Chapter 7

What are the main trends in particulate matter in the UK?

Key points

- The decline in coal use during the second half of the 20th century has led to declines in the rural concentrations of a wide range of trace elements in suspended PM.

- The phase-out of lead in petrol has exerted a significant influence on particulate lead concentrations throughout the UK from the 1980s onwards. Particulate lead concentrations have declined by more than an order of magnitude over the last 20 years in urban locations.

- Annual mean black smoke concentrations in London appear to have fallen by about a factor of 50 over the 80 years during which black smoke records have been kept, largely as a result of the phase-out of coal burning.

- The average black smoke concentration over the 197 available sites fell from 14.5 µg m^{-3} in 1990 to 7.1 µg m^{-3} in 2002. The annual trend was –0.67 µg m^{-3} per year or –5% per year. Over this same period, black smoke emissions are estimated to have declined by –6.8% per year.

- Black smoke levels declined at the Northern Ireland sites by –1.1 µg m^{-3} per year or –5% per year. The trends would indicate that substantial progress is being made with smoke control and that, if maintained, black smoke levels in Northern Ireland will, within the next 6 years or so, become comparable to those in the rest of the UK.

- Between 1990 and 2002 London sites showed some of the smallest black smoke trends of all the regional groupings: only –0.5 µg m^{-3} per year or –2.8% per year due to a significant contribution to black smoke emissions from diesel road traffic.

- Particulate sulphate levels in rural locations are falling with highly statistically significant annual trends that lie in the range of –4 and –9% per year. Over the period from 1990 to 2002, particulate sulphate levels appear to have fallen by more than a half, with the UK average concentration falling from 1.2 µg S m^{-3} to 0.5 µg S m^{-3}. The observed trends in particulate sulphate levels are significantly smaller than the decline in UK SO_2 emissions over the same period, which averaged about –11% to –12% per year. This may point to an increasing fraction of the emitted SO_2 being oxidised and present in the atmosphere as particulate sulphate.

- Across the UK, 48 PM_{10} monitoring sites have long enough records for trend determination and all but three of the sites show downwards trends in annual mean PM_{10} concentrations, with about half being highly statistically significant. The steep decline in urban PM_{10} levels observed from 1992 to 1999 has given way to a flattening out and a slight increasing trend from 2000 to 2003.

- From 1997 to 2003, network average annual mean PM_{10} concentrations declined from 23.4 μg m^{-3} to 20.5 μg m^{-3}, indicative of a trend of about –0.5 μg m^{-3} (–2%) per year. Some sites in the London area have exhibited trends distinctly smaller than the network average or close to 0.

- The observed downwards trends in PM_{10} reported for rural sites are significantly larger than those trends that could be explained solely by the observed trends in particulate sulphate over the same period. There must, therefore, be significant proportions of primary or other secondary PM in the PM_{10} size range, reaching these rural sites, and these components must also have been showing significant downwards trends.

- There is only one long-running time series of $PM_{2.5}$ measurements in the UK and these are for Hodge Hill, Birmingham. Annual mean $PM_{2.5}$ concentrations over the 7-year period 1995–2002 have shown a statistically significant downwards trend of –0.4 μg m^{-3} (3%) per year. On the basis of the observed trends in particulate sulphate at Stoke Ferry, a downwards trend of –0.4 μg PM_{10} m^{-3} per year (by scaling up to $(NH_4)_2SO_4$ from sulphur) would be anticipated, close to that observed at the Hodge Hill site. This would point to the trends in secondary sulphate particles as the main driving force behind the $PM_{2.5}$ trends at the Hodge Hill site.

- Over the 7-year period, PM_{coarse} levels at the Hodge Hill, Birmingham site have shown a highly statistically significant downwards trend of –0.7 μg m^{-3} (–13%) per year.

- Normalised monthly mean PM_{10} trends within the LAQN have shown a 35% decrease by 2000 relative to 1st January 1996, followed by a period of substantial increase so that by the end of 2003, PM_{10} levels have declined by only 19%. The normalised trends of PM_{10} and NO_x are diverging increasingly because the normalised trends in NO_x are monotonically downwards.

- Particulate cadmium and copper trends during 1990 and 2002 in the multi-element network have shown highly statistically significant downwards trends at Glasgow, London Cromwell Road and Motherwell. Chromium trends have been upwards at London Brent and Glasgow. Trends in manganese have been downwards at five sites, but London Brent has shown no overall trend. Nickel and vanadium have shown downwards trends at all six sites.

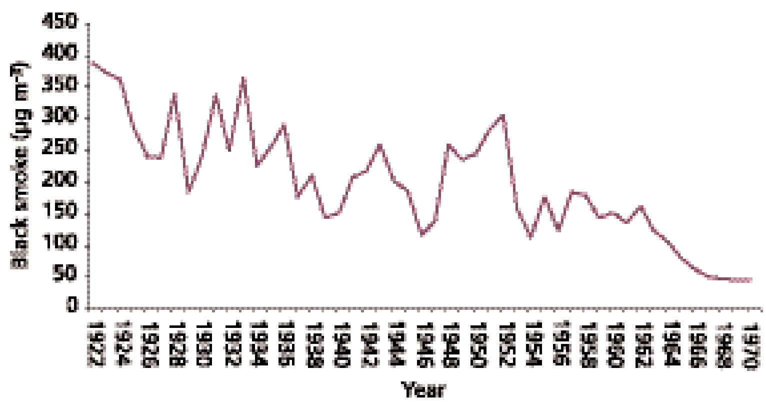
- Particulate iron levels have shown highly statistically significant downwards trends at Motherwell, Leeds and London Cromwell Road from 1990 to 2002. The upwards trend in particulate iron observed at the London Brent site could indicate a steady increase in non-exhaust traffic emissions due to traffic. This could explain why the observed PM_{10} trends at the London Brent and London Marylebone Road were significantly smaller than those observed elsewhere in the UK.

7.1 Historical setting

702. Particulate monitoring began over 80 years ago using the black smoke method and has continued through to the present day, making it the most heavily and continuously monitored of all the urban pollutants. Figure 7.1 presents the time series of black smoke measurements made at the Kew Observatory from 1922 through to the end of the 1960s, when the site closed. Black smoke appears to have been declining throughout the five decades during which these measurements were made, a decline which has been barely influenced by the passage of the Clean Air Act in 1956. The downwards trend must have been influenced by economic factors such as the change of London from a manufacturing to a trading centre, the substitution of coal burning for electricity and town gas and, more recently, the use of oil and natural gas instead of coal. Similar trends in changes to black smoke levels will have been seen in all the major population and industrial centres throughout the UK.

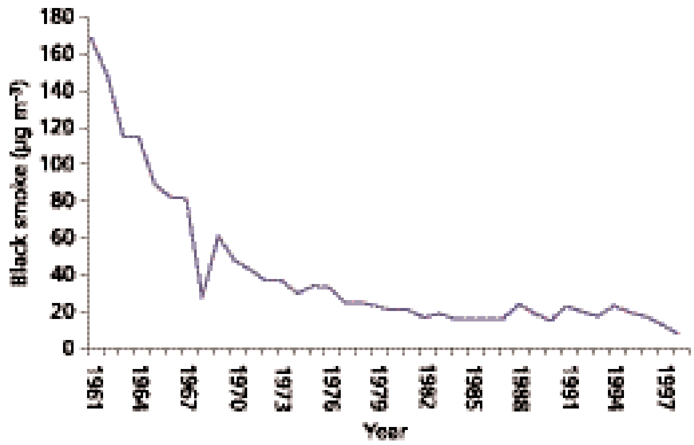
703. Figure 7.2 shows how black smoke concentrations have continued to decline since 1961 through to the present day. Marked downwards trends have been seen during the 1990s in London; these are the subject of a more detailed analysis below. Again, similar trends during the 1960s to 1990s have been reported across the UK. The network of black smoke monitoring sites reached its peak during the 1960s with over 1200 monitoring sites. The network has been shrinking in size during the 1990s but complete records are still available for 197 sites. Although absolute measurements using the black smoke method are of limited validity, they still serve as a useful indicator of long-term trends in suspended PM. Current (2002) annual mean black smoke concentrations in London are now about 6–9 µg m^{-3} compared with historic (1922) levels of about 380 µg m^{-3}, a decrease of about a factor of 50 over the 80 years of the measurement record.

Figure 7.1 Black smoke concentrations at the Kew Observatory, London between 1922 and 1970.



704. The decline in coal use during the second half of the 20th century has influenced not only black smoke levels but has also led to declines in the concentrations of a wide range of trace elements in suspended PM. Table 7.1 shows an example of the trends in iron aerosol at four rural locations during 1972–1981 and 1982–1991 (Cawse *et al*., 1994). Similar trends have been reported for other trace elements at these locations, including particulate antimony, arsenic, caesium,

Figure 7.2 Black smoke concentrations in Lambeth, London between 1961 and 1998.



calcium, cerium, chromium, cobalt, copper, europium, indium, lanthanum, magnesium, manganese, nickel, potassium, samarium, scandium, selenium, silver and zinc.

Table 7.1 Rural concentrations of particulate iron at four locations between 1972 and 1991, showing mean concentrations and annual trends (Cawse *et al*. 1994).

Location	Mean	Concentrations (ng m^{-3})		
	1972–1981	Trend (% change per year)	Mean 1982–1991	Trend (% change per year)
Harwell, Oxfordshire	285	–6.0	172	3.3
Styrrup, Nottinghamshire	682	–11.2	460	–5.2
Trebanos, Glamorgan	372	–9.1	318	NST
Wraymires, Cumbria	242	NST	98	–4.2

NST, no significant trend.

705. In addition to the decline in coal usage, the phase-out of lead in petrol has exerted a significant influence on particulate lead concentrations throughout the UK from the 1980s onwards. Particulate lead concentrations have declined by more than an order of magnitude over the last 20 years in urban and rural locations. Levels have fallen from 770 ng m^{-3} in 1980 at the London Brent site to 22 ng m^{-3} in 2002, whereas at the rural site Chilton, Oxfordshire the fall has been from 110 ng m^{-3} to 8 ng m^{-3}. At the remote rural site, Eskdalemiur in Dumfries and Galloway, levels have fallen from 29 ng m^{-3} to 3 ng m^{-3}.

7.2 Trends during the 1990s and onwards

7.2.1 Black smoke trends

706. Trend analysis has been performed on a dataset of annual mean black smoke concentrations from 1990 to 2002 obtained from the longest running 197 UK sites. Each time series was subjected to two types of statistical analysis. The first tested for the presence of a monotonic increasing or decreasing trend with the nonparametric Mann-Kendall test; the second estimated the slope of a linear trend with the nonparametric Sen's method (Salmi *et al*. 2002). Of the 197 times series analysed, 15 showed upwards trends and 184 showed downwards trends. A total of 66 black smoke sites showed statistically significant trends at the 99.9% level of significance, 56 at the 99% level, 56 at the 95% and 9 at the 90% level, that is, 161 sites showed trends that were highly statistically significant. None of the upwards trends were highly statistically significant.

707. The network average black smoke concentration fell from 14.5 µg m^{-3} in 1990 to 7.1 µg m^{-3} in 2002. The trend was –0.67 µg m^{-3} (–4.8%) per year and this was highly statistically significant at the 99.9% level. Over this same period, according to the NAEI, UK national total black smoke emissions declined at –6.8% per year, a somewhat higher rate compared with the observed trend across the black smoke network.

708. The network average necessarily hides large variations across the UK and so the sites were assembled into regional groups to allow a more detailed analysis of the trends in black smoke levels. Table 7.2 summarises the trends in annual mean black smoke concentrations for the different regions across the UK, ranked in order of their 1990 average concentrations. The regional grouping with the highest annual mean concentrations at the start of the trend period was that encompassing the 38 sites identified as being located in the Yorkshire, Derbyshire, Durham and Lothian coalfields. These sites showed the largest downwards trends of all and almost all were highly statistically significant. These trends averaged –1.55 µg m^{-3}(–7.3%) per year and were large enough to bring this category from the most polluted regional grouping down to fourth most polluted. The phase-out of coal burning in these coalfield communities has clearly been associated with significant downwards trends in black smoke levels. The observed downwards trends agree closely with those reported in UK national total black smoke emissions over this same period in the NAEI.

709. Black smoke levels declined at the Northern Ireland sites by –1.1 µg m^{-3} (–5.3%) per year, see Table 7.2. These sites were ranked the second most polluted regional grouping at the start of the trend period and the most polluted at the end. Two-thirds of the observed trends were highly statistically significant. The trends indicate that substantial progress is being made with smoke control and that, if maintained, black smoke levels in Northern Ireland will, within 6 years or so, become comparable with those in the rest of the UK.

710. The West Midlands sites were the third most polluted regional grouping at the start of the trend period and, despite showing highly statistically significant downwards trends, remained with this ranking at the end of the period. The phase-out of coal combustion has halved black smoke levels, but they still remain significantly above the UK network average, presumably because of contributions from other sources, road transport and industry.

Table 7.2 Summary of the analysis of trends in the annual mean black smoke concentrations monitored in different regions of the UK between 1990 and 2002, showing the annual mean concentrations in 1990, the annual trends and the numbers of sites showing highly statistically significant trends for each region.

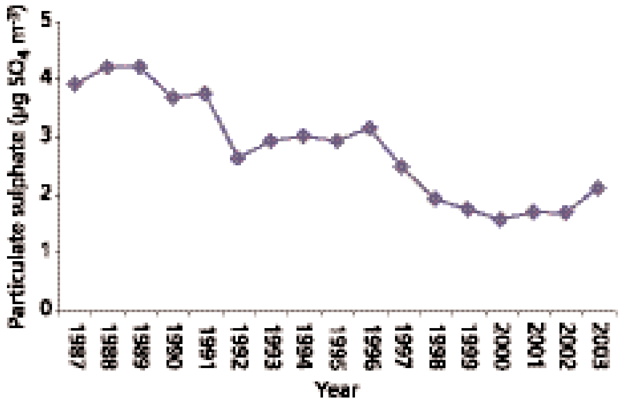
	Annual mean 1990 (μg m⁻³)	Annual trend 1990–2002		Number of sites				
		(μg m⁻³ per year)	(% per year)	Total	Statistical significance of annual trends			
					P < 0.001	*P* < 0.01	*P* < 0.05	*P* < 0.1
Coalfields	21.2	–1.55	–7.3	38	23	11	2	—
Northern Ireland	19.0	–1.10	–5.3	12	4	4	—	—
West Midlands	18.1	–1.13	–5.4	7	2	2	—	1
Northwest	14.5	–0.70	–4.6	35	15	9	—	3
Northeast	14.2	–0.66	–4.3	24	7	8	2	—
London	13.7	–0.52	–2.8	12	3	1	2	—
East Midlands	13.4	–0.70	–5.1	10	3	2	—	1
Southwest	11.4	–0.53	–4.5	4	1	2	—	1
Scotland	11.2	–0.69	–6.1	17	6	4	4	1
Southeast	10.7	–0.43	–3.9	11	2	1	3	1
Wales	9.2	–0.41	–4.5	5	—	—	3	—
Suburban and rural sites	4.7	–0.02	0.3	22	—	1	4	1
Network average or total	14.5	–0.67	–4.8	197	66	45	20	9

711. The London sites showed some of the lowest trends of all the regional groupings, only –0.5 μg m^{-3} (–2.8%) per year. This had the effect of taking the London region from close to the middle of the rankings at the start of the trend period to the second most polluted region after Northern Ireland at the end. This situation appears to have resulted from the phase-out of coal burning having taken place largely before the start of the trend period, leaving a significant contribution from other sources, particularly road transport. The most polluted London sites (Lambeth, Acton, London City, Ealing, Greenwich and Ilford) have shown downwards trends of between –6.7% per year and –3.7% per year. In contrast, the least polluted sites, with annual mean black smoke concentrations in the range 6–9 μg m^{-3}, have shown no significant trends during the period from 1990 to 2002. Since these levels are considerably higher than background black smoke levels (2 μg m^{-3} at Lerwick), the presence of a road traffic contribution is presumably the explanation for the lack of observed trends.

7.2.2 Particulate sulphate trends

712. Particulate sulphate levels have been falling steadily across Europe in response to a reduction in regional SO_2 emissions. Figure 7.3 shows the trend in the UK network average for the period 1987–2003 and indicates a substantial downwards trend. The annual average concentrations up to 2001 were calculated for the eight NETCEN Acid Rain Monitoring Network sites, extended to 2003 using the 12-site CEH network. Table 7.3 summarises the trend data from the rural particulate sulphate monitoring carried out as part of the Acid Rain Monitoring Network. UK particulate sulphate levels are falling with highly statistically significant annual trends that lie in the range of between –4.3% and –9.3% per year. Over the period 1990–2002, particulate sulphate levels appear to have fallen by more than half, with the UK average concentration falling from 3.7 µg SO_4 m^{-3} to 1.6 µg SO_4 m^{-3}.

Figure 7.3 Trends in the annual average particulate sulphate concentrations between 1987 and 2003.



713. The observed trends in particulate sulphate levels are somewhat smaller than the decline in UK SO_2 emissions over the same period, which averaged about –11% to –12% per year, and those of the EC by –10% per year. This would point to an increasing fraction of the emitted SO_2 being oxidised and present in the atmosphere as particulate sulphate. This may have resulted as a by-product of the decreased NO_x emissions and hence increased photochemical oxidation rate for SO_2 to sulphate aerosol.

714. This apparent increase in the oxidising capacity has been observed across Europe. Figure 7.4 shows the long-term measurements of sulphur dioxide and particulate sulphate at the EMEP GB02 site at Eskdalemuir, Dumfries and Galloway during the period 1977–2002. Although SO_2 concentrations have fallen by a factor 7, those

Figure 7.4 Long-term trends in sulphur dioxide and particulate sulphate (µg S m^{-3}) observed at Eskdalemuir, Dumfries and Galloway between 1977 and 2002.

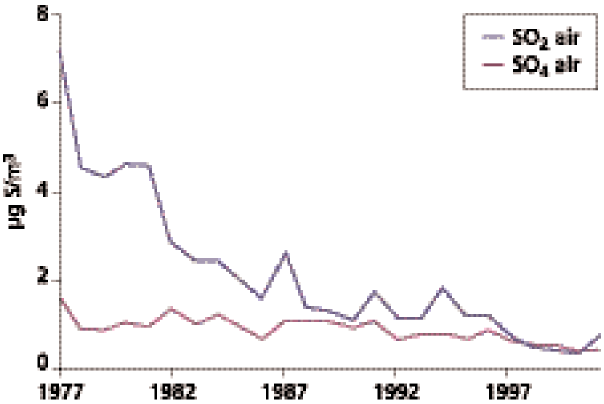


Table 7.3 Annual mean particulate sulphate concentrations at UK rural locations in 1990 and 2002 and their annual trends between1990 and 2002.

	Annual mean (μg S m^{-3})		Annual trend		
	1990	2002	μg S m^{-3} year 1	% per year	Statistical significance
Eskdalemuir	0.94	0.46	0.043	4.5	$P < 0.001$
Stoke Ferry[a]	1.66	0.72	0.093	5.4	$P < 0.001$
Lough Navar	0.81	0.40	0.046	4.9	$P < 0.01$
Barcombe Mills	1.48	0.71	0.076	4.7	$P < 0.01$
Yarner Wood	1.21	0.58	0.064	5.0	$P < 0.01$
High Muffles	1.33	0.59	0.068	5.0	$P < 0.001$
Strathvaich Dam[a]	0.87	0.32	0.044	5.9	$P < 0.01$
Glen Dye[a]	1.52	0.46	0.049	5.3	$P < 0.01$

[a]Data and trends refer to 1990–2001.

of particulate sulphate have fallen only by a factor 2. The fraction of airborne sulphur present as particulate has thus increased from ~0.18 to 0.33, a near doubling over the 25-year period.

7.2.3 PM_{10} trends

715. There are 12 long-running PM_{10} monitoring sites across the UK that allow trend determination over the period up to 2003; the time series of their annual mean PM_{10} concentrations are shown in Figure 7.5. All trend analyses for PM_{10} have used unscaled TEOM data. Also shown in Figure 7.5 is the average concentration for these long-running sites. Clear downwards trends are apparent in all cases. All these sites and their average showed highly statistically significant downwards trends over the period up to 2003. The observed downwards trends were between –0.3 and –1.7 μg m^{-3} per year or between –1.7 and –10.5% per year, see Table 7.4. The 12-site average annual mean concentration has shown a highly statistically significant downwards trend of –0.9 μg m^{-3} year 1 or –4.4% year 1. Interestingly, Figure 7.5 shows that the strength of this downwards trend has not remained constant over the 1992 2003 period. The downwards trends appears to have slowed significantly and there is a strong hint that it may have been reversed during 2000 to 2003.

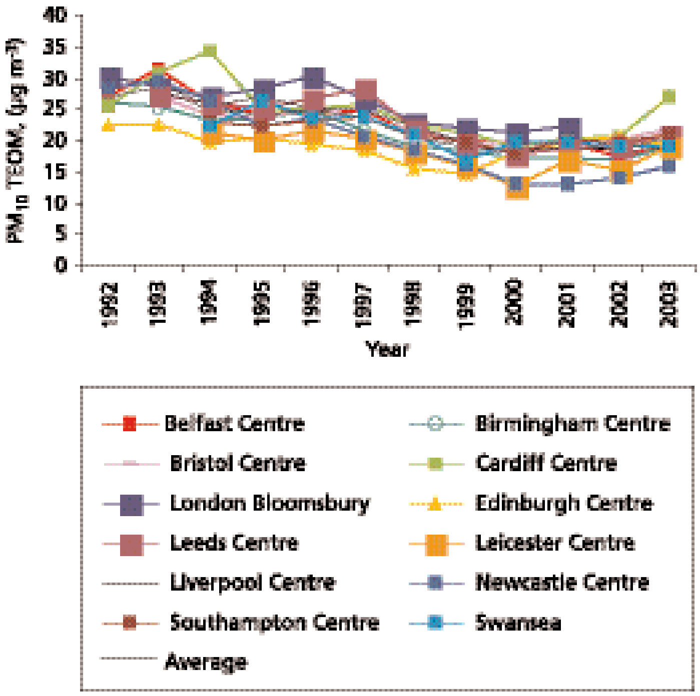
716 Across the UK, 48 PM_{10} monitoring sites have long enough records for trend determination during the 1997–2003 period and longer. The available trend data are summarised in Table 7.4, which gives the length of the monitoring record, the annual mean PM_{10} concentration in 2003, the annual trend in μg m^{-3} per year or percentage change per year and the statistical significance of the observed trend. All but three of the sites show downwards trends in annual mean PM_{10} concentrations, with just under half being highly statistically significant. The range of the observed downwards trends was from more than –14% per year to less than +3% per year. The network average annual mean PM_{10} concentrations declined from 23.4 μg m^{-3} to 20.5 μg m^{-3}, indicative of a trend of about 0.5 μg m^{-3} year^{-1} or –2% per year. Highly statistically significant downwards trends

Table 7.4 Annual mean PM_{10} concentrations and their recent trends, with their statistical significance at 48 locations, ranked in order of percentage change per year. (All analyses were performed on unscaled TEOM data.)

Site	First year	Last year	Annual mean 2003 (μg m^{-3})	Annual trend		Statistical significance
				μg m^{-3} year^{-1}	% change year^{-1}	
Stoke-on-Trent centre	1997	2003	11	–1.5	–14.0	$P < 0.01$
Reading	1997	2003	12	–1.5	–12.8	$P < 0.05$
Newcastle centre	1992	2003	16	–1.7	–10.5	$P < 0.001$
Stockport	1996	2003	15	–1.3	–8.4	$P < 0.01$
Liverpool	1993	2003	19	–1.2	–6.1	$P < 0.01$
Sheffield centre	1995	2003	21	–1.2	–5.9	$P < 0.05$
Norwich centre	1997	2003	18	–1.0	–5.8	—
Glasgow centre	1996	2003	16	–0.9	–5.7	$P < 0.05$
London A3 roadside	1997	2003	26	–1.4	–5.6	—
Leeds centre	1993	2003	21	–1.1	–5.0	$P < 0.05$
Birmingham centre	1992	2003	19	–1.0	–5.0	$P < 0.001$
Belfast centre	1992	2003	19	–0.9	–5.0	$P < 0.001$
Derry	1997	2003	18	–0.9	–4.9	—
Redcar	1997	2003	21	–1.0	–4.6	—
London Bloomsbury	1992	2001	23	–1.1	–4.6	$P < 0.01$
Glasgow kerbside	1997	2003	24	–1.0	–4.3	—
Narberth	1997	2003	14	–0.6	–4.1	—
Salford Eccles	1992	2003	18	–0.7	–4.1	—
Leicester centre	1994	2003	19	–0.7	–3.9	$P < 0.05$
Belfast Clara St	1994	2003	22	–0.8	–3.5	$P < 0.05$
Bristol centre	1993	2003	22	–0.7	–3.4	$P < 0.01$
Bradford centre	1997	2003	21	–0.7	–3.3	—
Swansea	1994	2003	19	–0.6	–3.3	$P < 0.05$
Middlesbrough	1995	2003	21	–0.7	–3.2	—
Nottingham centre	1996	2003	20	–0.6	–3.2	—
Cardiff centre	1992	2003	27	–0.8	–2.9	$P < 0.1$
Manchester Piccadilly	1995	2003	22	–0.6	–2.9	–
Port Talbot	1997	2003	24	–0.7	–2.8	$P < 0.05$
Camden kerbside	1996	2003	27	–0.8	–2.8	—
Leamington Spa	1996	2003	21	–0.6	–2.7	—
Haringey roadside	1996	2003	22	–0.5	–2.3	—

Site	First year	Last year	Annual mean 2003 (μg m^{-3})	Annual trend		Statistical significance
				μg m^{-3} year^{-1}	% change year^{-1}	
Hull	1994	2003	22	–0.5	–2.1	—
Plymouth centre	1997	2003	17	–0.4	–2.1	$P < 0.1$
Rochester	1996	2003	19	–0.4	–2.0	—
Bury roadside	1997	2003	27	–0.5	–1.9	—
Southampton centre	1994	2003	21	–0.4	–1.9	—
London N. Kensington	1996	2003	22	–0.4	–1.8	—
Edinburgh centre	1992	2003	19	–0.3	–1.7	$P < 0.1$
London Hillingdon	1996	2003	23	–0.4	–1.6	—
Wolverhampton centre	1995	2003	19	–0.3	–1.6	$P < 0.1$
Bolton	1997	2003	18	–0.2	–1.3	—
Birmingham East	1993	2003	17	–0.2	–1.2	—
Thurrock	1996	2003	23	–0.2	–0.8	—
London Brent	1996	2003	20	–0.1	–0.7	—
London Eltham	1996	2003	21	–0.1	–0.3	—
London Marylebone Road	1997	2003	37	0.0	0.0	—
Lough Navar	1996	2003	12	0.1	1.1	—
Scunthorpe	1997	2003	30	0.9	3.0	—

Figure 7.5 Time series of annual mean PM_{10} concentrations at the long-running urban background sites in the UK between 1992 and 2003.



during 1997–2003 were found at eight sites: Belfast centre, Birmingham centre, Glasgow centre, Plymouth centre, Port Talbot, Reading, Stockport and Stoke-on-Trent centre. The observed trends fell in the range of –0.4 to –1.5 μg m^{-3} per year. Slight upwards trends were reported for the Birmingham East, Edinburgh centre, Manchester Piccadilly, Middlesbrough and Scunthorpe sites.

717. The London conurbation appears to be showing somewhat different behaviour compared to the other network sites with regard to PM_{10} trends during the 1997–2003 period. At one site, London A3 roadside, the observed trend was higher than the 48-site trend. However, at all others either no trend was found (London Brent, London Eltham and London Marylebone Road) or small downwards trends in the range –1.0 to –1.8% per year were found (Camden kerbside, Haringey roadside, London Hillingdon, London North Kensington and Thurrock).

718. Urban PM_{10} trends have appeared to slow down during the period 2000–2003 compared to the 1993–1999 period. At some urban sites there is some evidence for increasing annual mean PM_{10} concentrations during 2000–2003. Bearing in mind that the year 2003 appears to have been influenced by some large-scale regional pollution episodes, it is an issue whether the trends during the 2000–2003 period have been overly influenced by 2003. The residuals in the trend analyses have been examined to see if they significantly concurred with the other years. For 33 of the sites in Table 7.4 it was apparent that 2003 did not stand out as particularly high in terms of the 2000–2003 period. However, for 15 sites, that is, about one-third, the year 2003 stood out as significantly different from the overall trend. There was, however, no clear spatial pattern in the distribution of the sites that showed the year 2003 as an outlier.

719. The rural sites at Narberth in Wales and Rochester in Kent show PM_{10} downwards trends of –0.6 μg m^{-3} per year and –0.4 μg m^{-3} per year, respectively. It is possible that the observed PM_{10} trends at these sites are driven by the simultaneous trends in particulate sulphate described in Section 7.2.2 above. The particulate sulphate trends estimated for Narberth and Rochester, accordingly, are –0.06 μg S m^{-3} per year and –0.08 μg S m^{-3} per year or –0.25 and –0.33 μg PM_{10} m^{-3} per year (by scaling up to $(NH_4)_2SO_4$ from S). It is clear, therefore, that the observed downwards trends in PM_{10} reported for these rural sites are larger (by factors of 1.2 to 2.5) than those trends that could be explained solely by the observed trends in particulate sulphate over the same period. There must therefore be significant proportions of primary or other secondary PM in the PM_{10} size range, reaching these rural sites and these proportions must have been showing significant downwards trends.

720. The rural site at Lough Navar, Northern Ireland shows the least PM_{10} trend of three rural sites, Lough Navar, Narberth and Rochester. The levels monitored at this site are the lowest reported for any of the 48 sites and have remained consistently in the range 9.4 to 9.9 μg m^{-3} for 5 of the 8 years that the site has been operational. This site might have been expected to show some evidence of a downwards trend if only from the decreasing long range transport of particulate sulphate, as shown by the Eskdalemuir site. The annual means for 2000–2003 have been consistently higher than the 9.4–9.9 μg m^{-3} range and it is clear from the residuals in the trend analysis that this reflects a reversal in trend rather than the influence of year-on-year variability caused by 2003.

721. Strong downwards trends are thus a widely observed feature of urban PM_{10} levels in the UK during the 1990s, before the apparent levelling off during the 2000–2003 period. Average PM_{10} concentrations are declining at about –4.4% per year, as indicated by the 12 long-running urban background sites during the 1990s, and this is close to the trend in PM_{10} emissions that have, over the same period, declined at about –5% per year, according to the NAEI.

722. Although PM_{10} emissions have continued to decline strongly from 1997 to 2003, this is not the case for urban PM_{10} concentrations. Only 12 of 48 sites achieved annual percentage trends during the 1997–2003 period that approached those shown by PM_{10} emissions. The 48-site average showed a trend of –2.2% $year^{-1}$, one-half of the trend shown by the estimated PM_{10} emissions over the same period. The PM_{10} emission inventory is therefore not giving a clear indication of the likely origin of the observed slowing up in the downwards trends in urban PM_{10} levels observed during 2000–2003.

7.2.4 $PM_{2.5}$ trends

723. There is only one long-running time series of $PM_{2.5}$ measurements in the UK and these are for Hodge Hill, Birmingham, a suburban background site operated by Birmingham City Council. Annual mean $PM_{2.5}$ concentrations were found to be 14 μg m^{-3} during 1995 and 12 μg m^{-3} in 2002. Over the 7-year period, $PM_{2.5}$ levels have shown a statistically significant (at the 90% level of significance) downwards trend of –0.4 μg m^{-3} (–3.1%) per year.

724. According to the NAEI, emissions of $PM_{2.5}$ have declined by –5.2% per year over the period 1995–2001. This decline in emissions is somewhat larger than the observed decline in $PM_{2.5}$ concentrations in Birmingham. This would suggest that primary emissions are not the main source of $PM_{2.5}$. Particulate sulphate is likely to be a component of the $PM_{2.5}$ fraction and is likely to be declining at about –0.09 μg S m^{-3} per year, based on the Stoke Ferry observations shown in Table 7.3. On this basis, a downwards trend of –0.4 μg $PM_{2.5}$ m^{-3} per year (by scaling up to $(NH_4)_2SO_4$ from S) would be anticipated, close to that observed at the Hodge Hill site. This would point to secondary sulphate particle trends as the main driving force behind the $PM_{2.5}$ trends at the Hodge Hill site and not primary emission trends.

Table 7.5 Observed trends in annual mean $PM_{2.5}$, PM_{10} and PM_{coarse} concentrations at four AURN sites between 1998 and 2003.

Site	$PM_{2.5}$ trends (μg m^{-3} $year^{-1}$)	PM_{10} trends (μg m^{-3} $year^{-1}$)	PM_{coarse} trends (μg m^{-3} $year^{-1}$)
London Marylebone Road	0.0	0.5	0.5
London Bloomsbury	–0.4	—	—
Harwell	0.3	0.0	0.0
Rochester	0.0	0.5	0.0

725. Co-located measurements of PM_{10} and $PM_{2.5}$ concentrations have been made at four AURN sites from 1998 onwards and their time series are just long enough to enable comments to be made about trends. Table 7.5 indicates that annual mean $PM_{2.5}$ concentrations have shown no overall trend at the London Marylebone Road and Rochester sites, an increasing trend at the Harwell site and a decreasing trend at the London Bloomsbury site, although none of these trends was highly statistically significant. The observed downwards trend in $PM_{2.5}$ at London Bloomsbury of –0.4 μg m^{-3} per year corresponds closely with the downwards trend of –0.4 μg $PM_{2.5}$ m^{-3} per year anticipated at Stoke Ferry on the basis of the observed trend in particulate sulphate.

7.2.5 PM_{coarse} trends

726. There is only one site in the UK for which long-running time series are available for both PM_{10} and $PM_{2.5}$ using co-located instruments, and this is the Hodge Hill, Birmingham site referred to in Section 7.2.4 above. Annual mean PM_{coarse} concentrations were monitored and found to be 9 μg m^{-3} in 1995 and 3 μg m^{-3} in 2002. Over the 7-year period, PM_{coarse} levels have shown a highly statistically significant (at the 99% level of significance) downwards trend of –0.7 μg m^{-3} (–13%) per year. There are no other time series that can be used to corroborate such a large and significant trend in particulate levels. The corresponding trend in PM_{10} levels observed at this site is –1.1 μg m^{-3} (–4.9%) per year. This is somewhat higher than the trends for the Birmingham centre and Birmingham East sites of –1.0 and –0.6 μg m^{-3} per year or –3.7 and –2.8% per year, reported for the same time period. On this basis, it would appear that at the Hodge Hill, Birmingham site, much of the observed trend in PM_{10} levels has been caused by the decline in PM_{coarse} levels.

727. The strong downwards trend in PM_{coarse} levels could point to coarse fraction secondary nitrate particles as the main source of the observed downwards trend in PM_{coarse} levels at the Hodge Hill site, through the influence of the halving of European regional scale NO_x emissions. An alternative explanation could be that there has been a steep decline in PM_{coarse} emissions during 1995–2002. The NAEI shows PM_{coarse} emissions declining by –4.4% $year^{-1}$ over during 1995 to 2001, thus providing some explanation for the large observed trend at the Hodge Hill, Birmingham site.

728. Table 7.5 indicates that at the four AURN sites from 1998 to 2003, annual mean PM_{coarse} concentrations have exhibited increasing trends at the London Marylebone Road and no trends at the rural sites (Harwell and Rochester), although none of these trends was highly statistically significant.

7.2.6 Ultrafine particle number trends

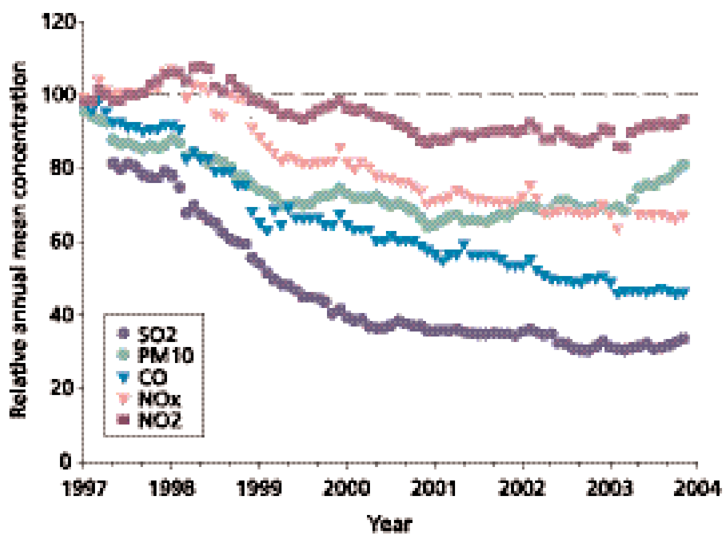
729. Particle number count is dominated by the ultrafine particle size range. An analysis of UK particle number data is presented in Chapter 6, but the time-series of measurements are insufficient to be able to comment on the long-term trends. Elsewhere in Europe, particle number measurements made each winter from 1991/1992 to 2000/2001 in Erfurt, Germany, have shown that, whereas the mass concentration of fine particles decreased substantially over this period (by 75%), the total number of ultrafine particles remained fairly constant. This was the consequence of a significant shift in the size distribution towards smaller sizes, with a twofold increase in the number concentration between 10 and 30 nm (Kreyling *et al*., 2003). In Helsinki, Finland, annual mean particle number concentration (for the 8–400 nm size range) declined by about 25% between

1997 and 2003 (Hussein *et al*. 2004). The very limited evidence, therefore, suggests that average ultrafine particle number concentrations are declining and shifting to smaller sizes, but are likely to show a different trend to PM_{10}. It is important to remember that particle number concentrations exhibit wide fluctuations in short-scale spatial and temporal variability about longer term means.

7.2.7 Normalised monthly mean PM_{10} trends within the LAQN

730. Figure 7.6 shows the trends in different pollutants in London from the beginning of 1996. Annual data have been averaged at monthly intervals and hence show a start date of 1997. Concentrations have been normalised to 100 at the beginning of the dataset to highlight the relative change in the concentrations of the different species. A range of site types and sites mostly from the LAQN has been used for the different species, depending on data availability and the need for a consistent dataset since 1996. Most pollutants have shown a steady decline in concentration since 1996. The most significant reductions have been observed for SO_2 and CO, where concentrations have declined by over 50% between 1996 and 2003. For PM_{10}, the annual mean concentration declined rapidly until 1999, but has seen a significant increase during 2003 due to the frequency of pollution episodes in that year. Since 1996 PM_{10} concentrations have declined by 19% by the end of 2003.

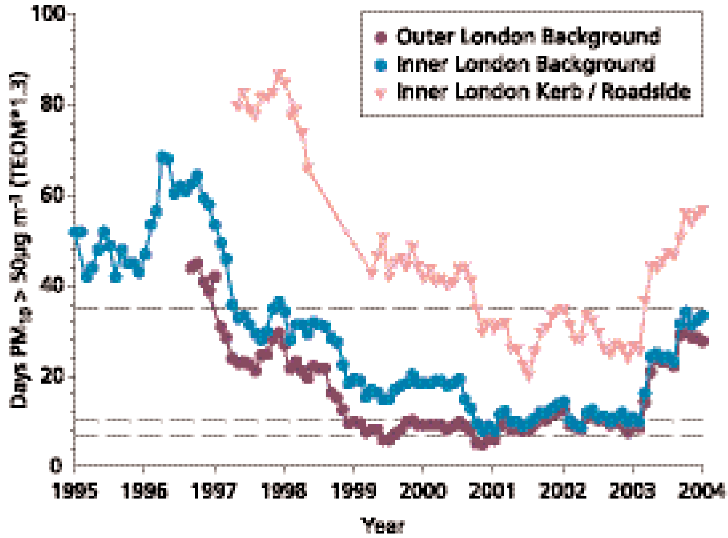
Figure 7.6 Relative annual mean concentration (monthly intervals) for a selection of monitoring sites in London. (Data for 2003 are provisional.)



731. The sites in the LAQN, therefore, seem to be showing the same behaviour in PM_{10} trends as the other urban background sites throughout the UK. Annual mean PM_{10} trends were strongly downwards during the 1990s but flattened out during 2000–2003 and have shown a slight increase during 2003. Figure 7.6 shows that the PM_{10} and NO_x curves followed each other closely during 1997 2000 and then began to diverge increasingly from 2001 onwards. This divergence has been maintained through 2003, a year associated with large regional pollution episodes.

732. Figure 7.7 shows how the daily mean concentrations >50 μg m^{-3} have varied at three different locations in London. These data have been compiled in a similar

Figure 7.7 Days when the daily mean PM_{10} concentration was >50 μg m^{-3} (TEOM * 1.3) for different categories of monitoring sites in London. Annual data have been averaged at monthly intervals.



way to that used for Figure 7.6. It is clear that there is significant interannual variability in this statistic, which highlights the importance of different episodes in different years. Of note are the episodes during 1996 and 2003. The most recent pollution episodes during 2003 resulted in the inner London roadside and kerbside sites exceeding the 35 days objective, with the inner London background sites being very close to this limit. Also shown in Figure 7.6 are the levels of the different limits and objectives shown at 35, 10 and 7 days.

7.2.8 Trends at sites close to motorways

733. TRL operates two long-running sites, on behalf of the Highways Agency, by the side of the M4 and M25 motorways. The M4 site measures PM_{10} and the M25 site measures both PM_{10} and $PM_{2.5}$. During 1996–2003 the annual mean PM_{10} concentrations showed downwards trends of –0.33 and –0.85 μg m^{-3} $year^{-1}$ at the M4 and M25 sites, respectively, although neither were highly statistically significant. Interestingly, both sites showed strong downwards trends at the start of the period, which flattened out and then showed signs of an increase during 2000–2003, as exhibited by many urban background and roadside sites.

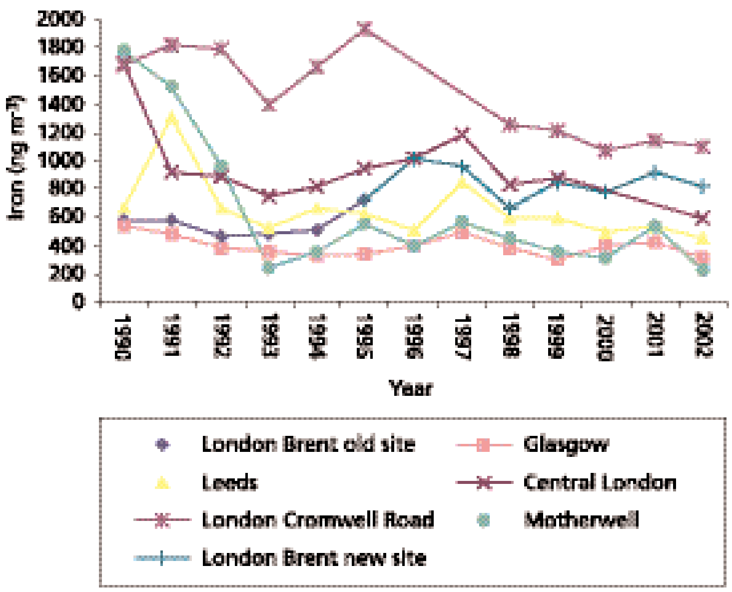
734. During 1999–2003 there were no significant trends in the annual mean PM_{10}, $PM_{2.5}$ or PM_{coarse} concentrations at the M25 site.

7.2.9 Trends at the multi-element sites

735. Up to eight trace elements are monitored at six sites in the multi-element network. During 1990 to 2002, highly statistically downwards trends were observed for cadmium at the Glasgow, London Cromwell Road and Motherwell sites, but no trends were observed at the Leeds, Central London and London Brent sites. Highly statistically significant upwards trends were reported for chromium at the London Brent and Glasgow sites. Upwards trends were also observed for chromium at the Leeds, London Cromwell Road and Motherwell sites and downwards at the Central London site, but these were not highly statistically significant. Highly statistically significant downwards trends were observed for copper at the Glasgow, London Cromwell Road and Motherwell sites. Manganese showed highly statistically significant downwards trends at five of the six sites, with no trend detected at the London Brent site. Nickel and vanadium showed highly statistically significant downwards trends at all six sites.

736. Iron is an important trace element in its own right. It is also an important indicator species in source attribution analyses. It is appreciably enriched in roadside air and appears to be associated with the non-exhaust emissions from road traffic (Harrison *et al.*, 2003, 2004). Its origin is probably brake wear and corrosion products, although the relative amounts of direct emissions and non-exhaust traffic emissions are not known. It is also present in soils, but at much lower abundance. Figure 7.8 plots the time series for annual mean iron concentrations at the six multi-element sites for the period 1990 to 2002. Levels at the Motherwell site declined quickly during the early 1990s following the cessation of industrial activities close to the site. The Leeds and Glasgow sites showed steady declines throughout the 1990s. The behaviours observed at the three London sites were characteristically different from each other. The London Cromwell Road and Central London sites showed steady declines as seen for the Leeds and Glasgow sites, but the London Brent site showed a steady increase, although the site location was changed in 1995. Table 7.6 shows that this steady increase at the London Brent site is highly statistically significant.

Figure 7.8 Time series of the annual mean iron concentrations observed at the multi-element sites between 1990 and 2002.



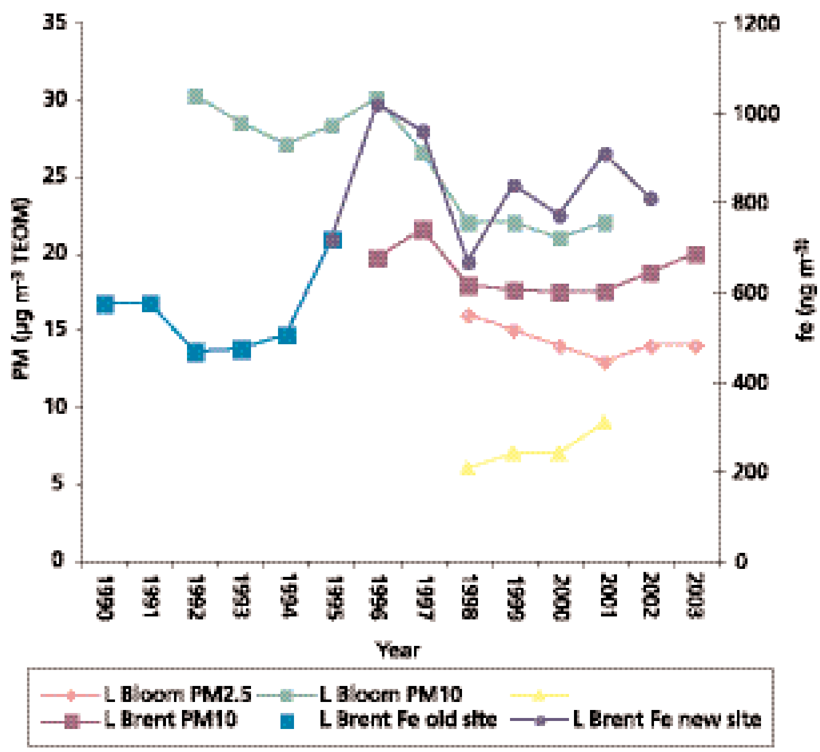
737. Figure 7.9 shows the time series for particulate iron and PM_{10} at London Brent from 1990 to 2003. Also shown are the PM_{10}, $PM_{2.5}$ and PM_{coarse} measurements for London Bloomsbury. The iron at London Brent shows a steady upwards trend with a major excursion during 1995 to 1997, although the site location was changed in 1995. The PM_{10} levels also show elevated levels during 1996–1997 and steady upwards trends thereafter exactly analogous to the behaviour shown by particulate iron. PM_{10} and particulate iron were both high during 1996–1997, which was marked by a number of intense regional pollution episodes.

Table 7.6 Details of the observed trends in the annual iron concentrations at the multi-element survey sites from 1990 to 2002.

Site	Start	Finish	Annual mean in 2002 ($ng\ m^{-3}$)	Annual trend ($ng\ m^{-3}\ year^{-1}$)	Annual trend (% change per year)	Statistical significance
London Brent[a]	1990	2002	810	32.6	4.0	$P < 0.05$
Central London	1990	2002	313	22.0	7.0	—
Glasgow	1990	2002	448	7.4	1.6	—
Leeds	1990	2002	598	17.1	2.9	$P < 0.05$
London Cromwell Road	1990	2002	1102	52.7	4.8	$P < 0.05$
Motherwell	1990	2002	224	69.9	31.2	$P < 0.05$

[a]Site change in 1995.

Figure 7.9 Time series for particulate iron and PM_{10} at London Brent and of PM_{10}, $PM_{2.5}$ and PM_{coarse} at London Bloomsbury, between 1990 and 2003.



738. The highly statistically significant upwards trend in iron observed at the London Brent site could indicate an upwards trend in non-exhaust traffic emissions due to increasing traffic levels. It could also explain why the observed PM_{10} trends at the London sites, particularly the London Brent and London Marylebone Road sites, were significantly smaller than those observed elsewhere in the UK.