

3.1 INTRODUCTION

Primary particulate matter comprises those particles emitted directly to the atmosphere from natural and anthropogenic sources. These sources are extremely diverse and the chapter will address firstly the inventory approach to estimating emissions and provide the latest information on UK and European emissions of particulate matter, and will consider projections to future years. Recent work on the validation of emissions inventories will be described.

Road traffic is a major contributor to primary particulate matter emissions and recent measurement data for PM_{10} and $PM_{2.5}$, and particularly that pertaining to traffic-influenced sites are analysed. Biological particles are a source not normally included in inventories, and a brief section reviews the major sources of biological particles and attempts a crude estimate of their contribution to atmospheric particle mass. Finally, two major studies of the chemistry of airborne particles, based in Leeds and South Wales, are reviewed and used to provide insights into the sources and nature of primary particulate matter. Such studies inevitably include secondary components. Further aspects of the Leeds study are considered in Chapter 4.

Emission inventories serve as a guide to atmospheric emissions but not necessarily to urban air quality. The distinction between emissions and urban air quality is important where the different sources have different heights of emissions or have special operational characteristics producing diurnal or seasonal variations. These factors are crucial for airborne particulate matter, with the result that the contributions from the different source categories to the primary emissions in current inventories do not reflect their respective contributions to urban air quality.

3.2 EMISSION INVENTORY

3.2.1 INTRODUCTION

This section describes the sources of primary particulate matter in the UK and quantifies their

annual emission rates. The current inventories of emissions on both national and urban scales refer only to particle mass in the PM_{10} size range as insufficient information has been available on the size distribution of particulate matter emissions from all the source sectors to allow a complete inventory time-series to be compiled for smaller size ranges. However, new emissions data for a number of sources have allowed the UK PM_{10} inventory to be improved and data have recently emerged on the mass fraction of particles below $2.5 \mu\text{m}$, $1 \mu\text{m}$ and $0.1 \mu\text{m}$ within the PM_{10} size range which has allowed inventories for UK emissions of $PM_{2.5}$, PM_1 and $PM_{0.1}$ to be estimated for the first time.

Particulate matter is emitted from a variety of sources besides fuel combustion which increases the difficulty and uncertainty in quantifying their emissions. Emissions from stationary combustion and road transport are certainly major sources, but there are also significant emissions from industrial processes, construction and quarrying and fugitive sources through mechanical break-up, abrasion and erosion (e.g. vehicle tyre and brake wear). These sources are particularly difficult to quantify and vary both spatially and temporally, particularly construction.

Whenever possible, an inventory uses measured emission rates from a particular source. Some measurements have been made for certain industrial processes, but since it is impossible to measure emissions from every source at all times, it is necessary to estimate the bulk of emissions using source-specific emission factors and activity statistics. For combustion sources, emission factors are usually fuel-related and are combined with the quantity of fuel used in the combustion process. However, for road transport, emission factors are traffic-related and are expressed in terms of mass of pollutant emitted per kilometre travelled; for construction, emission factors are expressed in terms of the area of floor space of construction and for quarrying on the amount of aggregate produced.

The main primary sources of PM₁₀ in the United Kingdom are:

Road transport. Emissions occur from all types of vehicles, not only from the exhaust, but also from tyre and brake wear. Exhaust emissions are considerably higher from diesel-engined vehicles than from those with petrol engines. Nationally, road transport contributed around 25% of PM₁₀ emissions in 1996. However, the contribution can be much higher in urban areas. For example, an urban inventory for London, where there are no significant emissions from industrial processes or stationary combustion, estimated that traffic contributes 77% of emissions. The traffic contribution has been estimated to be as low as 12% in certain areas such as Merseyside dominated by industrial sources.

Stationary combustion. Domestic coal burning has traditionally been the major source of airborne particles, but its decline has reduced the contribution to PM₁₀ emissions to around 14% nationally in 1996. However, it is still an important source in small towns and villages where domestic coal is still used and in Northern Ireland where natural gas is still not widely available. Emissions from power generation have also been declining due to the gradual switch from coal to gas for electricity generation and due to improvements in the performance of electrostatic precipitators at coal-fired stations. Public power generation is estimated to have been responsible for 16% of national PM₁₀ emissions in 1996.

Industrial processes. These include a range of different industrial processes leading to the formation and release of dust as well as construction, quarrying and mining activities. Nationally, it is estimated that these processes accounted for 28% of PM₁₀ emissions in the UK in 1996. However, their contribution can be much greater in some urban areas with large amounts of industrial

activity, for example in Merseyside where the PM₁₀ inventory shows an industrial contribution of 64%.

This section describes in more detail the primary sources of PM₁₀ and the methods used to quantify their emissions up to 1996. Emphasis will be given to the improvements and revisions made to the methodology used for the national inventory since the publication of the Third Report of the Quality of Urban Air Review Group in 1996 (QUARG, 1996). The time-series in emissions for the different sectors will be given for the period 1970 to 1996 from the UK's National Atmospheric Emissions Inventory (NAEI) (Salway *et al.*, 1997).

The mass fraction of PM₁₀ emitted in finer particle size ranges (PM_{2.5}, PM₁ and PM_{0.1}) are shown in Tables 3.13 - 3.15 for each emission source and fuel type, drawing on size distribution data from the USEPA and the Dutch TNO inventory, where these are considered applicable to sources in the UK. The mass fractions were used to derive UK emission inventories for PM_{2.5}, PM₁ and PM_{0.1} based on the reported inventory for PM₁₀. In general, combustion sources emit a higher proportion of the finer particles than mechanical sources and road transport makes a larger contribution to emissions as particle size decreases. For example, it accounted for 60% of UK PM_{0.1} emissions in 1996.

Until recently, emission inventories of particulate matter had been expressed only in terms of black smoke, defined by the simple monitoring method that had been used for many years as a measure of the blackening effect of airborne particles. Since the blackening effect of smoke depends on the fuel source, the relationship between black smoke and PM₁₀ mass emissions is not so clear as it was when coal combustion was the dominant source. Black smoke emissions are no longer measured and with so many sources making similar contributions to particulate emissions with different blackening effects, a black smoke inventory is now difficult to interpret.

The main uncertainties in the PM₁₀ emission estimates will be discussed and sources not included in the inventory will be highlighted, whether these are missing due to lack of data on emission factors for conditions pertinent to the UK or because of the lack of appropriate activity statistics for the source. In particular, resuspended road dust and emissions from agricultural sources such as ploughing, harvesting and livestock farming are identified as key areas not included in the inventory because of lack of UK source specific measurements of emission factors. Further measurements are also required of emissions from different mining and quarrying processes in the UK.

The most recent national and urban projections of emissions of PM₁₀ from road transport will be shown, based on the 1997 National Road Traffic Forecasts and the latest understanding of the impact of new fuel standards and European regulations on emissions from new vehicles post-2000.

Source apportionment of emissions and receptor modelling studies are facilitated by spatially disaggregating primary emissions of PM₁₀ and other primary pollutants involved in secondary particle formation. The UK emission inventory for PM₁₀ in 1996 has been spatially mapped on a 1 x 1 km grid for all the major sources. The results of the mapping study are shown in this Chapter. Since publication of the Third QUARG Report, urban emission inventories have been completed for nine major urban and industrial areas in the UK; the PM₁₀ inventories will be presented in this Chapter. These inventories, based on a “bottom up” approach, are more detailed than the national inventory and show significant variations in the relative emissions of PM₁₀ from the different sectors.

This section also discusses a primary particulate matter inventory which has recently been compiled for Europe by the TNO Institute of Environmental Sciences, Energy Research and Process Innovation in the Netherlands for the Dutch National Institute of Public Health and Environment (RIVM) (TNO, 1997). This inventory covers not only PM₁₀, but also PM_{2.5} and very fine particles PM_{0.1} for 26 European

countries. The figures for the UK in the TNO inventory are different to the UK's own national inventory because the former uses less detailed activity data and activity-based emission factors. Nevertheless, the data are still useful for comparing the sectorial breakdown of particle emissions for the different countries. The inventories have been spatially mapped on a 0.5 x 1° resolution.

3.2.2 PRIMARY EMISSION SOURCES OF PARTICULATE MATTER

This section describes the sources of primary PM₁₀ emissions in the UK and gives an indication of the mass fraction of each source emitted in the finer size ranges. The finer size ranges considered are those less than 2.5 µm (PM_{2.5}), less than 1 µm (PM₁) and the ultra-fine particle size less than 0.1 µm (PM_{0.1}). These size fractions were based on data from USEPA (1995) and TNO (1997) and have been applied to the emission inventory compiled for PM₁₀ to derive UK emission inventories for PM_{2.5}, PM₁ and PM_{0.1}.

3.2.2.1 Road Transport

Exhaust emissions

Road transport is a major source of PM₁₀ in urban areas. Many of the detailed characteristics of PM₁₀ emissions from motor vehicles were described in the Third QUARG Report. Both petrol and diesel vehicles must conform to specific emission limits for the pollutants NO_x, CO and hydrocarbons but only diesel vehicles must comply with limits for particulate matter. These limit values are laid down in EC type-approval legislation. Diesel engines emit particulate matter at a much higher rate than petrol engines. Consequently, there are considerably more emissions data available for diesel vehicles than for petrol vehicles. Nevertheless, the much greater number of petrol vehicles on the road means that their contribution to the road transport inventory for PM₁₀ is still significant.

Studies on the size distribution of particle emissions from diesel vehicle exhausts indicate that a significant

proportion of the particle mass is in the ultra-fine range and virtually all the mass is below 2.5 μm . Table 3.1 shows the mass fraction of particulates in the PM_{10} size range emitted below 2.5 μm , 1 μm and 0.1 μm diameter, taken from USEPA and TNO. For petrol vehicles, particulate mass emissions are undoubtedly lower than from diesel vehicles, but their size distribution is a little more unclear. Fewer measurements of the particle size distribution from petrol vehicle emissions have been made and most have used a size cut-off point of around 1 μm making it difficult to establish the mass fraction of PM_{10} emissions in the coarser size ranges. The general consensus is that the bulk of the particulate mass is below 2.5 μm , but there is a suggestion that a significant amount of the particulate mass emitted from non-catalyst petrol cars is in the 2.5 - 10 μm size range. This is reflected by the size fractions shown in Table 3.1 proposed for emission inventories by the USEPA (1995) and also used for the TNO European inventory. Particulate emissions from petrol cars equipped with a three-way catalysts are lower than from non-catalyst petrol cars and appear to have a finer particle size distribution.

Table 3.1 Mass fraction of PM_{10} emitted from road transport in the finer size ranges.

	<2.5 μm	<1 μm	<0.1 μm
Brake wear	0.4	0.1	0.08
Tyre wear	0.7	0.1	0.08
Petrol Exhaust – non-catalyst	0.8	0.65	0.25
– catalyst	0.9	0.85	0.5
Diesel Exhaust	0.9	0.85	0.5

USEPA (1995), TNO (1997)

Emission factors for exhaust emissions are expressed in grammes per kilometre and vary with vehicle speed. Equations relating PM_{10} emission factors to average speed have recently been formulated by Transport Research Laboratory for diesel vehicles manufactured before the European Stage I regulations came into effect in 1993/94 (Hickman, 1998); for diesel vehicles manufactured to the Euro Stage I standards emission factor - speed equations have been recommended by the Topic Report for COPERT II, the “Computer

Programme to Calculate Emissions from Road Transport” produced by the European Topic Centre on Air Emissions for the European Environment Agency (European Environment Agency, 1997). Both data sources are based on analyses of recent results from emission tests of on-road vehicles in test campaigns around Europe. The TRL and COPERT II equations are provided for different types of diesel vehicles in different bands of engine capacity, vehicle weights or axle configurations: cars, light goods vehicles (LGVs), heavy goods vehicles (HGVs) and buses and coaches.

In the UK’s National Atmospheric Emissions Inventory (NAEI) for road transport, these equations have been used to derive individual emission factors at average speeds for vehicles on urban, rural and motorway roads, weighted according to the proportion of the vehicle fleet in the different engine capacity and vehicle weight bands. The exhaust emission factors are shown in Table 3.2.

The different stages represent the different European Union Directives which have been in place up to 1997 limiting emissions of PM_{10} from new diesel vehicles, as well as emissions of other pollutants, NO_x , CO and hydrocarbons. Stage I refers to EC Directives 91/441/EEC for diesel cars, 93/59/EC for diesel LGVs and 91/542/EC for HGVs implemented in 1993/1994. Stage II refers to tighter EC Directives further limiting emissions from new vehicles implemented in 1996/1997 (94/12/EC for diesel cars).

Emission factors for pre-Stage I and Stage I diesel vehicles are based on measurements from surveys of in-service vehicles of different ages on a chassis dynamometer or engine test bed. There have been far fewer measurements of emissions from large diesel vehicles, especially buses and coaches, than from diesel cars, so the uncertainties in the emission factors for the larger vehicles must be regarded as higher than for cars. Some measurements have been taken from heavy duty vehicles while driven on the road (Hickman, 1997). Emission factors for on-road Stage II vehicles have not been measured because these vehicles have only

Table 3.2 PM₁₀ exhaust emission factors for diesel vehicles (g/km) used in the National Atmospheric Emissions Inventory, based on data from TRL, COPERT II.

		Cold Start (g/trip)	Urban	Rural Single Carriageway	Rural Dual Carriageway	Motorway
Diesel Cars	Pre-Stage I	0.171	0.185	0.155	0.149	0.202
	Euro Stage I	0.096	0.046	0.027	0.025	0.063
	Euro Stage II	0.055	0.020	0.012	0.011	0.028
Diesel LGV	Pre-Stage I	0.292	0.387	0.317	0.306	0.469
	Euro Stage I	0.169	0.123	0.086	0.085	0.165
	Euro Stage II	0.132	0.061	0.043	0.042	0.083
Rigid HGV	Pre-1988		1.831	1.129	1.033	1.033
	Pre-Stage I		0.895	0.540	0.491	0.405
	Euro Stage I		0.589	0.360	0.321	0.220
	Euro Stage II		0.310	0.189	0.169	0.116
Artic HGV	Pre-1988		1.520	0.910	0.710	0.710
	Pre-Stage I		1.296	0.798	0.722	0.542
	Euro Stage I		0.891	0.550	0.492	0.340
	Euro Stage II		0.343	0.212	0.189	0.131
Buses & Coaches	Pre-1988		1.600	1.400	1.400	0.710
	Pre-Stage I		0.848	0.488	0.434	0.308
	Euro Stage I		0.563	0.398	0.353	0.239
	Euro Stage II		0.347	0.153	0.136	0.092

recently come into service. For these vehicles, emission factors were estimated using scaling factors taken from COPERT II, based on the ratio of the Stage II/Stage I type approval limit values.

Cold start emission factors are for the additional emissions that occur when the vehicle is started with the engine cold. Cold start emission factors expressed in grammes per cold-started trip are shown in Table 3.2 for diesel cars and LGVs. Cold start emissions cannot be reasonably estimated for heavy duty vehicles at present, because there are very little experimental data available. However, with no clear trends in the experimental measurements of emissions from hot and cold engines, it appears that cold start emissions from heavy duty diesel engines are minimal (Boulter, 1996).

Data on emissions of PM₁₀ from petrol engine vehicles are very sparse. However, it has been

observed that mass emission rates from vehicles running on leaded petrol are higher than from those running on unleaded petrol, which in turn are higher than those running on unleaded petrol and fitted with a three-way catalyst (TWC) for the purpose of reducing emissions of NO_x, CO and VOCs. Table 3.3 shows estimated exhaust emission factors given by Gover *et al.* (1994) and taken from a survey of the literature by the Transport Research Laboratory. Insufficient information was available to distinguish emission rates under different driving modes and average speeds.

Table 3.3 PM₁₀ exhaust emission factors for petrol vehicles (g/km).

	Leaded	Unleaded without TWC	Unleaded with TWC
Cars	0.06	0.02	0.01
LGVs	0.08	0.04	0.02

Although on a per vehicle basis particulate emissions from petrol vehicles are low compared with diesel vehicles, the higher proportion of petrol cars on the road does merit further measurements of emission factors and size distribution of particulates emitted from on-road petrol vehicles for emission inventory and policy development purposes.

To calculate the national inventory for PM₁₀ emissions from road transport, the emission factors are combined with national traffic and fleet composition statistics. Traffic data are provided annually by the Department of the Environment, Transport and the Regions (DETR), based on their rolling census survey of the road network in Great Britain. The data provided are annual vehicle kilometres for cars, LGVs, HGVs with different axle configurations, buses and coaches and motorcycles for 11 different road classifications. Data are provided independently for Northern Ireland by the Northern Ireland Office. The composition of the vehicle fleet is based on vehicle licensing data and new vehicle registrations. This information defines the age distribution of the fleet and the percentage of diesel cars and LGVs.

The calculations have also taken account of the introduction of low sulphur diesel in 1996 which has

been estimated to reduce particulate emissions from existing HGVs and buses by 13% and to reduce emissions from light duty diesel vehicles by 2.4%.

Combining this information produces the UK inventory for PM₁₀ exhaust emissions from road transport, broken down by type of vehicle from 1970 to 1996 and shown in Table 3.4 and Figure 3.1. Figure 3.1 also shows emission projections based on the most recent National Road Traffic Forecasts and other assumptions that are discussed in Section 3.5. Table 3.5 and Figure 3.2 show the corresponding emissions data for UK urban roads based on traffic data for roads in built-up areas.

The figures show the dominance of diesel vehicles in emissions of PM₁₀. They also show how emissions were steadily rising from 1970 until 1991 due to the growth in traffic, but have been declining since 1992. This decline has been mainly due to the penetration into the fleet of diesel vehicles meeting the tighter emission standards, offsetting the growth in traffic which has continued over this period. Emissions from petrol vehicles have also been declining due mainly to the penetration of cars fitted with three-way catalysts in the fleet and the decline in the consumption of leaded petrol.

Table 3.4 UK exhaust emissions of PM₁₀ from road transport (ktonnes).

		1970	1975	1980	1985	1990	1991	1992	1993	1994	1995	1996
Cars	Petrol	9.3	11.1	13.1	15.3	15.3	14.5	13.5	12.4	11.5	10.6	9.8
	DERV	0.4	0.5	0.6	0.7	2.1	2.4	2.9	3.0	3.2	3.4	3.4
	All Cars	9.7	11.6	13.7	16.0	17.4	16.9	16.4	15.4	14.7	14.0	13.2
LGV	Petrol	1.3	1.4	1.5	1.6	1.7	1.5	1.3	1.1	1.0	0.9	0.7
	DERV	1.4	1.8	2.2	2.4	4.9	5.7	6.2	6.8	7.4	7.4	7.4
	All LGV	2.7	3.2	3.6	4.0	6.5	7.2	7.5	7.9	8.4	8.2	8.1
HGV	Artic	3.4	3.9	5.2	5.9	8.2	8.1	7.7	7.9	7.9	7.5	7.0
HGV	Rigid	22.3	23.7	24.1	23.0	23.7	23.1	21.6	20.0	18.9	16.8	14.6
ALL HGV		25.7	27.6	29.3	28.8	31.9	31.2	29.3	27.9	26.8	24.3	21.5
Buses		5.4	4.9	5.4	5.5	6.5	6.5	5.9	5.7	5.6	5.0	4.6
Motorcycles		0.4	0.5	0.7	0.7	0.6	0.5	0.4	0.4	0.4	0.4	0.4
All DERV		33.0	34.9	37.4	37.3	45.3	45.8	44.4	43.4	42.9	40.1	36.8
All Petrol		10.9	12.9	15.3	17.5	17.5	16.5	15.2	13.9	12.9	11.8	10.9
All Vehicles		43.9	47.8	52.6	54.9	62.8	62.3	59.5	57.3	55.8	51.9	47.7

Table 3.5 Urban UK exhaust emissions of PM₁₀ from road transport (ktonnes).

		1970	1975	1980	1985	1990	1991	1992	1993	1994	1995	1996
Cars	Petrol	4.8	5.4	6.5	7.1	6.9	6.5	6.0	5.6	5.3	4.9	4.5
	DERV	0.2	0.3	0.3	0.4	1.1	1.3	1.5	1.6	1.7	1.8	1.8
	All Cars	5.0	5.7	6.9	7.4	8.0	7.8	7.5	7.2	7.0	6.7	6.3
LGV	Petrol	0.6	0.7	0.7	0.7	0.7	0.7	0.6	0.5	0.5	0.4	0.3
	DERV	0.8	1.0	1.2	1.2	2.4	2.9	3.0	3.4	3.6	3.6	3.6
	All LGV	1.4	1.7	1.9	2.0	3.1	3.5	3.6	3.9	4.1	4.0	3.9
HGV	Artic	1.1	1.2	1.3	1.2	1.7	1.6	1.4	1.5	1.4	1.4	1.3
HGV	Rigid	10.3	11.3	11.5	10.0	10.3	10.1	9.5	8.8	8.5	7.5	6.7
ALL HGV		11.4	12.5	12.8	11.3	11.9	11.7	11.0	10.2	9.9	8.9	8.0
Buses		3.8	3.2	3.6	3.6	3.9	4.3	4.0	4.0	4.0	3.6	3.3
Motorcycles		0.4	0.5	0.7	0.7	0.6	0.5	0.4	0.4	0.4	0.4	0.4
All DERV		16.2	17.0	18.0	16.5	19.3	20.1	19.5	19.1	19.2	17.9	16.7
All Petrol		5.8	6.6	8.0	8.5	8.2	7.7	7.0	6.5	6.1	5.6	5.1
All Vehicles		21.9	23.5	25.9	24.9	27.5	27.8	26.5	25.6	25.3	23.5	21.8

Tyre wear

The USEPA gives an emission factor of 0.0012 g/km for PM₁₀ emissions from tyre wear on cars. Scaling up the emissions from HGVs and buses according to the number of wheels and combining with the traffic data gives an annual emission rate of 0.6 ktonnes for 1996 (Table 3.12). This compares with 47.7 ktonnes emitted from vehicle exhausts in 1996. Data from the USEPA suggest that particles from tyre wear are mainly of a coarser size range than exhaust emissions, as shown in Table 3.1.

Brake linings

The USEPA gives an emission factor of 0.00795 g/km for PM₁₀ emissions from wear of brake linings on cars, with 98% of particles in the <10 µm range. Again scaling up the emissions from HGVs and buses according to the number of wheels and combining with the traffic data gives an annual emission rate of 3.9 ktonnes for 1996 (Table 3.12).

Road dust

Resuspension of road dust can make a significant contribution to the local PM₁₀ concentration, as

shown by the roadside measurements at the London Marylebone Road site (Section 3.5). Resuspended particles are of a coarser size range than exhaust emissions.

Emission inventories cannot at present include this source due to the absence of emission factors for dust resuspension by traffic on roads and environmental conditions relevant to the UK. The USEPA have provided a formula for calculating emissions from this source, but this is based on old measurements made near exceptionally dusty roads, for example those running through an open desert-like environment and off dried mud-laden tracks. The emission factors and methodology are now viewed by the USEPA themselves as probably overestimating the extent of dust resuspension from more typical road conditions. For UK road conditions, the USEPA formulae are considered particularly inappropriate.

Clearly, further measurements of dust resuspension on UK-typical roads are required to provide data for emission inventory application. Until then, resuspension of road dust remains a major area of uncertainty in the UK particulate emission inventory.

Figure 3.1 UK road transport emissions of PM₁₀.

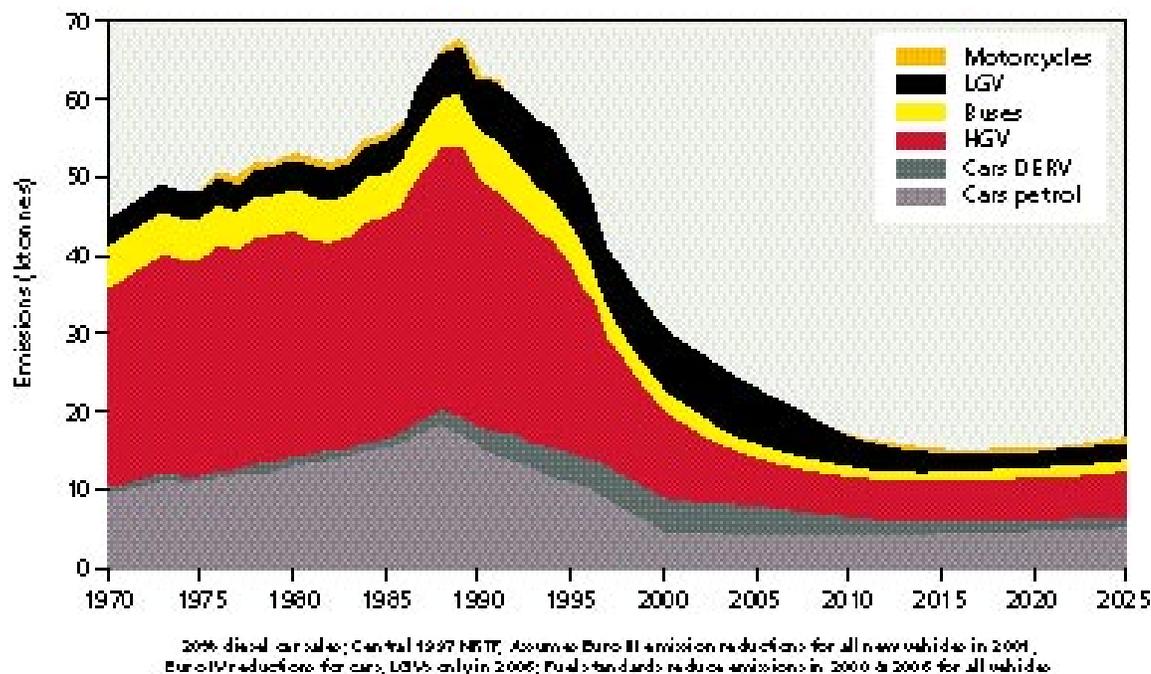
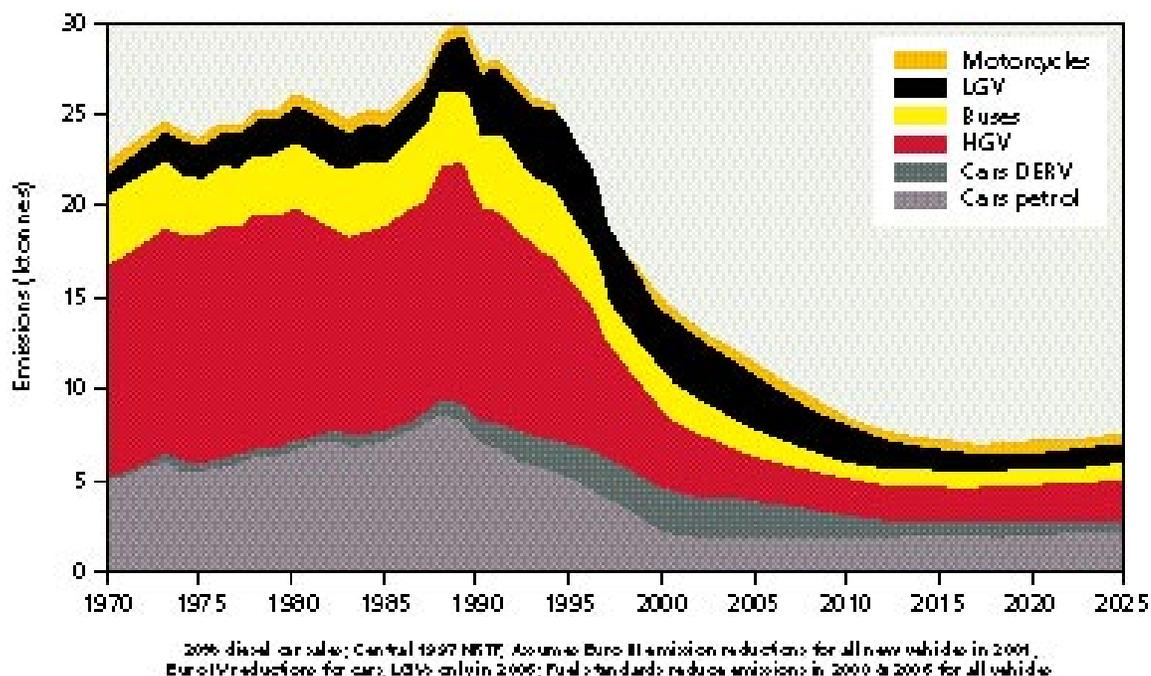


Figure 3.2 Urban UK road transport emissions of PM₁₀.



3.2.2.2 Other Transport and Off-Road Sources

Emissions from these sources have been estimated from fuel-related emission factors. Annual emissions of PM₁₀ from railways, shipping and aircraft are estimated to be around 2 ktonnes in 1996. Emissions from off-road sources include a variety of domestic, commercial, agricultural and forestry machinery such as lawn mowers, construction plant and cement mixers. Annual emissions from off-road sources are estimated to be around 2 ktonnes in 1996.

The size distribution of particulate emissions from these sources depends on the type of fuel used, a feature which applies to all other combustion sources. Table 3.6 shows the mass fraction of particulates in the PM₁₀ size range emitted below 2.5 µm, 1 µm and 0.1 µm diameter, taken from the USEPA and TNO. These are values typical for usage of these fuels by various machinery and in other transport sectors.

Table 3.6 Typical mass fraction of PM₁₀ emitted from other off-road transport and machinery in the finer size ranges.

	<2.5 µm	< 1 µm	<0.1 µm
Coke & SSF	0.26	0.04	0.02
Coal	0.38	0.32	0.08
Fuel oil	0.37	0.23	0.15
Gas oil	0.77	0.69	0.15
Petrol	0.94	0.85	0.25
Diesel	0.94	0.85	0.5

USEPA (1995), TNO (1997)

3.2.2.3 Stationary Combustion

PM₁₀ emissions from stationary combustion sources are calculated from fuel-based emission factors. Since the inventory presented in the Third QUARG Report was prepared, emission factors have been revised for coal and oil-fired power stations and for combustion of gas, coke and solid smokeless fuel (SSF) based on new USEPA work on particulate emissions and particle size distribution described in "Compilation of Air Pollutant Emission Factors, AP-42, Vol. 1 5th edition" (USEPA, 1995).

The size distribution of particulate emissions from stationary combustion processes depends on the type of fuel used as well as the type of combustion process. Particulate emissions from combustion of solid fuels are generally of a coarser size range than from combustion of petroleum fuels which in turn are coarser than particulates from gas combustion. Gas combustion produces less particulate mass emissions than combustion of solid and liquid fuels and the particulates produced are also entirely in the ultra-fine particle size range (<1 µm).

Public power generation

Power stations are the largest users of coal in the UK, but all coal-fired power stations are equipped with electrostatic precipitators (ESP) which remove around 99.0 to 99.8% of total particulate matter. Estimates of emissions from coal and oil-fired power stations are based on reported emissions of "dust" from coal and oil use in power stations and a USEPA estimate that 67% of this is in the PM₁₀ size range. Emissions from the combustion of Municipal Solid Waste (MSW) are also included. Overall, PM₁₀ emissions from public power generation have been declining due to the decreasing use of coal in favour of natural gas. Emissions of PM₁₀ in 1996 are estimated to be around 35 ktonnes (Table 3.12).

Table 3.7 shows the mass fraction of particulates in the PM₁₀ size range emitted below 2.5 µm, 1 µm and 0.1 µm diameter for different fuels, taken from USEPA and TNO. These refer to particulate emissions after any control device has been fitted.

Table 3.7 Mass fraction of PM₁₀ emitted from public power generation in the finer size ranges for different fuels.

	<2.5 µm	< 1 µm	<0.1 µm
Coke & SSF	0.26	0.04	0.02
Coal	0.44	0.19	0.08
Orimulsion	0.37	0.23	0.06
Municipal Solid Waste	0.82	0.73	0.15
Gas oil	0.43	0.18	0.15
Fuel oil	0.73	0.56	0.1
Gas	1	1	0.5

USEPA (1995), TNO (1997)

Industrial combustion

Particulate emissions from industrial combustion plant include those from coke production, petroleum refining, combustion in iron and steel and cement and lime industries including those for electricity generation by industries for their own consumption and other industrial combustion processes. Overall emissions have been declining due to the switch from coal to oil and now to natural gas as the fuel. Emissions of PM₁₀ in 1996 are estimated to be around 20 ktonnes (Table 3.12).

Particulate size fractions typical of many industrial combustion processes are shown in Table 3.8 for different fuels.

Table 3.8 Mass fraction of PM₁₀ emitted from industrial combustion processes in the finer size ranges for different fuels.

	<2.5 µm	< 1 µm	<0.1 µm
Coke & SSF	0.26	0.04	0.02
Coal	0.71	0.1	0.08
Gas oil	0.25	0.17	0.15
Fuel oil	0.65	0.42	0.15
Gas	1	1	0.5

USEPA (1995), TNO (1997)

Small combustion processes

Emissions from small combustion processes include those from domestic, commercial, public and agricultural combustion sources. The latter exclude emissions from field burning.

Domestic coal burning in open fires was nationally the major source of particulate matter in the 1950's and 1960's, but since the introduction of smoke control areas, smokeless solid fuels have replaced coal in domestic open fires. This and the switch to gas and electricity for domestic heating over much of the UK has reduced PM₁₀ emissions from the domestic sector from 216 ktonnes in 1970 to 30 ktonnes in 1996, accounting for around 14% of total emissions (Table 3.12).

Particulate size fractions typical of many small combustion processes are shown in Table 3.9 for different fuels.

Table 3.9 Mass fraction of PM₁₀ emitted from small combustion processes in the finer size ranges for different fuels.

	<2.5 µm	< 1 µm	<0.1 µm
Coke & SSF	0.26	0.04	0.02
Coal	0.38	0.32	0.02
Fuel oil	0.37	0.23	0.15
Burning oil	0.76	0.67	0.1
Gas oil	0.94	0.85	0.1
Gas	1	1	0.5

USEPA (1995), TNO (1997)

Emissions from domestic combustion may still be significant in certain small towns and villages where solid fuels are still widely used and in Northern Ireland where natural gas is still not widely available. CRE Group Ltd have recently carried out a study on behalf of Belfast City Council to measure PM₁₀ emission factors for different types of domestic solid fuel-fired appliances burning a range of different solid fuels (CRE, 1997). PM₁₀ was sampled in the flue gas of two different types of appliances burning nine different types of solid fuels. The results for two different domestic coals were found to be very similar to the emission factor used in the NAEI (10.4 g PM₁₀/kg fuel); for the smokeless solid fuels investigated, the emission factors were somewhat lower than the NAEI emission factors. However, the CRE study showed a great deal of variability in the emission factors even for the same type of appliance burning the same type of fuel. A closed type of appliance appeared to produce slightly higher emission factors than the open fire appliance, but given the variability in the data, the differences are not significant. Results from the CRE study suggest around 90% of the particle mass from the domestic appliances is in the PM₁₀ size range.

3.2.2.4 Production Processes

There are a wide variety of industrial processes which emit particulate matter from a range of dust

generating activities. Further emissions arise from construction and from mining and quarrying activities. These processes tend to emit particulates in the coarser particle size range (2.5 - 10 µm) compared with combustion sources.

Industrial processes

Using average emission factors and national production statistics, the National Atmospheric Emissions Inventory currently estimates that around 32 ktonnes of PM₁₀ were emitted from industrial processes in 1996 (Table 3.12).

The Environment Agency has compiled a Chemical Release Inventory (CRI) listing many processes that emit particulate matter. Figures for the 1995 CRI are shown in Table 3.10. Total emissions in the CRI for 1995 amount to 16 ktonnes compared with 40

ktonnes for the national PM₁₀ inventory covering the same sectors in 1995. However, these figures are not incompatible considering that the CRI:

- a) only includes emissions from a proportion of sites covered under Part A of the Environmental Protection Act (1990) and not from Part B processes;
- b) only includes emissions from sites which have been authorised;
- c) only covers England and Wales, and;
- d) relates to emissions of total suspended particulate matter, not PM₁₀.

Construction

The amount of dust emitted from a construction site depends on the size of the site and the amount of construction activity. It was explained in the

Table 3.10 Emissions of particulate matter in the 1995 Chemical Release Inventory.

Process	1995 CRI emissions (tonnes)
Carbonisation and associated processes	400
Gasification and associated processes	32
Petroleum processes	1575
Iron and steel	4751
Non-ferrous metals	49
Cement and lime manufacture	4515
Ceramic production	621
Glass manufacture and production	18
Other mineral fibres	586
Asbestos processes	9
Di-isocyanate processes	0.08
Paper and pulp manufacturing processes	0.5
Processes involving uranium	0.0004
Timber processes	0.003
Acid processes	218
Chemical fertiliser production	436
Inorganic chemical processes	234
Manufacture and use of organic chemicals	530
Pesticide production	0.01
Petrochemical processes	22
Processes involving halogens	80
Incineration	1912
Production of fuel from waste	8
Recovery processes	5
TOTAL	16000

Third QUARG Report that based on an American study and an assumption that 20% of total suspended particulate matter from construction activity is in the PM₁₀ size range, an emission factor of 0.538 tonnes/ha/month is derived for a drier climate like the US. The emissions are expected to be lower for the wetter UK climate, by around 50% based on the average number of days with rain each year in the UK. Using this climate-adjusted emission factor for the UK with statistics on the area of floor space of construction in the UK leads to an estimated mean emission rate of around 4 ktonnes in 1996, with an uncertainty range of 0.5 to 8 ktonnes (Table 3.12).

A time-series of emissions from construction has been estimated using the value of construction in the UK (at 1990 prices) as a surrogate statistic for trends in annual construction activity.

Although making a small contribution to the annual UK inventory, by its very nature, construction can dominate emissions locally over a period of time.

Table 3.11 shows the mass fraction of particulates from construction in the PM₁₀ size range emitted below 2.5 µm, 1 µm and 0.1 µm diameter. The data are taken from the USEPA and TNO. Most of the particulate mass emitted as PM₁₀ is in the coarse size range (>2.5 µm).

Table 3.11 Mass fraction of PM₁₀ emitted from construction, mining and quarrying processes in the finer size ranges.

	<2.5 µm	<1 µm	<0.1 µm
Mining and quarrying	0.29	0.08	0
Construction	0.31	0.11	0

USEPA (1995), TNO (1997)

3.2.2.5 Mining and Quarrying

Emissions from mining and quarrying are estimated using an emission factor of 0.1 g PM₁₀ per kilogramme of natural aggregate consumed,

taken from the USEPA Factor Information and Retrieval (FIRE) system (USEPA, 1995). This is an average emission factor covering a range of different mining and quarrying processes and its relevance to such processes in the UK can be questioned. Emissions from some procedures will have been reduced by implementing improved abatement measures (e.g. dry bag filters), but other procedures cannot be controlled so readily, for example dust emissions from movement of machinery on mining and quarrying sites. Clearly, further measurements of particulate emissions from mining and quarrying processes in the UK are required in a form suitable for emission inventory applications.

The USEPA emission factor has been combined with statistics on the consumption of natural aggregate (crushed rock, sand and gravel) taken from the UK Minerals Yearbook, published by the British Geological Survey (British Geological Survey, 1997) to produce the UK PM₁₀ inventory for this source. Emissions in 1996 are estimated to be around 24 ktonnes (Table 3.12).

Table 3.11 shows the mass fraction of particulates from mining and quarrying in the PM₁₀ size range emitted below 2.5 µm, 1 µm and 0.1 µm diameter. The data are taken from the USEPA and TNO. Most of the particulate mass emitted as PM₁₀ is in the coarse size range (>2.5 µm).

3.2.2.6 Waste Treatment and Disposal

Emissions from non-landfill waste treatment and disposal have been estimated from USEPA emission factors for incineration (USEPA, 1995). Emissions of PM₁₀ were estimated to be 6 ktonnes in 1996 and have been declining rapidly in recent years due to the closure of old incineration plants from 1995 and more stringent emission limits for new plants.

Data from the USEPA and TNO suggest that 82% of PM₁₀ emissions from waste incineration are in the <2.5 µm size range, 73% are in the <1 µm size range and 15% are in the <0.1 µm size range.

3.2.2.7 *Agriculture*

Emissions of particulate matter from agricultural activities are likely to come from a variety of sources, such as land preparation, fertiliser application and harvesting. These will include emissions of pollens. PM₁₀ emission factors are not currently available for agricultural activities typical of the UK, other than from fuel combustion by agricultural machinery, so PM₁₀ emissions from agriculture in the UK cannot yet be quantified. Stubble burning is now, except for a few limited exceptions, banned in England and Wales and is not thought to be significant in Scotland and Northern Ireland.

It is expected that harvesting is the main agricultural source of particulate matter as it is usually carried out when the crops are dry and so having a greater tendency for emitting particles. Clearly, emissions from harvesting will be very seasonal and will have an impact mainly in rural areas. TNO have estimated PM₁₀ emissions from agriculture and suggested significant emissions, including those of finer particles (PM_{2.5}), arise from livestock, particularly pig and poultry farming (TNO, 1997). However, all the particulate emission factors were measured for farming practices in the Netherlands where such agriculture is very intensive. These Dutch-specific emission factors are not expected to be appropriate for UK agriculture, but there is need for verification of this. Although agriculture may not make a significant contribution to the UK national inventory, emissions may be high locally and at certain times of the year. Ambient measurements of PM₁₀ and finer particulates in rural areas, near areas of harvesting, will help to verify this, but clearly further measurements of particulate emissions from different agricultural activities in the UK is required to provide data for emission inventory applications.

3.2.3 *TIME SERIES OF NATIONAL EMISSION ESTIMATES*

3.2.3.1 *PM₁₀*

The time series for UK emissions of PM₁₀ from all the sources are shown in Table 3.12, grouped into

the aforementioned sectors for which emissions can be quantified. These data are taken from the National Atmospheric Emissions Inventory which have been updated since the publication of the Third QUARG Report using more detailed emission factors and activity data. It can be seen how PM₁₀ emissions have declined significantly since 1970, mainly due to the decline in the use of coal for domestic and industrial combustion processes.

Emissions from electricity generation have also been declining since 1992 despite a 42% growth in electricity generated between 1970 and 1996. This is due to the move away from coal to natural gas and nuclear power for electricity generation and to improvements in the performance of electrostatic precipitators at coal-fired power stations.

The one sector which has shown significant growth in emissions since 1970 is road transport, its contribution to total UK emissions rising from 9% in 1970 to 25% in 1996. In urban areas with little industrial activity, where public power and industrial processes do not make a significant contribution, the contribution of road transport to emissions will be even higher; for example, as much as 77% in London (Table 3.21a). The main source of road transport emissions is exhaust from diesel engine vehicles. Emissions from diesel vehicles have been growing due to the growth in heavy duty vehicle traffic and the move towards more diesel cars, currently at around 10% of the fleet. Since around 1992, however, emissions from diesel vehicles have been decreasing due to the penetration of new diesel vehicles meeting tighter PM₁₀ emission regulations.

Among the non-combustion and non-transport sources, the major emissions are from a range of industrial processes and mining and quarrying whose emission rates have remained fairly constant.

Figure 3.3 shows trends in emissions taken from the NAEI broken down into the six main sectors: public power; industrial combustion, which includes all industrial combustion processes; small combustion (domestic and commercial); non-combustion

Figure 3.3 UK emissions of PM₁₀ : 1970-1996.

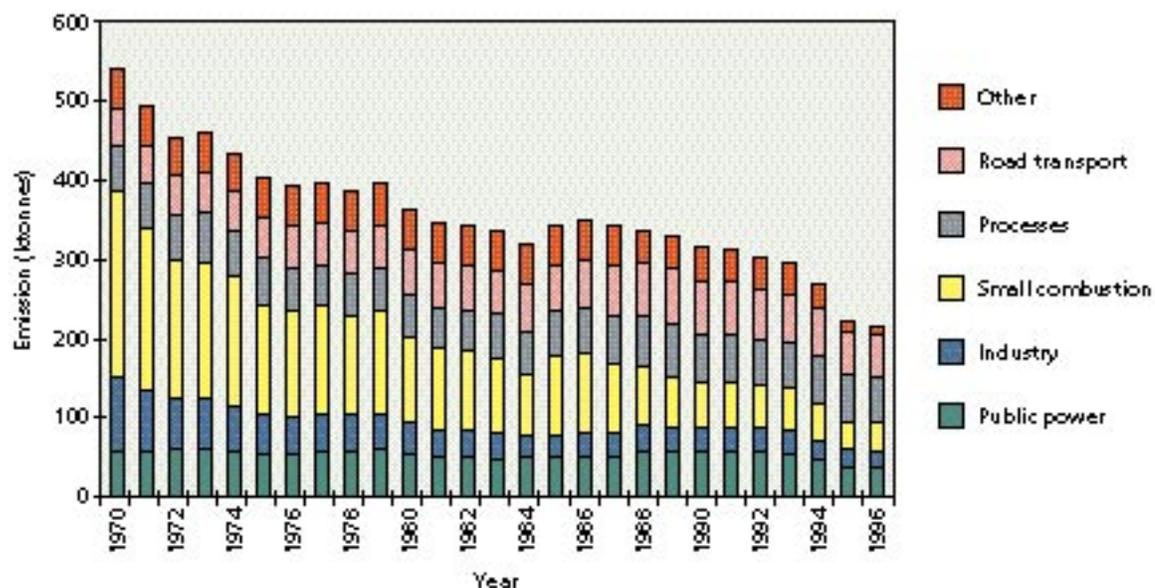
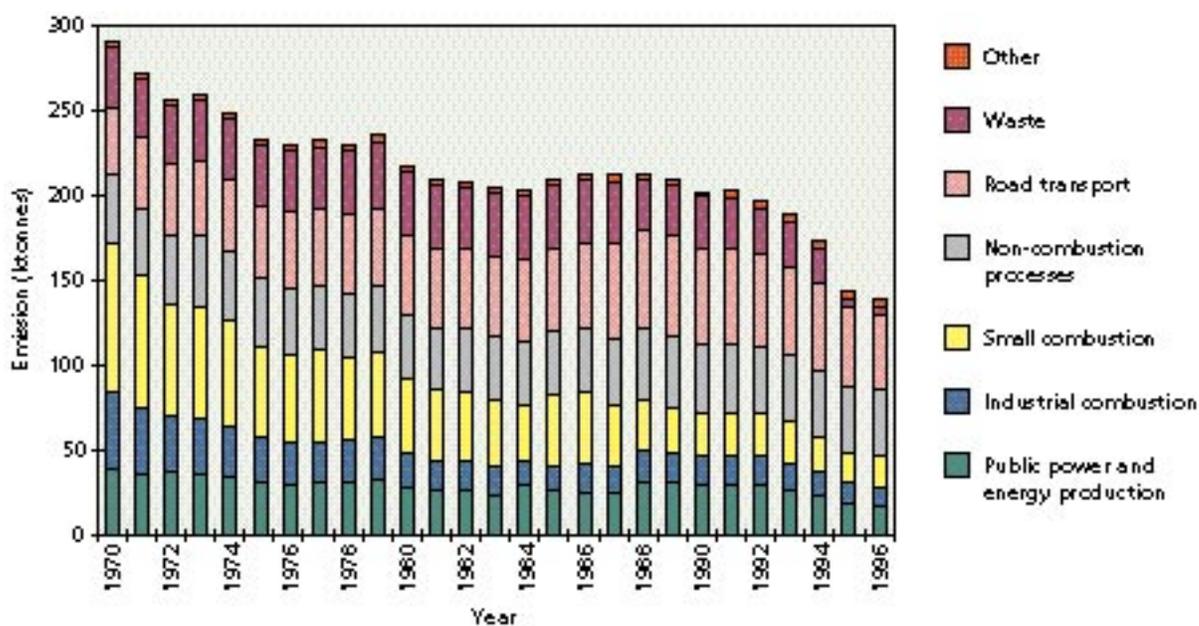


Figure 3.4 UK emissions of PM_{2.5} : 1970-1996.



processes in industry; road transport; and other sources which includes other transport and machinery and waste treatment.

3.2.3.2 PM_{2.5}, PM₁ and PM_{0.1}

Inventories for PM_{2.5}, PM₁ and PM_{0.1} have been estimated from the PM₁₀ inventory and the mass fractions in these size ranges available for different

emission sources and fuel types. A total of 33 different size distributions covering PM_{2.5} and PM₁ emissions from different source sectors were taken from the USEPA as being applicable to sources in the UK; a lesser number of sectors with size fractions in the PM_{0.1} range were available from the TNO study. Typical mass fractions of PM₁₀ emissions in these size ranges for different emission sources and fuel types were summarised in Tables 3.1, 3.6-3.9 and

Table 3.12 UK emissions of PM₁₀ by sector (ktonnes).

Source	1970	1975	1980	1985	1990	1991	1992	1993	1994	1995	1996	1996 as %
Combustion in energy production & transformation	78	61	60	53	60	59	59	57	49	40	38	18%
Public Power	58	54	54	49	57	56	55	53	46	36	35	16%
Other industrial	20	7	6	4	3	3	3	3	3	3	3	2%
Commercial, institutional & residential combustion	235	139	109	99	55	59	53	54	44	34	36	17%
Domestic	216	127	98	89	48	51	46	47	38	28	30	14%
Other	19	11	11	10	8	8	7	7	7	6	6	3%
Industrial combustion	74	43	33	25	26	26	27	25	23	20	17	8%
Iron & steel	9	5	2	1	1	1	1	1	1	1	1	1%
Other	65	38	30	24	24	25	26	24	22	19	16	8%
Non-combustion processes	57	58	54	56	63	60	58	59	61	59	59	28%
Industrial processes	33	32	31	32	32	32	31	31	32	32	32	15%
Mining & quarrying	21	23	20	22	28	25	23	24	26	24	24	11%
Construction	3	3	3	3	4	4	4	4	4	4	4	2%
Road transport	46	50	55	58	67	66	64	61	60	56	52	25%
Petrol exhaust	11	13	15	18	18	16	15	14	13	12	11	5%
Diesel exhaust	33	35	37	37	45	46	44	43	43	40	37	17%
Tyre wear	0	0	0	0	1	1	1	1	1	1	1	0%
Brake wear	2	2	2	3	4	4	4	4	4	4	4	2%
Other transport & machinery	5	5	5	5	5	5	5	4	4	4	4	2%
Waste treatment & disposal	44	44	46	45	38	37	34	33	26	7	6	3%
TOTAL	538	400	362	341	314	313	299	293	267	220	213	100%

3.11. In general, combustion processes emit a higher proportion of fine particles (<2.5 µm) than mechanical sources such as quarrying and construction. Gaseous fuels also tend to emit finer particles than petroleum and solid fuels.

Each of the detailed source sectors for which a PM₁₀ emission is estimated (a total of 139 individual sectors and sub-sectors) were allocated an appropriate size distribution and used to calculate emission inventories for PM_{2.5}, PM₁ and PM_{0.1}. The results are shown in Tables 3.13-3.15 in the same format as for the PM₁₀ inventory.

Figures 3.4-3.6 show trends in emissions of each particle size by source sector. The results show a general decline in emissions of each particle size since 1970, but with a slow down in the rate of decline as the particle size decreases. Between 1970 and 1996, UK emissions of PM₁₀ fell by 60%,

whereas emissions of PM_{2.5} are estimated to have fallen by 55%, PM₁ by 51% and PM_{0.1} by 34%. Also, there is a gradual change in the relative source contribution with particle size. This is illustrated more clearly in Figure 3.7 which shows the percentage contribution of each sector to PM₁₀, PM_{2.5}, PM₁ and PM_{0.1} emissions in 1970 and 1996. Road transport becomes an increasingly important sector as the particle size decreases. In 1996, it accounted for 25% of PM₁₀ emissions, but 60% of PM_{0.1} emissions. Emissions from non-combustion sources show the opposite trend.

The increased contribution of traffic to emissions of the finer particles also explains why the finer particles have shown a smaller fall in UK emissions between 1970 and 1996. It should be noted that for PM_{0.1} a large proportion of the fall occurred in 1995 due to controls and reductions in emissions from Municipal Solid Waste incinerators.

Figure 3.5 UK emissions of PM₁ : 1970-1996.

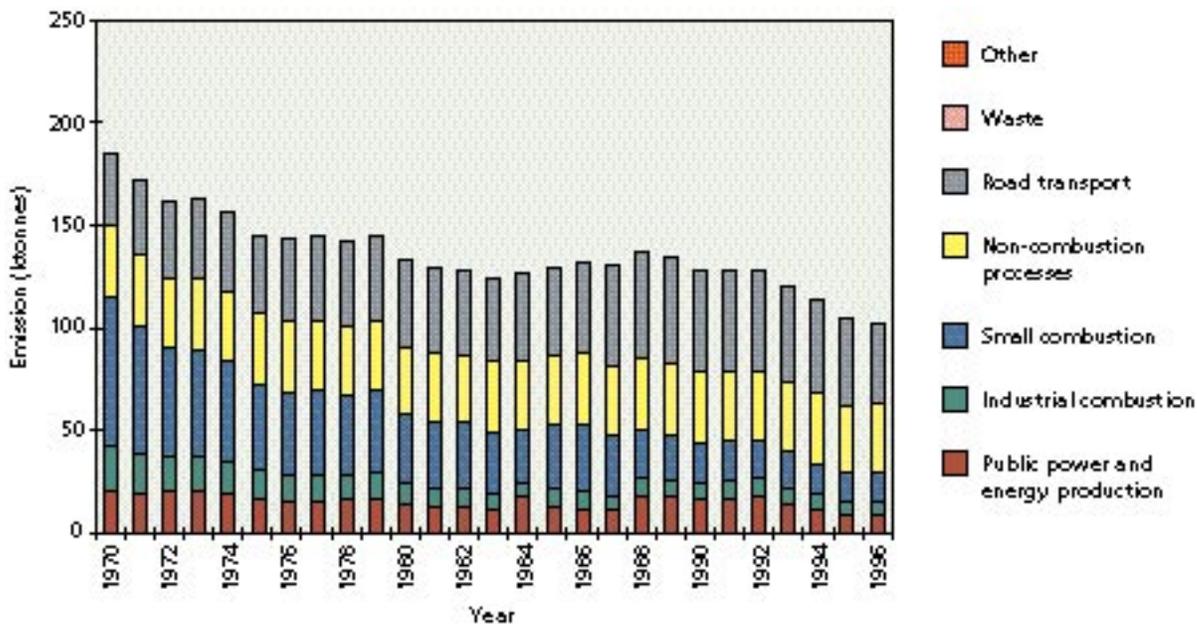
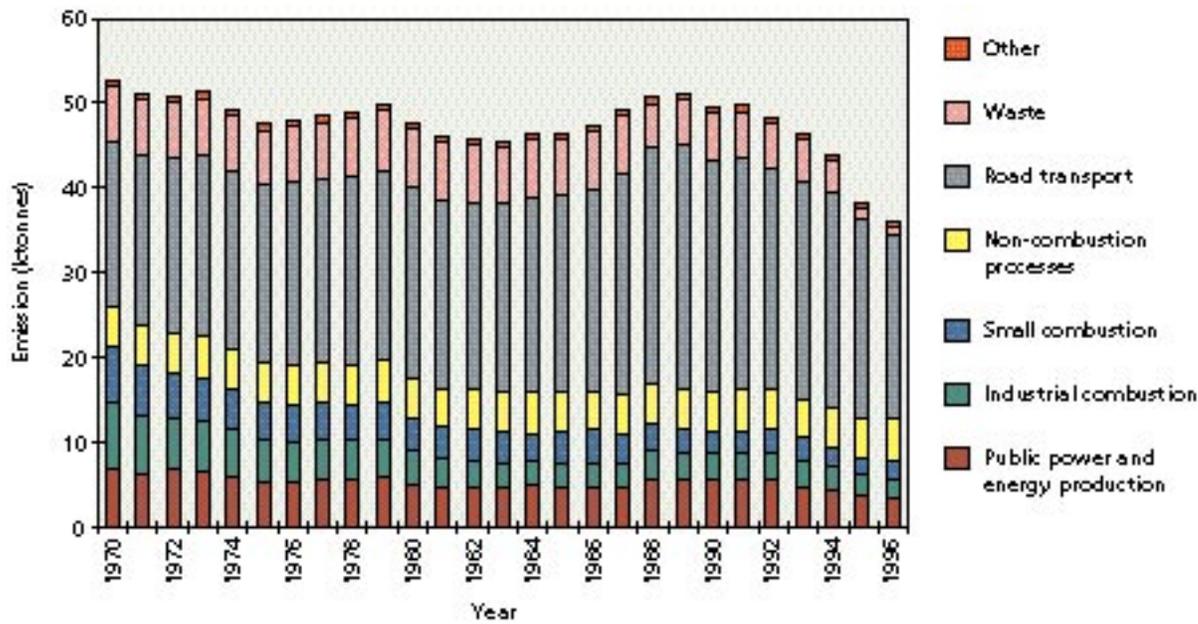


Figure 3.6 UK emissions of PM_{0.1} : 1970-1996.



Supplemented by falls in emissions from industrial combustion processes, these reductions in national emissions may have little effect on residential and commercial areas.

Clearly the approach adopted for estimating emissions of the smaller particle sizes, while it is currently the only one available, includes a number

of assumptions and uncertainties. The approach depends on the PM₁₀ emission rates estimated for each sector which themselves have great uncertainties. The inventories for the smaller particles will be even more uncertain as there are additional uncertainties in the size fractions and their applicability to individual emission source sectors. As pointed out, the relevance of US and

Dutch size fraction data to UK emission sources can be questioned. The figures should give a good indication of the finer particle sources and the relative magnitude of emissions, but there is clearly a need for more measurements of emission rates and particle size distributions of UK sources.

3.2.3.3 Black smoke

Until recently, emission inventories of particulate matter had been expressed only in terms of black smoke, defined by the simple monitoring method that had been used for many years as a measure of the blackening effect of airborne particles. The British Standard definition for black smoke (BS 1747), which has been used traditionally in the UK to assess the concentration of smoke particles, is based on a non-gravimetric reflectance method in which air is sampled through a filter and the resulting blackening measured.

Black smoke was calibrated for domestic coal smoke and when emissions come from similar sources, i.e. coal burning, as they had up to the 1960's, the degree of blackening should be proportional to mass concentrations. However, since smoke from different combustion sources has a different blackening effect depending on the fuel type, there is now no simple relationship between

black smoke and particle mass emissions. In particular, the increased contribution to smoke emissions from road transport makes this relationship unclear. Mass for mass, it has been estimated that diesel emissions have a blackening effect three to four times greater than coal emissions. Expressing black smoke emissions as the mass of smoke from uncontrolled coal combustion that would produce the same blackening effect on a filter, 1g of particle emissions from diesel combustion produces the equivalent of 3-4g of black smoke.

The NAEI is required to estimate black smoke emissions from all fuel combustion sources using fuel-based emission factors. However, the emission factors were measured more than 20 years ago when solid fuel combustion was the dominant source of particulate matter and black smoke emissions. Black smoke emissions are no longer measured and with other sources making a contribution to particulate emissions with different blackening effects and with different measures introduced to control their emissions, the black smoke inventory is now difficult to interpret. In particular, it is unclear how to quantify the effect of emission controls which have been introduced in recent years to reduce particulate emissions when the black smoke emission factors are based on such old measurements.

Figure 3.7 Percentage contribution of sectors to UK particulate emissions.

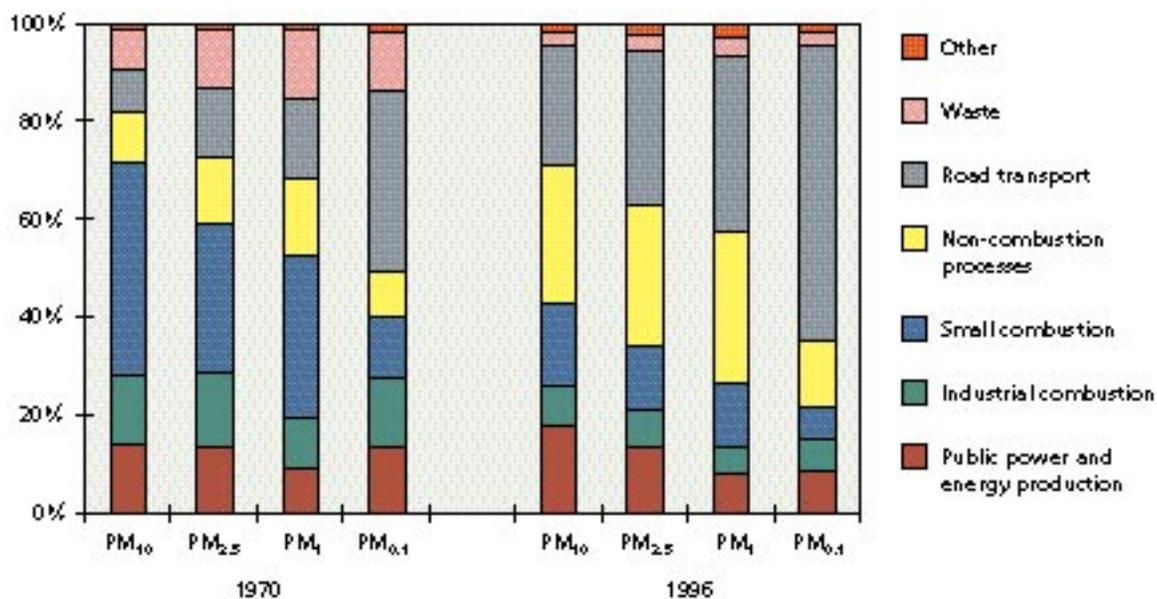


Table 3.13 UK emissions of PM_{2.5} by sector (ktonnes) estimated for the mass fraction of particles below 2.5 µm in each sector in the PM₁₀ inventory.

Source	1970	1975	1980	1985	1990	1991	1992	1993	1994	1995	1996	1996
Combustion in energy production & transformation	38	32	29	27	32	32	32	29	25	20	20	14%
Commercial, institutional & residential combustion	88	53	43	40	24	26	24	24	20	16	18	13%
Industrial combustion	49	28	21	16	16	17	17	15	14	13	10	8%
Non-combustion processes	38	38	37	38	40	39	38	38	39	39	39	28%
Industrial processes	31	31	30	30	30	30	30	30	30	30	30	22%
Mining & quarrying	6	7	6	6	8	7	7	7	8	7	7	5%
Construction	1	1	1	1	1	1	1	1	1	1	1	1%
Road transport	39	43	47	49	57	56	54	52	50	47	43	31%
Petrol exhaust	9	10	12	14	14	13	12	11	10	9	8	5%
Diesel exhaust	30	31	34	34	41	41	40	39	39	36	33	24%
Tyre wear	0	0	0	0	0	0	0	0	0	0	0	0%
Brake wear	1	1	1	1	1	1	1	1	1	2	2	1%
Other transport & machinery	5	5	4	4	4	4	4	4	3	3	3	2%
Waste treatment & disposal	36	36	39	38	32	31	28	27	22	6	5	4%
TOTAL	293	234	220	211	204	204	197	189	173	143	138	100%

Table 3.14 UK emissions of PM₁ by sector (ktonnes) estimated for the mass fraction of particles below 1 µm in each sector in the PM₁₀ inventory.

Source	1970	1975	1980	1985	1990	1991	1992	1993	1994	1995	1996	1996
Combustion in energy production & transformation	22	18	15	14	19	18	20	15	13	11	11	10%
Commercial, institutional & residential combustion	70	41	33	31	19	20	19	18	15	13	14	13%
Industrial combustion	22	13	10	7	6	7	7	6	6	5	4	4%
Non-combustion processes	32	32	32	32	33	32	32	32	33	32	32	30%
Industrial processes	30	30	30	30	30	30	30	30	30	30	30	28%
Mining & quarrying	2	2	2	2	2	2	2	2	2	2	2	2%
Construction	0	0	0	0	0	0	0	0	0	0	0	0%
Road transport	35	38	42	43	50	50	48	46	45	41	38	35%
Petrol exhaust	7	8	10	11	11	11	10	9	8	7	6	6%
Diesel exhaust	28	30	32	32	39	39	38	37	36	34	31	29%
Tyre wear	0	0	0	0	0	0	0	0	0	0	0	0%
Brake wear	0	0	0	0	0	0	0	0	0	0	0	0%
Other transport & machinery	5	4	4	3	3	3	3	3	3	3	3	3%
Waste treatment & disposal	32	32	35	34	28	28	25	25	19	5	5	5%
TOTAL	219	179	171	166	159	159	154	146	134	111	107	100%

Table 3.15 UK emissions of PM_{0.1} by sector (ktonnes) estimated for the mass fraction of particles below 0.1 µm in each sector in the PM₁₀ inventory.

Source	1970	1975	1980	1985	1990	1991	1992	1993	1994	1995	1996	1996
Combustion in energy production & transformation	7	6	5	5	6	6	6	5	5	4	4	10%
Commercial, institutional & residential combustion	6	4	4	4	3	3	3	3	2	2	2	7%
Industrial combustion	8	5	4	3	3	3	3	3	2	2	2	6%
Non-combustion processes	5	5	5	5	5	5	5	5	5	5	5	13%
Industrial processes	5	5	5	5	5	5	5	5	5	5	5	13%
Mining & quarrying	0	0	0	0	0	0	0	0	0	0	0	0%
Construction	0	0	0	0	0	0	0	0	0	0	0	0%
Road transport	19	21	23	23	27	27	26	25	25	23	21	60%
Petrol exhaust	3	3	4	4	4	4	4	3	3	3	2	7%
Diesel exhaust	16	17	19	19	23	23	22	22	21	20	18	52%
Tyre wear	0	0	0	0	0	0	0	0	0	0	0	0%
Brake wear	0	0	0	0	0	0	0	0	0	0	0	1%
Other transport & machinery	1	1	1	1	1	1	1	1	1	1	1	2%
Waste treatment & disposal	7	7	7	7	6	6	5	5	4	1	1	3%
TOTAL	53	48	48	47	49	50	48	46	43	37	35	100%

3.2.4 UNCERTAINTIES IN EMISSION ESTIMATES AND SOURCES NOT COVERED

Although the primary UK emissions inventory for PM₁₀ is continuously being improved, the uncertainties in the estimates must still be considered high. These uncertainties stem from uncertainties in the emission factors themselves, the activity data with which they are combined to quantify the emissions and the size distribution of particle emissions from the different sources. There is also the possibility that not all the sources that exist have been considered and some sources which are known to exist have not been quantified because of the lack of relevant data for estimating the emissions.

Emission factors are generally based on a few measurements on an emitting source which is assumed to be representative of the behaviour of all similar sources. Emission estimates for PM₁₀ are based whenever possible on measurements of PM₁₀

emissions from the source, but sometimes measurements have only been made on the mass of total particulate matter and it has been necessary to convert this to PM₁₀ based on the size distribution of the sample collected.

The view was expressed in the Third QUARG Report that it was not possible to quantify the accuracy of the national emission estimates, but it was possible to give a qualitative indication of the overall reliability of the estimates and to rank them by source sector. This ranking of the reliability of emission estimates still holds. In order of decreasing reliability of emission estimates, the ranking is broadly:

- 1) Road transport
- 2) Stationary combustion
- 3) Industrial processes
- 4) Mining & quarrying and construction.

3.2.4.1 *Uncertainties in Road Transport Emissions*

Emission factors

Diesel cars are probably the source of PM₁₀ emissions which have had the largest number of measurements made. Even so, it is known that emissions from the same type and even model of car are very variable. Emission factors are based on the mean of results from surveys of on-road vehicles. Emission factors for diesel cars are probably reliable to within 30%.

There are fewer measurements of PM₁₀ emissions from heavy duty diesel vehicles, especially buses and even fewer for petrol vehicles. Emissions from HGVs are again variable, depending on vehicle maintenance and vehicle load. For petrol engined vehicles, the range of uncertainty is estimated to be around a factor of two for those running on unleaded petrol, but possibly as high as a factor of 5 - 10 for cars running on leaded petrol.

Activity data

The relevant activity data for estimating emissions from road transport are traffic and fleet composition statistics. The traffic data are based on surveys and the vehicle kilometre data scaled up to include the whole of the road network in Great Britain. The national vehicle kilometre data will therefore be subject to uncertainties, but these have not been quantified. Fleet composition data should be very reliable as these are based on DVLA statistics on vehicle registrations. Account is taken of the fact that newer vehicles travel greater mileage in a year than older vehicles using statistics from DETR.

3.2.4.2 *Uncertainties in Emissions from Stationary Combustion*

Emission factors

The uncertainties in emission factors for stationary combustion plant are fairly high because of the limited number of measurements made on the

many different plant and fuel types. However, for large combustion plant, the emission factor database has been expanding and the overall reliability of the data improving to probably within a factor of 2. Some of the factors for PM₁₀ will be based on measurements of total particulate matter sampled. Data for domestic combustion appliances using solid fuels are expected to be particularly uncertain and variable as demonstrated by the CRE study (1997).

Activity data

Fuel consumption statistics are the relevant activity data for estimating emissions from stationary combustion plant. These should on the whole be reliable, especially for large combustion plant (e.g. power stations), but probably less so for the many different types of small industrial, commercial and residential combustion plant.

3.2.4.3 *Uncertainties in Emissions from Production Processes*

Emission factors

Emissions data from the many non-combustion industrial, construction and mining and quarrying processes are very uncertain and again variable. There are very few measurements from industrial processes of which there are many different types. Much of the available data are of questionable validity for processes in the UK. This is especially true for construction and mining and quarrying processes where the available data are from US sources and would not be applicable to the wetter climate of the UK. The very nature of these fugitive sources means that they are likely to be variable and therefore using short period measurements of emissions to estimate annual emissions adds further uncertainty. The range of uncertainty for these processes is possibly as high as an order of magnitude.

Activity data

The appropriate activity data for these sources are usually production statistics for industrial

processes, aggregate production statistics for mining and quarrying and floor space for construction activity. Production statistics are probably reasonably reliable, but construction activity data much less so and again are expected to be variable in time and location. Construction activity is likely to be intense at a particular location only for a particular period in the year.

3.2.4.4 *Estimated Overall Uncertainty*

The overall uncertainty of the national emission inventory for PM₁₀ has not yet been quantified. However, TNO in their compilation of a European inventory of PM₁₀ (as well as for finer particle mass ranges) in 1990 and 1993 for the Dutch RIVM have attempted to quantify the uncertainties of their estimates for each of the 26 countries included in the inventory (TNO, 1997). The uncertainties were expressed as a standard deviation around an estimated mean value.

In their calculations, TNO assumed that the uncertainties in the activity rates were negligible compared with those in the emission factors. A USEPA approach was used and each factor was subjectively given a relative error reflecting the emission factor range at the 95% confidence level. This information was used to derive a standard deviation in the emission factor.

TNO concluded a total PM₁₀ emission rate for the UK in 1990 of 270 ktonnes compared with 314 ktonnes estimated for 1990 in the current UK national inventory. The agreement appears to be very good for that year although there are differences between the trends in PM₁₀ emissions in the two inventories as discussed in Section 3.2.7. The standard deviation which TNO estimate for their UK inventory was 27 ktonnes, i.e. 10%. This seems much smaller than anticipated for the national inventory and appears unrealistic.

3.2.4.5 *Emission Sources Not Included*

Although the major sources are thought to be included in the national PM₁₀ inventory, there are

still some sources which have not been included either because of lack of emission factor measurements for conditions pertinent to the UK or because of lack of appropriate activity data. Sources that are notably missing from the inventory are:

- ***Entrainment of road dust.*** *Whilst USEPA data exist for calculating emissions from this source, they are not considered appropriate for UK road conditions. Further measurements of dust resuspension on UK-typical roads are required to provide data for emission inventory application. Until then, resuspension of road dust remains a major area of uncertainty in the UK particulate emission inventory.*
- ***Agricultural sources.*** *Emissions from ploughing and harvesting activities may be significant sources. Certain livestock farming activities may also be important. Emission factors relevant to UK agricultural activities are not available to estimate emissions from these sources at present. Further measurements of particulate emissions from different agricultural activities in the UK are required to provide data for emission inventory applications.*
- ***Biological sources.*** *Emission factors are not currently available for this source.*

Inventories currently provide only an annual emission rate. They do not take account of episodic emission events nor the temporal variability of certain sources, yet these would be important for receptor modelling and source apportionment studies and for interpreting ambient concentration measurements of airborne particles. The classic episodic event, as far as primary emissions of particulate matter is concerned, is the period around Bonfire Night. No attempt has yet been made to estimate emissions on Bonfire Night, but this would clearly be useful for the interpretation of ambient PM₁₀ concentration measurements which show air quality episodic behaviour at this time. This is an area that merits further investigation.

Seasonality in emissions of particulate matter is also of interest. Emissions from stationary combustion sources would be expected to be higher in winter months. Agricultural and biological sources would show different seasonal emission trends.

3.2.5 PROJECTED EMISSIONS OF PM₁₀ FROM ROAD TRANSPORT

Forecasting trends in PM₁₀ emissions depends on making judgements and assumptions on future activities and emissions performance of each source. This has been carried out for road transport exhaust emissions, both nationally and in urban areas.

The latest projections on emissions from road transport take account of the new and more detailed national estimates of the growth in traffic in the UK, assumptions about the composition of the vehicle fleet based on fleet turnover and new vehicle sales and the latest understanding on new fuel standards and emission limits for new vehicles manufactured after 2000 enforced by forthcoming European Community Directives.

The projections are based on the central growth rate of the revised and more detailed National Road Traffic Forecasts produced by the DETR (the 1997 NRTF) (DETR, 1997a). The NRTF provides traffic forecasts for different types of vehicles and roads from 1996 to 2026. The central growth rate is lower than the mean of the previously estimated growth rates.

It is assumed in the projections that future diesel car sales will remain at a rate of 20%, close to the level they have already reached. Based on current Government policy, it is also assumed that consumption of leaded petrol will virtually cease by 2000.

The age distribution of the vehicle parc in future years is calculated from projections of new vehicle sales and on the assumption that the survival rates of vehicles remain at the mean levels of recent years. Projections of new vehicle sales are based on the NRTF and output from DETR's Vehicle Market Model.

The projections are also based on the implementation of proposed European Community Directives which will further limit emissions of PM₁₀ from new diesel vehicles marketed after 2000. These are referred to as the Euro III reductions. Further emission reductions are also proposed for new diesel vehicles from 2005. These are referred to as the Euro IV reductions, but although proposed for light duty and heavy duty vehicles, they are assumed in the projections to come into effect only for light duty vehicles (cars and vans) as both the dates of implementation and the extent of emission reductions are far more uncertain for heavy duty vehicles.

Emission factors for Euro III and Euro IV diesel vehicles are calculated by scaling the emission factors for Euro II vehicles according to the ratio of the Euro III/Euro II and Euro IV/Euro II Type Approval limit values. The appropriate scaling factors and assumed UK implementation dates based on current Government thinking are shown in Table 3.16. It is assumed that the on-road emission factors are scaled down by the same amount as the Type Approval limit values are scaled relative to Euro II limit values. It is further assumed that the emission factors are scaled down by the same amount at all speeds which is an assumption which can be questioned.

The effect on UK emissions of these European Directives on emissions from new vehicles in 2000 and 2005, reflected by the scaling factors shown in Table 3.16, will occur gradually from their implementation dates, as the new vehicles meeting these emission standards gradually penetrate the vehicle parc and older vehicles are scrapped. In addition to the reductions in emissions from new diesel vehicles post-2000, it is also assumed that PM₁₀ emissions from vehicles currently in the fleet will be reduced due to the implementation of new EU fuel standards in 2000 and 2005. Current indications are that these standards will reduce PM₁₀ emissions from pre-Euro III vehicles in 2000 and 2006 by the amounts shown in Table 3.17 for PM₁₀. The effect of the fuel directive will occur as a step change in UK emissions, as improvement to fuel quality will affect emissions from old as well as new vehicles in the fleet.

Table 3.16 PM₁₀ emission scaling factors for new diesel vehicles relative to Euro II vehicles and implementation dates assumed in projections. The on-road Euro II emission factors are scaled down by the same scaling factors at all speeds.

	Euro III		Euro IV*	
	Implementation date	Scaling factor	Implementation date	Scaling factor
Cars	1/1/2001	0.63	1/1/2006	0.31
LGV < 1250kg	1/1/2001	0.63	1/1/2006	0.31
LGV 1250-1700kg	1/1/2002	0.63	1/1/2006	0.31
LGV >1700kg	1/1/2002	0.63	1/1/2006	0.31
HGVs**	1/10/2001	0.7	–	–
Buses**	1/10/2001	0.7	–	–

* *Indicative values*

** *No Euro IV reductions are assumed for these vehicles in the projections as the Euro IV implementation dates and scaling factors are currently too uncertain.*

It should be noted that the reductions in emissions arising from new vehicle and new fuel standards from 2000 assumed for the projections are only indicative, based on the current proposals being negotiated by the EU Member States up to March 1998¹. Both the standards and their effects on emissions are subject to change. This is especially true for the Stage IV and 2005 fuel standards which are referred to as indicative standards.

In addition to the new European vehicle and fuel standards, it is also assumed in the projections that improved vehicle technologies and fuels will reduce the rate that emissions from new vehicles in the fleet after 2000 will increase with vehicle age. The current assumptions are that PM₁₀ emissions from diesel cars and LGVs increase by a factor of 2 over 80,000 km, but this degradation rate will be reduced to a factor of 1.2 for Euro III vehicles based on type approval limits linked to durability.

The projected emissions of PM₁₀ from road vehicle exhausts up to 2025 based on these assumptions are shown in Table 3.18 and Figure 3.1. Table 3.19 and Figure 3.2 show the corresponding projections for UK urban roads. The results show a 56% reduction in UK emissions and a 53% reduction in urban UK

emissions by 2005 relative to 1995 levels.

The decline in emissions from petrol cars and LGVs from 1990 is due to the decrease in the consumption of leaded petrol and the penetration of vehicles equipped with three-way catalysts in the vehicle parc. These changes both lead to lower vehicle emission factors (g/km). As noted in Section 3.2.2.1, measurements of particulate emissions from petrol engined vehicles are currently very sparse, so the emission factors given by Gover *et al.* (1994) which were used for the inventory are highly uncertain. Nevertheless, they should indicate a realistic trend in emissions from these vehicles.

The assumptions made for these projections represent a “business as usual” scenario, following the Government’s expectations in the demand for fuel, vehicle ownership and passenger and freight transport by road, combined with future European Union legislation on vehicle emissions. Other factors which can affect the emission projections include the switch to vehicles running on alternative fuels (e.g. LPG, CNG, City Diesel); retrofitting of particulate traps; new vehicle technologies such as electric powered vehicles and hybrids; fiscal measures to encourage scrappage of older vehicles; and public transport and traffic management schemes in urban areas. Many

¹ *EC directives on fuel quality standards for 2000 and 2005 and vehicle emission standards for passenger cars and light vans also for 2000 and 2005 have recently been adopted. However, the precise impact of these directives could not be assessed in time for this publication.*

Table 3.17 Assumed reduction in PM₁₀ emissions from pre-2000 diesel vehicles resulting from new fuel standards.

		% reduction
Pre-Euro III reductions from 1/1/2000	Cars and LGVs	0.25%
	HGVs and buses	1.5%
Pre-Euro III reductions from 1/1/2006	Cars and LGVs	0.5%
	HGVs and buses	2.5%

Table 3.18 Projected UK exhaust emissions of PM₁₀ from road transport (ktonnes).

ktonnes		1990	1995	1996	2000	2005	2010	2015	2020	2025
Cars	Petrol	15.3	10.6	9.8	4.5	3.9	3.9	4.2	4.5	4.6
	DERV	2.1	3.4	3.4	3.8	3.4	2.0	1.1	0.9	0.9
	All Cars	17.4	14.0	13.2	8.3	7.3	5.9	5.3	5.3	5.5
LGV	Petrol	1.7	0.9	0.7	0.3	0.1	0.1	0.1	0.2	0.2
	DERV	4.9	7.4	7.4	7.0	6.3	3.6	2.0	1.7	1.9
	All LGV	6.5	8.2	8.1	7.3	6.5	3.7	2.2	1.9	2.0
HGV	Artic	8.2	7.5	7.0	4.0	2.5	2.1	2.3	2.6	2.9
HGV	Rigid	23.7	16.8	14.6	7.7	4.2	3.3	3.2	3.4	3.5
ALL HGV		31.9	24.3	21.5	11.7	6.7	5.4	5.6	6.0	6.4
Buses		6.5	5.0	4.6	2.8	1.8	1.2	1.1	1.1	1.2
Motorcycles		0.6	0.4	0.4	0.4	0.4	0.4	0.5	0.5	0.5
All DERV		45.3	40.1	36.8	25.1	18.1	12.2	9.8	9.7	10.4
All Petrol		17.5	11.8	10.9	5.2	4.4	4.5	4.8	5.1	5.3
All Vehicles		62.8	51.9	47.7	30.4	22.6	16.7	14.6	14.7	15.6

measures such as these are being promoted or considered specifically to have a beneficial effect on particulate emissions from road transport.

3.2.6 SPATIALLY MAPPED UK EMISSIONS AND URBAN INVENTORIES OF PM₁₀

3.2.6.1 Spatially Mapped Emissions of PM₁₀ in the UK

The national emissions inventory has been spatially mapped on a 1 x 1 km grid square resolution for a number of pollutants (Goodwin *et al.*, 1997). Figure 3.8 shows the UK map for primary PM₁₀ emissions in 1996 expressed as tonnes per square km in 1 x 1 km squares.

The map was produced by representing emissions from different sectors as point sources, area sources

or line sources. Emissions from power stations and large industrial plant are represented by points located by a grid reference. Emissions are either reported for a particular plant as total particulate matter and a PM₁₀ size fraction applied to derive the emissions or fuel use data and fuel-based emission factors were used to calculate emissions from the plant. For sectors such as industrial combustion, the national emission estimate is distributed among each site using an appropriate surrogate statistic. Emissions from domestic, commercial and small industrial combustion processes, off-road transport, agricultural machinery and construction were treated as area sources and spatially distributed over the UK using surrogate statistics available for each grid square, e.g. population, employment and land use. Emissions from quarrying were mapped according to levels of quarrying activity in different regions of the country.

Table 3.19 Projected urban UK exhaust emissions of PM₁₀ from road transport (ktonnes).

ktonnes		1990	1995	1996	2000	2005	2010	2015	2020	2025
Cars	Petrol	6.9	4.9	4.5	2.0	1.7	1.8	1.9	2.0	2.0
	DERV	1.1	1.8	1.8	2.1	2.0	1.2	0.6	0.5	0.5
	All Cars	8.0	6.7	6.3	4.2	3.7	2.9	2.5	2.5	2.6
LGV	Petrol	0.7	0.4	0.3	0.1	0.1	0.1	0.1	0.1	0.1
	DERV	2.4	3.6	3.6	3.4	3.1	1.8	1.0	0.9	0.9
	All LGV	3.1	4.0	3.9	3.6	3.2	1.8	1.1	0.9	1.0
HGV	Artic	1.7	1.4	1.3	0.7	0.4	0.3	0.4	0.4	0.4
HGV	Rigid	10.3	7.5	6.7	3.6	2.0	1.6	1.6	1.6	1.7
ALL HGV		11.9	8.9	8.0	4.3	2.4	1.9	1.9	2.0	2.1
Buses		3.9	3.6	3.3	2.1	1.4	1.0	0.9	0.9	1.0
Motorcycles		0.6	0.4	0.4	0.4	0.4	0.4	0.5	0.5	0.5
All DERV		19.3	17.9	16.7	11.9	8.9	5.9	4.5	4.3	4.5
All Petrol		8.2	5.6	5.1	2.6	2.2	2.2	2.4	2.5	2.6
All Vehicles		27.5	23.5	21.8	14.5	11.1	8.1	6.8	6.8	7.1

Emissions from road transport were mapped by treating major roads and minor roads separately. Major roads were treated as line sources using DETR traffic flow data for 1996 on around 14,000 road links on the major road network. Emissions were calculated on each link using emission factors, traffic flow for the different types of vehicles and assuming a distribution of vehicle speeds on each link appropriate for the type of vehicle and type of road and hence related to the speed limit. Emissions for each link were then mapped according to its Ordnance Survey grid reference.

As traffic flow data is not available for the minor roads, these roads were treated as area sources. Emissions from minor roads were calculated using land use and population data as surrogate statistics.

The map clearly shows the major conurbations and emissions from a number of major trunk roads. Around these areas, emissions are fairly widespread from domestic and commercial combustion processes, traffic on minor roads, construction and use of agricultural machinery.

3.2.6.2 Urban Emission Inventories

Emission inventories for PM₁₀ have been compiled for nine major urban and industrial areas in the UK

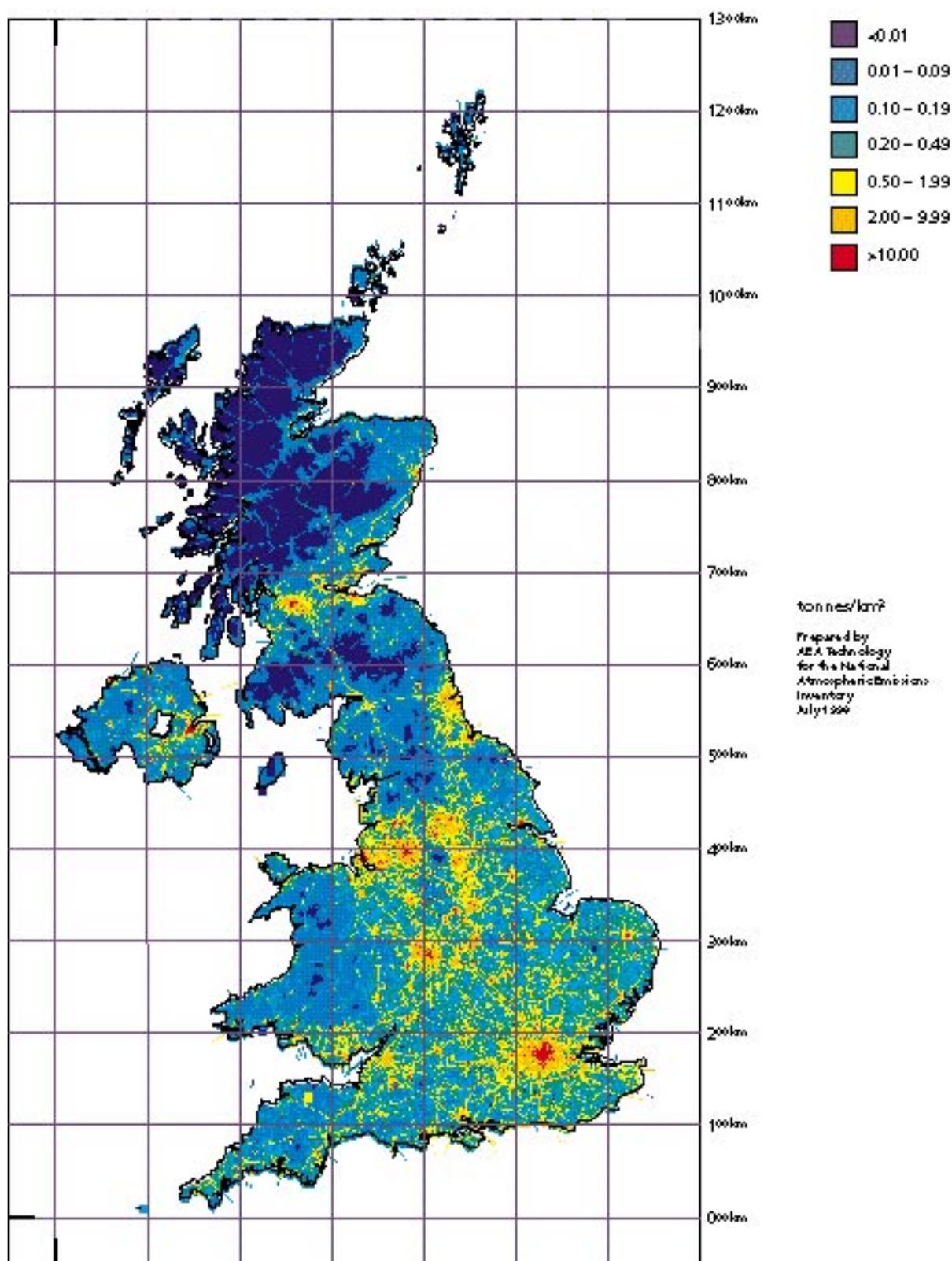
by the London Research Centre (LRC) (Hutchinson and Clewley, 1996; Buckingham *et al.*, 1997a; Buckingham *et al.*, 1997b; Buckingham *et al.*, 1998a; Buckingham *et al.*, 1998b). Urban inventories are compiled using a more detailed “bottom up” approach in which local data are gathered for the inventory. For a particular area, they should always provide a more accurate representation of emissions than can be achieved from spatially disaggregating the national inventory by a “top down” approach. In fact, the two inventories are complementary.

Urban inventories have so far been compiled for (London Research Centre, 1998):

- *West Midlands (1994)*
- *Greater Manchester (1995)*
- *Merseyside (1996)*
- *Bristol (1996)*
- *Southampton & Portsmouth (1996)*
- *Swansea & Port Talbot (1996)*
- *London (1996)*
- *Middlesbrough (1996)*
- *Glasgow (1996)*

The year refers to the base year of the inventory, although they sometimes cross more than one calendar year.

Figure 3.8 1996 Emissions of PM₁₀ mapped across the UK.



The results of the urban PM₁₀ inventories for these areas are summarised in Table 3.20. These inventories have each been spatially disaggregated by LRC on a 1 x 1 km square grid.

Figure 3.9 displays as percentages the relative contribution of the sources to PM₁₀ emissions in each area and compares these with the contribution for the UK national inventory. The variation in the relative contribution of the different sectors appears quite striking, reflecting the variation in industrial and urban activity in each area, with London and Merseyside being at the two extremes. Emissions in London are dominated by road transport (77%) because of the absence of major industrial activity in the area. Conversely, emissions in Merseyside are dominated by industrial processes (85%, including industrial combustion and sources involving solvent use) with road transport only making a 12% contribution. It should be noted that emissions from road transport in the urban areas include tyre and brake wear, but does not include road dust resuspension.

Comparing urban inventories, however, requires a note of clarification if misleading interpretation is to be avoided. First, the contribution of different emission sources depends on where the boundaries of the urban area are fixed and this can be rather

arbitrary. The inventory for a particular urban area could be dominated by one industrial point source away from the main urban centre. The inventories for Merseyside and Swansea & Port Talbot illustrate this, where PM₁₀ emissions are dominated by a few industrial processes. Conversely, the Glasgow inventory is strongly influenced by a busy motorway passing through the area included in the inventory. Excluding emissions from all industrial processes and the industrial combustion and the solvent use categories in the urban inventories (Table 3.20), the extreme variability in the road traffic contribution to PM₁₀ emissions is somewhat smoothed out. Caution should also be exercised when comparing the urban inventories for different cities because each one was prepared using the best reported data available at the time. Less detailed emissions data were available when the first urban inventories were prepared (e.g. West Midlands), so the uncertainties of the older inventories will be higher than those compiled more recently (e.g. Middlesbrough). Finally, it should also be recognised that when considering the impact of a source or source type upon local air quality at ground-level, the height of emissions is important with ground-level sources (e.g. road traffic) having a far greater impact within a few tens of metres than emissions from high stacks whose impact would not be localised, but at some distance from the source where the emission plume comes to ground level.

Figure 3.9 Comparison of urban PM₁₀ primary emissions in 1994/96 (Urban data from London Research Centre, UK data from NAEI (Salway *et al.*, 1997).

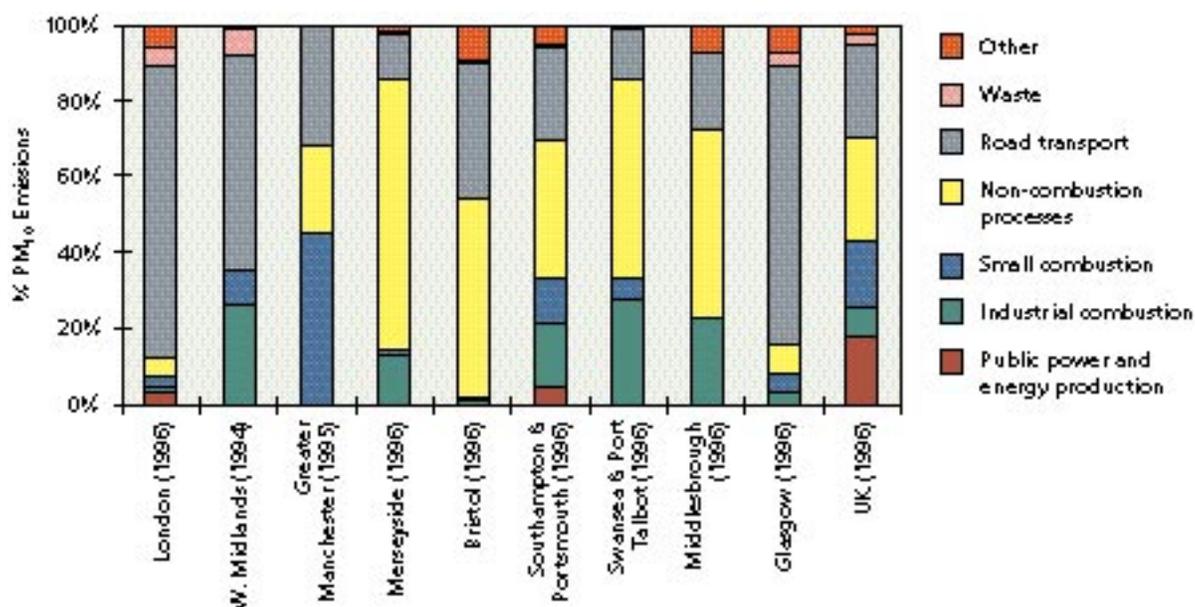


Table 3.20 Urban PM₁₀ emission inventories by category (tonnes). Data from London Research Centre.

	London	W. Midlands	Greater Mancheser	Merseyside	Bristol	Southampton & Portsmouth	Swansea & Port Talbot	Middlesbrough	Glasgow
Combustion in energy production	343					102			
Commercial and residential combustion	238	340	5082	102	19	252	125	2	46
Industrial combustion	182	1093		1241	9	367	641	505	35
Industrial processes	400	19	2572	6039	600	764	1212	1083	84
Solvent use	47			674	2	21	1		
Road transport	7649	2349	3494	1100	413	514	299	426	748
Cars & taxis	1554	785	570	226	70	169	55	76	100
LGVs	632	238	309	67	40	87	26	35	47
HGVs	4871	1052	2124	384	227	209	193	273	494
Buses	570	264	485	418	75	44	23	40	105
Motorcycles	22	10	6	5	1	5	2	1	2
Other transport and machinery	537	27	31	144	110	114	33	154	76
Waste treatment and disposal	491	293	9	82	8	13	1	10	33
TOTAL	9887	4121	11188	9381	1161	2147	2312	2226	1022
% Road transport – All sources	77%	57%	31%	12%	36%	24%	13%	19%	73%
% Road transport – Exc. ind. processes	83%	78%	41%	77%	75%	52%	65%	67%	83%

The figures for London, Merseyside and Swansea and Port Talbot have been very slightly revised by LRC (personal communication, 1998) from those originally published due to re-examination of certain reported data. Road transport does not include road dust resuspension.

Table 3.21 shows other emission statistics for the urban inventories useful for the interpretation of monitoring data and understanding the source apportionment of airborne particles. For seven of the urban areas studied by LRC, the ratio of total primary PM₁₀ to total CO and NO_x emissions for the area are shown and the contribution of road transport as a percentage of the total emissions of each of these pollutants in each urban area is summarised. The same statistics are also shown for the UK inventory.

Once again, caution needs to be exercised when interpreting and comparing data like these for different urban areas for the reasons mentioned earlier. Trends in the emissions of one particular pollutant can be skewed by the positioning of the

urban boundary and the contribution of a large point source. Table 3.21(a) shows that the ratios in total emissions of these pollutants in each area is quite variable, emphasising the variation in industrial and urban activity. It can also be seen that in London, where traffic makes the dominant contribution to emissions of each of these pollutants, the primary PM₁₀/CO and PM₁₀/NO_x emission ratios are relatively low, whereas the ratios are considerably higher in regions like Merseyside and Swansea and Port Talbot where other sources of these pollutants are more apparent. Excluding emissions from industrial processes and industrial combustion from the analysis tends to smooth out the major differences in the primary PM₁₀/CO and PM₁₀/NO_x emission ratios between each urban area, as can be seen in Table 3.21(b). The emission

Table 3.21 Ratio of urban emission inventories for PM₁₀, CO and NO_x and the contribution of road traffic to emissions of each pollutant based on data from LRC. Figures for the UK inventory shown for comparison, taken from the National Atmospheric Emissions Inventory for 1996.

(a) Ratio in emissions from all sources

		London	W. Midlands	Greater Mancheser	Merseyside	Bristol	Southampton & Portsmouth	Swansea & Port Talbot	Middlesbrough	Glasgow	UK
Total emissions	PM ₁₀ /CO	0.018	0.026	0.063	0.110	0.037	0.042	0.128	0.025	0.028	0.046
	PM ₁₀ /NO _x	0.067	0.089	0.148	0.211	0.114	0.093	0.147	0.069	0.086	0.103
% Road transport	PM ₁₀	77%	57%	31%	12%	36%	24%	13%	19%	73%	25%
	CO	97%	98%	95%	91%	97%	93%	83%	22%	95%	71%
	NO _x	75%	85%	63%	42%	61%	47%	28%	17%	76%	47%

(b) Ratio in emissions from all sources excluding industrial processes and combustion

		London	W. Midlands	Greater Mancheser	Merseyside	Bristol	Southampton & Portsmouth	Swansea & Port Talbot	Middlesbrough	Glasgow	UK
Total emissions	PM ₁₀ /CO	0.017	0.019	0.049	0.018	0.018	0.020	0.026	0.030	0.025	0.030
	PM ₁₀ /NO _x	0.065	0.067	0.115	0.057	0.066	0.060	0.080	0.053	0.078	0.072
% Road transport	PM ₁₀	83%	78%	41%	77%	75%	52%	65%	67%	83%	38%
	CO	97%	99%	96%	97%	98%	97%	87%	96%	96%	72%
	NO _x	78%	88%	64%	74%	74%	66%	77%	46%	78%	51%

ratios for the UK inventory are within the range of those for the urban inventories.

Urban inventories are currently being compiled by LRC for West Yorkshire and Belfast.

3.2.6.3 Primary PM₁₀ emissions in Greater Belfast

A primary PM₁₀ emission inventory for Greater Belfast has recently been compiled by Cambridge Environmental Research Consultants as part of a study for Belfast City Council on modelling airborne PM₁₀ and SO₂ concentrations (CERC, 1998). Emissions data were taken from the NAEI (Salway *et al.*, 1997) and the CRE study (CRE, 1997) on domestic solid fuel burning appliances. These data were combined with a domestic fuel usage survey for Belfast carried out by Market Research Northern Ireland (DETR, 1997b). Results of the CERC inventory expressed as an annual rate of emissions over the 1995/1996 winter period are summarised in Table 3.22.

Table 3.22 Annual emission rate of primary PM₁₀ from sources in Belfast over the 1995/1996 winter period. Based on data taken from CERC (1998).

	Emission rate 1995/1996 (tonnes/year)
Power stations	3000
Household fuel use	430
Other point sources	220
Road traffic	150
TOTAL	3800

Emissions are dominated by two coal-fired power stations which both emit through tall stacks 200m and 73m in height. Over the winter period of the CERC modelling study, domestic fuel consumption contributed around 11% to primary emissions overall, but its contribution to near-ground level sources (i.e. excluding the power station emissions) was around 54%.

The domestic fuel usage survey indicated that solid fuel is used in 23% of households in Belfast, 17% as the main fuel. It also showed that about 18% of

solid fuel users may be burning unauthorised, higher emitting solid fuels.

3.2.7 EUROPEAN EMISSIONS INVENTORY OF PM₁₀, PM_{2.5} AND PM_{0.1}

TNO Institute of Environmental Sciences, Energy Research and Process Innovation in the Netherlands have compiled a primary PM₁₀ emissions inventory for Europe for the years 1990 and 1993 (TNO, 1997). This inventory covers not only PM₁₀, but also PM_{2.5} and very fine particles PM_{0.1} for 26 European countries. Inventories were produced for each country, broken down into eight main source categories, and the data combined to produce the European inventory. The total emissions for Europe were also spatially distributed for 1990 on a 0.5° x 1° grid.

The 1993 data for each country are summarised in Tables 3.23 to 3.25.

Figure 3.10 displays emissions of PM₁₀ per capita for each country in different country groups (Western Europe, Central and Eastern Europe and the former USSR). Figure 3.11 shows the averaged PM₁₀ emissions per capita for each of these country groups in 1990 and 1993. It can be seen that per capita emissions are generally higher for the Central & Eastern European (CEE) countries than for the Western European countries and higher still for the former USSR countries. The reason for this is the greater use of solid fuels for stationary combustion in the CEE countries and former USSR and lack of effective emission abatement measures. It can also be seen how emissions per capita have been falling in Western Europe from 1990 to 1993, but slightly rising in Central & Eastern Europe.

Table 3.23 European inventory for primary PM₁₀ in 1993 in ktonnes. Data from TNO (1997).

	Power generation	Industrial combustion	Small combustion	Processes	Road transport	Other transport	Agriculture	Waste processing	Total
Albania	0.5	4.5	0.9	0.8	1.3	0.0	0.4	0.0	8
Austria	1.3	2.4	9.7	7.5	10.0	1.0	9.1	0.0	41
Belgium	1.6	7.4	14.0	21.0	21.0	1.0	17.0	0.7	84
Bulgaria	76.0	14.0	7.5	24.0	11.0	1.0	8.8	0.0	140
Switzerland	0.1	1.2	12.0	2.3	5.2	0.0	4.3	0.3	25
Former Czechoslovakia	140.0	150.0	110.0	64.0	35.0	1.0	18.0	0.0	520
Former GDR	250.0	0.4	44.0	39.0	37.0	0.0	9.1	0.0	380
Former Western Germany	35.0	79.0	19.0	80.0	160.0	0.0	56.0	0.7	430
Germany	285.0	79.4	63.0	119.0	197.0	0.0	65.1	0.7	810
Denmark	3.1	2.1	9.7	3.4	11.0	1.0	25.0	0.2	56
Spain	14.0	18.0	12.0	30.0	65.0	6.0	48.0	1.5	190
Finland	2.0	4.1	5.7	6.5	12.0	0.0	3.3	0.0	34
France	1.5	40.0	54.0	49.0	120.0	0.0	53.0	78.0	400
UK	21.0	32.0	41.0	44.0	99.0	1.0	31.0	2.8	280
Greece	12.0	5.1	6.5	6.4	20.0	2.0	4.7	0.0	57
Hungary	31.0	3.9	16.0	21.0	17.0	0.0	22.0	0.0	110
Ireland	1.7	1.2	8.7	1.3	5.7	0.3	4.1	0.0	23
Italy	35.0	26.0	23.0	49.0	110.0	0.0	35.0	16.0	290
Luxembourg	0.0	0.9	0.7	3.5	1.1	0.0	0.0	0.0	6
Norway	0.0	3.2	1.3	6.9	5.2	2.5	1.9	0.1	21
Poland	510.0	210.0	150.0	85.0	38.0	3.0	45.0	0.0	1000
Portugal	4.4	3.1	2.1	5.5	12.0	0.0	8.0	0.0	35
Romania	86.0	29.0	6.2	66.0	14.0	1.0	25.0	0.0	230
Former USSR (European part only)	740.0	950.0	1900.0	1300.0	210.0	100.0	170.0	0.1	5400
Sweden	0.6	4.6	6.3	8.9	9.3	0.5	5.9	0.2	36
Former Yugoslavia	170.0	26.0	37.0	35.0	15.0	0.0	18.0	0.0	300
Europe, exc. former USSR	1400	670	600	660	830	30	450	100	4800

Table 3.24 European inventory for primary PM_{2.5} in 1993 in ktonnes. Data from TNO (1997).

	Power generation	Industrial combustion	Small combustion	Processes	Road transport	Other transport	Agriculture	Waste processing	Total
Albania	0.4	2.0	0.6	0.3	0.9	0.0	0.2	0.0	4
Austria	0.8	1.5	5.8	3.9	6.8	0.4	4.4	0.0	24
Belgium	0.9	3.9	7.9	13.0	14.0	1.0	8.3	0.5	49
Bulgaria	47.0	6.3	3.5	16.0	7.5	0.2	3.9	0.0	85
Switzerland	0.1	0.7	7.2	1.1	3.4	0.0	2.0	0.2	15
Former Czechoslovakia	89.0	64.0	47.0	41.0	24.0	0.0	8.7	0.0	270
Former GDR	160.0	0.2	18.0	27.0	27.0	0.0	4.4	0.0	240
Former Western Germany	22.0	38.0	8.8	48.0	100.0	10.0	27.0	0.5	250
Germany	182.0	38.2	26.8	75.0	127.0	10.0	31.4	0.5	490
Denmark	1.7	1.2	7.3	1.7	7.3	0.7	12.0	0.2	33
Spain	8.7	8.8	7.1	16.0	43.0	5.0	23.0	1.0	110
Finland	1.2	2.4	3.6	4.0	7.9	0.4	1.6	0.0	21
France	0.9	20.0	34.0	27.0	80.0	2.0	22.0	54.0	240
UK	12.0	16.0	21.0	26.0	65.0	5.0	13.0	1.9	160
Greece	7.7	2.7	4.2	2.7	13.0	2.0	2.0	0.0	34
Hungary	20.0	1.8	7.6	15.0	11.0	1.0	9.2	0.0	65
Ireland	1.1	0.7	4.8	0.5	3.8	0.2	1.9	0.0	13
Italy	23.0	13.0	15.0	27.0	70.0	1.0	15.0	11.0	170
Luxembourg	0.0	0.5	0.4	2.2	0.7	0.0	0.0	0.0	4
Norway	0.0	1.7	0.8	4.1	3.4	2.3	0.9	0.0	13
Poland	300.0	95.0	69.0	50.0	25.0	2.0	22.0	0.0	560
Portugal	2.7	1.9	1.6	2.2	8.1	0.1	3.6	0.0	20
Romania	58.0	12.0	3.6	42.0	9.3	0.7	12.0	0.0	140
Former USSR (European part only)	440.0	430.0	900.0	830.0	140.0	70.0	77.0	0.1	2900
Sweden	0.4	2.8	3.7	5.7	6.1	0.5	2.8	0.2	22
Former Yugoslavia	110.0	11.0	17.0	24.0	10.0	0.0	8.4	0.0	180
Europe, exc. former USSR	870	310	300	400	550	30	210	70	2700

These differences are also highlighted in Figure 3.12 which displays as percentages the relative contributions of the different sources to PM₁₀ emissions for each group of countries in 1993. The contribution of stationary combustion was around 37% for Western Europe compared with 76% in the CEE countries and 67% in the former USSR. Conversely, the contribution of road transport is about 30% higher in Western Europe.

Table 3.26 shows how the total emissions estimated for the UK in the TNO inventory compare with the UK National Atmospheric Emissions Inventory (NAEI) figures for PM₁₀. As far as the total emissions are concerned, the agreement between the two inventories is remarkably, and probably fortuitously, good considering the uncertainties of each inventory and the different sources of emission factors and activity data used. Emission factors

used for the TNO inventory were generally not country-specific and there are obvious differences between the TNO and NAEI inventories for the UK when examined sector-by-sector. For example, for power generation the NAEI figures (53 ktonnes in 1993) are based on reported particulate emissions from coal and oil-fired stations and a USEPA estimate of the mass fraction emitted from these sources in the PM₁₀ size range. The TNO data for UK power generation (21 ktonnes in 1993) would be based on emission factors and activity data. The TNO estimate for PM₁₀ emissions from road transport in the UK (99 ktonnes in 1993) would not be based on such detailed traffic activity and fleet composition data, nor use UK-vehicle fleet specific emission factors as were used for the NAEI estimate (61 ktonnes in 1993). Another major difference between the two inventories is that TNO include emissions from agricultural activities not included

Table 3.25 European inventory for primary PM_{0.1} in 1993 in ktonnes. Data from TNO (1997).

	Power generation	Industrial combustion	Small combustion	Processes	Road transport	Other transport	Agriculture	Waste processing	Total
Albania	0.0	0.1	0.0	0.1	0.3	0.0	0.0	0.0	0.6
Austria	0.1	0.2	0.7	1.0	2.0	0.2	0.0	0.0	4.1
Belgium	0.1	0.4	1.1	3.5	4.1	0.4	0.0	0.1	9.7
Bulgaria	3.2	0.5	0.3	3.3	2.2	0.1	0.0	0.0	9.4
Switzerland	0.0	0.1	1.2	0.2	0.9	0.0	0.0	0.0	2.5
Former Czechoslovakia	4.5	3.3	2.7	9.0	7.6	0.0	0.0	0.0	27.0
Former GDR	6.8	0.0	0.9	6.0	8.1	0.0	0.0	0.0	22.0
Former Western Germany	1.4	2.1	0.5	12.0	29.0	1.0	0.0	0.1	47.0
Germany	8.2	2.1	1.4	18.0	37.1	1.0	0.0	0.1	69.0
Denmark	0.3	0.1	0.8	0.4	2.1	0.3	0.0	0.0	4.0
Spain	0.8	0.9	1.0	3.7	13.0	2.0	0.0	0.2	22.0
Finland	0.1	0.2	0.6	1.2	2.2	0.2	0.0	0.0	4.5
France	0.1	1.4	4.4	7.5	24.0	1.0	0.0	12.0	50.0
UK	1.8	1.3	1.8	7.0	18.0	2.0	0.0	0.4	33.0
Greece	0.4	0.2	0.6	0.6	4.1	0.7	0.0	0.0	6.7
Hungary	0.9	0.1	0.6	3.8	3.7	0.1	0.0	0.0	9.2
Ireland	0.1	0.1	0.4	0.1	1.1	0.1	0.0	0.0	1.9
Italy	3.5	1.1	2.1	6.9	21.0	1.0	0.0	2.5	38.0
Luxembourg	0.0	0.0	0.1	0.8	0.2	0.0	0.0	0.0	1.1
Norway	0.0	0.1	0.1	1.2	1.0	1.1	0.0	0.0	3.6
Poland	32.0	4.4	3.4	9.4	7.8	0.3	0.0	0.0	57.0
Portugal	0.4	0.2	0.2	0.6	2.5	0.1	0.0	0.0	4.0
Romania	4.7	0.7	0.3	8.2	2.9	0.5	0.0	0.0	17.0
Former USSR (European part only)	66.0	21.0	54.0	150.0	37.0	24.0	0.0	0.0	350.0
Sweden	0.1	0.3	0.6	1.6	1.8	0.2	0.0	0.0	4.6
Former Yugoslavia	4.0	0.8	1.2	5.1	3.1	0.1	0.0	0.0	14.0
Europe, exc. former USSR	65.0	19.0	26.0	93.0	160.0	10.0	0.0	16.0	390.0

Figure 3.10 Emissions of primary PM₁₀ per capita in Europe in 1993 (Data from TNO, 1997).

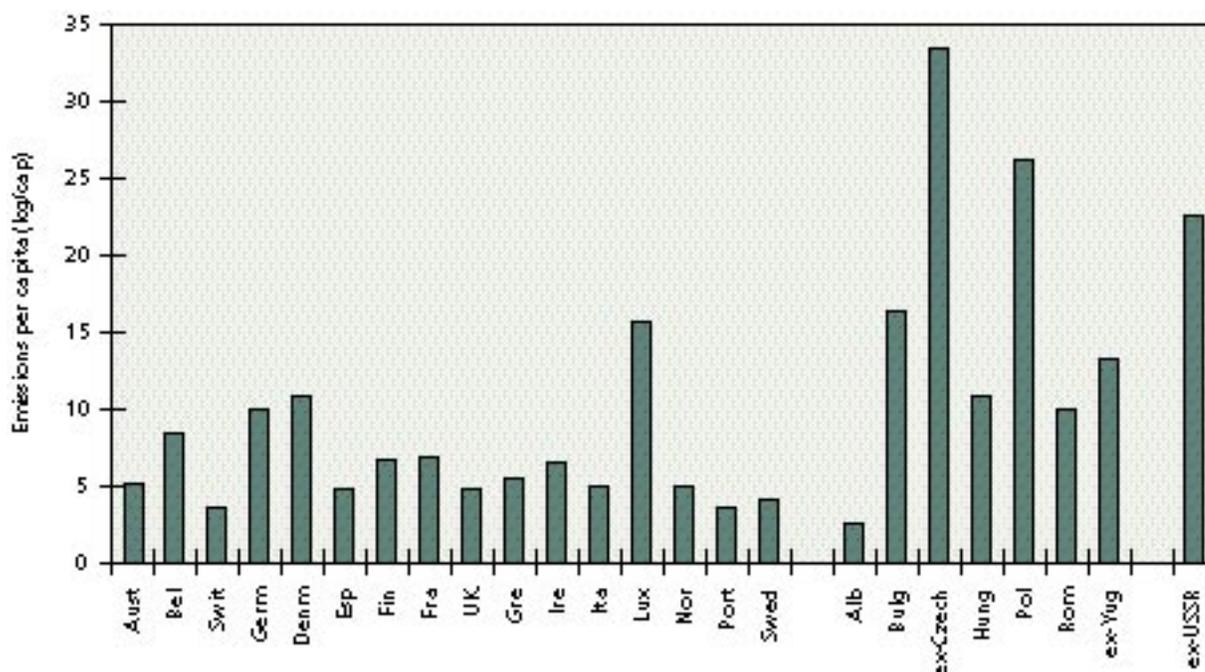


Figure 3.11 Emissions of primary PM₁₀ per capita by region in Europe (Data from TNO, 1997).

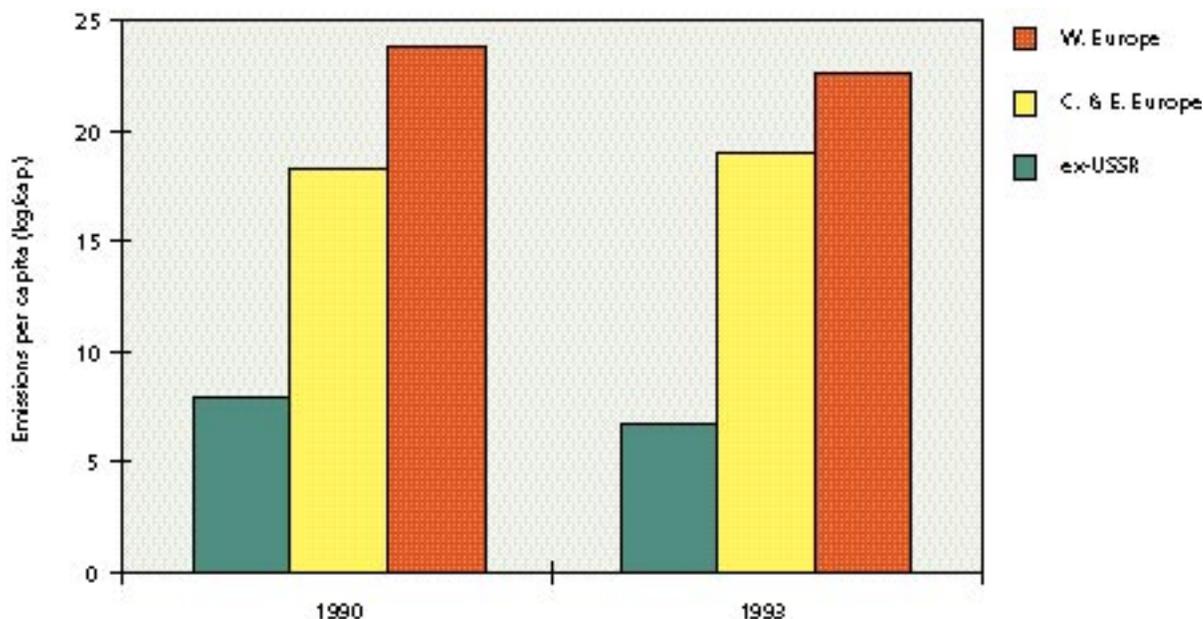
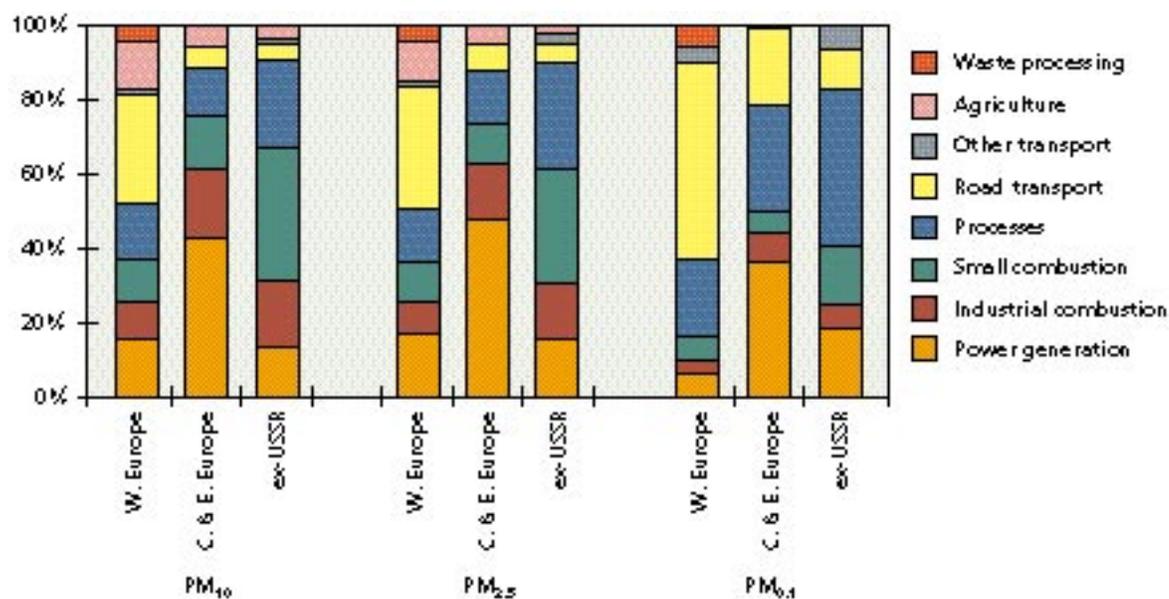


Figure 3.12 Emissions of primary PM₁₀, PM_{2.5} and PM_{0.1} by sectors in Europe in 1993 (Data from TNO, 1997).



in the NAEI because the Dutch-specific emission factors are not considered appropriate for UK agricultural activities. This difference is largely compensated for by the fact that the NAEI estimates much higher emissions from waste treatment in the UK in 1993 than estimated by TNO, due to emissions from old incinerators which have since closed.

Table 3.26 Comparison of TNO and NAEI inventories for PM₁₀ emissions in the UK (ktonnes).

	1990	1993
TNO	270	280
NAEI (1996)	314	293

TNO estimated a standard deviation of 10% for their UK inventory, but this would seem to be optimistic. The uncertainty in the NAEI inventory

could be as much as a factor of two, but the NAEI figures for specific sectors in the UK should be more reliable than the TNO inventory given that the NAEI uses emission factors thought more appropriate for UK sources and more detailed UK statistics for activity data.

In spite of these obvious differences in the two inventories, it is still difficult to explain why the TNO data show PM₁₀ emissions increasing between 1990 and 1993 for the UK, whereas the NAEI figures show a decrease in emissions over this period. The NAEI shows that emissions are declining from all the source sectors, including road transport, albeit at different rates. This is due to the decline in the use of coal as well as the introduction of various abatement measures in stationary combustion processes and road transport. The TNO figures for the UK show that emissions increased from 1990 to 1993 from industrial and small combustion sources and also from road transport, at variance with the NAEI data.

These comparisons suggest that interpretation of data for individual countries in the TNO inventory is probably not valid, but the data are useful for country comparisons and emission mapping at a Europe-wide level.

The contribution of the different sectors to emissions of fine particles, PM_{2.5} and PM_{0.1}, in Europe in 1993 is also shown in Figure 3.12 for comparison with the PM₁₀ data. These clearly show for Western Europe the higher contribution of road transport to the ultra-fine particles.

Table 3.27 shows a comparison of the NAEI and TNO estimates for UK PM₁₀, PM_{2.5} and PM_{0.1} emissions in 1993, excluding agricultural emissions from the TNO inventory as this sector is not included in the NAEI. For 1993, the NAEI gives higher emission rates for the UK than the TNO inventory does for all the particle sizes, the differences becoming proportionately larger for the smaller particle size ranges. However, given the uncertainties and different sources of data

used for both inventories, these differences are not irreconcilable.

Table 3.27 Comparison of TNO and NAEI Inventories for PM₁₀, PM_{2.5} and PM_{0.1} Emissions in the UK in 1993. Emissions from agriculture excluded from the TNO inventory as this source is not included in the NAEI (ktonnes).

	PM ₁₀	PM _{2.5}	PM _{0.1}
TNO	249	147	33
NAEI (1996)	293	189	46

3.3 INVENTORY VERIFICATION

An evaluation of emission inventories with respect to measured air quality can provide a semi-quantitative verification of the emission estimates. The relationship between emissions from major point sources, such as power stations, and measured air quality is best investigated using dispersion models. The relationship between measured air quality and emissions inventory estimates of low-level area and line sources is however more straightforward as low-level emissions impact on air quality at close range. This type of comparison has enabled the validity of the spatial distribution of these sources for a range of pollutants within the National Atmospheric Emissions Inventory to be investigated (Goodwin *et al.*, 1997). The local emissions inventories prepared by the London Research Centre (LRC) have provided estimates of PM₁₀ emissions at a 1 km x 1 km grid scale for the first time:

- *West Midlands (Hutchinson and Clewley, 1996)*
- *Greater Manchester (Buckingham *et al.*, 1997a)*
- *London (Buckingham *et al.*, 1998a)*
- *Merseyside (Buckingham *et al.*, 1997b)*
- *Bristol (Buckingham *et al.*, 1997b)*
- *Southampton and Portsmouth (Buckingham *et al.*, 1997b)*
- *Swansea and Port Talbot (Buckingham *et al.*, 1997b). Data not available for this comparison*
- *Glasgow and Middlesbrough (Buckingham *et al.*, 1998b). Data not available for this comparison.*

These local and national emission estimates are compared with ambient PM₁₀ measurements in this section.

Measured PM₁₀ concentrations (annual means) can be considered to be made up of three parts:

- *A contribution from local primary emissions. Work on the estimation of air pollutant concentrations from emission related parameters (Stedman et al., 1997) studied the spatial scale at which local emissions seem to influence urban background ambient air quality has been studied. Estimates of emissions in an area of 25 km² centred on the monitoring site were found to provide the most robust relationships with ground level concentrations. Emissions from the 25 individual 1 km x 1 km grid squares surrounding each automatic monitoring site have therefore been summed to provide an estimate of local emissions.*
- *Secondary particles. The concentration of secondary particles for 1996 at each automatic monitoring site has been estimated by interpolation from measurements of annual mean sulphate concentrations at eight rural sites (see Section 4.2.1).*
- *'Other' particles. The annual mean concentration of 'other' particles is assumed to be roughly constant at all sites and is represented by the intercept of the graphs presented here.*

Measured urban background ambient background annual average PM₁₀ concentrations are given by the following expression:

$$\text{Measured PM}_{10} (\mu\text{g m}^{-3}) = \text{secondary PM}_{10} (\mu\text{g m}^{-3}) + k_1 \times \text{emissions (Tonnes per 25 km}^2 \text{ per year)} + C (\mu\text{g m}^{-3})$$

where: $k_1 \times$ emission represents primary particle concentration, *secondary PM₁₀* represents secondary particle concentration, and *C* represents the coarse particle concentration (see Section 5.6)

Figures 3.13 to 3.16 show comparisons of measured annual mean PM₁₀ concentrations with emissions estimates for urban background (not roadside or industrially influenced or coal burning influenced) sites for which emissions estimates are available from LRC. PM₁₀ measurement data for 1996 were used where available, otherwise data for other years were used. These sites are listed in Table 3.28 along with the identification number used in the graphs. All of the sites are from the national monitoring networks except for the Joint Environment Programme site at Hall Farm for which data were provided by National Power.

Table 3.28 PM₁₀ urban background measurement sites used for inventory verification.

Site name	Year	Ident. no.
London Bloomsbury	1996	31
Birmingham Centre	1996	35
Bristol Centre	1996	38
Liverpool Centre	1996	39
Birmingham East	1996	40
Southampton Centre	1996	43
London Bexley	1996	44
Manchester Piccadilly	1996	47
Wolverhampton Centre	1997	49
London Brent	1997	50
Kensington and Chelsea 1	1997	54
London Eltham	1997	60
London Hillingdon	1997	62
Stockport	1997	69
Hall Farm, Essex	1995	88
Bolton	1997	188
Salford Eccles	1997	189

Table 3.29 lists the regression coefficients, intercepts and correlation coefficients for the relationships between emissions estimates and

Table 3.29 Regression coefficients, intercepts and correlation coefficients for the relationships between emissions estimates and ambient PM₁₀ concentrations.

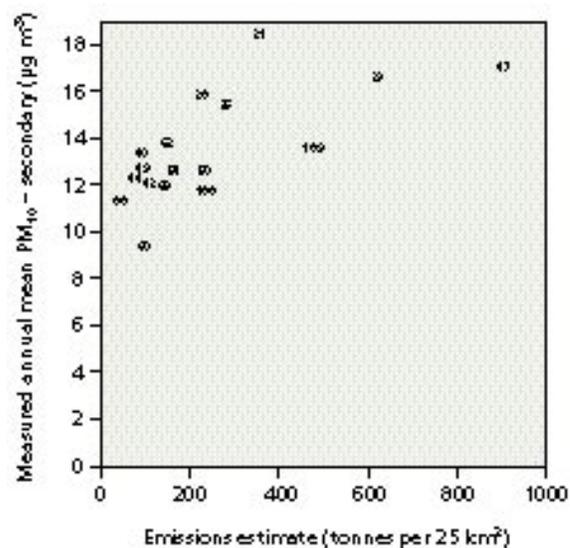
LRC emission estimates	Figure no.	Sites excluded	k_1	<i>C</i>	<i>r</i>
all emissions	3.13	none	0.007	11.8	0.69
all emissions except Part 'A' processes	not shown	Liverpool	0.020	9.9	0.75
all emissions except Parts 'A' and 'B' processes	3.14	Liverpool	0.024	9.8	0.81
road transport only	3.15	Liverpool	0.033	9.5	0.81
NAEI road transport only	3.16	Kensington	0.040	9.9	0.64

measured concentrations (with the secondary particle concentration subtracted) i.e.:

$$\text{Measured PM}_{10} (\mu\text{g m}^{-3}) - \text{secondary PM}_{10} (\mu\text{g m}^{-3}) = k_1 \times \text{emissions (Tonnes per 25 km}^2 \text{ per year)} + C (\mu\text{g m}^{-3}).$$

Figure 3.14 shows the good agreement between the spatial distribution of all emissions except Part 'A' and Part 'B', i.e. low level emissions, and primary particle concentrations. The large intercept of about $10 \mu\text{g m}^{-3}$ represents the sum of UK regional and local coarse particle concentrations and any primary particles from sources which have not been represented by the inventory. Emissions of primary particles from elevated sources, such as Part 'A' and Part 'B' regulated processes do have an impact on annual mean ground level ambient PM_{10} concentrations. Liverpool Centre, in particular, has a higher PM_{10} than would be expected due to low level sources alone. In general, the contribution that these emissions make to the annual mean concentration is lower per tonne emitted than for low-level emissions (compare Figures 3.13 and 3.14), due to more effective dispersion and dilution in the atmosphere before the emissions come to ground.

Figure 3.13 Comparison of annual mean PM_{10} , urban background PM_{10} concentrations minus annual mean secondary, with London Research Centre PM_{10} emission inventory: all sources.



Whilst the validity of the spatial distribution of the emission estimates is supported by the correlation coefficient, the reliability of the absolute magnitude of the emission estimates can be assessed by considering the coefficient, k_1 . The coefficient obtained from a similar comparison of low-level emissions estimates of a range of pollutants from the NAEI with ambient background concentrations (Stedman, 1998) were typically in the range 0.018 to 0.034, expressed in the same units. The NAEI estimates of low level emissions exclude most Part 'A' processes but include the majority of Part 'B' processes. Chamberlain *et al.* (1979) calculated a value for k_1 by considering the annual average concentration at the centre of a circular urban area of specified radius containing ground-level emissions with a uniform source strength. For an urban area the size of Birmingham and meteorological conditions typical of central England a value of 0.026 for k_1 was obtained in good agreement with the empirically derived factors. The values for the coefficients obtained for the local inventory data are therefore in reasonably good agreement with these other empirically or theoretically derived coefficients, indicating that the magnitudes of the emissions estimates are fairly accurate. If the magnitude of low level emissions is

Figure 3.14 Comparison of annual mean PM_{10} , urban background PM_{10} concentrations minus annual mean secondary, with London Research Centre PM_{10} emission inventory: all sources except Part 'A' and 'B'.

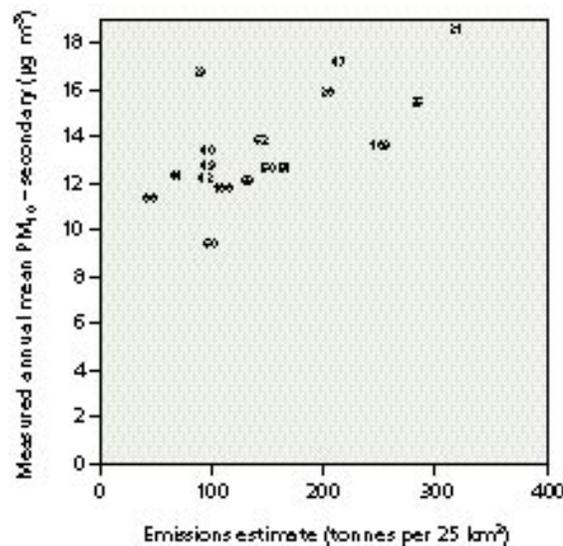
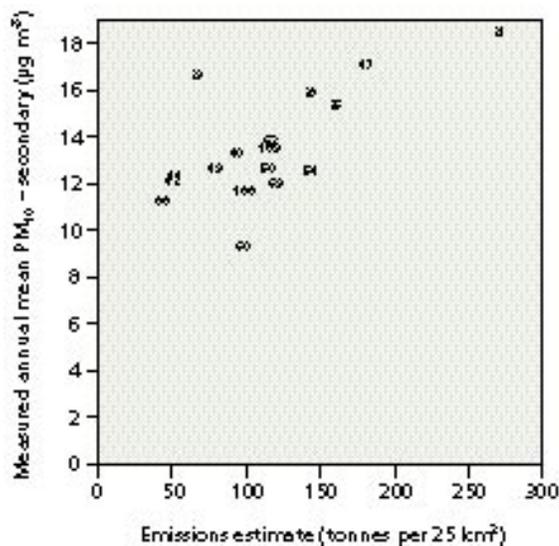


Figure 3.15 Comparison of annual mean PM_{10} , urban background PM_{10} concentrations minus annual mean secondary, with London Research Centre PM_{10} emission inventory: road transport.

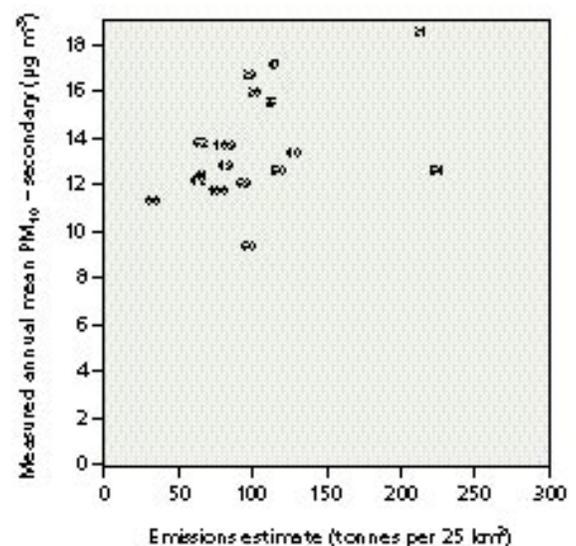


underestimated this leads to an unrealistically high k_1 coefficient as obtained by the regression analysis for traffic emissions only. An anomalously low k_1 coefficient is obtained if emission for high level point sources are included because of the smaller contribution that high sources make to ground level PM_{10} sources per tonne of emission. k_1 coefficients closest to the theoretical value are obtained if Part 'A' process emissions are not included in the regression analysis.

Figure 3.16 shows that the relationship between the NAEI nationally derived estimates of road transport emissions for PM_{10} and ambient concentrations is weaker than for the locally derived road transport emission estimates. This is to be expected since the locally derived inventories will have incorporated emission estimates from a more complete major road network, leaving less of the minor road emissions to be distributed according to surrogate statistics such as population.

Not only does this analysis confirm the quality of the local inventories, it shows clearly the impact of local road traffic emissions upon annual average PM_{10} concentrations at urban sites. It should be noted, however, that only the emission estimates for locations within a few km of automatic

Figure 3.16 Comparison of annual mean PM_{10} , urban background PM_{10} concentrations minus annual mean secondary, with National Atmospheric PM_{10} Emissions Inventory: road transport.



monitoring sites has been examined. The quality of the local inventory estimates at places more distant from monitoring sites cannot be assessed using this method.

3.4 TRANSPORT OF PRIMARY PARTICULATE MATTER FROM MAINLAND EUROPE TO THE UK

3.4.1 MODELLING THE LONG-RANGE TRANSPORT OF PRIMARY PARTICULATE MATTER

A simple trajectory model has been developed to assess the imported contribution of primary particulate emissions in Europe to PM_{10} concentrations in the UK. This has been used with the European inventory compiled by TNO and described in Section 3.2.7. The particulate material is divided into three different size categories in accordance with the PM_{10} , $PM_{2.5}$ and $PM_{0.1}$ classification though the latter contributes little to mass. The model has been used in a receptor oriented mode with straight line trajectories in different sectors weighted according to a geostrophic wind rose. A constant mixing layer depth of 800 metres was assumed, and a mean wind of 8 m s^{-1} . The emissions have been mapped

on to the EMEP grid for each country (assuming the same approximate spatial distribution within a country as for NO_x); this makes it possible to estimate the relative contributions from different countries. After emission into an air column, dry and wet deposition processes are introduced at different rates depending on particle size (dry deposition velocities of 0.1 cm s^{-1} for the finer $\text{PM}_{2.5}$ fraction, and 1.5 cm s^{-1} for the 2.5 to $10 \text{ }\mu\text{m}$ component). Wet deposition is modelled statistically with transition between dry periods (average length 54 hours) and wet periods (average length 6 hours) in accordance with the approach adopted by Smith (1998).

The resulting modelled annual average concentrations of imported primary PM_{10} are in the range 0.8 to $2 \text{ }\mu\text{g m}^{-3}$ over most of the UK, with the highest values in the south-east. This is reflected in Table 3.30 where a break-down of these concentrations for different size classes is given for London and Edinburgh. The largest contributions appear to come from Germany, followed by France and Poland.

Table 3.30 Modelled annual average concentrations of primary particulate material imported from Europe based on 1992 emissions data.

	PM_{10} $\mu\text{g m}^{-3}$	$\text{PM}_{2.5}$ $\mu\text{g m}^{-3}$	$\text{PM}_{0.1}$ $\mu\text{g m}^{-3}$
London	1.9	1.6	0.27
Edinburgh	1.0	0.95	0.16

NB The $\text{PM}_{0.1}$ is overestimated in that the transfer to larger particles by coagulation is not yet included.

Because of the position of the UK on the North-western coast of Europe, the concentrations are highly dependent on wind direction and when trajectories have travelled across major emission sources in Europe. Thus the model implies much higher imported primary particulate concentrations with winds from the east and south-east sectors as compared with the annual average values. Thus values for individual wind sectors are as high as $15 \text{ }\mu\text{g m}^{-3}$ for imported PM_{10} over London and $6 \text{ }\mu\text{g m}^{-3}$ for Edinburgh. Although the model is very simple, this clearly implies that

primary PM_{10} imported from Europe can be an important contribution in anticyclonic situations in relation to the UK air quality standard for PM_{10} of $50 \text{ }\mu\text{g m}^{-3}$ 24-hour running mean.

An example of the relative contributions from local roads, and the background concentrations from city sources and longer range contributions in air imported into London in such episodic conditions, is provided by recent modelling studies (H. ApSimon, Imperial College, London). These concern primary PM_{10} in anticyclonic conditions with low wind speeds giving residence times of 12 or more hours for air over London below a persistent inversion at 200 metres. In such conditions the imported contribution from Europe, using the simple trajectory model and TNO European emission inventory as in section 3.4, implies concentrations of the order of 15 to $30 \text{ }\mu\text{g m}^{-3}$, equivalent to a 24 hour averaging period. The contribution from London emissions using the URBPM model (similar to the URBNOX model of O’Keeffe, 1997) to simulate dispersion of London emissions above the city in these inversion conditions, implies an additional 30 to $40 \text{ }\mu\text{g m}^{-3}$ (equivalent to a 24 hour averaging period) at street level. These may be compared with the contribution close to a major road such as the Cromwell Road, with traffic flows of the order of 10,000 vehicles per hour. Here a local scale model, such as the CALINE 4 model, predicts road side concentrations of around $12 \text{ }\mu\text{g m}^{-3}$ in average meteorological conditions, but more complex Computational Fluid Dynamics modelling of street canyons and junctions implies localised hot-spots of concentration up to an order of magnitude higher.

These results apply only to primary PM_{10} and ignore secondary particulate contributions. However they clearly show how even primary PM_{10} alone can lead to episodes of high concentration (i.e in excess of $50 \text{ }\mu\text{g m}^{-3}$) and emphasise the importance of the background contribution, both imported from outside the city, and generated from emissions within a large city such as London, in addition to local sources.

3.4.2 ANALYSIS OF RURAL BLACK SMOKE MEASUREMENTS IN NORFOLK

Further indications of the likely contribution of long range transport of primary particles to measured PM₁₀ concentrations are available from measurements of black smoke in rural areas. The local sources of primary combustion particles in rural areas are expected to be small relative to more distant sources. This can be examined from the daily measurements of black smoke concentrations at Bacton which is a rural site close to the coast of Norfolk where measurements have been carried out by Phillips Petroleum Company.

A comparison of the daily measurements of black smoke concentrations for 1996 at this site with an average value for London, calculated as the mean of all black smoke monitoring sites in London, is shown in Figure 3.17. Black smoke concentrations are, as expected, generally higher in the winter than in the summer. The highly elevated black smoke concentrations in London on 15th and 16th January are much higher than those recorded at Bacton. The prolonged periods with black smoke concentrations in the range 20 to 30 µg m⁻³ in late January and mid March are however seen at both Bacton and London, with the day to day variation being very similar. These periods were characterised by easterly winds and corresponded

with significantly elevated secondary particle concentrations (see section 5.6). The elevated black smoke concentrations measured in London during November and December 1996 did not coincide with strongly elevated concentrations at Bacton.

An analysis of the mean black smoke concentrations at Bacton for each transport direction is shown in Table 3.31. The transport direction was defined by calculating 96-hour back trajectories for each day and assigning 45 degree sectors on the basis of the dominant transport direction. It is clear that the highest concentration days at this site are associated with transport from the east with mean concentrations for the east and south east directions being of the order of 10 µg m⁻³. Indeed, during the January to March 1996 period daily mean black smoke concentrations at Bacton were in excess of 15 µg m⁻³ on 12 days when the transport direction was invariably between north east and south east. These easterly days contribute about 1.5 µg m⁻³ to the annual mean black smoke concentration of 4 µg m⁻³ at this site. This analysis is consistent with modelling results for the long range transport of primary particles from European sources (see section 3.4.1), with the limitation that black smoke measurements can only be considered as indicative of primary PM₁₀ concentrations.

Figure 3.17 Comparison of daily mean black smoke concentrations at Bacton (Norfolk) and London (average of all monitoring sites) during 1996 (µg m⁻³).

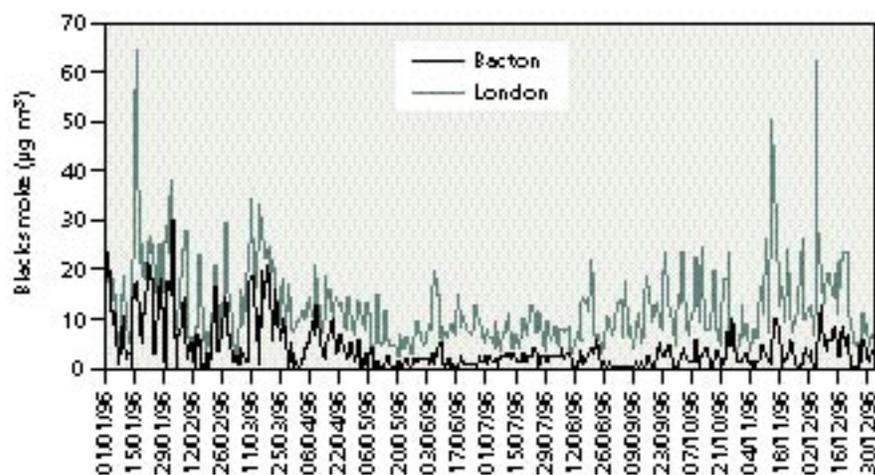


Table 3.31 Sector analysis of black smoke concentrations at Bacton for 1996 ($\mu\text{g m}^{-3}$).

	Mean concentration	Number of days
N	2.98	56
NE	2.60	24
E	9.64	34
SE	11.00	11
S	6.18	18
SW	2.85	33
W	2.20	50
NW	1.88	38
UNDEFINED	3.21	100

3.5 ANALYSIS OF $\text{PM}_{2.5}$ / PM_{10} DATA

As a step to source apportionment of atmospheric measurements, a useful first subdivision is into separate estimates of $\text{PM}_{2.5}$, which arises predominantly from combustion sources and high temperature industrial processes, and coarse particles. These coarse particles are the fraction between PM_{10} and $\text{PM}_{2.5}$, and have sources associated with mechanical disintegration processes

which include such activities as quarrying and building construction, as well as natural contributors such as sea spray, wind blown soil and surface dust and fungal spores. In the Third Report of QUARG, an analysis was conducted of the then rather small data set from the TEOM instrument at the Birmingham, Hodge Hill site operated by Birmingham City Council. The site is located close to ground level, horizontally 60m from the edge of the elevated section of the M6 motorway in north Birmingham. Because of the prevailing wind directions, the site is frequently unaffected by the motorway, and even when downwind, much of the motorway-derived pollution carries above the site. Therefore measurements of PM_{10} and NO_x show the pollution climate of the site to be broadly similar to that of the East Birmingham and Birmingham Central stations of the Automatic Urban Network, and the site differs little from the urban background category in terms of these pollutants. Data from the Hodge Hill site appear in Table 3.32, summarizing a full three-year period from October 1994 to September 1997. Table 3.33 shows the results of regression analysis of both daily and

Table 3.32 Summary of Concentration data for PM_{10} , $\text{PM}_{2.5}$, $\text{PM}_{\text{coarse}}$, and NO_x at Birmingham, Hodge Hill, October 1994 - September 1997.

		PM_{10} ($\mu\text{g m}^{-3}$)	$\text{PM}_{2.5}$ ($\mu\text{g m}^{-3}$)	Coarse ($\mu\text{g m}^{-3}$)	NO_x (ppb)
Whole period	1/10/94-30/11/97	22.5	14.4	8.2	53.2
Annual	1/10/94-31/8/95	22.8	13.5	9.4	55.3
	1/9/95-31/8/96	23.8	15.7	8.2	52.4
	1/9/96-31/8/97	21.0	14.0	6.9	52.0
Season (aggregating whole period)	Autumn (94,95,96&97)	22.4	15.1	7.4	60.7
	Winter (94/95,95/96&96/97)	20.9	14.5	6.5	61.5
	Spring (95,96&97)	24.5	15.8	8.6	52.1
	Summer (95,96&97)	22.3	12.9	9.4	40.9
Single Season	Autumn (10/94-11/94)	25.2	18.3	7.1	70.3
	Winter (12/94-02/95)	17.0	12.0	5.4	57.7
	Spring (03/95-05/95)	24.9	14.4	10.4	56.9
	Summer (06/95-08/95)	25.4	12.3	13.2	45.3
	Autumn (09/95-11/95)	23.2	14.7	8.7	59.3
	Winter (12/95-02/96)	26.3	17.6	8.8	63.8
	Spring (03/96-05/96)	24.5	17.5	6.9	47.1
	Summer (06/96-08/96)	21.3	13.0	8.4	39.8
	Autumn (09/96-11/96)	21.0	13.2	7.6	55.9
	Winter (12/96-02/97)	19.3	14.0	5.4	63.0
	Spring (03/97-05/97)	24.1	15.6	8.4	52.5
	Summer (06/97-08/97)	20.3	13.6	6.7	37.5
Autumn (09/97-11/97)	20.2	14.3	6.1	57.4	

Table 3.33 Result of regression analyses of PM_{2.5} and PM₁₀ at Birmingham, Hodge Hill, October 1994 - September 1996.

Daily Data		PM _{2.5} = m PM ₁₀ + c	r	PM _{coarse} = m PM ₁₀ + c	r
Annual-1	1/10/94-31/8/95	PM _{2.5} = 0.63 PM ₁₀ - 0.68	0.87	PM _{coarse} = 0.38 PM ₁₀ + 0.72	0.75
Annual-2	1/09/95-31/8/96	PM _{2.5} = 0.77 PM ₁₀ - 2.73	0.95	PM _{coarse} = 0.24 PM ₁₀ + 2.58	0.69
Annual-3	1/09/96-31/8/97	PM _{2.5} = 0.72 PM ₁₀ - 0.99	0.95	PM _{coarse} = 0.28 PM ₁₀ + 0.99	0.76
Season (aggregating three years)	Autumn (94,95,96&97)	PM _{2.5} = 0.76 PM ₁₀ - 2.17	0.95	PM _{coarse} = 0.25 PM ₁₀ + 1.98	0.71
	Winter (94/95,95/96&96/97)	PM _{2.5} = 0.75 PM ₁₀ - 1.15	0.97	PM _{coarse} = 0.25 PM ₁₀ + 1.33	0.79
	Spring (95,96&97)	PM _{2.5} = 0.77 PM ₁₀ - 2.64	0.91	PM _{coarse} = 0.25 PM ₁₀ + 2.31	0.60
	Summer (95,96&97)	PM _{2.5} = 0.53 PM ₁₀ + 1.10	0.86	PM _{coarse} = 0.47 PM ₁₀ - 1.06	0.83
Hourly Data					
Annual-1	1/10/94-31/8/95	PM _{2.5} = 0.61 PM ₁₀ - 0.38	0.86	PM _{coarse} = 0.39 PM ₁₀ + 0.40	0.74
Annual-2	1/09/95-31/8/96	PM _{2.5} = 0.73 PM ₁₀ - 2.06	0.94	PM _{coarse} = 0.26 PM ₁₀ + 2.07	0.70
Annual-3	1/09/96-31/8/97	PM _{2.5} = 0.70 PM ₁₀ - 0.60	0.94	PM _{coarse} = 0.30 PM ₁₀ + 0.60	0.77
Season (aggregating three years)	Autumn (94,95,96&97)	PM _{2.5} = 0.78 PM ₁₀ - 2.57	0.96	PM _{coarse} = 0.22 PM ₁₀ + 2.58	0.68
	Winter (94/95,95/96&96/97)	PM _{2.5} = 0.72 PM ₁₀ - 0.72	0.95	PM _{coarse} = 0.27 PM ₁₀ + 0.74	0.75
	Spring (95,96&97)	PM _{2.5} = 0.69 PM ₁₀ - 0.87	0.89	PM _{coarse} = 0.31 PM ₁₀ + 0.87	0.66
	Summer (95,96&97)	PM _{2.5} = 0.47 PM ₁₀ + 2.54	0.83	PM _{coarse} = 0.53 PM ₁₀ - 2.55	0.86

hourly data for PM_{2.5} and PM₁₀, both of which were measured with TEOM instruments. The data are broadly consistent with those presented in the Third Report of QUARG showing that within individual seasons PM_{2.5} and PM₁₀ are strongly correlated but the percentage of PM₁₀ comprised by PM_{2.5} shows a strong seasonal dependence. This is seen clearly in gradients of the relationships of PM_{2.5} and PM₁₀ and between PM_{coarse} and PM₁₀ in which the proportion of coarse particles is greatest in the spring and summer, and least in the wetter autumn and winter seasons. In the Third Report of QUARG, this was interpreted as being most probably the result of a better dispersion of pollutant emissions in the hotter months leading to lower concentrations of PM_{2.5} and more effective wind-driven suspension of coarse dusts in the dryer months of the year leading to higher concentrations of PM_{coarse}. An examination of the data in Table 3.32 shows the latter effect clearly and indicates that inter-annual variability in the weather can have strong influences on PM_{2.5} causing the large difference in winter mean between 1994/95 (11.84 µg m⁻³) and 1995/96 (17.39 µg m⁻³). By far the

highest seasonal mean for coarse particles was that in summer 1995 (13.04 µg m⁻³) which comprised 52% of the summer average PM₁₀ summer concentration for that year.

Since the summer of 1997, the Department of Environment, Transport and the Regions has funded the operation of four additional sites measuring both PM_{2.5} and PM₁₀. These are as follows:

- London, Marylebone Road. This is the so-called "super-site" located within the street canyon on this busy central London highway.
- London, Bloomsbury. A central urban background location within a small park in the centre of a heavily trafficked square.
- Rochester. A site in Kent to the east of London generally characterised as suburban, but frequently showing a relatively rural pollution climate.
- Harwell. On the site of the research laboratory in rural Oxfordshire but subject at times to influence of the London urban plume and the adjacent busy A34 highway.

Table 3.34 Summary of concentration data for PM₁₀, PM_{2.5}, PM_{coarse} and NO_x at sites operated by DETR.

	PM ₁₀ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)	Coarse (µg m ⁻³)	NO _x (ppb)
<i>Whole period</i>				
LM (20/6/97 - 28/2/98)	37.90	23.57	13.88	221.91
LB (20/6/97 - 28/2/98)	26.12	18.20	8.85	87.61
RO (20/6/97 - 28/2/98)	19.32	14.18	6.32	20.53
HAR (28/9/97 - 28/2/98)	16.60	12.50	4.47	20.68
<i>Summer period</i>				
LM (20/6/97 - 31/8/97)	40.73	24.57	14.30	188.51
LB (20/6/97 - 31/8/97)	26.34	19.14	8.42	61.76
RO (20/6/97 - 31/8/97)	21.34	14.71	8.07	12.88
HAR (20/6/97 - 31/8/97)	-	-	-	-
<i>Autumn period</i>				
LM (1/9/97 - 30/11/97)	39.59	23.34	14.30	245.34
LB (1/9/97 - 30/11/97)	28.10	19.67	10.12	104.91
RO (1/9/97 - 30/11/97)	18.84	13.77	5.92	22.17
HAR (28/9/97 - 30/11/97)	16.63	13.29	4.34	24.17
<i>Winter period</i>				
LM (1/12/97 - 28/2/98)	34.51	22.80	13.16	218.06
LB (1/12/97 - 28/2/98)	23.84	15.82	7.85	90.58
RO (1/12/97 - 28/2/98)	17.97	14.23	5.08	25.06
HAR (1/12/97 - 28/2/98)	16.57	12.04	4.54	18.67

LM - London Marylebone; LB - London Bloomsbury; RO - Rochester; HAR - Harwell; '-' indicates that data is not available.

Table 3.35 Result of regression analyses of PM_{2.5} and PM₁₀ data at sites operated by DETR for Hourly averages.

	PM _{2.5} = m PM ₁₀ + c	r	PM _{coarse} = m PM ₁₀ + c	r
<i>Whole period</i>				
LM (20/6/97 - 28/2/98)	PM _{2.5} = 0.67 PM ₁₀ - 1.40	0.86	PM _{coarse} = 0.33 PM ₁₀ + 1.41	0.60
LB (20/6/97 - 28/2/98)	PM _{2.5} = 0.58 PM ₁₀ + 2.83	0.66	PM _{coarse} = 0.46 PM ₁₀ - 3.78	0.60
RO (20/6/97 - 28/2/98)	PM _{2.5} = 0.57 PM ₁₀ + 2.48	0.59	PM _{coarse} = 0.44 PM ₁₀ - 2.41	0.49
HAR (28/9/97 - 28/2/98)	PM _{2.5} = 0.81 PM ₁₀ - 1.27	0.90	PM _{coarse} = 0.19 PM ₁₀ + 1.27	0.33
<i>Summer period</i>				
LM (20/6/97 - 31/8/97)	PM _{2.5} = 0.69 PM ₁₀ - 1.67	0.84	PM _{coarse} = 0.31 PM ₁₀ + 1.67	0.53
LB (20/6/97 - 31/8/97)	PM _{2.5} = 0.59 PM ₁₀ + 3.07	0.71	PM _{coarse} = 0.41 PM ₁₀ - 2.69	0.57
RO (20/6/97 - 31/8/97)	PM _{2.5} = 0.63 PM ₁₀ + 0.26	0.66	PM _{coarse} = 0.38 PM ₁₀ - 0.30	0.43
HAR (20/6/97 - 31/8/97)	-	-	-	-
<i>Autumn period</i>				
LM (1/9/97 - 30/11/97)	PM _{2.5} = 0.65 PM ₁₀ - 1.24	0.86	PM _{coarse} = 0.35 PM ₁₀ + 1.25	0.63
LB (1/9/97 - 30/11/97)	PM _{2.5} = 0.54 PM ₁₀ + 3.80	0.63	PM _{coarse} = 0.48 PM ₁₀ - 4.05	0.61
RO (1/9/97 - 30/11/97)	PM _{2.5} = 0.48 PM ₁₀ + 4.20	0.51	PM _{coarse} = 0.53 PM ₁₀ - 4.15	0.58
HAR (28/9/97 - 30/11/97)	PM _{2.5} = 0.80 PM ₁₀ - 0.84	0.92	PM _{coarse} = 0.20 PM ₁₀ + 0.86	0.41
<i>Winter period</i>				
LM (1/12/97 - 28/2/98)	PM _{2.5} = 0.67 PM ₁₀ - 1.35	0.87	PM _{coarse} = 0.33 PM ₁₀ + 1.37	0.61
LB (1/12/97 - 28/2/98)	PM _{2.5} = 0.68 PM ₁₀ + 0.12	0.70	PM _{coarse} = 0.32 PM ₁₀ + 0.32	0.36
RO (1/12/97 - 28/2/98)	PM _{2.5} = 0.73 PM ₁₀ - 0.12	0.75	PM _{coarse} = 0.26 PM ₁₀ + 0.39	0.30
HAR (1/12/97 - 28/2/98)	PM _{2.5} = 0.82 PM ₁₀ - 1.48	0.89	PM _{coarse} = 0.18 PM ₁₀ + 1.48	0.30

Table 3.34 shows a summary of the concentration data measured at these sites, disaggregated where possible by season. The effect of road traffic is very clearly seen in the substantial elevations of both PM_{2.5} and PM₁₀ at London Marylebone Road, relative to the nearby London Bloomsbury urban background site. Over an eight month period an

elevation of approximately 12 µg m⁻³ of PM₁₀ and 5.5 µg m⁻³ in PM_{2.5} is observed. If a notional regional background of about 10 µg m⁻³ of secondary PM_{2.5} is subtracted, the local elevation at Marylebone Road is seen to be very appreciable (almost 50% of primary pollutant background). Perhaps most notable is the substantial increment in

Figure 3.18 Comparison of daily mean PM₁₀ and NO_x concentrations at roadside and background sites, June 1997 - September 1998 ($\mu\text{g m}^{-3}$, contains provisional data).

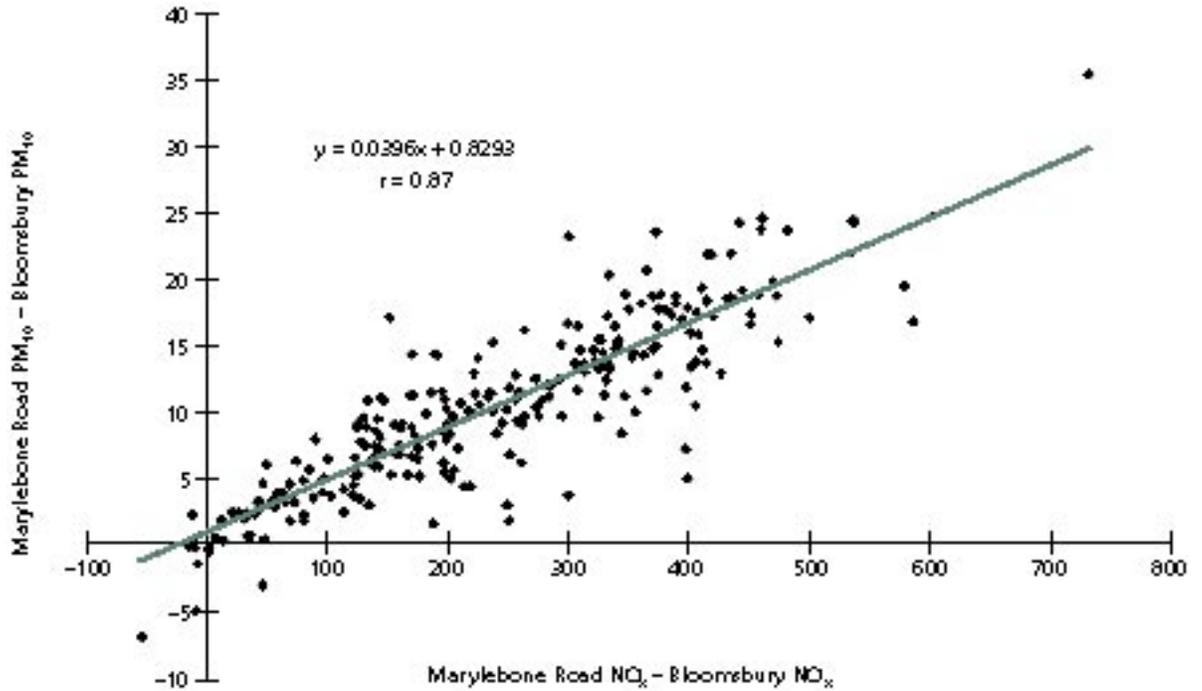


Figure 3.19 Comparison of daily mean PM_{2.5} and NO_x concentrations at roadside and background sites, June 1997 - September 1998 ($\mu\text{g m}^{-3}$, contains provisional data).

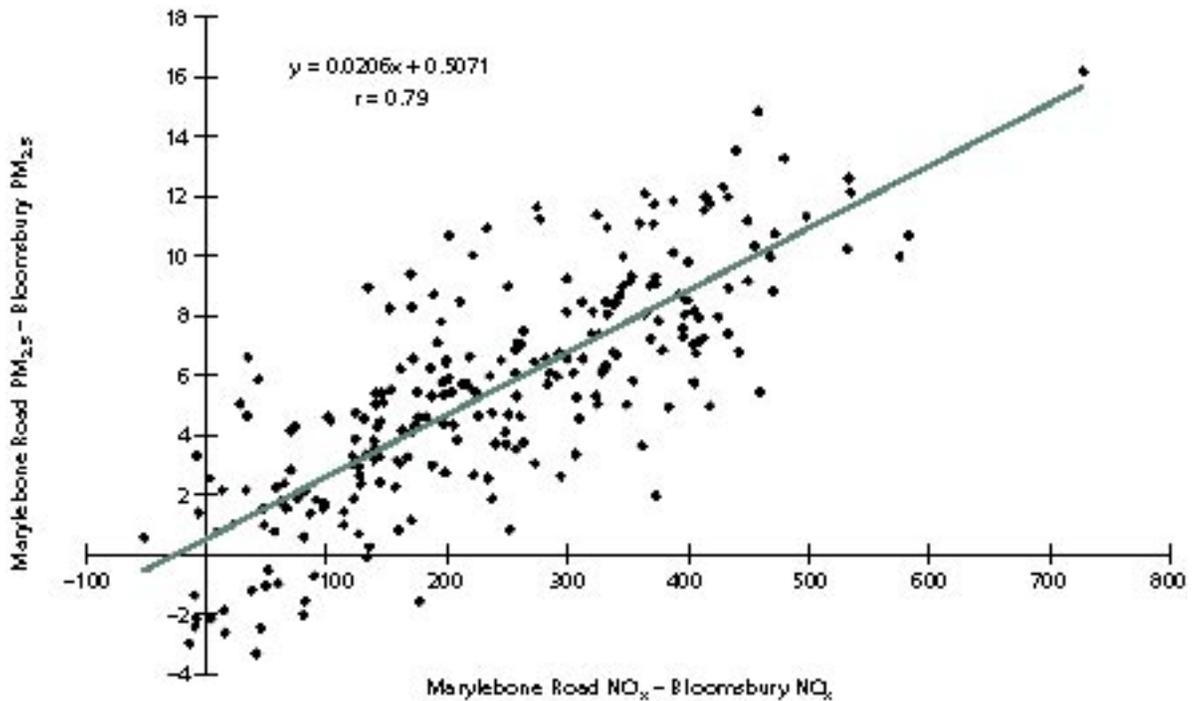
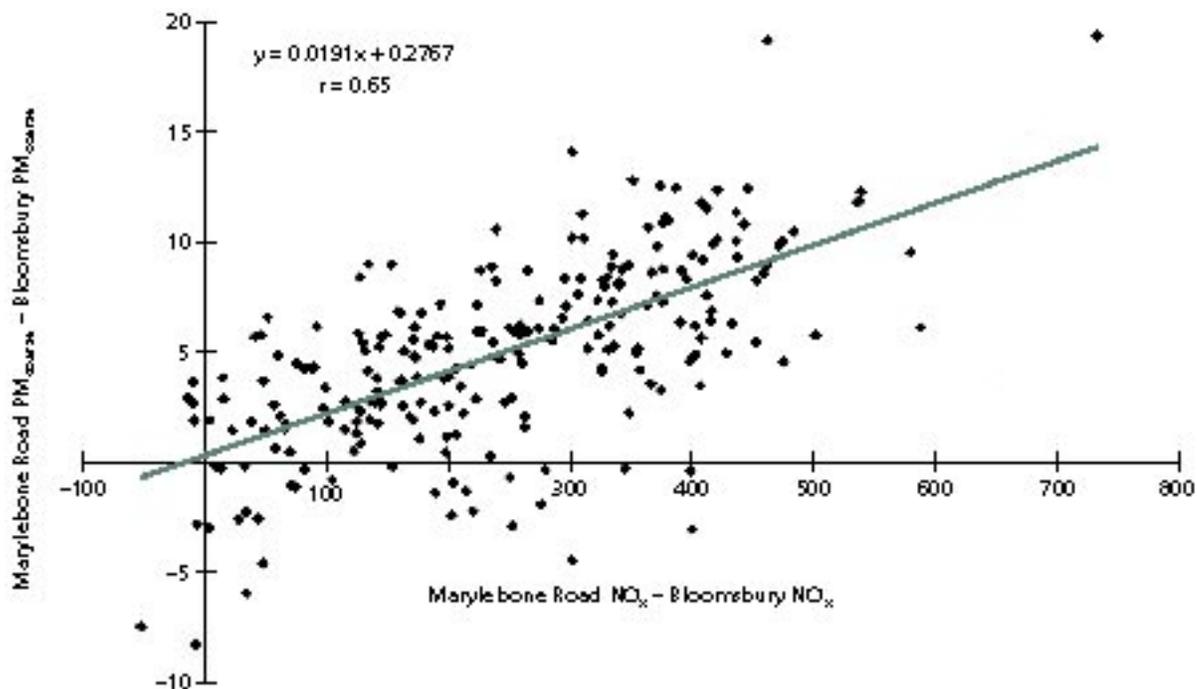


Figure 3.20 Comparison of daily mean PM_{coarse} and NO_x concentrations at roadside and background sites, June 1997 - September 1998 ($\mu g\ m^{-3}$, contains provisional data).



coarse particles between London Marylebone Road and London Bloomsbury. This is attributable to resuspension due to traffic-induced turbulence at the road-side location and is analysed further in Section 3.5.1. Comparing London Bloomsbury with Rochester and Harwell, the impact of local sources of both fine ($PM_{2.5}$) and coarse particles within London is very clearly seen.

Table 3.35 shows correlations between $PM_{2.5}$ and PM_{10} and between PM_{coarse} and PM_{10} at the four sites derived from hourly data. These demonstrate the generally strong correlation between $PM_{2.5}$ and PM_{10} in both the seasons for which measurements are currently available.

3.5.1 COMPARISON OF DAILY MEAN CONCENTRATIONS OF PM_{10} AND $PM_{2.5}$ AND NO_x AT MARYLEBONE ROAD

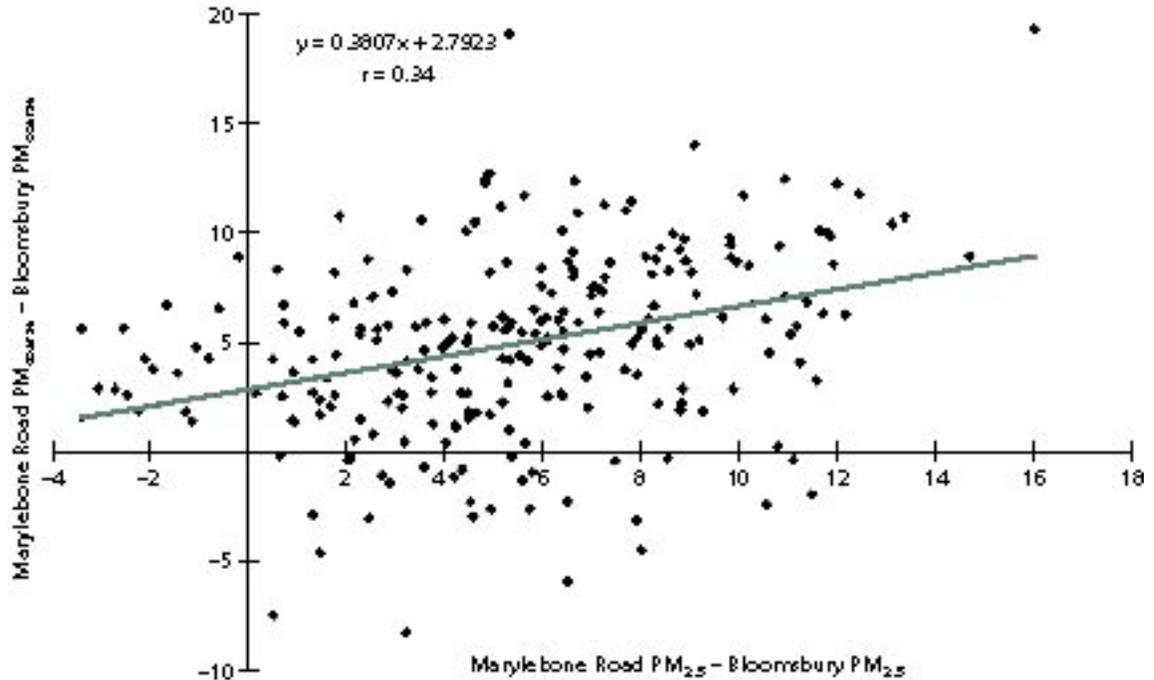
The relationships between the measured concentrations of PM_{10} and $PM_{2.5}$ and coarse particles at Marylebone Road have been investigated further by comparing daily mean concentrations

with those measured at London Bloomsbury. Daily mean concentrations of PM_{10} and $PM_{2.5}$, coarse particles and NO_x have been calculated for Marylebone Road for the period from June 1997 to September 1998. Daily mean background concentrations from London Bloomsbury were then subtracted to give the daily 'roadside enhancement' of the concentration of each species.

$$\text{roadside enhancement} = \text{roadside measurement} - \text{background measurement}$$

Figures 3.18, 3.19 and 3.20 show scatter plots of the daily mean enhancement of particle concentrations and NO_x concentrations. There is a clear correlation between PM_{10} and NO_x ($r = 0.88$), the relationship is weaker for $PM_{2.5}$ ($r = 0.79$) and weaker still for coarse particles ($r = 0.65$). The intercepts are also very small, indicating that London Bloomsbury is an appropriate background monitoring site for this analysis. The gradient of the linear regression line for PM_{10} and NO_x is about 0.04, approximately twice the value of the gradients for $PM_{2.5}$ and coarse particles. This is consistent with the roadside enhancement of daily mean PM_{10}

Figure 3.21 Comparison of daily mean PM_{coarse} and $PM_{2.5}$ concentrations at roadside and background sites, June 1997 - September 1998 ($\mu g m^{-3}$, contains provisional data).



concentrations at Marylebone Road consisting of, in roughly equal halves fine exhaust derived particles and coarse resuspended material. The relationship between the daily mean enhancements of $PM_{2.5}$ and NO_x concentrations is quite good, reflecting a common source (exhaust emissions) and similar day to day variation in dispersion characteristics within this street canyon environment. The relationship is weaker between coarse particles and NO_x due to the difference between the exhaust and resuspension sources (see section 5.3). Figure 3.21 shows that the relationship between the daily mean enhancements of $PM_{2.5}$ and coarse particles at Marylebone Road is weak ($r = 0.34$).

An analysis of PM_{10} , $PM_{2.5}$ and $PM_{10}-PM_{2.5}$ diurnal curves for monitoring data at Marylebone Road can also be used to derive an estimate of the likely contribution to roadside PM_{10} from resuspension. Comparison of the diurnal variation of particulates, NO_x and CO concentrations indicates that traffic-induced resuspension is a significant source.

3.6 ANALYSIS OF DATA FROM ROADSIDE SITES

Only recently have data on roadside PM_{10} concentrations become available. These data have been reviewed either by comparing two nearby sites, one of which is near a busy road and one of which is representative of the urban background, or by using modelling results which predict the concentration of particles generated by vehicles travelling along the road.

The annual average roadside contribution may be assessed from the difference in concentration at two sites when one site is adjacent to the road, and one site is thought not to be strongly affected by the road. The TSP data collected by Clarke *et al.* (1996) in Leeds illustrate typical results:

Roadside Conc. (Road+Background)	Urban Background	Road Contribution
5 m above ground 3 m from kerb	25 m above ground	Calculated from difference
70.2 $\mu g m^{-3}$	44.8 $\mu g m^{-3}$	25.4 $\mu g m^{-3}$

In this case sampling of the total suspended particulate matter took place over 9h daytime periods. The traffic flow along the road was 3000 vehicles/hour during the period of peak traffic flow, or 15,300 vehicles per nine-hour day. The local contribution from traffic to the measured concentration is very sensitive to the actual location of the sampler inlet. The reported concentrations may be compared with the Leeds Centre PM₁₀ annual average concentration of 25 µg m⁻³ in 1995. Further analyses of annual average PM₁₀ concentrations at roadside sites in 1997 using the same approach have been collated and these are summarised below (Table 3.36). All the sites are part of the UK National Network, apart from the M25 site, and TEOM instruments were used at all sites.

The estimated road contribution to annual average concentrations from the seven kerbside sites in the UK National Network is up to 13 µg m⁻³. Despite an exceedingly high traffic flow, the M25 monitoring site (operated by the Transport Research Laboratory) indicates that the annual average road contribution is less than 10 µg m⁻³ (see Section 3.6.1). Dilution caused by the wakes of moving vehicles may be significant when vehicles travel at motorway speeds and may explain the

apparently modest roadside concentration. Additionally the absence of roadside buildings favours good dispersion.

There is evidence from sites operated by Westminster City Council in London that in special circumstances the annual average contribution to PM₁₀ from the local road may be higher. This evidence comes from sites where Partisol analysers have been used to make PM₁₀ measurements and therefore direct subtraction of concentrations measured by TEOM analysers may not be valid. However, the possibility of enhanced local concentrations along roads carrying a high proportion of diesel vehicles, such as London's Oxford Street, or where a street canyon effect is likely to be important, such as in Regent Street, should be recognised.

The comparisons above are of annual average concentrations only. When considering short-term concentrations, such as daily averages, one expects the contribution from a nearby road to be more pronounced, since peak short-term concentrations occur on occasions when local factors, such as traffic flow, wind direction, wind speed and atmospheric stability, combine to produce high

Table 3.36 Comparison of longer term average PM₁₀ concentrations at roadside and urban background sites for 1997*.

Roadside Site		Background Site		Road Contribution	
Name	µg m ⁻³	Name	µg m ⁻³	Annual Average Daily Flow of traffic	µg m ⁻³
Sutton 1	24	Bexley/Eltham	22	20000	2
Haringey	26	Brent	22	17100	4
Camden 1	32	Brent/Kensington	23	45100	9
Glasgow Kerbside	30	Glasgow Central	20	15000	10
Bury	30	Bolton/Manchester Piccadilly	22	26100	8
A3	29	Bexley	23	104000	6
Marylebone Road	38	Kensington	25	71200	13
M25	27	Bexley/Eltham	22	184000	5

* where more than one site is given the data are average concentrations. Annual average daily traffic flows are both monitored and estimated. Distance from the sampler to the roadside for the sites were as follows: Sutton 3.5 m, Haringey 5 m, Camden <1 m, Glasgow Kerbside < 1 m, Bury 30 m, A3 < 1 m, Marylebone Road 1 m. Data are annual average concentrations for all sites except in the comparison between Glasgow Kerbside and Glasgow Central (March - December 1997), London A3 and Bexley (July - December 1997) and Marylebone Road and Kensington (July 1997 - December 1997).

Table 3.37 Measured and modelled results for Cambridge.

Cambridge Monitoring Site	Measured roadside $\mu\text{g m}^{-3}$	Maximum of primary vehicle generated PM_{10} predicted by three models* $\mu\text{g m}^{-3}$
Annual average PM_{10}	24	11
99 th percentile running 24h average PM_{10}	63	22

* models included ADMS-urban, AEOLIUS and AEOLIUSF.

concentrations immediately downwind of the road. Some of these meteorological conditions may also produce elevated concentrations from traffic emissions from the surrounding urban area. At urban background sites the traffic related fraction of the daily PM_{10} concentration during periods of high PM_{10} concentration was estimated by regression analysis using CO concentrations to be up to about 70% (see Section 5.2).

The determination of roadside concentrations by taking differences between concentrations measured at locations adjacent to and some distance away from a busy road is straightforward for annual average concentrations, for which the road, urban background and rural contributions to PM_{10} may be added directly. To determine the contribution from the road to short-term concentrations (such as daily averages) or for determining the influence of the road on percentiles is a much more complex procedure. In this case one needs to subtract the concentrations on an hour-by-hour basis taking into account local wind direction when the roadside site is downwind of the road. In many situations, reliable information on the local wind pattern is not available for such an analysis to be attempted.

Another approach to interpreting kerbside concentrations is to investigate the roadside contribution theoretically using a model. Factors such as local meteorology and distance from road differ from site to site and can, in principle, be investigated using a dispersion model.

A model cannot be expected to predict hourly concentrations accurately because of hourly changes of traffic flow, local meteorology and

urban background concentrations. In Cambridge, a PM_{10} monitor (Beta-attenuation monitor) was placed 5.8 m above ground in a street carrying 10,000 vehicles per day for the period September 1996 to February 1997. The street had a horizontal dimension of 17.5m and a vertical dimension of 12.5 m. The maximum road contribution predicted by three models was less than half the measured annual average concentration. The prediction was even less accurate when the models were used to estimate incidents of high pollution (Beeching, 1997). In addition, the model was used to predict primary PM_{10} only and modelled data shown in Table 3.37 have not had a secondary or coarse component added for comparison to the monitored concentrations.

The conclusion to be drawn from the Cambridge results is that within the annual average concentration of PM_{10} at least $13 \mu\text{g m}^{-3}$ arises from sources not considered by the model, and the contribution from more distant sources may be even greater to short-term peak concentrations.

During 1997 seven kerbside and roadside sites within the AUN, have become operational (Sutton, Haringey, Camden, Glasgow, Bury, London A3 and London Marylebone Road). If seven urban background sites associated with these roadside sites, two motorway sites along the M4 and M25, the Cambridge site and two of the Westminster City Council sites are included, one has a data set of 19 sites with 1997 PM_{10} concentrations which may be used to compare predictions from a dispersion model with measurements. Using the model GRAM (Fisher and Sokhi, 1998) which has been developed to provide a screening assessment of pollutants within the National Air Quality

Strategy and takes into account concentrations from road and urban background sources, the annual average and the 99th percentile of daily PM₁₀ concentrations may be predicted. These concentrations have been compared with annual and peak daily PM₁₀ concentrations.

The relationship between predicted and measured annual average PM₁₀ concentrations takes the following form:

$$\begin{aligned} \text{Mean_measured } PM_{10} &= 14.2 + 1.09 \\ \text{Mean_calculated } PM_{10} & (\mu\text{g m}^{-3}) \\ \text{where } r & \text{ equals } 0.823. \end{aligned}$$

The relationship between the predicted and measured 99th percentile daily PM₁₀ concentrations was found to be:

$$\begin{aligned} \text{Peak_measured } PM_{10} &= 41.4 + 0.28 \\ \text{Peak_calculated } PM_{10} & (\mu\text{g m}^{-3}) \\ \text{where } r & \text{ equals } 0.783. \end{aligned}$$

The model is able to explain much of the variation in annual average concentrations and the 99th percentile of daily PM₁₀ concentrations. However, a sizeable extra PM₁₀ concentration arising from regional sources mainly of secondary origin has to be included. In this regard, the model is consistent with the roadside modelling studies considered earlier. For peak concentrations, the GRAM model overestimates the contribution from road and urban background sources by a large margin. In this regard it is different to the earlier modelling studies. It may be considering situations which describe dispersion in a way which is unduly pessimistic. The large intercept of 41.4 μg m³ represents the contribution from regional, largely secondary sources. It is nearly three times the magnitude of the annual average PM₁₀ concentration arising from regional sources.

Episodes of high secondary PM₁₀ concentration may occur during the year as well as episodes of high urban and roadside concentrations. The effect of episodes of high concentration is attenuated within the annual average concentrations. The

meteorological conditions that cause episodes, namely light, generally easterly winds, may be associated with meteorological conditions which can lead to high urban and roadside concentrations (King and Dorling, 1997, Stedman, 1997; see Section 5.6.3).

Analysis of atmospheric measurement data indicates that the local contribution from a road is superimposed upon background primary and secondary particles. Modelling is not able to come to definitive conclusions as to the relative importance of each. At very busy kerbside sites, the ratio of the local road concentration to the urban background concentration to the rural background concentration, expressed as annual averages, is very roughly 1:1:1, but considerable site to site variation is to be expected. A traffic related contribution exists in all three components.

Assessing the relative contributions from local road, urban background and rural background concentrations to periods of high PM₁₀ concentration is not straightforward. Each of the components may make at different times, on its own or in combination, an important contribution to high PM₁₀ concentrations. The modelling results suggest that within adverse meteorological conditions very high PM₁₀ concentrations can occur near busy roads. However, the empirical results suggest that the local contribution is not as high as predicted and that regional effects assume considerable importance. The impact of traffic emissions on air quality close to major roads cannot be ignored in air quality reviews and assessments. Traffic emissions may still be the most significant source close to roads carrying very heavy traffic or which are in street canyons.

3.6.1 ANALYSIS OF DATA FROM TWO MOTORWAY SITES

3.6.1.1 Introduction

The Transport Research Laboratory operates a small network of air pollution monitoring stations for the Highways Agency. Two of these are located close to motorways (within a few

metres of the hard shoulder), and thus give an opportunity to examine air pollution in traffic-dominated situations relatively uninfluenced by other local sources. At the beginning of 1996, each of these motorway sites was equipped with a TEOM instrument configured to measure PM₁₀, and this section briefly describes the pollution conditions recorded at the two sites during 1996 and 1997. Most attention is given to the PM₁₀ measurements, but their interpretation is aided by considering levels of other compounds. Total oxides of nitrogen and carbon monoxide data are presented to characterise primary pollutants emitted by the motorway traffic; ozone and nitrogen dioxide are chosen to represent secondary compounds giving some insight into the local photochemical conditions.

In addition to the pollutant concentrations, the monitoring stations are equipped to measure traffic flows and some meteorological parameters. Because these variables play an important part in the formation and dispersion of the pollutants, they are also used in the examination of the data.

3.6.1.2 Site Descriptions

Site 1, M4 Motorway Theale (OS grid reference SU 4699 1695)

This site is located adjacent to the eastbound carriageway of the M4 motorway between junctions 11 and 12. The immediately surrounding area is predominantly rural. The nearest large town is Reading, the centre of which is approximately 4 km to the north. The villages of Theale and Shinfield are 5 km to the west and 4 km to the east respectively. The motorway at this location has three lanes in each direction and carries an average daily flow of about 100,000 vehicles with peak hour flows over 8000 vehicles. Congestion on the road is unusual, and the traffic normally travels at the high speeds typical of motorway driving.

Standard analysers are installed for the continuous measurement of carbon monoxide, total oxides of

nitrogen, nitric oxide, nitrogen dioxide (by difference), ozone, total hydrocarbons, methane, non-methane hydrocarbons (by difference) and PM₁₀. Traffic is counted in each direction using inductive loops installed between the same two junctions. Wind speed, wind direction, air temperature and relative humidity are also recorded.

Air samples for the gas analyses are taken from a point at the edge of the hard shoulder, at a height of 1.6 m; the TEOM takes its sample from a roof-mounted PM₁₀ inlet head 3.5 m from the hard shoulder, at a height of 3 m. Wind data are measured at 3.5 m height, and temperature and humidity at 2 m.

Site 2, M25 Motorway Staines (OS grid reference TQ 5027 1737)

This site is near the clockwise carriageway of the M25 between junctions 13 and 14. Again, the immediate surroundings are largely rural, including several nearby reservoirs. There are many conurbations in the area of the site: to the northwest are Old Windsor, Windsor and Slough, at distances of 4, 7 and 8 km; to the south and southeast are Egham, Staines and Ashford, between 2 and 10 km from the site; to the east are Stanwell (3 km), Feltham (8 km) and the westerly part of Greater London; and about 5 km to the north-east is Heathrow Airport. The motorway has 4 lanes in each direction; traffic flows average approximately 180,000 vehicles per day with peak hourly flows of 12,000 vehicles. This section is one of the busiest on the M25, and is often congested during peak periods. Thus, while vehicles mainly travel at typical motorway speeds, there are times when the traffic is slower (a variable speed limit system is in operation which restricts maximum speeds to 50 or 60 mph depending on traffic conditions).

Instrumentation at this site is similar to that at the M4. Sampling arrangements and locations are also similar to those at the M4 site.

3.6.1.3 Data summaries

Figures 3.22 and 3.23 show average pollutant concentrations for each hour of the day at the two monitoring sites. Tables 3.38 and 3.39 provide additional statistical information, partly in relation to the objectives of the National Air Quality Strategy.

Table 3.38 Summary of pollutant concentrations at the M4 site, 1996 and 1997.

Statistic	CO (ppm)	NO ₂ (ppb)	NO _x (ppb)	PM ₁₀ (µg m ⁻³)
1996				
max 1-h	5.3	393	962	152
98%ile 1-h		123	412	
annual av	0.58	31	129	24.4
max 8-h	3.3			
97%ile 8-h				
max 24-h				101
99%ile 24-h				65.3
1997				
max 1-h	4.8	302	830	173
98%ile 1-h		115	468	
annual av	0.68	27	130	23.1
max 8-h	3.0			
97%ile 8-h				
max 24-h				94
99%ile 24-h				82.7

Table 3.39 Summary of pollutant concentrations at the M25 site, 1996 and 1997.

Statistic	CO (ppm)	NO ₂ (ppb)	NO _x (ppb)	PM ₁₀ (µg m ⁻³)
1996				
max 1-h	22.2	129	1156	184
98%ile 1-h		75	580	
annual av	1.65	30	202	31.2
max 8-h	17.0			
97%ile 8-h				
max 24-h				125
99%ile 24-h				99
1997				
max 1-h	8.2	112	937	164
98%ile 1-h		60	538	
annual av	0.90	26	170	26.6
max 8-h	5.8			
97%ile 8-h				
max 24-h				82
99%ile 24-h				70

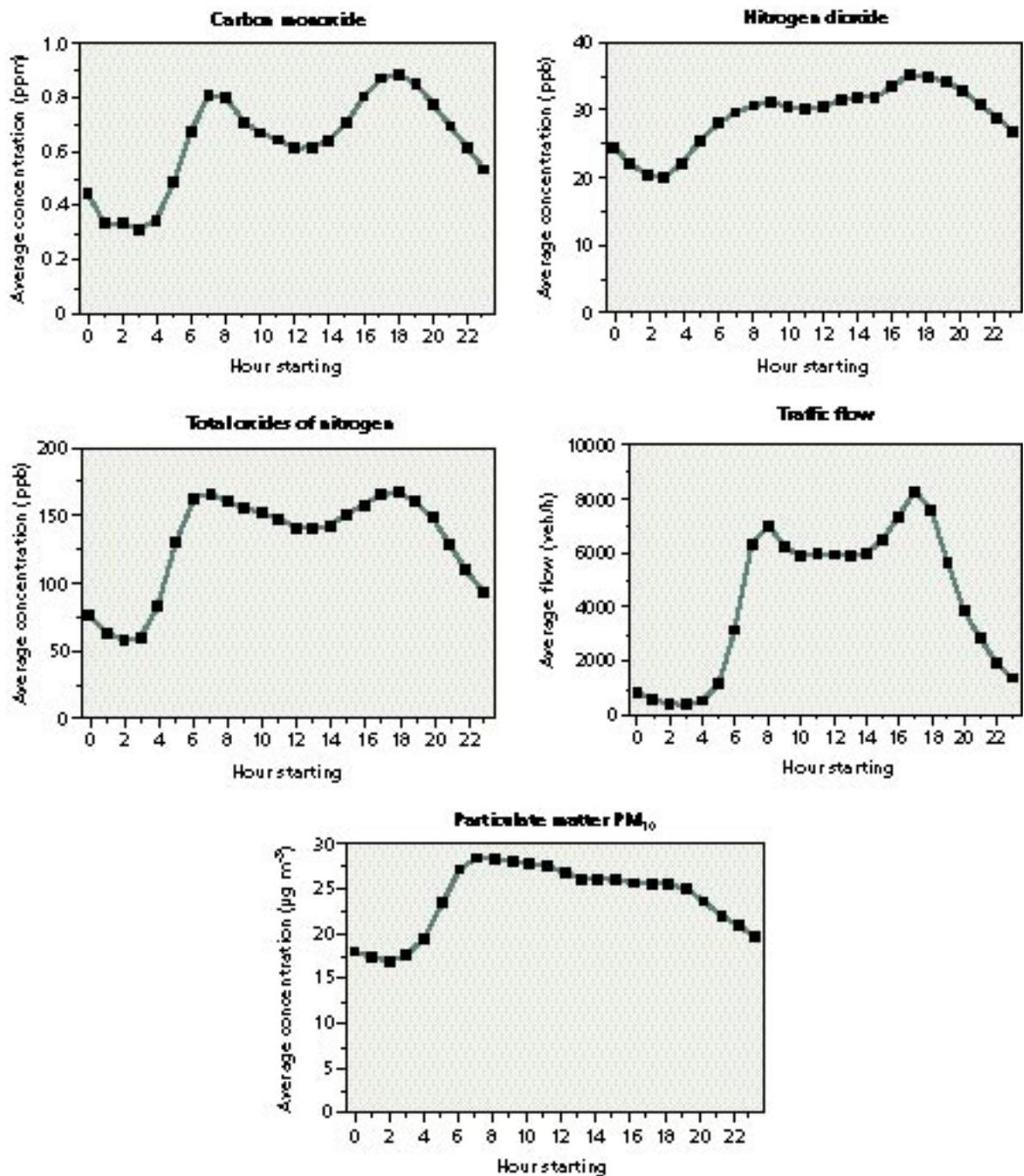
The daily concentration profiles shown in the figures show a general relationship between all of the pollutants and the traffic flow. This is most apparent for the primary compounds (CO and NO_x), but also clear for NO₂ and PM₁₀. This link is also demonstrated by the absolute values recorded for the primary compounds: the ratio of average CO concentrations during the morning peak hour is (M25:M4) 2.1:1, that for NO_x is 1.5:1, while the equivalent ratio of traffic flows is 1.8:1. Some concentrations recorded for NO₂ at the M4 site were surprisingly high and there is no apparent explanation for this. The probability of congestion is greater on the M25 than on the M4, with the result that emission rates of CO per vehicle are likely to be somewhat higher and those of NO_x lower on the M25 during the peak hour. The equivalence, therefore, of the pollution concentration and traffic ratios is very close. On the other hand, NO₂ and PM₁₀, pollutants with a significant secondary or non-traffic component behave rather differently. Ratios of peak hour concentrations are 0.9:1 for NO₂ and 1.2:1 for PM₁₀. Nitrogen dioxide concentrations in these locations are principally controlled by the equilibrium between NO, NO₂ and O₃. Thus, even though NO_x concentrations are higher at the M25, those of NO₂ are, on average, a little lower. Concentrations of PM₁₀ at the M25 are higher than at the M4, but by a smaller amount than suggested by the relative traffic flows. This reflects the significant contribution of particles from sources other than the local traffic. A notable feature of Figure 3.22 and 3.23 observed also at some other heavily trafficked locations is that the diurnal profile of PM₁₀ parallels that of NO_x more closely than CO. This suggests different patterns of petrol and diesel traffic, the latter being the main determinant of PM₁₀ and NO_x.

3.6.1.4 Sources of PM₁₀

Local traffic emissions

Using the average data presented in Figures 3.22 and 3.23, a simple regression analysis has been undertaken to examine the relationship between

Figure 3.22 Average pollution concentrations and traffic flow by hour of day, M4, Jan - Dec, 1996 and Jan - Dec, 1997.



the annual average PM₁₀ concentrations and the traffic on the motorways. Linear regressions yield the following equations:

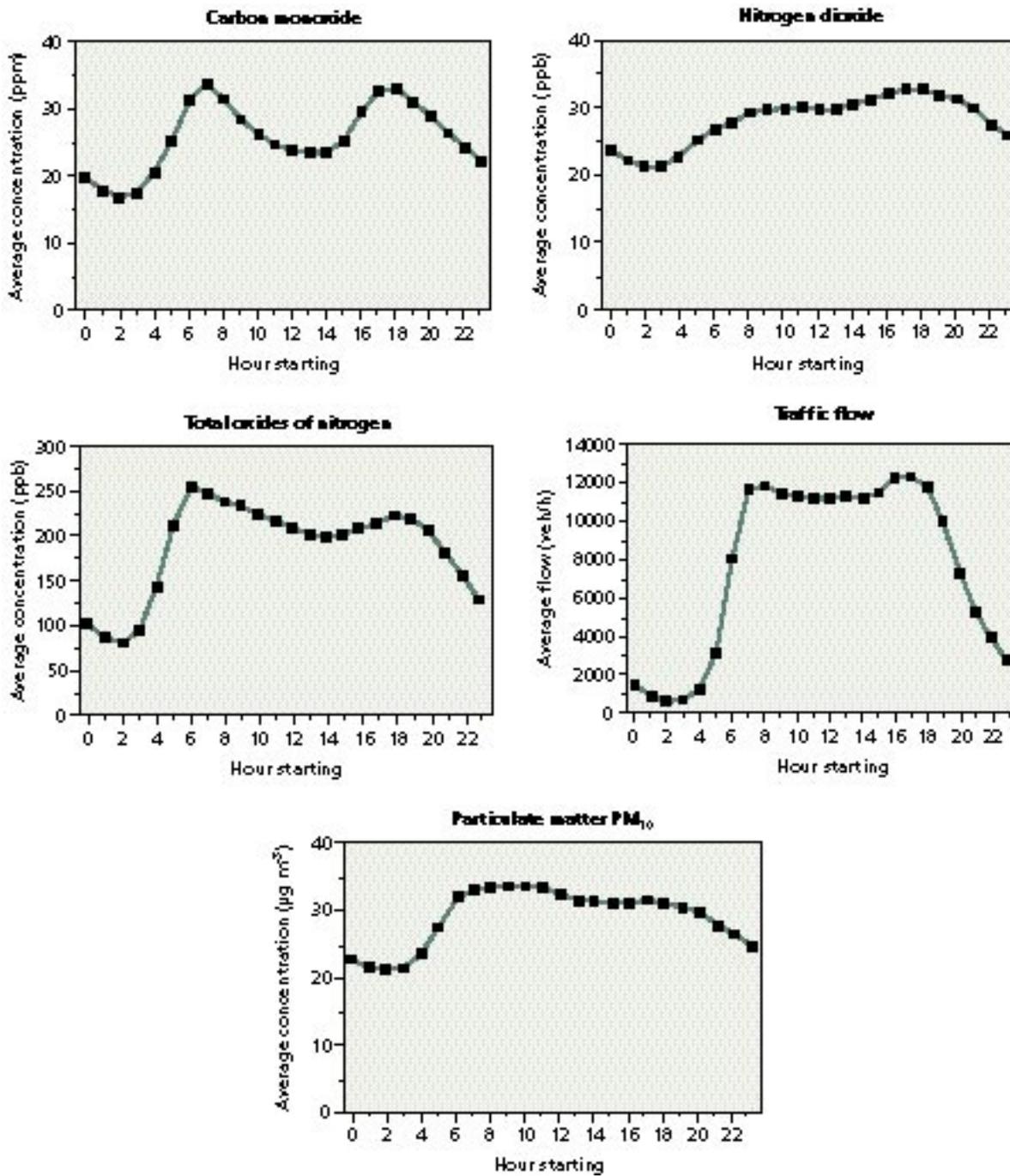
at the M4 $PM_{10} = 18.6 + 0.00122T$

and at the M25 $PM_{10} = 22.1 + 0.000901T$

where PM₁₀ is the PM₁₀ concentration in µg m⁻³ and T is the traffic flow in veh/h.

Although slightly different at each site, a simple interpretation of these results is that the annual average background concentration of about 20 µg m⁻³ is increased by about 1 µg m⁻³ for each 1000 veh/h.

Figure 3.23 Average pollution concentrations and traffic flow by hour of day, M25, Jan - Dec, 1996 and Jan - Dec, 1997.



Applying the same procedure to the measured CO concentrations shows the traffic contribution to be approximately 0.05 ppm, or 60 µg m⁻³ per 1000 veh/h. An intercomparison of vehicle emission rates for carbon monoxide and PM₁₀, weighted according to the traffic composition on the motorways and allowing for the typical

traffic speeds is broadly consistent with these observations. The latest vehicle emission factors compiled by TRL and NETCEN suggest average rates of emission for these traffic conditions of 6.3 g/veh.km of CO and 0.09 g/veh.km of PM₁₀, in very close agreement with the 60:1 ratio estimated from the atmospheric concentrations.

Other contributions

Even when there is little traffic on the motorways, the average concentrations of PM_{10} do not fall much below $20 \mu\text{g m}^{-3}$. Clearly there are sources other than the local traffic contributing to the observed pollution levels. These are examined with reference to Figures 3.24 and 3.25. They show the product of the hourly average PM_{10} concentration and the wind speed as a function of the wind direction for periods of high and low traffic flow on the motorways. The main factors determining the concentration not far downwind of a near ground-level source are source strength, wind speed, depth of cloud through which the pollution has mixed and wind direction. The concentration is proportional to the source strength and inversely proportional to the wind speed and cloud depth. It is therefore plausible to expect that a peak in the product of concentration and wind speed in a certain directional sector surrounding the monitoring site is an indication that a significant source lies in that sector.

At the M4 site (Figure 3.24), there are two clear wind directions that give rise to high levels of PM_{10} . One is when winds are from a direction slightly to the east of north. This wind direction is in a direct line from Reading town centre to the monitoring location, and there is little doubt that the elevated concentrations arise from particle emissions in Reading. The second, broader wind sector producing high PM_{10} levels is from approximately south to west: winds in this sector blow over the M4 towards the monitoring site which suggests that the particles are from the traffic on the motorway.

The situation at the M25 site is rather less clear. During periods of both high and low traffic flows, the highest PM_{10} concentrations occur when winds are in the north-east to east sector (Figure 3.25). This direction will transport particles from Greater London towards the monitoring position, and also from the M25, since the monitoring station is to the west of the motorway. However, when traffic flows are high, there is also evidence of increased

concentrations during periods when the wind is from the south. This may indicate a contribution from activities in the nearby towns of Staines and Egham, but may also show an accumulation of particles from the motorway traffic: a southerly wind is roughly parallel with the road and will direct emissions towards the monitoring site.

3.6.1.5 Other roadside PM_{10} monitoring

In addition to the long term continuous monitoring discussed above, TRL have conducted shorter period surveys at several other roadside locations. Their results are summarised below.

A40, Cheltenham

A new monitoring station was established in Cheltenham during 1997. From mid-November, a TEOM has been in operation at this site together with a range of gas analysers similar to those at the motorway locations. The station is situated on a roundabout on the A40 trunk road. The TEOM sample is taken at a position 1.5 m from the road, at a height of 2 m; gas samples are taken 3 m from the road at a height of 3.5 m. Traffic counting equipment has not yet been installed, but short term manual counts suggest an average flow of the order of 30,000 veh/day.

Average concentrations of CO , NO_x and PM_{10} recorded during November and December 1997 are shown in Figure 3.26. Although the estimated traffic flow at this site is well below those on either motorway, average levels of CO are higher. This is attributable to the high rates of CO emission produced by the slow moving traffic at this location. Typically, CO emissions from petrol cars travelling in the speed range below 30 km/h are 5 to 20 times higher than those from cars at 100 km/h. Oxides of nitrogen concentrations are lower than at the motorway sites: urban emission rates may be around twice as high as those on motorways, but this is offset by the lower traffic flow. Levels of PM_{10} are comparable with those found at the motorway sites, reaching an average concentration of

Figure 3.24 PM₁₀ concentrations during different wind and traffic conditions, M4, Jan - Dec, 1997 (1 hour averages).

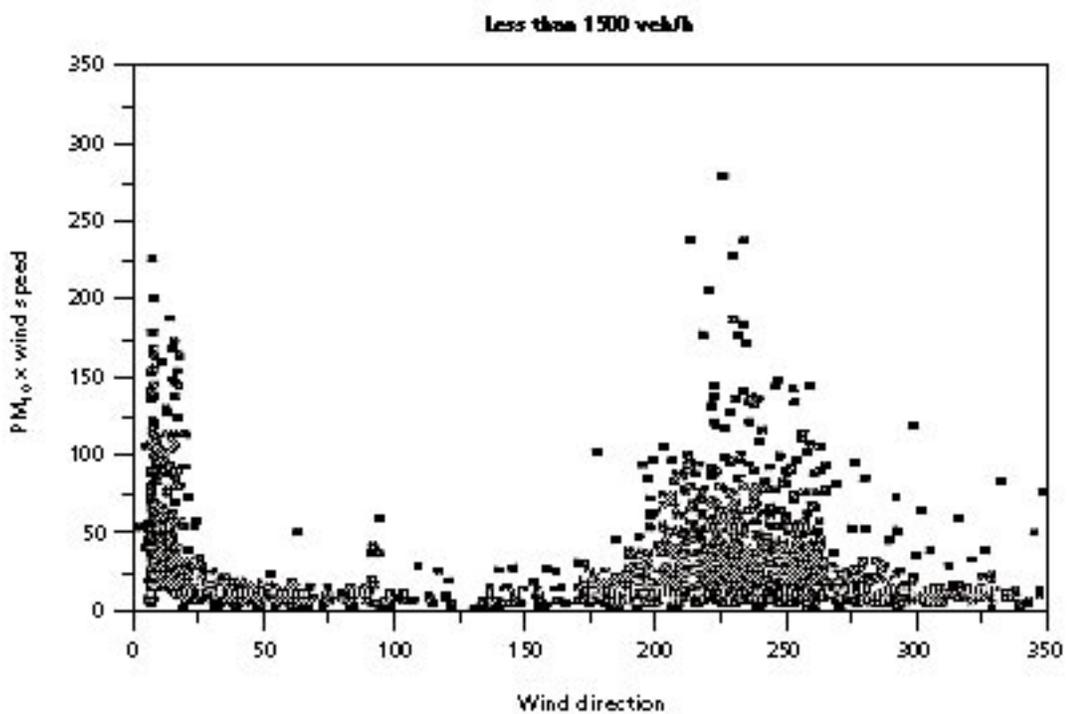
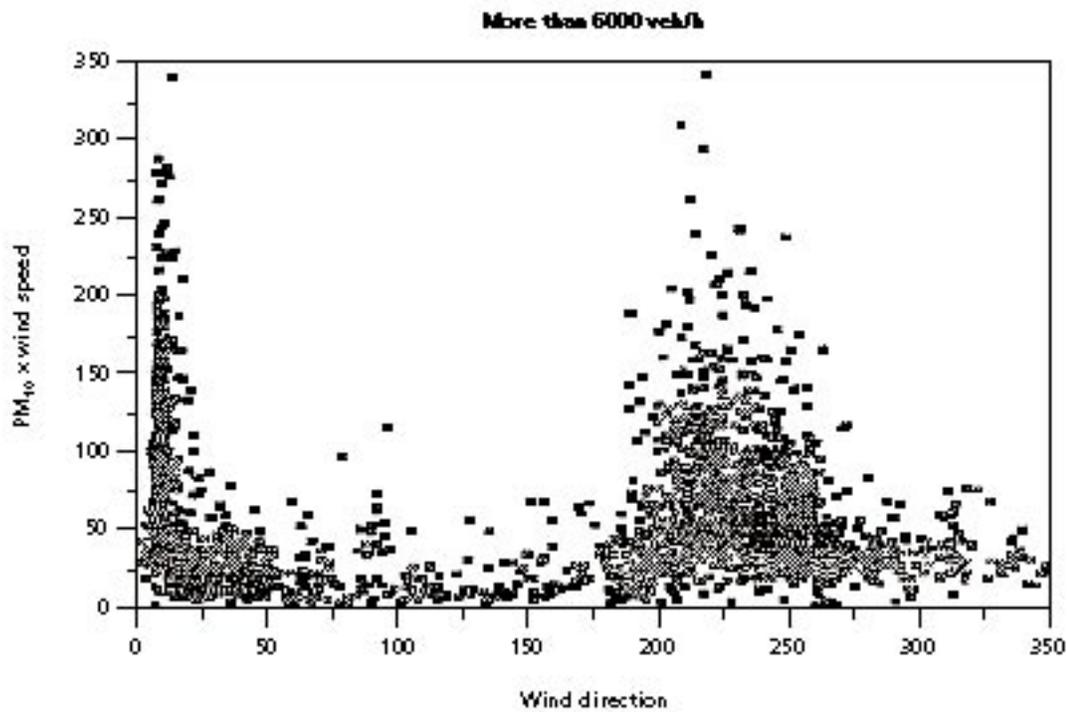
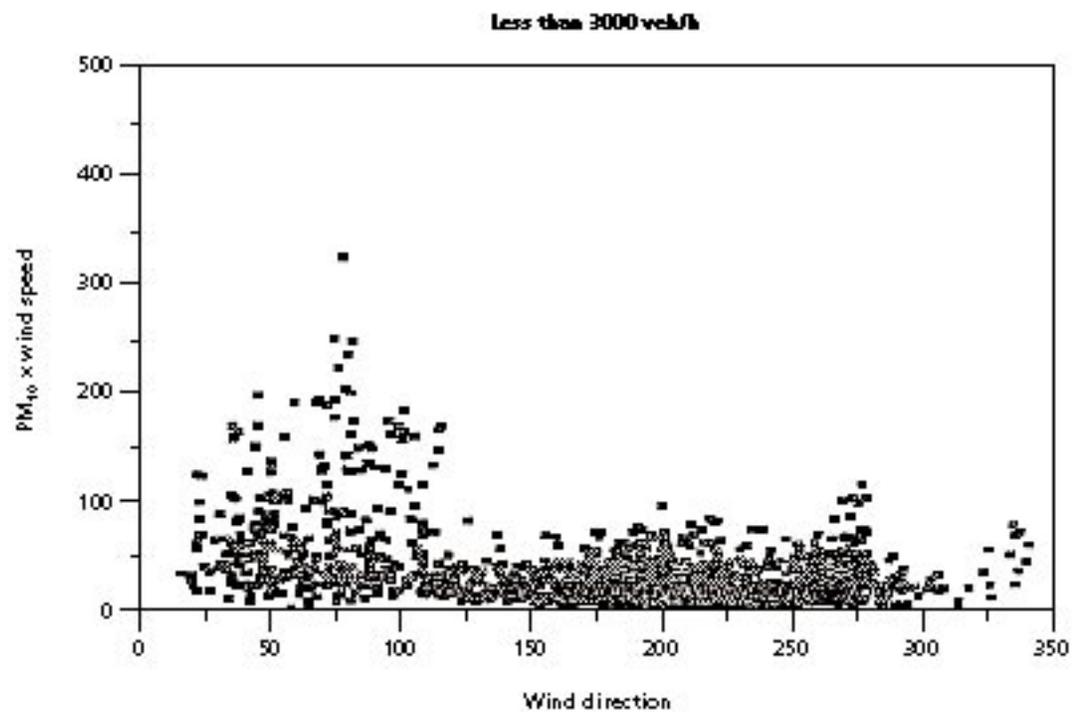
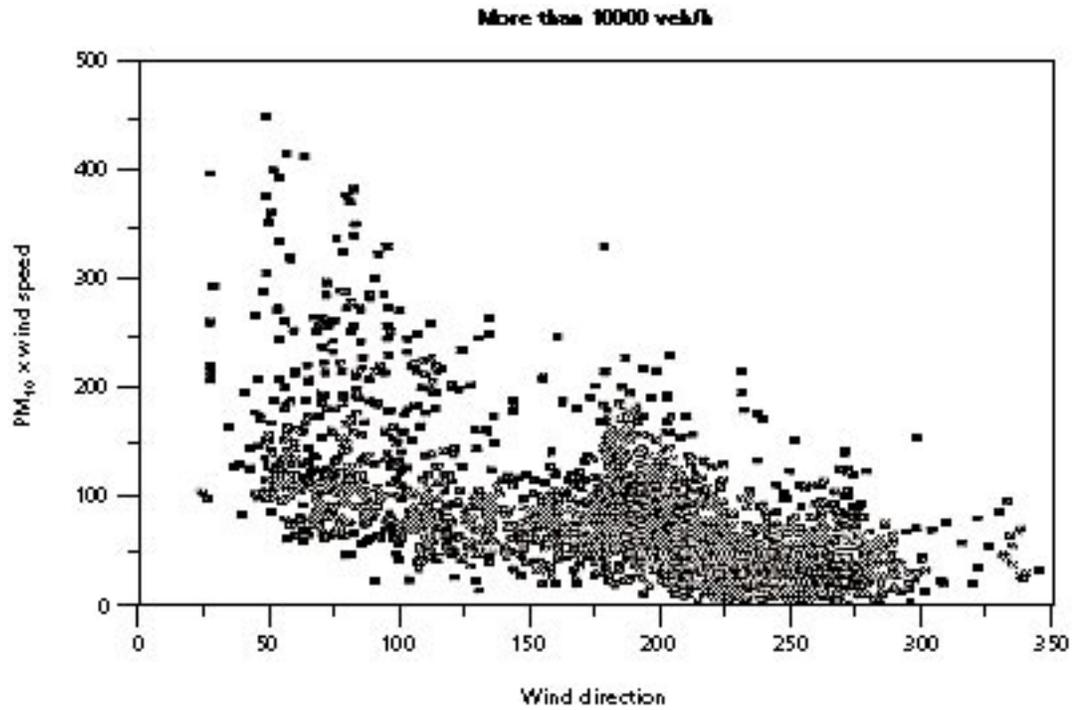


Figure 3.25 PM_{10} concentrations during different wind and traffic conditions, M25, Jan - Dec, 1997 (1 hour averages).



around $30 \mu\text{g m}^{-3}$ during times of peak traffic. Again, low speed urban driving causes higher rates of emission, but the high levels relative to the traffic flow in comparison with the motorway sites may also reflect contributions from other traffic and non-traffic sources in Cheltenham.

Victoria Street, Central London

Daily average PM_{10} concentrations have been measured in Victoria Street since November 1997. Samples are taken using a Partisol system and determined gravimetrically. The sampler is mounted on the pavement, with its inlet approximately 0.5 m from the road at a height of a little over 1 m. The road carries an average of about 20,000 veh/day. To date, a total of 106 samples have been processed. The average PM_{10} concentration shown by these samples is $45 \mu\text{g m}^{-3}$, with a maximum daily concentration of $105 \mu\text{g m}^{-3}$.

Because of the difference in sampling and analysis, these results are not directly comparable with TEOM measurements.

A465, South Wales

A short survey was conducted near the A465 in South Wales during May and June 1997. For a total of 26 days, MiniVol PM_{10} samplers were operated simultaneously at 4 locations. The sites were in two pairs, each combining a position near to the road with one at a greater distance. In one case the positions were 2 and 54 m from the kerbside and the others were at 4 and 100 m. All samples were taken at a height of about 1 m. In this area, which is mainly rural, the A465 carries a flow of about 15,000 veh/day.

Results from this survey are summarised in Table 3.40. As with the Partisol data, because the sampling method was different, they cannot be compared directly with TEOM data. Whilst they are rather limited, these data do illustrate the effect of increasing distance from the road on PM_{10} concentrations.

Figure 3.26 Hourly average pollution concentrations by time of day, Cheltenham (Nov - Dec, 1997).

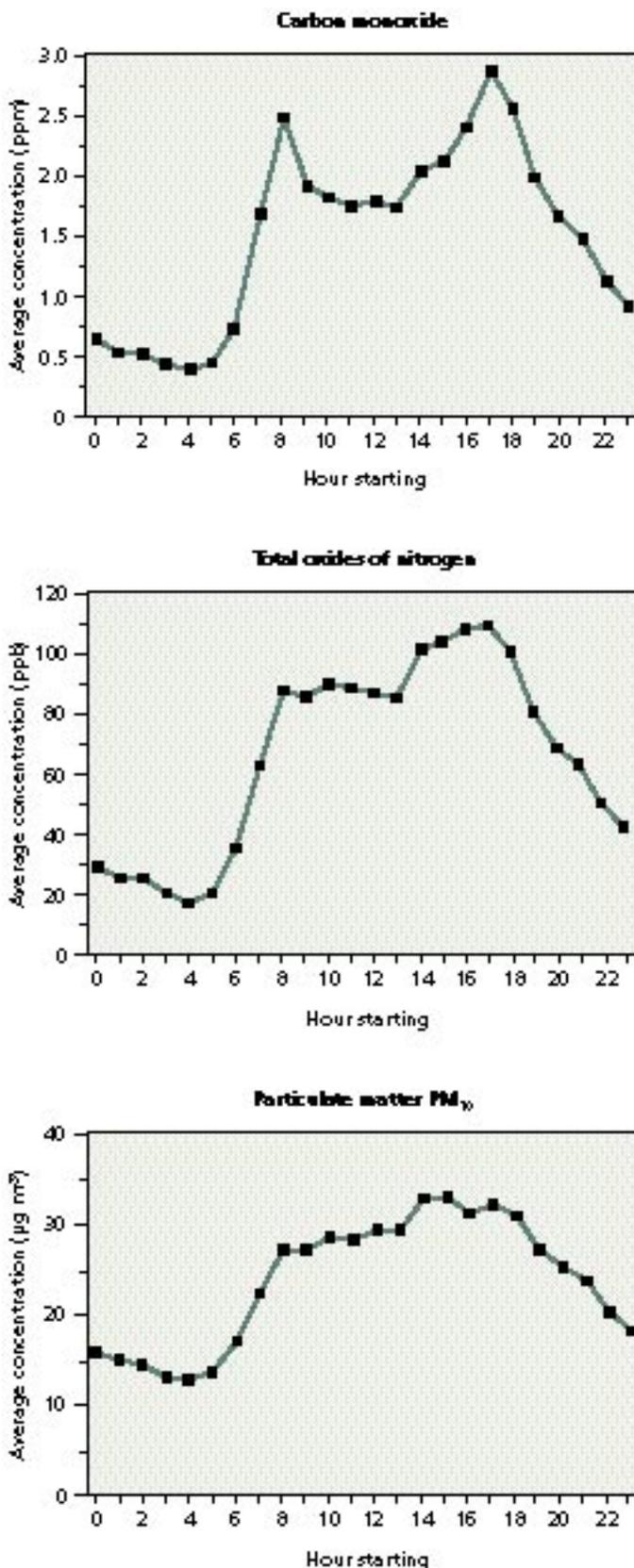


Table 3.40 PM₁₀ concentrations measured near to the A465*.

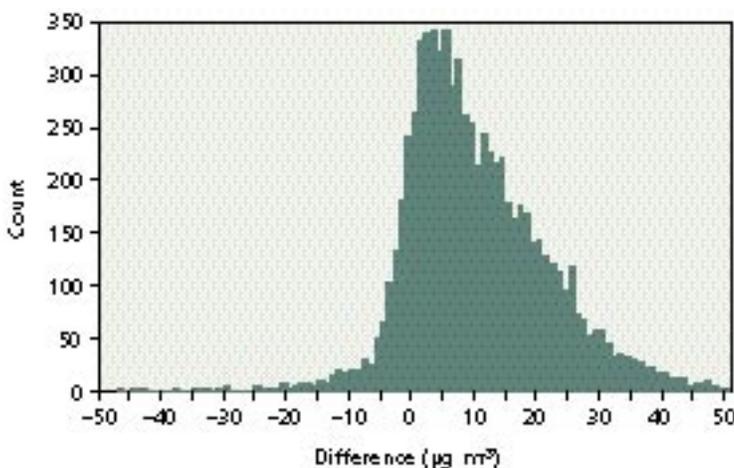
Position number	Distance to A465 (m)	PM ₁₀ concentrations (µg m ⁻³)	
		Average	Maximum
1a	2	32	76
1b	54	22	55
2a	4	28	55
2b	100	19	46

*Samples were discreet 24 hour samples over a total of 26 days. The maximum is the highest 24 hour value and the average is the mean of the 26 days. MiniVol samplers were used.

3.6.2 TRAFFIC CONTRIBUTION TO PM₁₀

The establishment of kerbside monitors enables an examination of the contribution of PM₁₀ from traffic related sources. The site at Marylebone Road, London commenced in July 1997. Differences in concentrations on an hour by hour basis between levels recorded at Marylebone Road and the urban background site at London Bloomsbury are shown in Figure 3.27 for the period July 1997 - May 1998 (contains provisional data). This frequency distribution indicates that in general concentrations of PM₁₀ were larger at the kerbside site when compared to the urban background site. However, there were some occasions when the opposite occurred which may be related to weather conditions or occasions

Figure 3.27 Frequency Distribution showing the difference in hourly concentrations from London Marylebone Road and London Bloomsbury (July 1997 - May 1998 - contains provisional data) (µg m⁻³).



during the night when emissions were likely to have been lowest at Marylebone Road. Most typically, concentrations of PM₁₀ were about 5 µg m⁻³ higher at the kerbside site compared to the urban background site but, in addition, there were numerous occasions when this difference was up to about 30 mg m⁻³ and a few occasions when the difference in concentrations exceeded 30 µg m⁻³. This emphasises both the magnitude and the variability of the traffic related contribution to measured PM₁₀.

3.7 PARTICLE EPISODES AROUND BONFIRE NIGHT

An analysis of the concentrations of PM₁₀ at numerous sites in the national automatic monitoring network during the bonfire night period indicates strong differences from 1994 to 1997 (Figure 3.28). Episodes occurred during 1994 and 1995 when November 5th fell on a weekend when most of the celebrations took place causing an increase in PM₁₀ concentrations. In 1995 the highest concentrations of PM₁₀ occurred in Leeds urban background national monitoring site during the period 4 - 6th November, where low temperatures with light winds combined to trap the particles close to their sources thus causing the observed build up. The maximum hourly concentration of 782 µg m⁻³ occurred at 11 pm when most of the bonfires would have subsided and were smouldering (Clark, 1997). In 1995 there was no obvious pattern in the geographical distribution of the maxima in PM₁₀. Since high and low concentrations were found in most regions of the country it is therefore likely that PM₁₀ concentrations measured on Bonfire Night were most strongly influenced by local conditions such as how close the monitoring sites were to bonfires, and local meteorology.

However, such widespread episodes were not evident during 1996 and 1997 when November 5th fell on a Tuesday and Wednesday, respectively. It is likely in these years festivities occurred over a number of days including November 5th itself and

Table 3.41 Exceedance of 50 µg m⁻³ standard for 24 hour running means from November 1st - November 10th in the years 1994 - 1997.

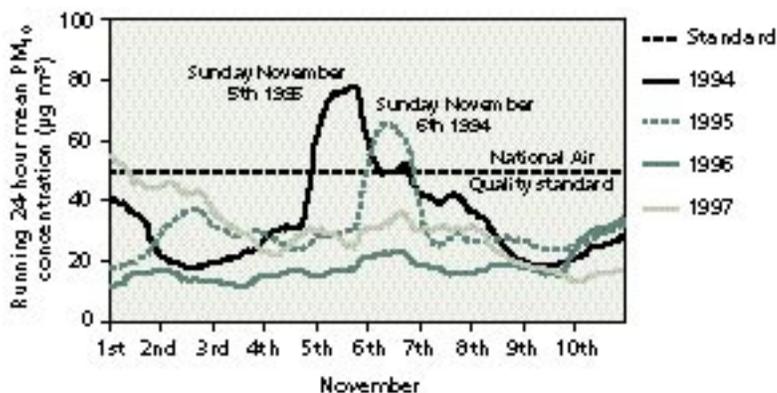
Site Name	1994		1995		1996		1997	
	Exc	Days	Exc	Days	Exc	Days	Exc	Days
London Bloomsbury	4	1	72	5	0	0	32	2
Edinburgh Centre	0	0	0	0	0	0	0	0
Cardiff Centre	43	4	60	4	0	0	43	2
Belfast Centre	20	1	8	1	2	1	70	5
Birmingham Centre	24	2	24	2	19	1	19	1
Newcastle Centre	22	3	47	3	0	0	13	1
Leeds Centre	36	3	42	3	0	0	34	4
Bristol Centre	19	1	64	5	0	0	45	2
Liverpool Centre	24	2	54	4	0	0	0	0
Birmingham East	26	2	48	3	23	1	23	1
Hull Centre	25	2	19	2	0	0	47	4
Leicester Centre	21	1	0	0	0	0	22	1
Southampton Centre	12	1	90	6	0	0	9	2
London Bexley	21	1	65	4	0	0	53	3
Swansea Centre	-	-	70	5	0	0	0	0
Middlesbrough	-	-	27	2	21	2	26	2
Manchester Piccadilly	-	-	-	-	0	0	23	1
Sheffield Centre	-	-	-	-	0	0	17	2
Wolverhampton Centre	-	-	-	-	18	1	9	1
London Brent	-	-	-	-	0	0	33	3
Sutton Roadside	-	-	-	-	0	0	64	3
London North Kensington	-	-	-	-	0	0	52	3
Haringey Roadside	-	-	-	-	0	0	37	3
Camden Roadside	-	-	-	-	0	0	43	3
London Eltham	-	-	-	-	0	0	39	3
London Hillingdon	-	-	-	-	0	0	45	3
Glasgow Centre	-	-	-	-	0	0	0	0
Leamington Spa	-	-	-	-	0	0	12	2
Nottingham Centre	-	-	-	-	14	1	25	2
Thurrock	-	-	-	-	0	0	36	3
Stockport	-	-	-	-	-	-	11	1
Port Talbot	-	-	-	-	-	-	0	0
Bury Roadside	-	-	-	-	-	-	38	2
Bolton	-	-	-	-	-	-	41	3
Glasgow Kerbside	-	-	-	-	-	-	16	2
Salford Eccles	-	-	-	-	-	-	20	1
Stoke On Trent Centre	-	-	-	-	-	-	22	1
London A3 Roadside	-	-	-	-	-	-	71	3
Derry	-	-	-	-	-	-	46	3
Redcar	-	-	-	-	-	-	7	1
London Marylebone Road	-	-	-	-	-	-	63	5
Norwich Centre	-	-	-	-	-	-	30	2
Reading Centre	-	-	-	-	-	-	35	2
Plymouth Centre	-	-	-	-	-	-	0	0

Key:

Exc= number of exceedances.

Days= number of days with exceedances.

Figure 3.28 Bonfire Night Episodes 1994 to 1997
Running 24-Hour Mean Network Averaged Data (all sites).



the preceding and following weekends. Consequently, emissions from bonfire and fireworks were likely to have been more diffuse than during 1994 and 1995.

Exceedances of the national air quality standard of $50 \mu\text{g m}^{-3}$ as a 24 hour running average occurred at numerous sites during the period 1st to 10th November, 1994 to 1997 (Table 3.41). Exceedances tended to be fewer in 1996 than in other years due to the higher windspeeds enhancing dispersion. The national air quality PM_{10} objective is set at the 99th percentile which 'allows' exceedances over 4 days (DoE, 1997). During 1994, 1995 and 1997 exceedances over 4 or more days occurred at at least one site in the national monitoring network. Indeed, in 1995 the current national air quality objective would not have been met at 25% of the monitoring sites in operation (exceedances > 4 days). No additional exceedances throughout the year would have been possible at a further 19% of monitoring sites if the objective was to have been met (exceedances = 4 days). It is not considered likely that emissions from the Bonfire Night celebrations will significantly change in future years and consequently this source alone is likely to contribute significantly to primary sources of PM_{10} . The number of exceedances caused by this additional source of emissions is likely to be highly dependent on meteorological conditions at that time, as well as the day of the week which November 5th falls.

3.8 LOCAL SOURCES OF PARTICLES

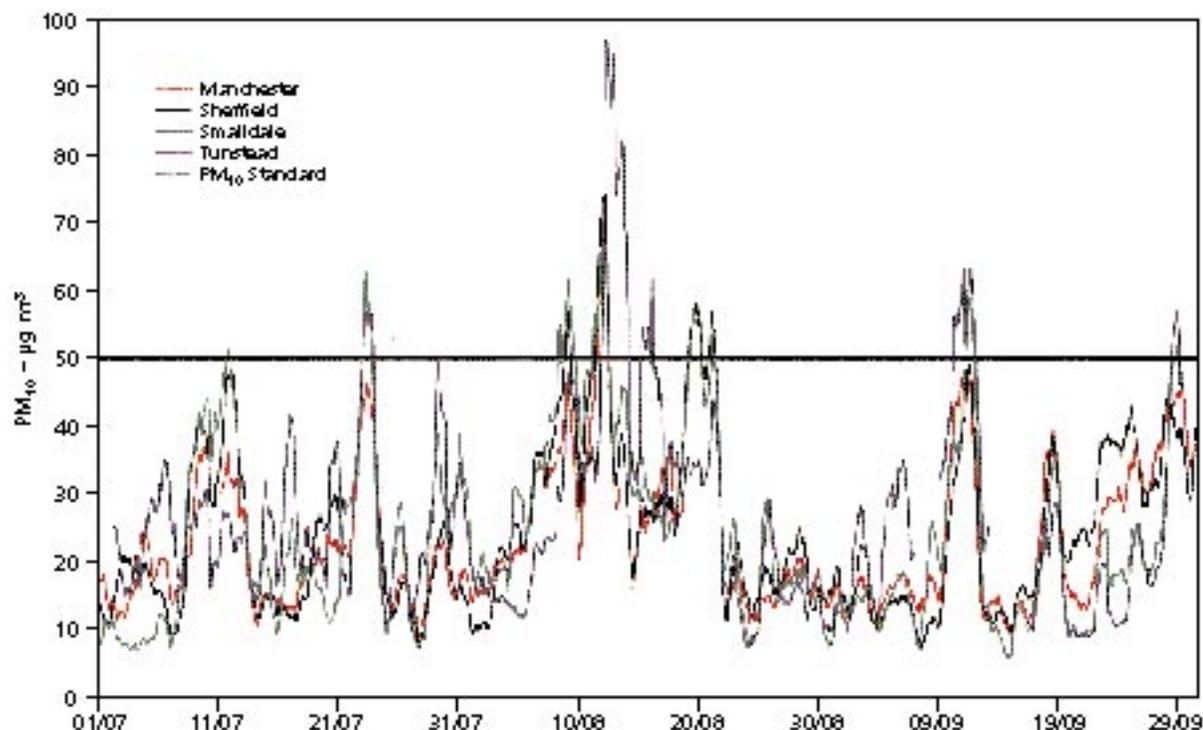
3.8.1 INTRODUCTION

The atmospheric particle concentrations measured at a particular location will be made up from particles from a range of sources. The concentrations of secondary particles, such as sulphates and nitrates are likely to be fairly uniform across a local authority area (QUARG, 1996) (see Chapter 4). The concentrations of particles derived from vehicles can be expected to vary with local and regional traffic movements. The concentrations of particles from local industrial or other sources have been examined by several Local Authorities who undertook pilot studies as part of the 'First Phase Authority' work (DETR *et al.*, 1999). Several authorities undertook measurements of PM_{10} concentrations in areas where it was thought likely that local particle sources could make contributions. Some key results from the monitoring of PM_{10} concentrations in areas with sources specific to particles are discussed below.

3.8.2 HIGH PEAK: PM_{10} IN THE VICINITY OF QUARRY ACTIVITIES

The High Peak authorities investigated the influence of mineral processing activities on local PM_{10} concentrations (DETR *et al.*, 1999). Four monitoring sites were established, three within 400 m of the boundaries of large limestone quarries in the Buxton area, together with a background site more than 5 km from the nearest mineral process. Instruments deployed at these sites were Casella APM950 devices based on a light-scattering measurement technique. Measurements were carried out between December 1996 and September 1997 and hourly and running 24-hour average PM_{10} concentrations were calculated and compared with city centre PM_{10} monitoring results from the national Automatic Urban Network (AUN) sites in Sheffield and Manchester. Weekly concentrations of a variety of elements from the particles collected were also measured by scanning electron microscopy.

Figure 3.29 Running 24-hour average PM_{10} concentrations at Tunstead, Derbyshire, and AUN sites for the period 1 April 1996 - 30 June 1997 ($\mu\text{g m}^{-3}$). A TEOM instrument was used at Manchester and Sheffield but at the other sites a Casella APM950 instrument was used.



Exceedances of the national air quality standard of $50 \mu\text{g m}^{-3}$ 24 hour running mean were recorded at all of the monitoring sites in this study, with the background site at Litton showing the smallest number of days. Exceedances of the $50 \mu\text{g m}^{-3}$ air quality standard during the winter at these sites generally occurred at more than one site and coincided with high concentrations at the Sheffield and/or Manchester Automatic Urban Network sites. These exceedances were associated with cold, still conditions, with the likelihood of temperature inversions. The sulphur concentrations of the particles associated with this type of episode were generally more significant than calcium, indicating that the quarry related activities may not have made a significant contribution.

Summer-time exceedances were different, with calcium more dominant, indicating the importance of local quarry sources and/or lime or cement processes. Exceedances were almost always due to calm overnight conditions. Quarry crushing and screening processes do not operate at night but lime and cement processes continue. When wind speeds

increased in the morning, PM_{10} concentrations decreased. These exceedances also often coincided with increased concentration at the Sheffield and Manchester AUN sites. Two episode periods at the Tunstead site that were clearly strongly influenced by the local mineral process emissions are illustrated in Figure 3.29. Concentrations at this site were significantly elevated above those measured at the AUN sites on the 11th and 12th of August and again on the 10th and 11th of September. The scanning electron microscopy analysis results for the weekly filter samples collected over these periods clearly indicate the importance of calcium (Figure 3.30).

3.8.3 CORNWALL: PM_{10} IN THE VICINITY OF CHINA CLAY WORKINGS

The authorities participating in the Cornwall Air Quality Forum made measurements of PM_{10} concentrations at sites within the St Austell china clay area in order to provide new information on the likely impact of mineral extraction activities on local air quality. Monitoring took place at

Figure 3.30 Scanning Electron Microscope analysis from measurements made at Tunstead, Derbyshire, for the period July - September 1997 ($\mu\text{g m}^{-3}$).

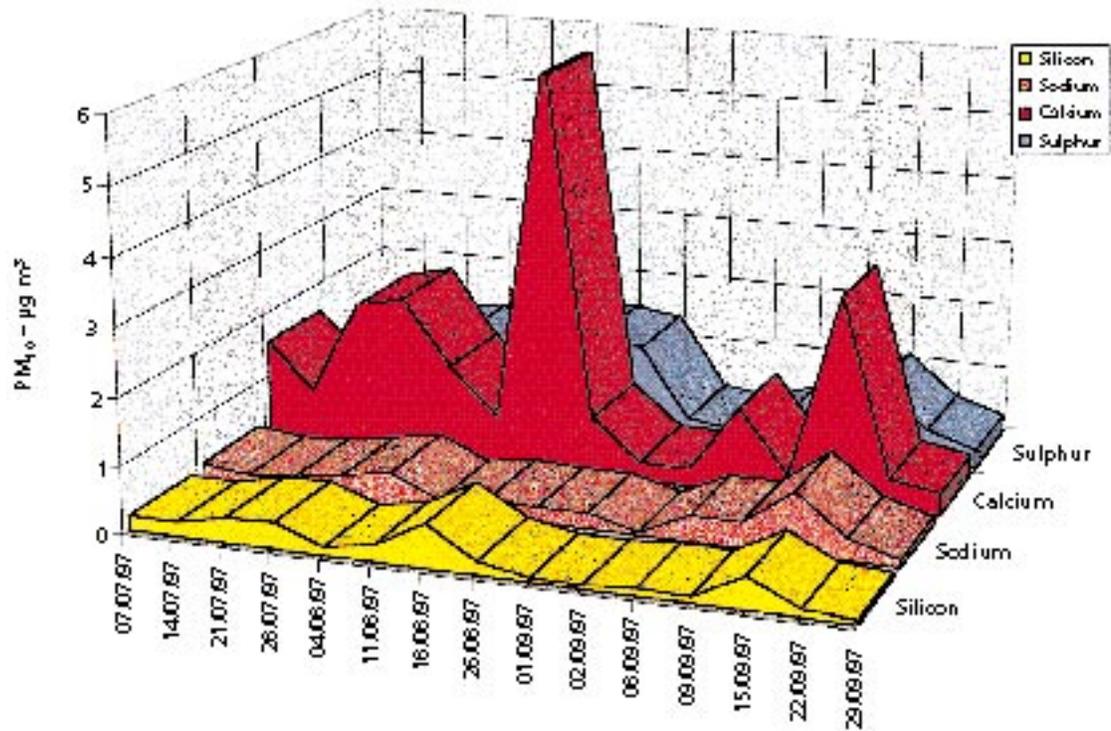
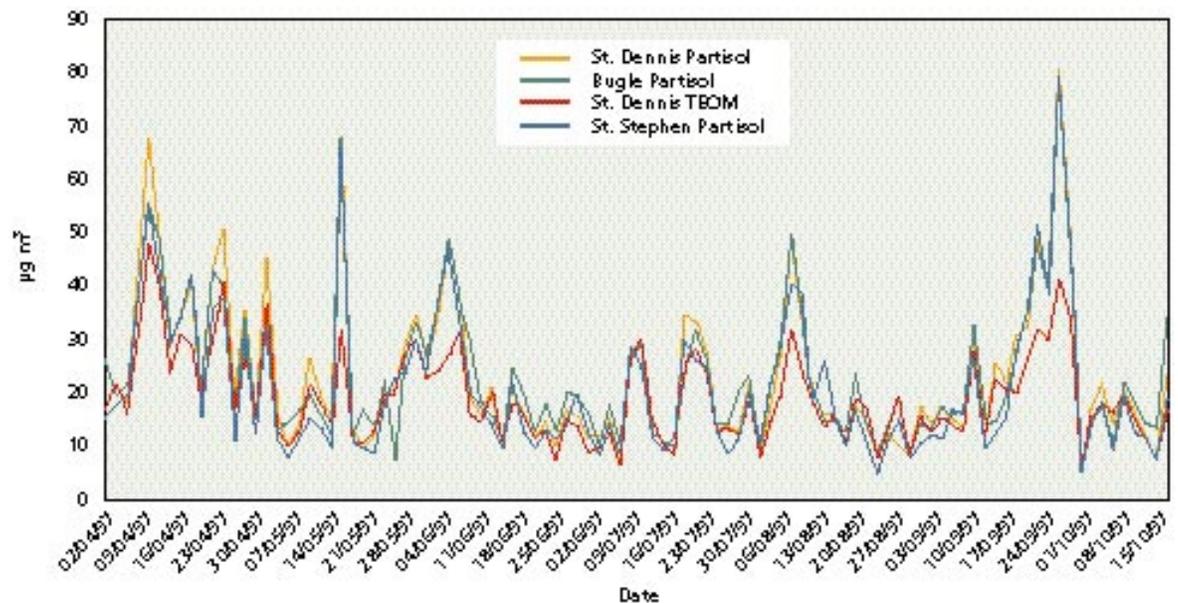


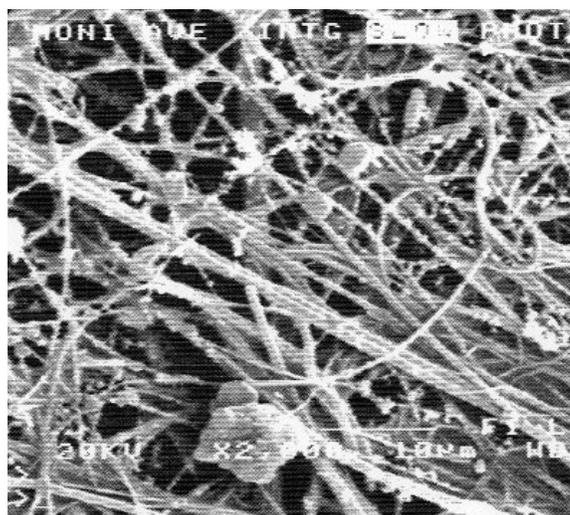
Figure 3.31 Daily PM_{10} concentrations measured at four sites in Cornwall for the period April - October 1997 ($\mu\text{g m}^{-3}$).



three sites from April to October 1997 with partisol instruments (TEOM and Partisol instruments were used at one site, St Dennis) (DETR *et al.*, 1999). Figure 3.31 shows a

comparison of the daily PM_{10} concentrations at these sites. The large mineral extraction related particles causing visible nuisances in the area will not have contributed to the measured PM_{10}

Figure 3.32 Scanning Electron Microcopy pictures of particulate matter collected at St Dennis, Cornwall in March 1997.



concentrations due to their large size. Tentative conclusions from scanning electron microscopy and X-ray fluorescence studies on a small number of the filter samples collected indicated the possible presence of fine particles (less than 2.5 µm in diameter) from the mineral extraction activities. Figure 3.32 shows two SEM pictures from the same filter. The first shows a large agglomeration, which has tentatively been identified with combustion material, while the second includes a number of small particles, which are thought to be kaolinite.

There were no exceedances of the national air quality standard of 50 µg m⁻³ within the data set from the TEOM instrument at St Dennis. There were a number of daily means over 50 µg m⁻³ but since these were measured using a Partisol instrument these cannot be compared against the current national air quality standard.

3.8.4 NORTH DERBYSHIRE: PM₁₀ IN THE VICINITY OF A RANGE OF SOURCES

The authorities in the North Derbyshire area carried out a campaign of PM₁₀ monitoring using Partisol gravimetric samplers at a total of six sites. The locations of these sites were chosen in order to investigate the variations in particle concentrations across a transect of about 8 km between Chesterfield and Bolsover in the vicinity of open cast coal mines, the M1 motorway and a solid fuel processing plant. The sites are listed in Table 3.42 along with the number of days with measured daily mean PM₁₀ exceeding 50 µg m⁻³ and the mean value for the period. Samples were collected on alternate days from November 1996 to October 1997.

Both the period mean concentrations and the number of days with concentrations greater than 50 µg m⁻³ were very similar at all sites except

Table 3.42. Results of North Derbyshire PM₁₀ monitoring November 1996 to October 1997.

Site	Location	Period mean (µg m ⁻³)	Days with concentration >= 50 µg m ⁻³	Number of measurement days
Hasland	urban background	26	22	157
Arkwright	close to opencast mine	18	12	125
Longcourse	close to opencast mine	28	23	153
Duckmanton	close to M1	26	20	137
Shuttlewood	close to solid fuel plant	28	19	116
Castle	urban housing	26	22	130

Table 3.43 Distribution of threshold exceedances for 24 hour average PM₁₀ concentration in Tyne and Wear, March to September 1997.

Number of exceedances	Mill	Souter	NT1	Sund1	Sund2	P6
50 - 99 µg m ⁻³	10	14	6	5	8	0
100 - 149 µg m ⁻³	1	1	0	0	2	0
150 - 199 µg m ⁻³	0	1	0	0	1	0
200 - 249 µg m ⁻³	0	0	0	1	0	0
250 - 299 µg m ⁻³	0	0	0	0	0	0
300 - 349 µg m ⁻³	0	0	0	0	0	0
above 350 µg m ⁻³	0	0	0	0	1	0

Arkwright where the slightly sheltered position of the monitor may have influenced the results. Apart from this, the variation in concentrations from day to day was very consistent across these sites. This indicates that PM₁₀ concentrations are strongly influenced by regional scale meteorological conditions and emissions. Concentrations were generally higher than at the Sheffield city centre national AUN site but Partisol values are generally higher than those monitored using TEOM instruments (see Chapter 2).

3.8.5 MEASUREMENTS AROUND INDUSTRIAL SOURCES OF PM₁₀ IN TYNE AND WEAR

Gravimetric (Partisol) PM₁₀ samplers were installed around individual industrial processes in Tyne and Wear:

1. Process 1 was an opencast and reclamation site. Three monitoring sites were used: "Sund 2" was the nearest residential property in the prevailing downwind direction; "Sund 1" was the nearest residential area on the opposite side of the site.
2. Process 2 was an oil platform construction and repair site using shotblasting to prepare surfaces for coating. Two monitoring sites were used: "NT1", a source of a recent complaint and "Mill", the nearest residential location.
3. Process 3 was a limestone quarry. One monitoring site was used, "Souter" the nearest residential location.
4. A control site, "P6" was installed at the council offices in Washington Village.

A summary of exceedances of various thresholds for the 24 hour average PM₁₀ concentrations is given in Table 3.43. SEM-EDX (scanning electron microscopy - energy dispersive X-Ray) Spectroscopic analysis of some of the filters showed that industrial source particles were clearly present. For example, at the Sund 1 and Sund 2 sites, 10 to 30% of the particles by number were shale particles typical of those from the opencast coal overburden. At Mill, 45% by number of the particles in one sample were iron particles probably derived from shotblasting, although another sample from a day with a similar PM₁₀ concentration was dominated by carbon, sulphate and biological particles.

These data emphasise the importance of local sources to the measurement concentrations of PM₁₀ in certain areas of the UK. It is apparent that very local activities can lead to an exceedance of the current national air quality standard for PM₁₀.

3.8.6 PM₁₀ MEASUREMENTS IN BELFAST

It has been noted that concentrations of PM₁₀ in Belfast are higher than those recorded similarly sized UK cities, for example in 1996 the annual average concentrations in Belfast was 24 µg m⁻³ and the maximum hourly concentration was 280 µg m⁻³ compared with those recorded in Edinburgh 19 µg m⁻³ (annual average) and 130 µg m⁻³ (maximum hourly). These high levels of PM₁₀ in Belfast are probably the result of different fuel usage patterns compared to other UK cities, with greater use of coal for domestic and commercial purposes. In addition, the topography of the area is such that Belfast is surrounded on three sides by hills, preventing effective dispersion of polluted air.

3.9 BIOLOGICAL PARTICLES IN THE ATMOSPHERE

The atmosphere contains a wide range of biological particles. These vary enormously in size and require very different techniques for sampling and

assay according to their size and nature. The most prevalent biological components are thought to comprise the following:

Pollens

Most pollen grains are well in excess of 10 µm diameter, and even allowing for the fact that their aerodynamic diameter may be different from their geometric diameter (for example because they are hollow), few intact pollen grains are likely to be sampled within PM₁₀. There is clear evidence, however, that pollen grains can break up whilst in the atmosphere generating fragments in the PM₁₀ range extending right down to below 1 µm. Whilst these can be assayed by biochemical techniques, knowledge of their atmospheric mass is lacking (Emberlin, 1995).

Fungal Spores

Many fungal spores also exceed 10 µm in diameter, but spores of some abundant species do extend in size into the 5-10 µm range, and hence contribute to coarse particle mass in the atmosphere. There is a typical seasonal pattern in fungal spore abundance with a maximum in the summer and a minimum in the winter.

Bacteria and Viruses

These are mostly in the fine particle range with bacteria being about 1 µm diameter and viruses considerably smaller. Whilst number concentrations in the atmosphere can be appreciable, the mass represented is relatively modest.

Matthias-Maser and Jaenicke (1995) have quantified biological aerosol in the size range from 0.4 – 80 µm diameter at a site on the outskirts of Mainz in Germany. They express their results as the number distribution of total aerosol particles and of biological particles. The percentage biological particles across the range 0.4 – 10 µm diameter is typically about 10% with little seasonal variation. If it is assumed that the density of biological particles and other aerosol particles are similar, this percentage should apply to mass also. Extrapolation to an estimation of a contribution to mass in the less than 2.5 µm fine particle size fraction is difficult due to the limited range of the measurements of biological particles.

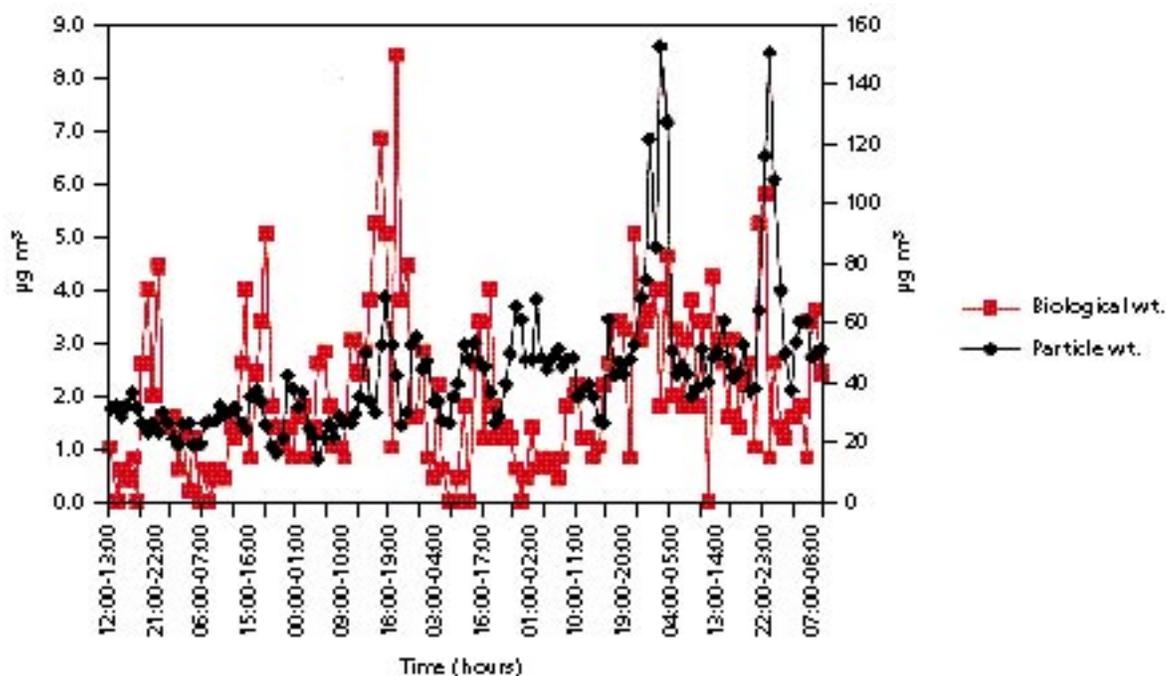
However, in the 2.5 – 10 µm coarse particle range, a figure of 10% of mass at the Mainz site being due to biological particles would appear appropriate. Caution must be used in applying this observation to measurements with the TEOM analyser since much of the volume of a biological particle is likely to be comprised of water which will presumably be largely removed in the drying stage of the TEOM.

In a study conducted in suburban Birmingham, Phillips (1997) used a Burkard spore trap to collect particles for identification with a light microscope. Over a three week period of measurements fungal spores were counted. These comprised about one in four particles in the 5 – 10 µm size range, and more than half of particles in the size range greater than 10 µm. The contribution to PM₁₀ mass is critically dependent upon the relative sizes of the biological and non-biological particles and since this was not recorded, it is not possible to make any convincing estimate of the contribution of spores to PM₁₀. Since fungal spore numbers tend to peak in the UK in the summer, and this study was carried out in August 1997, it is likely that the results represent an upper limit to the contribution to total particles in any case.

Biological particulates in central London were monitored over the period 16th - 22nd July, 1996, as part of a larger study which seeks to characterise, enumerate and apportion to sources all particle types down to 0.5 µm diameter on a seasonal basis between 1996 - 1997. Particles were collected using a Burkard spore trap, which is co-located with the AUN central London hydrocarbon monitor site and is 300 m from the AUN site at Russell Square.

Fungal spores have an average density of about 1.1 µg cm⁻³ (Gregory, 1973), and this has been used to estimate mass concentration values. Within the July period, hourly mass concentration of fungal spores ranges between 0.2 µg m⁻³ up to a maximum 8.4 µg m⁻³, with an average of 2 µg m⁻³. The graph below shows hourly changes in estimated mass concentrations of fungal spores (primary y axis) plotted alongside total PM₁₀ mass

Figure 3.33 Estimated biological mass and measured particle mass (by TEOM) for the period July 16th - 22nd, 1996 at the AUN site in Russell Square, London.



(secondary y axis), as measured by the TEOM at the AUN site in Russell Square (Figure 3.33).

There is very little overall correlation between the two graphs, although several total PM_{10} weight peaks are coincident with increases in biological particle mass. The dominant spore type present during this period was *Cladosporium*, exhibiting daily increases related to increasing daytime temperatures (Mackay, 1998). These increases are relatively small however, in comparison to increases in the total PM_{10} fraction, but the full data set for this week of Summer 1996 indicates that biological material contributed on average 5% of total PM_{10} mass and up to 12% of hourly average PM_{10} mass. These figures are likely to represent the upper limits of mass contribution, given that spore numbers are greatest during the summer.

3.10 COMPOSITION OF PARTICLES

3.10.1 INTRODUCTION

The chemical composition and size distribution of specific chemical components of airborne particulate matter can provide valuable insights into the sources of

such particles. Such information can be used both in qualitative identification of sources and in quantification of the contribution of such sources to the mass of particulate matter. Chemical data are therefore crucial to understanding primary and secondary particulate matter and to the receptor modelling studies described in Chapter 5. In this chapter two comprehensive studies of the composition of particulate matter conducted in Leeds and South Wales are described, with particular reference to the results pertaining to primary components of the particles.

3.10.2 MEASUREMENTS FROM LEEDS

3.10.2.1 Introduction

The chemical composition and size distributions of atmospheric particles were measured at four sites in and around Leeds during the period March 1995 to March 1996. Weekly samples were taken using Andersen cascade impactors which collect 9 size ranges - 8 impactor stages with 50% cut-off diameters ranging from 0.43 to 9 μm plus a back-up filter representing particles finer than 0.43 μm . The impactors were fitted with pre-separators which make them approximately equivalent to a PM_{10} sampler.

The impactor sites were:

1. University of Leeds - Roadside. A ground floor window, 5 m above busy road near traffic lights.
2. University of Leeds - 5th floor. Almost vertically above site 1 at a height of approximately 25 m.
3. Leeds AUN Site - On the roof of the hut adjacent to the TEOM air intake. This site is about 1 km down the road from the University sites and 30 m from the A660 trunk route out of Leeds.
4. Rural Site - Haverah Park. 20 km N of Leeds, 7 km W of Harrogate. About 3 m above ground on the roof of a hut in open moorland, 0.5 km from minor road.

In addition to particle mass concentration, analyses were undertaken for anions, ammonium, acidity, metals (Pb, Fe, Zn, Ca), PAH (the 16 US EPA priority list) and, in addition, some information about volatile matter and carbon content was obtained. Not all analyses were attempted each week.

3.10.2.2 Total Mass Concentrations

There was strong consistency in the total mass collected at the three urban sites with the usual order being ground floor > fifth floor \approx AUN site. The roadside enhancement relative to the urban background was 5-10 $\mu\text{g m}^{-3}$ on a weekly-average basis but previous work (Clarke *et al.*, 1996) showed enhancements of 25-30 $\mu\text{g m}^{-3}$ (as weekly averages) during weekday daytimes associated with heavy traffic. (But note, those were TSP not PM_{10} measurements). The rural site collected on average about 70% of the mass collected by the urban sites representing an urban enhancement of about 15 $\mu\text{g m}^{-3}$ as a weekly average.

The impactors appeared to collect up to a factor of two more mass than the Leeds AUN TEOM. It is suggested that the differences relate partly to moisture or organic vapour collection by the impactor substrates that is not compensated for in

the normal drying/weighing procedure and the data for Figures 3.34-3.37 below have been subject to a correction for this effect based on auxiliary experiments. The differences also partly relate to volatile matter loss in the TEOM. The TEOM operates at 50°C and there is significant mass loss relative to any sampler operating at ambient temperature - sometimes as low as 0°C during the Leeds survey (see Chapter 2).

3.10.2.3 Particle Size Distribution by Mass.

On average 10-20% of the urban mass concentration is in the finest particles <0.43 μm , 50% below about 1.5 μm , and 80% of the PM_{10} mass is below 5 μm . The average cumulative frequency distributions for 17 weeks when data were available at all sites are shown in Figure 3.34. The balance between coarse (2.1 - 10 μm) and fine particles ($\leq 2.1\mu\text{m}$) is shown in Figure 3.35. Typically 60-70% of the PM_{10} mass is in the $\text{PM}_{2.1}$ fraction and 30-40% in the coarse range with the larger proportions of coarse material in the summer months. This is consistent with the work of Harrison *et al.*, (1997) in Birmingham who found about 60% of PM_{10} was $\text{PM}_{2.5}$ overall but they observed a sharper summer to winter variation (summer about 50% and winter about 80% $\text{PM}_{2.5}$).

Figure 3.34 Cumulative size distributions for the mass concentration of particulates from the Leeds four site survey (average of 17 weekly samples).

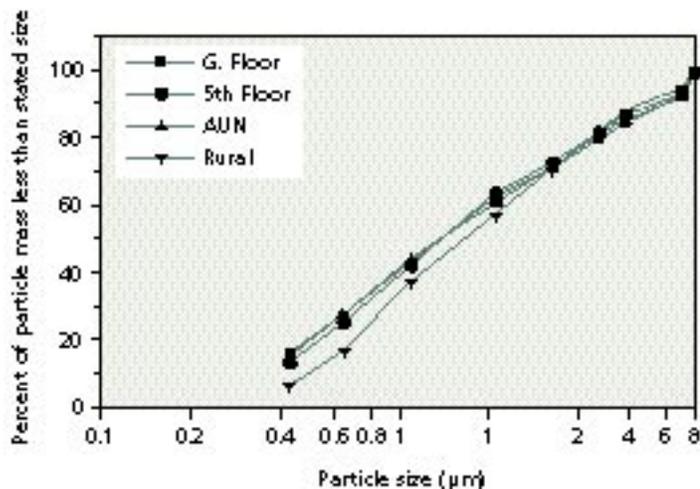


Figure 3.35 Variations in the weekly average coarse (2.1-10 µm) and fine (<2.1 µm) particle contributions to PM₁₀ from the Leeds 1995/6 survey.

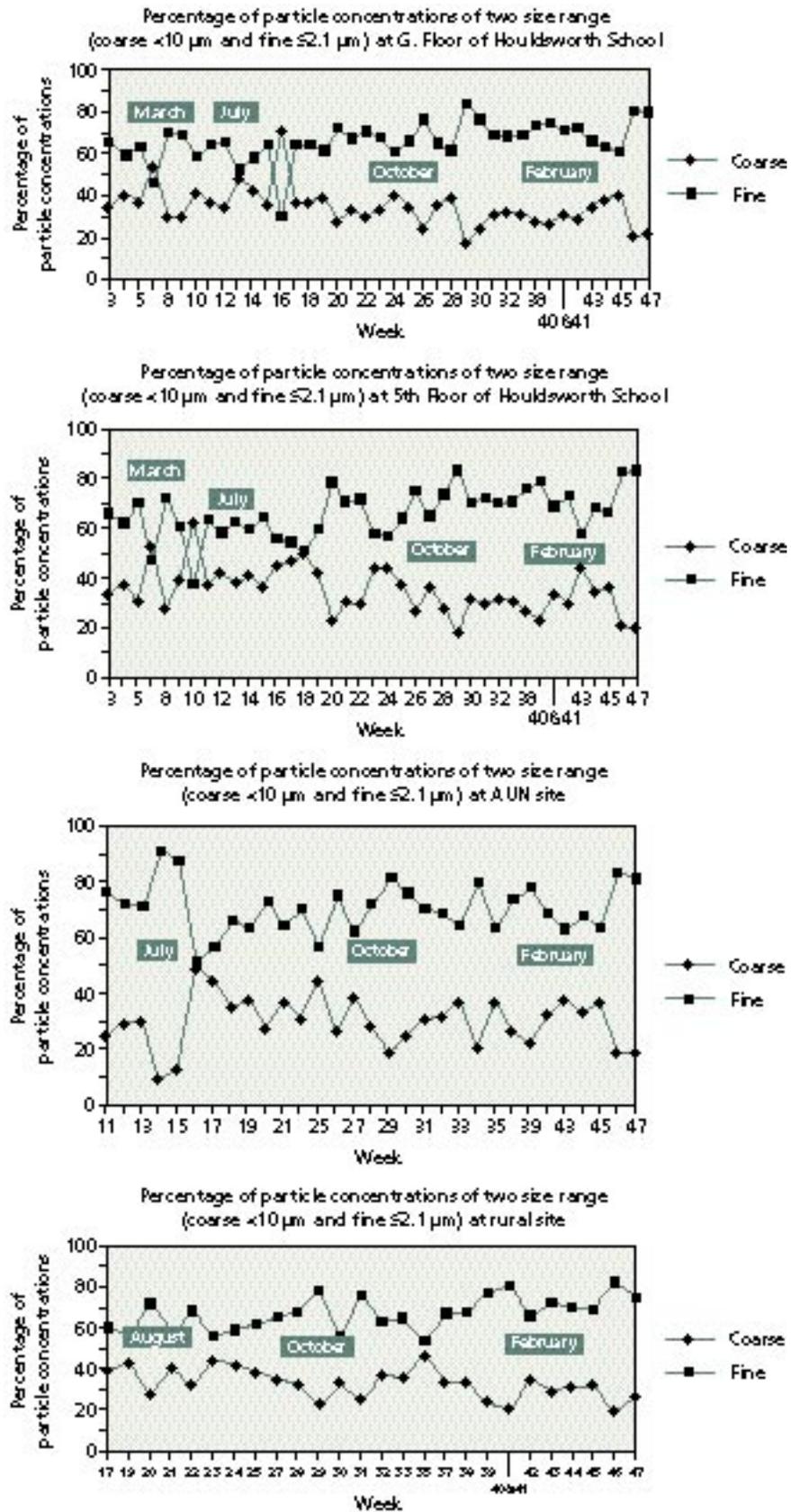
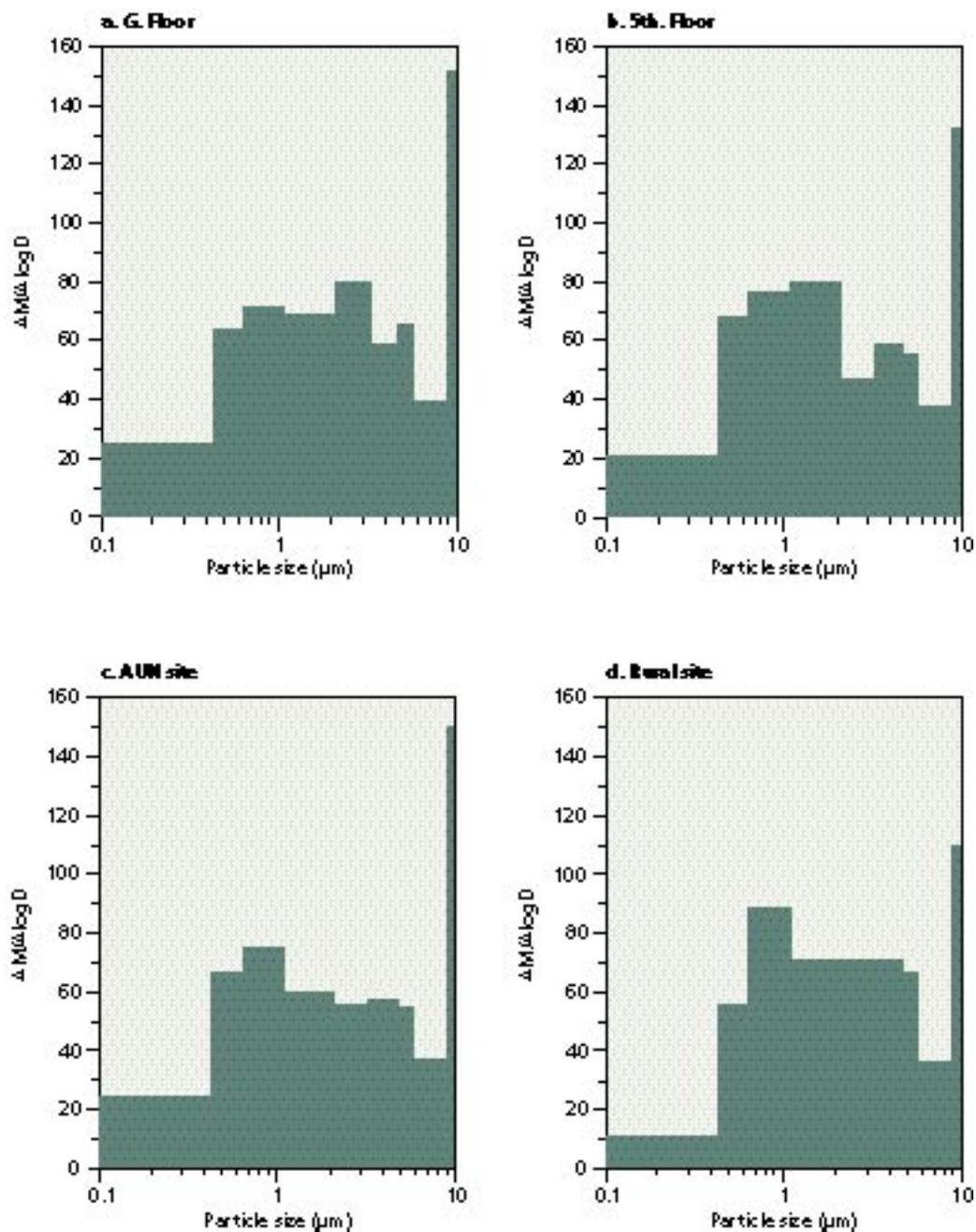


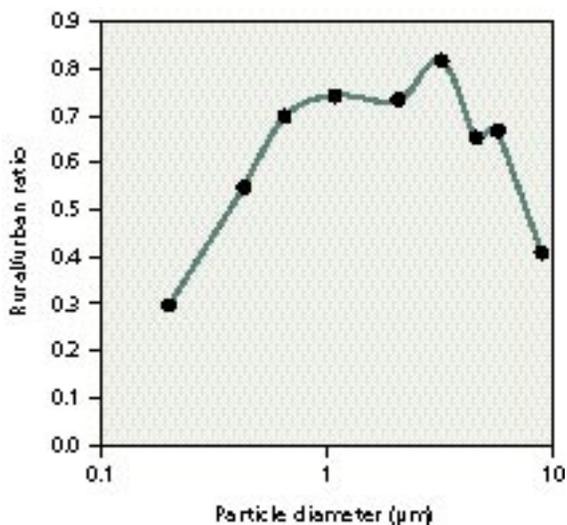
Figure 3.36 Size distribution of total particulate mass based on percentage of mass in each size range at four sites in the Leeds 1995/6 survey.



1. Since there is no lower size limit to the particles collected on the back-up filter an arbitrary value of 0.1 μm has been assumed. The first band on these plots covers the range 0.1 - 0.43 μm .

2. In the plots of $\Delta M / \Delta \log D$ a peak at 9-10 μm often appears. The data presentation is based on the assumption that the pre-separator removes all material >10 μm and the top stage of the impactor removes material >9 μm . In practice, the top stage probably collects some particles larger than 10 μm which have found their way through the pre-separator. ΔM (the incremental mass concentration) is increased while $\Delta \log D$ (where D is the aerodynamic diameter of the particle) is very small for this stage resulting in the anomalous appearance of the graph.

Figure 3.37 Rural to urban ratio of particle mass concentration as a function of particle size from the Leeds 1995/6 survey.



Plots of $\Delta M/\Delta \log D$ (ΔM is the incremental mass concentration and D is the aerodynamic diameter of the particle) are shown in Figure 3.36. Sometimes a clear bimodal pattern is seen as in the averaged data for the 5th floor site, with one peak around $1\mu\text{m}$ and another around $4\text{-}6\mu\text{m}$. Sometimes the two modes are merged.

The rural site has lower concentrations across the whole range but this is most noticeable in the finest and coarsest size ranges, Figure 3.37. Urban aerosols have a higher proportion of vehicular (and possibly industrial) emissions which are in the very fine size range compared to the rural site. The rural aerosol is also depleted in the larger particles which correspond to the effects of human activity including road dust raised by vehicular motion, construction activities and industrial emissions.

3.10.2.4 Carbonaceous Content

An assessment of volatile matter and elemental carbon has been made using a thermo-gravimetric analyser (TGA). Volatile matter is defined as the sample mass loss up to 500°C under nitrogen. 'Volatiles' therefore includes not only hydrocarbons but also all ammonium salts and possibly some additional moisture not removed in

the filter drying procedure. The elemental carbon is then the subsequent mass loss on switching the gas to air. The technique was difficult to apply because it requires about 1 mg of particulate sample on a small area of filter in order to be able to mount it in the TGA apparatus and special methods had to be used to obtain useful data.

For the one full set of data analysed the amount of elemental carbon as a fraction of the total collected mass of PM_{10} was as follows:

Ground floor:	21%;
Fifth floor	17%;
AUN site	27%;
Rural site	9%.

Harrison *et al.* (1997) found the average fraction of elemental carbon in Birmingham PM_{10} to be 18%. The carbon is predominantly in the finer size ranges. The percentile points of the cumulative frequency distributions shown in Figure 3.38 for the urban sites were: 50% below $0.6\mu\text{m}$ and 80% below $3\mu\text{m}$. The rural site seems to lack the very fine carbon component since the back-up filter showed no measurable carbon.

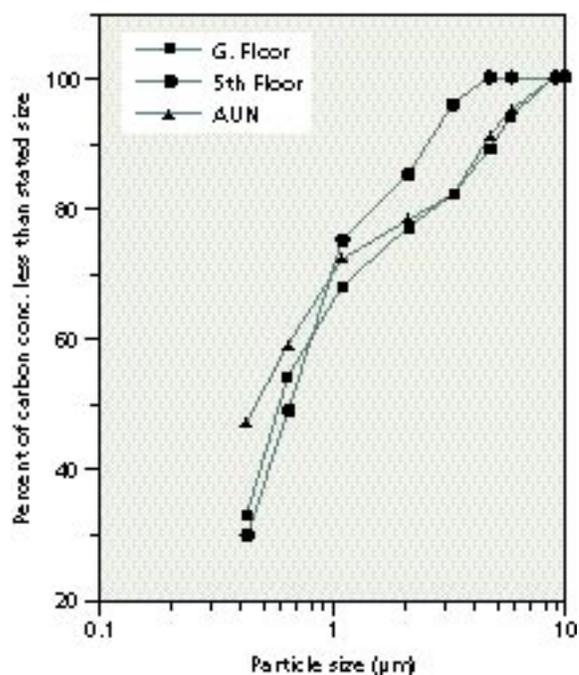
The elemental carbon would be expected to correlate with optical measurements of black smoke since the smoke measurement is dominated by highly absorbing black particles, of which the most important are from diesel smoke.

3.10.2.5 Sea-Salt and Road Salt

There has been no systematic survey of sea salt aerosols in the UK. Some of the early data up to 1983 are surveyed in the paper by Willison *et al.*, (1989) and these are combined with recent data on the size distribution of chloride aerosols from the Leeds survey in this section. Further information on the sea salt contribution appears in Sections 5.5.1 and 5.5.2 and information on sea-salt displacement in Section 4.1.3).

Not all the particulate chloride measured in the atmosphere is marine in origin. Some arises from

Figure 3.38 Carbon cumulative size distribution for three urban sites from the Leeds 1995/6 survey.



reactions of gaseous HCl which is emitted from coal fired power stations, incinerators and some chemical processes. One *secondary* chloride arising from these reactions is ammonium chloride but this is highly volatile. In winter it is detectable in the fine aerosol fraction, in summer it disappears. To assess the marine fraction of the chloride it is conventional to measure the sodium content and to estimate the marine chloride contribution using the seawater Cl/Na ratio. The UK surveys covered in the Willison paper indicated marine chloride fractions ranging from 43-76%.

Another potential source of chloride is the resuspension of road salt used in winter. For example in February 1983 a peak daily value of $7.3 \mu\text{g Cl m}^{-3}$ was recorded in Leeds some 4-5 times the annual average concentration. There was no corresponding peak at a nearby rural site. However this was an exceptionally long cold spell with dry conditions resulting in visible white deposits on the roads.

Even under normal conditions with no potential for a road salt contribution there appears to be a

slight urban excess of chloride which may relate to local HCl emissions or possibly the resuspension of sea-salt deposited on roads or other paved areas. The previous Leeds survey (Willison *et al.*, 1985) gave annual average levels of $2.0 \mu\text{g Cl m}^{-3}$ (urban) and $1.5 \mu\text{g Cl m}^{-3}$ (rural) for 1982-3. More recent data are indicated in Table 3.44 which shows results for weekly samples. For three of the weeks the rural chloride is 75-80% of the urban value consistent with the earlier data. For the fourth week in January 1996 the difference is much larger - indicating an additional road salt contribution of the order of $1 \mu\text{g Cl m}^{-3}$.

Taking a nominal 75% of the measured chloride as marine in origin and the sea-salt/chloride mass ratio as 1.816, the corresponding sea-salt contributions to the atmospheric PM_{10} mass can be estimated (Table 3.44). It amounts to $1\text{-}3 \mu\text{g m}^{-3}$ for the inland area of Leeds and is 2-3 times higher in winter than in summer. At a coastal location such as Lancaster the 1979-81 average was $4.2 \mu\text{g Cl m}^{-3}$ (76% marine) corresponding to over $5 \mu\text{g m}^{-3}$ of sea-salt (Harrison and Pio, 1983).

Table 3.44 Urban and rural chloride concentrations in PM_{10} mass (weekly sampling period).

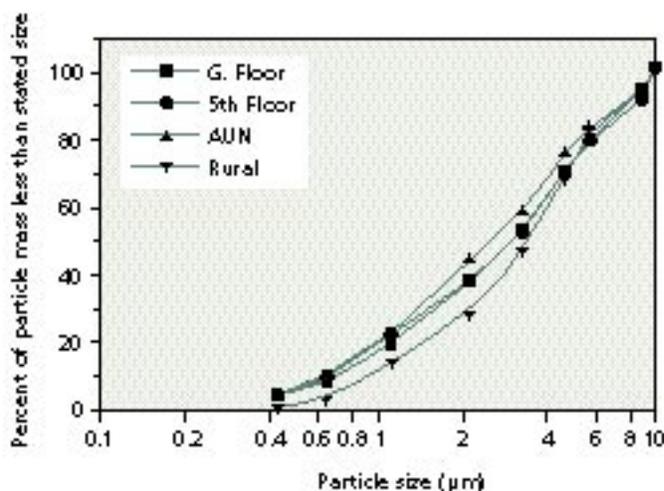
Week beg.	Urban Chloride+	Rural Chloride	Urban Sea salt mass*
	$\mu\text{g m}^{-3}$	$\mu\text{g m}^{-3}$	$\mu\text{g m}^{-3}$
15-May-95	0.52		0.7
21-Jun-95	0.53		0.7
05-Jul-95	1.17		1.6
26-Jul-95	0.5		0.7
09-Aug-95	0.62	0.5	0.8
18-Oct-95	1.27	0.99	1.7
15-Nov-95	2.6	1.94	3.5
17-Jan-96	2.89	1.26	see text
8 week Average			
1995-1996	1.26	-	1.7
Annual Average**			
1982-1983	2.0	1.5	2.7

+ average of three sites

* Sea-salt mass: $0.75 \times 1.816 \times \text{Cl}$ (assumes 75% of Cl is marine and total salt/Cl ratio= 1.816)

** These figures are actually for PM_{15} not PM_{10}

Figure 3.39 Cumulative size distribution for chloride from the Leeds 1995/6 survey.



Sea-salt aerosols are relatively coarse with a peak in the size distribution in the 4-6 µm range. Since the fine chloride is minimal in summer, but significant in winter, it is probably a good approximation to apportion all PM₁₀ chloride >2.5 µm to sea salt and all fine chloride <2.5 µm to NH₄Cl. This can amount to around 20% of the total chloride in winter. Results from the recent Leeds survey are shown in the cumulative frequency curves (Figure 3.39) and ΔM/ΔlogD plot (Figure 3.40). The overall average urban chloride size distribution gave 20% below 1.2 µm, 50% below 3.2 µm and 80% below 6.0 µm.

3.10.2.6 Tyre Dust

Very little experimental information is available for the contribution of tyre wear to ambient particulate concentrations. Emission inventories have tended to rely on emissions factors derived in the USA which are of questionable relevance to the UK.

Measurements of tyre dust in airborne particles and in road dust were made in Leeds during summer 1997 (Pollard, 1997). The air samples were taken using an open-faced 37mm GF/F glass fibre filters shielded by an M-type sampler hood. The particles collected will therefore include some which are coarser than PM₁₀. Road dust samples were swept from approximately 1m² of road in

various locations on occasions when there had been at least 4 days without rain, sieved to remove coarse grit and then crudely size segregated to <10 µm by drawing the particles through the pre-separator of an Andersen cascade impactor and collecting the dust passing through on the back-up filter.

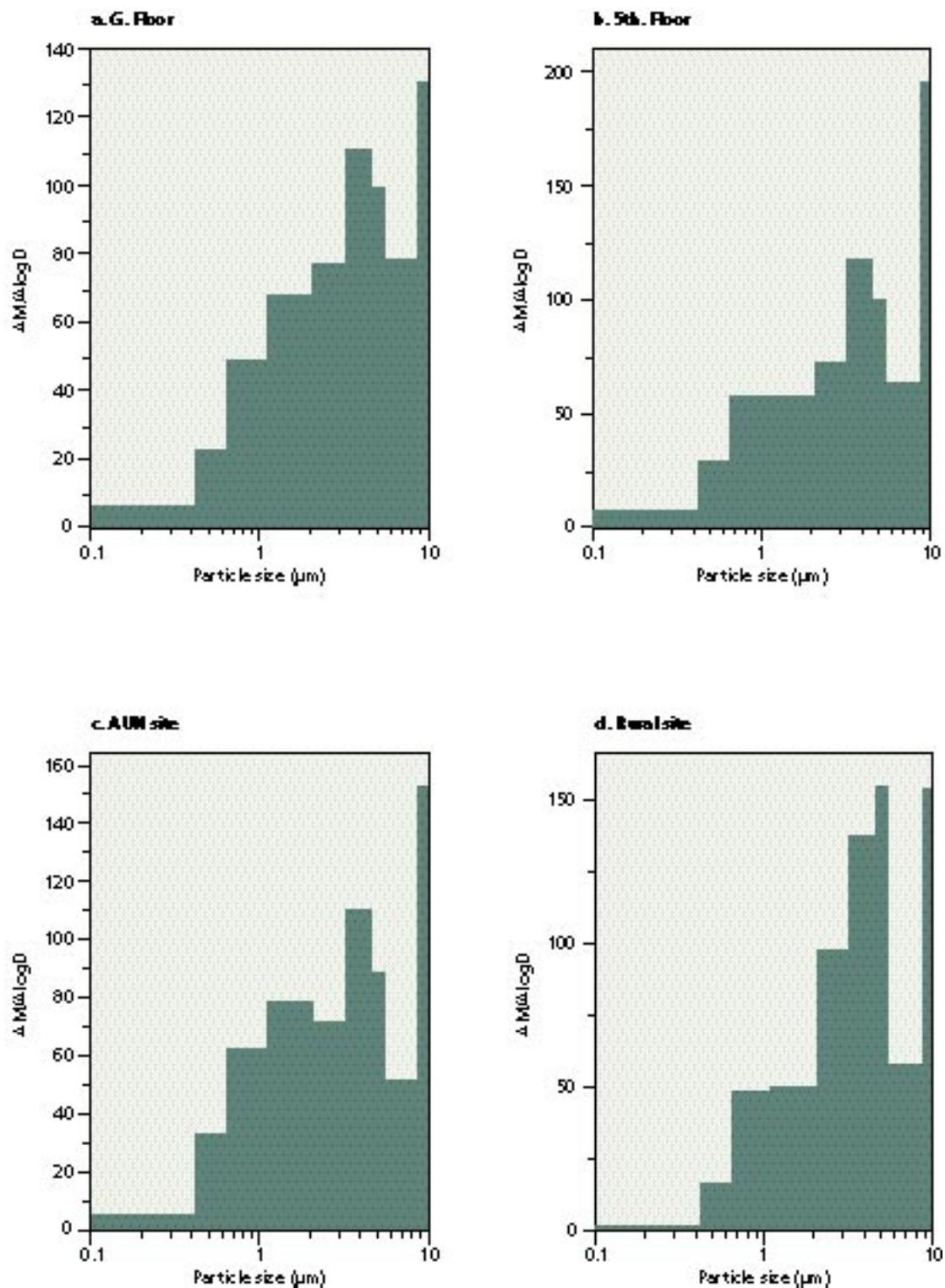
The analytical technique used is pyrolysis Gas Chromatography (Kim *et al.*, 1990). Samples are heated rapidly to 600°C in the pyroprobe causing organic compounds to be vapourised. Generally, the most noticeable components are the fuel + lubricating oil hydrocarbons from C₁₅ upwards, plus a small amount of light gaseous compounds C₂-C₄ which arise from thermal decomposition processes. In the presence of tyre dust two specific compounds are readily resolved - di-pentene C₁₀H₁₆ (or limonene) and styrene C₈H₈. Di-pentene is consistently observed in all GC traces from airborne particles and arises from natural rubber or isoprene-based synthetic rubbers used for heavy duty tyres on buses and trucks. Styrene appears irregularly and arises from styrene/ butadiene rubbers used mainly for automobile tyres. By measuring amounts of the characteristic compound released into the GC by a known mass of the pure rubber compound it becomes possible to calculate the fraction of tyre rubber in the original dust sample.

The results based on isopentene analysis are as follows:

Airborne dust rubber fraction	0.38% (range 0.19 to 0.73%, 8 samples)
Airborne rubber concentration	0.28 µg m ⁻³ (range 0.12 to 0.73 µg m ⁻³)
Road dust rubber fraction	0.61% (range 0.31 to 0.83%, 6 samples)

The national emissions inventory currently quotes emissions of 0.6 ktonne per annum for tyre wear compared with total exhaust emissions of 51.9 ktonne, a fraction of just over 1%. Contributions to airborne particulate matter of the order of a few tenths of a percent are therefore to be expected and this is consistent with the Leeds data.

Figure 3.40 Size distribution of chloride based on percentage of Cl in each size range at four sites in the Leeds 1995/6 survey.



3.10.3 MEASUREMENTS FROM SOUTH WALES

The composition of airborne particulate matter is complex and one approach to its characterisation is to employ sequential analytical methods and a variety of techniques to obtain as complete a chemical and physical picture as possible of the material before an attempt is made to attribute its origin to particular sources.

In the South Wales study the approach has been to subdivide sampled airborne particulate material into four fractions for the purpose of analysis. These fractions conveniently divide the samples into component parts which are useful for identifying the sources from which they most probably originated. They consist of:

- (i) The aqueous soluble fraction
- (ii) The fraction soluble in an organic solvent
- (iii) The insoluble carbonaceous fraction
- (iv) The insoluble inorganic fraction

The water soluble fraction is analysed by ion chromatography and inductively coupled plasma emission spectroscopy to identify its main components. Water soluble organic material can also be determined by aqueous carbon analysis. The mass of dissolved material is determined using the weight loss of treated samples, while the quantities of dissolved species can be combined for comparison with the latter figure. The insoluble carbonaceous fraction is quantified in samples after removal of the water and organic solvent soluble material. It is determined as a weight loss from samples after treatment by low temperature plasma ashing. These weight loss values can be compared with readings of carbon content obtained from carbon analysis equipment.

The residue of samples after treatment with water and organic solvents followed by low temperature ashing provide samples of insoluble inorganic material for analysis. A variety of analytical techniques can be applied to this residue to identify its components, including quantitative analytical

electron microscopy, X-ray fluorescence and diffraction procedures.

Having obtained the analysis of the component parts of a sample, the results can then be utilised to assess their origin.

3.10.3.1 Source Apportionment

The main components of the water soluble material detected in most urban air particulate samples are sulphate, chloride, sodium and ammonium ions, whilst magnesium, nitrate and heavy metals may also be present in significant quantities. These ionic species derive from salts originating from the sample leading to a charged balanced neutral solution in which the cationic and anionic equivalents offset each other.

When the composition of water soluble extracts from a number of air particulate samples is compared with the semiquantitative composition of sea water, it can be seen that the contribution of this source to the aqueous soluble fraction may be significant. The sea as a source can explain the high quantities of chloride and sodium present, while the ammonium and sulphate is more probably derived from secondary aerosols.

The organic solvent soluble fraction of the suspended particulate material from urban air is known to contain a large number of organic compounds. This soluble organic fraction is present in significant amounts often in excess of 40% of the total sample weight. The source of this material can be attributed almost solely to traffic in urban areas. In a similar manner the source of the insoluble carbonaceous fraction of urban airborne particle samples can also be related directly to traffic situations, both the soluble organic and insoluble carbonaceous material being intimately associated in the suspended particles.

The inorganic insoluble fraction is often a complex mixture of material from a variety of sources, either natural or anthropogenic. In certain circumstances a small number of major industrial sources may define the composition of this fraction. In others it will be a

very heterogeneous mixture of many local and imported sources. Table 3.45 contains a listing of the major oxide weight percentage compositions of two inorganic insoluble fractions of airborne particles. Also included in the table is a list of an average composition of the earth's crust. It can be seen that the contribution from natural sources will be predominantly silicate rich and that industrial sources may add significantly to the non-silicate component. It is possible with this fraction to partition the chemistry of the sample with reasonable accuracy between various sources.

Table 3.45 Semi-quantitative composition of insoluble inorganic fractions expressed as elemental oxides (weight %).

	Earth's Crust	Sample 1	Sample 2
Na ₂ O	3	1	1
MgO	4	2	2
Al ₂ O ₃	16	14	10
SiO ₂	60	42	22
K ₂ O	3	2	1
CaO	6	3	9
TiO ₂	1	2	<1
V ₂ O ₅	<1	-	<1
Cr ₂ O ₃	<1	1	<1
MnO	<1	1	1
FeO	7	29	51
CoO	<1	-	<1
NiO	<1	-	<1
CuO	<1	-	1
ZnO	1	1	1
Pb ₃ O ₄	<0.002	0.1	0.46

Samples collected:

1. Cardiff City Centre
2. In the vicinity of a steel works, Margam, South Wales

The estimated overall composition of a Cardiff urban airborne particle sample collected with a High Volume sampler and PM₁₀ sampling head over a five day period is given in Table 3.46. This was produced by drawing together all the partial compositions of the particle sample, including both soluble and insoluble fractions. The results are expressed in milligrams per gram of material sampled, and represent a semiquantitative presentation of particle chemistry with factors such as moisture content being neglected. Further confirmation of the source of material in the insoluble inorganic fraction can be provided by the analysis and morphology of individual particulate

material and the quantification of the crystalline components it may contain.

In terms of apportionment, the most difficult fraction to subdivide between sources is the water soluble fraction. Although its composition is not too complex, it owes the origin of its components to a variety of sources, e.g., ammonium sulphate may originate as a primary component of aerosols and also as a secondary product.

The carbonaceous plus soluble organic fractions will possess the most stable composition. The relative proportions of the two fractions together with the composition of the latter provide a direct link to the major sources of this material.

Applications of this total approach to the analysis of airborne particulate matter are scarce, but it provides many insights into the contributions to airborne particle mass.

Table 3.46 Semi-quantitative composition of a gram of airborne particulate matter collected in the Cardiff area (mg) over a period of a week.

	Soluble Material	Insoluble Material	Total
Sol. org.+ carb.	-	-	555
Na	14	2	16
NH ₄ ⁺	8	-	8
Mg	3	6	8
K	11	8	19
Ca	-	12	12
F	<1	-	<1
Cl	19	-	19
NO ₃ ⁻	3	-	3
SO ₄ ²⁻	41	-	41
Al	<1	49	49
Si	<1	144	144
Ti	-	8	8
Cr	<1	4	4
Mn	-	4	4
Fe	<1	100	101
Co	<1	-	<1
Ni	<1	-	<1
Cu	-	1	1
Zn	<1	4	4
Pb	-	3	3

* The Insoluble fraction components are expressed as elemental oxides.

3.10.4 COARSE PARTICLES - ORIGINS AND SPECIATION

The coarse particle fraction ($>2.5\mu\text{m}$) is shown elsewhere (see section 3.5 and 3.10.2) to contribute 30-50% of the total PM_{10} . Typical levels of the most common constituents are presented here with a discussion of their origins.

Sea salt is predominantly in the coarse mode and contributes typically $1\text{-}3\ \mu\text{g m}^{-3}$. Of the secondary components nitrate (e.g. NaNO_3) is predominantly in the coarse mode, especially in summer while sulphate (e.g. $\text{CaSO}_4\cdot 2\text{H}_2\text{O}$) is distributed more evenly between coarse and fine modes (Figure 4.13). The long range transport of both natural and industrial particles may occasionally be significant, for example, when material from Saharan dust storms reach the UK. However, such events are infrequent.

Experimentally it is quite difficult to distinguish local industrial particles, eg. coal ash, construction-derived particles such as cement, limestone or gypsum $\text{CaSO}_4\cdot 2\text{H}_2\text{O}$, and components of windblown soils. All of these may deposit on the ground and be resuspended, and they contribute to road dust. Analysis may be undertaken for the *elements* by a variety of techniques or for the specific chemical *compounds* by X-Ray Diffraction (XRD).

The largest body of available data in the UK is for trace metals including Pb, Fe and Zn. One of these, iron, is expected to be predominantly from crustal sources. Typical long term average concentrations of iron in UK cities are from 0.3 to $1\ \mu\text{g m}^{-3}$ (M-type samplers). Recent Hi-Vol data for Leeds (Tolebi, 1997) which were analysed for metals including Fe and Al showed that in this particular urban area

- the Al levels are slightly higher than the Fe levels
- the Fe to Al ratios varied within a factor of 0.5 to 1.5 of the accepted crustal ratio
- night-time levels of both components were approximately half the day-time values.

These findings are indicative of local sources with a crustal or similar composition.

Taking the concentration of Fe as 5.6% in crustal material the total mass would be 18 times the iron mass. Taking this factor with the Fe levels indicated above gives atmospheric mass concentrations, measured as total suspended particulates, of crustal material of $5\text{ - }18\ \mu\text{g m}^{-3}$. The contribution to PM_{10} would be somewhat less than this. Whilst this type of calculation gives the right order of magnitude it is unfortunately too uncertain to be of great predictive value. For example, data for Cardiff (Table 3.47) are indicative of much higher proportions of iron in atmospheric particles (relative to other metals) than are found in Leeds. On the other hand a recent analytical survey of the resuspendable fraction of road dust in Leeds gave iron contents of 2-3% - only half the global figure for crustal material.

Specific chemical components of the coarse aerosol can be obtained by XRD techniques. Data for Toronto (Sturges *et al.*, 1989) identified the following components of TSP in addition to sea salt and secondary components:

Component	Winter 1986 $\mu\text{g m}^{-3}$	Summer 1986 $\mu\text{g m}^{-3}$
Quartz	0.51	1.37
Calcite	1.19	2.43
Dolomite	0.71	1.41
Gypsum	0.32	0.13
Feldspar	0.21	1.07
TSP	19.4	26.2

Data for Leeds confirm typical calcite concentrations of about $1\ \mu\text{g m}^{-3}$ (Clarke and Karani, 1992) and confirm that it is predominantly in the coarse mode. The total calcium levels recorded in the Leeds impactor survey also ranged up to about $1\ \mu\text{g m}^{-3}$. Examination of size fractionated suspended particulate material in the Cardiff area has shown increasing levels of calcium, silicon, aluminium and iron, with increase in aerodynamic particle size. This is especially true of the transition in size from $2.5\ \mu\text{m}$ and above.

These increases are accompanied by corresponding reductions in the levels of sulphur, chlorine and elemental carbon content of size fractions. Increases in silicon and aluminium contents have been shown by XRD to be associated with the presence of silicate minerals including muscovite, kaolinite and quartz. The calcium content of coarse fractions are closely related to the carbon content of these fractions in the form of carbonate minerals which have also been identified by X-ray diffraction results.

The relative concentration of a number of elemental components from suspended airborne particulate samples collected in Cardiff is contained in the Table 3.47 which illustrates the compositional variations described.

Table 3.47 Relative concentrations of elemental components in size fractions of a suspended airborne PM₁₀ sample from Cardiff.

Component	>2.5 µm	1 - 2.5 µm	<1 µm
SiO ₂	22.32	6.64	3.26
Al ₂ O ₃	6.14	2.44	1.19
CaO	11.50	2.88	0.69
FeO	23.88	7.75	2.27
SO ₃	20.09	72.43	83.99
Cl	9.38	0.78	0.69
C (total including CO ₃)	6.70	7.09	7.91

In summary, the coarse fraction contains significant amounts of mineral compounds which may have natural or industrial sources. These compounds are normally fairly local in origin and may be transported from the sources either directly, through the atmosphere, or indirectly, following deposition and resuspension. The QUARG Third Report indicated that these contributions could be just over one half of the coarse particle fraction and it is not possible, with current knowledge, to give a more precise estimate.

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