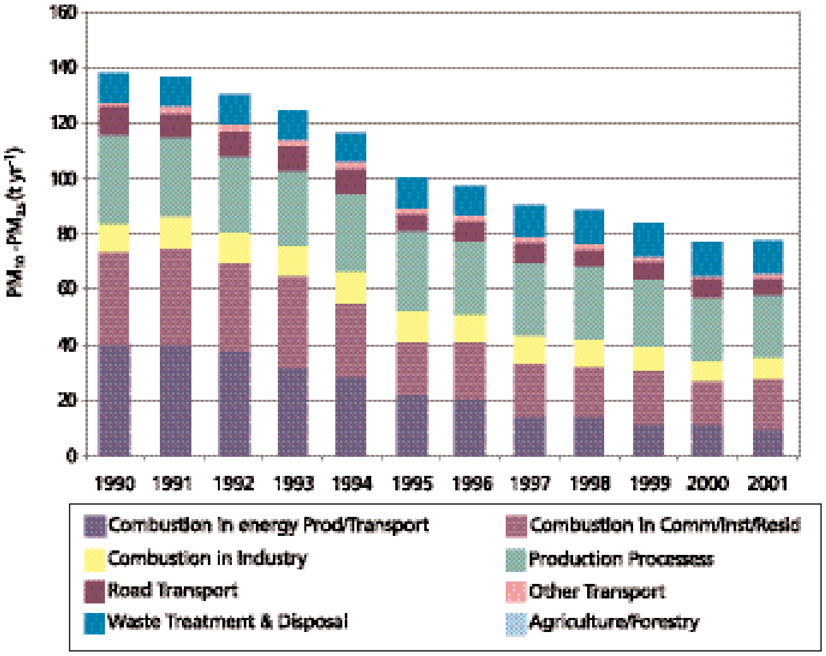


Figure 4.14 Trends in PM_{10}–$PM_{2.5}$ from 1990 to 2001.



4.5.5 Trends in UK precursor emissions

308. This section discusses the temporal trends in UK emissions of the precursor pollutants to PM in the atmosphere, as indicated by the 2001 NAEI.

4.5.5.1 Temporal trends of NO_X emissions

309. Since 1990 there has been a 39% reduction in total NO_X emissions; however, this decrease in emissions has not been constant (Figure 4.15). Up to 1984 the NO_X emission profile was relatively flat with small peaks in 1973 and 1979, as seen previously for CO_2; these peaks were due largely to the cold winters during those years. However, from 1984, emissions rose markedly as a result of the growth in road traffic reaching a peak in 1989. Since 1989, total emissions have declined by 40% as a result of a 51% reduction from power stations and 40% decrease from road transport. The decrease in road transport emissions since 1990 has been due mainly to the introduction of catalytic converters on petrol cars and stricter regulations on emissions from diesel vehicles.

310. Emissions from power stations were relatively constant in the 1970s, but have been declining since 1979. In the 1980s this was due to the increased use of nuclear power and an increase in the average efficiency of thermal power stations. However, since 1988, the electricity generators have adopted a programme of progressively fitting low-NO_X burners to their large coal-fired stations. The introduction of modern combined cycle gas turbine (CCGT) plants burning natural gas in the early 1990s and the decreased share of coal for power generation in favour of gas further reduced NO_X emissions from this sector up until 1999. Emissions of low NO_X gas turbines are lower than those of pulverised coal-fired plants, even when these are fitted with low-NO_X burners. However, this trend was broken in 2000 when output from coal-fired stations grew again at the expense of nuclear power production. Further growth in electricity output from coal-fired stations occurred in 2001, this time at the expense of gas, possibly due to the increased price of natural gas supplies. The trend in NO_X emissions has been discussed in detail (AQEG, 2004). Emissions from industry have declined by 26% since 1990, again mainly due to the decline in coal use in favour of gas.

Figure 4.15 Trend in UK total NO_X emissions by sector.

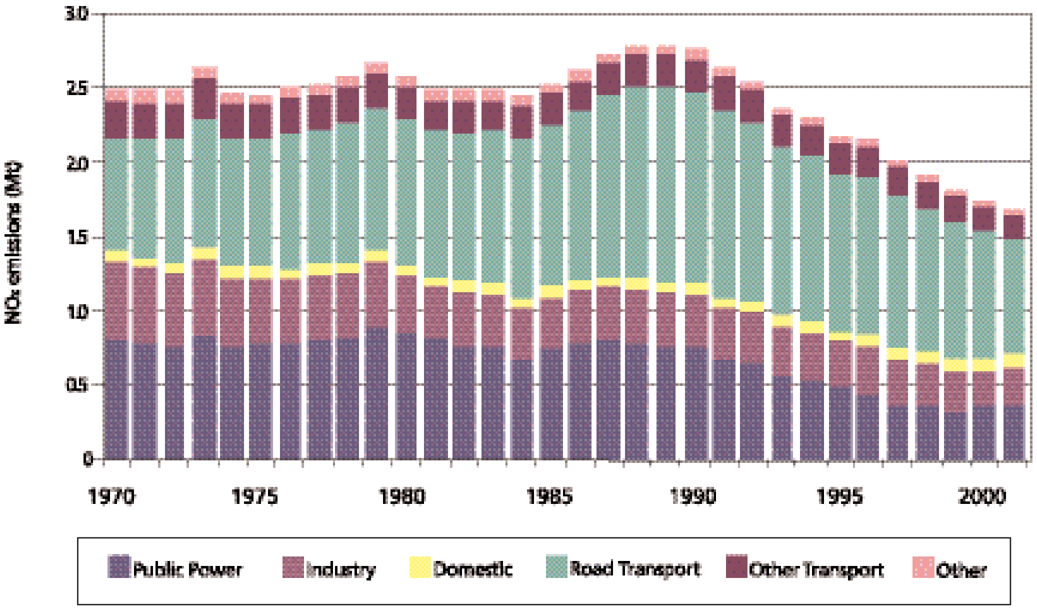
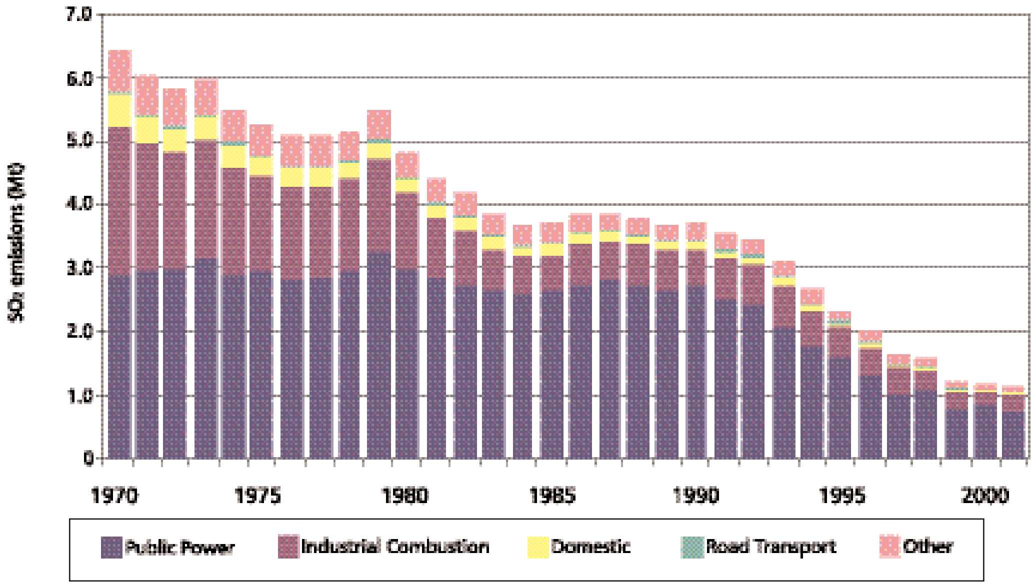


Figure 4.16 Trend in UK total SO_2 emissions by sector.



4.5.5.2 Temporal trends of SO_2 emissions

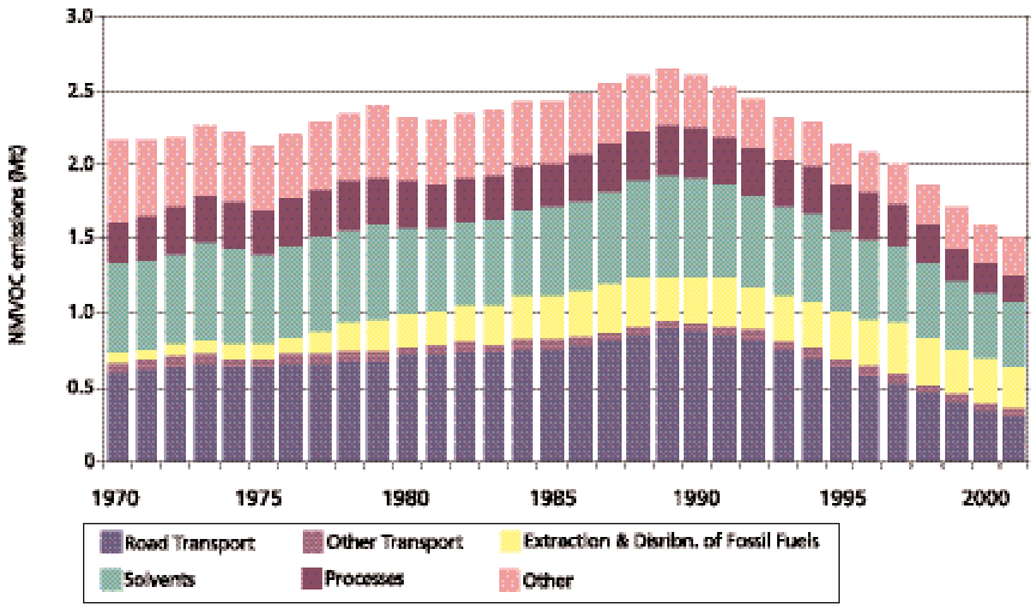
311. Since 1970 there has been a substantial overall reduction of >83% in SO_2 emissions (Figure 4.16). Emissions have been falling fairly steadily over this period. Power stations are the largest contributors to SO_2 emissions, accounting for 64% of UK emissions in 2001. However, emissions from power stations have fallen by 74% since 1970. During the 1980s, the decrease was due to the increased use of nuclear power at the expense of coal and an increase in average efficiency of thermal power stations. Since 1990, this decline has accelerated because of the increase use of CCGT stations and other gas-fired plants. CCGTs are more efficient than conventional coal and oil stations and have negligible SO_2 emissions. Most recently the FGD plants constructed at Drax and Ratcliffe power stations have had a significant effect on emissions.

312. SO_2 emissions from industry fell by 90% from 1970 to 2001. This was due to a decline in the use of coal and oil in favour of natural gas for combustion as well as a decline in the energy intensive iron and steel industry and other heavy industries. Emissions from domestic and other commercial/institutional sectors have declined substantially during the period 1970–2001, reflecting the major changes in fuel mix from oil and coal to gas.

4.5.5.3 Temporal trends of NMVOC emissions

313. Emissions of NMVOCs have fallen by 42% since 1990, when they were near their peak, as shown in Figure 4.17. The road transport sector has been mainly responsible for this fall. Emissions from road transport have decreased by 66% since 1990, mainly due to the penetration of cars fitted with three-way catalysts, which are very efficient at reducing NMVOC emissions from exhausts. The fitting of carbon canisters on cars and the decline in the volatility of summer fuel blends have reduced evaporative emissions from vehicles, also contributing to the fall in NMVOC emissions from road transport. Emissions from solvent use have decreased by 37% since 1990. This has been partly due to emission controls, technological changes and reduced manufacturing output in some sectors. Emissions from the domestic use of solvents have also declined due to a trend

Figure 4.17 Trend in UK total NMVOC emissions by sector.



towards formulating products such as paints and aerosols with lower solvent contents. Emissions from various production processes have fallen by 46% since 1990. This has been mainly due to reductions in emissions from petroleum refineries and the chemical industry. Emissions from food and drink manufacture have risen slightly over this period. Emissions from offshore oil and gas have increased since 1990 and are now contributing 11% of total NMVOC emissions.

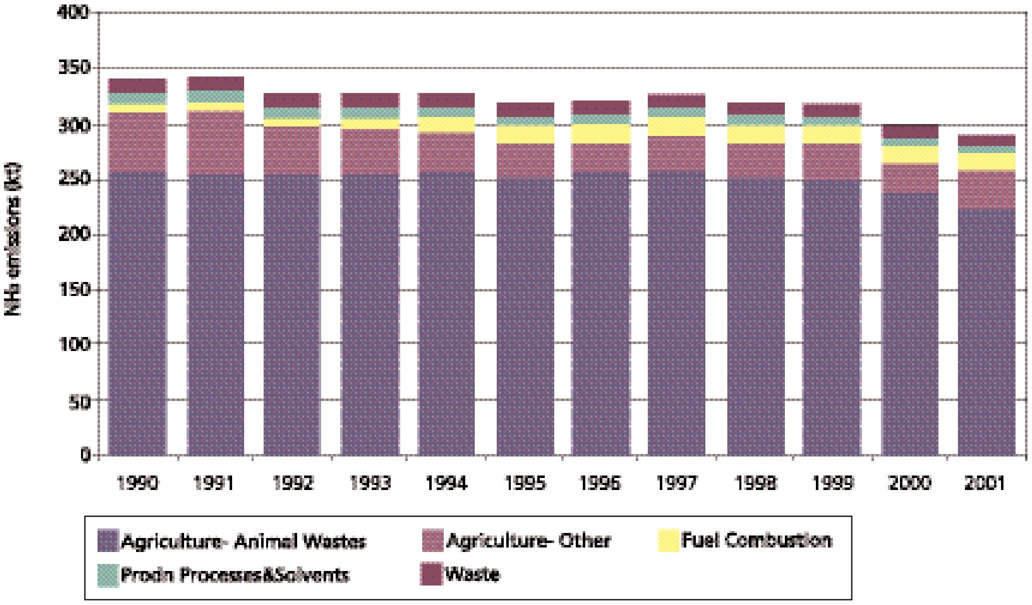
4.5.5.4 *Temporal trends of NH_3 emissions*

314. Ammonia emissions have fallen by 15% since 1990, as shown in Figure 4.18. Emissions are dominated by agricultural sources with emissions from livestock and their wastes comprising 77% of the total emission. These have been decreasing as animal numbers have been declining. During 2001 the foot and mouth outbreak had a significant impact on the animal livestock numbers. However, a degree of restocking occurred after the outbreak had finished. As a result, across an annual period, the impact of the foot and mouth outbreak does not stand out as being a particularly strong feature in the time series. The other agricultural sources contributing to NH_3 emissions include fertiliser use, crops and decomposition of agricultural vegetation. A decline in fertiliser use has led to a reduction in emissions from this sector. The non-agricultural sources are diverse and contribute 11% to the total emissions. However, emission estimates for these sources are very uncertain due to a lack of data. Emissions of ammonia from road transport, although relatively small, are increasing as a result of the increasing number of three-way catalysts in the vehicle fleet.

4.5.6 Trends in the PM_{10} to NO_X ratio

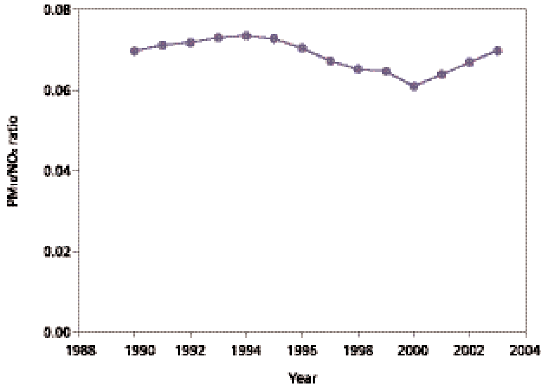
315. It can be useful to consider trends in the ratio of pollutants since they can provide some information on the validity of emissions inventories compared with atmospheric measurements. The ratio between PM_{10} and NO_X is important because this ratio is used in some receptor-based modelling approaches. Figure 4.19 shows the ratio of PM_{10}:NO_X since 1990 based on the NAEI 2002 methodology for calculating road transport emissions. The PM_{10} emissions include

Figure 4.18 Trend in UK total NH_3 emissions by sector.



exhaust emissions, tyre and brake wear, but no resuspension estimate. The 2002 NAEI has revised the estimates of particle emissions of tyre and brake wear and assumes that tyre wear is more important than previous inventory estimates. Figure 4.19 shows how the ratio declined from 1994 to 2000, but since 2000 has begun to increase again. The upward trend since 2000 mostly results from the increased purchase and use of diesel cars, which has kept urban PM_{10} emissions almost constant during 2000–2003, whereas emissions of NO_X have declined.

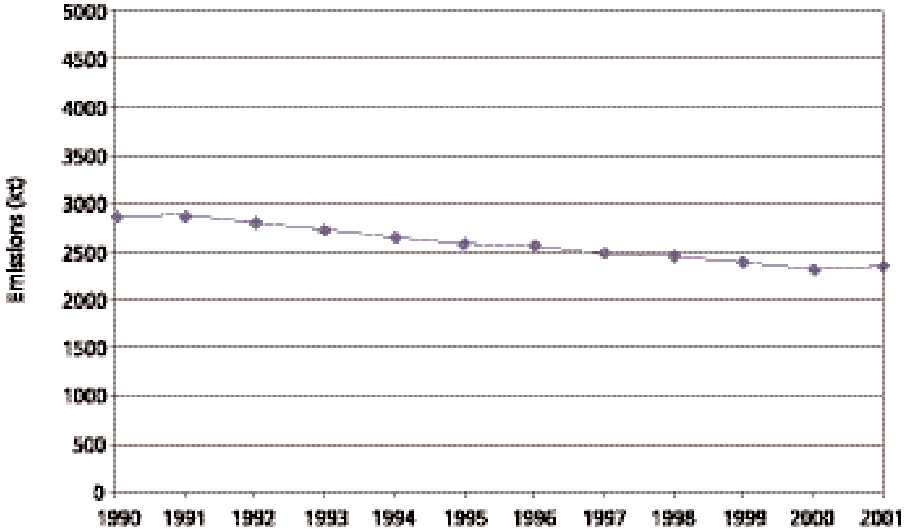
Figure 4.19 PM_{10}:NO_X emissions ratio for urban road transport emissions using the NAEI.



4.5.7 Emissions of primary particulates in Europe

316. An indication of European emissions and time trends for PM_{10} and particulate precursor species can be obtained from the national and sectoral emissions data officially reported by countries to UNECE/CLRTAP. A detailed analysis of European emission sources and time-series trends has also recently been prepared by the EC's CAFE Working Group on Particulate Matter (CAFE WG, 2003). The following analyses are based on the most recent data available, which were reported in 2003. Where countries did not report data, estimates of primary PM_{10} national totals were obtained from the Auto-Oil II programme (European Commission, 2000) for the EU15 countries (the 15 countries that were EU members prior

Figure 4.20 European PM_{10} emissions 1990–2001.



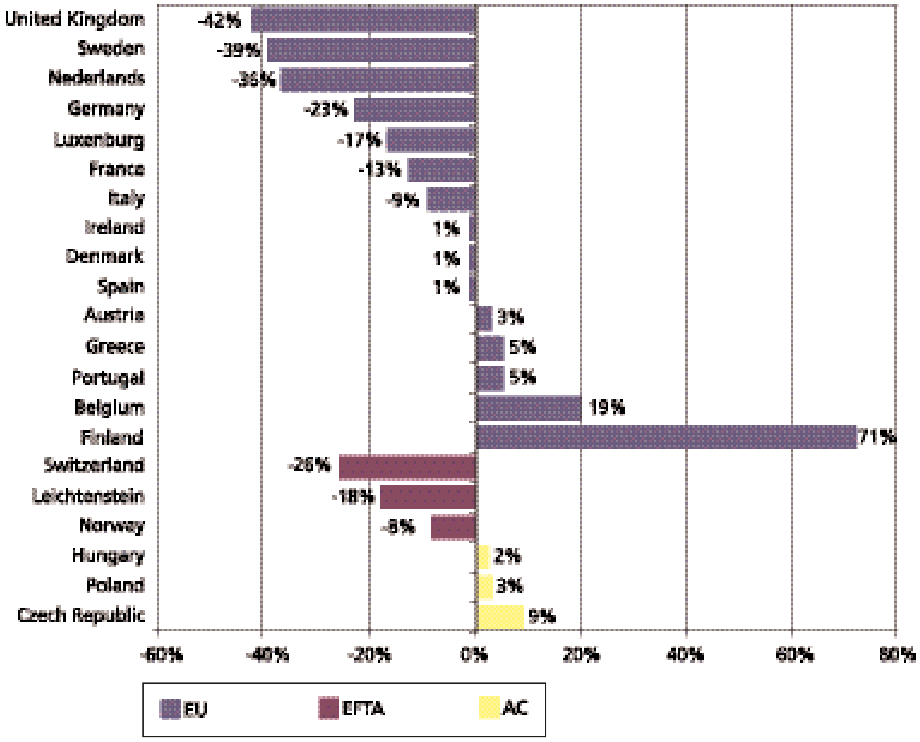
to the 2004 expansion) and from CEPMEIP (2001) for other countries. Gaps in reported time-series data have been filled using the methodology employed by the European Topic Centre on Air and Climate Change (ETC-ACC). Figure 4.20 illustrates the reduction in emissions of PM_{10} during 1990–2001 for the EU15 countries.

317. Emissions across all countries have decreased by 15% between 1990 and 2001. The vast majority of this decrease is primarily due to reductions that have occurred in the EU15, where emissions have decreased by 18% during this period. Emissions from the accession countries are around 21% of the EU15 emissions in 2001. However, this is based on an estimate for 1995 and the time series of emissions from the accession countries (and levels in 2001) is not certain due to lack of reported data. The EFTA4 countries (Liechtenstein, Norway and Switzerland only – Iceland did not report emissions) show only a relatively small level of emissions compared to the other country groupings. In 2001, emissions from this region accounted for 3% of the European total. Figure 4.21 shows the percentage change in PM_{10} emissions on a country-by-country basis. As the figure shows, most EU countries showed a significant decrease in primary PM_{10} emission reductions over the period 1990–2001.

318. The reasons why the Finnish emissions have increased steeply over this period are not currently known, and the emissions reported by Finland to UNECE/CLRTAP submission in 2003 are consistent with the values reported in previous years (such as 2002). In comparison with other European countries, the UK has shown the largest percentage decrease during this period (–42%) and, in absolute terms, the second largest emissions decrease among European countries (–131 kt), the first largest being Germany (–264 kt).

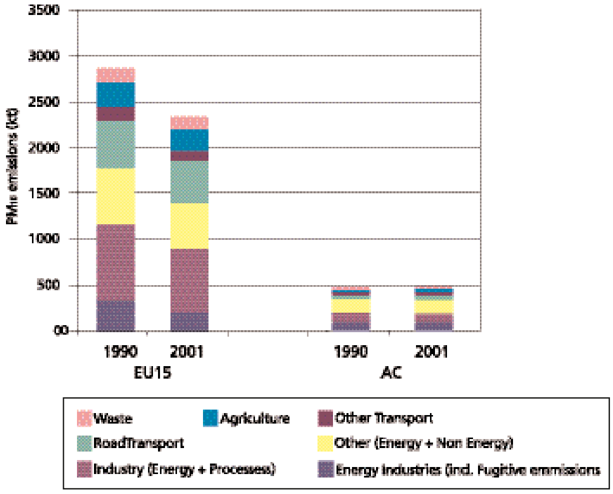
319. In the EU15, PM_{10} emissions are dominated by sectors that include road transport, industry and 'other' (for example, fuel combustion in the residential sector) (Figure 4.22). In the accession countries, the largest sectoral contribution of PM_{10} emissions comes from the 'other' (energy and non-energy) sector, with only a small contribution from road transport. This difference is probably due to the smaller vehicle fleets, the greater relative use of solid fuels (in comparison to natural gas in power generation) and the lower use of effective abatement technologies in the power generation sector.

Figure 4.21 Percentage change in PM_{10} emissions by country, 1990–2001.



320. Sectors contributing most to the reduction in PM_{10} emissions between 1990 and 2001 have been the energy industries and industry sectors and road transport (Figure 4.22). In energy industries fuel switching to natural gas and the use of and improvements to technologies such as electrostatic precipitators have contributed to the decrease in emissions observed. In the road transport sector, tighter PM_{10} emission regulations have been introduced in Europe for heavy duty vehicles and for cars.

Figure 4.22 Emissions of PM_{10} by sector, 1990 and 2001.



4.5.8 Emissions of PM precursor gases in Europe

321. Figure 4.23 illustrates the European (EU15, EFTA4 and AC10) emission trend between 1990 and 2001 for the particulate precursor species SO_2, NO_X, NH_3 and NMVOCs (AC10 refers to the ten accession states that joined the EU in 2004). The most significant reduction in emissions during this period was for SO_2, which was predominantly due to fuel switching from high sulphur solid and liquid fuels

Figure 4.23 Emissions of particulate precursor gases 1990–2001 in Europe (EU15, AC10 and EFTA4 countries).

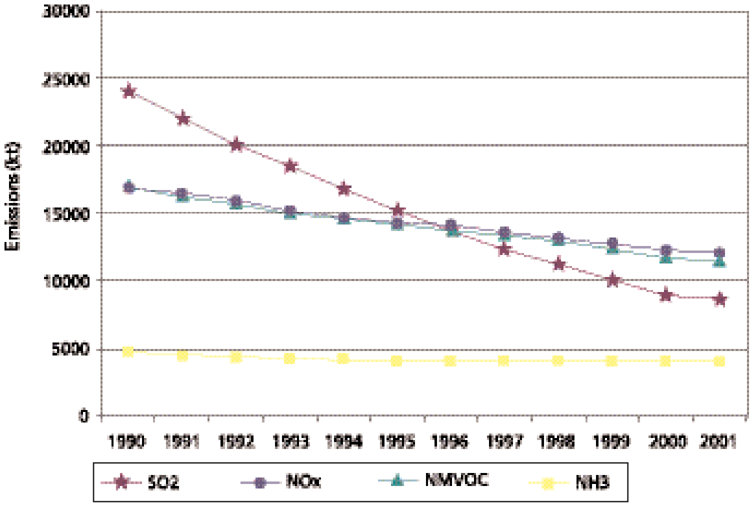


Figure 4.24 Emissions of NO_X by sector, 1990 and 2001.

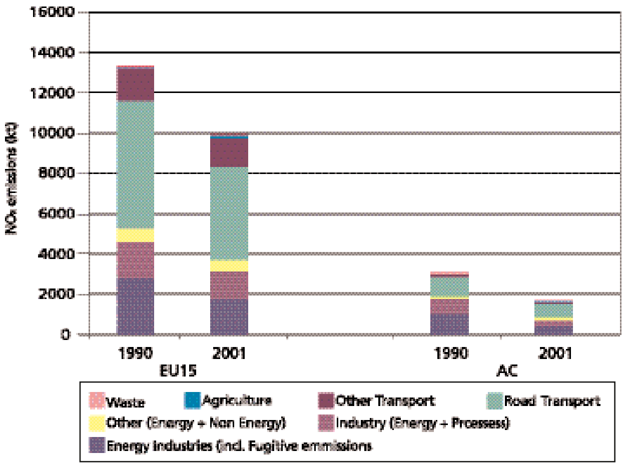


Figure 4.25 Emissions of SO_2 by sector, 1990 and 2001.

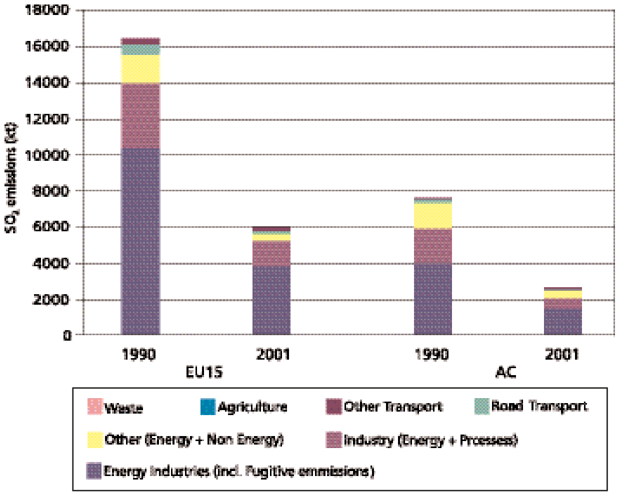


Figure 4.26 Emissions of NMVOC by sector, 1990 and 2001.

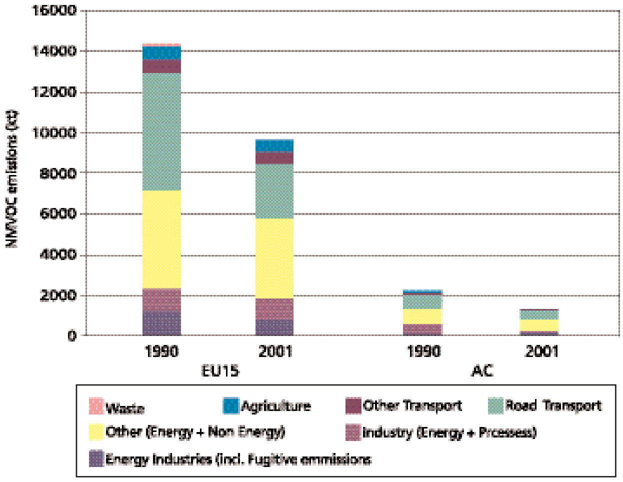
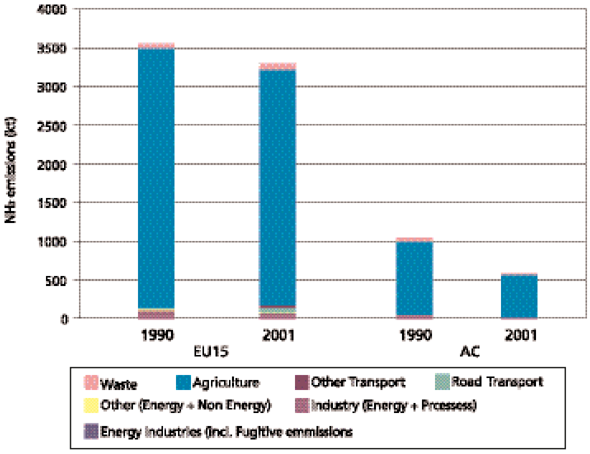


Figure 4.27 Emissions of NH_3 by sector, 1990 and 2001.



to natural gas in the energy industries and industrial and domestic sectors. The construction of new, more efficient power plants coupled with the use of low sulphur coal and FGD have also contributed to the lower emissions observed. The relative sector contribution to emissions for each of the respective particulate precursor species in 1990 and 2001 are shown in Figures 4.23 to 4.26.

322. To allow UK emissions to be placed in context with those from other European countries, Table 4.15 shows the emissions of PM_{10} together with those of the particulate precursor species in 2001, the most recent year for which data is available.

4.5.9 UK emission projections

323. Projections of UK emissions are compiled by the NAEI to enable comparisons with international commitments and to aid in the development of national strategies and policies. The 2001 base year projections were recently produced for the four National Emission Ceiling Directive Pollutants and PM_{10}. To produce a projection each activity is linked to an activity driver. The majority of these drivers have been obtained from either Energy Paper 68 (EP68) or Oxford economic forecasting (OEF) or, as is the case for road transport, vehicle kilometre and fleet data. In addition to changes in activity-influencing emissions, improvement in abatement measures – often the result of legal requirements – will reduce emissions. Therefore, the emission factors, where relevant, have been varied to account for this.

Table 4.15 Emissions of primary PM_{10} and particulate precursor species, 2001 (in kt).

Country grouping	Country	PM_{10}	NO_X	SO_2	NMVOCs	NH_3
AC	Cyprus	0.6	22	52	14	9
AC	Czech Republic	33	332	251	220	79
AC	Estonia	33	38	92	33	9
AC	Hungary	30	184	401	166	66
AC	Latvia	13	42	13	59	12
AC	Lithuania	20	55	49	71	50
AC	Poland	305	909	1564	577	313
AC	Slovakia	41	106	129	89	28
AC	Slovenia	13	57	66	39	19
AC	Iceland	—	28	27	10	—
AC	**Total**	**490**	**1772**	**2643**	**1277**	**586**
EFTA4	Liechtenstein	0.1	0.3	0.1	0.6	0.2
EFTA4	Norway	64	221	25	385	25
EFTA4	Switzerland	24	92	21	147	68
EFTA4	**Total**	**88**	**313**	**46**	**533**	**92**
EU15	Austria	48	199	37	232	54
EU15	Belgium	77	323	176	259	85
EU15	Denmark	22	207	26	125	102
EU15	Finland	53	220	85	152	33
EU15	France	550	1411	610	1674	779
EU15	Germany	887[a]	1592	650	1606	607
EU15	Greece	41	331	485	267	74
EU15	Ireland	19	125	131	90	123
EU15	Italy	204	1373	758	1336	451
EU15	Luxembourg	5	17	3	13	7
EU15	Netherlands	51	423	89	268	143
EU15	Portugal	21	397	301	492	108
EU15	Spain	115	1404	1425	1533	384
EU15	Sweden	69	248	57	303	54
EU15	**UK**	**178**	**1680**	**1125**	**1336**	**290**
EU15	**Total**	**2342**	**9950**	**5960**	**9687**	**3294**

Source: UNECE/CLRTAP (2003) with additional data for PM_{10} taken from Auto Oil II (European Commission, 2000) for EU15 countries and from AC countries from CEPMEIP (2001), where values were not reported.
[a]Note: In 2003 Germany did not report values for PM_{10} emissions to UNECE/CLRTAP and so the value of 887 kt for emissions of PM_{10} in Germany is taken from the Auto-Oil II programme (European Commission, 2000), as was done for other EU countries that did not report. However, Germany did report a total suspended particle(TSP) emission of 251 kt for 2000 (from which an estimate of 175 kt PM_{10} can be derived). There is a clear discrepancy between these two emission estimates for PM_{10}.

324. For road transport, a fairly detailed emission forecasting approach is used, adopting the latest traffic forecasts reflecting current Government policies on transport, fleet turnover and the penetration of vehicles meeting the tougher European vehicle emission directives and the impact these will have on emissions from in-service vehicles.

4.5.9.1 Assumptions for non-road transport sources

325. For most of the sectors, other than road transport, the main activity driver is forecasts in fuel consumption according to the central growth/high fuel price scenario in EP68, provided by the DTI. These estimates incorporate an assumed growth in economic activity of about 2.5% per year and the continuation of current trends towards greater use of natural gas and cleaner technologies (DTI, 2000). The industrial and service share sector projections have been based on analysis by OEF using the 1998 version of their UK industry model. The OEF model is not currently used to forecast beyond 2010. To project beyond 2010, OEF assumes that industries will have adjusted to their long-term growth paths by 2010 and that they will continue on these paths.

326. Agricultural forecasts have been provided by Defra on a sector-by-sector basis. The estimates exclude possible effects of common agricultural policy (CAP) reform on animal numbers and farm practice.

327. At the present time the Large Combustion Plant Directive is not transposed into UK law and it is still not decided how the UK is going to do this. The EP68 analysis was based on an assumption that plants without FGD will make use of the 20,000 h opt-out. For plants with FGD, no Large Combustion Plant Directive controls have been applied.

328. Other regulations that have been taken into account include the Sulphur Content of Liquid Fuels Regulations 2000 and the IPPC Directive.

329. In general, the projections are based on a number of assumptions: that measures are introduced when required by legislation and not earlier; and that all operators comply with this legislation and there is no improvement in environmental efficiency other than in response to legislation. Therefore, the projections produced tend to be conservative and are likely to over estimate the actual emissions.

4.5.9.2 Assumptions for road transport sources

330. The base emission projections for road transport are currently from a 2001 base year and are calculated from a combination of road traffic activity projections and knowledge of the expected emission characteristics of the vehicle fleet in the future. The changes in emission factors for vehicles in the projections are largely driven by the legislative emission standards set in the European Directives for Euro III and Euro IV vehicles applying to conventional petrol and diesel powered vehicles. This means that no further improvements in new vehicle emissions occur beyond the limits set for introduction in 2006 for light duty vehicles and 2008 for heavy duty vehicles. In other words, the penetration of advanced vehicle types and technologies currently under development, such as fuel cell, hybrids and other electric-powered vehicles are not considered in the base projections, neither are alternative fuels such as compressed natural gas (CNG).

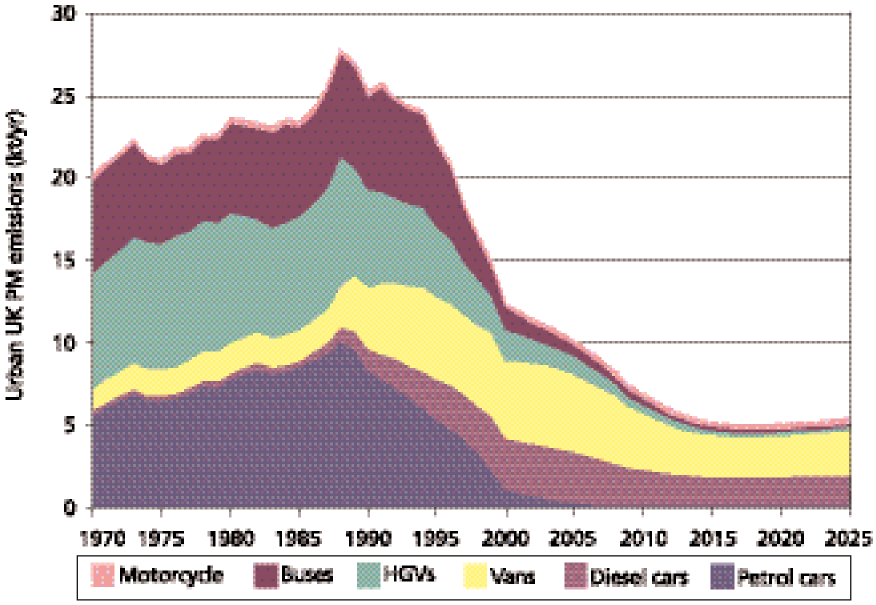
331. The projections are based on emission factors for vehicles up to and including Euro IV standards as shown in Table 4.1. The assumptions and methods for calculating the road transport emission projections were explained in detail in the AQEG report on NO_2 (AQEG, 2004). However, there have been several important changes made to some of the key assumptions since that report was published and these are described in this section.

- The projections now use the latest set of figures from DfT in 2003 on traffic growth, taking account of the impact of the Ten Year Plan for Transport. The mean of the high and low demand traffic growth scenarios is used for traffic in 2010. Projections beyond 2010 use the figures from DfT's older set of 1997 National Road Traffic Forecasts, which provided vehicle km data in 5-year intervals from 1996 to 2031. The vehicle kilometre data in the National Road Traffic Forecasts are rescaled to the Ten Year Plan figures for 2010.

- The rate of penetration of diesel cars in the fleet is important in the emission projections for PM. The 2001 base projections assumed a growth of diesel car sales from actual levels of 18% of all new car sales in 2001 to 30% by 2010.

- As described in Section 4.1.2.2, detailed account is taken of the emission durability requirements of the Directive covering Euro III and IV light duty vehicles and consequently changes in the emission degradation rate for new vehicles assumed for the 2001 NAEI projections were made. Changes to the assumptions made concerning the percentage of failed catalyst cars in the fleet were also described earlier.

Figure 4.28 Time series of PM_{10} emissions from UK urban road transport by vehicle type.



- London-specific bus fleet forecast data from TfL are incorporated into the national emission projections, taking account of measures introduced by London Transport Buses to reduce emissions from the bus fleet in London.
- The NAEI projections now take account of the introduction of sulphur-free fuels (with a sulphur content below 10 ppm) required under Directive 2003/17/EC. The Directive requires their introduction by 1 January 2009, but the NAEI projections assume their penetration into the UK market sooner than this, starting at 15% in 2005 to 100% by 2009. The sulphur-free fuel penetration rates used in the NAEI are supplied by the DfT. The effect of these sulphur-free fuels on emissions is based on studies carried out by SENCO (2000).

4.5.9.3 Baseline PM emission projections for urban UK road transport

332. Figure 4.28 and Table 4.16 demonstrate the trends in urban UK road transport emissions of PM_{10} by vehicle type from 1970 to 2025 as forecast by the 2001 NAEI.

Table 4.16 Urban UK exhaust emissions of PM_{10} (kt) from road transport projected to 2025.

	1970	1980	1990	1995	2000	2005	2010	2015	2020	2025
Petrol cars	5.71	7.83	8.39	5.39	1.16	0.31	0.15	0.13	0.14	0.14
Diesel cars	0.28	0.38	1.28	2.41	3.04	3.09	2.17	1.77	1.75	1.82
Vans	1.33	1.84	3.73	5.03	4.76	4.74	3.45	2.53	2.51	2.74
HGVs	6.96	7.85	5.92	4.28	1.82	1.07	0.51	0.27	0.22	0.23
Buses	5.69	5.47	5.67	5.03	1.36	0.70	0.37	0.23	0.18	0.17
Motorcycles	0.20	0.33	0.29	0.21	0.23	0.26	0.27	0.29	0.30	0.32
Total	**20.18**	**23.70**	**25.27**	**22.34**	**12.36**	**10.16**	**6.92**	**5.21**	**5.10**	**5.42**

333. Urban exhaust emissions have declined by 51% from 1990 to 2000 mainly as a result of the reduction in emissions from the large fleet of petrol cars due to the phasing out of leaded petrol and the introduction of cars fitted with three-way catalysts. However, the tighter standards on emissions of PM from diesel vehicles introduced in the 1990s have also contributed to this decline. The fact that emissions from diesel cars and vans appear to have reduced less rapidly than emissions from large diesel HGVs and buses is because of the faster growth in diesel car and van activity over this period somewhat offsetting the gains achieved by technology improvements.

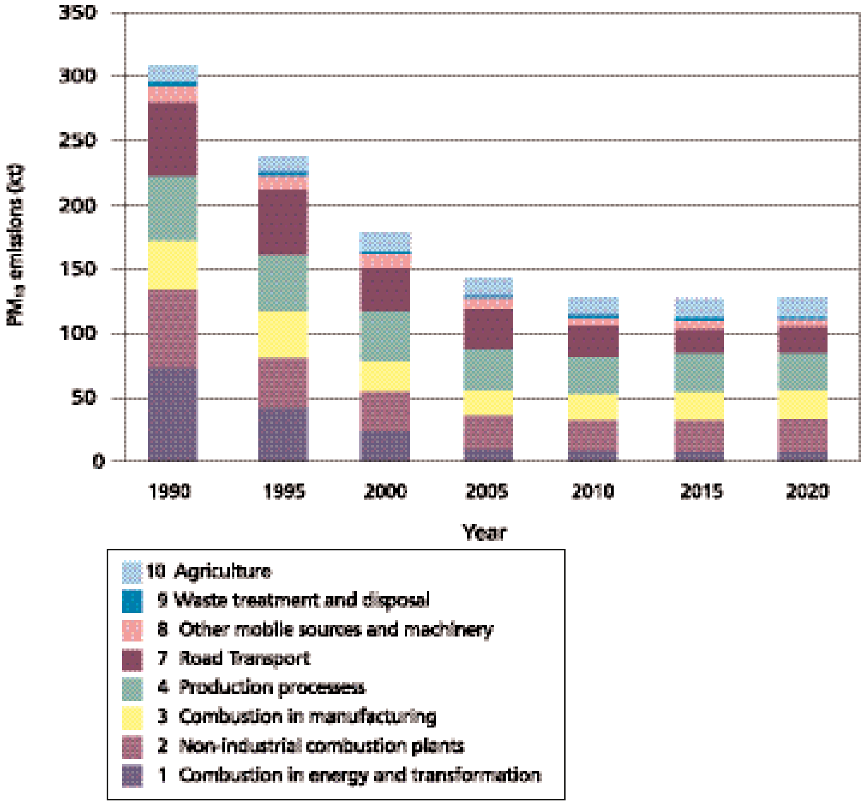
334. Emissions are expected to continue to decline until around 2018 due to the growth of lower-emitting diesel vehicles in the fleet that comply with tighter emission regulations. After this date, without any further improvements in technology beyond those necessary to meet Euro IV standards, emissions start to rise again as the continuous growth in traffic starts to offset the gains achieved by the penetration of cleaner vehicles in the fleet.

335. By 2010, urban exhaust emissions are predicted to reduce by 44% compared to 2000 levels and by 2020 by 59% compared to 2000 levels. By 2010 the largest PM emitters are expected to be diesel cars and vans due to their increased growth in activity, although this is tempered by the fact that new vehicles entering the fleet will be lower emitting. Very tight standards on PM emissions from larger diesel vehicles (HGVs and buses) ensure the reduction in emissions from these vehicles continues. The introduction of cleaner diesel fuels with lower sulphur content will also contribute to the decrease in PM emissions.

4.5.10 Baseline PM emission projections for all UK sources

336. Figure 4.29 shows the time series of PM_{10} emissions in the UK from 1990 projected to 2020 at 5-year intervals. Emissions are expected to decline by 28% between 2000 and 2010. This is primarily due to the fitting of FGD at power stations and reductions in the road transport.

Figure 4.29 Historic and projected UK PM_{10} emissions.



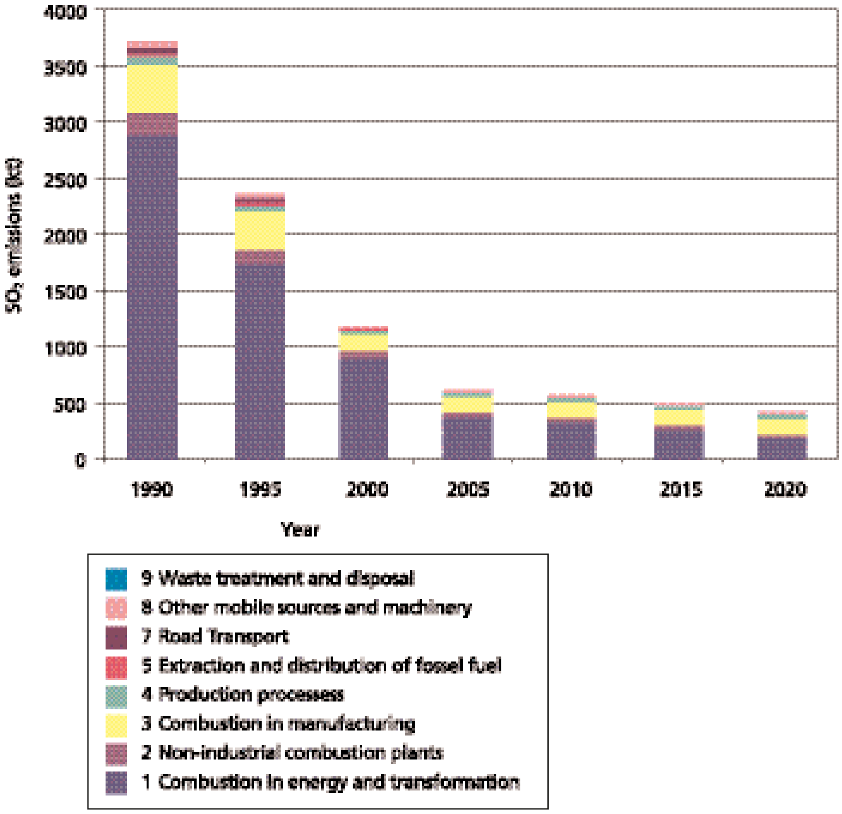
4.5.11 Baseline emission projections for PM precursor pollutants from all UK sources

4.5.11.1 SO_2

337. Figure 4.29 shows the time series of SO_2 emissions in the UK from 1990 projected to 2020 at 5-year intervals. Emissions of SO_2 are predicted to decline by approximately 52% between 2000 and 2010. This is largely due to the Sulphur Content of Liquid Fuels Regulations 2000 (1999/32/EC). For gas oil, a sulphur content of no more than 0.2% was required by 2003 and no more than 0.1% by 2008. For fuel oil, the requirement was that from 2003 the sulphur content of the oil must be less than 1%. In addition, a large decline is seen in SO_2

emissions from the power station sector. This is because a switch away from coal to lower sulphur fuels such as gas and the fitting of FGD at further coal-fired power stations are forecast.

Figure 4.30 Historic and projected UK SO_2 emissions by sector.



4.5.11.2 NO_X

338. Figure 4.31 shows the time series of NO_X emissions in the UK from 1990 projected to 2020 at 5-year intervals. UK NO_X emissions are predicted to decline by ~30% between 2000 and 2010. NO_X emissions from the road transport sector are forecasts to decline by 54% as a result of vehicles meeting tighter emission standards penetrating the UK fleet. A decline is also expected in the power station sector as coal consumption with higher NO_X emissions per joule of energy is forecast to decline, whereas natural gas consumption with lower NO_X emissions per joule of energy is expected to increase.

4.5.11.3 NMVOCs

339. Figure 4.32 shows the time series of NMVOC emissions in the UK from 1990 to 2020 at 5-year intervals. NMVOC emissions are predicted to decline by ~28% between 2000 and 2010. This is largely a result of a reduction in emissions from industrial processes due to more stringent emission controls resulting from the Solvent Emission Directive (1999/13/EC). In addition NMVOC emissions from road transport are expected to decline. This is as a result of an increasing number of vehicles meeting tighter emission standards requiring catalytic converters, fuel switching from petrol to diesel cars, lower volatility of summer petrol fuel blends and better evaporative control systems on cars leading to lower fuel evaporative losses.

Figure 4.31 Historic and projected UK NO_X emissions by sector.

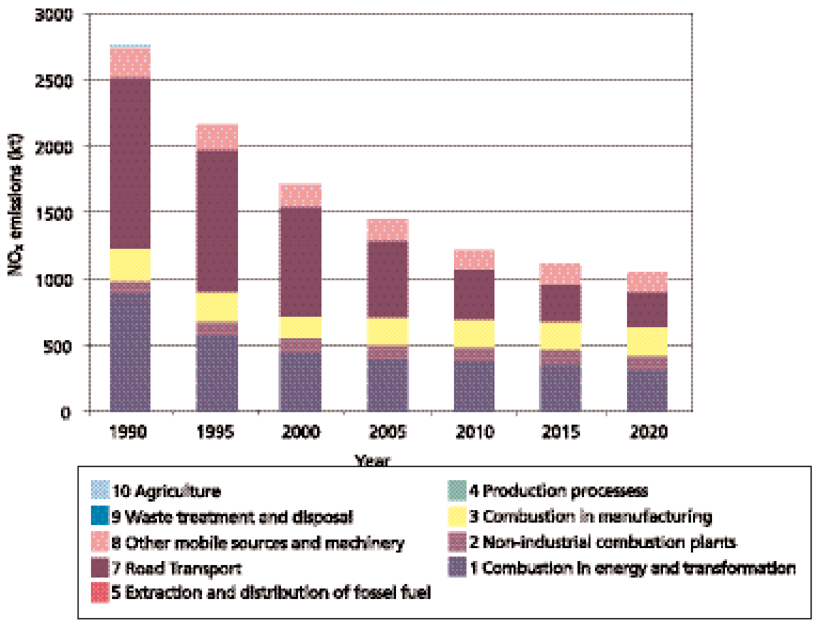
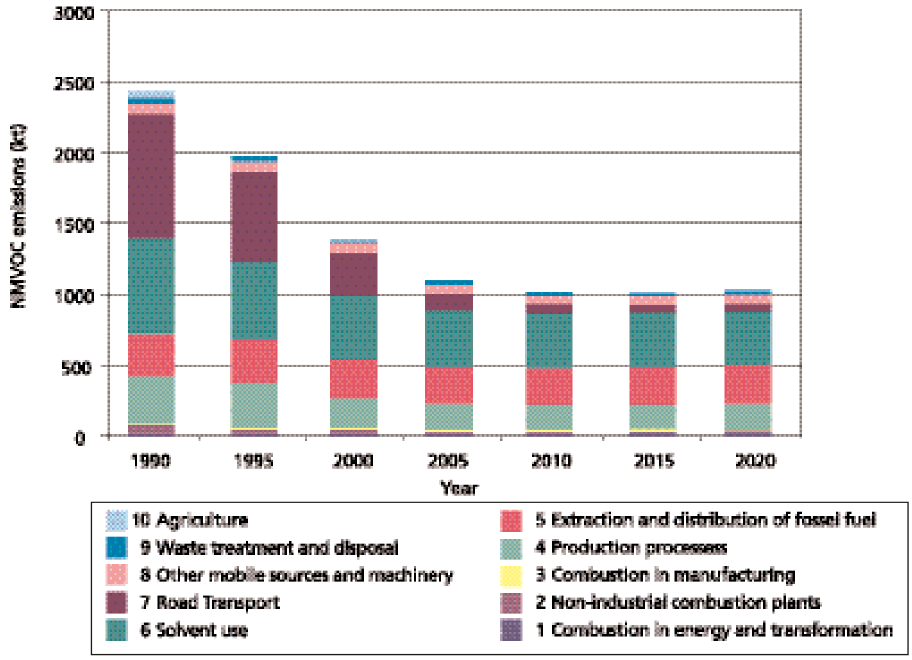


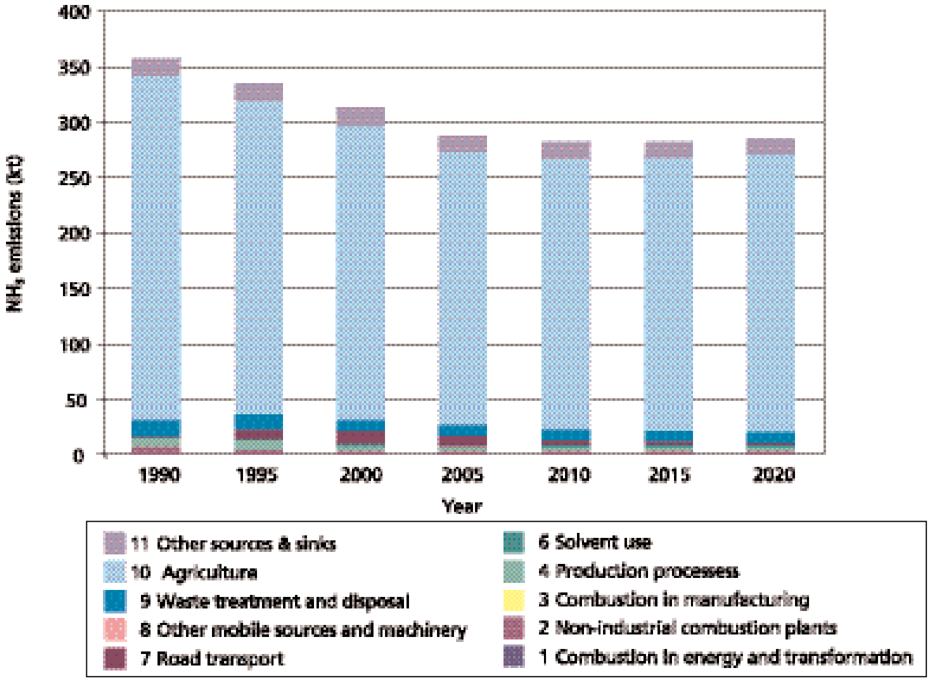
Figure 4.32 Historic and projected UK NMVOC emissions by sector.



4.5.11.4 NH_3

340. Figure 4.33 shows the time series of NH_3 emissions in the UK from 1990 to 2020. In 2000, 85% of NH_3 emissions were from agriculture. A 10% decline in total NH_3 emissions is predicted between 2000 and 2010. This is largely due to decreasing animal numbers and a decline in fertiliser use. NH_3 emissions from road transport increased between 1990 and 2000. This was caused by the increased use of catalytic converters. Post 2000 NH_3 emissions from road transport are predicted to decline due to the increasing number of diesel vehicles and improvements in technology.

Figure 4.33 Historic and projected UK NH_3 emissions by sector.



4.6 Emissions controls and abatement technologies

4.6.1 Abatement of industrial emissions of particles

341. Strategies for reducing emissions of PM from industrial sources may be subdivided into three general approaches:

- process changes;
- process management; and
- end-of-pipe arrestment.

A detailed description of abatement methods is beyond the scope of this report, but this section gives a brief overview of some of the main control strategies for industrial emissions of PM. Further information can be found in the IPPC BREF documents that can be found at http://eippcb.jrc.es/pages/FActivities.htm and in the *Environment Agency Guidance Notes*.

342. Not all of the approaches will be available for each source of particles but all three can contribute to significant reductions in national emissions of PM.

343. Process changes involve modifications to raw materials, process technology, operational parameters or products, which reduce or prevent emissions occurring from a process. Examples of process changes range from relatively simple measures such as burning 'cleaner' fuels or switching fuels used in an existing plant or modifying individual pieces of equipment such as burners (to improve combustion efficiency) through to more fundamental changes such as replacing complex plant with less polluting technology, for example, replacing a coal-fired power station with a CCGT power station.

344. Process management refers to improvements to the operation of an existing process leading to reduced emissions. Process management measures can be very ‘low-tech’, for example, ensuring that spillages of dusty materials from processes are cleaned up rapidly before they can be suspended by wind, keeping doors closed to minimise fugitive dust emissions or ensuring that process equipment is maintained in good condition. At the other end of the scale, computerised control of large combustion processes can ensure efficient combustion and potentially lower emissions of PM. Monitoring of PM abatement plant efficiency in order to prevent high emissions occurring during periods of equipment failure is also an important process management option.

345. Both process change and process management methods have been applied by UK industry and are particularly important for those sources where fugitive emissions are significant. Both strategies have made a major contribution to reducing particle emissions over the past three decades, however, the scope for further reductions via these means is probably fairly limited (except in the case of major changes in process technology). Currently, the main option for reducing emissions of particles is the use of end-of-pipe arrestment technology to recover, or occasionally destroy, dust in waste gas streams. The four most important technologies are electrostatic precipitators (ESPs), fabric filters, scrubbers and cyclones, with the first three being most important for large industrial (Part A) processes and cyclones used as the main abatement measure mostly for smaller (Part B) processes.

346. ESPs work by using a high-voltage electrical current to impart an electrical charge to PM passing through the device and then collecting the charged particles on collection plates. The collection plates are periodically rapped or vibrated to dislodge the PM into a hopper. ESPs typically have two or three stages of collection. ESPs can operate at relatively high temperatures and can treat both wet and dry waste streams. Their performance is affected by the electrical properties of the dust and collection efficiency is markedly lower for finer particles (especially <1 µm).

347. Fabric filters employ a fabric membrane that is gas permeable but traps PM. The layer of dust that builds up on the fabric also acts as a filter and increases the efficiency of collection but must be removed periodically to control the pressure drop over the filter. Fabric filters can operate to moderately high temperatures but are sensitive to moisture. They are very efficient even for fine PM. Dry or semi-dry absorbents can be added to the waste gas upstream of a fabric filter and, being collected with the PM, can remove gaseous pollutants such as SO_2 or mercury. Ceramic and other sinter materials have been developed for filtration of gases in high temperature applications.

348. Wet scrubbing involves the entrainment of dust particles in a liquid. The liquid can be water or an aqueous solution of an acid, alkali or oxidising agent where gaseous pollutants are also being abated. Various designs exist for the contact stage of scrubbers and venturi scrubbers are the most efficient at collection of PM but are less good at absorbing gaseous pollutants. Generally, therefore, the choice of scrubber design will depend upon whether the control of gaseous or particulate pollutants is more important. Wet scrubbing techniques can be used for wet and dry waste streams and are able to treat high temperature waste streams. They are less efficient at collecting fine particles. Wet FGD is an example of a wet scrubbing technology used for control of gaseous pollutants that will capture PM as well. However, because wet FGD systems cannot handle high levels of PM, a primary abatement device such as an ESP must be fitted upstream.

349. Cyclones are inertial separation devices and can achieve high collection efficiencies for larger particulate material ($>PM_{10}$). For particles smaller than 10 µm and particularly below 2.5 µm their efficiency is substantially poorer than ESPs and bag filters. They are rarely applied as final abatement on major industrial (Part A) processes but are used as a first stage abatement device. The technology is still widely used in some industries that generate coarse and/or ‘difficult’ PM (for example, wood machining processes and food processes). Although the emissions may be small in a national context, such processes may have low height discharges and other factors that may make the emissions more significant for local air pollution.

350. ESPs are the dominant technology for utility boilers and other large combustion plants and are also widely used in industrial processes such as cement and lime kilns. Fabric filters are also common for the control of industrial process emissions and are increasingly important for combustion processes as well. Fabric filters can be regarded as the likely candidate technique for most new industrial processes, although ESPs can also be considered BAT for some industrial sectors, e.g. for cement and lime kilns. The use of wet scrubbing techniques is declining. However, they are still important where waste streams containing mixed gaseous and PM emissions must be treated. Cyclones are too inefficient to be used alone for large industrial sources of PM, but are still common for smaller processes.

4.6.2 Abatement of emissions from road vehicles

351. During the 1990s, reductions in particulate emissions from diesel vehicles were mainly achieved through improvements to engine design and fuel systems. These were further aided by a simultaneous reduction in the sulphur content of diesel fuels from levels close to 2000 ppm in the early 1990s to around 50 ppm by the end of the decade.

352. However, to meet the even more stringent particulate emission standards coming into effect, diesel vehicles – especially the larger ones – will need exhaust after-treatment processes to further reduce emissions. Many vehicles will be fitted with some form of diesel particulate filter (DPF) on the exhaust, as these are extremely efficient at reducing emissions of particles, including the ultrafines.

353. There are a number of DPF designs on the market, all generally requiring some form of continuous or periodic filter regeneration capability built into them. DPFs capture particulates in the exhaust stream through a combination of surface-type and deep-bed filtration mechanisms. The most commonly used type of DPF in the UK are those that convert NO_X to NO_2 in the exhaust stream and use the NO_2 to continuously oxidise the particulates held on the filter, thus regenerating the trap. The system uses a ceramic wall-flow filter to trap particulates. An oxidation catalyst converts NO_X in the exhaust to NO_2 upstream of the filter. The efficiency of these DPFs at reducing PM emissions is typically around 90%. A potential problem is the release of a higher fraction of NO_X as NO_2 from the exhaust unless the system is optimised to reduced NO_2 slippage. The negative implications of primary NO_2 emissions from diesel exhausts were discussed in the AQEG report on

NO_2 (AQEG, 2004). This type of DPF requires use of ultra-low sulphur fuel (<50 ppm), but this fuel has been freely available in the UK since around 2001, well ahead of legislative requirements mandating its introduction by 2005. Some new diesel vehicle models are now equipped with this type of DPF. DPFs of this nature have also been developed for retrofit application, and schemes such as the Clean UP programme – run by the Energy Savings Trust – promote and offer grants towards the cost of fitting DPFs on diesel vehicles.

354. Fuel additives have also been developed to enable the regeneration of DPF systems. These are usually organometallic fuel additives, which oxidise the particulates held on the filter through a catalytic process (Cook and Richards, 2002). The most common additives use iron and cerium, but others have used sodium and strontium.

355. Diesel oxidation catalysts have been fitted on some vehicles for some time, especially HGVs and buses and other vehicles in 'captive fleets'. These help to reduce emissions of CO and hydrocarbons, but also reduce the soluble organic fraction of diesel particulates. However, they will also oxidise SO_2 present in the exhaust leading to the generation of sulphate particulates, which can lead to an increase in mass emissions of PM from vehicles running on fuels with high sulphur content. This is less of a problem with fuels of very low sulphur content, and modern oxidation catalysts are optimised to minimise CO and hydrocarbon emissions with low SO_2 conversion.

356. Without any after-treatment technology, the decrease in sulphur content of fuel alone will slightly reduce PM emissions from diesel vehicles. Directive 2003/17/EC mandates the provision of sulphur-free fuels (<10 ppm) by 2009, but the uptake of this fuel in the UK is expected to commence earlier than this date. It is estimated that running a diesel vehicle on sulphur-free fuel leads to around a 5% reduction in PM emissions relative to emissions from the same vehicle running on current ultra-low sulphur grade fuel (~50 ppm sulphur).
The introduction of ultra-low sulphur diesel itself led to a 510% reduction in PM emissions compared to emissions from vehicles running on 500 ppm sulphur diesel, typical of the grade of fuel sold in the mid to late 1990s.

357. Water-diesel emulsion fuels have also shown significant reductions in both emissions of PM and NO_X. The uptake of this fuel in the UK has been limited so far.

358. Switching vehicles from running on diesel to alternative transport fuels such as LPG and CNG led to significant reductions in PM emissions. The uptake of these fuels has not been significant in the UK, but targeted fleets have the potential to switch to CNG, with significant benefits in terms of reduction in PM emissions.

359. Among the precursor pollutants, transport only makes a major contribution to emissions of NO_X and NMVOCs. Emissions of these pollutants have been significantly reduced over the last decade as a result of the fitting of three-way catalysts on petrol vehicles. Tighter reductions on emissions of these pollutants from both petrol and diesel vehicles will lead to further reductions in the future. Measures for reducing NO_X emissions were discussed in detail elsewhere (AQEG, 2004). Emissions of NMVOCs have also been reduced by the fitting of carbon canister devices on petrol cars to curb evaporative emissions of fuel vapour from vehicles, and the reduction in the volatility of fuel blends sold during the summer months, as required by European directives. Controls at filling stations have also helped to reduce emissions during the refuelling stages.

360. Transport is not a significant source of ammonia, but emissions have been increasing with the fleet penetration of cars with catalytic converters, as ammonia is a by-product of the NO_X reduction process on the catalyst. However, better catalyst and engine management systems are expected to tackle this problem, leading to lower emissions from cars in the future (AQEG, 2004). Selective catalytic reduction using urea to control NO_X emissions from heavy duty diesel vehicles can lead to ammonia slip, but again systems can be optimised to minimise emissions (AQEG, 2004).

361. Road transport makes a very small contribution to emissions of SO_2 in the UK; emissions are being further reduced with the uptake of fuels with lower sulphur content.

4.7 Impact of policies on particle emissions

4.7.1 The impact of increased diesel car penetration

362. There has been a rapid rise in the sale of new diesel cars in the UK in recent years. Sales increased from 14% in 2000 to almost 24% by the end of 2002 and 27% by 2003. It is believed that the reason for this rapid increase is that motor manufacturers, in order to meet the Voluntary Agreement on CO_2 emissions, have been making diesel cars more attractive through reducing the previous cost penalty over commensurate petrol cars. Additionally, the performance and driveability of diesel cars has improved to the extent that the stigma attached to them by motorists in the past has been diminishing. Diesel car sales in the UK are still lower than the rest of Europe, but are expected to catch up, although still not reach parity. The current forecast of 30% diesel car sales by 2010 used in the NAEI emission projections, originally set in 2002, now looks doubtful and the DfT and Society of Motor Manufacturers and Traders (SMMT) now forecast sales in the UK will reach 42% by 2010.

363. The NAEI has modelled the sensitivity of projections for PM_{10} and NO_X emissions from UK road transport in response to increased diesel car sales relative to the original base case assumption of 30% diesel car sales by 2010. Emissions were calculated for a growth in car sales to 42% by 2010 (the DfT/SMMT estimate), 35% by 2010 and 50% by 2010. The results for road transport emissions in urban areas are shown in Table 4.17. PM emissions are more sensitive to the increase in diesel car sales than NO_X emissions. By 2015, PM emissions would be almost 10% higher for the 42% scenario than is currently forecast for that year; for the 50% scenario, PM emissions would be 15% higher than is currently forecast.

364. Table 4.18 shows how increases in diesel car penetration rates affect the predicted rate of decline in PM and NOX emissions for each year relative to 2000. It shows that whereas a growth in diesel car sales to 30% in 2010 is associated with a 57% reduction in PM10 emissions compared to 2000 levels by 2015, that reduction becomes 53% if diesel car sales reach 42% by 2010.

Table 4.17 Urban UK exhaust emissions in kt from road transport for different diesel car penetration rates. Base case assumes 30% sales by 2010.

		2000	2005	2010	2015	2020	2025
PM	Original base case	12.08	9.95	6.82	5.19	5.09	5.41
	35% diesel by 2010	12.08	9.98	6.94	5.40	5.36	5.70
	42% diesel by 2010	12.08	10.01	7.08	5.70	5.73	6.10
	50% diesel by 2010	12.08	10.02	7.17	5.96	6.14	6.55
NO_X	Original base case	274.1	196.6	129.5	95.5	89.4	91.5
	35% diesel by 2010	274.1	197.3	130.3	96.5	90.5	92.7
	42% diesel by 2010	274.1	197.9	131.4	97.8	92.0	94.2
	50% diesel by 2010	274.1	198.1	132.0	99.0	93.7	96.1

Table 4.18 Percentage change in urban UK exhaust emissions from road transport relative to emissions in 2000 for different diesel car penetration rates. Base case assumes 30% sales by 2010.

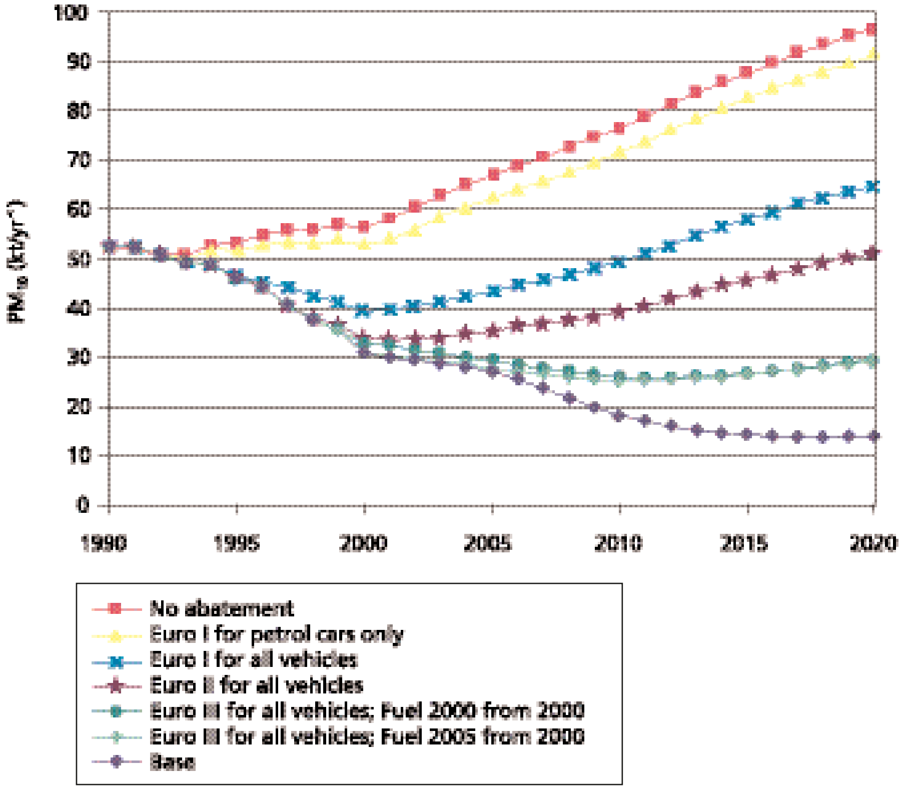
		2000	2005	2010	2015	2020	2025
PM	Original base case	0%	18%	44%	57%	58%	55%
	35% diesel by 2010	0%	17%	43%	55%	56%	53%
	42% diesel by 2010	0%	17%	41%	53%	53%	50%
	50% diesel by 2010	0%	17%	41%	51%	49%	46%
NO_X	Original base case	0%	28%	53%	65%	67%	67%
	35% diesel by 2010	0%	28%	52%	65%	67%	66%
	42% diesel by 2010	0%	28%	52%	64%	66%	66%
	50% diesel by 2010	0%	28%	52%	64%	66%	65%

4.7.2 An evaluation of policies and regulations affecting emissions from the road transport sector in the UK

365 Netcen carried out a study to evaluate the impact of air quality policy over a period of major change – the last decade – to help provide information on which policies have been successful and which have not. The study helped to improve our knowledge of the quality of modelled outputs and showed how the response to legislation can sometimes be different to that which was originally anticipated. The study focused on the road transport sector, in particular the Euro standards on vehicles and fuel standards and how they affected emissions and air quality.

366 The analysis considered the emission benefits of individual policies and combined policies compared to a scenario of no policies, the 'no abatement' scenario. The no abatement scenario assesses what would have happened in the absence of the policies (that have since been implemented). It is based on the conditions in 1990, but takes account of the economic/activity growth over the period. It should be pointed out that the study involved projections from an older version of the inventory (2000 NAEI), with slightly less up-to-date assumptions than those generally discussed in this report. The absolute emission reductions for PM_{10} and NO_X are shown in Figures 4.34 and 4.35. In general, the greatest single benefit has been achieved by the Euro I emission standard. However, the pattern of behaviour is different with regard to the effectiveness of the Euro standards on these two pollutants.

Figure 4.34 Effect of Euro emission standards on trends in UK PM_{10} emissions from road transport.

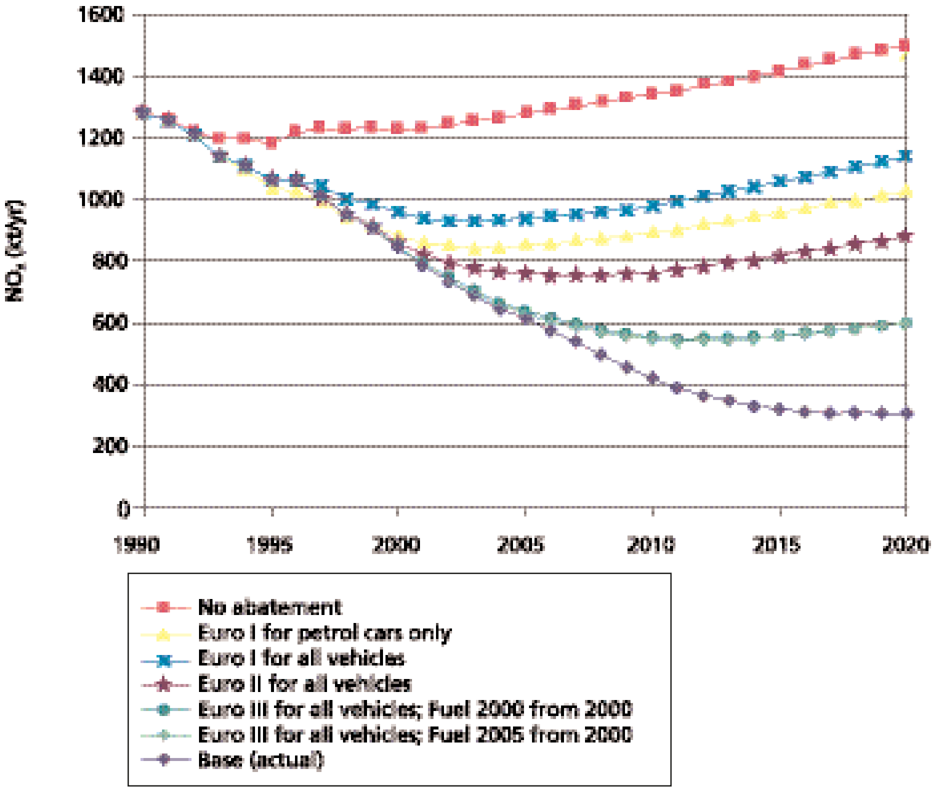


367. For NO_X, a large proportion of the improvements seen are from the introduction of Euro I technology to petrol vehicles (around a 30% improvement compared to 'no abatement'). However, the NO_X emission factors for Euro I heavy duty vehicles were higher than for pre-Euro I vehicles (this issue was discussed elsewhere (AQEG, 2004)). Consequently, the Euro I emission standards appear to be detrimental for NO_X emissions from diesel vehicles. This spreads the benefits of NO_X reductions more evenly over Euro I to IV standards. The analysis here shows that the approximate (cumulative) benefits of successive technical standards are 30% reduction for Euro I, 40% for Euro II, 60% for Euro III and 70–80% for Euro IV, relative to the 'no abatement' scenario.

368. A different pattern emerges for PM_{10}. The effect of introducing Euro I technology to petrol vehicles was small (7%). The largest benefits arose from the introduction of Euro I technology to diesel vehicles (30%), but there were also progressive improvements through the Euro standards: compared to the 'no abatement' scenario, the cumulative reductions were 50% for Euro II, 65–70% for Euro III and 75–85% for Euro IV.

369. These conclusions are important because when all emissions are considered, it is the Euro I standard (for petrol cars) that seems to have provided the dominant benefit. However, when the focus is switched to the two primary pollutants of most concern for local air quality management, NO_2 and PM_{10}, the benefits are attributable to all the Euro standards applied to all vehicles.

Figure 4.35 Effect of Euro emission standards on trends in UK NO_X emissions from road transport.



370. This pattern is slightly different to that anticipated at the start of the study. A previous evaluation (undertaken in 2001 with an older set of emission factors) showed a much greater overall benefit attributed to Euro I in reducing NO_X emissions and lower benefits from later Euro standards. The new emission factors, therefore, show more benefits attributable to later Euro standards. This mitigates against the expected effect of diminishing returns, that is, as emission standards are tightened, it becomes progressively more difficult to get large emission reductions at reasonable cost. It should be stressed, however, that the Euro III and Euro IV factors are not based on measurement data (whereas values for Euro I and II are) and that there are, therefore, some uncertainties in these conclusions and the predicted outcomes.

4.7.3 Low emission zones

371. A feasibility study that considered the effects of a low emission zone (LEZ) in London has recently been carried out; its aim was to quantify the benefits of reducing road vehicle emissions of NO_X and PM_{10} (Watkiss *et al*., 2003). The study highlighted the difficulty in determining which vehicles would be affected by the scheme for several reasons; these included the reliability of emission factors, the costs of enforcement of the scheme and the impacts on the operators of vehicle fleets. Nevertheless, three recommended schemes were identified. It was stated that by 2006/7 all lorries buses and coaches that are Euro II (that is, vehicles built before late 2001) or older would need to fit abatement equipment (for example, a particle filter) in order to enter the zone, but Euro III or Euro IV vehicles would be allowed to enter. In 2010, two schemes emerged as the best options. The first would require all heavy vehicles that are Euro III (that is, those built before late 2006) or older to fit pollution control equipment in order to enter the zone but would allow Euro IV vehicles to enter. The second would in addition restrict vans and taxis more than 10 years old from entering the zone. The study recommended the scheme should be enforced within the Greater London boundary.

372. It was assumed that vehicles fitted with pollution control equipment used continuously regenerating traps (CRTs). It was further assumed that these traps reduced total PM_{10} emissions by 95%. For 2007 it was calculated that total road transport emissions of PM_{10} would be reduced by 9.0% compared with baseline 2007 emissions. For 2010 it was estimated the LEZ would reduce PM_{10} emissions by 19.0% – by 23% if vans and taxis were included, – compared with baseline 2010 emissions.

4.7.4 The London Congestion Charging Scheme (CCS)

373. The London CCS started in February 2003. The CCS aims to reduce road traffic congestion in central London by charging each vehicle £5 to enter the zone on weekdays during 7.00 am – 6.30 pm (excluding bank holidays). The CCS zone covers approximately 22 km^2 of central London (<1.5% of the area covered by the Greater London boundary). Certain important vehicle types, such as buses and taxis, are exempt from the charge, as are vehicles using alternative fuels such as gaseous fuels and electricity. An initial assessment of the impact of the scheme has been undertaken to determine the effect on emissions and the concentrations of NO_X, NO_2, PM_{10}, CO_2 and fuel use (Barratt *et al.*, 2004); the following summary is based on that assessment. Overall, the scheme has been at the high-end of TfL's expectations in terms of reducing traffic congestion (TfL, 2003).

374. Table 4.19 summarises the changes in emissions brought about by the introduction of the CCS by vehicle type (Beevers and Carslaw, 2005). These emissions were calculated using the currently available emission factors described in Section 4.2.2.4 and London-specific vehicle type and age profiles, which are also used in the LAEI. Use was made of detailed vehicle flow and speed data that were collected as part of the CCS monitoring programme. Changes in the total emissions of PM_{10} depend on the vehicle type considered and vary between a +5.0% increase for motorcycles and a –22.4% for rigid HGVs.

Table 4.19 Effect of congestion charging on calculated annual emissions of PM_{10} (% change).

	Motor-cycles	Taxis	Cars	Buses and coaches	LGV	Rigid HGVs	Artic HGVs	Total
CCS zone	5.0	3.3	–36.1	–13.4	–14.8	–22.4	–20.8	–11.9
Inner ring road	6.0	–4.6	–2.3	–0.7	4.0	–6.7	–6.0	–1.4

375. One important outcome from the analysis is the effect of vehicle speed. Because vehicle speeds in central London are low, any increase in vehicle speed caused by reduced congestion can have a large effect on calculated emissions. For example, taxi flows increased by 13% in the CCS zone but total emissions of PM_{10} only increased by 3.3%. The small increase is a result of the increased vehicle speed on roads in the CCS zone, which offsets the increased flows of both taxis and buses. Overall, the effect of increased vehicle speed has reduced PM_{10} emissions in the zone by around a factor of two compared with the reduction of vehicle flows. Figure 4.36 shows the percentile speed distribution of hourly vehicle speeds on major roads before and after the introduction of the CCS and highlights that the most significant changes in speed have occurred in the lower speed range below 20 km h^{-1}. It should also be stressed that the analysis might underestimate the benefit of the CCS in terms of the total reduction in PM_{10} emissions because no account has been taken of the potential reduction in tyre and brake wear and resuspended PM_{10} resulting from a reduction in vehicle flows (particularly of HGVs). As is discussed in Section 4.2.2.12, resuspended material from the road surface (and tyre and brake wear) can be important sources of PM_{10}.

Figure 4.36 Percentile vehicle speed distribution for hourly speed data on major roads before and after the introduction of the CCS.