

# **Emission estimation and emission factor development work during 2000-2001**

A report produced for the Department of the Environment, Transport and the Regions; the National Assembly of Wales; the Scottish Executive; and the Department of Environment in Northern Ireland

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# Executive Summary

This report has been prepared under the DETR research programme 'Emission Factors and Cost Curves for Air Pollutants' (reference EPG 1/3/134). This programme includes research aimed at improving the estimation methods used in the National Atmospheric Emissions Inventory (NAEI).

A review of the data in the NAEI identified a number of areas where focused research was likely to yield improved data quality:

- VOCs: petrol distribution, solvent use and other emission sources;
- PAHs: off-road vehicles, manufacture and use of tar and bitumen.

Improvements regarding VOCs are:

- an improved methodology for petrol distribution taking into account the effect of emission abatement measures in place;
- improved emission estimates for film coating, textile coating, paper coating and seed oil extraction processes through a survey of information held by regulatory bodies;
- updated solvent consumption estimates from the Solvents Industry Association (SIA);
- revised emission estimates for the coating of metal packaging;
- a revised methodology for painting and printing, leading to improved emission factors and emission estimates;
- a revised estimate of emissions from other solvent usage;
- new data on adhesives usage and solvent consumption in the adhesives sector;
- a revised methodology for estimating emissions from the use of non-aerosol cosmetics and toiletries;
- an improved emission factor for bread making;
- a revised methodology for estimating emissions from sugar production based on limited emission measurements;
- updated data for emissions from the chemicals sector.

Improvements regarding PAHs are:

- newly estimated emissions from off-road vehicles by analogy with equivalent emissions from on-road vehicles;
- a detailed investigation of likely PAH emission sources and first estimation of emissions from tar and bitumen manufacture and use.



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# 1 Introduction

This report has been prepared under the DETR research programme ‘Emission Factors and Cost Curves for Air Pollutants’ (reference EPG 1/3/134). This programme includes research aimed at improving the estimation methods used in the National Atmospheric Emissions Inventory (NAEI).

A review of the data in the NAEI identified a number of areas where focused research was likely to yield improved data quality:

- VOCs: petrol distribution, solvent use;
- PAHs: off-road vehicles, manufacture and use of tar and bitumen.

This report summarises the main findings from each of the above areas (Sections 2 & 3 respectively), with supporting Appendices which contain further details. Recommendations for improved emission factors to use in the NAEI are given in Section 4.



## 2 Improvements to VOC emission factors

Specific objectives regarding VOCs were to:

- improve the methodology for petrol distribution by taking into account the effect of emission abatement measures in place;
- improve emission estimates for film coating, textile coating, paper coating and seed oil extraction processes through a survey of information held by regulatory bodies;
- obtain updated solvent consumption estimates from the Solvents Industry Association (SIA).

New information for some other VOC sources were also identified and recorded for inclusion in the NAEI:

- coating of metal packaging;
- painting and printing;
- other solvent usage;
- adhesives;
- non-aerosol consumer products;
- bread;
- sugar;
- chemicals.

The following sections summarise the work undertaken; further details are given in **Appendix 1**.

### 2.1 PETROL DISTRIBUTION

The existing methodology for petrol distribution did not take account of abatement. Research was therefore aimed at improving the methodology to take account of the increasing use of vapour return systems in accordance with the requirements of the Environmental Protection Act. Relevant trade associations were contacted and asked for data. The Institute of Petroleum (IoP) was able to provide a draft methodology which better reflects current circumstances and which takes into account the changes in petrol volatility and petrol density which have occurred recently; and the extent of and type of abatement systems.

Improved emission estimates have been generated in an Excel spreadsheet so that all assumptions, inputs, outputs and calculations may be verified or amended at a later date. Appendix 1 contains a summary of the revised emission estimates.

## 2.2 FILM COATING, TEXTILE COATING, PAPER COATING, AND SEED OIL EXTRACTION

A number of English local authorities, and SEPA were contacted, and information sought on emissions from film coating, textile coating, paper coating and seed oil extraction processes. Similar contacts made with local authorities in 1997-98 had obtained a lot of information on film coating, textile coating, and seed oil extraction processes. In general, where the earlier survey had shown that emissions from a particular site were relatively small then no further contact was made with the relevant local authority. As a result of the two surveys, emissions estimates were made for the period 1996-1999 in the case of paper coating processes, and the period 1993-1999 in the case of the remaining three sectors. **Table 1** summarises data for 1996 and 1999.

**Table 1 Solvent usage and emission estimates for industrial solvent using processes**

Sector	No. of processes	Data available <sup>1</sup>	Solvent usage (tonnes)		Solvent emission (tonnes)	
			1996	1999	1996	1999
Film coating	36	35	15315	15030	15315	8681
Textile coating	87	86	6780	6769	6780	4284
Paper coating	44	42	14023	12303	8079	3953
Seed oil extraction	5	5	- <sup>2</sup>	- <sup>2</sup>	2897	2354

1 – data available for at least part of time series

2 – Solvent is recycled within the process and usage figures have not been estimated

A default emission factor for seed oil extraction of 1 kg solvent / tonne of oil seeds and nuts processed has been derived. This emission factor has been demonstrated for the period 1993 to 1999, and so may reasonably be applied to earlier years as a default emission factor.

## 2.3 COATING OF METAL PACKAGING

Revised emission estimates have been derived for the years 1993-1999 based on information provided by the Metal Packaging Manufacturers Association (MPMA) and by local authorities. Estimates for 1996 and 1999 are shown in **Table 2**.

**Table 2 Solvent usage and emission estimates for metal packaging processes.**

Sector	No. of processes	Data available <sup>1</sup>	Solvent usage (tonnes)		Solvent emission (tonnes)	
			1996	1999	1996	1999
Metal packaging	41	5	14821	9224	13824	6796

1 – data available for at least part of time series

Emission factors have been estimated using coating sales statistics provided by the British Coatings Federation (BCF).

## 2.4 PAINTING AND PRINTING

Some changes have been made to the methodology used to calculate emissions from the use of paints and inks using new data received from the BCF. Appendix 1 summarises the emission factors and emission estimates for each sector following the methodological changes.

## **2.5 OTHER SOLVENT USAGE**

The emission estimate for this sector has been revised, following the provision of new data by the Solvent Industry Association (SIA). On average, SIA estimates of solvent consumption provided in four triannual surveys are 46 ktonnes higher than independently derived estimates for known source sectors, and this figure is used for as an estimate of solvent consumption in unassigned sectors for all years. Further details are given in Appendix 1.

## **2.6 ADHESIVES**

Some minor changes have been made using an SIA estimate of solvent usage by the sector in 1999, and some additional data on UK adhesives usage taken from the web-site of the European trade association ([www.feica.com](http://www.feica.com)). In view of the nature and variety of adhesives it is not possible to derive a single emission factor.

This sector is a priority for further investigation, with the collection of a full series of consistent activity data a priority.

## **2.7 NON-AEROSOL CONSUMER PRODUCTS**

The existing methodology for non-aerosol consumer products has been modified using value of sales data for cosmetics and toiletries covering the years 1994-1999. These data have been used to derive estimates of the volume of sales of each type of cosmetic or toiletry. Again due to the variety of products covered, it is not useful to develop a default emission factor.

## **2.8 BREAD**

The Institute of Bakers provided an improved emission factor of 1 kg/tonne bread based on measurements at two UK plant bakeries, which may replace the previously employed emission factor of 3 kg/tonne bread which had been based on measurements at bakeries in California.

## **2.9 SUGAR**

A revised emission estimate has been extrapolated from measurements at one sugar beet processing plant - see Appendix 1.

A default emission factor of 0.01895 kg VOC per tonne of sugar produced from sugar beet. Sugar production statistics are available back to at least 1970 from the Annual Abstract of Statistics, so applying the new default emission factor would enable emission estimates to be made for each year relevant to the NAEI.

## **2.10 CHEMICALS**

Updated data have been collected and entered into the NAEI as in previous years:

- from the Environment Agency's Pollution Inventory for England and Wales;
- from SEPA for Scotland;
- directly from plant operators in Scotland and Northern Ireland.

This information relates to sources where individual circumstances makes the development of default emission factors inappropriate.

# 3 Improvements to PAH emission factors

Specific objectives for improving the NAEI estimates of PAH emissions and emission factors were:

- derive emission factors and emissions for off-road vehicles using analogous on-road vehicle data;
- make estimates of PAH emissions and emission factors from the manufacture and use of tar and bitumen.

The work carried out in these areas is summarised below; further details are given in **Appendix 2**.

## 3.1 OFF-ROAD VEHICLES

A review of published literature did not identify documented PAH emission factors for agricultural power units, domestic house and garden appliances, aircraft support vehicles and industrial off road machinery. So that these emissions may be included in the NAEI, the PAH emission factors for each of these sources have been equated to on-road vehicles in urban driving conditions as shown in **Table 3**.

**Table 3: Off-road PAH sources, and on-road sources designated as equivalent**

Off road sector	Designated equivalent on-road source
Agricultural power units (petrol)	LGV non catalyst (petrol)
Agricultural power units (gas oil)	LGV, (derv)
Domestic house and garden (petrol)	Car non catalyst, (petrol)
Domestic house and garden (derv)	LGV, (derv)
Aircraft support (gas oil)	HGV, (derv)
Industry off road (petrol)	LGV non catalyst, (petrol)
Industry off road (gas oil)	LGV, (derv)

Fuel consumption data were available for each of the off-road sectors which allowed total emissions to be calculated.

## 3.2 MANUFACTURE AND USE OF TAR AND BITUMEN

An extensive literature and internet search was undertaken along with communication with experts from industry and trade associations. Emission factors have been derived, these are summarised in **Table 4** – see Appendix 2 for further details.

**Table 4: Emission factors for PAHs from the tar and bitumen industry**

PAH	Pollcode	Emission Factor (kg/Mtonne)
Phenanthrene	34	561.07
Anthracene	33	13.06
Fluoranthene	35	23.44
Pyrene	36	79.28
Chrysene	38	119.10
Benzo( <i>a</i> )anthracene	37	31.89
benzo( <i>k</i> )fluoranthene	40	0.92
Benzo( <i>a</i> )pyrene	41	5.53
Benzo( <i>ghi</i> )perylene	44	9.77

## 4 International exchange of information

International exchange of information is supported as a means of deriving best value for primary research work - where processes operated overseas are essentially the same as those in the UK, default emission factors are equally applicable to both cases.

An important forum for exchange of information is the UNECE / European Environment Agency Emission Inventories Task Force which is co-chaired by the UK.

During the past year, Canada has made available results of work on PAH emissions from wood burning stoves - see **Table 5**.

**Table 5: Emission factors for PAH emissions from wood burning stoves**

Pollutant	Stove Type Emission Factor (kg/tonne)			
	Conventional	Noncatalytic	Catalytic	Pellet
<b>PAH</b>	<b>kg/tonne</b>			
Acenaphthene	0.005	0.005	0.003	ND
Acenaphthylene	0.106	0.016	0.034	ND
Anthracene	0.007	0.0045	0.004	ND
Benzo(a)anthracene	0.01	<0.050	0.012	ND
Benzo(b)fluoroanthene	0.003	0.002	0.002	0.000013
Benzo(g,h,i)fluoroanthene	ND	0.014	0.003	ND
Benzo(k)fluoroanthene	0.001	<0.050	0.001	ND
Benzo(g,h,i)perylene	0.002	0.01	0.001	ND
Benzo(a)pyrene	0.002	0.003	0.002	ND
Benzo(e)pyrene	0.006	0.001	0.002	ND
Biphenyl	ND	0.011	ND	ND
Chrysene	0.006	0.005	0.005	0.0000376
Dibenzo(a,h)anthracene	BM	0.002	0.001	ND
7,12-Dimethylbenz(a)anthracene	ND	0.002	ND	ND
Fluoranthene	0.01	0.004	0.006	0.0000274
Fluorene	0.012	0.007	0.007	ND
Indeno(1,2,3,cd)pyrene	BDL	0.01	0.002	ND
9-Methylanthracene	ND	0.002	ND	ND
12-Methylbenz(a)anthracene	ND	0.001	ND	ND
3-Methylcholanthrene	ND	<0.050	ND	ND
1-Methylphenanthrene	ND	0.015	ND	ND
Naphthalene	0.144	0.072	0.093	ND
Nitronaphthalene	ND	BDL	ND	ND
Perylene	ND	0.001	ND	ND
Phenanthrene	0.039	0.059	0.024	0.0000166

Pollutant	Stove Type Emission Factor (kg/tonne)			
	Conventional	Noncatalytic	Catalytic	Pellet
<b>PAH</b>	<b>kg/tonne</b>			
Phenanthrol	ND	BDL	ND	ND
Phenol	ND	<0.050	ND	ND
Pyrene	0.012	0.004	0.005	0.0000242
<b>PAH Total</b>	<b>0.365</b>	<b>&lt;0.250</b>	<b>0.207</b>	<b>0.000119</b>



## 5 Way forward

It is recommended that the various new methodologies, emission factors and emission estimates described in this report should be incorporated into the NAEI.

Items of further work in the areas covered by this report which would be of value:

### VOCs

**Petrol distribution:** key assumptions should be discussed with UKPIA.

**Textile coating:** development of an improved methodology for calculating emissions during 1970 to 1992.

**Film coating:** development of an improved methodology for calculating emissions during 1970 to 1992.

**Other solvent use:** the methodology adopted should be validated through discussion with the Solvents Industry Association.

**Adhesives:** this sector is a priority for further investigation, with the collection of a full series of consistent activity data a priority.

**Non-aerosol consumer products:** further effort is advised for household products and car-care products, with the emphasis placed on the more important sources such as slow-release air fresheners, toilet blocks, antifreeze, de-icer, and windscreen wash solutions.

**Chemicals:** in future years it is recommended that selected process operators are contacted to try to verify the assumptions made.

### PAHs

**Off-road vehicles:** the assumptions made should be tested by a limited set of emission measurements.

**Tar & bitumen sources:** the assumptions made should be tested by a limited set of emission measurements.

# Appendices

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Appendix 1	Details of new VOC research
Appendix 2	Details of new PAH research

# Appendix 1

## Details of new VOC research

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### PETROL DISTRIBUTION

The existing methodology for petrol distribution did not take account of abatement. Research was therefore aimed at improving the methodology to take account of the increasing use of vapour return systems in accordance with the requirements of the Environmental Protection Act. Relevant trade associations were contacted and asked for data. One of these organisations - the Institute of Petroleum (IoP) - was able to provide a draft methodology for estimation of petrol distribution emissions. This had a number of advantages over the existing methodology:

- it takes account of abatement systems such as vapour return;
- it takes account of changes in petrol volatility;
- it includes emissions from sources such as spillages and working losses from petrol retailer's storage tanks, unlike the previous approach;
- as an industry standard, the methodology draws upon industry knowledge and will, presumably, find support within the industry.

Values for a number of parameters are needed in order to calculate emissions using the method. These are:

- Petrol volatility;
- Petrol density;
- Extent of and type of abatement systems.

Currently, good data are available on petrol densities and overall petrol consumption. Petrol volatilities are available for the period 1970 – 1994, although no more recent values have been obtained. The use of Stage 1 and 2 vapour recovery systems is covered in an IoP retail survey. The proportion of petrol stations with Stage 1 equipment is put at 60%, and UKPIA have suggested that this will probably equate to at least 80% of petrol being distributed through petrol stations with Stage 1 equipment, since a large number of the petrol stations without abatement systems will have small throughputs. The main uncertainties are the

proportion of stations with abatement in earlier years, and the relative proportion of petrol loaded into road tankers using top and bottom loading.

Revised emission estimates are summarised for the years 1990-1999 in **Table A1.1**.

**Table A1.1: Summary of improved VOC emission estimates from petrol distribution**

Emission source	Units	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
Terminals, storage (leaded)	ktonnes	4.1535	3.6608	3.3043	2.9133	2.5041	2.1008	1.8566	1.6166	1.0517	0.5251
Terminals, storage (unleaded)	ktonnes	2.0854	2.4931	2.8465	3.1588	3.325	3.494	3.8477	4.0422	3.8516	3.6795
Terminals, tanker loading (leaded)	ktonnes	11.046	9.835	8.9671	7.8886	6.7355	5.6132	4.9037	4.2253	1.7904	0.2453
Terminals, tanker loading (unleaded)	ktonnes	5.7454	6.8503	7.8001	8.7008	9.0997	9.5019	10.343	10.761	6.5269	1.662
Service stations, loading (leaded)	ktonnes	28.918	25.859	23.679	20.922	18.034	15.174	13.385	11.647	8.7293	1.2212
Service stations, loading (unleaded)	ktonnes	15.042	18.012	20.598	23.076	24.365	25.687	28.233	29.662	31.817	8.2938
Service stations, breathing losses (leaded)	ktonnes	3.911	3.4974	3.2026	2.8296	2.4391	2.0523	1.8103	1.5752	1.1806	0.6997
Service stations, breathing losses (unleaded)	ktonnes	2.0343	2.436	2.7858	3.1209	3.2952	3.474	3.8184	4.0117	4.3031	4.7323
Vehicle refuelling, loading (leaded)	ktonnes	43.495	38.895	35.616	31.469	27.125	22.824	20.133	17.518	13.13	7.7422
Vehicle refuelling, loading (unleaded)	ktonnes	22.624	27.091	30.981	34.709	36.647	38.635	42.465	44.615	47.856	52.366
Vehicle refuelling, drips and spillages (leaded)	ktonnes	2.6073	2.3316	2.135	1.8864	1.626	1.3682	1.2069	1.0501	0.7871	0.4664
Vehicle refuelling, drips and spillages (unleaded)	ktonnes	1.3562	1.624	1.8572	2.0806	2.1968	2.316	2.5456	2.6745	2.8687	3.1549

## FILM COATING, TEXTILE COATING, PAPER COATING, AND SEED OIL EXTRACTION

A number of local authorities, and SEPA were contacted, and information sought on emissions from film coating, textile coating, paper coating and seed oil extraction processes. Similar contacts made with local authorities in 1997–98 had obtained a lot of information on film coating, textile coating, and seed oil extraction processes. In general, where the earlier survey had shown that emissions from a particular site were relatively small then no further contact was made with the relevant local authority. As a result of the two surveys, emissions estimates were made for the period 1996–1999 in the case of paper coating processes, and the period 1993–1999 in the case of the remaining three sectors. **Table A1.2** summarises data for 1996 and 1999.

**Table A1.2: Solvent usage and emission estimates for industrial solvent using processes**

Sector	No. of processes	Data available <sup>1</sup>	Solvent usage (tonnes)		Solvent emission (tonnes)	
			1996	1999	1996	1999
Film coating	36	35	15315	15030	15315	8681
Textile coating	87	86	6780	6769	6780	4284
Paper coating	44	42	14023	12303	8079	3953
Seed oil extraction	5	5	- <sup>2</sup>	- <sup>2</sup>	2897	2354

<sup>1</sup> – data available for at least part of time series

<sup>2</sup> – Solvent is recycled within the process and usage figures have not been estimated

Emission factors can be calculated for seed oil extraction processes using statistics given in the Annual Abstract of Statistics. This gives annual tonnages of oilseeds and nuts processed. An emission factor, calculated from the emission estimate for 1993, is used for all previous years back to 1970.

No activity data have been found for textile coating and film coating. Instead, DTI indices of manufacturing output for textiles and textile products, and pulp, paper, printing, and publishing respectively are used to extrapolate from the 1993 figure back to 1970. In the long term, the development of an improved methodology for calculating the earlier part of the NAEI time series should be a priority.

In the case of paper coating processes, different approaches have to be used for various subsectors of the paper coating sector (the sector has been divided into wallpaper manufacture, vehicle air filters, and speciality coated paper products. The Wallpaper Manufacturers Association (WMA) have provided data on UK sales and exports of wallpapers for the period 1981–1999. Wallpaper sales for the period 1996–1999 are used to derive emission factors from the local authority data and the 1996 emission factor is combined with WMA statistics to obtain emission estimates for the period 1985–1995. The WMA data for 1981–1984 are on a different basis to the later data and have not been used for the NAEI – instead the 1985 emission estimate has been extrapolated back to 1970 using the DTI index of manufacturing

output for pulp, paper, printing, and publishing. The 1996 emission estimates for vehicle air filters and speciality coated paper products are extrapolated back to 1970 using DTI indices of manufacturing output for transport equipment, and pulp, paper, printing, and publishing respectively.

## COATING OF METAL PACKAGING

In 1997, local authorities which regulated metal packaging processes were contacted for information on emissions during the period 1993-1996. In 1999, the Metal Packaging Manufacturers Association (MPMA) provided an emission estimate for the sector during 1998. This figure was based on a survey of their members. Following discussions with the MPMA last year, they agreed to provide an emission estimate for 1999 as well, following a further survey of members. The data from the local authorities and from the MPMA have been used to derive emission estimates for each process over the period 1993 to 1999. Data for 1996 and 1999 are shown in **Table A1.3**.

**Table A1.3: Solvent usage and emission estimates for metal packaging processes**

Sector	No. of processes	Data available <sup>1</sup>	Solvent usage (tonnes)		Solvent emission (tonnes)	
			1996	1999	1996	1999
Metal packaging	41	5	14821	9224	13824	6796

<sup>1</sup> – data available for at least part of time series

The British Coatings Federation (BCF) provide coating sales statistics back to 1988 and earlier sales are estimated by extrapolation from the 1988 sales using the DTI index of manufacturing output for food, drink and tobacco. The BCF data are used to generate emission factors from the detailed emission estimates for the years 1993-1999 mentioned above. The emission factor calculated for 1993 is also used for earlier years.

## PAINTING AND PRINTING

Some changes have been made to the methodology used to calculate emissions from the use of paints and inks. This has occurred as a result of new data received from the BCF. These data include improved estimates of UK consumption of coatings and new estimates of the solvent usage/emission for each coating type. **Table A1.4** summarises the emission factors and emission estimates for each sector following the methodological changes.

**Table A1.4: Emission factors and emission estimates for painting and printing**

Sector	Emission factor (kg/tonne)		Emission (ktonnes)	
	1988	1999	1988	1999
Decorative paint (retail decorative)	124	95	30.6	22.7
Decorative paint (trade decorative)	170	112	37.5	25.6
Industrial coating (automotive)	358	259	9.9	10.4
Industrial coating (vehicle refinishing)	717	664	14.1	7.2
Industrial coating (commercial)	717	664	9.4	4.8
Industrial coating (marine)	898	600	8.0	6.3
Industrial coating (high performance)	444	342	7.0	6.1
Industrial coating (packaging)	588	224	16.2	6.8
Industrial coating (coil)	43	15	0.7	0.4
Industrial coating (drum)	714	557	2.2	1.5
Industrial coating (wood)	752	549	14.6	13.0
Industrial coating (aircraft)	764	675	1.6	1.1
Industrial coating (agricultural and construction equipment)	596	495	7.8	2.7
Industrial coating (general industrial)	664	586	41.4	26.5
Printing (heatset web offset)	125	26	5.0	0.9
Printing (other offset)	50	19	0.8	0.3
Printing (newspaper)	50	22	1.9	0.9
Printing (publication gravure)	72	72	2.8	0.9
Printing (flexible packaging)	1733	1011	28.0	23.3
Printing (other flexography)	100	100	0.4	0.7
Printing (metal decorating)	123	136	0.1	0.0
Printing (screen printing)	1040	949	2.4	1.0
Printing (overprint varnishes)	350	153	1.2	2.9
Printing (print chemicals)	124	103	1.7	1.1

## OTHER SOLVENT USAGE

The emission estimate for this sector has been revised, following the provision of new data by the Solvent Industry Association (SIA). The SIA have been encouraged to provide data for use in the NAEI for many years and have, since 1992, provided estimates of solvent consumption in each of a series of sectors for four different years – 1991, 1993, 1996 and 1999. Emissions from other solvent use are calculated by comparing these SIA estimates with estimates of solvent consumption from other sources. Previously, the estimate was based on a comparison of the 1996 SIA data with independent estimates used in the 1997 version of the NAEI which suggested that 44 ktonnes of solvent were emitted from the sector. The current revision is based on comparing data from all four sets of data from the SIA, with independent estimates taken from the 1999 version of the NAEI.



Before comparing the SIA numbers with estimates from other sources, it was decided to remove estimates of solvent consumed in certain sectors from both sides, on the basis that it was believed that the scope of these estimates was the same or similar in both cases (the sectors so treated were painting, printing, wood preservation, metal degreasing, dry cleaning, adhesives, rubber processing, textiles, aerosols, cosmetics, and pesticides). In addition, the SIA estimate of solvent use in the chemical/pharmaceutical industries was also ignored, since the independent estimate of emissions from these industries used in the NAEI does not split out solvents from other organic emissions (such as reactants or products). The remaining numbers were compared and the difference between the two was assumed to be the quantity of solvent consumed by sectors not covered by the independent estimates. The results of the comparisons are shown in **Table A1.5**.

**Table A1.5: Comparison of solvent consumption data from SIA and other sources**

	1991	1993	1996	1999	Mean
Consumption estimated from SIA	166	198	109	127	150
Consumption estimated from other sources	100	105	109	103	104
Difference	66	93	0	24	46

The difference varies greatly from one year to another. Because of the ad-hoc nature of the surveys used by the SIA to collect their data, it is likely that inconsistencies in the way in which solvent sales are assigned to each of the SIA's reporting categories account for some of these variations. On average, the SIA estimates are 46 ktonnes higher than the independent numbers and this figure is used for all years. Further investigation is needed in order to clarify whether this estimate is reasonable.

## ADHESIVES

The estimation of emissions from adhesives use has always been difficult due to the absence of a consistent set of activity statistics and the poor quality of emission factors. Previously, emissions have been estimated by taking SIA estimates of solvent emissions for the adhesives sector in 1991, 1993 and 1996 and using DTI Business Monitor data on adhesives production for the years 1987 to 1992 and the DTI index of output for manufacturing to extend the time series.

Some minor changes have been made, in that the SIA have provided an estimate of solvent usage by the sector in 1999, and some additional data on UK adhesives usage have been collected from the web-site of the European trade association ([www.feica.com](http://www.feica.com)).

This sector is a priority for further investigation, with the collection of a full series of consistent activity data a priority.

## **NON-AEROSOL CONSUMER PRODUCTS**

The existing methodology for non-aerosol consumer products has been modified somewhat. Previously, the existing, detailed, estimate for 1993 had been extrapolated to other years on the basis of population, numbers of households, or vehicle numbers. Research has been carried out to identify data which would allow more accurate calculation of the time series. The most significant success has been the collection of value of sales data for cosmetics and toiletries covering the years 1994-1999. These data have been used to derive estimates of the volume of sales of each type of cosmetic or toiletry. Further effort is advised for household products and car-care products, with the emphasis placed on the more important sources such as slow-release air fresheners, toilet blocks, antifreeze, de-icer, and windscreen wash solutions.

## **BREAD**

The Institute of Bakers was contacted and invited to comment on the methodology used in the 1998 NAEI for bread baking. The Institute was able to provide background information on the UK bread industry and an emission factor based on measurements at two UK plant bakeries (1 kg/tonne bread). This is likely to be more reliable than the figure of 3 kg/tonne bread, used in the 1998 inventory because the latter was based on measurements at bakeries in California. Bakeries in the USA employ a different baking process, involving a longer resting time of typically 3 hours, compared with the UK where doughs would be used immediately upon mixing. This difference is likely to lead to higher emissions of ethanol in US bakeries.

## **SUGAR**

The UK sugar industry can be divided into production from sugar cane and production from sugar beet. Only the latter is considered a source of VOC emissions and the emission actually occurs during drying of sugar beet residues following extraction of the sucrose. The emission factor used in the 1998 NAEI was reviewed. This factor is taken from the UNECE Emission Factor Guidebook and is 10 kg/tonne of sugar produced. This factor is assigned a data quality of E (the lowest possible) in the Guidebook. The factor was of concern because, at approximately 1% of the sugar content of the beet processed, it seemed high.

Emissions from beet pulp driers are undoubtedly odorous but most organic components are present in trace quantities. However, the literature does not give any information on the total organic content of waste gases.

The sole UK company involved in sugar beet processing was approached and they indicated that the only measurements that they had made were at their Kidderminster factory (Authorisation number AA3328). An emission estimate was submitted to the Pollution Inventory and this number has been used to calculate an emission estimate for the whole industry by extrapolation on the basis of capacity.

## **CHEMICALS**

Data has been collected from the Pollution Inventory as in previous years. Because of the way in which data are reported, there is a need to make a lot of assumptions about what is meant. For example emissions may be reported as individual species such as benzene, as groups of compounds such as hydrocarbons, as total VOC of specific types (Class A or B), or as total VOC (as C). Frequently, it appears from the data that emissions might be reported in more than one form e.g. as individual species and total VOC (as C). For each authorisation it is necessary therefore to estimate how the data were reported – in future years it is recommended that selected process operators are contacted to try to verify these assumptions.

Data for Scottish and Irish plants have been collected from process operators and, in the case of Scotland, from SEPA (notably SEPA East), who have provided data from an emissions database which is under development. Data are not available for all Scottish plants (including at least two which use solvents and therefore might have significant emissions).

# Appendix 2

## Details of new PAH research

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### OFF-ROAD VEHICLES

A review of published literature did not identify documented PAH emission factors for agricultural power units, domestic house and garden appliances, aircraft support vehicles and industrial off road machinery. So that these emissions may be included in the NAEI, the PAH emission factors for each of these sources have been equated to on-road vehicles in urban driving conditions as shown in **Table A2.1**.

**Table A2.1: Off-road PAH sources, and on-road sources designated as equivalent**

Off road sector	Designated equivalent on-road source
Agricultural power units (petrol)	LGV non catalyst (petrol)
Agricultural power units (gas oil)	LGV, (derv)
Domestic house and garden (petrol)	Car non catalyst, (petrol)
Domestic house and garden (derv)	LGV, (derv)
Aircraft support (gas oil)	HGV, (derv)
Industry off road (petrol)	LGV non catalyst, (petrol)
Industry off road (gas oil)	LGV, (derv)

Fuel consumption data were available for each of the off-road sectors which allowed total emissions to be calculated – see **Table A2.2**.

**Table A2.2: Calculated emissions of PAHs from off-road vehicles**

	Napthalene	Acenaphthene	Acenaphthylene	Fluorene	Anthracene	Phenanthrene	Fluoranthene	Pyrene	Benz-a-anthracene	Chrysene	Benzo-b-fluoranthene	Benzo-k-fluoranthene	Benzo-a-pyrene	Dibenz_ah_anthracene	Indeno_1,2,3-cd_pyrene	Benzo_ghi_perylene
<b>1999</b>																
Industry off road - petrol	1,277.8	166.7	305.6	333.3	138.9	361.1	222.2	194.4	166.7	194.4	138.9	138.8	166.7	30.6	27.8	516.7
Industry off road - Gas oil	517.1	67.8	127.1	118.7	50.9	152.6	84.8	93.2	59.3	59.3	42.38	50.86	50.86	10.17	7.63	172.9
agricultural power units - petrol	1,277.8	166.7	305.6	333.3	138.9	361.1	222.2	194.4	166.7	194.4	138.9	138.8	166.7	30.6	27.8	516.7
agricultural power units - gas oil	517.1	67.8	127.1	118.7	50.9	152.6	84.8	93.2	59.3	59.3	42.4	50.9	50.9	10.2	7.6	172.9
domestic house and garden - petrol	2,794.0	381.0	657.4	702.2	295.1	821.8	474.4	466.9	366.1	399.7	302.6	310.0	321.2	63.9	57.5	1,048.1
domestic house and garden - DERV	517.1	67.8	127.1	118.67	50.86	152.60	84.77	93.24	59.34	59.30	42.38	50.86	50.86	10.17	7.63	172.9
aircraft support - gas oil	471.9	59.0	118.0	98.3	59.0	137.6	78.6	78.7	59.0	59.0	39.3	39.3	39.3	9.8	7.9	161.2
<b>1998</b>																
Industry off road - petrol	1,277.8	166.7	305.6	333.3	138.9	361.1	222.2	194.4	166.7	194.4	138.9	138.8	166.7	30.6	27.8	516.7
Industry off road - Gas oil	517.1	67.8	127.1	118.7	50.9	152.6	84.8	93.2	59.3	59.3	42.4	50.9	50.9	10.2	7.6	172.9
agricultural power units - petrol	1,277.8	166.7	305.6	333.3	138.9	361.1	222.2	194.4	166.7	194.4	138.9	138.8	166.7	30.6	27.8	516.7
agricultural power units - gas oil	517.1	67.8	127.1	118.7	50.9	152.6	84.8	93.2	59.3	59.3	42.4	50.9	50.9	10.2	7.6	172.9
domestic house and garden - petrol	3070.4	401.5	705.6	808.9	327.7	868.0	513.7	501.9	395.6	434.0	327.7	339.5	354.3	69.7	63.5	1150.5
domestic house and garden - DERV	517.1	67.8	127.1	118.7	50.9	152.6	84.8	93.2	59.3	59.3	42.4	50.9	50.9	10.2	7.6	172.9
aircraft support - gas oil	471.9	59.0	118.0	98.3	59.0	137.6	78.6	78.7	59.0	59.0	39.3	39.3	39.3	9.8	7.9	161.2
<b>1997</b>																
Industry off road - petrol	1,325.6	186.04651	302.32558	372.1	162.7907	372.1	232.5	209.3	186	186.0465	139.5	162.8	162.8	32.558	27.9	539.5

	Napthalene	Acenaphthene	Acenaphthylene	Fluorene	Anthracene	Phenanthrene	Fluoranthene	Pyrene	Benz-a-anthracene	Chrysene	Benzo-b-fluoranthene	Benzo-k-fluoranthene	Benzo-a-pyrene	Dibenz_ah-anthracene	Indeno_1,2,3-cd_pyrene	Benzo_ghi_perylene
Industry off road - Gas oil	637.8	88.3	166.8	137.4	68.7	196.3	107.9	117.8	68.7	78.5	58.87	58.87	68.69	12.76	9.81	213.9
agricultural power units - petrol	1,325.6	186.04651	302.32558	372.1	162.7907	372.1	232.5	209.3	186	186.0465	139.5	162.8	162.8	32.558	27.9	539.5
agricultural power units - gas oil	637.8	88.3	166.8	137.4	68.7	196.3	107.9	117.8	68.7	78.5	58.9	58.9	68.7	12.8	9.8	213.9
domestic house and garden - petrol	3,092.5	400.3	707.0	831.1	337.1	859.2	517.4	501.0	400.3	437.8	330.1	344.1	358.2	70.7	64.4	1,173.1
domestic house and garden - DERV	637.8	88.3	166.8	137.4	68.68	196.3	107.9	117.8	68.69	78.5	58.87	58.87	68.69	12.76	9.81	213.9
aircraft support - gas oil	588.7	78.5	157.0	137.4	58.9	176.6	98.1	98.1	58.9	58.9	58.9	58.9	58.9	11.8	7.9	194.3
<b>1996</b>																
Industry off road - petrol	1,444.4	185.2	333.3	388.8	166.7	388.9	240.7	240.7	185.2	203.7	166.6	166.6	185.2	35.2	31.5	583.3
Industry off road - Gas oil	757.2	189.3	105.2	168.3	84.1	231.4	126.2	136.7	84.1	84.1	63.1	73.6	73.6	14.7	11.6	254.5
agricultural power units - petrol	1,444.4	185.2	333.3	388.8	166.7	388.9	240.7	240.7	185.2	203.7	166.6	166.6	185.2	35.2	31.5	583.3
agricultural power units - gas oil	757.2	189.3	105.2	168.3	84.1	231.4	126.2	136.7	84.1	84.1	63.1	73.6	73.6	14.7	11.6	254.5
domestic house and garden - petrol	3,035.9	398.0	699.3	818.7	341.1	843.3	507.9	492.7	396.1	434.0	327.8	343.0	358.2	70.7	63.5	1,175.5
domestic house and garden - DERV	757.2	189.3	105.2	168.3	84.1	231.4	126.2	136.7	84.14	84.14	63.1	73.6	73.6	14.72	11.57	254.5
aircraft support - gas oil	661.6	85.4	170.7	149.4	64.0	192.1	106.7	128.1	64.0	85.4	64.0	64.0	64.0	12.8	10.7	224.0
<b>1995</b>																
Industry off road - petrol	1,539.7	206.3	365.1	428.6	190.5	428.6	254.0	254.0	206.3	222.2	174.6	174.6	190.5	36.5	33.3	623.8
Industry off road - Gas oil	890.3	118.7	225.5	201.8	95.0	273.0	154.3	154.3	95.0	106.8	83.1	83.0	95.0	16.6	13.1	296.8
agricultural power units - petrol	1,539.7	206.3	365.1	428.6	190.5	428.6	254.0	254.0	206.3	222.2	174.6	174.6	190.5	36.5	33.3	623.8
agricultural power units - gas oil	890.3	118.7	225.5	201.8	95.0	273.0	154.3	154.3	95.0	106.8	83.1	83.0	95.0	16.6	13.1	296.8

	Napthalene	Acenaphthene	Acenaphthylene	Fluorene	Anthracene	Phenanthrene	Fluoranthene	Pyrene	Benz-a-anthracene	Chrysene	Benzo-b-fluoranthene	Benzo-k-fluoranthene	Benzo-a-pyrene	Dibenz_ah_anthracene	Indeno_1,2,3-cd_pyrene	Benzo_ghi_perylene
domestic house and garden - petrol	3,004.2	393.9	694.2	813.6	342.2	829.8	502.0	485.9	393.9	431.0	324.5	342.2	358.4	70.7	63.1	1,176.3
domestic house and garden - DERV	890.3	118.7	225.5	201.8	95.0	273.0	154.3	154.3	95.0	106.8	83.1	83.0	95.0	16.6	13.1	296.8
aircraft support - gas oil	720.7	87.4	174.7	152.9	87.4	218.4	131.0	131.0	87.4	87.4	65.5	65.5	65.5	13.1	10.9	240.2
<b>1994</b>																
Industry off road - petrol	1,610.4	207.8	376.6	441.6	194.8	441.6	272.7	259.7	207.8	233.8	181.8	181.8	194.8	39.0	33.8	649.4
Industry off road - Gas oil	1,058.7	137.5	275.0	233.7	110.0	316.2	178.7	192.5	110.0	123.7	96.2	96.2	110.0	20.6	15.1	356.0
agricultural power units - petrol	1,610.4	207.8	376.6	441.6	194.8	441.6	272.7	259.7	207.8	233.8	181.8	181.8	194.8	39.0	33.8	649.4
agricultural power units - gas oil	1,058.7	137.5	275.0	233.7	110.0	316.2	178.7	192.5	110.0	123.7	96.2	96.2	110.0	20.6	15.1	356.0
domestic house and garden - petrol	2,887.2	378.3	666.8	787.5	332.7	792.9	481.7	465.6	379.7	415.9	312.6	331.4	347.5	68.4	60.8	1,140.4
domestic house and garden - DERV	1,058.7	137.5	275.0	233.7	110.0	316.2	178.7	192.5	110.0	123.7	96.2	96.2	110.0	20.6	15.1	356.0
aircraft support - gas oil	775.7	114.1	205.3	182.5	91.3	228.1	136.9	136.9	91.3	91.3	68.4	68.4	68.4	16.0	11.4	260.0
<b>1993</b>																
Industry off road - petrol	1,694.1	223.5	388.2	470.6	200.0	458.8	282.4	270.6	223.5	247.0	188.2	200.0	211.8	41.2	36.5	682.4
Industry off road - Gas oil	1,179.5	166.1	299.0	265.8	132.9	365.5	199.4	216.0	132.9	132.9	99.7	116.3	116.3	23.3	18.3	397.0
agricultural power units - petrol	1,694.1	223.5	388.2	470.6	200.0	458.8	282.4	270.6	223.5	247.0	188.2	200.0	211.8	41.2	36.5	682.4
agricultural power units - gas oil	1,179.5	166.1	299.0	265.8	132.9	365.5	199.4	216.0	132.9	132.9	99.7	116.3	116.3	23.3	18.3	397.0
domestic house and garden - petrol	2,774.1	364.2	643.1	758.3	323.9	759.5	462.2	447.2	366.5	401.1	302.0	321.5	335.4	66.3	58.5	1,105.6
domestic house and garden - DERV	1,179.5	166.1	299.0	265.8	132.9	365.5	199.4	216.0	132.9	132.9	99.7	116.3	116.3	23.3	18.3	397.0
aircraft support - gas oil	834.1	119.2	214.5	190.6	95.3	262.1	143.0	143.0	95.3	95.3	71.5	71.5	95.3	16.7	11.9	281.2

	Napthalene	Acenaphthene	Acenaphthylene	Fluorene	Anthracene	Phenanthrene	Fluoranthene	Pyrene	Benz-a-anthracene	Chrysene	Benzo-b-fluoranthene	Benzo-k-fluoranthene	Benzo-a-pyrene	Dibenz_ah_anthracene	Indeno_1,2,3-cd_pyrene	Benzo_ghi_perylene
<b>1992</b>																
Industry off road - petrol	1,857.1	241.8	428.6	516.5	219.8	505.5	307.7	296.7	241.8	263.7	197.8	219.78	219.8	43.956	39.56	741.76
Industry off road - Gas oil	1,233.9	170.8	303.7	265.8	132.9	379.7	208.8	208.8	132.9	151.9	113.9	113.9	113.9	22.8	19.0	410.0
agricultural power units - petrol	1,857.1	241.8	428.6	516.5	219.8	505.5	307.7	296.7	241.8	263.7	197.8	219.8	219.8	44.0	39.6	741.8
agricultural power units - gas oil	1,233.9	170.8	303.7	265.8	132.9	379.7	208.8	208.8	132.9	151.9	113.9	113.9	113.9	22.8	19.0	410.0
domestic house and garden - petrol	2,808.8	367.8	648.8	773.5	329.7	764.0	467.1	451.2	370.9	405.8	305.4	325.5	340.3	67.2	59.4	1,122.3
domestic house and garden - DERV	1,233.9	170.8	303.7	265.8	132.9	379.7	208.8	208.8	132.9	151.9	113.9	113.9	113.9	22.8	19.0	410.0
aircraft support - gas oil	881.8	126.0	226.8	201.6	100.8	277.1	151.2	151.2	100.8	100.8	75.6	75.6	75.6	17.6	12.6	292.3
<b>1991</b>																
Industry off road - petrol	1,969.7	252.5	444.4	545.5	232.3	525.3	323.2	313.1	259.6	282.8	212.1	222.2	232.3	46.5	41.4	778.8
Industry off road - Gas oil	1,272.2	166.9	312.8	292.0	125.1	375.4	208.6	229.4	146.0	146.0	104.3	125.0	125.0	25.0	18.8	425.5
agricultural power units - petrol	1,969.7	252.5	444.4	545.5	232.3	525.3	323.2	313.1	259.6	282.8	212.1	222.2	232.3	46.5	41.4	778.8
agricultural power units - gas oil	1,272.2	166.9	312.8	292.0	125.1	375.4	208.6	229.4	146.0	146.0	104.3	125.0	125.0	25.0	18.8	425.5
domestic house and garden - petrol	2,869.1	369.3	655.5	800.1	335.4	771.4	476.0	458.5	376.5	411.3	309.8	330.3	344.7	68.0	60.7	1,136.6
domestic house and garden - DERV	1,272.2	166.9	312.8	292.0	125.1	375.4	208.6	229.4	146.0	146.0	104.3	125.0	125.0	25.0	18.8	425.5
aircraft support - gas oil	933.7	116.7	233.4	210.0	93.4	280.1	163.4	163.4	93.4	116.7	70.0	93.4	93.4	18.7	14.0	308.1
<b>1990</b>																
Industry off road - petrol	1,969.7	252.5	444.4	545.5	232.3	525.3	323.2	313.1	259.6	282.8	212.12	222.2	232.32	46.46	41.41	778.78
Industry off road - Gas oil	1,203.9	165.2	306.9	259.7	118.0	377.7	212.4	212.5	141.6	141.6	118.0	118.0	118.0	23.6	18.9	406.0
agricultural power units - petrol	1,969.7	252.5	444.4	545.5	232.3	525.3	323.2	313.1	259.6	282.8	212.1	222.2	232.3	46.5	41.4	778.8



	Napthalene	Acenaphthene	Acenaphthylene	Fluorene	Anthracene	Phenanthrene	Fluoranthene	Pyrene	Benz-a-anthracene	Chrysene	Benzo-b-fluoranthene	Benzo-k-fluoranthene	Benzo-a-pyrene	Dibenz_ah_anthracene	Indeno_1,2,3-cd_pyrene	Benzo_ghi_perylene
agricultural power units - gas oil	1,203.9	165.2	306.9	259.7	118.0	377.7	212.4	212.5	141.6	141.6	118.0	118.0	118.0	23.6	18.9	406.0
domestic house and garden - petrol	2,910.3	364.0	652.2	828.2	335.7	767.5	481.3	461.1	377.2	412.6	309.4	330.7	344.8	68.0	61.6	1,134.6
domestic house and garden - DERV	1,203.9	165.2	306.9	259.7	118.0	377.7	212.4	212.5	141.6	141.6	118.0	118.0	118.0	23.6	18.9	406.0
aircraft support - gas oil	897.7	131.4	218.9	197.0	87.6	262.7	153.3	153.3	87.6	109.5	87.6	87.6	87.6	17.5	13.1	299.9

# MANUFACTURE AND USE OF TAR AND BITUMEN

An extensive literature and internet search was undertaken along with communication with experts from industry and trade associations.

## Description of sources

### Tar

Tar has been defined as a black viscous liquid, having waterproofing and adhesive properties, obtained from the destructive distillation of coal, oil etc.

There are two main types of coal tar:

- Low temperature tar- the condensation product obtained from the cooling of the gas evolved from the carbonisation of coal at temperatures up to 700°C
- High temperature tar – the condensation product obtained from the cooling of the gas evolved from the carbonisation of coal at temperatures above 700°C (Typically 1000-1350°C), these tars are usually produced as by-products of the manufacturer of coke for blast furnaces.

### Processes

Coal tar is produced by the destructive distillation of coal during the manufacture of coke. This process produces a product that contains a greater proportion of PAHs than bitumen in fact many orders of magnitude greater therefore if used in similar applications to bitumen the emissions will be greater.

From communications with Shell it became apparent that tar was no longer produced or used in significant quantities within the UK.

### Bitumen

Bitumen is a viscous liquid or solid, consisting essentially of hydrocarbons and their derivatives. Bitumen is soluble in trichloroethylene, is substantially non-volatile and softens gradually when heated. It is black or brown in colour and possesses waterproofing and adhesive properties. It is obtained from petroleum using refinery processes.

### Processes

Bitumen products are formed from the residue from vacuum distillation of selected crude oil. The nature of the crude oil can effect the process required to produce different grades of bitumen. In the US bitumen is referred to as asphalt, in the UK a mixture of bitumen and mineral matter is called asphalt.

There are three main types of bitumen these are; penetration grade bitumen, hard bitumen and oxidised bitumen.

**Penetration grade bitumens** are produced by vacuum distillation of selected crude oil partially oxidised, solvent precipitation, or a combination of these processes depending on the characteristic required. Penetration grades are principally used for road surfacing but also used for industrial applications and roofing.

**Hard bitumens** are produced in a similar way to penetration grades and are used in the manufacture of paints and enamels.

**Oxidised bitumens** are produced by vigorous air blowing of the residues of the vacuum distillation under controlled conditions; this process produces a more rubbery product which is reported to have reduced temperature susceptibility and greater stress resistance. The main use of oxidised or air blown grade bitumen are in roofing felts, waterproofing and electrical goods.

The oxidation of bitumen can be carried out by three slightly different methods, these are bitumen oxidation, air rectification and catalytic oxidation. These processes can be carried out in either continuous or batch operations, it is however more common to have a continuous process. Bitumen is oxidised by blowing air through a straight run of bitumen. If the oxidation reactor is not at a refinery site bitumen can be delivered by rail or road. It is usually held in storage until required, then fed into the reactor as the feedstock. The reactor which is kept at a constant temperature of about 230°C. The oxidised product is continuously removed from the reactor and stored in heated tanks. The oxidised product may then be poured into kegs or silicone lined boxes for storage and transported to customers once cool. The oxidised material may however be delivered directly to the customer whilst hot.

The air rectification process is used to produce harder grades of bitumen from softer petroleum residues. The process is similar to that of the oxidation process however the temperatures and air flow are lower. Air emissions usually less than for the oxidative process.

Catalytic oxidation can also be used to produce oxidised bitumen, the process can use a variety of catalysts including ferric chloride however this process is not presently in operation within the UK.

### **Bitumen Derivatives**

There are four common derivatives of Bitumen these are cutback bitumen, blended grades, bitumen emulsions, and modified bitumen.

**Cutback Bitumen** is produced by mixing penetration grade bitumens with a solvent such as kerosene with an aim of reducing the viscosity. Generally the amount of cut back bitumen produced and used is declining as it is increasingly being replaced by bitumen emulsions which have better environmental and safety

performance. Cut back bitumen is usually used for road construction and repair industry.

**Bitumen emulsions** are a fine dispersion of bitumen in water. They are usually produced by high shear devices and normally involve the addition of an emulsifier. The bitumen content is usually in the range 30-70% by weight. Bitumen emulsions have similar uses to cutback emulsions but can be used at ambient temperature or with minimal heating.

**Blended grade bitumens** are produced by controlled mixing of different grades of bitumen and solvent extracts to create a bitumen with particular properties.

**Modified bitumens** contain adhesives (3-15% by weight) to modify the properties of the bitumen, they are commonly used in road construction and roofing.

## **Secondary product manufacture**

### Joists fillers/sealant

The manufacture of joints fillers/sealants is low in volume and is a specialised industry. The manufacture is usually a batch blending process, oxidised bitumen is blended with rubber and filler. The process involves mixing in a closed system in a similar manner to the manufacture of polymer modified bitumens.

### Manufacture of roofing felts

Conventional roofing felt consists of a fabric impregnated with hot penetration grade bitumen, surface coating is applied to both sides using oxidised bitumen or polymer modified bitumen, with added filler. A binding coat of sand or fine chippings is added to one side. Different grades of felt are prepared by varying the fabric used in the process. The manufacture process involves a roll of fabric being passed continuously through an open bath of hot penetration grade bitumen. The bath is kept topped up and kept at about 140°C. The coating layer is then applied by passing the sheet through a curtain of oxidised or polymer bitumen with fillers. The final coat of sand or other fine mineral aggregate is sprayed onto one side of the sheet.

### **Manufacture of Mastic Asphalt**

Mastic asphalt consists of bitumen and finely divided limestone. The process usually uses hot bitumen being mixed with mixed grade limestone. The blend is agitated in a heated mixing tank for 2 to 5 hours at 230°C. Once the mixing process is completed the mastic is poured into moulds to cool.

Although some mastic is taken straight to sites for immediate use, most is re-heated when required using oil or gas fired cauldrons for flooring and roofing applications.

## **Air Emissions**

### **Bitumen production for road construction and roofing manufacture**

The processes where emissions may be released from the bitumen manufacture process are: during the feedstock delivery, during storage, loading the product, fugitive emissions from leakage at pumps seals, pipe flanges, combustion of flue gas, kegging of the oxidised bitumen.

### **Road Manufacture**

During road manufacture the emissions to air of PAHs depend on the temperature at which the road is laid. Roads are usually laid at about 150°C. The emissions take place during the following operations.

Delivery of the raw material bitumen: Raw bitumen is delivered hot from the bitumen plant. Bitumen is usually delivered by road or rail tankers:

Tankers usually vent to atmosphere and are discharged either by air blowing, by pumps, or by gravity transfer to storage tanks or for direct use. It is regarded that pumped discharge to storage tanks produce least emission to air. Air discharge for off loading road tankers is most common, on board compressors are used to expel the hot liquid. This process gives rise to greater emission than the pumped emission, particularly during the depressurising step at the end of the discharge process where fumes are emitted from the tanker.

Emission will also take place if the bitumen is not stored before use.

### **Road Construction process**

During the road construction process emissions of Fumes from the bitumen are inevitable. The emission rate will be dependant on the temperature of the bitumen during the laying process. Road paving usually takes place at about 150°C. As the paving cools the rate of emission of fumes will decrease dramatically until essentially no fumes are being emitted. An industrial source indicated that the emission of PAHs would be virtually zero below 110°C.

### **Mastic manufacture**

The emissions from mastic asphalt production are from feedstock delivery, storage, the manual mixing of the feedstock and limestone. As the majority of the systems are not automated the emissions are usually around fill points. The fumes arising are emitted to the atmosphere through vents or hoods. Where automated systems are used the emissions are vented through the roof although the fumes arising through the fill points may be reduced.

### **Use of Mastic Asphalt**

Although generally the proportion of mastic asphalt used in the UK is much less than bitumen used in road construction the relative emissions from laying the mastic asphalt is greater than laying a similar amount of bitumen for road paving. The reason for this is that mastic asphalt is laid at 250°C compared to 150°C for bitumen for paving and therefore will inevitably cause higher emissions of PAHs.

### **Manufacture of Joints filler/sealants**

The emissions from this process is similar in type to emissions from bitumen production processes.

### **Manufacture of roofing felt**

The parts of the production processes which cause emissions are the delivery of the feedstock, storage of the feedstock, fumes arising of the blending tanks of the filler bitumen and polymer bitumen, the saturation tank and the coating processes. The amount of fuming is dependent on the temperature and surface area. The felt is kept at the minimum temperature possible to reduce heating costs. Emissions from all of these processes are vented through extraction hoods to the atmosphere.

### **Roofing laying**

Emissions from the process of laying roofing are generally greater than that of laying a similar amount of bitumen during road construction. The reason for this is that roofing is generally laid at 200°C compared to 150°C for bitumen for paving and therefore will inevitably cause higher emissions of PAHs.

Emission processes are summarised in **Table A2.3**.

**Table A2.3: Summary of emission sources for PAHs**

	Emission Processes				
	Delivery of Raw Material	Handling and Storage of raw materials	Process	Product Storage/cooling	Loading/use
Manufacture of Bitumen	✗	✗	✓ from process usually vented and burnt, fugitive emissions, leaks, keggings and handling of product	✓ fumes while product is stored at temperature usually vented to the atmosphere	✓ during transfer of the product for transportation for use
Manufacture of Roofing materials	✓ delivery of bitumen	✓ storage of delivered bitumen	✓ usually fugitive emissions, emissions possibility of abatement	✓ fumes whilst product is cooling	See roof laying
Road construction and repair	✓ delivery of the bitumen	✓ storage of the bitumen prior to use	✓ during the construction/repair process	✓ during the cooling process	N/A
Roofing construction	✗ usually raw materials are heated on site so emissions only take place during the roof laying process	✗ usually raw materials are heated on site so emissions only take place during the roof laying process	✓ during the laying process from the heating of the bitumen used for securing the roofing felt	✓ during the cooling process of the roofing material	N/A

### Derivation of emission factors

There is relatively little information available concerning the actual emissions of PAH from bitumen production and uses. Industrial contacts have indicated that the major emissions will occur in the areas of use of bitumen in the road, airfield and roof laying processes. It was indicated that the production of bitumen and its products could release PAH to the atmosphere but 'off-gases' from the blowing process are usually incinerated under conditions which should preclude the emission of PAH during manufacture.

Therefore the most appropriate approach concerning air emissions is to concentrate on the emissions from the use of the bitumen in road and roofing industry, as there are no abatement techniques used.

The emission profile for bitumen use has been estimated using technical assistance from industrial contacts and using Concawe product dossier. It was assumed that 85% of bitumen is used in road and airfield applications and 15% is used in other

applications of which the majority is utilised in the roofing industry. It was assumed that the PAH content in the fume is proportional to the emission profiles shown in the two sets of heating experiments of bitumen in the Concauwe dossier. It is possible to estimate the proportion of each PAH measured in the fume in the experiments shown in the Concauwe dossier. This has been done for both grades of bitumen used for both road/airfield applications and roofing applications. The major difference between the two applications is that roofing applications heat bitumen to over 200–250°C whereas road construction usually involves heating in the range 150–160°C. It was estimated that the loss on heating of bitumen was between 0.01% and 0.2% during the heating process therefore it was assumed that the average loss on heating was the best estimate (0.105%).

The emissions profile calculation is shown below:

$$\text{Emission of PAH(X)} = (0.105/100) \times (\% \text{ individual PAH in fume}/100)$$

Using this equation it is possible to get an emission profile for each of the PAH covered in the heating experiments in the Concauwe dossier. From these it was possible to create an overall emission profile assuming that the bitumen use was split between road and airfield applications and roofing application in the percentages 85% to 15% respectively.

The emission factors that this estimation process generated are shown in **Table A2.4** below.

**Table A2.4: Emission factors for PAHs from the tar and bitumen industry**

PAH	Pollcode	Emission Factor (kg/Mtonne)
Phenanthrene	34	561.07
Anthracene	33	13.06
Fluoranthene	35	23.44
Pyrene	36	79.28
Chrysene	38	119.10
Benzo( <i>a</i> )anthracene	37	31.89
benzo( <i>k</i> )fluoranthene	40	0.92
Benzo( <i>a</i> )pyrene	41	5.53
Benzo( <i>ghi</i> )perylene	44	9.77

The emission factors shown are for individual PAH in kg per Mtonne of bitumen used in the UK.

These emission factors are displayed in the national emissions inventory and the source data is stored in the NAEI database referenced as 881, (Naei99/rawdata/datafmt/bit\_fumecc1.xls).



## **Limitations of the emission profiles**

The limitations of the profile for the emissions of PAH from the tar and bitumen industry are:

- There have been no emission measurements taken at the production sites of bitumen or its secondary product manufacturing sites.
- The assumption that the majority of the air emissions occur at the end-use and that abatement or combustion of the 'off gases' limit the emissions at the manufacturing plant.