623. In conclusion, it is clear that construction activities can give rise to elevated PM₁₀ concentrations, both in terms of annual mean and 24-h exceedences of 50 μ g m⁻³. This is certainly the case within 10 m of relatively small construction sites and may extend out to 50–100 m from major construction sites. It would seem that beyond about 100 m, construction sites are likely to make only a small contribution to local PM₁₀ concentrations. The evidence is that vehicle movements are a key source of the PM₁₀, including byway or resuspended material tracked out of the site onto public roads. This track out of dust and dirt could extend the area of significant impact further than 100 m from the site boundary. The situation occurring at Manor Road, Bexley in London, where dust and dirt are being tracked out from light industrial sites onto the local roads by lorries, is similar (see Section 6.4.7). The study of monitoring sites across the southeast of England suggests that there might be fairly widespread contributions to PM₁₀ made by fugitive, in many cases construction-related, sources and that these incidents can contribute to the number of days with concentrations $>50 \ \mu g \ m^{-3}$. No specific studies have been identified that show the contributions construction activities can make to PM₂₅ concentrations, although the results for the Marylebone Road site suggest that PM_{2.5} concentrations are affected by construction activities, although to a lesser extent than PM_{coarse} concentrations.

6.4 Use of correlations, concentrations or chemistry to elucidate particle sources

6.4.1 Comparison of PM concentrations with measurements of other pollutants

- **624.** Measured PM_{10} and $PM_{2.5}$ concentrations have been compared with the measured concentrations of other air pollutants at Marylebone Road and Harwell. Figures 6.77 to 6.79 show comparisons of hourly PM concentrations during 2002 with NO_x , benzene and CO. PM concentrations are more closely correlated with NO_x than with benzene or CO. This is consistent with the expected emission factors for road vehicles, which are higher for PM and NO_x for diesel vehicles and higher for benzene and CO for petrol vehicles. It confirms that traffic emissions are an important PM source at this site. The emissions of PM_{10} are greater than the emissions of $PM_{2.5}$ (the gradient of the line is greater), although this analysis does not directly inform as to whether the additional emissions of PM_{coarse} are exhaust or other emissions.
- **625.** The gradients for PM₁₀ and PM_{2.5} are much more similar at the rural site at Harwell (Figures 6.80 and 6.81) and the correlation coefficients are lower because this site is less strongly influenced by traffic sources. Figures 6.84 and 6.85 show the relationships between PM concentrations and ozone. The daily maximum running 8-h ozone concentration has been compared with daily mean PM concentrations (hourly PM and ozone concentrations tend to be anticorrelated because primary PM emissions are correlated to NO emissions and thus anti-correlated with measured ozone concentrations). Figure 6.82 shows that there is some tendency for high PM concentrations to be associated with high ozone concentrations in the summer at Harwell, which reflects the photochemical generation of secondary particles. This is not the case in winter when the weak anti-correlation suggests that the higher PM days tend to be associated with lower ozone concentrations, when poor dispersion leads to a build up of primary

Figure 6.77 Marylebone Road hourly NO_x against PM_{10} and $PM_{2.5}$ (TEOM) in 2002.



Figure 6.78 Marylebone Road hourly benzene against PM_{10} and $PM_{2.5}$ (TEOM) in 2002.



particles and more efficient titration of ozone with NO. Figure 6.83 shows that the titration of ozone with locally emitted NO dominates measured ozone concentrations at Marylebone Road leading to PM and ozone concentrations being anti-correlated in both winter and summer.

626. Figures 6.84 and 6.85 show the diurnal variation of PM and other pollutant concentrations at these two sites in 2002. These plots confirm the conclusions of the scatter plot analysis. There is also evidence that NO_x and PM emissions at Marylebone Road are greatest during the morning peak and that CO and benzene emissions are greatest during the evening peak. This is consistent with a greater proportion of goods vehicle activity during the morning, as is shown in Figure 6.86.



Figure 6.79 Marylebone Road hourly CO against PM₁₀ and PM₂₅ (TEOM) in 2002.

- **627.** The diurnal variation of the measured CPC particle counts is also included in Figure 6.84 (for the period January 2001 to March 2002). The diurnal variation in the particle count is more similar to that for PM and NO_x than to the diurnal variation for CO and benzene. Figures 6.87 and 6.88 show the correlation between and PM and CPC particle count and CPC particle count and NO_x concentrations at Marylebone Road. The particle count appears to be more closely correlated with NO_x than with PM.
- **628.** In the final analysis in this section, Figure 6.89 and 6.90 show the correlation between the roadside increments of hourly PM concentrations at Marylebone Road in 2002 with the roadside increments of NO_x and CO concentrations (the roadside increment is the measured roadside concentration with the measured value from a nearby background site subtracted. In this case we have chosen London Bloomsbury, where both PM_{10} and $PM_{2.5}$ concentrations are measured.) The correlation coefficients for this roadside increment analysis are not necessarily better than for the total roadside concentrations due to the uncertainties associated with the subtraction of background concentrations, but the intercepts are smaller and the gradients of the lines should be more directly comparable with emission factors.



Figure 6.80 Harwell hourly NO_x against PM_{10} and $PM_{2.5}$ (TEOM) in 2002.





Figure 6.82 Relationship of PM_{10} and $PM_{2.5}$ with daily maximum running 8-h mean ozone; Harwell 2002 and 2003.



Figure 6.83 Relationship of PM_{10} and $PM_{2.5}$ with daily maximum running 8-h mean ozone; Marylebone Road 2002 and 2003.







Figure 6.85 Harwell diurnal concentrations normalised by average.



Figure 6.86 London Marylebone Road diurnal traffic flow data, normalised by average, local time.



Figure 6.87 PM_{10} and $PM_{2.5}$ versus CPC at Marylebone Road, January 2001 to March 2002.



Figure 6.88 CPC versus NO_x (paired hourly data) at Marylebone Road.



Figure 6.89 Marylebone Road PM_{10} and $PM_{2.5}$ increment against NO_x .



Figure 6.90 Marylebone Road roadside PM₁₀ and PM_{2.5} increment against CO.



629. Table 6.16 shows the gradients determined in this comparison of hourly roadside increments and the ratios of the NAEI estimates of road traffic exhaust emission for Marylebone Road in 2001 and for total UK urban traffic emissions in 2001. The measured gradients are lower for $PM_{2.5}$ than for PM_{10} . The implied ratio of PM_{10} and NO_x traffic emission at Marylebone Road is somewhat lower than the NAEI estimates for this road but similar to the ratio for total UK urban emissions. This is a little unexpected, as it suggests that exhaust emissions are sufficient to account for the measured roadside increment of PM_{10} , without any contribution from non-exhaust traffic emissions. The observed ratio of PM_{10} :CO concentrations is lower than suggested by the comparisons of traffic emissions.

Table 6.16 Empirically determined gradients and ratios of emissions(dimensionless).

	PM _{2.5} :NO _x	PM _{2.5} :CO	PM ₁₀ :NO _x	PM ₁₀ :CO
Marylebone Road measurements 2002	0.036	0.0040	0.048	0.0065
Marylebone Road emission 2001	_		0.055	0.0116
UK urban emissions 2001	_		0.046	0.0079

6.4.2 Weekday/weekend differences at traffic-influenced sites

630. Table 6.17 shows the results of a comparison of PM concentrations at Marylebone Road on weekdays (Monday to Friday) and Sundays. Measured PM concentrations were higher at Marylebone Road on weekdays than on Sundays. The weekday-Sunday difference was greater for PM₁₀ than for PM_{2.5} and was similar for both Partisol and TEOM measurements. The percentage difference was also very consistently ~20% for the different monitoring methods and PM_{2.5}, PM₁₀ and PM_{coarse} fractions. The weekday-weekend difference was smaller at the nearby

background sites (North Kensington and London Bloomsbury) but was still evident (North Kensington was chosen as the most appropriate background site for this analysis, but TEOM PM_{2.5} measurements are not available at this site.) The weekday-Sunday difference for the roadside increment was therefore somewhat smaller than for the total roadside concentration but generally larger in percentage terms. The weekday-Sunday difference for particle count for the period January 2001 to March 2002 was also calculated for comparison and was ~25%. Interestingly, the total traffic count was similar on weekdays and Sundays at Marylebone Road but there was more light traffic on Sundays than on weekdays and the count for heavy traffic was much lower on Sundays. This suggests that heavy traffic had the strongest influence on the difference in measured concentrations between weekdays and Sundays.

Table 6.17 Average PM concentrations for weekdays and Sundays during January 2002 to October 2003 at Marylebone Road (μg m³).

We	eekdays	Sundays	Weekdays-Sundays difference
PM _{2.5} (Partisol)	29	23	5 (19%)
PM ₁₀ (Partisol)	47	39	8 (18%)
PM _{2.5} (TEOM)	22	17	4 (20%)
PM ₁₀ (TEOM)	38	30	8 (20%)
PM _{coarse} (Partisol)	17	13	4 (22%)
PM _{coarse} (TEOM)	16	13	3 (21%)
North Kensington PM _{2.5} (Partisol)	20	18	2 (10%)
North Kensington PM ₁₀ (Partisol)	28	24	3 (12%)
London Bloomsbury PM _{2.5} (TEOM)	14	12	2 (14%)
North Kensington PM ₁₀ (TEOM)	21	19	3 (14%)
North Kensington PM _{coarse} (Partisol)	8	7	2 (19%)
North Kensington PM _{coarse} (TEOM)	7	6	1 (16%)
Roadside increment PM _{2.5} (Partisol) Roadside increment PM ₁₀ (Partisol) Roadside increment PM _{coarse} (Partisol) Roadside increment PM _{2.5} (TEOM) Roadside increment PM ₁₀ (TEOM) Roadside increment PM _{coarse} (TEOM) CPC (cm ³) (January 2001 to March 2002) 1,3	9 18 7 16 9 25,265	6 14 7 5 11 7 961,576	3 (32%) 4 (25%) 2 (18%) 2 (31%) 5 (29%) 2 (26%) 36,3689 (27%)
Light traffic (count per day)	67556	69699	–2143 (–3%)
Heavy traffic (count per day)	9337	4093	5244 (56%)
Total traffic (count per day)	76896	73478	3418 (4%)

631. The weekday-Sunday difference was a similar magnitude to Marylebone Road at Bury roadside and M25 Staines (Table 6.18). The total traffic counts was much lower at Bury roadside on Sundays than on weekdays; the difference was rather smaller at M25 Staines. The variation in rural sulphate concentrations by day of the week was also investigated but no consistent patterns were found.

	Weekdays	Sundays	Weekdays-Sundays difference
Bury roadside PM ₁₀ (TEOM) Bury roadside total traffic	26	20	6 (23%)
(counts per day)	146,869	97,456	49,413 (34%)
M25 Staines PM _{2.5} (TEOM)	14	12	3 (18%)
M25 Staines PM ₁₀ (TEOM) M25 Staines PM _{coare} (TEOM)	24 9	7	5 (23%) 3 (30%)
M25 Staines total traffic (counts per day)	197,849	167,410	30,438 (15%)

Table 6.18 Average PM concentrations for weekdays and weekends January 2002 to December 2003 at Bury roadside and M25 Staines ($\mu g m^3$).

6.4.3 Influence of air mass back trajectory and weather type on PM concentrations

Daily concentrations of PM₁₀ in Edinburgh for the period 1992–2000 vary 632. significantly with the geographical sector of the back trajectory calculated for the arriving air mass (Figure 6.91). Trajectories were categorized objectively by statistical hierarchical clustering using mean distance squared and average linkage clustering on the vectors of the 30 variables comprising: distance north of Edinburgh; distance south of Edinburgh; and pressure level at 12, 24, 36...120 h prior to arrival at Edinburgh. There was no a priori expectation for the 'best' number of clusters into which to classify the trajectories, which was determined by examining plots of RMS and r^2 as a function of number of trajectories for major discontinuities in slope. Overall, five major clusters emerged, together accounting for >94 % of the total number of trajectories. Median PM_{10} on days when air mass originated from continental Europe or from local circulation about the British Isles (23 and 18 μ g m³ (TEOM), respectively) were significantly elevated compared with days when air mass originated from Atlantic SW, Atlantic W and the Arctic (15, 15 and 16 μ g m³ (TEOM), respectively). This means that air-mass source can be associated with increases of up to 50%, on average, of receptor PM_{10} in the UK.

Figure 6.91 Apportionment of 24 h PM₁₀ concentrations in Edinburgh (1992–2000) by geographical sector of air mass back trajectory. (Cluster 1, Local British Isles; Cluster 2, Atlantic SW, Cluster 3, European; Cluster 4, Arctic; Cluster 5, Atlantic W. The box edges define lower and upper quartiles; lines extend to 5th and 95th percentiles.)



Figure 6.92 Apportionment of 24 h PM_{10} and $PM_{2.5}$ concentrations in Edinburgh (1999–2000) and $PM_{2.5}$: PM_{10} ratio by geographical sector of air mass back trajectory.



- **633.** Data for colocated PM_{10} and $PM_{2.5}$ for the shorter period 1999–2000 show a similar, but accentuated, trend for $PM_{2.5}$ (Figure 6.92). For this more recent period, median PM_{10} on days when air mass originated from continental Europe or from local circulation about the British Isles (20 and 16 µg m³ (gravimetric), respectively) was still significantly elevated compared with days when air mass originated from Atlantic SW, Atlantic W and the Arctic (12, 13 and 14 µg m³ (gravimetric)). Likewise, $PM_{2.5}$ was significantly elevated (by 3–4 µg m³ (gravimetric) or >50%, on average) on days with air masses from continental Europe and the British Isles. Importantly, the ratio of $PM_{2.5}$ to PM_{10} was also significantly elevated on these days, as shown by the numbers at the bottom of Figure 6.92. This demonstrates the influence in the UK of sources of anthropogenically derived fine particles, independent of local scale air quality management areas.
- **634.** Similar findings were noted by Buchanan *et al.* (2002), who divided data according to the classification of daily regional scale synoptic weather maps into Jenkinson weather types to show that PM₁₀ in Edinburgh was significantly higher for anticyclonic, southerly and southeasterly Jenkinson weather types, indicative of air flow from continental Europe.

6.4.4 Analysis of trends in PM concentrations by wind direction

635. Figures 6.93 and 6.94 show an analysis of the trend in mean PM₁₀ concentration at London Bexley by wind direction. An examination of the mean concentrations in each 10° bin has indicated that the concentration when the wind is from directions between 4° and 120° (east) were generally higher than the mean of all wind directions (due to the elevated secondary PM concentration in easterly winds). Previous analysis of particulate sulphate measurements at rural sites in the UK has shown that concentrations are highest for easterly flow (RGAR, 1999). Concentrations when the wind was from the southwest (190° to 270°) were generally lower than the mean. The mean of concentrations when the wind was from all other wind directions.

Particulate Matter in the United Kingdom

- **636.** The unusually high secondary PM concentration in 1996 and 2003 are clearly shown in Figure 6.93. Concentrations in the southwesterly direction showed a decline from 1994 to 1998 and were then relatively constant. Concentrations from the east decline to 2000 and then increased. Figure 6.94 shows the contributions from the wind directions to the annual mean concentrations. The contribution from the east was highest in 1996, 1997 and 2003. The contribution from the southwest was reasonably constant throughout. Figure 6.95 shows the frequency of wind directions within these categories. The frequency of 'other' directions was relatively consistent throughout the period. The frequency of wind from the southwest was correspondingly low in these years. 1996 and 2003 were the years with the highest concentration in easterly winds and the greatest frequency of winds from this direction.
- **637.** Figures 6.96 to 6.98 show a similar analysis for Belfast Centre. Mean concentrations for the easterly direction were relatively constant between 1999 and 2003 but the contribution to the annual mean from this wind direction was much greater in 2003, due to the greater prevalence of easterly winds. The frequency of winds from the southwest decreased over the period leading to a decline in the contribution from the southwesterly direction seen in Figure 6.97. Overall the frequency of winds from the east was lower than in London.
- **638.** Figures 6.99 to 6.101 show the results of the analysis for the Birmingham Hodge Hill site. The PM_{10} concentration in winds from the east shows a decline over the period but a rise between 2002 and 2003. Concentrations when the wind was from the east were particularly low in 1999. The PM_{10} concentration for southwest winds also shows a decline. The relative frequencies of the wind directions at this site were reasonably consistent with those for Bexley with less winds from the east in 1998, 1999 and 2000 and a higher frequency of winds from the southwest in these years. The contribution from winds from the east to annual mean PM_{10} concentration was highest in 1995, 1996 and 1997, with a rise from 2002 to 2003.
- **639.** $PM_{2.5}$ concentrations have also been measured at Birmingham Hodge Hill and this enables the analysis of the concentrations measured for different wind directions to be carried out for both $PM_{2.5}$ and PM_{coarse} , in addition to PM_{10} . Figures 6.102 and 6.103 show the results for $PM_{2.5}$. Once again concentrations were greater for winds from the east. The trends with time were similar to those for PM_{10} but the overall decline in concentrations was less steep. Figures 6.104 and 6.105 show the results for PM_{coarse} . The PM_{coarse} concentrations showed a decline for all wind directions. The PM_{coarse} concentrations showed less variation in concentration with wind directions than $PM_{2.5}$. The decline in annual mean $PM_{2.5}$ concentrations at Birmingham Hodge Hill has, however, not been consistently observed across the other UK monitoring sites, as is illustrated in Figure 6.106.





Figure 6.94 Bexley time series PM₁₀ contribution by year and direction.







Figure 6.96 Mean PM_{10} concentration by wind direction at Belfast centre (µg m³).















Figure 6.100 Birmingham Hodge Hill time series PM_{10} contribution by year and direction.







Figure 6.102 Mean $\text{PM}_{2.5}$ concentration by wind direction, Birmingham Hodge Hill (µg m³).



Figure 6.103 Birmingham Hodge Hill time series PM_{10} contribution by year and direction.







Figure 6.105 Birmingham Hodge Hill time series PM_{coarse} contribution by year and direction.



6.4.5 Coarse particles at Marylebone Road: sources and concentrations

6.4.5.1 Concentrations measured at Marylebone Road

640. As shown in Figure 6.107, hourly concentrations of PM_{coarse} measured at Marylebone Road from July 1998 to August 2001 were highly variable. They ranged from 1 to 595 μg m⁻³ and followed lognormal distributions. High particle mass concentrations in comparison to urban background sites are measured in Marylebone Road. More than 25% of hourly PM_{coarse} concentrations were above 15 μg m³. Some very high concentrations (above 100 μg m⁻³) were occasionally measured. They were generally associated with demolition or construction activities. No seasonal variation was observed at Marylebone Road. This suggests that there is no significant temporal variation of influential meteorological parameters and sources of particulate matter at Marylebone Road on a seasonal timescale, or that the effects of different factors cancel one another.

Figure 6.106 TEOM measurements for (a) PM_{10} , (b) $PM_{2.5}$ and (c) PM_{coarse} during 1991–2003.





Figure 6.107 (a) Basic statistics for $PM_{2.5}$ and PM_{coarse} concentrations measured at Marylebone Road and (b) relative frequencies of PM_{coarse} concentrations (logarithmic scale) when each size bin is 1 µg m³ in width. (Hourly data from July 1998 to August 2001 (n = 22,268).)

	ΡΜ _{2.5} (μg m³)	PM _{coarse} (μg m³)	ΡΜ ₁₀ (μg m³)	
Minimum	1	1	3	
Maximum	330	595	800	
Median	21	10	32	
Arithmetic mean	23.3	12.7	36.0	

(a)



641. Figure 6.108 presents the median weekly cycles of $PM_{2.5}$ and PM_{coarse} . The median weekly cycles of light duty traffic (cars and motorcycles) and of heavy duty traffic (lorries, buses, coaches) are also represented in this figure. Very similar patterns of traffic are observed from one week to another, and the median weekly cycles of traffic densities well represent the single weekly cycles. The median weekly cycle of $PM_{2.5}$ and PM_{coarse} concentrations at Marylebone Road shows similar strong daily and weekly variations that correlate with those of heavy duty traffic. Higher concentrations are measured during the daytime on work days (median >15 µg m³ for PM_{coarse}) and much lower concentrations during night-time and weekends. During the night, the median concentration of PM_{coarse} is <8 µg m³.

Figure 6.108 Median weekly cycles of $PM_{2.5}$ and PM_{coarse} concentrations and median weekly cycles of light duty and heavy duty vehicle counts.



642. The median $PM_{2.5}$: PM_{10} ratio at Marylebone Road was ~0.66 for the whole period of study, that is, on average one-third of the mass of the PM_{10} is within the PM_{coarse} fraction. Similar ratios are reported elsewhere (Ruellan and Cachier, 2001; Manoli *et al.*, 2002). This median mass ratio $PM_{2.5}$: PM_{10} at Marylebone Road is much lower than reported mass fractions from exhaust emissions (in the APEG report it was 0.8 for non-catalyst petrol exhaust, 0.9 for catalyst petrol exhaust and 0.9 for diesel exhaust (APEG, 1999). This suggests that there is an excess of coarse particles in comparison to exhaust emissions.

6.4.5.2 Local sources of PM_{coarse} at Marylebone Road

6.4.5.2.1 Traffic

- **643.** The weekly cycle of PM_{coarse} suggests that PM_{coarse} , as well as $PM_{2.5}$, is related to the traffic. The highest concentrations are measured during the midday period of the work days when the heavy duty traffic is at a maximum level and reaches 20% of the total traffic flow. The lowest concentrations are measured on Sundays and a minor peak in concentration is seen on Saturday mornings. Both $PM_{2.5}$ and PM_{coarse} cycles are clearly closer to the heavy duty traffic cycles than to the light duty traffic cycle (see Figure 6.108). The stronger wind and air turbulence during the day would lead to the same PM_{coarse} concentration patterns on work days as on weekends if the coarse particulate matter were only mechanically induced by the wind; this is clearly not the case. A high Spearman rank correlation coefficient is also found between both the $PM_{2.5}$ and the heavy duty traffic (r = 0.80). A high Spearman rank correlation coefficient is also found between both the $PM_{2.5}$ and the heavy duty traffic (r = 0.77).
- **644.** The relationship between traffic data and particle data was also examined using a hierarchical cluster analysis (Wards' method and square Euclidean distance as a similarities measure). This multivariate method separates observations into groups according to their similarities and differences. The median weekly cycle of concentrations is used to smooth the variability of concentrations due to the variability of the meteorological parameters. Only the concentrations measured when the wind direction favoured the measurement of on-road emissions were included in the analysis, that is to say, the following wind directions were considered: (i) wind parallel to the street (accumulation of on-road emissions) and (ii) from southerly directions (vortex favouring the measurement of on-road emissions). The two tracers, NO_x and CO were simultaneously examined. The results obtained with this method are presented in Figure 6.122.

Figure 6.109 Dendogram representing the result of a hierarchical cluster analysis (Wards' method, square Euclidean distances as a similarities measure).



645. According to Figure 6.109, two clusters are clearly identified:

- $PM_{2.5}$, PM_{coarse} and NO_x concentrations are clustered with the heavy duty traffic.
- CO concentrations are clustered with the light duty traffic.

646. These results confirm that PM_{coarse} concentrations as well as PM_{2.5} concentrations are associated with the heavy duty traffic (lorries, coaches and buses). The clustering of NO, with the heavy duty traffic and of CO with the light duty traffic (mainly petrol vehicles) gives confidence in the results. The heavy duty traffic is mainly comprised of diesel vehicles that have high emission factors for particle mass; this result was expected for the fine fraction of particulate matter. The clustering of the coarse PM with the heavy duty traffic suggests that the substantial turbulence induced by the heavy duty traffic is responsible for a greater magnitude of resuspension of particles from the road than the light duty traffic. Another explanation is the larger amounts of dust emitted by the heavy duty traffic due to stronger abrasion processes such as tyre wear and brake linings. Sternbeck et al. (2002) found a strong correlation between particulate barium (which is a common element in brake linings and is not used as a fuel additive in Sweden) and heavy duty traffic, suggesting that heavy duty vehicles are responsible for larger brake wear particulate emissions than light duty vehicles. Large vehicles have higher brake wear emission rates for airborne particles than small vehicles and these emissions would include both mechanically generated coarse particles and the formation of finer particles due to high temperatures at the brake/rotor interface (Garg et al., 2000). These results are in agreement with other studies that have shown a link between coarse PM and traffic flow (Janssen et al., 1997; Ruellan and Cachier, 2001; Gehrig and Buchmann, 2003).

6.4.5.2.2 Construction and demolition activities

647. Analyses of the Marylebone Road data in regard of construction and demolition activities appear in Section 6.3.6.

6.4.6 Estimation of the non-exhaust road transport particle emissions at Marylebone Road

648. Data from the Marylebone Road kerbside site have been analysed to determine the diurnal variations in NO_v and different PM components. Consideration of hourly means of the coarse fraction shows that it is highly variable on this timescale and has a poor relationship with other pollutants such as NO_x or vehicle flows. Because of its highly variable nature, longer term averages are required to reveal how it varies against other parameters. Consideration of Figure 6.110 highlights several important features. First, there is a time lag of 1 h between winter and summer diurnal plots of all pollutants, including the coarse fraction. This time lag is indicative of road traffic activity sources where the lag is observed when Greenwich Mean Time (GMT) changes to British Standard Time (BST). These results strongly indicate that the majority of the coarse fraction observed at Marylebone Road is related to road traffic. Another feature of Figure 6.110(d) is that there is very little difference between PM concentrations in summer and winter, which, unlike NO_x, is clearly lower in summer due to increased dispersion. The similarity between the magnitude of the summer and winter coarse fraction suggests that either the source strength is higher in summer or the dispersion processes are different compared with NO_x.

Figure 6.110 Summer and winter diurnal variations in (a) NO_x , (b) PM_{10} , (c) $PM_{2.5}$ and (d) PM_{10} -PM_{2.5} at Marylebone Road (May 1998–December 2002).



- **649.** The diurnal variation in the coarse fraction shown by Figure 6.110(d) is different from the variation in NO_x and $PM_{2.5}$, indicating different origins for these pollutants.
- **650.** A consideration of how the coarse fraction varies over these periods strongly suggested a link with the flow of HGVs since the coarse fraction is lowest during weekend periods (particularly Sundays) and the diurnal variation through each day was similar to that for HGVs. A multiple regression was carried out to determine whether the observed variation by hour of the day and day of the week of the coarse fraction could be related to other parameters such as other pollutants and vehicle flows. The multiple regression took the form:

$$PM_{coarse} = a * LGV + b * HGV + c.$$

Where LGV and HGV are the hourly mean flows of LGVs and HGVs by day of the week; a, b and c are constants to be derived from the regression. The results from the multiple regression are shown in Table 6.19. These results suggest an HGV emission factor approximately 15-times higher than LGVs. The statistics for the regression also suggest that all calculated constants are significant at the 95% CI, but the relationship with HGVs is much stronger than LGVs. The multiple regression also yielded a high r^2 value of 0.88. The intercept of 3.9 µg m⁻³ is for the coarse fraction that cannot be related to vehicular activity and is presumably related to wind-blown PM and other sources of coarse particles in the urban environment.

	Coefficients	Standard error	t statistic
Intercept c	3.932492	0.299579	13.1
a	0.00085	0.000112	7.6
b	0.012306	0.000534	23.0

Table 6.19 Results from the multiple regression.

651. Figure 6.111 shows the comparison between the measured and predicted coarse fraction. Overall the agreement is very good, but predicted values tend to be too high on Sundays. One likely explanation for the poor comparison on Sundays is the use of broad categories of vehicle types, which mask important changes for individual vehicle types. For example, an analysis of manual count traffic data in London suggests that rigid and articulated vehicles are ~30% of their weekday flows on Sundays, whereas the continuous traffic count data suggests a value of 43%. It is likely the traffic counter at Marylebone Road categorizes other non-rigid or articulated HGVs as heavy vehicles.

Figure 6.111 Comparison between measured and predicted coarse particles at Marylebone Road (May 1998 to December 2002).



652. The results above can be used to estimate the emissions factors in g km⁻¹ for coarse particles from light and heavy vehicles. Emission rates of NO_x have already been calculated for light and heavy vehicles using Marylebone Road (see the AQEG NO₂ report). A multiple regression of light and heavy vehicles with NO_x suggests that over the period covered by the dataset, heavy vehicles were responsible for ~150 µg m⁻³, for a mean emission rate of 9.52 g km⁻¹. Using these data with the results from the coarse fraction multiple regression it is possible to estimate the emission rates for light and heavy vehicles of 0.415 g km⁻¹ and 0.028 g km⁻¹ for light vehicles. The mean rate for the mix of vehicles using Marylebone Road is 0.067 g km⁻¹. The coarse particle emission rates for light and heavy vehicles are very similar in magnitude to the estimated exhaust emissions, suggesting that at this location particle resuspension and exhaust emissions are similar in magnitude.

6.4.7 Particle resuspension at the Manor Road site, London

653. Manor Road lies in Erith in the north of the London Borough of Bexley. Manor Road provides access to an industrial area with vehicles having to pass through a residential section of the road. The roadside TEOM situated on Manor Road consistently exceeds the 2005 EU limit value. The TEOM has measured between 78 and 131 daily means above 50 μ g m³ (TEOM * 1.3) per year since installation in 1999. Consequently, Manor Road has been declared an AQMA. Hourly mean PM₁₀ at the site shows a distinct diurnal and day of week elevation above background, with Manor Road PM₁₀ being elevated during working hours on weekdays and on Saturday mornings. This is shown in Figure 6.112.

Figure 6.112 Diurnal and day of week mean PM₁₀ at Manor Road, compared to background, during July to October 2001.



- **654.** Analysis of the measurements of elevated PM_{10} and NO_x above background suggests that the elevated PM_{10} is due to traffic using Manor Road. The effective PM_{10} :NO_x ratio at the site is over ten-times higher than the average factor at other sites in London. Manor Road is often visibly silted from material largely carried onto the roadway from a waste transfer site and vehicles using Manor Road often carry open and dusty loads (Fuller *et al.*, 2001; Bexley Council, 2003). It is likely that resuspended PM_{10} and perhaps dust lifted directly from vehicles accounts for the disproportionately high PM_{10} at this location. Similar elevated PM_{10} levels have been measured at the Hastings 1 site in East Sussex where over 50 daily means >50 µg m³ per year were recorded during 2002 and 2003. Again visible road silting is also present near the site, arising from access to a nearby landfill. Although further investigation is required, it is likely that similarly elevated local PM_{10} may arise at other locations with substantial road silting. Such sites may include other landfills and waste transfer sites and also large construction sites and exits from unmade roads.
- **655.** Examination of the elemental composition of individual particles sampled from roadside air at Manor Road and sampled from the surface of Manor Road (Jansz *et al.*, 2004) confirms that the airborne particles are similar to the material on the road during times when the roadside PM_{10} concentration is elevated. In wind directions where roadside PM_{10} concentrations at Manor Road are similar to those at nearby background sites, the airborne particle composition is different to that on the road surface.

- **656.** Deposit gauge measurements at varying distances from the road and the waste transfer station confirm the raised levels of airborne particulate matter are associated with the road and not airborne dispersion of fugitive emissions directly from the waste transfer site.
- **657.** Detailed examination of the time series shown in Figure 6.112 potentially provides some indication that the non-exhaust emissions are raised from the road surface by all vehicles, not just from the soiled wheels or spilling of load of the vehicles exiting the waste transfer station. The daily opening and closing hours of the waste transfer station and the different diurnal and weekly variability of total traffic flow on Manor Road allows the respective contributions of the two parts of the vehicle fleet to be separated using modelling, including analysis of the temporal variability of the sediment loading on the road surface. This modelling is currently in progress (Jansz *et al.*, 2004).

6.4.8 Contribution to ambient particle concentrations from power stations

- The power generators as part of their Joint Environmental Programme (JEP) 658. undertake routine, continuous monitoring at a number of rural sites in the vicinity of coal-fired and oil-fired power stations. These sites provide a valuable supplement to the limited number of monitoring sites in the national network. PM₁₀ and PM₂₅ monitoring results using TEOMs have been summarised in a report (Webb et al., 2001). This report has been updated by a more recent report on monitoring in 2001 (Webb et al., 2002). There is one site at Cliffe in the vicinity of Yorkshire power stations, two monitoring sites at Bottesford and Thorney in the Trent valley and two sites at Hall Farm and Wingham in the Thames Valley. Given that major industrial sources, whose emissions are regulated by the Environment Agency, are a small fraction of total national primary PM_{10} emissions (~10%) and that releases from tall chimneys are likely to be diluted near the ground, industrial sources are generally not thought to make a large contribution to primary particulate matter concentrations at ground level. Using co-located SO₂ measurements as an indicator of the presence of a power station plume, from measurements from the network it was concluded that the primary particulate emissions contributed much less than 10% of the daily PM₁₀ threshold of 50 μ g m⁻³ and ~0.25 μ g m⁻³ to the annual mean PM₁₀ concentration.
- Power stations and other industrial processes can also make a contribution 659. to PM through the conversion of gaseous releases of SO_2 and NO_2 to secondary particulate. Studies of the formation of secondary particles in plumes containing high concentrations of acid gases have been reviewed (Griffiths and Hill, 2000; Wright and Cocks, 2001). The particles would initially be in the ultrafine range and combine together with other particles to form fine agglomeration mode particles. To supplement the reviews and theoretical understanding, field studies have been attempted. One set of measurements of ultrafine particulates in power station and urban plumes used a differential mobility particle sizer (DMPS) during three episodes (Griffiths et al., 2002). Power station source attribution is difficult; using SO₂ and NO_x as plume markers and considering correlation coefficients, estimates of the contribution of a power station plume to particle numbers at ground level were made. In another study two particle size analysers (APS and ELPI) were installed near Didcot Power Station and operated for one month (Brooke and Wright, 2002). The particle size distributions from the two

instruments were rather different. In conclusion, there is no clear evidence for rapid particle formation in the vicinity (out to some 100 km) of coal- and oil-fired power stations under typical British atmospheric conditions.

6.4.9 Industrial sources: Port Talbot case study

660. There are few industrial sources that give rise to significant PM_{10} contributions. One that has been recognised from the early days of PM_{10} monitoring is the Corus steel works at Port Talbot. A monitoring site was established in 1997 in the town, which lies just to the east-northeast of the works. Frequent exceedences of 50 µg m⁻³ as a 24-h concentration (TEOM * 1.3) were measured, which are comparable to the London Bloomsbury urban centre site (Table 6.20). The results for this site illustrate the difficulties associated with the default TEOM scaling factor. A KFG reference gravimetric sampler was subsequently operated alongside the TEOM analyser at this site for three years, showing a relationship which was close to unity. The TEOM results for Port Talbot are, therefore, shown in Table 6.20 as unscaled as well as scaled to the default 1.3 factor. This factor of 1.0 at Port Talbot is consistent with a dominant contribution of non-volatile particles being emitted from the steel works. With this factor, there would be no exceedences of the objective.

Table 6.20 PM₁₀ concentrations in Port Talbot and the urban centre site at London Bloomsbury during 1997 and 2003.

	1997	1998	1999	2000	2001	2002	2003
Exceedences of 50 µg m³ Port Talbot days							
>50 µg m³ (TEOM) Port Talbot days	22	28	26	27	18	5	16
>50 μ g m ³ (TEOM * 1.3)	59	59	62	56	39	24	43
$>50 \ \mu g \ m^3$ (TEOM * 1.3)	43	20	20	11	17	(21) ^a	(14) ^a
Annual mean							
Port Talbot annual mean (TEO) Port Talbot	M) 27	27	26	26	23	21	24
annual mean (TEOM 1.3) London Bloomsbury	35	35	34	33	30	28	32
annual mean (TEOM 1.3)	35	30	28	28	29	n/aª	n/aª

^aThere was low data capture for 2002 and 2003 at this site.

661. On the basis of the monitoring carried out near the steel works, and using the default 1.3 scaling factor for the TEOM, Neath Port Talbot County Borough Council declared an AQMA covering the residential areas to the north and east of the steel works. In order to inform its Review and Assessment work and subsequent Action Plan, the Council commissioned various studies to identify the sources of the high PM₁₀ concentrations (Neath Port Talbot CBC, 2000). The monitoring site lies 75 m from the M4 motorway and 700 m from the steel works, both these sources being in opposite directions (Figure 6.113). It was, therefore, necessary to establish the relative roles of the motorway and the steel works and within the steel works to identify the relative roles

of the different processes and/or fugitive PM from the various stockpiles on site. The first investigations involved analysis against wind direction (Figure 6.114). This showed that elevated PM_{10} concentrations, above 50 µg m³ (TEOM * 1.3) were associated with winds from the southeast through to west-northwest and, in particular, with southwest and west-southwest winds. The higher CO concentrations were associated in particular with the peak PM_{10} directions of southwest and west-southwest winds. This was consistent with the steel works being the most important contributor to elevated PM_{10} , with the sources widely distributed across the site, although with one or more major sources to the southwest and west-southwest of the monitoring site, which were likely to be combustion related given the associated carbon monoxide elevation. The motorway appeared to be influencing the nitrogen dioxide concentrations, perhaps increasing them by some 10–20 µg m⁻³ (assuming a background for non-motorway directions of around 20 µg m⁻³). This pattern has been seen in each year of monitoring.

Figure 6.113 Layout of Port Talbot monitoring site in relation to the steel works and the M4 motorway.



- **662.** Samples were also collected using an ACCU sampler attached to the TEOM. This was set up to collect samples on eight different filters according to the wind direction. Samples were then analysed for chemical composition and examined for particle morphology using an analytical transmission electron microscope. The particle examination identified an abundance of spherical particles associated with the winds from the south and southwest. These particles were compared to samples of the emissions from the coke works, the BOS plant, the sinter plant and the blast furnaces. The semi-quantitative chemical analysis showed that iron oxide dominated in the ambient samples. The composition profile and the particle morphology of the ambient samples provided the best match with the blast furnace particles.
- **663.** Confirmation of the role of the steel works processes came from the unfortunate incident that took place on 8 November 2001, when an explosion occurred in furnace 5 (Kent and Willis, 2004). Hourly average concentrations, which had shown frequent peaks of 50 150 μg m⁻³, were essentially eliminated after the accident (Figure 6.115). Table 6.20 shows that annual average concentrations and

Figure 6.114 PM_{10} (TEOM * 1.3), carbon monoxide and nitrogen dioxide concentrations at Port Talbot AURN site 1989.



24-h exceedences of 50 μ g m⁻³ were also much lower in 2002 than 2001. The steel works is though still contributing to PM concentrations measured locally, as discussed below.

- A directional analysis of 2003 size fractionated monitoring data for the AURN 664. site at Port Talbot provides further insight into industrial concentrations. Daily concentrations of PM_{10} and PM_{25} have been measured using a Partisol (gravimetric) sampler, which has also allowed PM_{coarse} concentrations to be calculated. A KFG, reference PM₁₀ (gravimetric) sampler, has also been used, and the site is equipped with a TEOM sampler. The directional analysis of the 24-h samplers will inevitably smooth the pollution rose, as the 24-h concentration is applied to each hour of the day. The PM_{10} (TEOM) pollution rose shows a pattern similar to that previously shown (Figure 6.116), with emphasis on winds from the southwest; however it is notable that the PM₁₀ (gravimetric) pollution rose measured with the KFG sampler (Figure 6.117) shows a much more even distribution, further illustrating the problems with the TEOM. The problem arises because of loss of volatiles from the PM coming from the northeast, broadly the direction of the motorway on the basis of the nitrogen dioxide pollution rose (Figure 6.114), but also the direction for imported air from England and continental Europe. The TEOM is under-reading by ~40% for the winds from the northeast, whereas from the southwest there is a virtual 1:1 agreement. The latter is consistent with mineral particles from the steel works.
- **665.** The $PM_{2.5}$ and PM_{coarse} results from the Partisol sampler (Figure 6.118) show that the particles from the east are dominated by $PM_{2.5}$, whereas from the southwest the PM_{coarse} particles dominate. This would suggest that the steel works is predominantly contributing coarse PM. This is a significant observation if the air quality criterion is to be changed from PM_{10} to $PM_{2.5}$, as it shows that less attention would be paid to industrial PM emissions from a steelworks.
- **666**. The emerging technique of single particle aerosol mass spectrometry is yielding useful insights into the sources and behaviour of airborne particles. Two complementary mass spectrometry techniques have been deployed for the characterisation of aerosols in the UK atmosphere. One instrument, referred to as an Aerosol Time-of-Flight Mass Spectrometer (ATOFMS) provides real time size and mass spectral information from which chemical composition can be inferred on individual atmospheric particles. The other technique using an instrument referred to as an Aerosol Mass Spectrometer (AMS) provides real-time size resolved information on the concentrations of specific non-refractory components of airborne particles.

Figure 6.115 Hourly average PM_{10} (TEOM) concentrations from August 2001 to January 2002 at Port Talbot. (An explosion took place on 8 November 2001 at the steel works.)



Figure 6.116 PM₁₀ (TEOM) at Port Talbot in 2003, by wind direction.



667. Beddows *et al.* (2004) deployed an ATOFMS instrument at Eskdalemuir, a rural location in southern Scotland. A total of 51,980 particles in the size range 0.3–7.4 µm were detected. The individual particle mass spectra were analysed for the presence of individual elements and of ion fragments characteristic of specific molecular composition. A wide range of chemical species was identified with an abrupt change in composition coincident with a transition from a continental to a maritime airmass. Whilst the technique has great potential for application to source apportionment of particles in polluted atmospheres, there are major issues of data interpretation to be addressed before fully quantitative data are obtainable.

Figure 6.117 PM₁₀ (gravimentric, KFG) at Port Talbot in 2003, by wind direction.



Figure 6.118 $\rm PM_{2.5}$ and $\rm PM_{coarse}$ (Partisol) at Port Talbot in 2003, by wind direction.



The AMS instrument has been deployed by Allan et al. (2003) for the 668. measurement of fine particle chemical composition in Edinburgh and Manchester. The results from the AMS are more readily quantifiable than those from the ATOFMS, but the instrument is limited to measuring a smaller number of chemical components. The most powerful application of the instrument is to the high time resolution measurement of the size distributions of chemical components such as sulphate, nitrate and carbon. By careful interpretation of the mass spectral data, the instrument allows differentiation between primary and secondary organic carbon components of airborne particles. Allan et al. (2003) reported that the observed nitrate, sulphate and organic activity appeared to be confined to two modes during all campaigns. The lower diameter mode had a mass peak at an aerodynamic diameter of 100–200 nm and mainly consisted of organic chemicals with little oxidation which appeared to originate from motor vehicle emissions. The larger diameter mode appeared at various diameters between 300–800 nm and contain nitrate, sulphate and organic chemicals with a notable degree of oxidation, as would be expected from secondary aerosol.