Assessment of benzo[a]pyrene atmospheric concentrations in the UK to support the establishment of a national PAH objective

A report produced for the Department for Environment, Food and Rural Affairs, the National Assembly for Wales, the Scottish Executive and the Department of the Environment for Northern Ireland

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Executive Summary

This report presents the results of a number of assessments designed to inform the development of a possible National Air Quality Strategy objective for polycyclic aromatic hydrocarbons (PAHs). PAH exposure has been associated with a range of cancer endpoints including lung, skin and bladder cancers. Some evidence supports a link between lung cancer and PAH exposure through inhalation. The Expert Panel on Air Quality Standards has recommended setting a national Standard for PAHs using annual average concentrations of benzo[a]pyrene (BaP) as a marker for the human health effects of PAHs. EPAQS has recommended an annual average concentration of 0.25 ng/m^3 as a guideline. A European Commission working group has recommended a limit value between 0.5 – 1.0 ng/m^3.

In 1999, the most recent year for which emissions estimates have been calculated, total UK emissions of BaP were 10.3 t. The main sources of emissions were from domestic coal and wood burning (39%), fires (28%), anode baking (19%) and vehicles (8%). Emissions of BaP have declined markedly since 1990 when the national total emission is estimated to have been 68 t.

Emissions of BaP are predicted to decline to 6.4 t in 2010, thereafter showing a slight increase to 6.6 t in 2020. The sectors contributing to the decline in emissions are road transport, the production of anodes for the aluminium industry and domestic coal use. The sectors whose emissions are predicted to increases are principally aluminium production and domestic wood burning.

 The emission inventory for BaP is subject to considerable uncertainty. This is since few emission measurements have been made both nationally and internationally and the relevant activity statistics in some cases are not collected regularly. The diffuse nature of some sources also increases the uncertainty in the emissions estimates. In particular, further work is required to understand better the magnitude and spatial pattern of emissions from accidental, malicious, demolition and natural fires, bonfires and domestic solid fuel use particularly in urban and rural communities without access to natural gas.

AEA Technology's National Environmental Technology Centre (NETCEN) operates the PAH network (15 sites) and TOMPs network (6 sites) measuring concentrations of PAHs in ambient air on behalf of DEFRA and the devolved administrations. The concentrations of BaP in 2000 ranged between 0.04 ng m^{-3} at rural sites to 2.28 ng m^{-3} at an industrial site. Concentrations of BaP at 7 of the 12 urban/industrial sites operating in 2000 equalled or exceeded 0.25 ng m^{-3}. However only at Kinlochleven (2.28 ng m^{-3}), Lisburn (0.93 ng m^{-3}) and Scunthorpe (1.17 ng m^{-3}) were concentrations significantly greater than the EPAQS guideline. The aluminium smelter at Kinlochleven closed in June 2000. The Lisburn site is in an area of Northern Ireland which is not a smoke control area and does not yet have a supply of natural gas. The Scunthorpe site is near both a major integrated steel works and a coal tar processing plant.

Statistical analysis of the measurements from those sites at which robust pollution trend analysis is possible suggests that there has been a significant decline in concentrations during the 1990s. The rate of decline has been greatest at urban sites. The measured concentrations of BaP also show significant seasonality. Concentrations are typically greater in winter than in summer by factors of between 1.8 at industrially influenced sites, 4.2 at urban sites and 4.5 at rural sites.

The projected emissions were used to predict concentrations in 2010 at the sites which were operating during 1999 and 2000. This assessment suggested that by 2010 concentrations may be less than 0.25 ng m^{-3} at all sites with the exception of Kinlochleven, Lisburn and Scunthorpe. The projection methodology is conservative and it is probable that with the closure during 2000 of the Kinlochleven smelter the concentrations there will decline more rapidly than predicted.

Maps of predicted UK BaP concentrations in 1999 and 2010 have been prepared. It is expected that the uncertainty in the modelled concentrations will reflect the inherent uncertainty in the emission estimates and the assumptions used in modelling emissions from point sources. Other sources of uncertainty include using 1999 meteorological data to predict concentrations in 2010. The concentration maps suggest that while areas of the country are likely to continue to exceed the 0.25 ng m^{-3} criterion in 2010 the area affected will be significantly smaller than in 1999. The area greater than the EPAQS guideline will decline from 1562 to 551 km^{2}. The area with concentrations greater than 1.0 ng m^{-3} is predicted to decline from 20 to 11 km^{2}. The population weighted annual average concentration is predicted to decline from 0.156 to 0.131 ng m^{-3}.

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Appendices

Abbreviations used

AEAT	AEA Technology plc
BaP	Benzo[*a*]pyrene
DEFRA	Department for Environment, Food and Rural Affairs
DETR	Department of the Environment, Regions and Transport [1]
DTI	Department of Trade and Industry
EPAQS	Expert Panel on Air Quality Standards
IARC	International Agency for Research on Cancer
MSW	Municipal Solid Waste
PAH	Polycyclic Aromatic Hydrocarbon
SSF	Solid Smokeless Fuel
WHO	World Health Organisation
WSL	Warren Spring Laboratory

g	gramme		
kg	kilogramme	=	10^{3} gramme
mg	milligramme	=	10^{-3} gramme
µg	microgramme	=	10^{-6} gramme
ng	nanogramme	(1 ng =	1×10^{-9} g)
pg	picogramme	(1 pg =	1×10^{-12} g)
fg	femtogramme	(1 fg =	1×10^{-15} g)

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[1] The predecessor Government Department with responsibility for Air Quality

1 Introduction

1.1   PURPOSE OF THIS REPORT

This report presents the results of a number of assessments designed to inform the development of a possible National Air Quality Strategy objective for polycyclic aromatic hydrocarbons (PAHs), (DEFRA, 2001). PAH exposure has been associated with a range of cancer endpoints including lung, skin and bladder cancers (EPAQS, 1999). Some evidence supports a link between lung cancer and PAH exposure through inhalation. The Expert Panel on Air Quality Standards has recommended setting a national Standard for PAHs using annual average concentrations of benzo[*a*]pyrene (BaP) as a marker for the human health effects of PAHs.

This report details the approach and outcomes from a range of assessments to improve understanding of the sources, levels, trends and future concentrations of BaP in the UK including:

- Development of updated emission inventories and improved projections of future emissions including the spatial distribution of releases (Chapter 2).

- A review of ambient monitoring results including determination of the seasonal variation and long term trends in pollution levels (Chapter 3).

- Site specific analysis of future BaP concentrations to 2025 using a combination of monitoring, and emissions data and use of dispersion models (Chapter 4).

- Derivation of maps of BaP concentrations in 1999 and 2010 including estimation of population exposure (Chapter 5).

- Conclusions regarding the principal findings (Chapter 6).

1.2   DEVELOPMENTS IN PROPOSED HEALTH-BASED STANDARDS FOR PAHs

The 1995 Environment Act requires the development of a National Air Quality Strategy including standards and objectives for a range of pollutants. The current Strategy (DETR, 2000) was published in January 2000 and the Government has announced it will implement a rolling programme to periodically review and update the standards and objectives included with the Strategy. In 1999, the UK Expert Panel on Air Quality Standards (EPAQS, 1999) issued a report concerning the health effects of PAHs. EPAQS concluded that present evidence supports the use of BaP as a representative marker for the health effects of PAHs and recommended that an annual average concentration for BaP in air as 0.25 ng m^{-3} would make the risk to human health from inhalation of PAHs insignificant. In the 2000 Strategy, the Government announced it was considering whether to set an objective for PAHs in light of the Panels recommendation. This report inputs into the Government's considerations on an Objective for PAHs.

A European Commission working group has been reviewing evidence in order to recommend a limit value for PAHs for inclusion in a future air quality daughter directive. The working group has, like EPAQS, proposed a limit value based on a maximum annual average concentration of BaP as a marker for PAH. The working group has recommended a limit value between 0.5 – 1.0 ng/m^3.

2 Emission Estimates

This section of the report discusses how the current emission inventory was derived, how projections of emissions of BaP up to 2010 were obtained and how the 1999 and 2010 emissions were spatially and temporally distributed to allow dispersion modelling.

2.1   EMISSION TRENDS

The BaP inventory has been updated to incorporate new information regarding emission factors and activity data on sources of BaP. The new inventory is significantly different from the most recently produced national emission inventory, prepared as part of the National Atmospheric Emission Inventory (NAEI) programme (Goodwin et. al. 2000). Total UK emissions in 1999 were about a third lower than those previously estimated.

Table 2.1 shows the updated emissions estimates of BaP since 1990. In 1999, the most recent year for which emissions estimates have been calculated, the main sources of BaP were from domestic coal and wood burning (39%), fires (28%), anode baking (19%) and vehicles (8%). Since 1990, total emissions have declined from 68 t to an estimated 10.3 t in 1999.

The most notable changes to the BaP inventory are:

- Significantly reduced emissions from some industrial sources as a result of new abatement controls and plant closures;

- Smaller road transport emissions.

The updated emission inventory for BaP still contains considerable uncertainty. This is since there has been more limited emissions testing for both vehicles and industrial sources of BaP than for other pollutants included within the Strategy. Many of the significant sources of BaP are also highly dispersed and difficult to quantify accurately. Emissions from the important sources of BaP, particularly fires (accidental, malicious, bonfires and forest fires) and domestic solid fuel burning are especially difficult to estimate.

2.1.1   Road Transport Emission Trends

 The road vehicle emission factors used in the 1999 NAEI were based on a reconciliation of emissions data reported in four main literature sources from the 1980s and early 1990s which showed a fairly large range among vehicles of similar types (Rogge et al (1993), Westerholm (1991), USEPA (1985) and Volkswagen (1989). et al. The measurements were limited to fairly old vehicle models, many being from the U.S. From these sources, average emission factors were estimated for petrol cars with and without a catalyst and for a small diesel truck equivalent to a pre-Euro I model. Data were not available for different types of diesel vehicles (cars, LGVs, large trucks and buses) of different ages (hence Euro standards), so PAH emission factors were estimated by scaling them relative to the value for a rigid HGV according to the way the exhaust emissions of particulate matter (PM) scale. (Goodwin et al, 2000).

Table 2.1 Historic and Projected Emissions of Benzo[a]pyrene

UK Benzo[a]pyrene emissions (kg)	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2005	2010	2015	2020
Road transport (diesel)	618	614	595	579	571	521	486	445	385	352	303	170	91	62	58
Road transport (petrol)	4709	3996	3338	2655	2092	1584	1176	869	642	487	385	168	148	155	162
Fires (natural, agricultural, controlled and accidental)	31197	27141	20539	3254	2880	2880	2880	2880	2880	2880	2880	2880	2880	2880	2880
Creosote use	59	59	59	59	59	59	59	59	59	59	59	57	52	49	46
Aluminium production	1920	1827	1729	1635	1541	1448	1355	1022	316	162	175	239	250	262	273
Anode baking	22705	21842	20980	20118	19256	14833	3850	2897	2730	1930	1667	353	369	386	403
Coke and SSF production	160	149	139	130	132	133	132	133	133	126	125	119	113	108	104
Domestic wood combustion	1170	1170	1170	1170	1170	1170	1170	1170	1169	1169	1225	1509	1702	1920	2165
Industrial wood combustion	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Domestic coal, coke, anthracite and SSF combustion	5149	5330	4822	4635	3323	2203	2385	2261	2403	2878	2514	689	487	354	260
Industrial coal and coke combustion	418	389	292	279	251	213	221	226	131	91	101	151	142	132	124
Other sources	222	226	218	212	201	182	174	161	153	141	143	150	156	160	163
Total	68329	62744	53882	34725	31476	25226	13889	12123	11002	10277	9578	6485	6392	6469	6639

Note : the total may differ slightly from the sum of the individual sectors as a result of rounding in the values displayed

The BaP emission factors used in the 1999 NAEI for different vehicle types and for the different European emission standards which prevailed at the time the vehicle was first registered are shown in Table 2.2.

Changes in the emission estimates for road transport in the updated inventory arise from use of new information from a range of authoritative studies on the concentration of BaP in vehicle exhaust emissions. This has led to the revised emission factors shown in Table 2.2 in comparison with the 1999 NAEI factors.

Table 2.2 BaP emission factors for road vehicles (in μg/km)

Vehicle Type	Emission Class	**1999 NAEI factors**	**Revised factors**
Petrol Cars	Pre-1983	23.0	43.5
	ECE 15.04	23.0	2.38
	Euro I	1.1	0.26
	Euro II	1.1	0.26
Diesel cars	Pre-Euro I	4.5	2.85
	Euro I	1.1	0.63
	Euro II	0.5	0.38
Petrol LGVs	Pre-Euro I	23.0	2.38
	Euro I	1.1	0.26
	Euro II	1.1	0.26
Diesel LGVs	Pre-Euro I	9.5	5.83
	Euro I	3.0	1.80
	Euro II	1.5	1.08
Rigid HGVs	Pre-Euro I	21.9	7.09
	Euro I	14.4	4.06
	Euro II	7.6	1.98
Artic HGVs	Pre-Euro I	31.6	8.72
	Euro I	21.8	5.45
	Euro II	8.4	2.09
Buses & coaches	Pre-Euro I	20.7	11.96
	Euro I	13.8	7.95
	Euro II	8.5	4.53

The high value for pre-1983 petrol cars stems from the work of Rogge et al (1993) which included measurements on old, mainly U.S. cars. The lower value for more recent pre-catalyst cars (equivalent to ECE 15.04 standards) is the mean of emission factors reported in 5 studies

and covers a range from 0.3 to 4.2 µg/km. These are from Westerholm et al (1988), Volkswagen (1989), Westerholm et al (1992), Mi et al (1996) and Koufodimos (1999). The value for cars with three-way catalyst (91/441/EEC (Euro I) standards and beyond) is the mean of emission factors from three studies covering a smaller range from 0.10 to 0.42 µg/km. These are from Volkswagen (1989), Westerholm et al (1992) and Koufodimos (1999). The reduction in the average emission factor for petrol cars with a catalyst implied by these figures is in accord with the report by CONCAWE (1998) which concludes that a car with a three-way catalyst is highly efficient (>80%) in reducing both vapour and particulate phase PAH species. In the absence of any information, emission factors for petrol LGVs are assumed to be the same as for petrol cars.

The revised BaP emission factors for diesel cars are taken from the COPERT III compilation of emission factors for road vehicles published by the European Environment Agency (EEA, 2000). This reports two values for light duty diesel vehicles, one for vehicles with direct fuel injection (DI) and the other for vehicles with indirect fuel injection (IDI). For this work, the higher IDI emission factor was assigned to pre-Euro I diesel cars and the lower DI emission factor assigned to Euro I diesel cars.

COPERT III reports a single BaP emission factor value for all types of heavy duty diesel vehicles with DI engines. The reported value is only slightly higher than the value for DI light duty diesel vehicles. However, particulate matter emissions from heavy duty diesel vehicles are significantly higher than those from light duty diesel vehicles. Therefore, a conservative assumption was made in this study, in which emissions of BaP from diesel LGVs, HGVs and buses were assumed to be higher than emissions from a Euro I diesel car by the same proportion that the particulate emissions (PM_{10}) were higher. This leads to progressively higher emissions with increasing vehicle size. Emission factors for the different Euro standards were also scaled according to the trends in the PM_{10} emission factors.

Insufficient information is available to make a distinction between emission factors over different drive cycles or speeds, so all these factors refer to bulk emission factors.
Use of the revised vehicle emissions factors for the inventory shown in Table 2.1 indicates that vehicles do not represent a significant source of BaP nationally contrary to previous evidence. Monitoring data from the Marylebone Road, kerbside monitoring site in 1998 and 1999 supports the new inventory and shows that levels of BaP were not significantly higher at this kerbside location than at an urban background site in London. These is also no evidence of a significant roadside increment at monitoring undertaken recently in Lille, France (Leoz-Garziandia, 2001). In the early 1990's, a significant roadside increment compared to urban background locations was found in Exhibition Road, London (EPAQS, 1999). This may suggest, as discussed below, that as vehicle emissions have become cleaner and fuel quality improved, BaP emissions from vehicles have significantly declined.

2.2   EMISSIONS PROJECTIONS

Table 2.1 also shows projected future emissions from 2000 to 2010 for a baseline 'implementation of current policy' scenario. By 2010, emissions are projected to decline from 10.3 t in 1999 to 6.4 t. The principal source sectors in which further significant reductions in emissions are forecast are road transport and domestic coal consumption. Increased emissions are projected from the aluminium industry and from domestic wood combustion.

The projections of emissions are based on a common set of activity drivers used throughout the NAEI which are related to DTI forecasts of energy demand. Emissions sectors for which information is not available such as accidental and natural fires have been left constant. These projected activities are combined with emissions factors that incorporate, where necessary, changes arising from the implementation of existing industry commitments under the Integrated Pollution Control (IPC) and Integrated Pollution Prevention and Control (IPPC) regimes.

The projections of future emissions of BaP are subject to considerable uncertainty. This uncertainty arises from three sources;
- The uncertainty in the base inventory.
- The uncertainty in changes in activity within industrial sectors.
- The uncertainty introduced from possible changes in process.

The scale of activity within particular industrial sectors is driven by macroeconomic factors which may not follow the long-term pattern for the UK economy e.g. an increase in the price of aluminium has led to production from the Lynemouth smelter doubling in recent years. This can lead to step changes in production volumes in either direction whereas the emission projections assume a continuous change based on projected energy demand. Emissions of BaP are principally associated with industries with heavy energy costs, changes in the price of energy can rapidly alter energy demand. Emission factors will probably be subject to less uncertainty as large capital investments take time to be implemented and are normally signalled in advance through plant improvement programmes agreed between the relevant regulator and the operator. However new processes that lead to significant changes in emission can be imagined on the 20 year timescale of the projections. Hence while the historic emission estimates for BaP are subject to significant uncertainty, the projections of emissions are subject to a large uncertainty the size of which will increase with time from the present.

2.2.1   Road Transport Emission projections

Emission projections for road transport are based on the same assumptions about traffic growth and changes in the vehicle fleet as are used for the base emission projections for the other pollutants. Traffic is assumed to grow at rates defined in the Ten Year Plan for Transport 'Plan' scenario for England, the Central Scotland Transport Model (CSTM3) and the 1997 National Road Traffic Forecasts.

PAH emissions are not subject to regulation by the European Directives on vehicle emissions that will tighten Type-Approval limit values on emissions of other pollutants from new vehicles in the next 5 years. Nevertheless, BaP emissions might be expected to fall as a result of new technologies aimed at reducing the regulated pollutant emissions, especially particulates from diesel vehicles. The CONCAWE (1998) report reviews the effects of a number of exhaust after-treatment measures on particulate-phase PAH emissions, including diesel oxidation catalysts and particulate traps. They tend to indicate that the effectiveness of these measures on particulate-PAH abatement from diesel vehicles is similar to their effectiveness on total particulate mass. BaP emission factors for future diesel vehicles meeting the tighter Euro III and Euro IV standards are therefore assumed to be reduced by the same amount as their PM emission factors. Appropriate scaling factors for PM emissions from Euro III (2001) and Euro IV (2006) diesel vehicles relative to emissions from Euro II vehicles are used for the BaP projections, taken from Murrells (2000). These are summarised in Table 2.3.

Table 2.3 Scaling factors for BaP emissions from Euro III and Euro IV diesel vehicles relative to emissions from Euro II vehicles.

	Directive	Standard	Date of implementation	BaP emission scaling factor
Light duty diesel	98/69/EC	Euro III	1 January 2001	0.6
		Euro IV	1 January 2006	0.37
Heavy duty diesel	1999/96/EC	Euro III	1 October 2001	0.72
		Euro IV	1 October 2006	0.15

Like particulates, emissions of BaP may also be expected to be reduced by changes in fuel formulation. Reductions in the sulphur content of diesel reduce emissions of PM, but are not likely to affect emissions of BaP. BaP emissions are more likely to be influenced by changes in the polyaromatic content of the fuel. However, the current polyaromatic content of diesel fuels in the UK is already within the limits set by future European fuel directives, therefore significant changes in fuels of the future may not be expected. Consequently, the projections assume that BaP emissions will not be affected by anticipated changes in fuel formulation.

2.3 SPATIAL DISTRIBUTION OF UK EMISSIONS

In order to map concentrations of BaP, it is necessary to distribute the UK national total emissions spatially. This was achieved by allocating each source category to one of 32 surrogate statistics. The situation is further complicated by the need for modelling purposes to split emission sources into categories of either area or point sources to allow for differences in discharge height. To model UK concentrations it is also important to consider the temporal variation in emissions and this is considered in Section 2.4.

The spatial distribution of each separately modelled category in 2010 was based on the 1999 distribution. The individual categories are then added together to give the maps shown in Figures 2.1 and 2.2 for 1999 and 2010 respectively.

2.3.1 Area Emission Sources

Table 2.4 shows the emissions allocated to the source descriptions as modelled. Tables 2.5 and 2.6 provide a further breakdown of the "Light industrial activity" and "Area other" source sectors.

Table 2.4 Breakdown of BaP emissions from area sources

Area source sector	1999 Emission (kg)
Roads[1]	840
Light industrial activity	98
Domestic	4047
Area other	2916
Total	7900

Notes:
1. Emission from roads include petrol, diesel and tyre wear

Table 2.5 Breakdown of BaP emissions from Light Industrial Activity

Source	1999 Emission (kg)
MSW (power generation)	1.7
SSF Coke	3.6
Collieries	7.8
Creosote	59.2
Off road vehicles	19.0
Unattributed area source	6.3

Table 2.6 Breakdown of BaP emissions from "Area Other" Sources

Source	1999 Emission (kg)
Miscellaneous	14.0
Public Services	0.7
Agriculture (stationary sources)	6.5
Agriculture (mobile sources)	5.7
Domestic House and Garden	4.4
Aircraft support	0.0
Shipping	4.1
Incinerators	0.4
Fires[1]	2880

Note

1. The emission from the fire category have been assumed to arise from urban and natural fires. 50% from natural fires and 50 % from urban fires. Section 5.6.1 contains a sensitivity analysis investigating the appropriate spatial distribution.

2.3.2   Point Emission Sources

In 1999, emissions of BaP from industrial processes such as aluminium production, metallurgical coke and smokeless fuel production, were estimated to be 2366 kg. Table 2.7 provides a breakdown in the amount of BaP emission from each of the major source sectors in the United Kingdom. Anode baking, coke production and aluminium production were the largest point source emitters.

Emissions from large industrial plant such as refineries, cement plant, power stations and the processes at steel plant have not been modelled. An initial scoping study established that these were small sources of BaP and that with effective dispersion from their relatively tall stacks, they made an insignificant contribution to annual average ambient BaP concentrations. In consequence emission from these point sources were excluded from the modelling assessment but included within their contribution to the assumed background concentration. Table 2.7 shows that there are differences between the total emission from all sectors and those sources that were specifically modelled of only 124 kg out of a total of 10.3 t of BaP released in 1999.

Table 2.8 provides the estimated BaP emission for those industrial processes for which releases were separately modelled (as described in Section 4). Emission estimates for each plant were obtained from the Pollution Inventory, the National Atmospheric Emission Inventory or assessment of abatement costs study carried out on behalf of DTI (ENTEC 2001).

Table 2.7 Emission Estimates for 1999 from Industrial Processes in the United Kingdom

	Total emission from sector (kg)	Emissions specifically modelled (kg)	Number of sources	Number of sources modelled
Coking plant	122	122	8	7
Other industry combustion	18.09	0.48	178	
Power generation	3		20	
Refineries	46		12	
Chemical industry	38	32	4	
Aluminium production	162	158	4	3
Cement manufacture	12		25	
Anode baking	1930	1930	2	2
Steel production (processes)[1]	35		4	
Crematoria	0		240	
Total	2366	2242	495	12

Note:
1. Processes from the iron and steel sector include sinter plant, blast furnace and basic oxygen furnace.

Table 2.8 BaP Emission Estimates for 1999 from Specific Industrial Point Sources

Process	Industrial Source Modelled	Emission Modelled (kg)
Aluminium Production	Alcan Aluminium Smelter (Ashington, England)	46
	Anglesey Aluminium Metal Ltd (Holyhead, Wales)	1907
	Alcan Plant (Kinlochleven, Scotland)	135
Coking Plant	Corus (Llanwern, South Wales)	12
	Corus (Port Talbot, South Wales)	19
	Corus (Scunthorpe)	21
	Corus (Middlesbrough)	28
	Cwm Coking Works (South Wales)	10
	Monckton Coke Works (Royston, near Barnsley)	32
Solid fuel production	Coalite Coking Plant (Derbyshire)	0.4

1999 BaP kg / 1x1km
- 3e-008 - 0.0006
- 0.0006 - 0.002
- 0.002 - 0.01
- 0.01 - 0.03
- 0.03 - 0.06
- 0.06 - 0.3
- 0.3 - 1910
- No Data
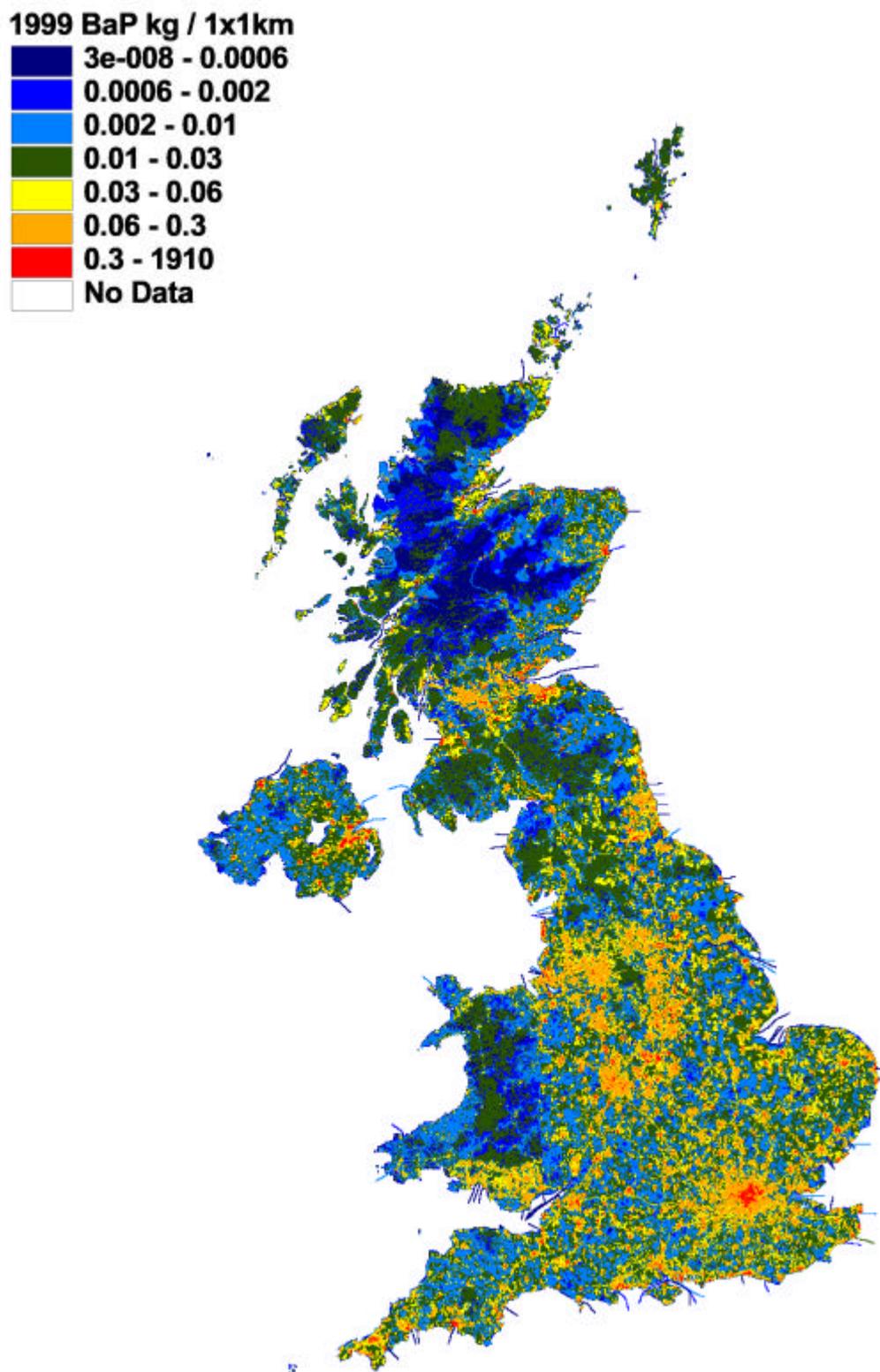
Figure 2.1 Projected Emissions of Benzo[a]pyrene in the UK in 1999
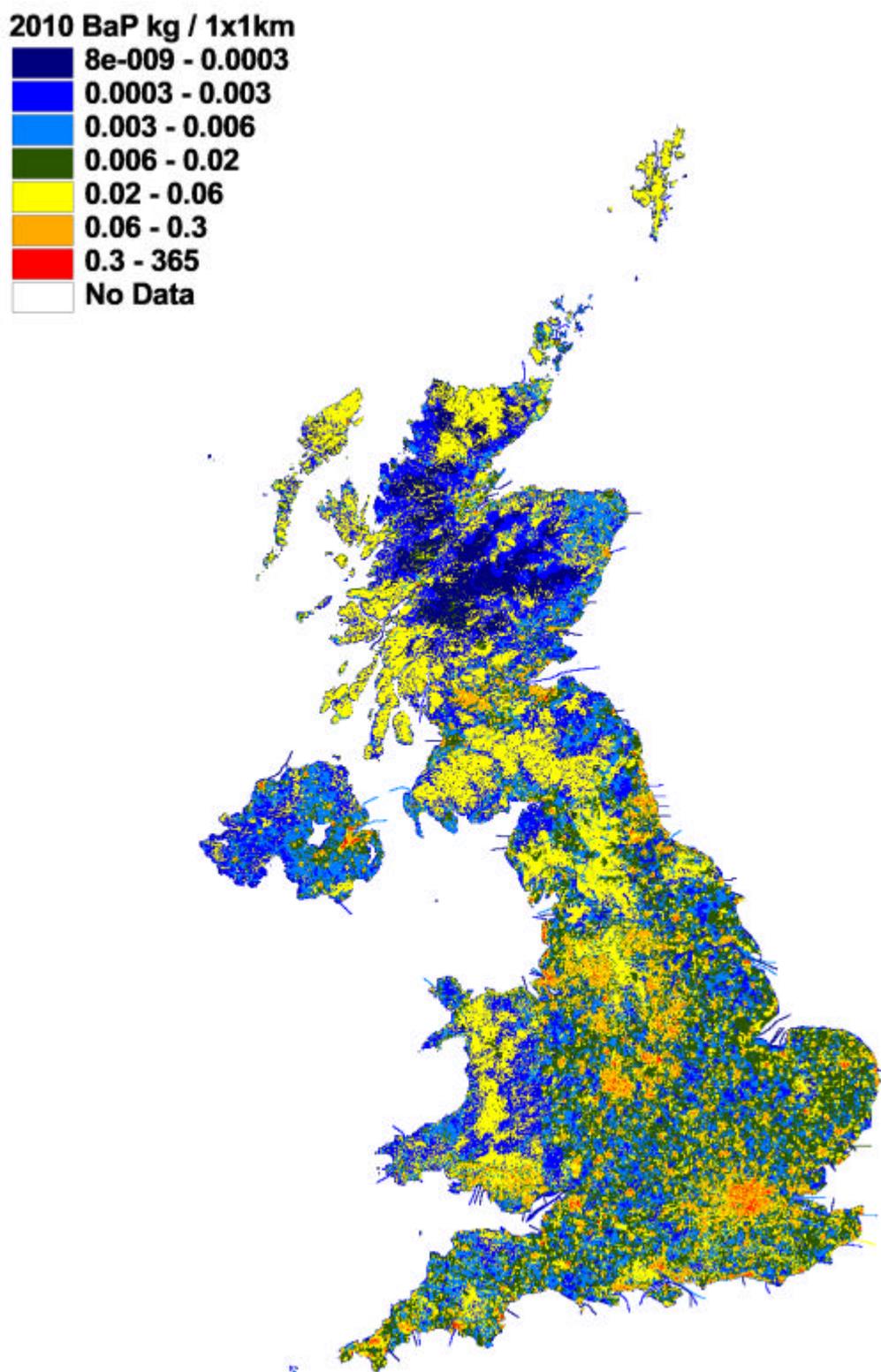
Figure 2.2 Projected Emissions of Benzo[a]pyrene in the UK in 2010

2.4 TEMPORAL PROFILES OF EMISSION SOURCES

The temporal variation in emissions should be considered in order to model robustly current and future concentrations of BaP as dispersion conditions vary both seasonally and diurnally. Hence it is important to understand not just the quantity of BaP released from a source sector but when the pollution is released as this will affect the resulting modelled concentration at ground level. Road transport emissions were modelled using a diurnal pattern of traffic derived from road transport statistics. Domestic emissions were given temporal profiles which included both a diurnal variation and a seasonal dependence of emission release that takes into account higher emissions in the winter time than in summer. In the absence of more detailed information, emissions from the point sources and the "industrial area" and "area other" sectors were assumed to have a constant emission rate.

2.4.1 Road transport profile

The diurnal patterns of traffic flows within the district were taken from Road Traffic Statistics (DETR, 1999). The normalised flows are presented in Figure 2.3. They show the pronounced morning and evening week day rush hours. Typically the dispersion conditions are less effective in the morning than during the evening. The figure also shows the pattern of traffic at weekends.

Figure 2.3   Diurnal traffic flows used in the modelling study
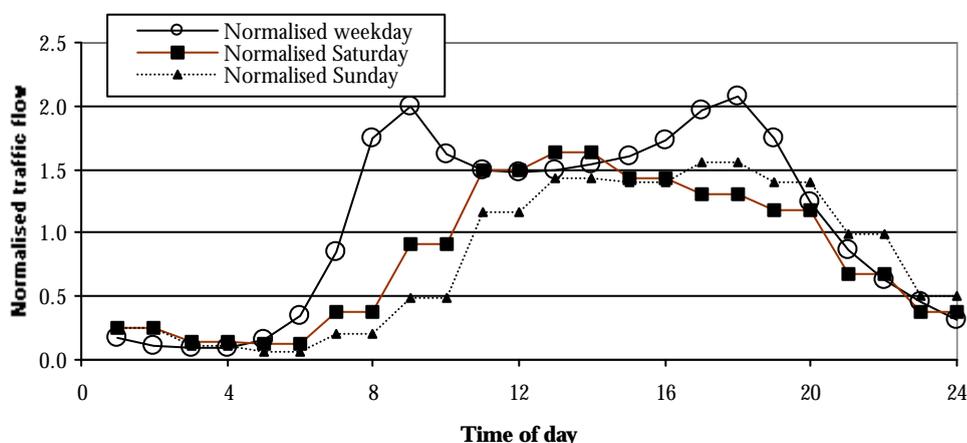
2.4.2 Domestic profile

Emissions from the domestic emission source were weighted with both seasonal and diurnal emission patterns. The seasonal emission pattern was obtained from the Building Research Establishment Domestic Energy Model (BREDEM) (BRE, 1985). The pattern was derived using formulae that allow a degree day to be calculated. The degree day provides a method to weight emissions to the colder periods of the year. Figure 4.4 shows the seasonal profile derived from the BREDEM model for the meteorological station at Waddington in 1999. A seasonal profile for modelling domestic emissions in Great Britain were derived from the Waddington meteorological data; for Northern Ireland 1999 Aldegrove data was used.

A diurnal emission pattern was superimposed on top of each of the degree day emission weightings. This emission pattern was obtained from a diurnal pattern used in modelling sulphur dioxide emissions in Belfast (CERC, 1998). The emission profile is presented in Figure 2.5. The application of temporal emissions data for modelling the spatial distribution and future levels of BaP concentrations is discussed in Sections 4 and 5.

Figure 2.4 Degree days calculated for each day at the Waddington sampling site in 1999 (the base temperature used was 15.5 °C)
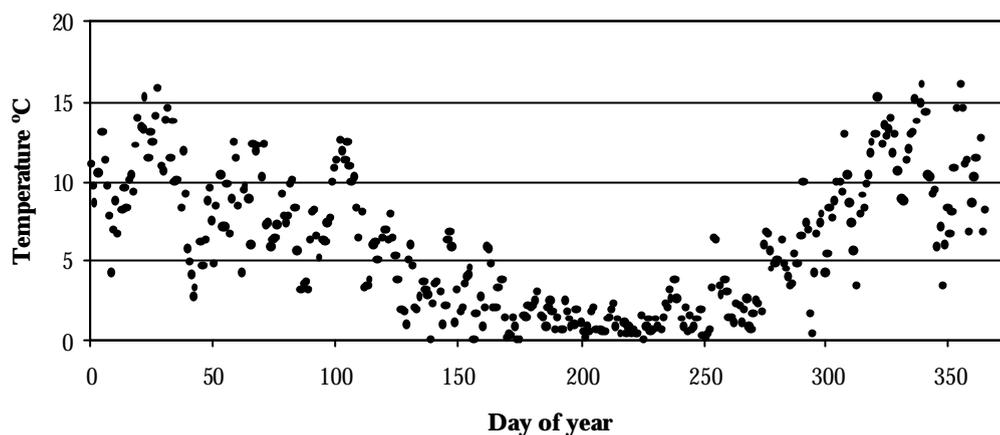
Figure 2.5 Diurnal emission profile for domestic emissions
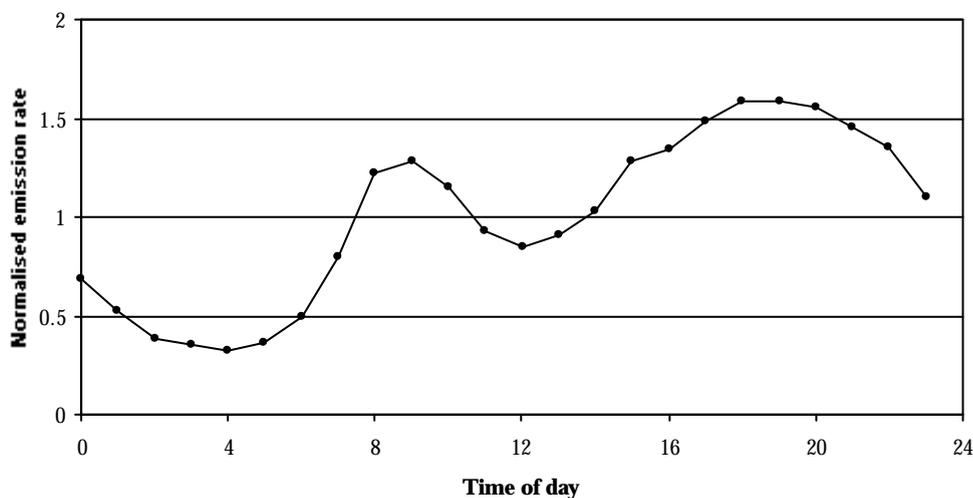
3 Concentrations and trends in BaP

This section discusses the monitoring networks and measurements of BaP in the UK. It also provides a statistical analysis of the trends in BaP concentrations measured in ambient air samples.

3.1   MONITORING SITES

DEFRA operates two national monitoring networks measuring BaP:
- TOMPS Network (Total Organic Micro-pollutants Network)
- PAH Network.

The original network of TOMPs sites (Hazelrigg, London, Manchester and Middlesbrough) was established in the early 1990's with the objective of providing information on the long term trends in ambient concentrations of dioxins, PCBs and PAHs. Two new sites (High Muffles and Stoke Ferry) were added in 1996 to investigate more fully the concentrations found in rural locations.

The PAH Network is a more recent development designed to improve knowledge of the levels of PAHs throughout the UK. The PAH Network currently comprises of fifteen further monitoring sites, some near industrial sites of types that were identified as significant sources of PAHs. Other sites are located in an area of high domestic solid fuel use (Lisburn) at a roadside site (Bromley) and others in major urban areas (Belfast, Birmingham, Glasgow, Leeds, Liverpool, Newcastle). Samplers were installed near the two aluminium anode baking works in the UK (Holyhead and Ashington), the only aluminium smelter using the Soderberg process (Kinlochleven)[2], three of the large metallurgical coke works (Newport[3], Port Talbot and Scunthorpe) and one of the three smaller carbonisation works (Bolsover). These sites were installed and commissioned between December 1998 and March 2001. A further four monitoring sites will be installed during summer 2001 in the London Borough of Brent, Brighton, Cardiff and Edinburgh. Information on the PAH monitoring sites is given in Table 3.1.

3.2   SAMPLING AND ANALYTICAL METHODS

Details of the sampling and analytical techniques used have been given in previous reports (AEAT, 1999) and can be found on the DEFRA air quality website (http://www.aeat.co.uk/netcen/airqual /reports/home.html).
Samples are collected using polyurethane foam plugs and filter papers to capture PAHs in the volatile and non-volatile phases. The fortnightly samples are extracted using dichloromethane and then bulked to give a single sample. The concentrations of 32 individual PAHs, including BaP, are determined from the quarterly samples using gas chromatography with mass spectral detection.

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[2] The aluminium smelting operations at Kinlochleven were stopped in June 2000.
[3] The coke works at Newport closed in June 2001.

Table 3.1 Details of the Sites where PAHs are measured

Site name	Network	Site type	Monitoring	Location	OS Grid Reference	Latitude	Longitude
Ashington	PAH	urban industrial	February 1999 -	Roof of council buildings in town centre	NZ 272 878	55º 11' 1" N	1º 34' 22" W
Belfast	PAH	urban	January 2001 -	Clara Street Depot	J 362 735	54º 35' 28" N	5º 53' 36" W
Birmingham	PAH	urban	January 2001 -	Ward End School	SP116 889	52º 29' 51" N	1º 49' 45" W
Bolsover	PAH	industrial	February 1999 -	Ground level on the edge of housing near Bolsover	SK 469 726	53º 14' 53" N	1º 17' 49" W
Bromley	PAH	roadside	January 2001 -	Roadside adjacent to Crystal Palace Park	TQ 334 713	51º 25' 28" N	0º 4' 26" W
Cardiff	TOMPs	urban	December 1990 – March 1994	Roof of the University of Cardiff	ST 183 772	51º 29' 14" N	3º 10' 42" W
Glasgow	PAH	urban	May 1999 -	Roof of the City Hall, George Street	NS 595 653	55º 51' 37" N	4º 14' 42" W
Hazelrigg	TOMPs	semi-rural	September 1992 -	University of Lancaster's environmental field station	SD 493 579	54º 0' 49" N	2º 46' 26" W
High Muffles	TOMPs	rural	January 1997 -	Ground level on Forestry Commission land in a remote location in the North York Moors	SE 776 939	54º 20' 5" N	0º 48' 23" W
Holyhead	PAH	urban industrial	February 1999 -	On police station roof in centre of Holyhead	SH 246 827	53º 18' 43" N	4º 37' 59" W
Kinlochleven (1)	PAH	urban industrial	February 1999 – June 2000	On Social Club roof near Aluminium smelter	NN 186 620	56º 42' 52" N	4º 57' 51" W
Kinlochleven (2)	PAH	urban industrial	June 2000-	on edge of village near former Aluminium smelter	NN 193 619	56º 42' 50" N	4º 57' 10" W
Leeds	PAH	urban	January 2001 -	Potternewton Environmental Health Office	SE 307 366	53º 49' 28" N	1º 32' 1" W
Lisburn	PAH	urban	May 1999 -	Dunmurray High School	J 286 675	54º 32' 22" N	6º 0' 46" W
Liverpool	PAH	urban	March 2001 -	St Christopher's School	SJ438 835	53º 20' 42" N	2º 50' 39" W

Site name	Network	Site type	Monitoring	Location	OS Grid Reference	Latitude	Longitude
London (1)	TOMPs	urban	January 1991 – March 1998	Roof of DETR Romney House	TQ 300 791	51º 29' 43" N	0º 7' 37" W
London (2)	TOMPs	urban	September 1998 -	Roof of DEFRA, Ashdown House	TQ 292 792	51º 29' 47" N	0º 8' 18" W
Manchester	TOMPs	urban	January 1990 -	Roof of the Law Courts in central Manchester	SJ 834 982	53º 28' 48" N	2º 15' 1" W
Middlesbrough	TOMPs	urban	May 1992 -	Roof of Longlands College in the town centre	NZ 505 194	54º 34' 2" N	1º 13' 8" W
Newcastle on Tyne	PAH	urban	January 2001 -	Newcastle Civic Centre	NZ251 649	54º 58' 41" N	1º 36' 28" W
Newport	PAH	urban industrial	February 1999 -	School roof near Llanwern coke works	ST 355 880	51º 35' 12" N	2º 55' 52" W
Port Talbot	PAH	urban industrial	February 1999 -	Ground level in hospital grounds near coke works	SS 780 882	51º 34' 44" N	3º 45' 39" W
Scunthorpe	PAH	urban industrial	February 1999 -	Roof of council depot near steel plant	SE 905 107	53º 35' 5" N	0º 37' 58" W
Stevenage	TOMPs	urban	January 1990 – April 92	Roof of Warren Spring Laboratory	TL 237 224	51º 53' 9" N	0º 12' 9" W
Stoke Ferry	TOMPs	rural	January 1997 -	Ground level at a rural water treatment plant near Kings Lynn	TL 700 988	52º 33' 36" N	0º 30' 29" E

Notes:
- CC   City Council
- BC   Borough Council

3.3   ANNUAL-AVERAGED CONCENTRATIONS

Figure 3.1 shows the annual-averaged concentrations of BaP at all the sites where measurements have been made. The concentrations are presented on a logarithmic scale. Table 3.2 contains the annual mean concentrations of BaP measured in 1999 and 2000.

A visual inspection of the data at the longer-running sites suggest that concentrations of BaP measured in urban and rural areas are, in general, declining as emissions from area sources such as traffic have declined. A year-on-year decline is not always apparent, and variations in the annual mean will be caused by changes in the strengths of the sources of BaP and the prevailing meteorology.

A small number (less than 5% of the total) of the measured quarterly concentrations from the PAH network were excluded from the quarterly series of data because of quality control reasons. In these cases, the quarterly concentration has been interpolated from the concentrations in the preceding and succeeding quarters. Where concentrations of BaP were below the detection limit, the concentration has assumed to be equal to the detection limit, which results in a conservative estimate of the concentration. A duplicate sampler was added to the site at Ashdown House in London from the beginning of 2000. The BaP concentrations reported for 2000 and thereafter are the averages of the two measurements.

Table 3.2 Annual-averaged Concentrations of Benzo[a]pyrene in 1999 and 2000

Site	(site type)	1999 Annual Mean Concentration ($ng\ m^{-3}$)	2000 Annual Mean Concentration ($ng\ m^{-3}$)
Ashington	(Urb-Ind)	0.20	0.17
Holyhead	(Urb-Ind)	0.11	0.11
Kinlochleven	(Urb-Ind)	6.78	2.28
Newport	(Urb-Ind)	0.23	0.35
Port Talbot	(Urb-Ind)	0.24	0.59
Scunthorpe	(Urb-Ind)	0.37	1.17
Bolsover	(Ind)	0.24	0.25
Glasgow	(Urb)	0.19	0.12
Lisburn	(Urb)	0.74	0.93
London	(Urb)	0.34	0.13
Manchester	(Urb)	0.15	0.24
Middlesbrough	(Urb)	0.24	0.28
Hazelrigg	(Semi-Rur)	0.06	0.06
High Muffles	(Rur)	0.06	0.04
Stoke Ferry	(Rur)	0.11	0.09

Notes:

Urb-Ind	Urban Industrial	Semi-Rur	Semi rural
Ind	Industrial	Rur	Rural
Urb	Urban		
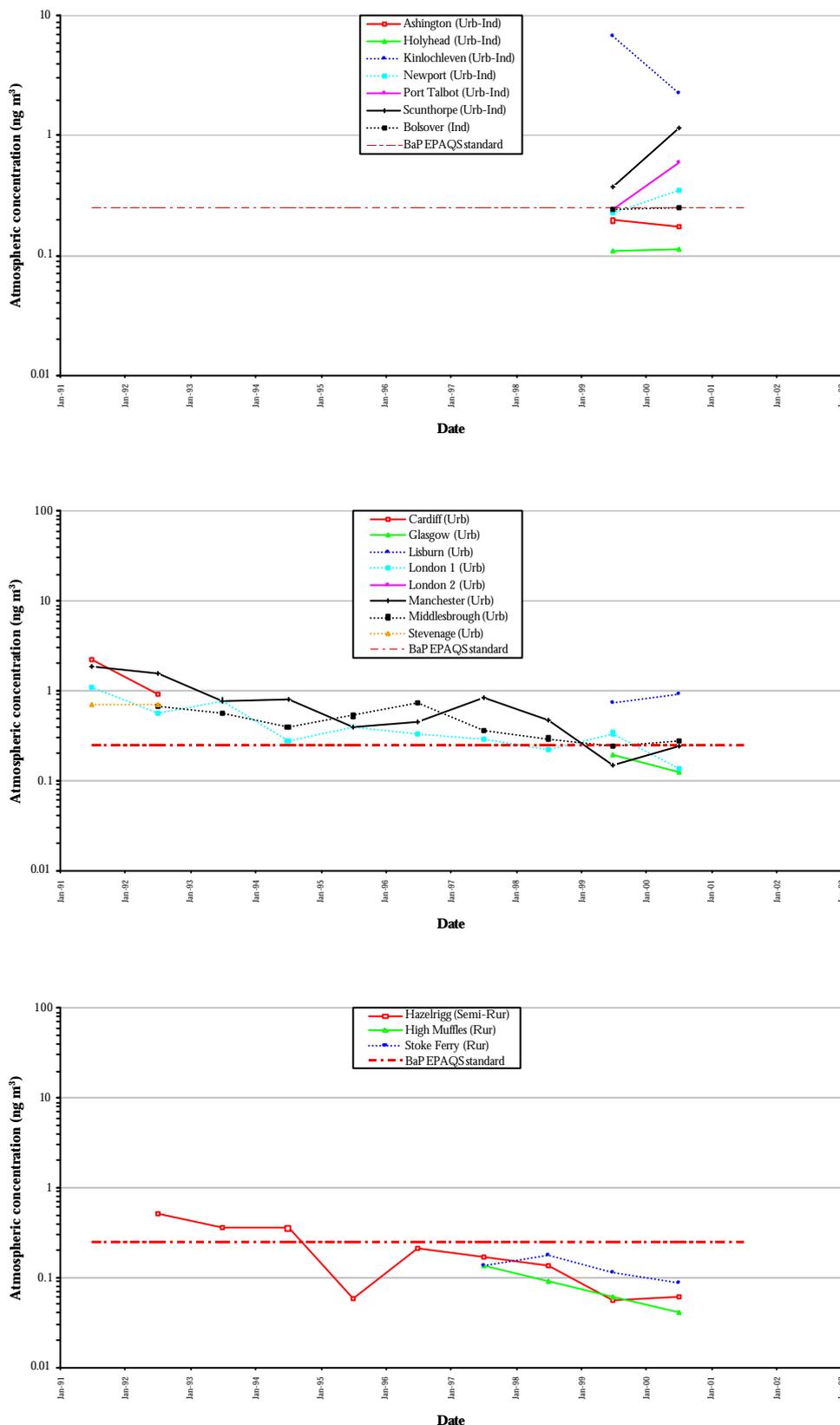
Figure 3.1 Mean Annual Concentrations of Benzo[a]pyrene at UK PAH Monitoring Sites (Note logarithmic scales)

3.4 COMPARISON OF RECENT ANNUAL AVERAGE CONCENTRATIONS OF BAP TO THE PROPOSED EPAQS STANDARDS

The UK Expert Panel on Air Quality Standards has (EPAQS, 1999) has recommended that an annually-averaged concentration of BaP in air as 0.25 ng m^{-3} would make the risk to human health from inhalation of PAHs insignificant.

The proposed EPAQS standard is also shown in Figure 3.1. In 2000, annual mean concentrations of BaP were equal to or were greater than 0.25 ng m^{-3} at seven of the urban-industrial, industrial and urban sites shown in Table 3.1. The annual mean concentrations of BaP at Kinlochleven (now closed), Lisburn (a coal burning area) and Scunthorpe (close to a major industrial source) were significantly greater than the proposed EPAQS standard. There were no exceedences of the proposed EPAQS recommended air quality standard at rural locations. Table 3.3 presents the measurements as a ratio to the proposed EPAQS standard.

Table 3.3   Ratio of benzo[a]pyrene concentrations in 1999 and 2000 to the proposed EPAQS BaP standard of 0.25 ng m^{-3}

Site	(site type)	Ratio of concentration to the EPAQS standard (1999)	Ratio of concentration to the EPAQS standard (2000)
Ashington	(Urb-Ind)	0.8	0.7
Holyhead	(Urb-Ind)	0.4	0.5
Kinlochleven	(Urb-Ind)	**27**	**9.1**
Newport	(Urb-Ind)	0.9	**1.4**
Port Talbot	(Urb-Ind)	**1.0**	**2.3**
Scunthorpe	(Urb-Ind)	**1.5**	**4.7**
Bolsover	(Ind)	**1.0**	**1.0**
Glasgow	(Urb)	0.8	0.5
Lisburn	(Urb)	**2.9**	**3.7**
London	(Urb)	**1.3**	0.5
Manchester	(Urb)	0.6	**1.0**
Middlesbrough	(Urb)	**1.0**	**1.1**
Hazelrigg	(Semi-Rur)	0.2	0.2
High Muffles	(Rur)	0.2	0.2
Stoke Ferry	(Rur)	0.5	0.3

Notes:

Urb-Ind	Urban Industrial	Semi-Rur	Semi rural
Ind	Industrial	Rur	Rural
Urb	Urban		

Values greater than or equal to one in **bold**

3.5 LONG TERM TRENDS - STATISTICAL ANALYSIS OF CHANGES IN BAP CONCENTRATIONS WITH TIME

Long-term trends in pollution levels were assessed following the approach used in previous reports (AEAT, 1999). This involved use of the Seasonal Kendall Test (Hirsch *et al.*, 1982). This is a non-parametric test specially tailored to assess seasonally variable data. The test is a development of the standard Mann-Kendall test to determine the significance of the trend (assuming a linear decline in atmospheric concentrations with time). The test has been applied to the quarterly air concentrations of BaP at:

- London (Urban)
- Manchester (Urban)
- Middlesbrough (Urban)
- Hazelrigg (Semi-Rural)
- High Muffles (Rural)
- Stoke Ferry (Rural)

There were sufficient measurement data at these sites for the statistical analysis.

The estimated slope calculated was used to determine the extent of any trend, together with a method that produces estimates of the confidence levels on the result (Gilbert, 1987). Although originally formulated in terms of monthly values, the test was amended slightly to deal with the quarterly results that are available. The tools were implemented partly by using a Fortran77 code given in the literature (Smith *et al.*, 1982). The results of the test are summarised in Table 3.4.

Table 3.4 Results of the Seasonal Kendall statistical test assuming a linear decline in concentrations of benzo[a]pyrene with time

Site	Number of years annual mean data	Seasonal Kendall Test (τ)	Significance level (α)	Slope (ng/m^3 per 3 months)	95% confidence on slope (upper)	95% confidence on slope (lower)
London (Urb)	10	-0.467	0.0004	-0.0358	-0.0800	-0.0200
Manchester (Urb)	10	-0.561	0.0004	-0.0660	-0.1500	-0.0325
Middlesbrough (Urb)	9	-0.302	0.030	-0.0334	-0.070	-0.0050
Hazelrigg (Semi-Rur)	9	-0.308	0.034	-0.0100	-0.0400	0[†]
High Muffles (Rur)	4	-0.667	0.009	-0.0200	-0.0600	-0.0050
Stoke Ferry (Rur)	4	-0.458	0.063	-0.0050	-0.0700	0[†]

Notes:

τ   is the Seasonal Kendall Test statistic (-1 for all negative differences between season-specific data values, +1 for all positive differences)

α   is the significance level (two-tailed) on the value of τ representing the probability that the true slope is actually zero

†   as a result of the non-parametric nature of the test a slope of 0 occurs with a range of probabilities which includes the 95th percentile.

3.6   DATA USED IN THE ANALYSES

A small number (less than 5% of the total) of the measured quarterly concentrations from the PAH network were excluded from the quarterly series of data because of quality control reasons. In these cases, the quarterly concentration has been interpolated from the concentrations in the preceding and succeeding quarters. Where concentrations of BaP were below the detection limit, the concentration has assumed to be equal to the detection limit, which results in a conservative estimate of the concentration. The concentrations in the second and third quarter concentrations in 1997 at Middlesbrough were received as a single value, and so the concentrations in those two quarters have been assumed equal.

A duplicate sampler was added to the site at Ashdown House in London from the beginning of 2000. The BaP concentrations reported for Quarter 1 2000 and thereafter are the averages of the two measurements.

Concentrations have been used from the start of the measurements at each site up to and including concentrations recorded in the last quarter of 2000.

3.6.1   Significance of declines in concentrations

In Table 3.4, α is the significance level (two-tailed) on the value of τ representing the probability that the true slope, fitted through the atmospheric concentrations data, is actually zero. The larger the value of α, the greater the chance that the slope of the line is actually zero; in other words, small values of α indicate that there is a significant change in atmospheric concentration with time (increase or decline).

The values of α for all the sites are small and this suggests that there has been a significant change in atmospheric concentrations of BaP with time. The slopes of the fitted linear models are all negative, and this indicates that there has been a decline in concentrations of BaP with time.

3.6.2   Rate of decline in concentrations

The slope in Table 3.4 represents the rate of decline of the concentrations of BaP. The units of the slope are ng/m^3 per 3 months (or per quarter). The greatest rate of decline in BaP concentrations has occurred at Manchester, with smaller but similar rates of reductions in London and Middlesbrough. The rate of decline of BaP at the rural and semi-rural sites is smaller than at the urban sites, as the air mass sampled at these site is little affected by the reductions in BaP concentrations seen in the urban environments. The 95% confidence on the slopes is also given in Table 3.4.

3.7   SEASONALITY

Figure 3.2 shows the quarterly mean concentrations of BaP at all the sites where measurements of have been made. There is a considerable variation in the atmospheric concentrations of BaP, which vary by at least an order of magnitude according to time and between measurement sites.

The time series plots of the quarterly concentrations of individual PAHs suggested the atmospheric concentrations were seasonal. To examine this seasonality, mean quarterly concentrations of the BaP were calculated over the measurement periods and are shown, according to site type, in Figure 3.3.

All the available measurement data were used to calculate these ratios, and it is important to note that the number of quarterly measurements included in the calculations of each overall quarterly mean (Quarters 1, 2, 3 and 4) were different at the measurement sites. However, this simple analysis still allows the seasonality in the data to be revealed clearly. Table 3.5 shows the ratios of the mean 'winter' to 'summer' atmospheric concentrations of BaP at all the measurement sites (current and historic).

Nearly all the ratios are greater than unity, indicating that concentrations of BaP are raised during the 'winter' months. The differences in the concentrations in 'summer' and 'winter' are controlled by changes in the strengths of the sources of BaP and the prevailing meteorology. During the winter months, meteorological conditions (such as high-pressure systems) may restrict the mixing and dilution of released BaP, and concentrations of BaP may rise. The results indicate that at the rural and urban sites which typically have higher winter to summer ratios the concentrations are affected by a more altering seasonal source than at the industrially dominated sites.
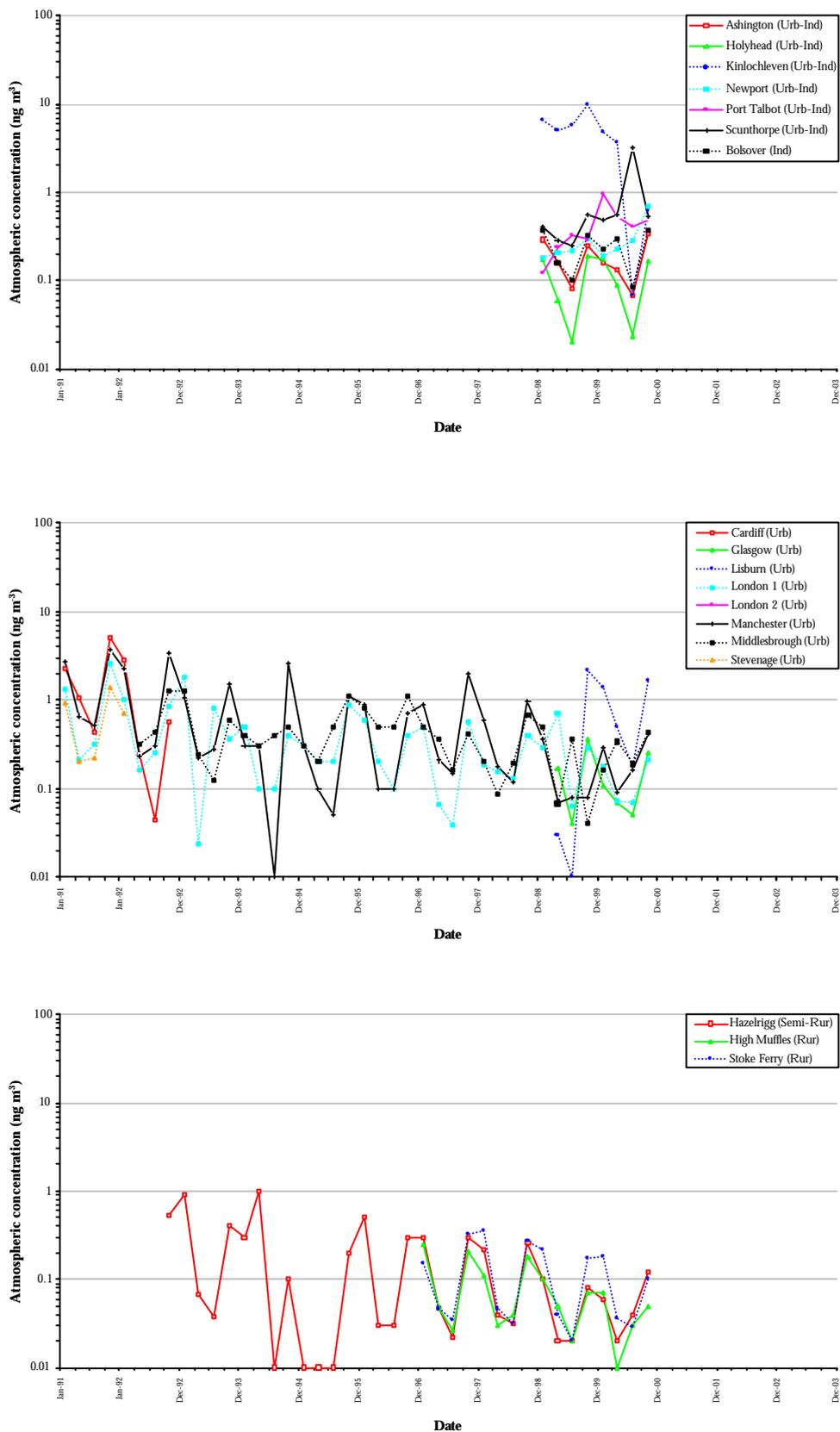
Figure 3.2 Mean Quarterly Concentrations of Benzo[a]pyrene Measured at UK PAH Monitoring Sites (Note logarithmic scales)
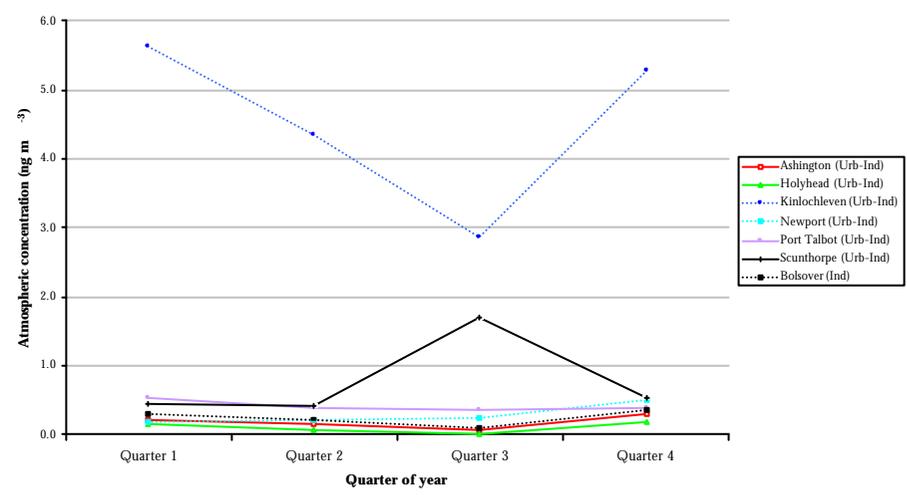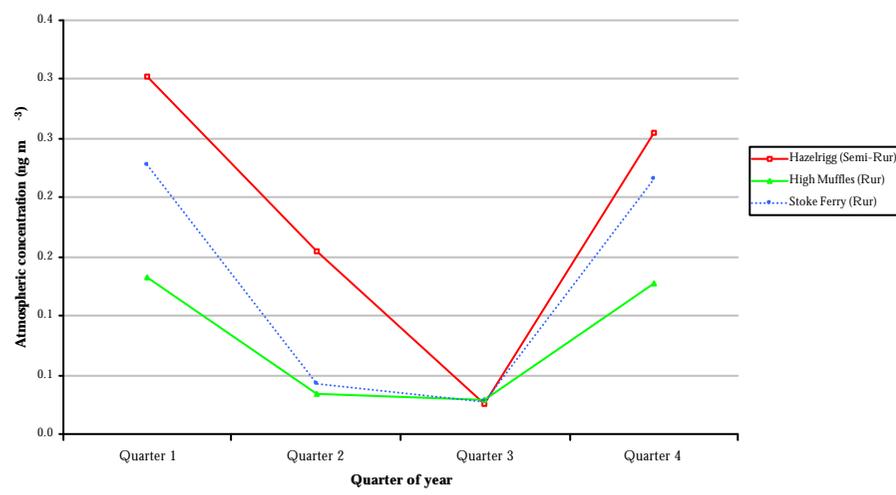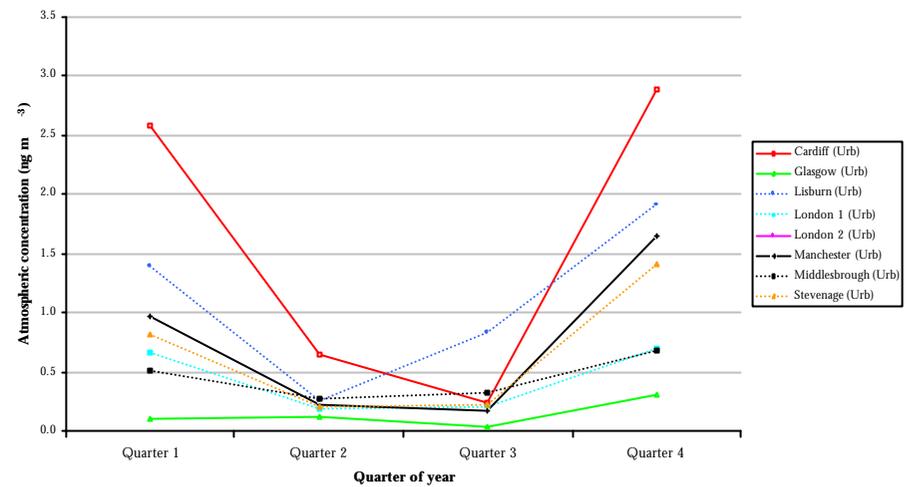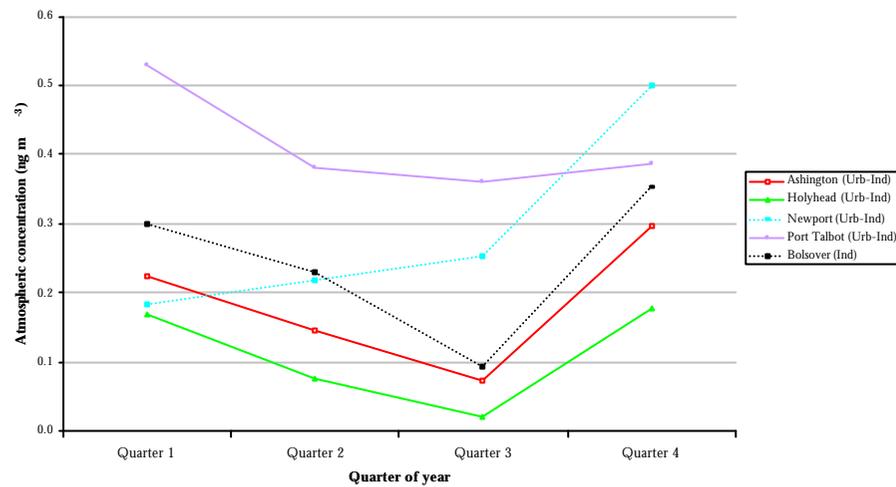
Figure 3.3 Period Means of the Quarterly Concentrations of Benzo[a]pyrene Measured at UK PAH Monitoring Sites

**Table 3.5 Ratio of mean 'winter' to 'summer'
atmospheric concentrations of benzo[a]pyrene**

Site	Site type	Mean 'winter' to 'summer' ratio
Ashington	urban industrial	2.4
Holyhead	urban industrial	3.6
Kinlochleven	urban industrial	1.5
Newport	urban industrial	1.4
Port Talbot	urban industrial	1.2
Scunthorpe	urban industrial	0.5
Bolsover	industrial	2.0
	Mean	**1.8**
Cardiff	urban	6.2
Glasgow	urban	2.6
Lisburn	urban	3.0
London	urban	3.4
Manchester	urban	6.7
Middlesbrough	urban	2.0
Stevenage	urban	5.2
	Mean	**4.2**
Hazelrigg	semi-rural	3.1
High Muffles	rural	4.1
Stoke Ferry	rural	6.3
	Mean	**4.5**

Notes:

- 'winter' is defined as Quarter 1 (January to March) and Quarter 4 (October to December)
- 'summer' is defined as Quarter 2 (April to June) Quarter 3 (July to September)
- All available data used to calculate the means, so the number of periods used for each site differ

4 Site-specific projections of BaP concentrations

This section describes the approach used to project forward concentrations of BaP at existing monitoring stations to 2025 and provides an indication of the likely improvement that can be anticipated in levels of BaP. There is considerable uncertainty in estimates of current and future emissions and these increase the uncertainty in the outcomes of the modelling assessment. The results presented in this chapter should therefore only be considered as indicative of possible future levels of BaP that remain uncertain.

4.1   METHOD

The method used to calculate site-specific projections of annual mean BaP concentrations was first implemented for the calculation of NO_2 and PM10 projections and is described in the Air Quality Strategy (DETR et al, 2000) and in some detail by Stedman et al (1998a) and Stedman (1999a and 1999b). The method has been implemented for BaP using monitoring data from 1999 and 2000 (Chapter 3) and the updated emissions inventory and projections (Chapter 2)

The projections are based on measurements carried out at sites within the PAH monitoring network (see Table 3.1 for details of the site locations and a list of sites included in this analysis and see www.aeat.co.uk/netcen/airqual for an archive of the monitoring results). The following steps were undertaken to project measured concentrations, and were carried out using the current baseline emissions scenario:

1. The measured BaP concentrations were divided into component parts, (rural, local and urban industrial components), as discussed above, for the following sectors:

> Domestic
> Industry
> Road transport
> Other
> Natural and accidental fires

    Local sources were summed within a 35 km x 35 km area centred on the monitoring site location. An ADMS based dispersion matrix was applied to weight emissions from individual grid cells according to distance and direction from the site location. It was assumed that the rural background contribution to the annual mean BaP concentration was 0.05 ng m^{-3}.

2. The contribution, in concentration units, from significant sources (coke and aluminium production processes) at industrial locations was derived from the dispersion modelling exercise for all significant point sources (see Section 4). Projected concentrations at these locations were based on a combination of national emission changes and detailed study of the processes activity data in operation as described in Section 2 above.

3. Each source component was then projected forwards from the measurement year through to 2025 according to the projected change in emissions from each sector and summed to give an estimate of annual mean BaP concentration.

Figures 4.1 to 4.4 present illustrative examples of site-specific projections of BaP concentrations for years between 1990 and 2025. Projected concentrations have been calculated from measured concentrations in 1999 and 2000. Monitoring data prior to 1999 are limited. For two sites with long running datasets Manchester and Middlesbrough, we present data for the 1997 calendar year that represents a projection based on poor atmospheric dispersion characteristics. Figures 4.1 and 4.2 show that there generally good agreement between the projections and the measured concentrations in earlier years, giving confidence in the emissions sector split and emissions estimates.

Figures 4.1 to 4.4 indicate that the EPAQS objective of 0.25 ngm^{-3} is likely to be met at by 2010 at the Manchester and Middlesbrough urban background locations and the Holyhead urban industrial location irrespective of atmospheric dispersion characteristics. The same objective is unlikely to be met at the Lisburn urban background location as a result of continued domestic coal usage.

4.2   BASELINE PROJECTIONS FOR 2010

Projected BaP concentrations for 2010 (using 1999 and 2000 base years) are presented in Table 4.1

Table 4.1 Annual mean BaP projections to 2010 based upon 1999 and 2000 base years

	Estimated annual mean BaP 2010 (ngm-3)	
	1999 base year	2000 base year
Ashington	0.14	0.13
Bolsover	0.15	0.16
Cardiff	0.42	0.23
Hazelrigg	0.05	0.06
Glasgow	0.11	0.08
High Muffles	0.06	0.04
Holyhead	0.07	0.08
Kinlochleven	1.93	0.28
Lisburn	0.42	0.56
London	0.14	0.08
Manchester	0.09	0.13
Newport	0.16	0.24
Port Talbot	0.18	0.40
Scunthorpe	0.36	0.79
Stoke Ferry	0.09	0.07

Table 4.1 indicates that by 2010, annual mean BaP concentrations will be lower than the EPAQS objective in all locations except industrial sites in Scunthorpe and Kinlochleven and the urban background location in Lisburn. Projected concentrations in 2010 for the Kinlochleven site are conservative. Projections have been derived from measurement data from base years during which the adjacent aluminium smelting facility was operational. It is reasonable to expect

concentrations in 2010 to be significantly lower following closure of this plant. Elevated concentrations in Lisburn and Scunthorpe continue to be driven by local emissions from domestic coal usage and steel works respectively.

Figure 4.1   Projected annual mean BaP concentrations at Manchester based on 1997, 1999 and 2000 monitoring data
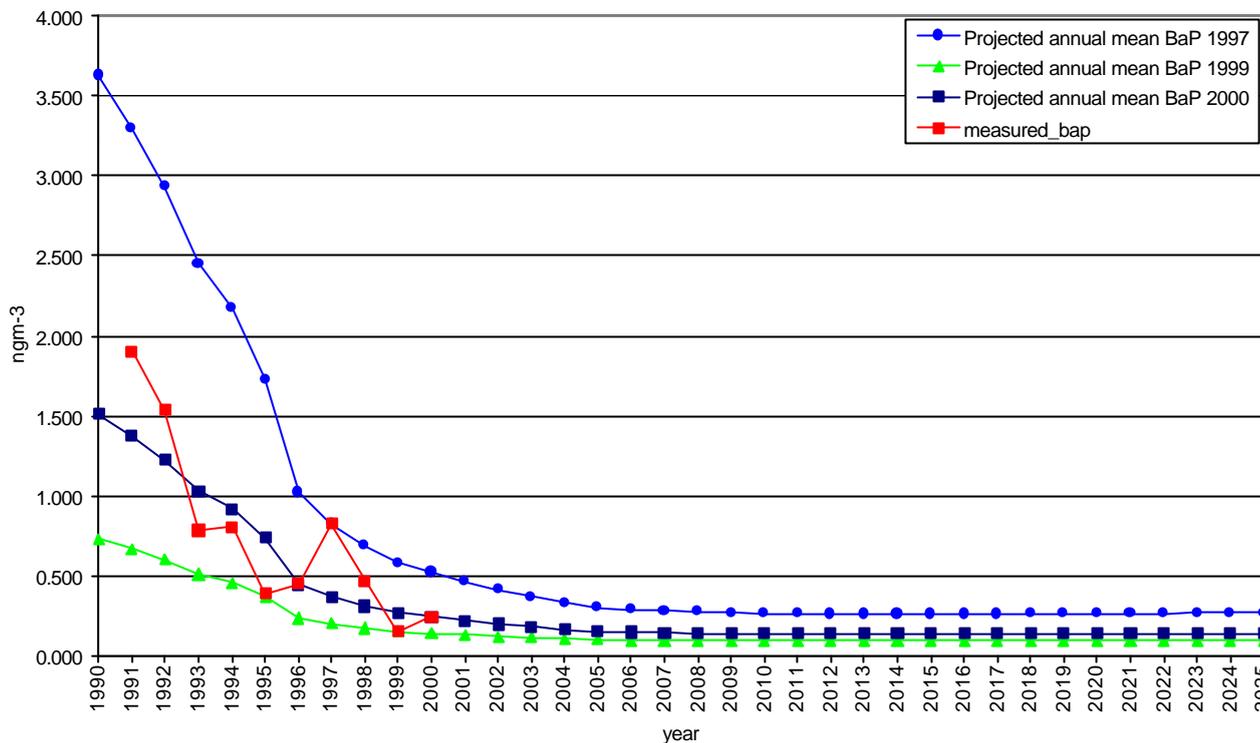
Figure 4.2 Projected annual mean BaP concentrations Middlesbrough based on 1997, 1999 and 2000 monitoring data
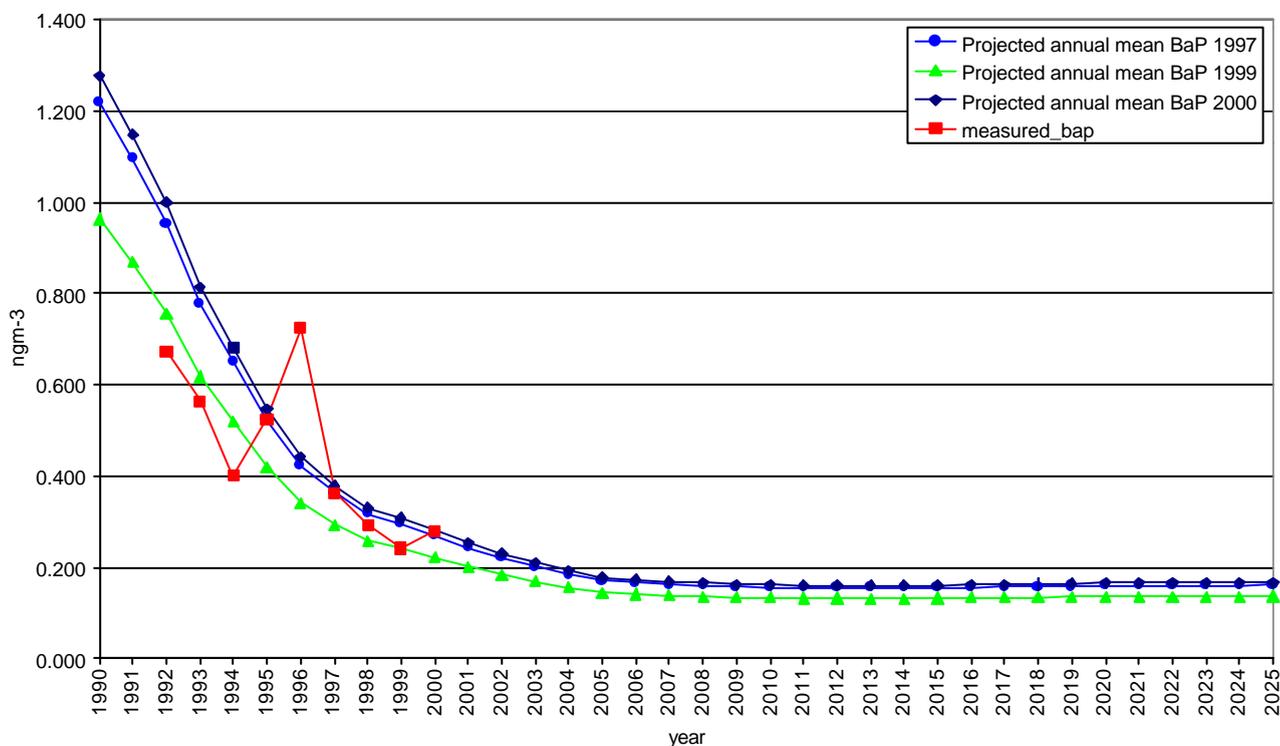
Figure 4.3 Projected annual mean BaP concentrations at Holyhead based on 1999 and 2000 monitoring data
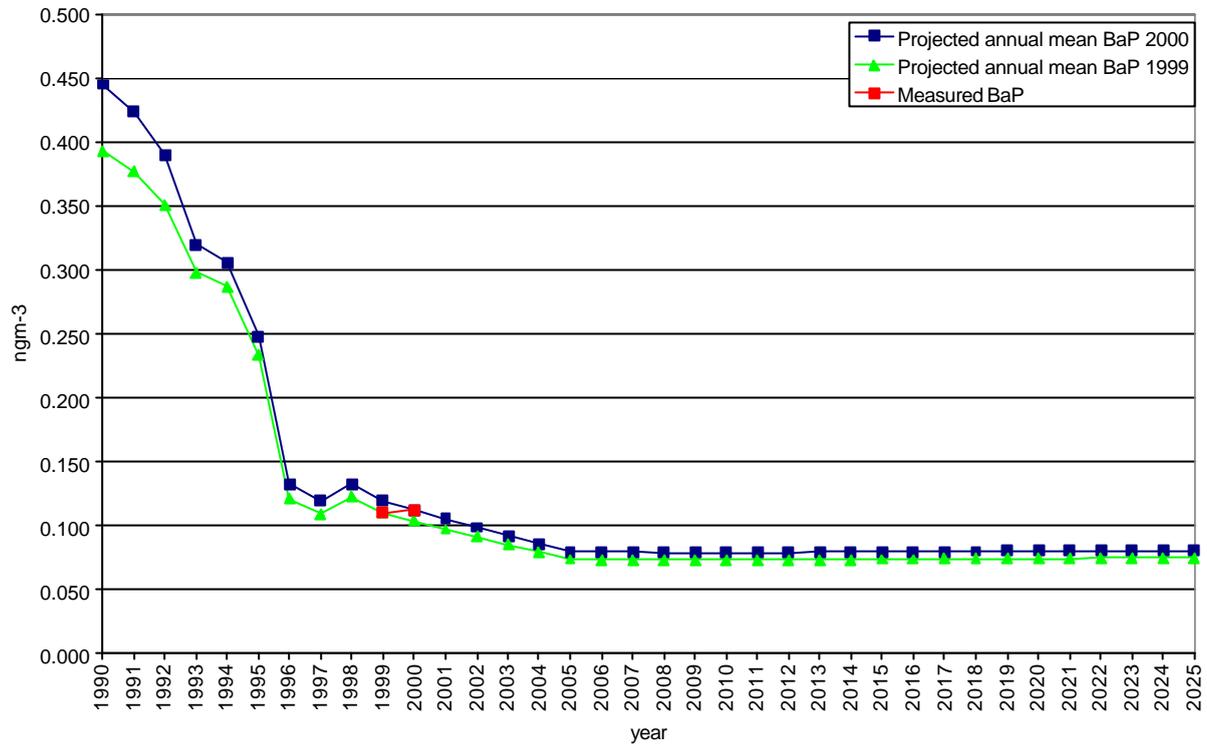
Figure 4.4 Projected annual mean BaP concentrations at Lisburn based on 1999 and 2000 monitoring data
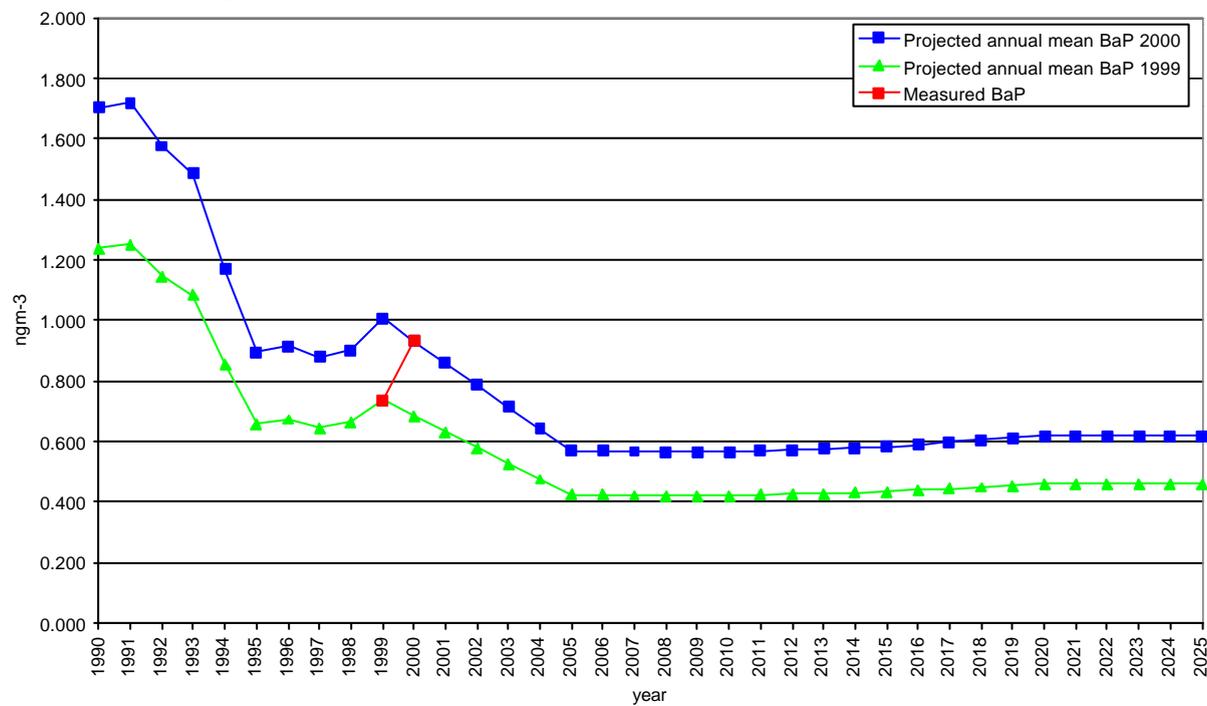
5 Mapping of current and future BaP concentrations

This section presents the results of a dispersion modelling approach to derive maps of BaP concentrations for 1999 and 2010. These maps have been combined with data on population to estimate the numbers of people exposed to different levels of BaP in these years. As with the previous assessments uncertainties are significant and the results should be treated as indicative only.

5.1 AREA SOURCE MODELLING

The NETCEN area source model has been used previously to predict annual sulphur dioxide and BaP concentrations (Abbott and Vincent, 1999; Abbott and Vincent, 2000). The current NETCEN area source model incorporates results from the dispersion model, ADMS-3 and calculates the annual average contribution from area sources on a 1 km receptor grid covering the country. To take account of contributions at each receptor from sources at distances greater than 15 km in the north-south or east-west directions a rural background concentration of 0.5 ng m^{-3} added. A uniform surface roughness of 1 m, corresponding to typical urban areas was used for the whole country. Wet and dry deposition were ignored on the basis of that they were shown to be insignificant in sensitivity studies (Abbott and Vincent, 2001).

BaP emissions for each 1 km square in the country for 1999 were obtained from the updated National Atmospheric Emission Inventory (NAEI) as discussed in Section 2. The emissions from each square was assumed to be uniformly distributed throughout the square at an initial height of 10 m: i.e., each 1 km square was represented by an emitting volume 1 km × 1 km × 10 m high. The estimate of 10 m is based on the height of a typical house and assumes that emissions will be entrained in the building wake.

5.2 POINT SOURCE MODELLING

The modelling of BaP emissions from point sources is complicated by the uncertainty in the scale of the emission source since many of these are fugitive in nature. For example, for a coking plant used to produce metallurgical coke, coal is placed into a hot oven which is sealed and heated to temperatures greater than 1300°C for periods of up to forty hours. The fugitive emissions which occur from seals and the oven door during this process are difficult to measure and in the absence of measurements difficult to quantify. Operators of coke ovens have made considerable efforts in recent years to reduce these fugitive emissions so that emissions may have been historically higher than current levels.

Typically, the following parameters were used to characterise the discharges from each plant:
- stack height;
- stack diameter;

- discharge velocity;
- discharge temperature.

The emission characteristics used in the modelling are provided in Appendix 1. Further details of the discharge characteristics of the emission sources were obtained from the respective Part A Authorisation, the Environment Agency or the plant operator. Footnotes to the Table in Appendix 1 provide further information relating to the assumptions made in the assessment.

5.3　　AREA SOURCES

A number of source categories were modelled as Area sources within each 1 km^2 grid square:

- roads;
- light industrial activity;
- coal and wood combustion in domestic grates;
- natural, controlled, accidental and malicious fires

Each of these sources were modelled separately. In the case of roads and coal and wood combustion in domestic grates the temporal variation in emissions (Section 2) was incorporated within the modelling assessment.

5.3.1　　Meteorological data for area scale modelling
The meteorological data used in the modelling assessment utilised information from sites throughout the UK that was divided into 14 separate geographical regions. The meteorological conditions for each region were characterised using one hourly sequential data for 1999 from the most suitable meteorological station. The geographical regions and meteorological data sets used are illustrated in Figure 5.1. Each data set included:

- The wind speed in 0.5 m s^{-1} increments;
- The wind direction in 10° increments
- An estimate of the cloud cover for each hour
- Boundary layer height
- Precipitation amount

5.4　　COMPARISON OF MODELLED AND MEASURED NO$_X$ CONCENTRATIONS

5.4.1　　Model validation using NO$_x$
To be confident of the suitability of the area source model to predict concentrations, NAEI emission estimates for oxides of nitrogen (NO$_x$) were modelled and compared against concentration measured at automatic monitoring stations.

Figure 5.1   Meteorological stations used in the modelling of benzo[a]pyrene concentrations

Region	Meteorological site
A	Hurn
B	Yeovilton
C	Heathrow
D	London Weather Centre
E	Shawbury
F	Coleshill
G	Wattisham
H	Valley
I	Ringway
J	Waddington
K	Aldergrove
L	Glasgow
M	Boulmer
N	Aviemore
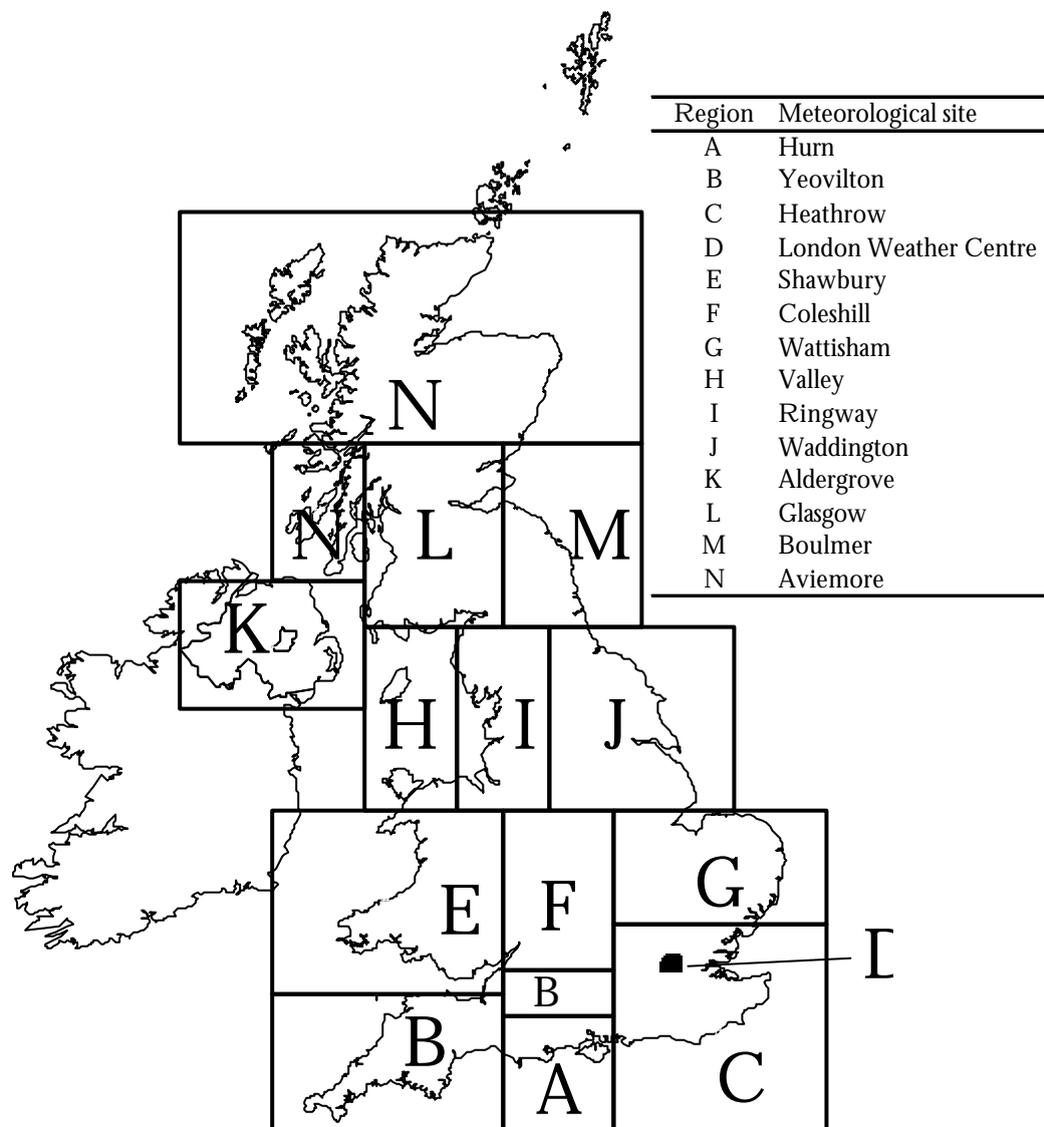
The modelling method used to model oxides of nitrogen concentrations was the same as that used to predict BaP concentrations. However, as a disaggregated NO_x inventory for 1999 is not yet available, emissions for 1998 were modelled using 1999 meteorology. The spatial variance in NO_x concentrations is principally controlled by the spatial distribution of traffic emissions, these change little from year to year. Source sectors similar to those for BaP were modelled.

A background component was added to the modelled concentrations. This background concentration was derived by interpolating the 1999 nitrogen dioxide concentrations measured by diffusion tube at sites belonging to the acid deposition monitoring network (Hayman *et al.,* 2001). This network was designed to monitor rainwater concentrations on a regional basis and hence the sampling sites are mainly situated away from major sources of pollution. For the purposes of this modelling study sites influenced by specific

point sources were excluded from the assessment. NO_x concentrations were obtained from the NO_2 concentrations using a multiplication factor of 1.2 (Stedman, 1998). Figure 5.2 shows that the modelled concentrations agree well for a variety of sampling site classification types (excluding those directly influenced by local sources, such as adjacent to roads).

Figure 5.2   Scatter plot showing predicted and modelled NOx concentrations
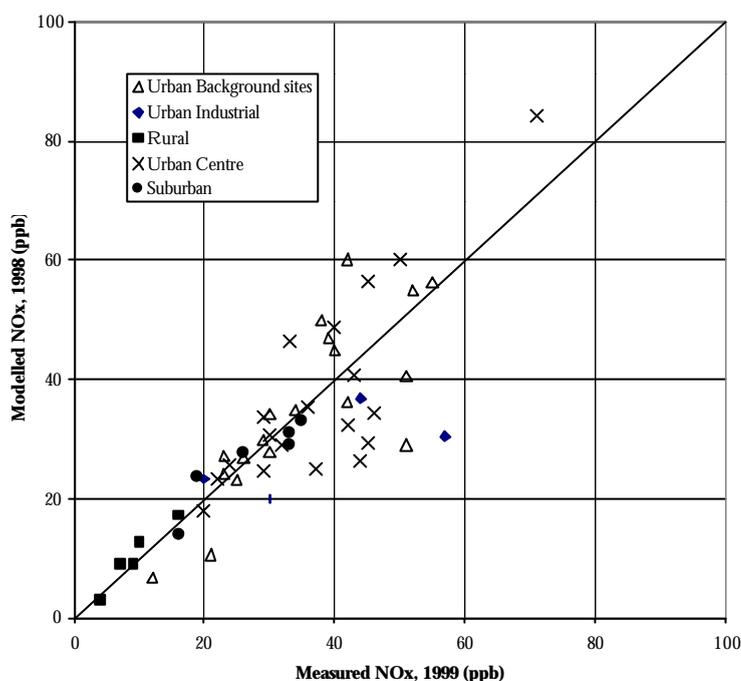
The relatively good agreement between measured and modelled NO_x concentrations indicates that the dispersion modelling approach works well for a conserved pollutant and provides confidence in predicting BaP concentrations. Emissions from road traffic sources are seen to be the dominant emission source (Figure 5.3).

5.5   PREDICTION OF BAP CONCENTRATIONS IN 1999

BaP concentrations in 1999 were predicted using the dispersion modelling approach described above.

5.5.1   Background concentrations

To take account of distant sources of BaP outside of the modelled 15km^2 a rural background concentration was added to modelled concentrations. Table 5.1 presents the annual mean BaP concentrations measured at the two rural sites at High Muffles and Stoke Ferry and at the semi rural site at Hazelrigg. Concentrations are approximately the same at Hazelrigg and High Muffles but are significantly higher in Stoke Ferry. In the absence of further information regarding concentrations at more remote sites a concentration of 0.05 ng m^{-3} was seen as a reasonable estimate of background concentrations at all locations.
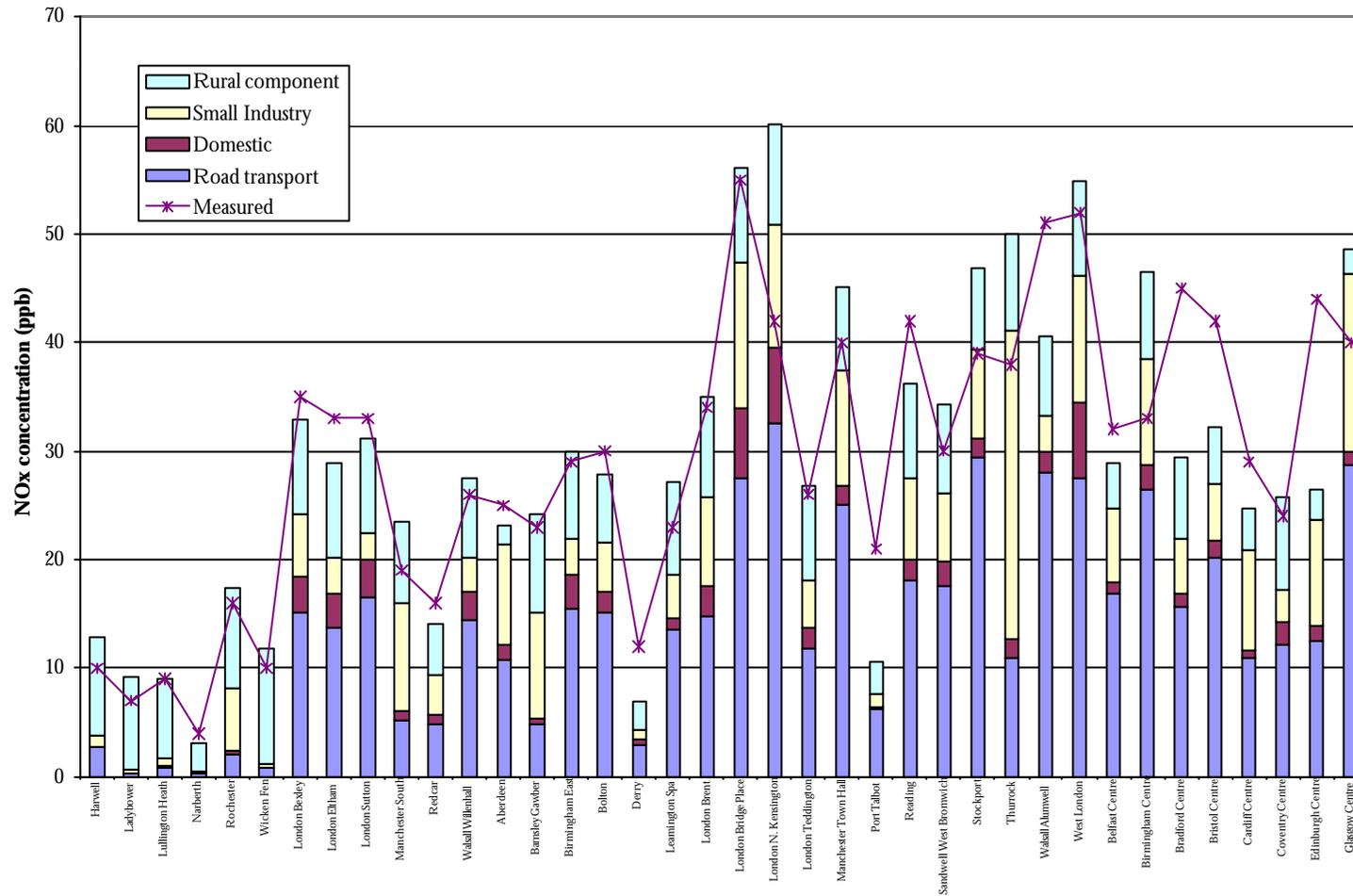
Figure 5.3 NO$_x$ source apportionment for rural, suburban, urban background and a selection of urban centre sites

Table 5.1 Annual mean concentrations of BaP (ng m^{-3})

Site	1999	2000
Hazelrigg	0.06	0.06
High Muffles	0.06	0.04
Stoke Ferry	0.11	0.09

Further work would be required to produce a spatially varying background concentration.

5.6   VALIDATION FOR BAP PREDICTED CONCENTRATIONS

Figure 5.4 shows a comparison of modelled and measured BaP concentrations at monitoring site locations. There is reasonably good agreement at all sites except Lisburn where the measured concentration is significantly under predicted. Further work is needed to reconcile measured and modelled concentrations of BaP in Lisburn.

Figure 5.4   Scatter plot showing predicted and modelled BaP concentrations
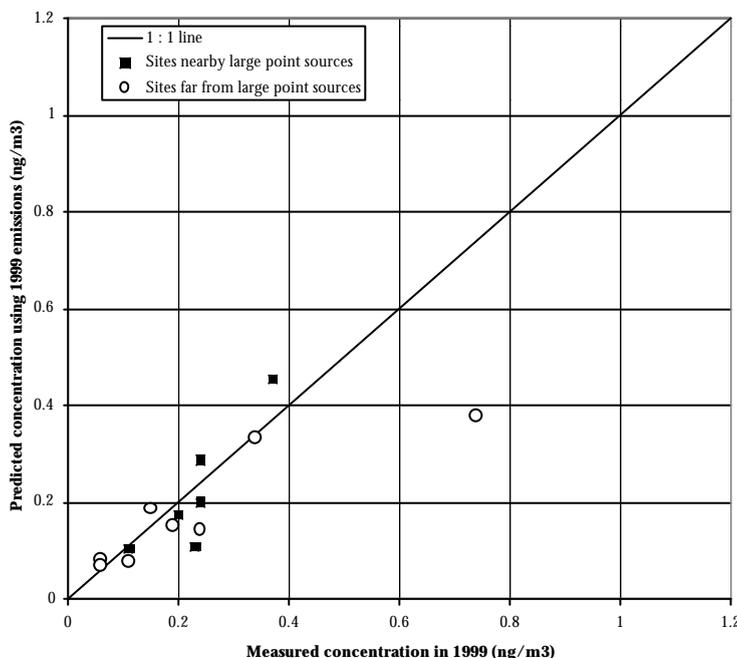
A source apportionment of modelled annual mean BaP concentration is shown in Figures 5.5 and 5.6. For most sites close to the large point sources the influence of the large emitter can clearly be seen. At other locations, such as at Lisburn, the domestic emissions is the most dominant source, for the site in London, the "area other" source – mainly fires dominates. For the rural and semi-rural sites the background component is controlling predicted concentrations.

Figure 5.5 BaP source apportionment at all BaP sampling sites
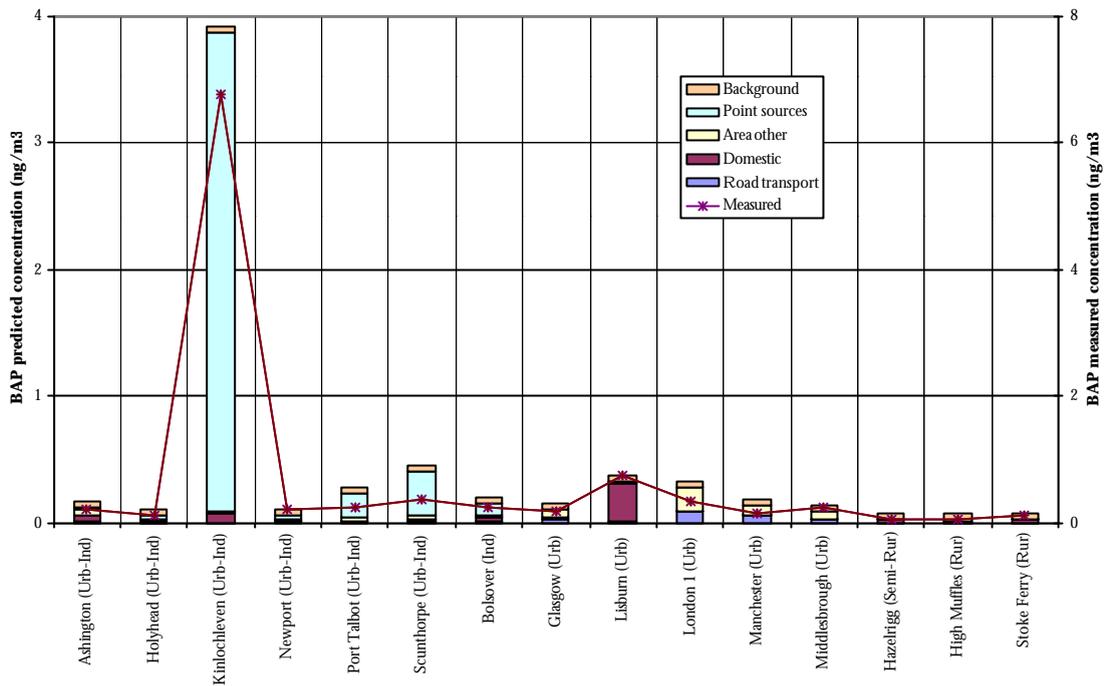
Figure 5.6 BaP source apportionment at sampling sites far from large point sources
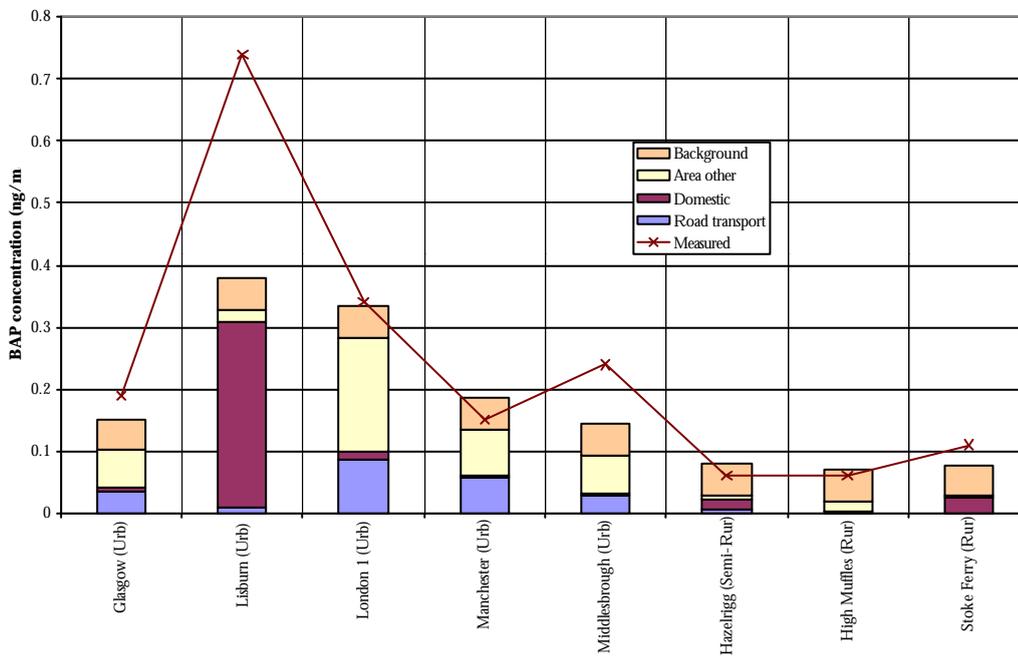
5.6.1 Sensitivity Analysis of the Spatial Distribution of Emissions from Fires

BaP emissions from fires in the United Kingdom were estimated to be 2880 kg in 1999 (28 % of the total). Fifty percent of this emission was assumed to originate from an urban sector and fifty percent from a rural sector. For the purposes of modelling the BaP emission, the fire sector was included within the "area other" category and was the major constituent of this category. Figure 5.7 shows that the area other source category was the most dominant source for sampling sites within cities like Glasgow, London, Manchester and Middlesbrough. A simplified analysis was performed to assess how sensitive the predicted BaP concentration was to the magnitude of the urban fire source.

When the urban fire emission was reduced by half to 720 kg, concentrations were under predicted at all Great Britain urban sites with the exception of Manchester (Figure 5.8). When the urban fire emission was doubled (to 2880 kg) concentrations were over predicted at all Great Britain urban sites with the exception of Middlesbrough (Figure 5.9).

Emissions from the fire sector are uncertain. The national urban emission total has been disaggregated based on population statistics- this had the effect of weighting the relatively large emission to urban centres. Figures 5.8 to 5.9 showed that the prediction of BaP is particularly sensitive to the proportion of BaP emission assigned to the urban fire category.

Further work to quantify the mass and spatial resolution of emissions from fires is necessary. The sensitivity analysis also demonstrates that the results of the modelling assessment must be treated as preliminary, contain considerable uncertainty and the results should therefore be treated with caution.

Figure 5.7 BaP source apportionment at sampling sites far from large point sources (urban fire component of "area other" has been scaled by 1.0)
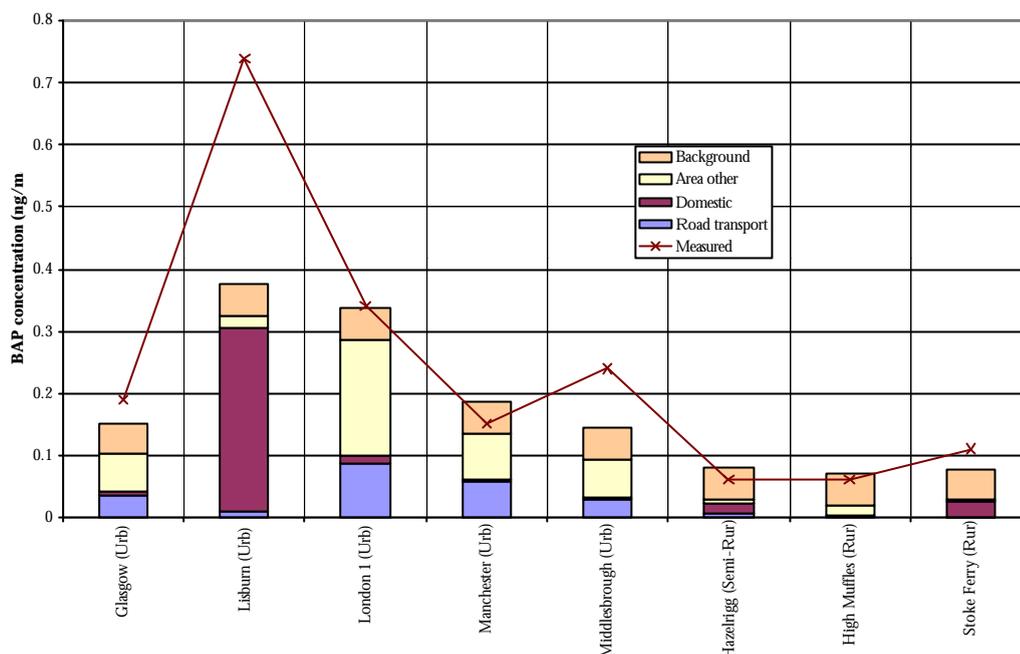
Figure 5.8 BaP source apportionment at sampling sites far from large point sources (urban fire component of "area other" has been scaled by 0.5)
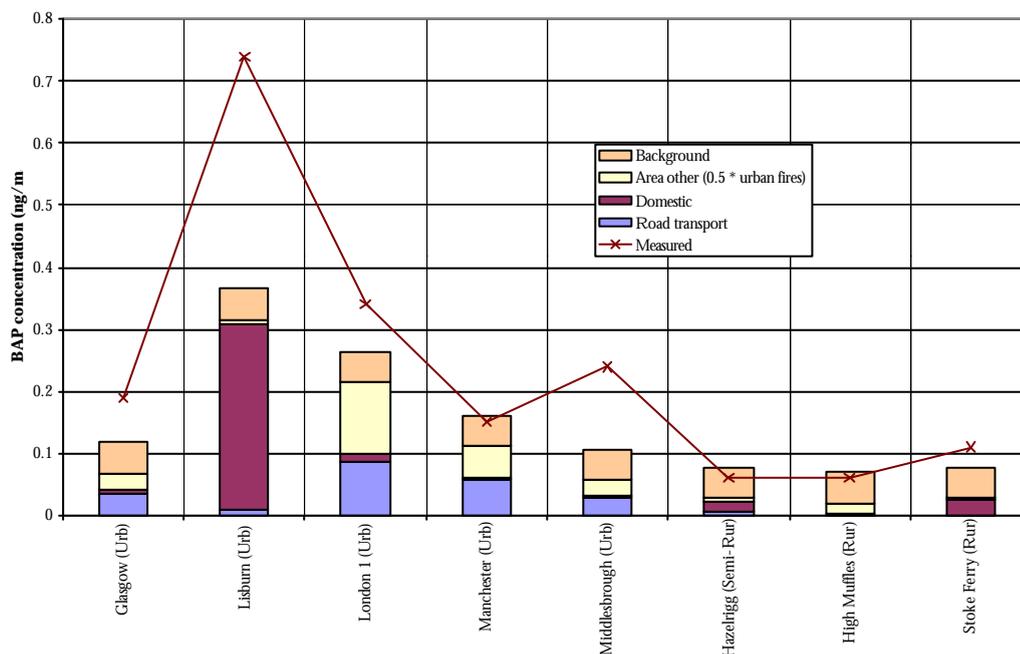
Figure 5.9 BaP source apportionment at sampling sites far from large point sources (urban fire component of "area other" has been scaled by 2.0)
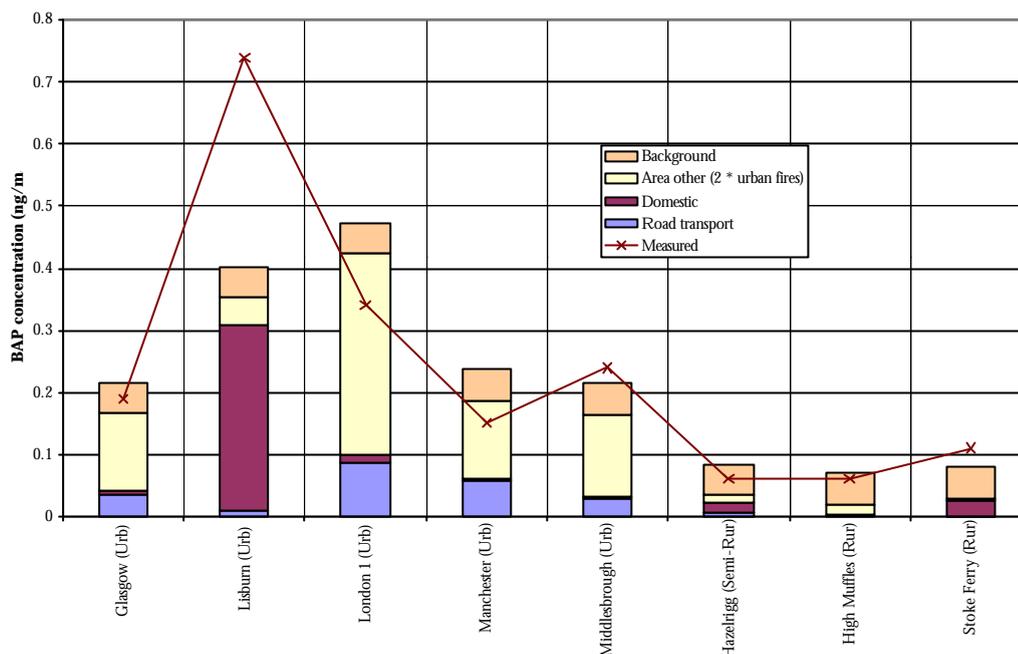
5.7 CONCENTRATION MAPS

The emissions modelled from the area and point sources were combined to produce BaP concentration maps for both 1999 and 2010. A background concentration of 0.05 ng m^{-3} was added for both years. Figures 5.10 and 5.11 show areas of the UK, in 1999 and 2010 estimated to exceed a BaP concentration of 0.25 ng/m^3. Where the 0.25 ng/m^3 concentration is exceeded, the overwhelming majority of 1 km squares do not exceed 0.5 ng/m^3.

A comparison of measured and modelled BaP concentration at the sampling site locations showed that the measured concentrations at most sites were reasonably well predicted (see Figures 5.5 and 5.6). However, it is expected that the modelled concentration will reflect the inherent uncertainty in emission estimates or assumptions used in modelling emissions from point sources. Other sources of uncertainty include the use of just fourteen meteorological data sets to characterise the entire United Kingdom surface area and using 1999 meteorological data to predict concentrations in 2010. It might be expected that a spatially varying background BaP concentration would improve prediction in rural areas.

Figure 5.10   BaP concentrations in 1999
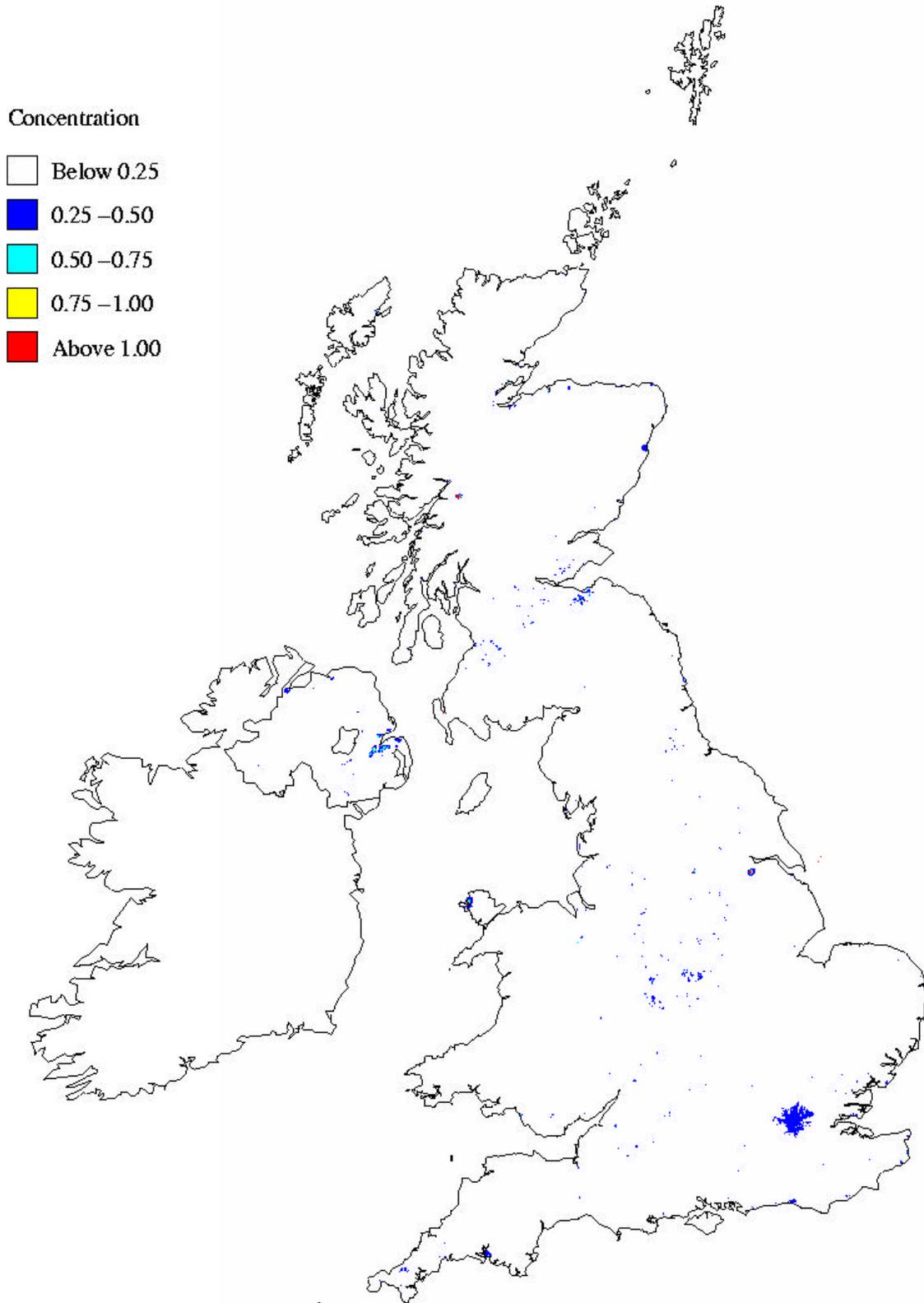
Figure 5.11 BaP concentrations in 2010
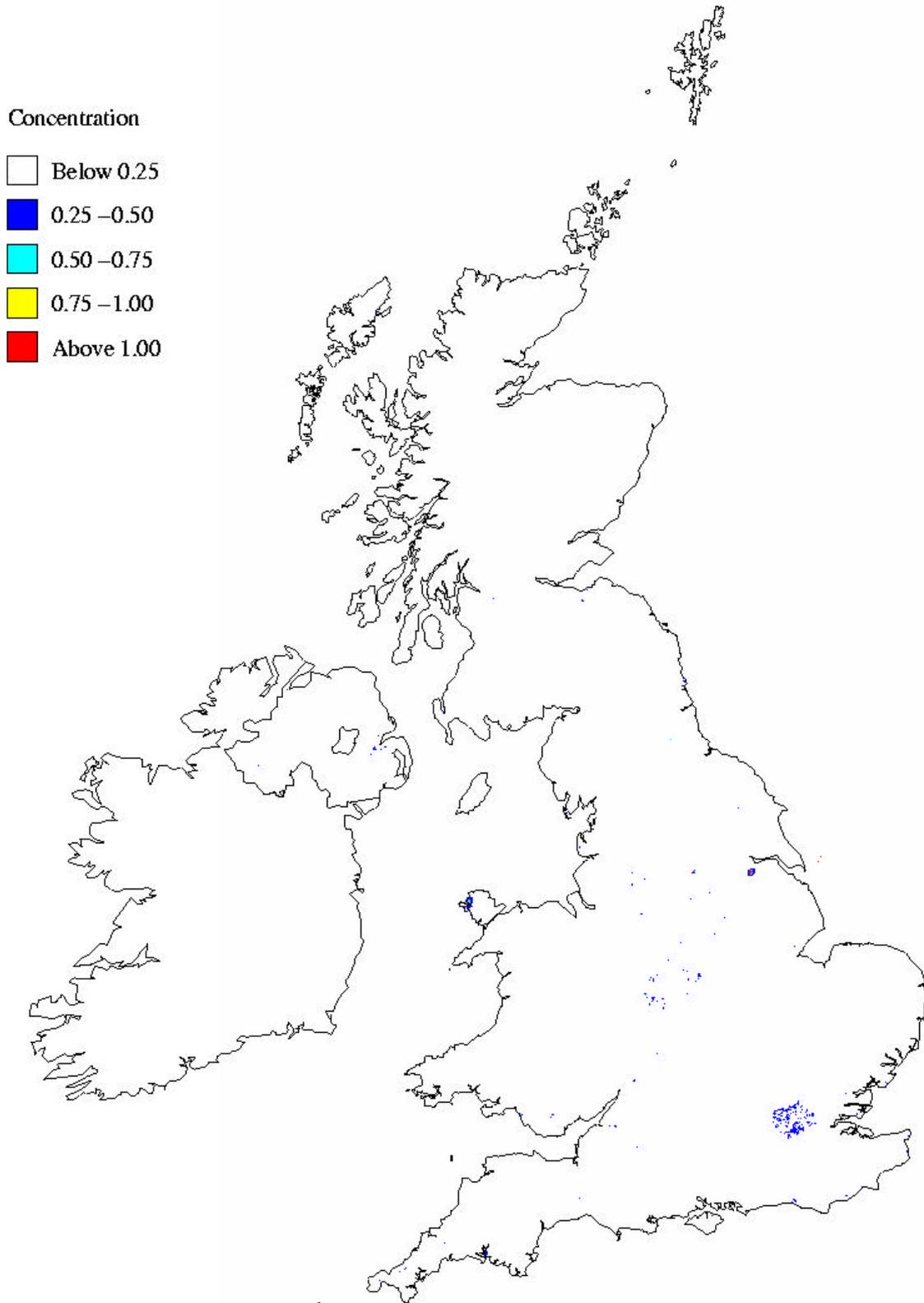
5.8   POPULATION EXPOSURES

Having derived BaP concentration maps for 1999 and 2010, population weighted mean concentrations for both 1999 and 2010 were obtained. The population weighted mean is calculated by multiplying the concentration in each 1 km grid square by the population, summing the results and dividing by the total population.

Populated weighted mean concentrations will by their nature be dominated by concentrations predicted in areas of high population density. Table 5.2 shows, assuming the population and its distribution in 2010 will be the same as in 1999, that the population weighted mean will decrease by 0.025 ng m^{-3}.

Table 5.2 also shows the number of people in the United Kingdom exposed to modelled BaP concentrations above certain concentration thresholds. Between 1999 and 2010 the number of people exposed to levels of BaP greater than 0.25 ng m^{-3} decreases by more than 60 %. For the 0.5 ng m^{-3} threshold, the decrease is much more dramatic- a decrease of 90 % is predicted. For the 1 ng m^{-3} threshold, the numbers of people exposed also decreases although the numbers of people potentially exposed to levels above this criterion are very small. The last component of Table 5.2 shows the area of the United Kingdom exposed to each concentration threshold.

Table 5.2   Comparison of BaP exposure in 1999 and 2010

Criteria	1999	2010
Population weighted mean concentration (ng/m^3)	0.156 ng/m^3	0.131 ng/m^3
Population exposed to BaP >0.25 ng/m^3	5817 000	2243000
Population exposed to BaP >0.5 ng/m^3	197000	18000
Population exposed to BaP > 1.0 ng/m^3	2900	1300
Number of 1 km squares in which BaP > 0.25 ng/m^3	1562	551
Number of 1 km squares in which BaP > 0.5 ng/m^3	122	40
Number of 1 km squares in which BaP > 1 ng/m^3	20	11

0.25 ng/m^3 Proposed standard
0.5 ng/m^3 Lower limit of range proposed by EC Working Group
1.0 ng/m^3 Upper limit of range proposed by EC Working Group

6 Conclusions

In 1999, the most recent year for which emissions estimates have been calculated, total UK emissions of benzo[*a*]pyrene (BaP) were 10.3 t. The main sources of emissions were from domestic coal and wood burning (39%), fires (28%), anode baking (19%) and vehicles (8%). Emissions of BaP have declined markedly since 1990 when the national total emission is estimated to have been 68 t.

Emissions of BaP are predicted to decline to 6.4 t in 2010, thereafter showing a slight increase to 6.6 t in 2020. The sectors contributing to the decline in emissions are road transport, the production of anodes for the aluminium industry and domestic coal use. The sectors whose emissions are predicted to increases are principally aluminium production and domestic wood burning.

The emission inventory for BaP is subject to considerable uncertainty. This is since few emission measurements have been made both nationally and internationally and the relevant activity statistics in some cases are not collected regularly. In particular further work is required to understand better the magnitude and spatial pattern of emissions from accidental, malicious, demolition and natural fires, bonfires and domestic solid fuel use particularly in urban and rural communities without access to natural gas.

AEA Technology's National Environmental Technology Centre (NETCEN) operates the PAH network (15 sites) and TOMPs network (6 sites) measuring concentrations of PAHs in ambient air on behalf of DEFRA and the devolved administrations. The concentrations of BaP in 2000 ranged between 0.04 ng m^{-3} at rural sites to 2.28 ng m^{-3} at an industrial site. Concentrations of BaP at seven of the 12 urban / industrial sites operating in 2000 equalled or exceeded 0.25 ng m^{-3}. However only at Kinlochleven (2.28 ng m^{-3}), Lisburn (0.93 ng m^{-3}) and Scunthorpe (1.17 ng m^{-3}) were concentrations significantly greater than the EPAQS guideline. The aluminium smelter at Kinlochleven closed in June 2000. The Lisburn site is in an area of Northern Ireland which is not a smoke control area and does not yet have a supply of natural gas. The Scunthorpe site is near both a major integrated steel works and a coal tar processing plant.

Statistical analysis of the measurements from those sites which robust pollution trend analysis is possible suggests that there has been a significant decline in concentrations during the 1990s. The rate of decline has been greatest at urban sites. The measured concentrations of BaP also show significant seasonality. Concentrations are typically greater in winter than in summer by factors of between 1.8 at industrially influenced sites, 4.2 at urban sites and 4.5 at rural sites.

The projected emissions were used to predict concentrations in 2010 at the sites which were operating during 1999 and 2000. This assessment suggested that by 2010 concentrations may be less than 0.25 ng m^{-3} at all sites with the exception of Kinlochleven, Lisburn and Scunthorpe. The projection methodology is conservative and it is probable that with the closure during 2000 of the Kinlochleven smelter the concentrations there will decline more rapidly than predicted.

Maps of predicted UK BaP concentrations in 1999 and 2010 have been prepared. It is expected that the uncertainty in the modelled concentrations will reflect the inherent uncertainty in the emission estimates and the assumptions used in modelling emissions from point sources. Other

sources of uncertainty include using 1999 meteorological data to predict concentrations in 2010. The concentration maps suggest that while areas of the country are likely to continue to exceed the 0.25 ng m^{-3} criterion in 2010 the area affected will be significantly smaller than in 1999. The area greater than 0the EPAQS guideline will decline from 1562 to 551 km^2. The area with concentrations greater than 1.0 ng m^{-3} is predicted to decline from 20 to 11 km^2. The population weighted annual average concentration is predicted to decline from 0.156 to 0.131 ng m^{-3}.

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Appendices

CONTENTS

Appendix 1
Point Source Release Details

CONTENTS

Table A1 Emission characteristics for each of the individual plant

Process type	Site	Stack Height	Diameter	Velocity	Temp. ^{o}C	Met. Station (1999 data)
Aluminium Production	Ashington[1]					Boulmer
	Fume Treatment Centre	32	1.960	16.1	109	
	FTC by-pass	46	2.125	13.0	131	
	Green mix scrubber	36	2.150	5.9	32	
	Holyhead[2]	121	9.807[2]	7.87[2]	82[2]	Valley
	Kinlochleven[3]					Aviemore
	Roof Vents	14	1.29	6.1	19	
	Fume Treatment Plant	25	1.78	8.2	16	
Steel Works[+] (Coking)	Llanwern Works	♣	♣	♣	♣	Shawbury
	Middlesbrough:	♣	♣	♣	♣	
	Southbank and Redcar					Boulmer
	Port Talbot:	♣	♣	♣	♣	
	Morfa and Grange					Shawbury
	Scunthorpe:	♣	♣	♣	♣	
	Dawes Lane and Appleby					Waddington
Coking (Smokeless fuel)	Bolsover (Coalite)[♠]	♣	♣	♣	♣	Waddington
Coking (Metallurgical)	Monckton CC[♣]	♣	♣	♣	♣	Waddington
	Cwm Coke Works	♣	♣	♣	♣	Shawbury

Notes

+ Each of the steel plant at Middlesbrough, Scunthorpe and Port Talbot have two sets of coking ovens with the emission split as follows: 33:67 between Southbank and Redcar works; 50:50 between Dawes Lane and Appleby works and 50:50 between Morfa and Grange works.

1. Dispersion characteristics for Ashington were provided by Alcan Smelting and Power UK. Personal Communication from John Clarkson to Peter Coleman, 9 th May and 15 th May 1999.
2. Information provided by Colin Hardman (EA), personal communication to Keith Vincent, 6 th July 1999. The stack at Holyhead exhausts gases from two separate processes; the carbon bake and the pot rooms. The stack consists of two concentric stacks, one inside the other with the exhaust from the pot room being the outer stack and carbon bake the inner one. BaP emissions are only expected to occur from the inner stack. As both processes are running at the same time it is necessary to take account of the influence of the buoyancy of the larger stack will have on the buoyancy of the emission from the inner stack. To do this the buoyancy and momentum of the both plumes were combined and an effective stack diameter and combined stack temperature was determined.

3. Dispersion characteristics for Kinlochleven were provided by Alcan Smelting and Power UK. Personal Communication from John Clarkson to Peter Coleman, 9 th May and 15 th May 1999. Although there are 77 roof fans extracting air from the cell room, emissions were modelled as a point source and was assumed to come from one fan located in the centre of the roof of the cell room.

♣ Each of the coke works was treated as a line source with dimensions taken from the relevant Ordnance Survey 1:25000 map (typically 80-225 m long and 15 m wide). Coke ovens require gas heating in order to overcome heat losses and maintain the pyrolysis reaction. At Cwm coke works, total fuel consumption is approximately 10500 $m^3 h^{-1}$. Assuming a heating value of the gas used of 4.7 MJ m^{-3} (Perry,1973), it is estimated that 13.6 MW is required to maintain the reaction. For this assessment, it has been assumed that all this heat is lost through the walls and roof of the coke ovens. This assumption

ignores the heat lost through battery stacks and heat lost or gained from endothermic or exothermic reactions: however, it is considered that the assumption is sufficient to provide order of magnitude estimates of the heat loss. The heat loss from the ovens is then assumed to be contained within the discharging plume from the ovens, providing buoyancy so that there is some limited plume rise. A nominal initial plume temperature of 115°C, specific heat 1012 J K^{-1} kg^{-1} and density 1.25 kg m^{-3} was used to estimate the initial volume flow rate of the plume. For other coke works, the heat loss and volume flow rate were estimated by scaling the estimates for Cwm coke works on the basis of the NAEI emissions estimates.