AEAT-3512/REMC/20459131/ISSUE 1

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Customer Department of the Environment, Transport and the Regions

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Executive Summary

There is growing concern over the polluting effects of Polycyclic Aromatic Hydrocarbons (PAHs), reflected in their gaining a higher profile in international environmental agreements and legislation such as the UNECE POPs Protocol. The main concern is the carcinogenic effects of some PAHs.

The UK Department of the Environment, Transport and the Regions (DETR) commissioned the National Environmental Technology Centre to carry out a study to estimate the speciated UK emissions of PAHs. The results of this study will assist in the development of policies which may be considered by DETR as necessary to reduce emissions of PAHs.

The main sources of PAH emissions in the UK in 1995 were :
- anode baking
- vehicles
- industrial combustion of coal

The majority of anode baking in the UK is carried out for the primary aluminium industry. It is estimated that PAH emissions from this source have significantly decreased since 1995 because of improved abatement measures at anode baking plant.

The key issues related to PAH emissions from vehicles are likely to be the significant emissions from poorly maintained vehicles and emissions during cold starting. The current estimates for PAH emissions from diesel vehicles are particularly uncertain.

The inventory presented in this report gives a more complete picture than is currently available elsewhere. Analysis of the uncertainties in the PAH emission estimates shows that the current inventory has a high uncertainty and further work is required to improve the reliability of the estimates. However, the PAH inventory has allowed the identification and prioritisation of the likely main emission sources in the UK.

Further research should concentrate on the main emission sources. In particular, improvements in the quality of emission factors for anode baking and vehicles (particularly diesel and unserviced vehicles) are needed. In addition to these, improvements to the inventory are required for the following areas :
- coke production;
- primary aluminium production;
- creosote use;
- open burning;
- other domestic and industrial combustion.

Emissions from bitumen production and use, a potentially significant source, have not been estimated due to a lack of data. Future work should also focus on this source.

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APPENDICES: - SECTOR PROFILES AND UNCERTAINTY ANALYSIS

1. Introduction

1.1 BACKGROUND TO PROJECT

This report is written for the Department of the Environment, Transport and the Regions. It presents the development of the first speciated polycyclic aromatic hydrocarbon (PAH) inventory for the UK. Sixteen PAHs, designated by the United States Environmental Protection Agency (US EPA) as compounds of interest under a suggested procedure for reporting test measurement results (US EPA 1988), have been examined in this report.

Table 1.1 - Categories of PAHS

	US EPA Priority pollutants (16 PAHs)	**IARC** Probable or possible human carcinogens (6 PAHs)	**Borneff** (6 PAHs)	**UNECE POPs Protocol** Indicators for the purposes of emissions inventories (4 PAHs)
Naphthalene	✓			
Acenaphthylene	✓			
Acenapthene	✓			
Fluorene	✓			
Anthracene	✓			
Phenanthrene	✓			
Fluoranthene	✓		✓	
Pyrene	✓			
Benz[a]anthracene	✓	✓		
Chrysene	✓			
Benzo[b]fluoranthene	✓	✓	✓	✓
Benzo[k]fluoranthene	✓	✓	✓	✓
Benzo[a]pyrene	✓	✓	✓	✓
Dibenz[ah]anthracene	✓			
Indeno[123cd]pyrene	✓	✓	✓	✓
Benzo[ghi]perylene	✓		✓	

Table 1.1 indicates the well-known categories of PAHs. The International Agency for Research on Cancer (IARC) identified a subset of six of these PAHs as probable or possible human carcinogens (IARC 1987). The Borneff 6 PAHs have been used in some emission inventory compilations. Table 1.1 also shows the 4 PAHs to be used as indicators for the purposes of emissions inventories under the United Nations Economic Commission for Europe (UNECE) Persistent Organic Pollutants (POPs) Protocol.

The rest of Section 1 gives further background on PAHs and inventories. Section 2 of this report describes in more detail the method used to compile an emission inventory and discusses the uncertainties involved and how these are analysed. The UK speciated PAH inventory is described in Section 3 along with the results of the uncertainty analysis. The conclusions which can be drawn from these results and the priorities for further work are discussed in Section 4. In the appendices, each source sector is described in detail.

1.2 DEFINITION OF PAHS

Polycyclic aromatic hydrocarbons (PAHs) are a group of compounds composed of two or more fused aromatic rings. Table 1.2 shows the molecular weight, formulae and structures of the sixteen PAHs included in the inventory in this report. Typical UK ambient mean concentrations of PAHs range from 0-5 ng m^{-3} for the higher molecular weight compounds (e.g. benzo[a]pyrene) to 0-80 ng m^{-3} for the lower molecular weight compounds (e.g. phenanthrene).

1.3 PROPERTIES OF PAHS

The main environmental impact of PAHs relate to their health effects, focusing on their carcinogenic properties. The most potent carcinogens have been shown to be benzo[a]anthracene, benzo[a]pyrene and dibenz[ah]anthracene. The semi-volatile property of PAHs makes them highly mobile throughout the environment via deposition and re-volatilisation between air, soil and water bodies. It is possible that a proportion of PAHs released in the UK are deposited in the oceans and/or subject to long range transport making them a global environmental problem.

1.4 USE OF EMISSION INVENTORIES

Emission inventories give a better understanding of environmental problems. They help to develop policy to reduce environmental impacts through identification of the main polluting sources. In addition an emission inventory helps to assess the effectiveness of control measures as they are updated and allows the reporting of emissions as part of international agreements.

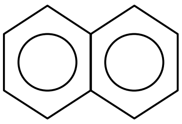
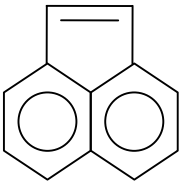
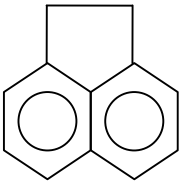
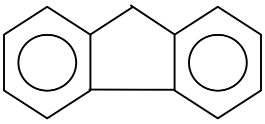
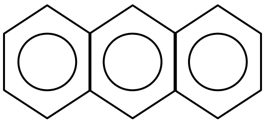
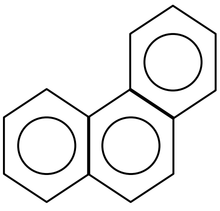
Inventories are subject to a certain amount of uncertainty both in the emission factors and the activity. However, the appropriate demonstration of this uncertainty can help to prioritise future work to improve the inventory.

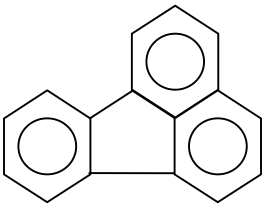
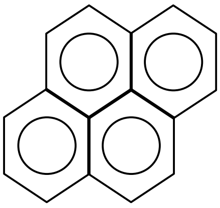
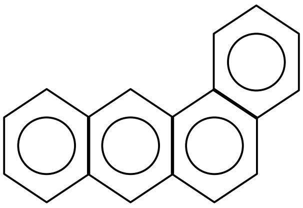
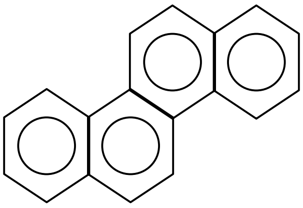
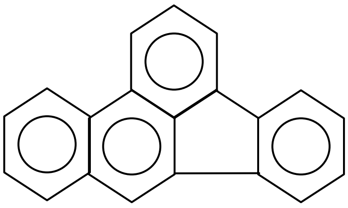
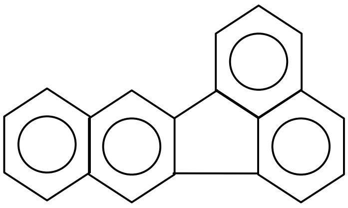
1.5 PREVIOUS PAH INVENTORY FOR THE UK

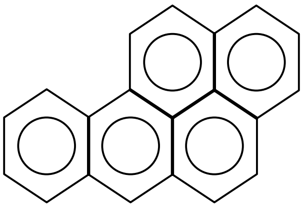
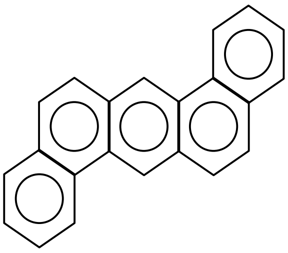
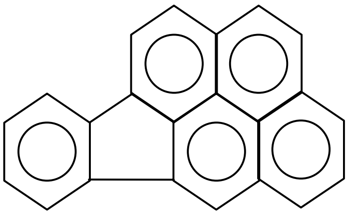
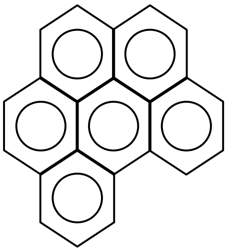
One of the first attempts to produce an inventory for PAH emissions in the UK is contained in the Air Pollution Abatement Review Group (APARG) report (DoE 1996) on the abatement of toxic organic micropollutants (TOMPS). Whilst this inventory was useful in prioritising sources of PAHs, it only reported on total PAHs giving limited information on the speciation of the PAHs emitted. In addition it was not always clear from the reference material used which PAHs were included in the ‘total’. Another estimate of PAH emissions was provided in a report by Wild and Jones (Wild 1985). This provided some speciation, but on fewer compounds, and covered fewer and broader source categories.

A speciated inventory is much more useful to prioritise sources of the most toxic PAHs in the light of international agreements and national regulations / standards, such as the UNECE protocols and the Expert Panel for Air Quality Standards (EPAQS) recommendations for air quality standards.

Table 1.2 - Molecular weight, formulae and structure of the sixteen PAHs classified by the US EPA as compounds of interest

PAH	Molecular Weight	Formula	Structure
Napthalene	128	$C_{10}H_8$	
Acenapthylene	152	$C_{12}H_8$	
Acenapthene	154	$C_{12}H_{10}$	
Fluorene	167	$C_{13}H_9$	
Anthracene	178	$C_{14}H_{10}$	
Phenanthrene	178	$C_{14}H_{10}$	

PAH	Molecular Weight	Formula	Structure
Fluoranthene	202	$C_{16}H_{10}$	
Pyrene	202	$C_{16}H_{10}$	
Benz[a]anthracene	228	$C_{18}H_{12}$	
Chrysene	228	$C_{18}H_{12}$	
Benzo[b]fluoranthene	252	$C_{20}H_{12}$	
Benzo[k]fluoranthene	252	$C_{20}H_{12}$	

PAH	**Molecular Weight**	**Formula**	**Structure**
Benzo[a]pyrene	252	$C_{20}H_{12}$	
Dibenz[ah]anthracene	278	$C_{22}H_{14}$	
Indeno[1,2,3-cd]pyrene	276	$C_{22}H_{12}$	
Benzo[ghi]perylene	276	$C_{22}H_{12}$	

2. Method

2.1 EMISSION INVENTORY

Emissions inventories are compiled through the use of emission factors (e.g. emission of a pollutant per unit of activity) which have been derived for each source sector from reported research. The emission factor is multiplied by a sector activity statistic (e.g. annual production or consumption) to give the estimated annual emission for that compound. When this is done for each source sector an inventory is built up.

In developing this PAH inventory there was little speciated information available for some source sectors. This resulted in the use of a profiling methodology, where the emission factors from one report are scaled according to, for example, an emission factor from a single PAH from another report. For example in the Coke Production sector, three reports on emission factors are used: Radian Corporation (1995), British Steel (1998) and the EMEP/CORINAIR Guidebook on Emissions Inventories (1998). The Radian Corporation report contained speciated emission factors for all the PAHs of interest, but for plant less representative of UK plant, whereas the other two contained a factor more representative of the UK, but for benzo[a]pyrene only. Assuming that the species profile will be similar for each plant where the measurements were made, the emission factors for the second two reports were derived by comparison of the benzo[a]pyrene factors and appropriate multiplication.

A wide variety of information sources was used in the compilation of this inventory. Emission factor data was obtained from research reports, journal articles and contact with industry. The activity data was mainly obtained from statistical digests, the National Atmospheric Emissions Inventory (NAEI) and contact with industry. Other sources of information on the source sectors include Best Available Technology (BAT) reports and Process Guidance (PG) Notes.

2.2 UNCERTAINTY

The estimates of emissions are open to a certain amount of uncertainty. Illustrating this uncertainty can serve an important function in helping to prioritise future work to improve the inventory. Some of the important factors leading to uncertainty are:

- uncertainties in the measurements used to derive emission factors;
- uncertainties in applying emission factors, which may be from a single or small number of measurements or sources, across a whole industry sector;
- for some sectors little is known about the level of abatement applied across the industry and how this compares to the plants at which measurements have been made;
- for some sectors it is very difficult to quantify production / consumption leading to uncertainty in the activity statistics.

In this report three ways of illustrating the uncertainty have been used. These are described below.

2.2.1 Data quality rating

The data quality rating system used in this study is that in the EMEP/CORINAIR Guidebook (1998). Emission estimates are given a data quality from A to E according to the following criteria :

A An estimate based on a large number of measurements made at a large number of sites fully representative of the sector.

B An estimate based on a large number of measurements made at a large number of sites representative of a large part of the sector.

C An estimate based on a large number of measurements made at a small number of representative sites, or an engineering calculation based on a number of relevant facts.

D An estimate based on a single measurement or an engineering calculation derived from a number of relevant facts and assumptions.

E An estimate based on an engineering calculation derived only from assumptions.

In addition, data quality F has been introduced for this study :

F Where estimates have been made by scaling to a profile, the data quality F has been assigned. These estimates are not necessarily less certain that data quality E, but the F category distinguishes those estimates made through use of profiles.

2.2.2 Range of emissions

In the appendices for each source sector a maximum and minimum value for the emission factor was applied by taking the highest and lowest of the more reliable of the reported values found for each compound. This gives a range of values to use in analysing the uncertainty. However, in some cases the emission factor used is taken from a single report and therefore the uncertainty range used in the analysis is based on the expert opinion of the authors.

2.2.3 Uncertainty analysis

More detailed uncertainty analysis can be carried out to demonstrate where within the ranges of emission estimates the actual emission is most likely to be.

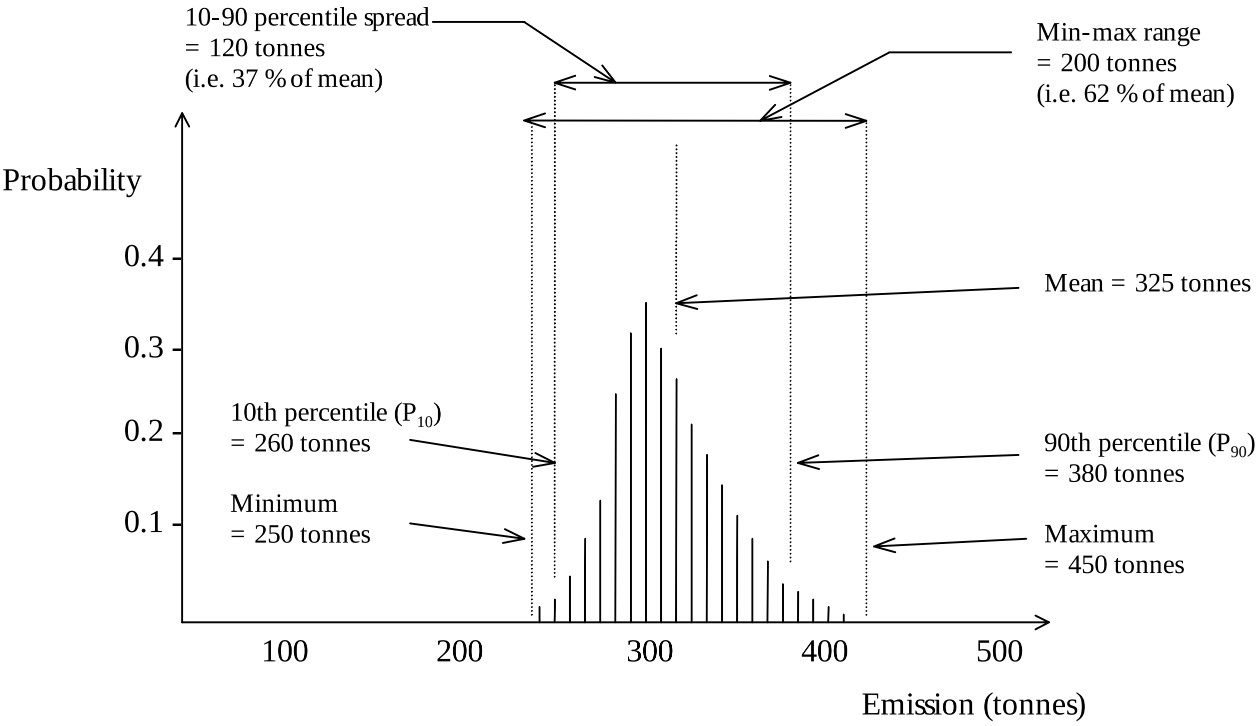
To carry out the sensitivity analysis, each of the 16 PAHs was examined separately. The contribution to the ‘best’ estimate of the emission of each compound from all of the source sectors was calculated as a percentage. Those sources which contributed 90 % of the emissions of each pollutant were subjected to uncertainty analysis, the ‘best’ estimate being deemed suitable for the remaining sources. The results of this analysis are presented in Appendix 12.

The uncertainty analysis was carried out by randomly sampling a set of input data from emission factor and activity statistic ranges and generating an estimate using these values. The estimate generated in this way is one possible forecast from an infinite number of possibilities. By repeating the sampling hundreds, thousands or even tens of thousands of times, a profile can be built up of the probability of a given emission occurring. This analysis was done using Palisade @Risk software in conjunction with Microsoft Excel 5.

As better data become available we would recommend that the analysis should be repeated to judge improvements in the inventories.

The results of the uncertainty analysis are shown graphically. In addition, a number of statistical parameters are given. The minimum, maximum and mean values are given for the emission of each compound. It should be noted that the mean will not necessarily be equal to the 'best' estimate. As an additional measure of dispersion, the 10th and 90th percentiles (P_{10} and P_{90}) are given. These values can be useful since distributions may be highly peaked with the majority of values covering a much smaller range than that between the minimum and maximum values. Figure 2.1 shows an annotated example output of the sensitivity analysis, indicating the statistical parameters discussed in Appendix 12.

Figure 2.1 - Annotated example output of sensitivity analysis for a 1990 emission inventory



3. Results

3.1 INVENTORY OF PAHS (1995)

The speciated PAH emissions inventory for 1995 is summarised in Table 3.1. All the emissions are shown in tonnes. This shows that naphthalene and phenanthrene contribute the largest mass to the total emission of PAHs; however they are not considered to be the most serious problem. It also shows that anode baking for the aluminium industry makes up by far the largest source of emissions in 1995. Anode baking is shown to be a significant source of all the PAHs studied.

The discussion of the results is split into three sections. The first covers benzo[a]pyrene only due to EPAQS' interest as an Air Quality Standard. The second section covers the four PAHs identified as indicator compounds in the UNECE POPs Protocol. The third section contains a general discussion on the remaining 12 PAHs.

Table 3.1 - Summary of Emissions (tonnes) of PAHs in UK in 1995

	Coke production	Electricity Supply Industry - sum	Industrial comb. - coal	Industrial comb. - wood	Domestic comb. - coal	Domestic comb. - wood	Iron and Steel Industry - sum	Aluminium prod. - pre-baked process	Aluminium prod. - VSS process	Anode baking	Waste Incin.	Wood treatment	Natural fires	Vehicles petrol	Vehicles diesel	Total
Naphthalene	53	1.0	389	9.3	181	81	13		19		0.19	18	72	48	9.5	
Acenaphthylene	4.5	0.021	23	7.87	10	71	1.1	0.022	2.1		0	5.0	4.6	11	2.41	
Acenaphthene	0.92	0.058	17	0.31	7.7	2.8	0.23		7		0	17	3.7	6.3	1.29	
Fluorene	3.1	0.31	51	0.84	24	7.5	0.78	0.0051	12		0	23	0.69	13	2.11	
Anthracene	2.7	0.10	5.8	0.65	2.7	5.9	0.66		15		0.032	13	0.52	5.5	1.02	
Phenanthrene	11	0.81	21	2.46	9.6	22	2.7		64		0.40	20	4.6	13	2.87	
Fluoranthene	4.8	0.41	9.5	0.69	4.4	6.2	1.2	0.20	32		0.48	2.6	1.5	8.0	1.6	
Pyrene	3.2	0.064	9.4	0.73	4.4	6.6	0.81	0.33	18		0.48	2.6	1.1	7.7	1.69	
Benz[a]anthracene	1.4	0.020	5.6	0.50	2.6	4.5	0.36	0.050	2.6		0.014	0.29	0.67	6.3	1.02	
Chrysene	0.48	0.093	5.3	0.38	2.4	3.4	0.12	0.071	6.6		0.062	0.67	0.68	6.9	1.1	
Benzo[b]fluoranthene	1.3	0.13	0.22	0.15	0.10	1.4	0.33		2.8		0.010	0.030	1.9	5.2	0.84	
Benzo[k]fluoranthene	0.67	0.057	0.073	0.051	0.034	0.45	0.17		1.4		0.010	0.030	0.92	5.5	0.91	
Benzo[a]pyrene	1.1	0.025	4.9	0.13	2.3	1.2	0.34	0.023	1.3		0.0023	0.059	3.0	5.7	0.95	
Dibenz[ah]anthracene	0.12	0.0026	5.6	0.0020	2.6	0.02	0.031	0.0045	0.5		0.011	0.030	0.059	1.1	0.17	
Indeno[123cd]pyrene	0.48	0.045	3.8	0.0089	1.7	0.08	0.12	0.012	0.7		0	0.030	1.1	1.0	0.14	
Benzo[ghi]perylene	0.46	0.052	2.6	0.10	1.20	0.92	0.11	0.011	0.7		0	0.055	0.58	19	3.18	
Total	89	3.2	552	24	257	215	22	0.72	185		1.7	103	97	163	31	

Refer to Appendices for details of references and data quality ratings

3.1.1 Benzo[a]pyrene

The major sources of benzo[a]pyrene emissions and their contribution to overall emissions are shown in Table 3.2. The source sectors shaded in grey make up 90 % of the emissions and are therefore subjected to uncertainty analysis (Appendix 12).

Table 3.2 - Sources of benzo[a]pyrene emissions (1995)

Benzo[a]pyrene Emissions	Emission (tonnes)	% Contribution to total emissions from sources in this table
Anode baking	15	
Industrial combustion - coal	4.9	
Natural fires	3	
Vehicles - leaded petrol	3	
Vehicles - unleaded petrol (no catalyst)	2.6	
Domestic combustion - bituminous coal	1.9	
Aluminium production - VSS process	1.3	
Domestic combustion - wood	1.2	
Coke Production	1.1	
Vehicles - diesel	0.95	
Domestic combustion - manufactured smokeless coal	0.36	
Iron and steel - sinter plant	0.26	
Industrial combustion - wood	0.13	
Vehicles (unleaded petrol with catalyst)	0.1	
Iron and steel - electric arc furnaces	0.075	
Wood treatment	0.059	
Domestic combustion - anthracite	0.048	
Aluminium production - pre-baked process	0.023	
Electricity supply industry - oil	0.019	
Electricity supply industry - coal	0.0064	
MSW incineration	0.002	
Clinical waste incineration	0.00018	
Sewage sludge incineration	0.000056	
Chemical waste incineration	0.00004	
Crematoria	0.0000044	
Total Emission (tonnes)	**37.06**	

It can be seen that anode baking was the largest source of benzo[a]pyrene in the UK in 1995. Emissions from this source are predicted to fall, however, because of improved abatement being applied to plant. Collectively, vehicles are the next biggest source of benzo[a]pyrene, contributing just over 20 % of the total emissions. Unserviced vehicles, whose emissions are much higher than recently serviced ones, are an important source of these emissions.

3.1.2 PAHs within the UNECE POPs Protocol

The largest sources of the four PAHs identified as indicator compounds in the UNECE POPs Protocol are shown separately and collectively in Table 3.3

Table 3.3 - Contributions of source sectors to total emission of each of the 4 UNECE POPs Protocol PAHs and the sum of the 4 PAHs (1995)

	B[a]P	B[b]F	B[k]F	I[123cd]P	Sum (tonnes)	%
Anode baking	40.47%	67.63%	57.24%	45.68%	69.9	54.66%
Vehicles - leaded petrol	8.09%	5.71%	11.07%	3.30%	9.17	7.17%
Industrial combustion - coal	13.22%	0.46%	0.28%	21.97%	8.993	7.03%
Vehicles - unleaded petrol (no catalyst)	7.02%	4.86%	9.54%	2.31%	7.8	6.10%
Natural fires	8.09%	4.02%	3.51%	6.36%	6.92	5.41%
Aluminium production - VSS process	3.51%	5.92%	5.34%	4.05%	6.2	4.85%
Coke Production	2.97%	2.75%	2.56%	2.78%	3.55	2.78%
Domestic combustion - bituminous coal	5.13%	0.18%	0.11%	8.10%	3.412	2.67%
Domestic combustion - wood	3.24%	2.96%	1.72%	0.46%	3.129	2.45%
Vehicles – diesel	2.10%	1.44%	2.79%	0.69%	2.84	1.81%
Iron and steel – sinter plant	0.70%	0.70%	0.65%	0.69%	0.88	0.69%
Domestic combustion - manufactured smokeless coal	0.97%	0.03%	0.02%	1.62%	0.6614	0.52%
Vehicles (unleaded petrol with catalyst)	0.27%	0.34%	0.42%	0.19%	0.403	0.32%
Industrial combustion - wood	0.35%	0.32%	0.19%	0.05%	0.3399	0.27%
Wood treatment	0.16%	0.06%	0.11%	0.17%	0.149	0.12%
Electricity supply industry - oil	0.05%	0.17%	0.06%	0.17%	0.146	0.11%
Electricity supply industry - coal	0.02%	0.11%	0.16%	0.08%	0.1134	0.09%
Domestic combustion - anthracite	0.13%	0.00%	0.00%	0.21%	0.08791	0.07%
Iron and steel – electric arc furnaces	0.20%	0.00%	0.00%	0.00%	0.075	0.06%
Aluminium production - pre-baked process	0.06%	0.00%	0.00%	0.07%	0.035	0.03%
MSW incineration	0.01%	0.02%	0.03%	0.00%	0.0202	0.02%
Clinical waste incineration	0.00%	0.00%	0.00%	0.00%	0.00182	0.00%
Sewage sludge incineration	0.00%	0.00%	0.00%	0.00%	0.000556	0.00%
Chemical waste incineration	0.00%	0.00%	0.00%	0.00%	0.00042	0.00%
Crematoria	0.00%	0.00%	0.00%	0.00%	4.4E-06	0.00%
Total Emission (tonnes)	**37.06**	**47.32**	**26.21**	**17.29**	**127.88**	

Again for this group of PAHs, anode baking is by far the largest source contributing almost 55 % of emissions. Collectively, vehicles are the second largest source contribution nearly 18 % of the total emissions. Other major sources are industrial coal combustion, natural fires, and primary aluminium production by the Vertical Stud Soderburg (VSS) process.

3.1.3 Total of 16 PAHs

As has been stated, anode baking is the largest source of PAH emissions in the UK, contributing 54 % of the total emissions of all PAHs. It is also the largest single source of the emissions of each of the individual PAHs studied, contributing between 37 % and 80 % of the emissions of all except 3 of the 16 US EPA PAHs. These are naphthalene where industrial coal combustion (35 %) is the largest source, acenapthylene where domestic wood combustion (42 %) is the largest source and benzo(ghi)perylene where vehicles (62 %) is the largest source.

Other major sources of all the PAHs are industrial and domestic coal combustion and domestic wood combustion. Collectively vehicles are a significant source of many PAHs with cars burning leaded petrol being the largest subgroup, both because of higher emission factors and a larger number of vehicle miles being driven. Leaded petrol is however rapidly falling off as a source of PAH emissions as its use is phased out. The vehicles miles is likely to mostly transfer to unleaded petrol.

Wood treatment is a significant source of some of the lighter PAHs such as acenapthene, fluorene and anthracene. The emissions from wood treatment might be higher than the estimates suggest as domestic use, where no abatement techniques are employed, has not been estimated separately due to a lack of data. Emissions from bitumen production and use have not been estimated due to a lack of data. It is likely that bitumen use is a significant source of benzo[a]pyrene and other PAHs.

3.2 UNCERTAINTY

The work to develop the PAH inventory has shown that there are significant uncertainties for the emission sources which are currently included. Sectors for which improvements in the inventory are a high priority are:
- coke production (US data suggest the emission could be higher than the UK estimate);
- vehicles (particularly diesel vehicles and unserviced vehicles);
- primary aluminium production (US data suggest the emission could be higher than the UK estimate);
- anode baking;
- creosote use (particularly domestic use);
- domestic and industrial combustion (particularly the activity data for wood combustion);
- natural fires and open agricultural burning.

An uncertainty analysis was carried out for each of the PAHs studied as described in Section 2.2.2 and 2.2.3. The results are presented in Appendix 12.

It is likely that the current PAH inventory in this report has covered the main sources in the UK. However, for some emission sources estimates have not been made because of the lack of reliable data. Although these are likely to be minor sources, this introduces additional uncertainty.

4. Conclusions

The main sources in the UK of PAH emissions are :

- anode baking
- vehicles
- industrial combustion of coal

These are the main sources for both the sum of the 16 PAHs designated by the US EPA and the sum of those identified in the UNECE POPs protocol.

The majority of anode baking in the UK is carried out for the primary aluminium industry. It is estimated that PAH emissions from this source have significantly decreased since 1995 because of improved abatement measures at anode baking plant.

The key issues related to PAH emissions from vehicles are likely to be the significant emissions from poorly maintained vehicles and emissions during cold starting. Emission estimates for vehicles are uncertain because most of the available emission data has been produced from tests on serviced vehicles, which are not representative of the UK vehicle fleet. The current estimated PAH emissions from diesel vehicles are particularly uncertain.

Analysis of the uncertainties in the PAH emission estimates shows that the current inventory has a high uncertainty and further work is required to improve the reliability of the estimates. However, the PAH inventory has allowed the identification and prioritisation of the likely main emission sources in the UK.

It is likely that the PAH inventory has covered the main sources in the UK. However, for some emission sources estimates have not been made because of the lack of reliable data. This introduces additional uncertainty. Bitumen use is likely to be a significant source of benzo[a]pyrene and other PAHs. Emissions from other sources for which estimates have not been made are likely to be minor.

Further research should concentrate on the main emission sources. In particular, improvements in the quality of emission factors for anode baking and vehicles (particularly diesel and unserviced vehicles) are required. In addition to these, improvements to the inventory are required for the following areas :

- coke production;
- primary aluminium production;
- creosote use;
- open burning;
- other domestic and industrial combustion;
- bitumen use.

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Appendices

CONTENTS

Appendix 1
Coke Production

KEY POINTS

- There are many potential sources of PAH emissions from coke oven plant. The dominant emissions sources are leakages from coke oven doors and charge hole lids, and emissions from charging operations.

- The fugitive nature of the majority of the emissions from coke production means that emissions will greatly vary between different coke production plant.

- Key emission control measures for coke production involve the effective maintenance of the plant and the effective management of operations. However, techniques for control of fugitive emissions vary in efficiency.

- Limited data are available on PAH emissions from coke production and there is considerable uncertainty associated with the estimated emission factors. The ranges in emission factors developed from the limited data that are available are very wide and illustrate the uncertainty.

SECTOR / PROCESS DESCRIPTION

Coke is principally used as a reducing agent in iron-making processes. The production of coke involves the heating of coal to temperatures as high as 1100^{o}C in the absence of oxygen. The main aim of this is to distil out tars and light oils and other compounds.

The manufacture of smokeless fuel by coking has been included in this section because of the similarities in the process to coke production for the UK plant.

The usual method of coking is by the gravity technique. The coal blend is charged into the oven chambers through a series of charge holes in the top of the oven. During heating the raw gas is extracted from the chambers through pipes, cooled with water sprays in a collecting main and processed in a gas purification and recovery plant. After 15-20 hours of heating the coke is pushed and usually quenched with water.

Several stages of the coke production process are potential PAH sources. These include :
- emissions from charging operations;
- leakages from coke oven doors and charge hole lids during carbonisation;
- coke oven gas, extracted during the carbonisation process;

- emissions from pushing operations;
- emissions from quenching operations;
- other sources, such as dust from coal crushing and transfer.

The fugitive nature of the majority of the emissions from coke production means that emissions will greatly vary between different coke production plants. The dominant emission sources are leakages from coke oven doors and charge hole lids, and emissions from charging operations.

Control of PAH Emissions

All coke manufacturing plants in the UK are regulated under IPC (Guidance Note S2 1.06). The pollutants for which achievable emission limits are defined include total PAHs, benzo[a]pyrene and tar fume.

Key emission control measures for coke production involve the effective maintenance of the plant and the effective management of operations. However, techniques for control of fugitive emissions vary in efficiency. The age and state of coking plants are very important with respect to emissions, as well as the abatement methods employed.

Emissions during charging operations are reduced by effective control over the rate and sequence of filling the charge holes combined with the practice of charging when the oven is connected to the coke oven gas collection main.

During carbonisation, leakages via coke oven doors and charge hole lids can be reduced by maintaining the sealing surfaces on the doors and frames, lids and rims in clean condition. Oven doors can become distorted by heat and mechanical stress, and although new oven door designs to reduce distortion are available, these are not suitable to all UK plants.

The coke oven gas, produced during carbonisation, is passed through heat regenerators to heat incoming combustion air, and the flue gas from this passes to atmosphere via a stack.

Emissions from pushing operations are increased if the charge has not been fully carbonised. Effective combustion control therefore reduces this emission. The most effective control during coke pushing operations involves fume collection via hoods over the coke collector car and the coke guide/pusher car.

Dust emissions from coal stockpiles can be reduced by water sprays; emissions from transfer conveyors reduced by covering / protecting these against the wind; and emissions from grinding processes by enclosure.

ACTIVITY

The quantity of coke produced in the UK has fallen from 7.5 Mt in 1990 to 6.2 Mt in 1995 (DTI 1996). In addition, it is estimated that approximately 0.4 Mt smokeless fuel was manufactured by coking in 1995.

EMISSIONS

The emission factors in Table A1.1 for benzo[a]pyrene are based on the EMEP/CORINAIR Guidebook (1998), Radian Corporation (1995) and British Steel (1998), TNO (1995) and Parma et al (1995).

Table A1.1 - Estimated PAH Emissions from coke production in the UK

PAH	**Emission factors**			**Emission (1)**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	g/t(2)	g/t(2)	g/t(2)	tonnes	
Naphthalene	0.55	369.4	7.99	52.7	F
Acenaphthylene	0.05	31.5	0.68	4.5	F
Acenaphthene	0.01	6.4	0.14	0.9	F
Fluorene	0.03	21.8	0.47	3.1	F
Anthracene	0.03	18.6	0.40	2.7	F
Phenanthrene	0.11	75.3	1.63	10.7	F
Fluoranthene	0.05	33.3	0.72	4.8	F
Pyrene	0.03	22.7	0.49	3.2	F
Benz[a]anthracene	0.01	10.0	0.22	1.4	F
Chrysene	0.01	3.4	0.07	0.5	F
Benzo[b]fluoranthene	0.01	9.1	0.20	1.3	F
Benzo[k]fluoranthene	0.01	4.7	0.10	0.7	F
Benzo[a]pyrene	0.011	7.4	0.16	1.1	E
Dibenz[ah]anthracene	0.001	0.9	0.02	0.1	F
Indeno[123cd]pyrene	0.01	3.4	0.07	0.5	F
Benzo[ghi]perylene	0.005	3.2	0.07	0.5	F
(1) Assumed activity = 6.6 Mt/year					

The emission factors in the document by Radian Corporation (1995) are based on calculations only. They are mainly for plant with no control. Radian Corporation do, however, include emission factors from all the potential sources of emission at coke production plant. These emission factors form the upper bound of the range, for all 16 PAHs in Table A1.1.

The technical paper to the TNO (1995) OSPARCOM Inventory, gives emission factors for the Borneff 6. These are much lower than the emission factors from Radian Corporation (1995). The emission factors represent some control such as improved operations and gas purification (washing).

The minimum emission factors for the 10 other PAHs have been estimated by scaling the minimum emission factor for benzo[a]pyrene using the profiles from Radian Corporation.

A limited amount of older information is also available on benzo[a]pyrene emissions from coke production from Josis (1989) and Smith (1984) and these emission factors are within the range in Table A1.1.

The emission factor range in Table A1.1 is very wide. In view of the fact that the upper bound emission factors represent plant with no control, the default emission factors for UK plant are likely to be nearer to the lower end of the range. Operational practices, for example improved maintenance of oven doors and charge hole lids, have been improved at UK plant.

The ‘best’ estimate emission factor for benzo[a]pyrene, 0.16 g/t in this report is taken from the EMEP/CORINAIR Guidebook (EMEP/CORINAIR 1998). The ‘best’ emission factors for other PAHs have been estimated by scaling the benzo[a]pyrene emission factor to the profile from Radian Corporation (1995) and assigning a data quality of F.

UNCERTAINTIES

The fugitive nature of the majority of the emissions from coke production means that emissions will greatly vary between different coke production plant and that the emission estimates, which are based only on the limited available data, are uncertain.

FURTHER WORK

There is a need to improve the reliability of the PAH emission inventory for this sector, which could be a significant source. In addition, the emission estimates for heavy metals need to be improved to confirm that coke production is not a significant source of these pollutants.

Appendix 2
Electricity Supply Sector

KEY POINTS

- This section covers combustion in coal-fired and oil-fired power plant.

- The large amounts of fuel burned in UK power stations make them a potentially significant source of PAHs. However, the efficient combustion conditions, as well as the increase of gas-fired plant, means that the electrical supply sector is responsible for only a minor contribution to total emissions of PAHs. Limited emission factor data are available.

SECTOR / PROCESS DESCRIPTION

Emissions of PAHs from power plant mainly depend on the combustion conditions and the fuel used. Coal has been the most important energy source in the UK, although the use of gas as a fuel is rapidly increasing. Oil-fired plant generally only operate at peak times. No data are available on emissions of PAHs from gas-fired plant, but these emissions are considered to be negligible.

In 1990 UK coal-fired power stations had particulate control technology such as electrostatic precipitators (ESPs). This would have reduced some PAH emissions. Oil-fired power stations generally did not have this control in 1990, except for the orimulsion plant.

Apart from particulate controls, the emphasis of emission control measures taken at power stations has been to reduce SO_2 and NO_x emissions, by flue gas desulphurisation and low NO_x burners, respectively.

ACTIVITY

The consumption of coal in UK power stations decreased from 83 Mt in 1990 to about 58 Mt in 1995. The quantity of oil burned in power stations also decreased, from 6.7 Mt in 1990 to 4.0 Mt in 1995 (DTI 1996).

EMISSIONS

Limited data are available on PAH emissions from power stations, especially for oil-fired plant.

The emission factors for coal-fired plant are taken from data obtained from NETCEN's TOMPs programme, and supported by data in Masclet et al. (1987), TNO (1995) and Parma et al. (1995). Some US data are also available, but the European data are likely to be more representative.

The emission factors for oil-fired power plant are taken from the TNO (1995) technical paper to the OSPARCOM Inventory. Only data for the Borneff 6 PAHs are available in the TNO report. This data is likely to be more representative of UK oil-fired plant than the only other data that has been obtained (Masclet et al. (1987) and Parma et al. (1995)). Limited profile data are available to estimate emissions of PAHs other than the Borneff 6 from oil-fired plant.

Table A2.1 - Estimated PAH Emissions from Power Stations in the UK

PAH	**Coal**			**Oil**		
	Emission factor	**Emission**	**Data quality**	**Emission factor**	**Emission**	**Data quality**
Unit	mg/t	tonnes		mg/t	tonnes	
Naphthalene	18	1.04	D		no data	
Acenaphthylene	0.37	0.02	D		no data	
Acenaphthene	1	0.06	D		no data	
Fluorene	5.3	0.31	D		no data	
Anthracene	1.8	0.10	D		no data	
Phenanthrene	14	0.81	D		no data	
Fluoranthene	2	0.12	D	73.5	0.29	D
Pyrene	1.1	0.06	D		no data	
Benz[a]anthracene	0.34	0.02	D		no data	
Chrysene	1.6	0.09	D		no data	
Benzo[b]fluoranthene	0.9	0.05	D	20.3	0.08	D
Benzo[k]fluoranthene	0.7	0.04	D	4	0.02	D
Benzo[a]pyrene	0.11	0.006	D	4.7	0.02	D
Dibenz[ah]anthracene	0.04	0.003	F		no data	
Indeno[123cd]pyrene	0.25	0.01	D	7.6	0.03	D
Benzo[ghi]perylene	0.2	0.01	D	10.2	0.04	D

(1) Assumed activity (1995)
- coal - 57.9 Mt
- oil - 4.0 Mt
(2) mg per tonne of coal or oil burned
(3) estimated range in emission factors for b[a]p from coal-fired power stations = 0.004 - 0.31 mg/t.

FURTHER WORK

A review and update of emission factors is required, but is of low priority compared with other sources. The POPs work by the power sector related to the Chemical Release Inventory should be taken into account.

Potential ranges in emission factors need to be determined so that uncertainties can be assessed.

Appendix 3
Industrial Combustion

KEY POINTS

- This sector covers industrial combustion of coal and wood.
- PAH emissions are likely to vary significantly between UK industrial combustion plant, depending on combustion conditions, the type of furnace / boiler and on any abatement technology fitted.
- The wide range in emission factors from industrial combustion shows the uncertainty in the estimated emissions.
- It is likely that the majority of industrial wood combustion plant employ only limited control.
- There is considerable uncertainty regarding the quantity of wood burned in the UK.

SECTOR / PROCESS DESCRIPTION

Emissions of PAHs from industrial combustion depend on the combustion conditions, fuel type, the type of furnace / boiler and on any abatement technology fitted. Therefore a large variation in emissions between different plant is expected as plant size, combustion conditions and emission control technology will vary.

Emissions of PAHs from wood combustion are likely to be higher if the wood has been treated with creosote.

Control of PAH emissions

Industrial wood combustion plant in the UK are generally much smaller than industrial coal combustion plant. PAH emissions are likely to vary significantly between UK industrial combustion plant, although it is likely that wood combustion plant employ only limited emission control technology.

ACTIVITY

The total quantity of industrial coal burned in the UK has decreased from about 8 Mt in 1990 to 6.3 Mt in 1995 (DTI 1996).

There is considerable uncertainty regarding the quantity of wood burned in the UK. The quantity of wood burned in industrial and commercial plant in the UK has been estimated to be constant at 0.2 Mt/year since at least 1990 (DTI 1996). There is a potential for industrial wood combustion to increase because of the landfill tax causing combustion to be a preferred option for disposal of waste wood.

EMISSIONS

Emission factors in Table A3.1 for industrial coal combustion are based on Radian Corporation (1995) and TNO (1995), supported by data in Smith (1984) and Parma et al (1995). The upper bound emission factors are equivalent to the ‘best’ estimates for domestic combustion of bituminous coal, assumed to represent industrial coal combustion plant with limited emission control.

Emission factors in the Table A3.1 are uncertain as no studies give data for all 16 PAHs. Therefore some estimations have been made using PAH profiles from Radian Corporation (1995).

Default (‘best’) emission factors have been estimated by taking the mid range values.

Table A3.1 - Estimated PAH Emissions from Industrial Coal Combustion in the UK

PAH	**Emission factor**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	mg/t	mg/t	mg/t	tonnes	
Naphthalene	71.78	123335	61703	388.73	F
Acenaphthylene	1.37	7154	3578	22.54	F
Acenaphthene	7.22	5237	2622	16.52	F
Fluorene	1.49	16209	8105	51.06	F
Anthracene	1.80	1848	925	5.83	D
Phenanthrene	8.40	6558	3283	20.68	D
Fluoranthene	24.00	2981	1502	9.47	D
Pyrene	1.90	2981	1491	9.40	D
Benz[a]anthracene	0.42	1788	894	5.63	D
Chrysene	1.10	1669	835	5.26	D
Benzo[b]fluoranthene	0.24	70	35	0.22	D
Benzo[k]fluoranthene	0.24	23	12	0.07	D
Benzo[a]pyrene	0.14	1550	775	4.88	D
Dibenz[ah]anthracene	2.66	1788	896	5.64	D
Indeno[123cd]pyrene	0.62	1192	596	3.76	D
Benzo[ghi]perylene	0.04	816	408	2.57	D
Assumed activity (1995) = 6.3 Mt					

Table A3.2 - PAH Emissions from Industrial Wood Combustion in the UK

PAH	**Emission factors**			**Emission**	**Data Quality**
	Minimum	Maximum	Best Estimate (default)		
Unit	mg/t	mg/t	mg/t	tonnes	
Naphthalene	2800	90030	46600	9.31	D
Acenaphthylene	90	78600	39300	7.87	D
Acenaphthene	10	3100	1600	0.31	D
Fluorene	30	8300	4200	0.84	D
Anthracene	30	6500	3300	0.65	D
Phenanthrene	230	24400	12300	2.46	D
Fluoranthene	40	6900	3400	0.69	D
Pyrene	30	7300	3700	0.73	D
Benz[a]anthracene	10	5000	2500	0.50	D
Chrysene	10	3800	1900	0.38	D
Benzo[b]fluoranthene	1	1500	800	0.15	F
Benzo[k]fluoranthene	10	0500	300	0.05	F
Benzo[a]pyrene	2	1300	650	0.13	D
Dibenz[ah]anthracene	1	20	10	0.00	F
Indeno[123cd]pyrene	2	90	40	0.01	F
Benzo[ghi]perylene	4	1000	500	0.10	D
(1) Assumed activity = 0.2 Mt/year					

The lower bound figures for the range of PAH emission factors from industrial combustion of wood in Table A3.2 are taken from US data for industrial wood-burning plant with end-of-pipe emission control technology (Radian Corporation 1995). The upper bound emission factors are the default emission factors developed in this study for domestic wood combustion, assumed to broadly represent industrial plant with poor combustion and no emission control. Emission factor data from TNO (1995), Smith (1984) and Ramdahl et al. (1983) have also been taken into account.

The default ('best') emission factors for UK plant have been taken as the middle of the emission factor range in Table A3.2. The width of the emission factor ranges shows the significant uncertainty with the estimated emissions.

UNCERTAINTY

Limited measurement data are available.

There is high uncertainty in the activity data for industrial wood combustion.

FURTHER WORK

Further work is needed on improving the PAH emission estimates from industrial plant.

Improvements to the activity data for wood combustion are required.

Appendix 4
Domestic Combustion

KEY POINTS

- PAH emissions are likely to be significant during the domestic combustion of coal and wood because of the poor combustion conditions.

- The PAH profiles in this report for wood and coal, differ considerably. However, the emission factors are uncertain and need to be verified by measurements. PAH emissions from the domestic combustion of wood are generally expected to be higher than those from bituminous coal combustion because less efficient combustion is likely.

- Emission factors from domestic combustion of bituminous coal are higher than manufactured smokeless fuel and emission factors from combustion of anthracite are lower.

- For many PAHs, data on emissions from domestic combustion are limited and data quality ratings for the emission estimates are D, E or F.

SECTOR / PROCESS DESCRIPTION

The combustion conditions for domestic burning of coal and wood are usually poor because of low flame temperatures with insufficient oxygen at the combustion surface. Emissions of PAHs are therefore important from this sector.

Emissions of PAHs from wood combustion are likely to be higher if the wood has been treated with creosote.

Control of PAH Emissions

PAH emissions from domestic combustion can be reduced by measures such as a change of fuel to gas or anthracite and other smokeless fuels, and to some extent by replacing open fires with stoves in which combustion conditions are improved. There are no emission limits on domestic combustion in the UK, although the burning of wood and coals is only allowed in smoke-reducing appliances in ‘smoke control areas’ under the Clean Air Act 1993.

ACTIVITY

The total quantity of domestic coal burned in the UK has decreased significantly from about 6 Mt in 1990 to 3.8 Mt in 1995 (DTI 1996). This has been largely due to the reduction in burning of bituminous coal (1.2 Mt in 1995); there has been a less significant reduction in burning of anthracite (1.5 Mt) and manufactured smokeless fuels (1.1 Mt).

Limited information is available on the quantity of domestic wood burned in the UK, which is estimated to have remained constant (0.9 Mt) since at least 1990 (DTI 1996).

The large majority of domestic burning is carried out in open appliances (some with back boilers).

EMISSIONS

Tables A4.1 to A4.3 give emission estimates for domestic coal combustion for bituminous coal, anthracite and manufactured smokeless fuel. Emission factors from combustion of bituminous coal are higher than manufactured smokeless fuel and emission factors from combustion of anthracite are lower.

The emission factors for bituminous coal are based on data from CRE (1992), Radian Corporation (1995) and Smith (1984), supported as appropriate by data from TNO (1995), Grimmer et al. (1983) and Ramdahl et al. (1983). For PAHs where limited data are available, profile data from Daisey et al. (1986) and Radian Corporation (1995) have been used to estimate emission factors, which have been given data quality F.

Limited data are available on PAH emissions from the domestic combustion of anthracite and manufactured smokeless fuels. The emission factors have therefore been estimated by multiplying the speciated emission factors for bituminous coal by the appropriate ratios of emission factors for total PAHs from APARG (1995). The speciated measurement data that are available for 8 PAHs from anthracite combustion (Radian Corporation 1995) are of the same order as the estimated emission factors in Table A4.2.

Table A4.4 gives PAH emission estimates from the domestic combustion of wood. The emission factors are expected to be higher than those from bituminous coal combustion. The emission factors for wood combustion are based on data from Radian Corporation (1995) and Smith (1984) with supporting data from Cooper (1980) and Ramdahl et al. (1983) used where appropriate. For PAHs where limited data are available, profile data from Daisey et al. (1986) and Khalili et al. (1995) have been used to estimate emission factors, which have been given data quality F. It should be noted that there is particular uncertainty over the emission factor for dibenz[ah]anthracene.

No account has been taken of the potential different emissions between combustion in stoves and open fireplaces. However, this uncertainty is less significant than the uncertainty in the emission factors.

Table A4.1 - Estimated PAH Emissions from Domestic Coal Combustion in the UK (Bituminous coal)

PAH	**Emission factors**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	g/t	g/t	g/t	tonnes	
Naphthalene	40	207	123.34	148.0	D
Acenaphthylene	2.3	12	7.15	8.6	F
Acenaphthene	1.7	8.8	5.24	6.3	F
Fluorene	5.2	27	16.21	19.5	D
Anthracene	0.6	3.1	1.85	2.2	D
Phenanthrene	2.1	11	6.56	7.9	D
Fluoranthene	1.0	5.0	2.98	3.6	D
Pyrene	1.0	5.0	2.98	3.6	D
Benz[a]anthracene	0.6	3.0	1.79	2.1	D
Chrysene	0.5	2.8	1.67	2.0	D
Benzo[b]fluoranthene	0.02	0.1	0.07	0.1	F
Benzo[k]fluoranthene	0.01	0.04	0.02	0.0	F
Benzo[a]pyrene	0.5	2.6	1.55	1.9	D
Dibenz[ah]anthracene	0.6	3.0	1.79	2.1	D
Indeno[123cd]pyrene	0.4	2.0	1.19	1.4	F
Benzo[ghi]perylene	0.3	1.4	0.82	1.0	D
(1) assumed activity = 1.2 Mt bituminous coal burned per year					

Table A4.2 - Estimated PAH Emissions from Domestic Coal Combustion in the UK (Anthracite)

PAH	**Emission factor**	**Emission**	**Data Quality**
Unit	g/t	tonnes	
Naphthalene	2.55	3.82	F
Acenaphthylene	0.15	0.22	F
Acenaphthene	0.11	0.16	F
Fluorene	0.33	0.50	F
Anthracene	0.04	0.06	F
Phenanthrene	0.14	0.20	F
Fluoranthene	0.06	0.09	F
Pyrene	0.06	0.09	F
Benz[a]anthracene	0.04	0.06	F
Chrysene	0.03	0.05	F
Benzo[b]fluoranthene	0.00	0.00	F
Benzo[k]fluoranthene	0.00	0.00	F
Benzo[a]pyrene	0.03	0.05	F
Dibenz[ah]anthracene	0.04	0.06	F
Indeno[123cd]pyrene	0.02	0.04	F
Benzo[ghi]perylene	0.02	0.03	F
(1) assumed activity = 1.5 Mt anthracite burned per year			

Table A4.3 - Estimated PAH Emissions from Domestic Coal Combustion in the UK (Manufactured smokeless coal)

PAH	**Emission factor**	**Emission**	**Data quality**
Unit	g/t	tonnes	
Naphthalene	26.23	28.86	F
Acenaphthylene	1.52	1.67	F
Acenaphthene	1.11	1.23	F
Fluorene	3.45	3.79	F
Anthracene	0.39	0.43	F
Phenanthrene	1.39	1.53	F
Fluoranthene	0.63	0.70	F
Pyrene	0.63	0.70	F
Benz[a]anthracene	0.38	0.42	F
Chrysene	0.36	0.39	F
Benzo[b]fluoranthene	0.01	0.02	F
Benzo[k]fluoranthene	0.00	0.01	F
Benzo[a]pyrene	0.33	0.36	F
Dibenz[ah]anthracene	0.38	0.42	F
Indeno[123cd]pyrene	0.25	0.28	F
Benzo[ghi]perylene	0.17	0.19	F
(1) assumed activity = 1.1 Mt manufactured smokeless fuel burned per year			

Table A4.4 - Estimated PAH Emissions from Domestic Wood Combustion in the UK

PAH	**Emission Factor Range**		**Emission factor**	**Emission (1)**	**Data Quality**
	Minimum	Maximum	Best Estimate		
Unit	g/t	g/t	g/t	tonnes	
Naphthalene	37	144	90.3	81.3	E
Acenaphthylene	51	106	78.6	70.7	E
Acenaphthene	1.3	5.0	3.1	2.8	E
Fluorene	4.7	12	8.3	7.5	E
Anthracene	6.0	7.0	6.5	5.9	E
Phenanthrene	9.8	39	24.4	22.0	E
Fluoranthene	3.7	10	6.9	6.2	E
Pyrene	2.6	12	7.3	6.6	E
Benz[a]anthracene	0.1	10	5.0	4.5	E
Chrysene	1.6	6.0	3.8	3.4	E
Benzo[b]fluoranthene	0.004	3.0	1.5	1.4	F
Benzo[k]fluoranthene	0.005	1.0	0.5	0.5	F
Benzo[a]pyrene	0.6	2.0	1.3	1.2	E
Dibenz[ah]anthracene	0.009	0.03	0.02	0.02	F
Indeno[123cd]pyrene	0.04	0.13	0.09	0.08	F
Benzo[ghi]perylene	0.04	2.0	1.0	0.9	E
(1) assumed activity = 0.9 Mt wood burned per year					

UNCERTAINTY

There is high uncertainty in the activity data for domestic wood combustion.

There is significant uncertainty in emission factors; the profiles for bituminous coal and wood differ considerably, but emission factors need to be verified.

FURTHER WORK

Improvements to the activity data for wood combustion are required.

Emission factors for PAHs are of low data quality and need improvement, through measurement.

Appendix 5
Iron and Steel Industry

KEY POINTS

- This sector covers sinter plant and electric arc furnaces in the iron and steel industry. Although the emission estimates are very uncertain, PAH emissions are unlikely to be significant; sinter plant and electric arc furnaces make a minor contribution (~1.5%) to the estimated total emissions of benzo[a]pyrene. No information is available on emissions from other processes in the iron and steel industry.

- Coke production is covered in Appendix 1.

- Further work is needed to quantify emissions from secondary steelmaking and casting operations.

SECTOR / PROCESS DESCRIPTION

Sinter plant

Sinter plants are an essential part of the iron-making process. The common practice of charging iron ore to the blast furnace requires a uniform size grading of the charge materials. Iron ore is therefore crushed and graded to 10-50 mm and the undersized fraction is often agglomerated by the sinter process. Sintering allows the use of fine materials, derived from coke, sinter and a large percentage of ore. It is an important pre-treatment as it improves the gas permeability of the blast furnace burden.

Sintering is carried out on a horizontal travelling grate (strand) with, for example, dimensions 3-5m wide by 40-100m long. The iron ore is mixed with coke breeze, limestone, sinter fines and water, and charged to the entry end of the strand. The strand is a chain of pallets moving continuously above a series of wind boxes, through which combustion air is continuously drawn from the ignition hood above. The grate travels forward, the flame front moves down through the bed fusing the mixture into a clinker until combustion is complete at the end of the strand. Combustion is maintained at a temperature of approximately 1500°C. Following combustion the pallets overturn and the sinter is discharged and then graded by screening before or after cooling.

As well as the treatment of ore, sintering plants serve for recycling of mill-scale and dusts produced in steel plants.

Off-gases from sinter processes are collected and passed through electrostatic precipitators (ESPs), which were fitted by 1990. There is potential for some fugitive emissions from operations such as material transfer, but gas collection equipment is used and the collected gases passed to ESPs.

Blast Furnaces

The blast furnace for iron production consists of a refractory lined steel shaft into which the charge is fed from the top and heated air (800-1000^{o}C) is blown from the bottom. The combustion of coke reduces the iron oxide to iron and provides the heat to melt this iron. Limestone and other fluxes are also added.

Electric arc furnaces

The electric arc furnace allows accurate control of the temperature and the time of the reaction for the production of the desired alloy composition. Considerable amounts of mild steel as well as high alloy steels are produced in electric arc furnaces. The close control of the reaction that can be achieved means that low grade scrap can be refined to meet the close specifications of the various steel alloys. A typical charge to an electric-arc furnace is mainly made up of cold scrap material (e.g. automobile scrap).

Other material is added depending upon the required composition of steel. Lime is usually a basic addition but others may include sand, fluorspar, iron ore, carbon, pig iron and other alloying elements.

Current is applied through electrodes and the charge is melted. Elements such as Silicon, Manganese, Carbon, etc. are oxidised from the metal and form slag which is separated. In the next stage, the reducing period, oxygen is removed from the metal through its reaction with carbon. Finally, alloying additions are made to bring the alloy within the required specification.

Basic Oxygen Steelmaking Furnaces

The basic oxygen steelmaking (BOS) furnace or ‘converter’ has in the past been a standard melting furnace for producing grey iron, but is now used as the main steel making process. Hot metal is the principal material used in the BOS process. The converter is tilted and charged, first with scrap, and then with molten iron. High purity oxygen is blown into the metal at high pressure. Lime is added as a flux to help carry off oxidised impurities as a floating layer of slag. After the steel has been refined, the converter is tilted and the steel tapped.

Emissions from charging and tapping operations at blast furnaces, electric arc and BOS furnaces are likely to be significant. Secondary extraction systems are therefore used. For many plant, off-gases are recycled as fuel and gas cleaning equipment is fitted to remove particulates from these gases.

Control of PAH emissions

Since 1990 improvements in emission control have been implemented at iron and steel works. These improvements include improved collection of fugitive emissions from charging and tapping operations, as well as improved particulate control of the collected gases.

ACTIVITY

The number of sinter plant in the UK has decreased since the 1980s. British Steel is the only company employing sinter plants. There are in total 5 sinter plants operating at the 4 UK integrated iron and steel sites. In 1995 the total quantity of sinter produced was about 15.6 Mt (British Steel 1998).

In 1995 a total of 12.2 Mt of iron was produced in UK blast furnaces, and a total of 17.5 Mt of crude steel was produced in the UK (NAEI 1998). The majority of crude steel is produced in basic oxygen steelmaking furnaces (13.0 Mt in 1995), the remainder mostly produced in electric arc furnaces (4.4 Mt in 1995), and a small amount produced in electric induction furnaces (0.1 Mt).

EMISSIONS

Estimated emissions for sinter plant and electric arc furnaces are given in the Table A5.1 and A5.2 respectively. No information is available on emissions form blast furnaces, BOS furnaces and secondary steelmaking and casting operations.

Sinter Plant

No data on emissions factors for sinter plant are available except for Ramdahl et al. (1983) for benzo[a]pyrene only. This emission factor is unreliable as the process details are unknown. It is assumed that the PAH profile from sinter plant is similar to that from coke production, and the emission factors for the other 15 PAHs have been estimated by scaling the emission factor for benzo[a]pyrene relative to those estimates mentioned in this study for coke production.

Table A5.1 - Estimated PAH Emissions from Sinter Plant in the UK

PAH	Emission factor	Emission	Data quality
Unit	g/t	tonne	
Naphthalene	0.85	13.21	F
Acenaphthylene	0.07	1.12	F
Acenaphthene	0.01	0.23	F
Fluorene	0.05	0.78	F
Anthracene	0.04	0.66	F
Phenanthrene	0.17	2.69	F
Fluoranthene	0.08	1.19	F
Pyrene	0.05	0.81	F
Benz[a]anthracene	0.02	0.36	F
Chrysene	0.01	0.12	F
Benzo[b]fluoranthene	0.02	0.33	F
Benzo[k]fluoranthene	0.01	0.17	F
Benzo[a]pyrene	0.017	0.26	E
Dibenz[ah]anthracene	0.002	0.03	F
Indeno[123cd]pyrene	0.01	0.12	F
Benzo[ghi]perylene	0.01	0.11	F
Assumed activity (1995) = 15.6 Mt			

Electric Arc Furnaces

No data on emissions factors for electric arc furnaces are available except for EMEP/CORINAIR Guidebook (1998) for benzo[a]pyrene only. This emission factor in the Guidebook is based on information from the Czech Republic and the appropriateness of this emission factor to UK plant is doubtful. Estimates of emissions of other PAHs have not been made because of the uncertainty in the benzo[a]pyrene estimate.

Table A5.2 - Estimated PAH Emissions from Electric Arc Furnaces in the UK

PAH	Emission factor	Emission	Data quality
Unit	g/t	tonne	
Naphthalene			
Acenaphthylene			
Acenaphthene			
Fluorene			
Anthracene			
Phenanthrene			
Fluoranthene			
Pyrene			
Benz[a]anthracene			
Chrysene			
Benzo[b]fluoranthene			
Benzo[k]fluoranthene			
Benzo[a]pyrene	0.017	0.07	E
Dibenz[ah]anthracene			
Indeno[123cd]pyrene			
Benzo[ghi]perylene			
Assumed activity (1995) = 4.4 Mt			

UNCERTAINTIES

Limited data are available on PAH emissions form iron and steel processes. The few emission estimates that have been made in this study are very uncertain.

FURTHER WORK

Improvements to PAH emission inventories for the iron and steel sector are required. In particular, further work is needed to quantify emissions from secondary steelmaking and casting operations. Activity statistics for casting operations are uncertain.

Appendix 6
Non-Ferrous Metal Production

KEY POINTS

- This sector covers primary aluminium production by the pre-baked process and the Vertical Stud Soderberg (VSS) process, and the production of anodes. No information is available on PAH emissions from other non-ferrous metal production.

- The production of primary aluminium is a potential source of significant PAHs emissions because of the use of carbon anodes.

- About 95 % of the primary aluminium produced in the UK is by the pre-baked anode process. The remainder is produced by the Vertical Stud Soderberg (VSS) production process. Anode production takes place at Lynemouth and Anglesey.

- Limited data are available on PAH emission factors from the aluminium production and anode production and there is considerable uncertainty in the emission inventory. The current estimates indicate that emissions from anode production are likely to be more significant than from aluminium production. Improved controls at the two anode production plant in the UK have reduced emissions since 1995.

- The emission factors form the VSS process are high, but production at the one UK plant is relatively small and the plant is likely to shut in 1999.

SECTOR / PROCESS DESCRIPTION

Emissions of PAHs from non-ferrous metal production in the UK are the most important for primary aluminium production and the production of anodes.

The other main non-ferrous metal industries in the UK are:
- secondary aluminium production,
- secondary copper production,
- primary lead production,
- secondary lead production, and
- primary zinc production.

However, no information is available on PAH emission from these processes. Only primary aluminium production and anode baking are discussed in this section.

Primary Aluminium Production

The production of primary aluminium has been widely reported to be a potential source of PAHs because of the use of carbon anodes. The majority of PAH emissions from primary aluminium production are likely to arise from the electrolysis process, although emissions from anode production are likely to be more significant.

There are two main types of process for the production of primary aluminium : the pre-baked anode process and the Soderberg process (Horizontal Stud Soderberg (HSS) or Vertical Stud Soderberg (VSS).

The main difference between the pre-baked and Soderberg processes is the method of preparation of the anode, which is made up of pitch and petroleum coke. In the Soderberg process the electrode paste is poured periodically into a steel shell, which is immersed in the electrolyte bath and baked by the heat of the reduction cell. The heat drives off the lower boiling point organic compounds and the new anode material is baked to the old anode. In this way the anode is continually replenished.

For the pre-baked process the anode is manufactured prior to placing it in the reduction cell.

Anode Production

For aluminium production by the ‘pre-baked anode’ process, the anode is manufactured prior to placing it in the reduction cell. The anode is produced from anthracite coal and petroleum coke with pitch as binder. This involves calcination followed by baking for about 21 days in a reducing and neutral atmosphere at high temperatures (1300-1400^{o}C). As a result of this process high boiling point organic compounds are emitted. A similar process is used to produce anodes for electric arc furnaces - one of the main steel-making processes.

Amongst the gases released during anode baking are PAH fumes, arising as products of combustion of natural gas and as coal tar pitch volatiles. As for coke production, significant PAH emissions are likely to be fugitive in nature.

The main PAH emission sources in the anode production process are :
- storage and handling of coal tar pitch;
- preparation operations (paste mixing, forming of electrode material etc.);
- leaks from baking ovens;
- extracted flue gas from baking ovens;
- charging and pushing operations.

The walls and floor (cathodes) of the aluminium production cells are imported.

Control of PAH Emissions

Primary Aluminium Production

The UK plants which use pre-baked electrodes remove fumes from the electrolysis process and these are passed through a dry scrubbing system with alumina feedstock. The main aim of this scrubber is to remove gaseous fluorides, and the alumina entrained by the cleaned gas is then removed by fabric filter. This gas-cleaning process, in place by 1990, therefore should also remove a high proportion of PAHs.

Releases from the electrolysis cells can be reduced by ensuring the process is kept as closed as possible and by effective operation and maintenance of the cell fume collection system.

At the Kinlochleven VSS plant, waste gases from the vertical stud Soderberg cells pass to a wet scrubber and ESP for removal of tar fumes and particulates.

Primary aluminium production in England and Wales is covered by IPC regulations.

Anode Production

PAH emissions from anode production are controlled under IPC regulations (Process Guidance Note IPR 6/2). These regulations include limits on emissions of total particulate matter, VOCs and benzo[a]pyrene.

Measures to reduce PAH emissions include improved maintenance of the fume extraction system from the baking ovens; improved operation, for example ensuring a negative pressure in the baking ovens so that the tar fumes are drawn back into the combustion zone; and end-of-process gas-cleaning techniques. In addition, the reduction in fugitive emissions from storage and handling of raw materials, by enclosure and dust extraction, is important.

Only limited emission controls were in place at the anode production plants in 1990. In 1997 scrubber gas-cleaning technology was fitted and operational improvements were made to reduce emissions from the anode production plant at Lynemouth.

ACTIVITY

Primary Aluminium Production

There are four primary aluminium production smelters in the UK. The three largest plants, at Anglesey (working capacity 145,000 tonnes/year), Lynemouth (working capacity 65,000 tonnes/year) and Lochaber (working capacity 40,000 tonnes/year) involve the pre-baked process, whereas the plant at Kinlochleven, with a capacity of only 11,000 tonnes/year is a vertical stud Soderberg process. At Lynemouth the total capacity is 130,000 tonnes/year, but there has been no production at one of the two pot lines since 1991. The VSS plant at Kinlochleven is running at about half capacity and is likely to shut in 1999, for environmental reasons (Alcan 1998)

The total quantity of primary aluminium produced in the UK has decreased from 290 Kt in 1990 to 229 Kt in 1995. The upgraded 60 Kt capacity production line at Lynemouth is likely to start production in 1999 (Alcan 1998), and total production will increase to 310 Kt**.**

Anode Production

Anode production is carried out at the Lynemouth and Anglesey aluminium production sites. In 1995, approximately 144 Kt of anode was produces at these two sites (Alcan 1998).

Anodes are also produced for electric arc furnaces. No data are available on the quantity produced, but it is estimated that the quantity is less than for primary aluminium production.

EMISSIONS

Limited data are available on PAH emission factors from aluminium production. Most available information is on emission factors from the horizontal stud Soderberg (HSS) production process which is not used in the UK.

Aluminium production - Pre-baked Process

The report from Radian Corporation (1995) gives emission factor data 10 of the 16 US EPA PAHs from aluminium production using the pre-baked anode process. The data is for a process using a dry scrubber gas cleaning system, which is similar to the emission controls used at the 3 plant in the UK. The Radian Corporation (1995) report gives much data from the HSS aluminium production process. However, this US data is significantly higher than data from TNO (1995) for the OSPARCOM inventory. The US data has been taken as the maximum estimates. For benzo[a]pyrene, the minimum and ‘best’ estimates have been taken from Alcan (1998) and OSPARCOM (1997) and estimates for other PAHs are scaled to the profile from Radian Corporation (1995).

Table A6.1 - Estimated PAH Emissions from aluminium production using the pre-baked anode process in the UK

PAH	**Emission factors**			**Emission[(1)]**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	g/t	g/t	g/t	tonnes	
Naphthalene			no data		
Acenaphthylene	0.03	8.1	0.09	0.02	F
Acenaphthene			no data		
Fluorene	0.007	1.9	0.02	0.05	F
Anthracene			no data		
Phenanthrene			no data		
Fluoranthene	0.26	74.0	0.9	0.20	F
Pyrene	0.43	124.0	1.4	0.33	F
Benz[a]anthracene	0.07	18.8	0.2	0.05	F
Chrysene	0.09	26.6	0.3	0.07	F
Benzo[b]fluoranthene			no data		
Benzo[k]fluoranthene			no data		
Benzo[a]pyrene	0.03	8.6	0.1	0.02	E
Dibenz[ah]anthracene	0.006	1.7	0.02	0.005	F
Indeno[123cd]pyrene	0.015	4.4	0.05	0.01	F
Benzo[ghi]perylene	0.014	4.1	0.05	0.01	F

(1) Assumed activity = 229 Kt in 1995

Aluminium Production - VSS Process

Limited emission factor data are available for speciated PAH emissions from the VSS process. The emission factor for benzo[a]pyrene in Table A6.2 are based on information form Alcan (1998). The emission factors are significantly larger than those from the pre-baked process. Emission factors for the other 15 PAHs have been estimated by scaling to profile data from Radian Corporation (1995). However, aluminium production from the VSS process makes up only a small proportion of total aluminium production in the UK, and the plant will close by 1999 for environmental reasons. Further work should focus on PAH emissions from anode production and aluminium production by the pre-baked anode process.

Table A6.2 - Estimated PAH Emissions from aluminium production from the VSS process in the UK

PAH	**Emission factors**			**Emission$^{(1)}$**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	g/t	g/t	g/t	tonne	
Naphthalene			2495	18.96	F
Acenaphthylene			277	2.11	F
Acenaphthene			968	7.36	F
Fluorene			1556	11.82	F
Anthracene			2034	15.46	F
Phenanthrene			8359	63.53	F
Fluoranthene			4155	31.58	F
Pyrene			2303	17.50	F
Benz[a]anthracene			340	2.58	F
Chrysene			869	6.61	F
Benzo[b]fluoranthene			369	2.80	F
Benzo[k]fluoranthene			178	1.36	F
Benzo[a]pyrene	148	195	172	1.30	D
Dibenz[ah]anthracene			72	0.55	F
Indeno[123cd]pyrene			91	0.69	F
Benzo[ghi]perylene			96	0.73	F
(1) Assumed activity = 7.6 Kt/year					

Anode Production

Limited data are available on the measurement of PAH emissions from the pre-baking of anodes. However, data on benzo[a]pyrene emissions and total PAH emissions at Lynmouth have been provided by Alcan (1998). The emission factors for the other 15 PAHs have been estimated using data from the HSS process (Radian Corporation 1995) and scaling to the profile relative to benzo[a]pyrene.

Equivalent data have been obtained for the Anglesey anode baking plant through the Environment Agency public register; these values have been obtained for 1996 and 1997 and are assumed the same for 1995 as no major changes have been made to the plant in the interim.

Although the emission factors are uncertain, the more volatile PAHs dominate the profile for anode production, as expected. These PAHs are less dominant in the profile for PAH emissions from aluminium production using the pre-baked process, as expected, because more of the volatiles have already been driven off during anode production.

Table A6.3 - PAH Emissions from Anode Production

PAH	Emission factors			Emission [1]	Data quality
	Minimum	Maximum	Best Estimate		
Unit	g/t	g/t	g/t	tonnes	
Naphthalene			1498	215.8	F
Acenaphthylene			167	24.0	F
Acenaphthene			581	83.7	F
Fluorene			934	134.5	F
Anthracene			1222	175.9	F
Phenanthrene			5020	722.9	F
Fluoranthene			2495	359.3	F
Pyrene			1383	199.2	F
Benz[a]anthracene			204	29.4	F
Chrysene			522	75.2	F
Benzo[b]fluoranthene			222	31.9	F
Benzo[k]fluoranthene			107	15.4	F
Benzo[a]pyrene			103	14.8	D
Dibenz[ah]anthracene			43	6.3	F
Indeno[123cd]pyrene			55	7.9	F
Benzo[ghi]perylene			57	8.3	F
(1) Assumed activity = 144 Kt in 1995					

It should be noted that the emission factor for benzo[a]pyrene from anode production at Lynemouth reduced from 103 g/t in 1995 to 19.7 g/t in 1997 (Alcan 1998) because of improved combustion controls and the introduction of fume treatment plant.

UNCERTAINTIES

There is significant uncertainty in the estimated emission factors and work should be carried out to improve emission estimates for anode baking and aluminium production by the pre-baked process.

No information is available to enable PAH emissions to be estimated form non-ferrous metal plant other than primary aluminium.

FURTHER WORK

Further work should focus on improving emission factors at UK anode production plant and aluminium production plant using the pre-baked process so that the emission inventory is improved.

Further work to improve PAH emission inventories should also focus on other parts of the non-ferrous metal industry, including emission factor development and research on the level of abatement at UK plant.

Appendix 7

Incineration

KEY POINTS

- This appendix covers municipal solid waste incinerators, clinical waste incinerators and other waste incinerators including chemical waste incinerators, sewage sludge incinerators and crematoria.

- The incineration sector is responsible for a minor contribution to total PAH emissions (about 0.005% for benzo[a]pyrene)

SECTOR / PROCESS DESCRIPTION

This appendix covers:

- Municipal Solid Waste Incineration,
- Clinical Waste Incineration,
- Chemical Waste Incineration,
- Sewage Sludge Incinerators, and
- Crematoria

Emissions from all types of incinerators greatly depend on the type of waste, the combustion conditions (e.g. combustion temperature, mixing of waste during combustion and residence time), and the methods of abatement. These factors can vary greatly for incineration plants.

Those incinerators with poor combustion conditions and poor abatement control are a potentially a significant source of emissions of HM/POPs.

Control of PAH Emissions

Municipal Solid Waste Incineration

Emission controls at MSW incinerators in 1990 mainly consisted of only electrostatic precipitators (ESPs), which gave moderate control of PAHs. Combustion control is more important with respect to PAH emissions.

MSW incinerators in England and Wales are regulated under IPC (Process Guidance Note S2 5.01 (1996)). The incinerators were required to comply with emission limits on other pollutants (including PCDD/Fs) by 1 December 1996.

Emissions from MSW incinerators have been reduced since 1990 by improvements in primary and secondary combustion control; installation of end-of-process gas-cleaning technology, such as acid gas scrubbers and improved ESPs or replacing ESPs with fabric filters for better particle control; or, for some plant, by closure. These controls were introduced to UK MSW incinerators between 1994 and 1997.

Clinical Waste Incineration

Major upgrades on clinical waste incinerators were required on many plant to comply with the emission limits at the 1995 or 1996 compliance dates.

The incinerators have reduced their emissions by improvements in combustion control; installation of end-of-process gas-cleaning technology, such as scrubbers and fabric filters; or, for some plant, by closure. All of these have helped to reduce the emissions of PAHs. It is assumed that these controls were introduced between 1994 and 1996.

Chemical Waste Incineration

The potential for emissions of other POPs from chemical waste incinerators has meant that they have held a high profile for several years and emission control was implemented at an earlier stage than for many other incinerators and industrial plants. Plant were fitted with particle emission control and wet scrubber end-of-pipe technology which give some control over PAH emissions.

Sewage Sludge Incineration

Nearly all sewage sludge incinerators are regulated by IPC. The compliance date for meeting IPC emission limits is 1 December 1996. Significant reductions in emissions from sewage sludge incinerators by the upgrading of plant have been made.

Crematoria

Emission controls at many crematoria in 1990 were limited and it is likely that in many cases combustion was poor. The UK crematoria have begun to act on regulations made under the EPA 1990. About 700 crematoria were in operation before 1990, and about 240-270 had been upgraded by 1994 (HMIP 1995). There are significant differences in plant configurations and combustion control between those that have and have not been upgraded.

ACTIVITY

In 1995 the following quantities of waste were incinerated:

- Municipal Solid Waste 2.9 Mt,
- Clinical Waste 0.26 Mt,

• Chemical Waste	0.29 Mt,
• Sewage Sludge	0.08 Mt, and
• Crematoria	440,000 cremations

EMISSIONS

Municipal Solid Waste Incinerators

For Municipal Waste Incinerators emission factors are based on the EMEP/CORINAIR Guidebook (1998) which are derived from Czech data (Parma et al. 1995). Other information is limited. Although the Czech emission factors are unlikely to be representative, emissions are within the expected range.

Clinical Waste Incineration

For Clinical Waste Incinerators emission factors are assumed to be the same as for MSW. This is a source of significant uncertainty.

Chemical Waste Incineration

Emission factors based on Czech data (Parma et al. 1995), the average for 2 plant burning liquid and solid waste. Other information is limited. Although the Czech emission factors are unlikely to be representative, emissions are within the expected range.

Sewage Sludge Incineration

No data are available for Sewage Sludge Incineration. Emission factors are assumed to be same as for MSW, which is a source of uncertainty.

Crematoria

The only data available on Crematoria are from the US (Radian Corporation 1995). Emission factors are lower than expected, particularly for some crematoria in the UK which might not have been upgraded in 1995. Further work required to check US emission factors, but this is not a high priority as the sector is likely to have a low contribution to total emissions.

Table A7.1 - Estimated PAH Emissions from MSW Incinerators in the UK

PAH	**Emission factor**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	mg/t	mg/t	mg/t	tonne	
Naphthalene			51.7	0.15	D
Acenaphthylene				no data	
Acenaphthene				no data	
Fluorene				no data	
Anthracene			9.1	0.03	D
Phenanthrene			110.5	0.32	D
Fluoranthene			145.4	0.42	D
Pyrene			147.4	0.43	D
Benz[a]anthracene			4.2	0.01	D
Chrysene			18.8	0.05	D
Benzo[b]fluoranthene			3.15	0.01	F
Benzo[k]fluoranthene			3.15	0.01	F
Benzo[a]pyrene			0.7	0.002	D
Dibenz[ah]anthracene			3.5	0.01	D
Indeno[123cd]pyrene				no data	
Benzo[ghi]perylene				no data	
Assumed activity (1995) = 2.9 Mt					

Table A7.2 - Estimated PAH Emissions from Clinical Waste Incinerators in the UK

PAH	**Emission factor**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	mg/t	mg/t	mg/t	tonne	
Naphthalene			51.7	0.01	D
Acenaphthylene				no data	
Acenaphthene				no data	
Fluorene				no data	
Anthracene			9.1	0.002	D
Phenanthrene			110.5	0.03	D
Fluoranthene			145.4	0.04	D
Pyrene			147.4	0.04	D
Benz[a]anthracene			4.2	0.001	D
Chrysene			18.8	0.005	D
Benzo[b]fluoranthene			3.15	0.001	F
Benzo[k]fluoranthene			3.15	0.001	F
Benzo[a]pyrene			0.7	0.0002	D
Dibenz[ah]anthracene			3.5	0.001	D
Indeno[123cd]pyrene				no data	
Benzo[ghi]perylene				no data	
Assumed activity (1995) = 0.26 Mt					

Table A7.3 - Estimated PAH Emissions from Chemical Waste Incinerators in the UK

PAH	**Emission factor**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	mg/t	mg/t	mg/t	tonne	
Naphthalene			34.5	0.01	D
Acenaphthylene				no data	
Acenaphthene				no data	
Fluorene				no data	
Anthracene			7.65	0.002	D
Phenanthrene			138.55	0.04	D
Fluoranthene			31.85	0.01	D
Pyrene			22.65	0.01	D
Benz[a]anthracene			0.55	0.0002	D
Chrysene			2.9	0.001	D
Benzo[b]fluoranthene			0.65	0.0002	F
Benzo[k]fluoranthene			0.65	0.0002	F
Benzo[a]pyrene			0.15	0.00004	D
Dibenz[ah]anthracene				no data	
Indeno[123cd]pyrene				no data	
Benzo[ghi]perylene				no data	
Assumed activity (1995) = 0.29 Mt					

Table A7.4 - Estimated PAH Emissions from Sewage Sludge Incinerators in the UK

PAH	**Emission factor**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	mg/t	mg/t	mg/t	tonne	
Naphthalene			51.7	0.004	E
Acenaphthylene				no data	
Acenaphthene				no data	
Fluorene				no data	
Anthracene			9.1	0.001	E
Phenanthrene			110.5	0.01	E
Fluoranthene			145.4	0.01	E
Pyrene			147.4	0.01	E
Benz[a]anthracene			4.2	0.0003	E
Chrysene			18.8	0.002	E
Benzo[b]fluoranthene			3.15	0.0003	F
Benzo[k]fluoranthene			3.15	0.0003	F
Benzo[a]pyrene			0.7	0.0001	E
Dibenz[ah]anthracene			3.5	0.0003	E
Indeno[123cd]pyrene				no data	
Benzo[ghi]perylene				no data	
Assumed activity (1995) = 0.08 Mt					

Table A7.5- Estimated PAH Emissions from Crematoria in the UK

PAH	**Emission factor**			**Emission**	**Data quality**
	Minimum	Maximum	Best Estimate		
Unit	mg/t	mg/t	mg/t	tonne	
Naphthalene			17.7	0.01	E
Acenaphthylene				no data	
Acenaphthene				no data	
Fluorene				no data	
Anthracene			0.093	0.00004	E
Phenanthrene			0.66	0.0003	E
Fluoranthene			0.059	0.00003	E
Pyrene			0.046	0.00002	E
Benz[a]anthracene			0.0038	0.000002	E
Chrysene			0.018	0.00001	E
Benzo[b]fluoranthene				no data	
Benzo[k]fluoranthene				no data	
Benzo[a]pyrene			0.01	0.000004	E
Dibenz[ah]anthracene				no data	
Indeno[123cd]pyrene				no data	
Benzo[ghi]perylene				no data	
Assumed activity (1995) = 440000 cremations					

UNCERTAINTIES

Emission factors for PAHs are uncertain because limited data are available. PAHs are not required to be routinely monitored from incinerators.

There is considerable uncertainty in the quantity of chemical waste incinerated in the UK.

Emission factors for crematoria for PAHs are also uncertain.

FURTHER WORK

Measurements are required to improve the emission factors for PAHs from incinerators and crematoria, although this would be a low priority relative to measurement requirements at many other source sectors.

Further work is required to estimate PAH emissions from the incineration of animal remains. Also, the emissions from the burning out of metal containers have not been estimated. Again, this work should be considered low priority relative to the updating of the PAH inventory for other sources.

Appendix 8
Wood Treatment

KEY POINTS

- PAHs have been considered to be an important pollutant from this sector because of the use of creosote.

- However, emissions of benzo[a]pyrene from wood treatment by creosote have been estimated to be 0.06 tonnes/year, which is a low contribution to total UK emissions. These estimates are uncertain because limited data are available. Estimates suggest that the more volatile PAHs are released in much greater quantities.

- The major PAH emission is likely to occur during the drying stage of industrial timber treatment processes and during domestic use (carbolineum).

- The use of creosote is declining in the UK and further work should first focus on the quantity of creosote used and predicted future use.

- Emissions during creosote production have not been estimated due to lack of information, but are likely to be small relative to emissions from creosote use.

SECTOR / PROCESS DESCRIPTION

Creosote is used for external applications such as transmission poles, railway sleepers, gates and fencing. It is used by industry and domestically. Its strong persistent odour makes it unsuitable for use inside buildings (DoE 1995).

Creosote has the potential to cause PAH emissions during its manufacture and use. The use of creosote is likely to be a more important source of PAH emissions than its production, for which emissions are easier to control. The more volatile PAHs are likely to be released in greater quantities during creosote use and drying.

Creosote is produced by the distillation of coal tar pitch. Creosote has a range of formulations, but it has been reported that PAHs account for 75-85% of its composition (IARC 1984, Wild and Jones 1995, DoE/HMIP 1992).

In the UK wood preservation industry, pressure treatment of timber by the vacuum process is a more common creosote application technique than non-pressure treatments such as immersion (DoE/HMIP 1992). In the vacuum process timber is treated in a pressurised chamber which is flooded with hot creosote for 1-3 hours. After draining, a vacuum is applied to draw off excess creosote and the timber is left to dry in open air.

Control of PAH Emissions

Timber processes are regulated by both IPC and LAAPC, depending on the process and chemicals used. PG Note 6/3 sets atmospheric emission limits for processes using impregnation or immersion methods for timber treatment with organic solvent-based preservatives and creosote.

The major control methods for industrial creosote application involve the reduction of fugitive emissions by improved general management of operations, such as enclosure of chemicals during storage, transport, loading etc., and improved application techniques.

Emissions from domestic application are harder to control, and are unlikely to be reduced unless changes in the type of preservatives used, such as water-based preservatives, are encouraged.

Most creosote production is likely to be covered by IPC, although emissions are easier to control than for application, and this source is likely to be less significant.

Levels of Benzo[a]pyrene in some types of creosote are restricted in the EU to 500 ppm for industrial use (14th amendment to the Marketing and Use Directive - Creosote (96/60/EEC)).

ACTIVITY

The Environment Agency ‘BAT report’ on ‘Pollution control for timber processes’ estimates that approximately 37,000 tonnes of creosote is used for wood treatment annually in the UK and the number of UK plant using creosote is estimated to be only 15 (1995, DoE/HMIP 1992(c)). The BAT report estimates creosote usage to be split into 19,000 tonnes used by industry and 18,000 tonnes used for retail purposes. These figures are uncertain, although the number of creosote applications is widely reported to be declining in the UK and therefore the estimated usage is likely to be an upper bound.

Creosote can be mixed with petroleum fractions to produce carbolineum. This can be brushed onto the surface of the wood and is mainly for private use.

EMISSIONS

Table A8.1 - Estimated PAH Emissions from Creosote Use in the UK

PAH	Emission factors			Emission	Data Quality
Unit	g/t	g/t	g/t	tonnes	
Naphthalene	178.47	791.2	484.8	17.9	F
Acenaphthylene	49.84	221.0	135.4	5.0	F
Acenaphthene	164.59	729.7	447.2	16.5	F
Fluorene	121.13	1108.1	614.6	22.7	F
Anthracene	78.60	648.6	363.6	13.5	F
Phenanthrene	293.13	810.8	552.0	20.4	F
Fluoranthene	97.00	45.9	71.5	2.64	E
Pyrene	87.85	54.1	70.9	2.6	F
Benz[a]anthracene	2.10	13.5	7.8	0.29	F
Chrysene	28.00	8.1	18.1	0.67	F
Benzo[b]fluoranthene	0.25	1.4	0.8	0.03	F
Benzo[k]fluoranthene	0.25	1.4	0.8	0.03	F
Benzo[a]pyrene	0.50	2.7	1.6	0.06	E
Dibenz[ah]anthracene	0.25	1.4	0.8	0.03	F
Indeno[123cd]pyrene	0.25	1.4	0.8	0.03	F
Benzo[ghi]perylene	0.25	2.7	1.5	0.05	E
assumed activity =37,000 tonnes creosote used per year					

It should be noted that there is significant uncertainty associated with the emission factors in Table A8.1.

The emission estimates are based on the EMEP/CORINAIR Guidebook (1998) (minimum estimates) and Gevao and Jones (1997) (maximum estimates). Profile data from IARC (1985) and Radian Corporation (1995) has been used for PAHs where no reliable data are available, a data quality F has been assigned in these cases.

Emission estimates from production of creosote are not included in Table A8.1 due to the lack of data. However, production is likely to be controlled and is likely to be a much less significant source of PAH emissions than the open use of creosote.

In addition, no separate estimates have been made for emissions from carbolineum production or use. Although it is assumed that the emissions from carbolineum use for domestic wood preservation are within the emissions in Table A8.1, the TNO (1995) emission factors for carbolineum are much higher than those from creosote use.

UNCERTAINTIES

The emission factors for the loss of active ingredient from previously treated wood are based on one limited study. Further work on this may greatly affect the scale and rate of decrease of emissions from this source. These emissions from creosote have not been estimated.

Little information is available beyond industry estimates of the extent of treated wood with these products.

The previous emissions before abatement measures were put in place are not well quantified. Hence the effectiveness of abatement and especially of the improvements in housekeeping which are taking place within the industry is unclear.

Limited data on emission factors from creosote use are available. The available data from The Netherlands (TNO 1995, EMEP/CORINAIR 1998), the USA (Radian Corporation 1995) and estimations for the UK in Gevao and Jones (1997) indicate wide ranges in emission factors for several PAHs. The TNO data give no details of process type, control etc. The US data give some emission factors for the application process with condensation control and some emission factors for open drying.

Creosote use in the UK ranges from uncontrolled domestic use to controlled application in industry. However, the major emissions are likely to be from open drying operations at industrial processes and from domestic use.

FURTHER WORK

The emission factors for timber treatment are subject to considerable uncertainty. It would greatly help the estimation of the emissions of PAHs if further work was carried out to assess the scale of these releases. Further work should first focus on the quantity of creosote used.

Appendix 9
Natural Fires and Open Agricultural Burning

KEY POINTS

- This sector includes natural fires (forest and moorland) and open agricultural burning, such as stubble burning.
- Limited emission factor data are available and reliable estimates of the amount of forest and moorland burned are difficult to make.
- Stubble burning has been banned in England and Wales since 1992, but was a significant source of PAHs before then.
- Although the extent to which natural fires can be controlled is limited, reliable emission estimates are useful to put in perspective the contribution to total emissions of other, more controllable sources such as vehicles, industry etc.

SECTOR / PROCESS DESCRIPTION

This section covers accidental and deliberate forest and moorland fires and open agricultural burning. Combustion conditions during these fires are generally poor and uncontrolled and there is therefore a high potential for the emissions of PAHs.

Annual emissions of PAHs from open biomass burning were in the past likely to be high in the UK because of straw/stubble burning practices. However, the emission from this source was greatly reduced in 1992 when stubble burning was banned in England and Wales with limited exceptions.

Other open agricultural burning (e.g. burning of wastes such as plastics) is likely to be a source of PAHs, although no activity data are available and emissions have not been estimated.

Control of PAH Emissions

The only controls over forest and moorland fires are the prevention of the fires and measures to improve extinguishing practices. The main emission control is the banning of stubble burning, which was implemented in 1992 in England and Wales.

ACTIVITY

The number and size of these fires will vary from year to year and the limited activity data that are available are therefore uncertain. It has been estimated that approximately 0.41 Mt of forest / moorland is burned each year (HMIP 1995).

The area of agricultural land burned as stubble in Scotland and Northern Ireland has not been estimated and no data are available on the quantities of other agricultural waste burned.

EMISSIONS

Estimated emissions are given in the Table A9.1 below.

The emission factors in Table A9.1 are based on US data in Jenkins et al. (1996) and Radian Corporation (1995). Although the Radian Corporation data is taken from a much older study, the emission factors in the two reports are a similar order of magnitude.

Information on emissions from natural fires in Ramdahl et al. (1983) and Wild and Jones (1995) was taken into account.

Table A9.1 - Emissions from natural fires in the UK

PAH	**Emission factors**			**Emission [1]**	**Data Quality**
	Minimum	Maximum	Best Estimate		
Unit	g/t	g/t	g/t	tonnes	
Naphthalene	1.3	348.0	174.7	71.61	E
Acenaphthylene	0.3	22.1	11.2	4.59	D
Acenaphthene	0.01	17.9	9.0	3.67	D
Fluorene	0.09	3.3	1.7	0.69	D
Anthracene	0.06	2.5	1.3	0.52	D
Phenanthrene	1.4	20.9	11.2	4.57	D
Fluoranthene	0.6	6.7	3.7	1.50	D
Pyrene	0.6	5.0	2.8	1.15	D
Benz[a]anthracene	0.2	3.1	1.6	0.67	D
Chrysene	0.2	3.1	1.7	0.68	D
Benzo[b]fluoranthene	0.02	9.3	4.7	1.91	D
Benzo[k]fluoranthene	0.3	4.2	2.3	0.92	D
Benzo[a]pyrene	0.2	14.3	7.2	2.96	D
Dibenz[ah]anthracene	0.01	0.3	0.15	0.06	D
Indeno[123cd]pyrene	0.6	4.8	2.7	1.11	D
Benzo[ghi]perylene	0.3	2.5	1.4	0.58	D
(1) Assumed activity = 0.41 Mt forest/moorland burned					

UNCERTAINTIES

The number and size of accidental / deliberate natural forest and moorland fires will vary from year to year and activity data are therefore uncertain.

Emission estimates are of low data quality and no estimates have been made for the stubble burning that continues in Scotland and Northern Ireland.

FURTHER WORK

The inclusion of emission estimates from stubble burning in Scotland and Northern Ireland and other open agricultural burning would reduce the uncertainly in this sector. The collection of information on the extent of emissions from bonfires and their contribution to air quality would be of use.

Appendix 10
Road Transport

KEY POINTS

- Vehicles are an important source of PAHs. Factors such as design and age of the engine, fuel quality, driving mode, oil consumption, cold starts and choking have important effects on the emissions of PAHs from vehicles. The influence of servicing is particularly important.

- The majority of the available emission factor data is from measurements on serviced vehicles. This does not provide a realistic representation of the UK vehicle fleet and emission factors for unserviced vehicles have generally been derived from calculations rather than measurement data.

- The wide ranges in the estimated PAH emission factors show the high uncertainty. The lower end of the ranges represent emission factors from serviced vehicles, whilst the upper end represents unserviced vehicles.

- Emission factors from vehicles fitted with catalysts are significantly lower than those without catalysts. Emission data for diesel vehicles is particularly uncertain.

SECTOR / PROCESS DESCRIPTION

This section covers emissions from road transport in the following categories :
- cars running on leaded petrol;
- cars running on unleaded fuel - no catalytic converter;
- cars running on unleaded fuel - with catalytic converter;
- diesel cars
- diesel light-duty goods vehicles (LGV);
- diesel heavy-duty goods vehicles (HGV)
- diesel buses.

PAH emissions from vehicles arise either from their formation during combustion of the fuel components or from the survival during combustion of PAHs present in the fuel (Doel 1995). Factors such as design and age of the engine, fuel quality, driving mode, oil consumption, cold starts and choking have important effects on the emissions of these compounds. The aromatic (and PAH) content of the fuel has also been widely reported to have a direct influence on PAH emissions.

Control of PAH Emissions

There are several techniques available to reduce vehicle emissions. The key controls on PAH emissions from mobile sources are improved engine maintenance and three-way catalysts. All new gasoline cars are required to have three-way catalysts fitted. A number of other controls and factors will affect PAH emissions, including the replacement of older vehicles by new vehicles with more efficient engines.

ACTIVITY

Emissions were estimated using activity data (total number of km travelled in the UK each year by each vehicle category) based on statistics provided by DETR and used in the NAEI for estimating emissions of other pollutants (Salway et al., 1997).

EMISSIONS

In general, more research has been carried out on emissions of PAHs from vehicles than from other sources, but the majority of the dynamometer emission tests were carried out on serviced vehicles. These tests do not therefore give a true representation of the vehicles on UK roads, a large amount of which are not running in a recently serviced state. Furthermore, some of the measurements were made on vehicle models not representative of those found in the UK.

Petrol vehicles

Tests by Rogge et al. (1993) using the cold-start US Federal Test Procedure were carried out on vehicles in an ‘unserviced’ condition running on leaded petrol. Only particulate phase PAHs were collected. However, all other studies report data on emissions from serviced vehicles. There is a wide variation in measured PAH emissions from serviced and unserviced vehicles. The top of the emission factor range for cars running on leaded petrol in Table A10.1 is based on the results from Rogge et al. (1993), representing unserviced vehicles. However, it has to be noted that most of these were made on old (pre-1976) US models not typically found on UK roads with poor fuel economy. Profile data from Mi et al. (1996) have been used to estimate emission factors for the low molecular weight PAHs not measured by Rogge et al., and a data quality of F has been assigned in these cases. The lower end of the range represents emissions from serviced vehicles and is based on data from Volkswagen (1989), Mi et al. (1996) and Alsberg et al. (1985).

For non-catalyst cars running on unleaded petrol, no data are available on emissions from unserviced cars. The emission factors from Rogge et al. (1993) for unserviced US cars running on leaded petrol have therefore been assumed to represent the top of the emission factor range for non-catalyst cars running on unleaded petrol. The emission factors for serviced vehicles in Table A10.2 are based on data from Radian Corporation (1995), Mi et al. (1996) and Octel (1995). The emission estimates in Table A10.2 are uncertain and might be lower than the real values. This is because emissions of PAHs from vehicles running on unleaded fuel are expected to be higher than from vehicles running on leaded fuel due to the higher aromatic content in unleaded fuel, although this may be offset by the fact that tests using unleaded petrol will have been made on newer models made to tighter emission standards than the tests using leaded petrol.

As expected emission factors for cars fitted with a catalyst are lower than those with no catalyst. The emission factors in Table A10.3 are based on data from Volkswagen (1989) and Radian Corporation (1995). Profile data from Mi et al. (1996) were used for the PAHs where no information was available, and a data quality rating of F was assigned to these estimates.

For all vehicle categories the default emission factor has been taken as the middle of the range of emission factors for serviced and unserviced vehicles.

Diesel vehicles

There is limited information available on emissions of PAHs from diesel vehicles, but emissions will vary with size and load of vehicle. The majority of data which are available are from a series of measurements by Westerholm and coworkers (Westerholm et al. (1991), Westerholm and Li (1994)) carried out on a heavy duty diesel truck over an urban drive cycle. These measurements were assumed to represent the bottom end of a range of PAH emission factors. The emission factors for unserviced, leaded US vehicles (Rogge et al., 1993) were assumed to represent the top end of the range and a default figure for this size of truck at urban speeds was taken as the mean of the lower and upper end of the range.

Emissions of regulated pollutants over the drive cycle examined by Westerholm et al. (1991), especially for particulates, were compatible with the emission factors for a small, rigid-axled HGV at urban speeds used in the NAEI. Therefore, the default PAH emission factor was assigned to this size of diesel HGV and speed and the PAH emission factors for smaller diesel cars and LGVs and larger HGVs and buses were scaled according to the PM_{10} emission factor profile for these other vehicle types and speeds. The variation of PM_{10} emission factors from diesel vehicles of different sizes and average speeds has been well characterised in the NAEI.

Table A10.4 shows the average PAH emission factors for diesel cars, LGVs, HGVs and buses for the UK fleet and journey profile (average speed) of these vehicles in 1995. The calculated UK emission rates (in tonnes) are shown in Table A10.5

Table A10.1 - PAH Emissions from cars running on leaded petrol

PAH	Emission factors			Emission [1]	Data Quality
Unit	Minimum	Maximum	Best Estimate		
	μg/km	μg/km	μg/km	tonnes	
Naphthalene	109	307.0	208.0	26.9	F
Acenaphthylene	7.35	78.0	42.7	5.5	F
Acenaphthene	3.91	41.5	22.7	2.9	F
Fluorene	60.2	68.2	64.2	8.3	F
Anthracene	3.8	41.5	22.6	2.9	F
Phenanthrene	1.87	98.5	50.2	6.5	F
Fluoranthene	14.5	53.1	33.8	4.4	F
Pyrene	10.5	53.1	31.8	4.1	F
Benz[a]anthracene	5.36	45.5	25.4	3.3	F
Chrysene	6.76	49.3	28.0	3.6	F
Benzo[b]fluoranthene	3.57	37.9	20.7	2.7	D
Benzo[k]fluoranthene	3.93	40.7	22.3	2.9	D
Benzo[a]pyrene	2.47	43.5	23.0	3.0	D
Dibenz[ah]anthracene	0.72	8.3	4.5	0.6	D
Indeno[123cd]pyrene	2.4	6.4	4.4	0.6	D
Benzo[ghi]perylene	4.91	145.4	75.2	9.7	D
(1) Activity = 129.3 billion km.					

Table A10.2 - PAH Emissions from cars running on unleaded fuel - no catalytic converter

PAH	Emission factors			Emission [1]	Data Quality
Unit	Minimum	Maximum	Best Estimate		
	μg/km	μg/km	μg/km	tonnes	
Naphthalene	8.1	307.0	157.5	19.0	F
Acenaphthylene	7.91	78.0	43.0	5.2	F
Acenaphthene	10.7	41.5	26.1	3.1	F
Fluorene	1.8	68.2	35.0	4.2	F
Anthracene	0.1	41.5	20.8	2.5	F
Phenanthrene	2.6	98.5	50.6	6.1	F
Fluoranthene	1.4	53.1	27.2	3.3	F
Pyrene	1.4	53.1	27.2	3.3	F
Benz[a]anthracene	1.2	45.5	23.3	2.8	F
Chrysene	1.3	49.3	25.3	3.0	F
Benzo[b]fluoranthene	1	37.9	19.5	2.3	C
Benzo[k]fluoranthene	0.4	40.7	20.6	2.5	C
Benzo[a]pyrene	0.3	43.5	21.9	2.6	C
Dibenz[ah]anthracene	0.4	8.3	4.4	0.5	C
Indeno[123cd]pyrene	0.3	6.4	3.3	0.4	C
Benzo[ghi]perylene	0.6	145.4	73.0	8.8	C
(1) Activity = 120.4 billion km.					

Table A10.3 - PAH Emissions from cars running on unleaded fuel - with catalytic converter

PAH	**Emission factors**			**Emission** (1)	**Data Quality**
Unit	Minimum	Maximum	Best Estimate		
	μg/km	μg/km	μg/km	tonnes	
Naphthalene	16.2	23.5	19.8	2.0	F
Acenaphthylene	1.4	6.0	3.7	0.4	F
Acenaphthene	0.8	3.2	2.0	0.2	F
Fluorene	3.6	5.2	4.4	0.4	F
Anthracene	0.2	0.3	0.2	0.02	F
Phenanthrene	5.2	7.5	6.4	0.6	F
Fluoranthene	2.8	4.1	3.4	0.3	F
Pyrene	3.0	4.1	3.5	0.4	F
Benz[a]anthracene	0.2	3.5	1.8	0.2	F
Chrysene	0.3	3.8	2.0	0.2	F
Benzo[b]fluoranthene	0.3	2.9	1.6	0.2	D
Benzo[k]fluoranthene	0.1	2.0	1.1	0.1	D
Benzo[a]pyrene	0.2	1.9	1.1	0.1	D
Dibenz[ah]anthracene	0.1	0.3	0.2	0.02	F
Indeno[123cd]pyrene	0.2	0.5	0.3	0.03	D
Benzo[ghi]perylene	0.4	4.7	2.6	0.3	D

(1) Activity = 99.8 billion km.

Table A10.4 - Average emission factors for diesel vehicles in 1995

PAH (mg/km)	**Cars**	**LGV**	**HGV**	**Buses**
Naphthalene	22	72	190	254
Acenaphthylene	5.5	18.2	48.4	64.0
Acenaphthene	3.1	9.5	25.8	35.1
Fluorene	4.9	16.0	42.3	55.7
Anthracene	2.5	7.8	20.3	26.8
Phenanthrene	6.5	21.7	57.7	76.4
Fluoranthene	3.7	12.1	31.9	43.3
Pyrene	4.0	12.6	33.9	45.4
Benz[a]anthracene	2.5	7.8	20.3	26.8
Chrysene	2.5	8.2	22.3	28.9
Benzo[b]fluoranthene	1.8	6.5	16.8	22.7
Benzo[k]fluoranthene	2.2	6.9	18.1	24.8
Benzo[a]pyrene	2.2	7.4	19.0	24.8
Dibenz[ah]anthracene	0.3	1.3	3.5	4.1
Indeno[123cd]pyrene	0.3	1.0	2.8	3.7
Benzo[ghi]perylene	7.4	23.8	63.9	84.6

Table A10.5 - UK PAH Emissions from diesel vehicles in 1995

PAH	**Emission from vehicle category:**				**Total Emissions (all diesel vehicles)**	**Data Quality**
	Cars	**LGV**	**HGV**	**Bus**		
	tonnes	tonnes	tonnes	tonnes	tonnes	
Naphthalene	0.71	1.66	5.9	1.23	9.5	F
Acenaphthylene	0.18	0.42	1.5	0.31	2.41	F
Acenaphthene	0.1	0.22	0.8	0.17	1.29	F
Fluorene	0.16	0.37	1.31	0.27	2.11	F
Anthracene	0.08	0.18	0.63	0.13	1.02	F
Phenanthrene	0.21	0.5	1.79	0.37	2.87	F
Fluoranthene	0.12	0.28	0.99	0.21	1.6	F
Pyrene	0.13	0.29	1.05	0.22	1.69	F
Benz(a)anthracene	0.08	0.18	0.63	0.13	1.02	F
Chrysene	0.08	0.19	0.69	0.14	1.1	F
Benzo(b)fluoranthene	0.06	0.15	0.52	0.11	0.84	F
Benzo(k)fluoranthene	0.07	0.16	0.56	0.12	0.91	F
Benzo(a)pyrene	0.07	0.17	0.59	0.12	0.95	F
Dibenz(ah)anthracene	0.01	0.03	0.11	0.02	0.17	F
Indeno(123cd)pyrene	0.01	0.024	0.087	0.018	0.139	F
Benzo(ghi) perylene	0.24	0.55	1.98	0.41	3.18	F

UNCERTAINTY

The majority of dynamometer emission tests for PAHs have been carried out on a limited range of serviced vehicles or old US vehicles. These tests do not give a true representation of the vehicles on UK roads, a large amount of which are not running in a recently serviced state. Hence there is considerable uncertainty in the benzo[a]pyrene inventory for vehicles, particularly for diesel vehicles.

FURTHER WORK

Emissions of PAHs from off-road sources have not been estimated.

Emission estimates of PAHs from vehicles need to be improved. Measurements of emission factors for on-road vehicles and fuel types typical of the UK fleet are required to improve the inventory.

Appendix 11
Other Sources of PAHs

KEY POINTS

The following sectors are not covered in previous appendices and are also potential sources of PAHs

- Industrial oil combustion
- Domestic oil combustion
- Waste oil combustion
- Combustion of waste-derived fuel
- Petroleum processes
- Glass, ceramic and other mineral processes
- Bitumen and asphalt production and use
- Accidental fires
- Bonfires and other incidental fires

When prioritising inventory work, it is important to consider the potential contribution to total emissions (Table A11.1).

Table A11.1 - Potential contribution to total UK atmospheric emissions of PAH

Sources	PAHs
Bitumen and asphalt production and use	M
Accidental fires	M
Bonfires and other incidental fires	M
Industrial oil combustion	L
Domestic oil combustion	L
Waste oil combustion	L
Combustion of waste-derived fuel	L
Petroleum processes	L
Glass, ceramic and other mineral processes	L
H: High potential contribution to total emission M: Medium potential contribution to total emission L: Low potential contribution to total emission	

Considering the potential contribution of PAHs from the above sources, priority can be set for further work in order to improve the PAH inventories (Table A11.2).

Tables A11.2 - Priority for further work to improve HM/POPs inventories

Sources	PAHs
Bitumen and asphalt production and use	H
Accidental fires	H
Waste oil combustion	M
Glass, ceramic and other mineral processes	M
Bonfires and other incidental fires	M
Industrial oil combustion	L
Domestic oil combustion	L
Combustion of waste-derived fuel	L
Petroleum processes	L
H: High priority M: Medium priority L: Low priority	

It should be noted, that there are some other minor sources that have not been covered in the appendices.

Priority for further work to improve PAH inventories is likely to be higher for some of the major sources (Appendices 1-10).

Appendix 12
Uncertainty Analysis of the 1995 PAH Inventory

An uncertainty analysis was carried out for each of the PAHs studied as described in Section 2.2.2 and 2.2.3. The results are shown below in table and graphical form.

It is likely that the current PAH inventory in this report has covered the main sources in the UK. However, for some emission sources estimates have not been made because of the lack of reliable data. Although these are likely to be minor sources, this introduces additional uncertainty. The uncertainty analysis below does not include the additional uncertainty from sources for which emissions have not been estimated.

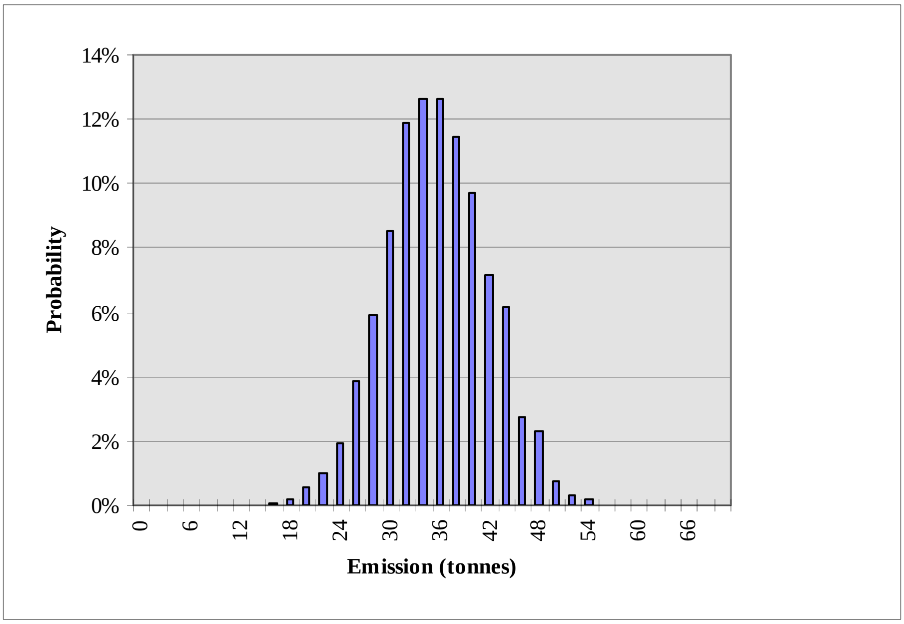
BENZO[A]PYRENE

The results of the uncertainty analysis for benzo[a]pyrene are shown in Table A12.1 and Figure A12.1. The parameters shown in Table A12.1 are shown in the example output in Figure 2.1.

Table A12.1 - Summary of uncertainty analysis for benzo[a]pyrene (1995) (Emissions in tonnes)

Minimum Emission	P_{10}	Mean Emission	P_{90}	Maximum Emission	Min-Max range	% of Mean	10-90% spread	% of Mean
17.71	28.91	36.68	44.82	54.97	37.26	102%	15.91	43%

Figure A12.1 - Graph showing B[a]P uncertainty



The data and the graph show that the uncertainty in the estimate for benzo[a]pyrene emissions is similar to a normal distribution. This fits with the knowledge that a variety of source sectors contribute to this uncertainty, and in the course of the analysis these uncertainties tend to cancel each other out leading to a lower uncertainty than might otherwise be expected. The 10-90 % spread of 43 % suggests that there is a large scope for improvement to both the emission factors and activity statistics of some of the major sources of benzo[a]pyrene emissions.

This graph shows a different picture to the uncertainty in the inventory for 1990 (Wenborn 1998). The reduction in benzo[a]pyrene emissions as a result of the ban on agricultural stubble burning in 1992 and the introduction of improved abatement in the anode baking process for aluminium production has reduced the uncertainty in the inventory.

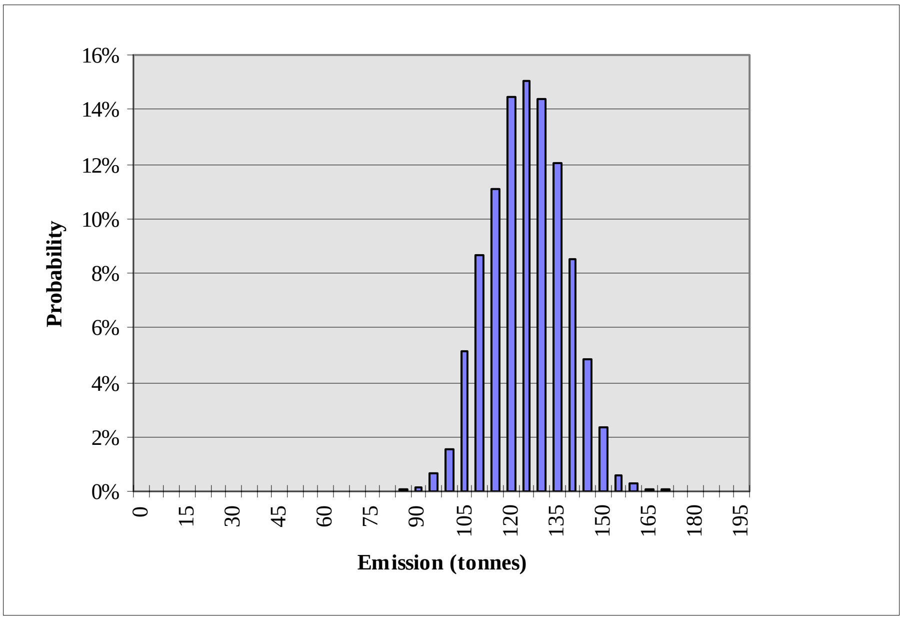
PAHS WITHIN THE UNECE POPS PROTOCOL

The results of the uncertainty analysis for the four PAHs identified as indicator compounds in the UNECE POPs protocol are shown in Table A12.2 and Figure A12.2. The parameters shown in Table A12.2 are shown in the example output in Figure 2.1.

Table A12.2 - Summary of the uncertainty analysis for the UNECE POPs Protocol indicator PAHs (1995)

	Minimum Emission	P10	Mean Emission	P90	Maximum Emission	Range	% of Mean	P90-P10	% of Mean
B[a]P	17.71	28.91	36.68	44.82	54.97	37.26	102%	15.91	43%
B[b]F	25.55	33.84	47.11	60.14	68.08	42.53	90%	26.30	56%
B[k]F	13.55	20.04	26.62	33.33	38.36	24.80	93%	13.29	50%
I[123cd]P	9.032	12.91	17.26	21.61	25.95	16.92	98%	8.694	50%
Total	86.62	111.6	127.7	143.6	170.7	84.11	66%	32.04	25%

Figure A12.2 - Graph showing uncertainty in the sum of the UNECE POPs Protocol indicator PAHs



The uncertainty in each of the PAHs is relatively large. However when the four are combined the large number of sources mean that some of the errors cancel out on average. This makes the figures for the total of the four less uncertain than might be expected.

REMAINING PAHS

The results of the uncertainty analysis for the remaining PAHs not discussed in the previous two sections is shown in Table A12.3. The parameters shown in Table A12.3 are shown in the example output in Figure 2.1.

Table A12.3 - Summary of the uncertainty analysis for the remaining PAHs (1995)

	Minimum Emission	**P10**	**Mean Emission**	**P90**	**Maximum Emission**	**Range**	**% of Mean**	**P90-P10**	**% of Mean**
Naphthalene	461.5	785.9	1119	1445	1753	1291	115%	659.5	59%
Acenaphthylene	101.6	140.9	169.3	198.6	238.6	137.0	81%	57.75	34%
Acenapthene	79.13	112.3	148.4	184.6	218.6	139.5	94%	72.34	49%
Fluorene	136.9	207.1	274.6	342.2	412.8	275.9	100%	135.1	49%
Anthracene	129.0	160.5	231.7	302.2	332.9	203.9	88%	141.6	61%
Phenanthrene	484.4	612.3	901.6	1191	1320	835.3	93%	578.5	64%
Fluoranthene	241.5	289.2	434.0	578.1	628.7	387.3	89%	288.9	67%
Pyrene	144.6	179.2	258.4	337.7	372.9	228.3	88%	158.5	61%
Benzo[a]anthracene	30.35	43.47	56.48	69.15	82.87	52.52	93%	25.68	45%
Chrysene	55.81	74.53	104.7	134.5	154.1	98.31	94%	60.00	57%
Dibenz[ah]anthracene	6.386	11.70	16.74	21.72	26.48	20.10	120%	10.03	60%
Benzo[ghi]perylene	13.51	29.63	40.40	51.44	67.45	53.94	134%	21.82	54%

The magnitude of the uncertainty is significant for all the PAHs, meaning that much improvement is needed in the estimates. Policy decisions should take into account the uncertainty in PAH emission estimates. The results can however be broken into two broad categories which can be illustrated by acenaphthylene at one end and fluoranthene at the other. Acenapthylene has a large number of sources making up 90 % of its emissions giving the uncertainty a shape similar to a normal distribution (Figure A12.3). Fluoranthene on the other hand has two sources which contribute 90 % of its emissions making the distribution much broader and flatter, even though the overall range in uncertainty is similar to that of acenaphthylene (Figure A12.4).

There is considerable uncertainty in all estimates for naphthalene because of the difficulties in the measurement of this PAH.

Figure A12.3 - Graph showing acenaphthylene uncertainty

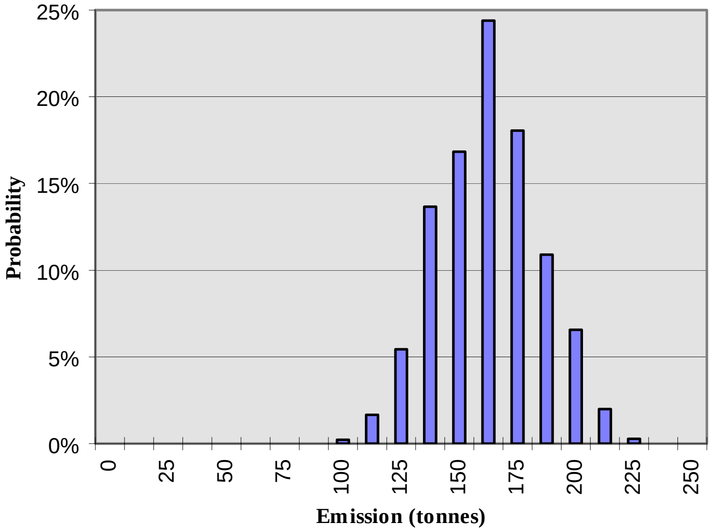


Figure A12.4 - Graph showing fluoranthene uncertainty