AEAT-6270 Issue 2

UK Particulate and Heavy Metal Emissions from Industrial Processes

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

Neil R Passant, Martin Peirce, Howard J Rudd, David W Scott, Ian Marlowe & John D Watterson

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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:

- emission factors for particulate matter and heavy metals emissions from industrial processes;
- species profiles for chromium, mercury and nickel for all emission sources.

This report is the result of that research and includes detailed recommendations for improvements to the emission factor data used in the NAEI in many areas including:

- particulate matter emission factors for solid smokeless fuel manufacture;
- particulate and heavy metal emission factors for sinter plants;
- particulate matter emission factors for blast furnaces;
- particulate matter, copper, chromium, and lead emission factors for basic oxygen furnaces;
- particulate matter emission factors for ferrous foundries;
- particulate matter emission factors for primary aluminium production;
- particulate matter and heavy metal emission factors for secondary aluminium production;
- particulate matter and heavy metal emission factors for glass production;
- particulate matter and heavy metal emission factors for bricks and ceramics;
- particulate matter and heavy metal emission factors for other chemical and non-ferrous metal processes.

Some changes to the NAEI were made during the compilation of the 1998 NAEI and particularly the 1999 NAEI based on the research described in this report. Further revisions have been made for the 2000 NAEI in order to complete the introduction of emission factors recommended in this report. While this did not lead to any major changes in national totals between the 1999 NAEI and the 2000 NAEI as a result of the revisions, changes made in the 1998 and 1999 NAEI, based on this work, did result in significant changes in the detailed estimates for these pollutants and, in five cases resulted in major changes in estimates of total emissions. In the case of cadmium, mercury and zinc, the main reason was the adoption of new emissions data for primary lead/zinc producer, while in the case of chromium and copper the main reason was the addition of emission estimates for chromium chemicals manufacture in the case of chromium and chemicals manufacture and copper alloys and semis production in the case of copper.

Table 1 Impact of revisions to NAEI estimates for 1997 based on this work (figures in tonnes)

Pollutant	Change due to addition of new factors	Change due to revisions to factors	Overall change	UK emission	Change as % of UK emission
Arsenic	0.08	-2.7	-2.6	52.6	5.0%
Cadmium	0.14	-7.0	-6.8	7.82	87.4%
Chromium	27	14	40	86.2	46.9%
Copper	10	2.0	12	66.2	18.3%
Mercury	0.24	-1.6	-1.4	12.2	11.2%
Nickel	6.6	9.1	16	223	7.0%
Lead	135	-143	-7.2	1180	0.6%
PM_{10}	6400	-260	6100	228000	2.7%
Selenium	0.1	-3.4	-3.3	78.6	4.2%
Vanadium	0.1	-2.1	-2.0	446	0.5%
Zinc	40	-280	240	655	36.7%

In addition, we propose species profiles for chromium, mercury and nickel which can be used to speciate the NAEI inventories for these metals, giving the following results.

Chromium	10% hexavalent chromium 90% trivalent chromium
Mercury	51% vapour phase metallic mercury 7% particulate-bound metallic mercury 42% divalent mercury compounds
Nickel	0.5% metallic nickel 47% oxidic nickel 50% soluble nickel compounds 0.02% nickel carbonyl 2.6% sulphidic nickel

Contents

1 Introduction & Approach

This report has been prepared under the DEFRA 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 NAEI was carried out at the start of the research (Passant and Wenborn, 2000. Options for Improvement of the National Atmospheric Emissions Inventory. AEAT-5015 Issue 2), which recommended a thorough review of particulate matter and heavy metal emission factors for industrial processes as these represent a substantial component of the NAEI inventory, and have high uncertainty. Subsequently, DEFRA have asked that this review should also include a re-assessment of speciation information for mercury, chromium and nickel. This report documents the review.

The review has been carried out by comparing the current NAEI (the ‘1999’ NAEI) methodology with emission factors and emissions data drawn from a number of sources, notably:

- the US EPA Compilation of Emission Factors for Air Pollutants, Fifth Edition (AP-42), available on the Air Chief CD, version 8.0, released in October 2000.
- the EMEP/CORINAIR Emission Inventory Guidebook, 2nd edition, published in 1999
- BREF notes produced by the European IPPC Bureau (EIPPCB), published during 2000 and 2001
- BAT notes produced by various consultants for the UK Environment Agency, published during the early 1990s
- the UK Environment Agency’s Pollution Inventory
- other data provided by the UK Environment Agency

Information was also solicited from industry, but with a few exceptions the response was poor.

This report includes an appendix on each industrial sector studied. Each appendix follows a standard format loosely based on the EMEP/CORINAIR Guidebook. Each appendix contains sections which:

- define the industry sector covered by the appendix;
- give details of the processes carried out;
- identify potential sources of particulate and heavy metal emissions;
- summarise abatement techniques applicable to the sector;
- describe the characteristics of the industry sector in the UK — number of plants, technologies in use, UK production levels, and types of abatement options chosen by UK operators;
- review the available information on emission factors and emissions data for particulate and heavy metal emissions from the information sources listed above;

- give details of the size distribution of particulate emissions so that factors for PM_{10}, $PM_{2.5}$ and other fractions can be calculated from the emission factors for total particulate;
- compare the available emission factors and emission estimates with the current NAEI methodology, and recommend any changes to NAEI practice considered appropriate.

Where US EPA emission factors are quoted, they are accompanied by the quality rating given to the emission factor by the US EPA. Ratings run from A through E, with A being the best.

The report also includes three appendices which describe the available information on the species present in emissions of mercury, nickel and chromium.

2 Summary of findings

2.1 PARTICULATE MATTER

In the 1999 NAEI industrial processes contributed 23% of total emissions, while transport and stationary combustion sources contributed 25% and 43% respectively. The remaining 8% is from waste disposal and agriculture.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plants
- blast furnaces
- cement production
- lime production
- glass production
- construction
- quarrying

Construction and quarrying are not covered by this report, but the review of emission factors for the remaining sources and other industrial production processes is covered in detail in Appendices A1 to A19.

Following the review we recommend that the NAEI be revised in a number of areas. In particular, we would recommend that the emission factors for solid smokeless fuels, sinter plants, blast furnaces, basic oxygen furnaces, primary lead/zinc production, and foundries be updated, as well as minor revisions to the methodologies used for coke ovens, electric arc furnaces, and glass production. Emission estimates should be added for alumina production and for the manufacture of bricks & ceramics. Finally, a large number of sources which have previously been grouped together as ‘chemical industry’ and ‘other non-ferrous metals’ should be included as separate sources and time series developed. These include secondary aluminium production, copper alloys and semis production, soda ash production and titanium dioxide production. For some industrial processes we have been able to recommend emission factors both for the current level of emissions control and also for the lower level of control in the years prior to the implementation of the Environmental Protection Act 1990 (EP Act). In the remaining cases, it has not been possible to recommend an emission factor to represent the situation before controls were tightened under the EP Act, due to an absence of information on the abatement systems in use at the time. This situation should ideally be resolved through further investigations, including consulting individual process operators, with the aim of obtaining historical emissions data. In addition, further information on current control measures would also be helpful in assessing the reasonableness of emission factors

PM_{10} emission factors can be calculated for all sources, using US EPA size distributions or data reported in the Pollution Inventory. An overall conclusion of this work, however, is that the existing data on the size of particulate matter emissions is subject to considerable uncertainty and that source monitoring is required to substantially improve this situation.

2.2 HEAVY METALS

Introduction

Emission factors for heavy metals are reviewed in detail in Appendices A1 to A19 and a large number of changes to the NAEI methodology are recommended. The following recommendations are generally applicable to all metals:

- separate emission estimates should be made for certain sources which were previously included in the general categories 'other non-ferrous metals' and 'chemical industry';
- the emission factors for primary lead/zinc production probably do not take account of fugitive sources and may therefore be 2 or 3 times too low;
- the issue of whether fugitive emissions are included in emission factors for other non-ferrous metal and from chemical processes has not been addressed, and may be an issue. Further discussions with regulators and process operators may help to establish whether existing emission factors take account of fugitive emissions;
- emission factors for solid smokeless fuels, sinter plants, and glass production should be revised and minor changes should be made to the calculation of emission factors for coke ovens, electric arc furnaces, and secondary lead production;
- better information is required on the level of emissions control, both in the current day, and also in the past, so that emission factors can be selected with more confidence;
- more information on the chemical composition of dusts would be helpful in assessing the reliability of emission factors for metals;
- information is needed on the distribution of metals between different size fractions of emitted particulate matter so that the impact of controls on metal emissions can be assessed with more confidence.

In addition, the following points can be made about the inventories for each metal.

Arsenic

In the 1999 NAEI industrial processes contributed 16% of total emissions, while stationary combustion sources contributed 84%. Emissions from other sources are negligible.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- blast furnaces
- basic oxygen furnaces
- electric arc furnaces
- primary lead/zinc production
- secondary copper production
- cement production
- glass production

The main concerns for the arsenic inventory are the emission factors for primary lead/zinc production which are probably too low, and glass production where the existing factors are very uncertain, but might be too high. Otherwise, the changes that we recommend should not impact very significantly on the emission inventory for arsenic.

Cadmium

In the 1999 NAEI industrial processes contributed 60% of total emissions, while transport and stationary combustion sources contributed 6% and 24% respectively. Incineration processes were responsible for 10% of emissions.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plant
- basic oxygen furnaces
- primary lead/zinc production
- secondary lead production
- glass production
- other processes (cadmium pigment manufacture)

Our review has found a number of areas where significant changes to the inventory are recommended. Emission factors for sinter plant and primary lead/zinc production are too low although some relatively minor sources such as cadmium pigments and phosphate fertiliser manufacture are probably overestimated.

Chromium

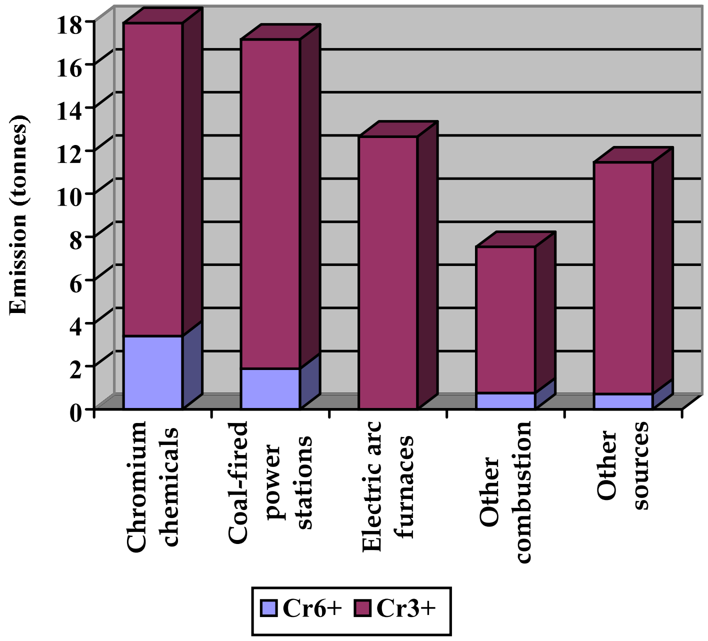
In the 1999 NAEI industrial processes contributed 60% of total emissions, while transport and stationary combustion sources contributed 1% and 39% respectively. Emissions from other sources are negligible.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- coke ovens
- sinter plants
- basic oxygen furnaces
- electric arc furnaces
- primary lead/zinc production
- cement production
- glass production
- chromium chemicals

Our review has found that emission factors for primary lead/zinc production are probably too low, although recommended figures for sinter plants and basic oxygen furnaces are lower than those currently in use in the NAEI. A previously uncharacterised source which might be significant is the manufacture of refractory materials based on chromium-containing minerals. Species profiles have been proposed for significant sources of chromium emissions and details are given in Appendix B1. Figure 1 summarises results obtained by applying these profiles to 1999 emission estimates for chromium, taken from the current (1999) NAEI. The speciation is split into hexavalent and trivalent chromium. The overall split is 10% hexavalent chromium and 90% trivalent chromium.

Figure 1. Speciation of UK emissions of chromium in 1999



Copper

In the 1999 NAEI industrial processes contributed 53% of total emissions, while transport and stationary combustion sources contributed 1% and 45% respectively.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plants
- blast furnaces
- electric arc furnaces
- secondary copper production
- copper alloys & semis
- chemicals

We have recommended a number of changes to copper emission factors which should have a relatively minor impact. These include any changes to the emission factor for primary lead/zinc production, as well as changes to the factors for basic oxygen furnaces and foundries.

Lead

In the 1999 NAEI industrial processes contributed 28% of total emissions, while transport and stationary combustion sources contributed 60% and 11% respectively. Incineration contributes the remaining 1%.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plants
- basic oxygen furnaces

- electric arc furnaces
- primary lead/zinc production
- secondary lead production
- glass production
- alkyl lead processes

Our review found that the factors for sinter plant are probably too low, but our proposed changes will lead to a relatively small change in the lead inventory. Factors for the glass industry should also be revised, possibly leading to significant changes.

Mercury

In the 1999 NAEI industrial processes contributed 32% of total emissions, while stationary combustion sources contributed 42%. Waste disposal contributed the remaining 26% while emissions from transport were negligible.

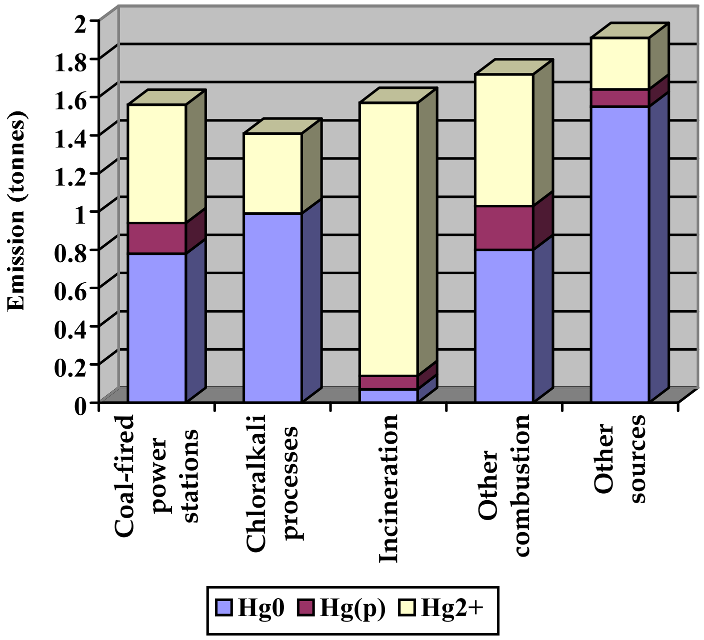
Amongst industrial processes, the major sources in the 1999 NAEI were:

- electric arc furnaces
- iron & steel foundries
- primary lead/zinc production
- cement production
- chloralkali processes

Probably the only significant change that we have recommended is that historical factors for chloralkali processes should be revised upwards, which might lead to a significant increase in the earlier part of the mercury inventory.

Speciated emission estimates have been developed for the top sources representing 90% of the mercury emission inventory; detailed information is given in Appendix B2. Figure 2 summarises results obtained by applying these profiles to 1999 emission estimates for mercury, taken from the current (1999) NAEI. The speciation is split into metallic mercury, particle-bound mercury and divalent mercury. The overall split is 51% vapour phase metallic mercury, 7% particle-bound metallic mercury and 42% divalent mercury.

Figure 2. Speciation of UK emissions of mercury in 1999



Nickel

In the 1999 NAEI industrial processes contributed 14% of total emissions, while transport and stationary combustion sources contributed 3% and 83% respectively.

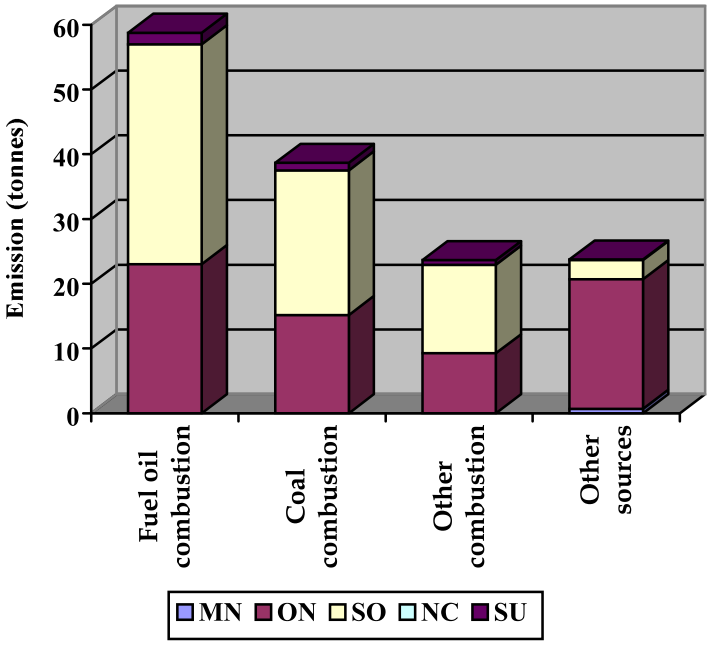
Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plants
- blast furnaces
- electric arc furnaces
- primary aluminium production
- glass production
- nickel production

None of the recommendations of this review are likely to lead to very significant changes to the nickel inventory.

Species profiles have been proposed for significant sources of nickel emissions and details are given in Appendix B3. Figure 3 summarises results obtained by applying these profiles to 1999 emission estimates for nickel, taken from the current (1999) NAEI. The speciation is split into metallic nickel (MN), oxidic nickel (ON), soluble nickel compounds (SO), nickel carbonyl (NC), and sulphidic nickel (SU). The overall split is 0.5% metallic nickel, 47% oxidic nickel, 50% soluble nickel compounds, 0.02% nickel carbonyl, and 2.6% sulphidic nickel.

Figure 3. Speciation of UK emissions of nickel in 1999



Selenium

In the 1999 NAEI industrial processes contributed 49% of total emissions, while transport and stationary combustion sources contributed 1% and 50% respectively.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plants
- blast furnaces
- secondary lead production
- cement production
- glass production

Glass production is by far the largest source, and our review proposes that emission factors for glass processes should be further reviewed and revised where necessary. This could lead to significant changes to the inventory.

Vanadium

In the 1999 NAEI industrial processes contributed 10% of total emissions, while transport and stationary combustion sources contributed 5% and 86% respectively.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- sinter plants
- blast furnaces
- glass production

None of the recommendations of this review are likely to lead to very significant changes to the vanadium inventory.

Zinc

In the 1999 NAEI industrial processes contributed 71% of total emissions, while transport and stationary combustion sources contributed 3% and 25% respectively. The remaining 1% is from incineration.

Amongst industrial processes, the major sources in the 1999 NAEI were:

- blast furnaces
- basic oxygen furnaces
- electric arc furnaces
- primary lead/zinc production
- secondary copper production
- glass production
- other processes (zinc alloys, chemicals)

The review recommends changes to the emission factors for primary lead/zinc production, which should lead to a small increase in the zinc inventory, otherwise no significant changes are expected.

3 Conclusions & Recommendations

We have reviewed literature sources and information from third parties regarding emissions of particulate matter and heavy metals from industrial processes. Resulting from this review, we have identified improved emission factors for many industrial processes, including:

- particulate matter emission factors for solid smokeless fuel manufacture;
- particulate and heavy metal emission factors for sinter plants;
- particulate matter emission factors for blast furnaces;
- particulate matter, copper, chromium, and lead emission factors for basic oxygen furnaces;
- particulate matter emission factors for ferrous foundries;
- particulate matter emission factors for primary aluminium production;
- particulate matter and heavy metal emission factors for secondary aluminium production;
- particulate matter and heavy metal emission factors for glass production;
- particulate matter and heavy metal emission factors for bricks and ceramics
- particulate matter and heavy metal emission factors for other processes (see Appendix A19 for details)

In addition, we have recommended a large number of changes in the detailed methodologies for most of the sectors considered, including consideration of whether higher emission factors, for example based on USEPA factors for uncontrolled processes, should be used for the earlier part of the time series.

Some changes to the NAEI were made during the compilation of the 1998 NAEI and particularly the 1999 NAEI based on the research described in this report. Further revisions have been made for the 2000 NAEI in order to complete the introduction of emission factors recommended in this report. While this did not lead to any major changes in national totals between the 1999 NAEI and the 2000 NAEI as a result of the revisions, changes made in the 1998 and 1999 NAEI, based on this work, did result in significant changes in the detailed estimates for these pollutants and, in some cases resulted in major changes in estimates of total emissions. Table 1 shows the change in emission estimates for the year 1997 when comparing data in the 1997 NAEI and the 2000 NAEI . In most cases, the change is less than 10% of the UK total, but in five cases (cadmium, chromium, copper, mercury and zinc) the changes are more than 10%. In the case of cadmium, mercury and zinc, the main reason was the adoption of new emissions data for primary lead/zinc producer, while in the case of chromium and copper the main reason was the addition of emission estimates for chromium chemicals manufacture in the case of chromium and chemicals manufacture and copper alloys and semis production in the case of copper.

Table 1 Impact of revisions to NAEI estimates for 1997 based on this work (figures in tonnes)

Pollutant	Change due to addition of new factors	Change due to revisions to factors	Overall change	UK emission	Change as % of UK emission
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Vanadium	0.1	-2.1	-2.0	446	0.5%
Zinc	40	-280	240	655	36.7%

There are still many areas where more data are needed. It has not been possible to fully address the issue of fugitive emissions from all processes (for example no estimate can currently be made for the UK primary lead/zinc producer). In addition, while emission factors for the current UK situation can be recommended, in the case of most of the industrial processes studied, it is not possible to recommend emission factors for the pre-1990s period with any confidence. Finally, more information is required on the current technologies and abatement systems for a number of sectors including foundries, glass-works, alumina production, ceramics production and lime processes. It is probable that data on emissions prior to the 1990s do not exist, and so further research is not recommended. However, we do recommend that further research be carried out to address the issues of fugitive emissions and current technology and abatement.

We propose species profiles for major sources of emissions of chromium, mercury and nickel, and recommend that these be adopted for use in the NAEI. However, these profiles are based on few data and further desk-based and/or measurement work is required before the profiles can be assigned a high level of confidence.

Appendix A1: Coke ovens

SNAP CODE:	**010406** **040201** **040204**
SOURCE ACTIVITY TITLE:	**Coke oven furnaces** **Coke oven (door leakage and extinction)** **Solid smokeless fuel**
NACE CODE:	**23**
PARCOM-ATMOS CODE:	**1.7**

ACTIVITIES INCLUDED

This chapter covers coke production for use in the iron and steel industry, in foundries and as smokeless fuel and the manufacture of other manufactured solid smokeless fuels such as Coalite using carbonisation processes.

GENERAL PROCESS DESCRIPTION

Coke is produced in ovens by the pyrolysis of coal. Most coke ovens are “by-product recovery” ovens (or slot ovens) and are built in a series or “battery” which contain up to 80 individual oven compartments. During operation, gas produced in some of the ovens is used to heat other ovens in the battery. The ovens are heated by an underfire furnace and the hot gases from the furnace pass through vertical heating flues in the walls of the ovens.

Coke production consists of a number of stages. Firstly, coal is prepared by crushing, screening, and blending. In some cases the coal is preheated before being charged. Charging is normally carried out using a mobile charging unit (larry car), which charges through holes in the top of the preheated ovens after which the charging ports are sealed with lids and proprietary sealants. The coal is heated for between 12 to 20 hours at temperatures of around 1100 ºC under air deficient conditions, causing volatile substances to be driven off (carbonisation or coking).

After coking is complete, the doors on both sides of the oven are removed and the coke is pushed out of the oven through a coke guide into a collecting (quench) car. The quench car moves along the battery to a quench tower where water is sprayed on to the hot coke to cool it. The car then discharges the coke onto a wharf to drain and continue cooling. When cooling is complete gates on the wharf are opened to allow the coke to fall on to a conveyor and the material is carried away for crushing and screening.

Approximately 1.6 tonnes of coal are used to produce 1 tonne of coke.

Some smokeless fuels are manufactured using similar carbonisation processes. Coalite is produced by low temperature (640°C) carbonisation of coal in cast iron retorts. As with coke production, evolved gases from the coal during carbonisation is used as a fuel, to heat the retorts and to raise steam. The retorts are lidded and a water seal is used to prevent gases escaping to atmosphere. After the carbonisation process is complete, the Coalite product is first allowed to cool in a cooling chamber below the retorts and then quenched with water sprays. Finally, the product is screened, given a final quench and sent for despatch. Other smokeless fuels are produced by blending and pressing of raw materials such as coke, anthracite and petroleum coke, sometimes followed by a low temperature (150-300°C) heat treatment stage.

SOURCES OF PARTICULATE EMISSIONS

The potential sources of particulate matter release to atmosphere in the coke production process are:

- Coal crushing
- Oven charging
- Oven door leaks
- Pushing operations
- Coke quenching
- Coke handling, crushing and screening
- Combustion of coke oven gas

The manufacture of solid smokeless fuels by carbonisation processes gives rise to emissions from similar sources. Other smokeless fuels, manufactured by blending and pressing are likely to give rise to relatively trivial emissions and are not considered further.

SOURCES OF HEAVY METAL EMISSIONS

Coal contains trace amounts of metallic species which are released during carbonisation. These elements can include arsenic, cadmium, chromium, copper, nickel, mercury, lead and zinc. The elements emitted and their quantities will vary between coal types and sources.

The potential sources of heavy metal emissions from carbonisation operations are similar to those identified for particulate matter emissions as the heavy metals will be predominantly bound to the particulate matter, particularly the smaller size fractions (e.g. $<PM_{10}$). Accordingly the abatement techniques identified for particulate matter will also largely apply to particle borne metallic compounds. However, some of the more volatile metallic species, notably mercury, may also have a significant presence in the vapour phase and hence may be released as fugitive emissions or in the coke oven gas.

ABATEMENT MEASURES AVAILABLE

Releases of particulate material from coal crushing operations are normally controlled by cyclones and for coal preheating operations (where this occurs), wet scrubbers and wet electrostatic precipitators have been used with good effect.

Abatement technology solutions for oven charging are more difficult although collection systems have been developed and are installed at US plants which utilise a wet scrubber. For many plants the answer has been to improve the operational strategy. Charging “on the main”, i.e. with the oven connected to the gas collecting main is the best method of control. This is most effective when coupled with a system of sequential charging, i.e. controlling the rate and sequence by filling through different charge holes.

Oven door and battery top leaks can contribute substantially to the release of particulate fugitive emissions. Good operational practice is essential in order to minimise leakage and therefore emissions. A great deal of effort has gone into oven door design in recent years to reduce leaks. The important control features include:

- use of good oven door and door frame design
- the use of effective insulation of doors and door frames
- keeping seals clean by use of water jets
- regular maintenance.

Pushing operations can give rise to a significant generation of dust. This can be made worse by the production of dark smoke and fume if carbonisation is not complete. Consequently paying stricter attention to the operational control of the coking process has proved to be a useful abatement strategy for reducing emissions for this process stage. There have also been some attempts at total enclosure on the coke side of the battery in the US and Germany but this has not been totally successful. Mobile collection and scrubbing systems mounted on the collection car are also in use. The most effective system currently employed involves the use of a fume collection hood mounted on the coke guide car and extending over the coke collection car. Fumes are ducted to a land based particulate removal system using fabric filters, electrostatic precipitator or venturi scrubber.

Uncontrolled quenching of the coke can potentially give rise to high releases of particulate matter. Dry quenching is carried out in some parts of Europe and the former USSR and this can produce significant emissions of dry coke dust. Water quenching is more normal and particulate release can be controlled by a number of techniques involving grit arrestors and automatic backwash sprays to keep the grit arrestors clear. Washing with clean water gives the best results.

Combustion stack emissions could contain significant amounts of particulate matter. These releases are normally controlled through efficient underfiring and attention to minimising cross-wall leakage.

Coke handling operations can give rise to small releases of coarse particulate matter. Emissions are normally minimised by enclosure and minimising transfer points.

UK SITUATION

UK plants

Most coke ovens operating in the UK are "by-product recovery" ovens (or slot ovens) as previously described. Most ovens are located adjacent to steelworks (Llanwern, Port Talbot, Scunthorpe and Teesside) and produce "metallurgical" coke, which is used as part of the steel-making process. There are also coke plants producing metallurgical coke at Llantwit Fardre, near Pontypridd, and Monckton, near Barnsley, and two plants producing smokeless fuel by a carbonisation process at Bolsover and Barnsley. There are no coke plants in Scotland or Northern Ireland.

UK activity statistics

In the UK the consumption of coal by and the production of coke and other manufactured smokeless fuels is reported annually in the Digest of UK Energy Statistics. During 1999, the UK production of coke oven coke was 5.837 Mt, production of coke breeze was 0.033 Mt, and the production of other manufactured smokeless fuels was 0.635 Mt.

Abatement measures used in UK plants

Most coal crushing operations in the UK are controlled using cyclones but some employ fabric filters. No UK facilities employ coal preheating. Oven charging operations are controlled using the "on-the main" approach and sequential charging, but additional installed abatement technology (e.g. wet scrubbers) is not employed. However, some installations employ smokeless charging facilities.

Oven door leaks are tightly controlled in the UK by the use of the good operational practice described earlier and demanding requirements are achievable for tops and door leak control factors.

Particle releases from pushing operations are controlled partly by means of good operational control of the coking process and partly by engineered measures, namely a fume collection hood which is mounted on the coke guide car and extends over the coke collection car. Fumes are ducted to a land based particulate removal system using fabric filters or venturi scrubbers. Draught hoods with water sprays are also used in the UK for coke side dust arrestment. These have much lower capital cost and use the natural draught, avoiding the large energy consumption of extracted systems.

Water quenching is used in the UK and particulate release is controlled by grit arrestors and automatic backwash sprays. Washing is carried out with clean water.

Combustion stack emissions are normally controlled by efficient under-firing and attention to minimising cross-wall leakage. Emissions from coke handling operations are normally minimised by enclosure and minimising transfer points.

Emissions from the manufacture of Coalite are controlled using cyclones (for emissions from quenching) and wet scrubbers (screening area).

PARTICULATE EMISSION FACTORS

For coke production, the Environment Agency's Pollution Inventory gives the total emission of particulate matter from all UK coke plants in 1999 as 559 t and the emission of PM_{10} as 328 t (figure for 6 out of 7 sites). The corresponding figures for 1998 were 610 tonnes and 317 tonnes (figures available for 6 out of 7 plants). The PM_{10} emissions from the remaining site can be estimated by applying a PM_{10} factor of 59% PM for 1999 and 54% for 1998, both figures being derived from the data for the 6 plants reporting both particulate matter and PM_{10} in each year. This gives revised PM_{10} emissions of 342 tonnes in 1999 and 330 tonnes in 1998. Given that the coke production from these plants was 5870 ktonnes in 1999 (6215 ktonnes in 1998), the Pollution Inventory data imply an emission factor of 95 g/tonne (coke produced) for total particulate matter and 58 g/tonne (coke produced) for PM_{10} (figures for 1998 are 98 g/t (coke produced) for total particulate matter and 53 g/t (coke produced) for PM_{10}).

The EIPPCB (2000a) presents a set of dust emission factors from various operations at coke oven plants (Table A1.1). The "overall" emission factor is based on a selection of 11 coke oven plants in four EU states. The emission factors for individual operations are compiled by EIPPCB from a number of reference sources.

Table A1.1 Dust emission factors (after abatement) for coke operations (EIPPCB, 2000a)

Process	**Abatement technology**	**Emission factor g/t (LS)**[a]	**Emission factor g/t (coke)**[a]
Overall	Current	17 – 75	50 – 220
Charging	"Old plants"	1 – 1.5	3 – 4.5
Charging	Not specified	0.1 – 3.5	0.3 – 1
Carbonisation: doors	"Old plants"	4	12
Carbonisation: doors	Not specified	0.1 – 2	0.3 – 6
Carbonisation: lids	Not specified	0.06 – 0.3	0.18 – 0.9
Carbonisation: ascension pipes	Not specified	< 0.07	< 0.2
Pushing	"Old plants"; unabated	135 – 200	400 – 600
Pushing	Not specified	< 2 70 0.3[b]	< 6 200 0.9[b]
Quenching	"Old plants"	20 – 40	60 – 120
Quenching	Not specified	up to 45 5[b]	up to 130 15[b]
Sieving/screening	Not specified	2	6

[a] LS = liquid steel (crude steel). Conversion factors (weighted average of all European blast furnaces and basic oxygen steelworks): 0.358 t (coke)/t (pig iron); 0.94 t (pig iron)/t (LS).
[b] EIPPCB (2000) quotes two or three values from different references.

Table A1.2 shows the total particulate emission factors quoted by US EPA (2000) for the various stages in the manufacture of coke and Table A1.3 lists the total particulate emission factors from the EMEP/CORINAIR (1999) Emission Inventory Guidebook. Although the US EPA data was issued in 2000, most of the source data considerably predates this. Where quoted, an estimate has been given for the PM_{10} and $PM_{2.5}$ emissions, after taking account of the abatement equipment employed. The EMEP/CORINAIR data is also fairly old, being taken from German, US and Dutch sources dating from 1989 and 1990.

Table A1.2 Coke manufacture particulate emission factors (US EPA, 2000)

Process	Abatement	Emission factors g/t (coke produced)			Quality rating[a]
		Total	PM10	PM2.5	
Coal crushing	Multicyclones	55	n.d.	n.d.	D
Coal preheater	Uncontrolled	1,750	1,700	1,000	C
	Scrubber	125	120	100	C
	Wet ESP	6	n.d.	n.d.	C
Oven charging	Uncontrolled	240	n.d.	n.d.	E
	Sequential charger	8	4	3	E
	Scrubber	7	n.d.	n.d.	E
Oven door leaks	Uncontrolled	270	n.d.	n.d.	D
Oven pushing	Uncontrolled	580	250	100	B
	ESP	225	n.d.	n.d.	C
	Venturi scrubber	90	80	70	D
	Baghouse	45	n.d.	n.d.	D
	Scrubber	36	12	11	C
Quenching (dirty water)	Uncontrolled	2,620	600	510	D
Quenching (clean water)	Uncontrolled	570	170	60	D
Quenching + baffles (dirty water)	Uncontrolled	650	210	130	B
Quenching + baffles (clean water)	Uncontrolled	270	30	20	B
Combustion stack (coke oven gas)	Uncontrolled	234	220	220	A
	ESP	55	n.d.	n.d.	D
	Baghouse	55	n.d.	n.d.	D
Combustion stack (blast furnace gas)	Uncontrolled	85	n.d.	n.d.	A
	ESP	46	23	20	B
Coke screening / handling	Multicyclones	3	3	3	D

[a] Quality rating for total particulate matter. Quality ratings for PM_{10} and $PM_{2.5}$ are given in Table A1.6.

Table A1.3 Emission factors for particulate from coke production (EMEP/CORINAIR 1999)

Process	Abatement	Emission factor g/t (coke produced)	Quality rating
All processes	–	500 to 900	E
All processes	–	53	E

Process	**Abatement**	**Emission factor g/t (coke produced)**	**Quality rating**
All processes	–	800 to 5,000	E
Coal handling	–	80 to 2,500	C
Coal handling	–	150	C

Comparing the various emission factors, there seem to be two sets of ranges: the US EPA and two of the EMEP/CORINAIR estimates are in the approximate range 500 – 1,500 g/t (coke), while the Pollution Inventory, EIPPCB and the third EMEP/CORINAIR reference are an order of magnitude lower, in the approximate range 50 – 200 g/t (coke). It is notable that the larger figures generally date from considerably earlier than the lower figures, and most likely simply reflect old plant and superseded regimes. The BREF note lends support to this, as it gives a range of 50 – 220 g/t (coke produced) for current plants and about 600 g/t (coke produced) for 'old' plants.

HEAVY METAL EMISSION FACTORS

For coke production, EMEP/CORINAIR provide emission factor ranges for a number of metals. These data are presented in Table A1.4 and appear to be predominately based on a range of European and American data dating from the 1978 to 1995.

Table A1.4 Heavy metal emission factors for coke works (EMEP/CORINAIR)

Compound	**Emission factor [g/t coke]**	**Data Quality**	**Abatement type**
Arsenic	0.003 - 0.03, 0.32	E	n. a.
Cadmium	0.0007 - 0.8	E	n. a.
Chromium	0.17 - 0.34	E	n. a.
Copper	0.09 - 0.05	E	n. a.
Mercury	0.004 - 0.04	E	n. a.
Nickel	0.065 - 0.19	E	n. a.
Lead	0.08 - 0.6, 2.85	E	n. a.
Zinc	0.22 - 0.58, 6.49	E	n. a.

Table A1.5 shows emission factors calculated for the NAEI by AEA Technology using Pollution Inventory data for 1999 compared with Corus' emission factors for PI reporting in 1998 and 1999 and expressed in terms of emissions per tonne of coke produced.

Table A1.5 Comparison of heavy metal emission factors derived from Pollution Inventory data and those supplied by Corus

Metallic species	**Corus 1998/1999 (g/tonne coke produced)**	**NAEI/Pollution Inventory (g/tonne coke produced)**
Arsenic	0.00632	0.00710
Cadmium	0.00654	0.00661
Chromium	0.304	0.323
Copper	0.0480	0.0486

Lead	0.548	0.551
Mercury	0.0037	0.00377
Nickel	0.181	0.181
Zinc	0.548	0.551

The figures derived from the Pollution Inventory and those provided by Corus agree closely with the CORINAIR data and it is likely that the Corus data are based on the CORINAIR figures. The EIPPCB do not include any heavy metal emission factors in their BREF note for the iron and steel industry (EIPPCB, 2000a). Similarly, the US EPA do not provide metal emission factors in AP-42 (US EPA, 2000).

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from the various stages of coke production. These are reproduced in Table A1.6..

There are no size distributions for several key processes and, where size distributions are available for significant processes, they vary considerably. This makes it difficult to come up with an overall size distribution. It may be estimated that PM_{10} makes up roughly half of total particulate (this also agrees with the Pollution Inventory data described above). The emissions of $PM_{2.5}$ are only slightly below those of PM_{10}.

Table A1.6. Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Coal preheating — uncontrolled	0.5	44	800	D
	1	48.5	800	
	2	55	1,000	
	2.5	59.5	1,000	
	5	79.5	1,400	
	10	97.5	1,700	
	15	99.9	1,700	
		100	1,700	
Coal preheating — venturi scrubber	0.5	78	100	D
	1	80	100	
	2	83	100	
	2.5	84	110	
	5	88	110	
	10	94	120	
	15	96.5	120	
		100	120	

Appendix A1: Coke ovens

Process	**Particle size (µm)**	**Cumulative mass % < stated size**	**Cumulative mass EF (g/t)**	**Quality rating**
Oven charging	0.5	13.5	1	E
	1	25.2	2	
	2	33.6	3	
	2.5	39.1	3	
	5	45.8	4	
	10	48.9	4	
	15	49.0	4	
		100	8	
Coke pushing — uncontrolled	0.5	3.1	20	D
	1	7.7	40	
	2	14.8	90	
	2.5	16.7	100	
	5	26.6	150	
	10	43.3	250	
	15	50.0	290	
		100	580	
Coke pushing — venturi scrubber	0.5	24	20	D
	1	47	40	
	2	66.5	60	
	2.5	73.5	70	
	5	75	70	
	10	87	80	
	15	92	80	
		100	90	
Mobile scrubber car	1	28	10	D
	2	29.5	11	
	2.5	30	11	
	5	30	11	
	10	32	12	
	15	35	13	
		100	36	
Quenching — uncontrolled (dirty water)	1	13.8	360	D
	2.5	19.3	510	
	5	21.4	560	
	10	22.8	600	
	15	26.4	690	
		100	2,620	
Quenching — uncontrolled (clean water)	1	4	20	D
	2.5	11.1	60	
	5	19.1	110	
	10	30.1	170	
	15	37.4	210	
		100	570	

Process	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Quenching — with baffles (dirty water)	1	8.5	60	
	2.5	20.4	130	
	5	24.8	160	
	10	32.3	210	
	15	49.8	320	
		100	650	
Quenching — with baffles (clean water)	1	1.2	3	D
	2.5	6	20	
	5	7	20	
	10	9.8	30	
	15	15.1	40	
		100	270	
Combustion stack — uncontrolled	1	77.4	180	
	2	85.7	200	
	2.5	93.5	220	
	5	95.8	220	
	10	95.9	220	
	15	96	220	
		100	230	

RECOMMENDED EMISSION FACTORS

Particulate matter

For coke production, the NAEI uses a PM_{10} emission factor of 56 g/tonne (coke produced) for 1999 and 63 g/t (coke produced) for 1998, based on the Pollution Inventory calculations given above. The same source has been used to yield emission factors for total particulate matter of 95 g/t (coke produced) for 1999 and 116 g/t (coke produced) for 1998. There is, however, a small error in the calculation of these factors and the Pollution Inventory data actually yield the factors quoted earlier. These factors are within the range of factors for modern plant found in EIPPCB (2000a), close to the mid-point. We recommend that the existing NAEI approach of using the Pollution Inventory data should be retained, although some corrections are needed to the calculation of emission factors. Some consideration will, however, need to be given as to whether the current NAEI practice of including Pollution Inventory data for coke production as well as calculating particulate matter emissions for fuel combustion by coke manufacturers using emission factors and fuel use data might lead to a double counting of emissions.

Consideration should be given to using a higher emission factor figure for the earlier part of the NAEI time series, particularly prior to the introduction of the Integrated Pollution Control regime. In the discussion above on particulate matter emission factors it was concluded that emission factors for old plant were within a range of 500 – 1500 g/tonne (coke produced). It is recommended that a particulate matter emission factor of 1000 g/tonne (coke produced) is used for the period to 1993 (1994 will be taken as the start date of improvements introduced under IPC). Based on the US EPA size distribution for uncontrolled pushing and uncontrolled quenching, it is recommended that a figure of 25% is adopted for the sub 10 micron fraction, giving an emission factor of 250 g/t (coke produced).

For solid smokeless fuel (SSF) production, the NAEI uses a PM_{10} emission factor of 210 g/t (SSF produced). This is derived from the US EPA emission factor for quenching with baffles and dirty water (see Table A1.2). This is not applicable to the current UK situation where clean water is used for quenching, and in any case the US data are rather old. It is therefore recommended that the current methodology be replaced by one based on use of data from the Pollution Inventory (although a lack of detailed production statistics may hinder the calculation of robust emission factors).

Heavy Metals

For heavy metals the NAEI uses emission factors derived from reported Pollution Inventory data and coke production data. The factors used show broad agreement with the CORINAIR data. Accordingly, continued use of the Pollution Inventory data for the NAEI is recommended. As with particulate matter factors some small corrections need to be made to the calculations for heavy metal emission factors. The emission factors calculated this way should also be extrapolated back using the same approach as recommended above for particulate matter to give a pre-1993 emission factor.

For production of solid smokeless fuels, the NAEI uses EMEP/CORINAIR emission factors for coke production. This approach is not particularly satisfactory, however there is currently no data in the Pollution Inventory with which to compile alternative factors. Some consideration might be given however to calculating factors from the existing Pollution Inventory data for particulate matter, by assuming that the metal content of dusts from solid smokeless fuel manufacture would be the same as dusts from coke manufacture.

Appendix A2: Sinter production

SNAP CODE:	**030301** **040209**
SOURCE ACTIVITY TITLE:	**Sinter & pelletizing plants**
NACE CODE:	**27**
PARCOM-ATMOS CODE:	**2.1.1**

ACTIVITIES INCLUDED

This chapter covers sinter plants in the iron and steel industry only. Some non-ferrous metal processes include sinter production and this has been included in the sections dealing with those processes.

GENERAL PROCESS DESCRIPTION

Blast furnaces need raw material with the following properties:

- It must have the form of large irregular lumps to so as to allow pathways for air flow through the fully charged furnace.
- The lumps must contain, intimately mixed together, all the reagents needed by the blast furnace reactions.
- The lumps must be mechanically strong enough not to be crushed under the weight of material in the fully charged furnace.

It is the sintering process that converts the fine-sized basic raw materials into this form.

In the sintering process iron ore, coke and limestone, together with small amounts of fluxes and recycled materials from the sinter plant and other parts of the steelworks, are crushed, sieved, mixed and placed on a continuous, travelling grate called a sinter strand. A burner at the beginning of the strand ignites the coke in the mixture after which the combustion is self supporting. Underneath the strand are windboxes that draw air and combustion gases down through the bed into a duct.

Fused sinter from the end of the strand is crushed and screened. Undersize material is recycled to the mixing mill and back to the strand. The remaining sinter product is cooled in the open air or in a cooler with mechanical fans. Air or water is used as the cooling medium in these coolers, with air being prevalent in newer plants and water being dominant in older plants (EMEP/CORINAIR 1999).

The cooled sinter is crushed and screened for a final time, then the fines are recycled, and the product is sent to be charged to the blast furnaces.

Table A2.1 shows typical data for plants operating in western Europe.

Table A2.1 Range of technical parameters of European sinter plants (EMEP/CORINAIR 1999)

Parameter	Range
Width of strand	2.5 – 4.5 m
Area of strand[a]	50 – 400 m^2
Specific wind box flue gas flows	1,800 – 2,000 Nm^3/t sinter
Flue gas flows	up to 1.5 million m^3/h
Height of sinter layer	approx. 250 – 650 mm
Coke input	38 – 55 kg/t sinter

[a] Some small operations are reported to be in operation in Poland, another one in Germany (sintering of iron containing return and filter materials)

An alternative process, currently available, is pelletisation. New technologies called “converted blast furnace” or “melting-reduction technology” are being developed but are not expected to be commercially operational before 2010. For this process sintering, pelletisation, and coke input will no longer be necessary (EMEP/CORINAIR 1999).

SOURCES OF PARTICULATE EMISSIONS

The main particulate emission sources in a sinter plant are gases from the windboxes — these contain considerable amounts of entrained particulate matter, some of which is released to air via the main stack, after passing through the particulate air pollution control (APC) equipment.

The steps for crushing, raw material handling, belt charging and discharging from the breaker and hot screens, all involve the generation of considerable amounts of entrained particulate matter. The potential emissions are normally ducted to a separate dust removal system and discharged through a separate stack, but they may be passed to the main control system and vented through the main stack. Some fugitive emissions could also occur.

Handling and transportation of the raw materials, including discharge of collected dust from the APC collection hoppers and of the cooled sinter — mainly fugitive emissions.

SOURCES OF HEAVY METAL EMISSIONS

The raw materials used in the sintering process contain heavy metals which may be volatilised (particularly lead, zinc and tin) and may form or be adsorbed onto fine particles during the sintering process. Accordingly the heavy metal emissions from sintering plant are predominantly associated with particulate emissions and therefore can be found concentrated in APC residues. However, those fine particles that are able to pass through the APC equipment may have a much higher content of these metals than the raw gas dust or the sinter mixture. In particular, during the sintering process, lead reacts with chlorides to form very fine crystals of lead chlorides which are able to pass through most ESPs.

ABATEMENT MEASURES AVAILABLE

Sinter strand windbox emissions are commonly controlled by cyclones, dry or wet electrostatic precipitators (ESPs), high pressure drop wet scrubbers, or baghouses. Crusher and hot screen emissions are usually controlled by ESPs.

UK SITUATION

UK plants

There are sinter plants at the four iron and steel plants in the UK: Llanwern, Port Talbot, Scunthorpe and Redcar, all operated by Corus UK Ltd. Table A2.2 lists these together with the number and size of the sinter strands in use.

Table A2.2. Sinter plant in existence at the end of 1997

Site	No of strands	Strand width (m)	Total useful grate area (m^2)
Redcar	1	4.0	336.0
Scunthorpe	2	4.0	582.0
Llanwern	1	2.9	180.0
Llanwern	1	2.5	120.0
Port Talbot	1	4.0	336.0

UK activity statistics

No production statistics are available for the UK, although pig iron production statistics are published annually by the Iron and Steel Statistics Bureau. We recommend that a suitable factor be sought from Corus in order to estimate sinter production from these data, although as a default, a value of 1.16 t (sinter)/ t (pig iron) be adopted, which is a weighted average of all European blast furnaces, given by the EIPPCB (EIPPCB, 2000a).

Abatement measures used in UK plants

UK plants all employ modern dry electrostatic precipitators (ESP) to control the air releases of particulate matter. The strand windbox emissions are passed through an ESP to vent at the main stack, whilst the other emissions (e.g. from the crusher) are gathered together and passed through a separate ESP and are normally discharged from a separate stack.

PARTICULATE EMISSION FACTORS

The Pollution Inventory contains emissions data for integrated steelworks however the emissions from the sintering process are not given separately so no emission factor can be calculated.

The EIPPCB (2000a) presents a set of emission factors from various sinter operations (Table A2.3). These are based on data from five sinter plants from five EU states. The total range for the whole sintering process is 230 – 1,200 g/t (sinter). Assuming sinter cooling is controlled with bag filters, a central value is roughly 310 g/t (sinter). This is for dust.

Table A2.3 Dust emission factors (after abatement) for sinter operations (EIPPCB, 2000a)

Process	Abatement technology	Emission factor g/t (LS)[a]	Emission factor g/t (sinter)[a]
Crushing/blending	Not specified ("after abatement")	<5	<4.5
Sinter strand	Not specified ("after abatement")	220 (100 – 480)	200 (90 – 440)
Discharge zone	Not specified ("after abatement")	10 – 270	9 – 250
Sinter cooling	Cyclones	100 – 450	90 – 410
Sinter cooling	Bag filters	40 – 110	37 – 100

[a] LS = liquid steel (crude steel). Conversion factors (weighted average of all European blast furnaces and basic oxygen steelworks): 1.16 t (sinter)/t (pig iron); 0.94 t (pig iron)/t (LS).

Table A2.4 lists various total particle emission factors quoted by the US EPA (2000) for the sintering process. Where quoted, an estimate has been given for the PM_{10} and $PM_{2.5}$ emissions, after taking account of the abatement equipment employed.

From the US EPA data it can be seen that all total particulate emission factors for controlled windbox releases are less than 1,000 g/t. The factor quoted for the dry ESP is surprisingly high at 800 g/t and is perhaps an error, given that the factor for venturi scrubbers is given as 235 g/t. A much more reasonable figure for the dry ESP would be expected to be around 200 g/t. Particulate matter will also be released at the sinter discharge, e.g. the crusher, and a range of particulate control technologies are employed in the US.

Given all this, the US EPA data suggest a particle emission factor for the whole sintering process in the US of roughly 400 g/t.

Table A2.4 Particulate emission factors for sintering as part of iron and steel production (US EPA, 2000)

Process	Abatement technology	Emission factor g/t (finished sinter)			Quality rating[a]
		Total	PM10	PM2.5	
Windbox	Uncontrolled (leaving gate)	5,560	830	280	B
Windbox	Uncontrolled (after coarse particles removal)	4,350			A
Windbox	ESP (dry)	800			B
Windbox	ESP (wet)	85	50	28	B
Windbox	Venturi scrubber	235	230	210	B
Windbox	Cyclone	500	370	260	B

Process	**Abatement technology**	**Emission factor g/t (finished sinter)**			**Quality rating**[a]
		Total	**PM10**	**PM2.5**	
Sinter discharge	Uncontrolled	3,400			B
Sinter discharge	Baghouse	50	16	6	B
Sinter discharge	Venturi scrubber	295			A
Wind box and discharge	Baghouse	150	100	40	A

[a] Quality factor for total particulate matter. Quality factors for PM_{10} and $PM_{2.5}$ are given in Table A2.8.

Table A2.5 lists total particulate matter emission factors from the EMEP/CORINAIR (1999) emission inventory guidebook. The total emission factor for the whole sintering process is about 450 g/t (sinter), assuming the best listed abatement technologies are used.

Table A2.5. European emission factors for dust (EMEP/CORINAIR 1999)

Process	**Abatement type**	**Abatement efficiency**	**Emission factor (g/t sinter)**	**Quality rating**
Sintering	Unabated	0	4,000	C
Sintering	Cyclones	60-70 %	1400	E
Sintering	ESP	>90 %	300	C
Cooling	Unabated	0	3,500	C
Cooling	Cyclones	60-70 %	3,000	E
Cooling	Multicyclone	>90 %	50	C
Handling	ESP, bag filters	> 90 %	100	D

The Corus (2000) suggest a typical total particulate emission factor of about 330 g/t, with a PM_{10} factor of about 250 g/t. These are based on the mean of the five UK sinter plants and making some assumptions about sinter cooler emissions.

HEAVY METAL EMISSION FACTORS

The Pollution Inventory contains emissions data for integrated steelworks however the emissions from the sintering process are not given separately so no emission factor can be calculated. However, Corus (2000) have provided emission factors based on source measurements for two sinter plant (Redcar and Scunthorpe) which are used to calculate the emissions for reporting to the Pollution Inventory.

The EIPPCB (2000a) provides a set of emission factors from various sinter operations. These are based on data from five sinter plants from five EU states and are provided for comparison with the Corus' data and current NAEI factors in Table A2.6.

Table A2.6 Metal emission factors from sinter plant (g/t liquid steel)

Element	Emission factor EIPPCB	Emission Factor Corus[c]	Emission Factor NAEI[c]
Arsenic	-	0.0068	0.0085
Antimony	-	0.0002	-
Beryllium	-	0.0001	-
Cadmium	0.002-0.04[a]	0.035	0.021
Chromium	0.005-0.05[a]	0.006	0.085
Copper	0.007-0.16[a]	0.20	0.34
Mercury	0.016-0.149[b]	0.02	0.0070
Manganese	0.02-0.4[a]	0.12	-
Nickel	0.002-0.04[a]	0.009	0.13
Lead	0.04-7[a]	3.42	0.70
Selenium	-	0.17	0.018
Thallium	0.005-0.03[a]	0.053	-
Vanadium	0.005-0.02	0.002	0.42
Zinc	0.002-1.8[a]	0.279	0.85

[a] Lower figure applicable to quench & fine scrubber system after ESP or ESP followed by bag filter

[b] Higher value when Hg is present in the iron ore in appreciable amounts

[c] Corus data were supplied as emissions per tonne of sinter and NAEI data were originally expressed as emissions per tonne pig iron. These have been converted to emissions per tonne of liquid steel assuming 1.16 t (sinter) / t (pig iron) and 0.94 t (pig iron) / t (liquid steel)

Comparison of the factors used by Corus with those from EIPPCB demonstrate that the Corus factors are generally within the ranges of the EIPPCB figures. However, the NAEI emission factors do not compare well with either of the other sources. The NAEI has used factors from a combination of sources. Where the NAEI emission factor exceeds the BREF and Corus factors, these factors are based on circa 1990's data and where the factors are lower than those given by BREF and Corus, they are based on data given by Entec (1996) with the exception of selenium which is again based on circa 1990's data.

The EMEP/CORINAIR Emissions Inventory Guidebook (1999) gives emission factors obtained from a range of sinter plants in the western European countries that are commonly equipped with particulate abatement facilities. These data are summarised in Table A2.7.

Table A2.7 Emission factors for heavy metals (EMEP/CORINAIR, 1999 which contains data deriving from *ca.* 1990)

Compound	Emission factor [g/ t sinter]	Data quality
Arsenic	0.05	C
Cadmium	0.2	C
Chromium	0.2	C
Copper	0.4	C
Mercury	0.05	C
Nickel	0.2	C
Lead	8	C

Selenium	0.02	C
Zinc	1	C

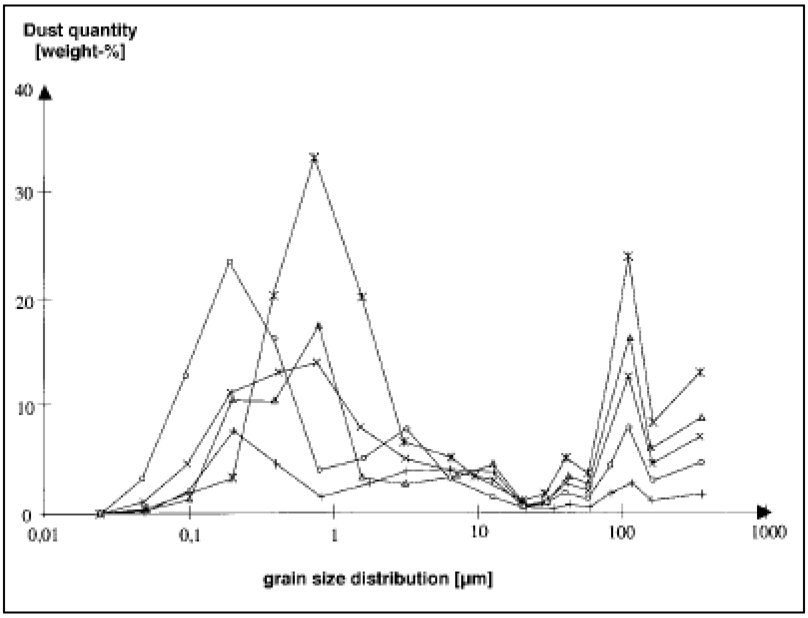
Further comparison of the Corus and EIPPCB emission factors for the non-volatile metal species with the EMEP/CORINAIR figures shows that the EMEP/CORINAIR emission factors are greater for the non-volatile metals (arsenic, chromium, copper and nickel). The EMEP/CORINAIR factors for the relatively volatile metals (selenium, lead, mercury and zinc) show broad agreement with the Corus and EIPPCB data, with the exception of the factor for cadmium which is at least an order of magnitude higher than that given by the Corus and EIPPCB data. These differences may be explained by improvements in particle abatement since the early1990's.

Further examination of the detailed data tables given within the EMEP/CORINAIR Guidebook show cadmium emission factors for European plant ranging from 0.22 to less than 0.001 g/t sinter. The value of 0.036 g/t steel used by Corus is within the middle of this range.

SIZE DISTRIBUTION

The EIPPCB (2000a) includes a graph showing the grain size and weight distribution of dust, based on samples from a number of sinter strands (Figure 2.1). There are two distinct maxima, one in the range 0.1 – 3 µm, one close to 100 µm. The coarse dust can be separated in ESPs with high efficiency. However, the composition of the fine dust, alkali chlorides, reduces the efficiency of ESPs.

Figure 2.1 Particle size distributions (EIPPCB, 2000a)



US EPA (2000) gives size distributions for particulate matter emitted from the various stages of sinter production. These are reproduced in Table A2.8. Ignoring the size distribution for uncontrolled windbox emissions, the average proportion of sub 10 micron material is 66% by weight and, in the absence of more applicable data, this factor could be used for the NAEI.

Table A2.8. Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Windbox	uncontrolled (leaving gate)	0.5	4	220	D
		1	4	220	
		2.5	6.5	280	
		5	9	500	
		10	15	830	
		15	20	1,110	
			100	5,560	
Windbox	ESP (wet)	0.5	18	15	C
		1	25	21	
		2.5	33	28	
		5	48	41	
		10	59	50	
		15	69	59	
			100	85	
Windbox	Venturi scrubber	0.5	55	129	C
		1	75	176	
		2.5	89	209	
		5	93	219	
		10	96	226	
		15	98	230	
			100	235	
Windbox	cyclone	0.5	25	130	C
		1	37	190	
		2.5	52	260	
		5	64	320	
		10	74	370	
		15	80	400	
			100	500	
Windbox	baghouse	0.5	3	5	C
		1	9	14	
		2.5	27	41	
		5	47	71	
		10	69	104	
		15	79	119	
			100	150	
Discharge breaker and hot screens	baghouse	0.5	2	1	C
		1	4	2	
		2.5	11	6	
		5	20	10	
		10	32	16	
		15	42	21	
			100	50	

RECOMMENDED EMISSION FACTORS

Particulate matter

The four data sources presented above broadly agree that the total emission of dust using the best technology is about 350 g/t (sinter), within a factor of 2. Information on PM_{10} is sparse.

The NAEI uses a PM_{10} emission factor of 197.5 g/t (pig iron). This is derived by assuming that emissions of particulate matter are 250 g/t (sinter) and that the PM_{10} fraction is 79% (see Table A2.8 above). The particulate matter emission factor is nominally taken from the US EPA, and represents an emission factor for total particulate for windbox and discharge controlled by baghouse. However, this is erroneous, since the actual US EPA figure would be 150 g/t.

Given the agreement in other data sources, the NAEI factor appears to be too low in any case. Although quantitative estimates of PM_{10} factors are sparse, applying a factor of 66% (see above) to a total particulate factor of 350 g/t, a figure of about 250 g/t seems reasonable. Data in the Pollution Inventory are too aggregated to use for specific iron and steel processes. The data supplied by the UK Steel Association (330 g/tonne (sinter) for particulate matter and 250 g/tonne (sinter) for PM_{10} agree closely with these figures and we recommend that these are used in the NAEI for recent estimates. For the earlier part of the time series, a higher figure might be more appropriate to take account of a lower degree of control, however more information is needed on control measures in place before a figure can be suggested.

Heavy metals

The emissions factors provided by EMEP/CORINAIR and largely used by the NAEI appear to be overestimates when compared with data provided by Corus and EIPPCB, and appear to be based on data from old plant with poorer particulate abatement. Conversely, where the NAEI have used data from Entec (1996), the emissions factors appear to be an underestimate. The EMEP/CORINAIR figure for selenium is also an order of magnitude lower than that reported by Corus, however there is no other available data with which to compare this figure.

Given the difficulty with disaggregating data from the Pollution Inventory and the age of the source data used in the EMEP/CORINAIR Guidebook, the use of Corus submitted factors are recommended as a basis for estimating emissions from this sector for the NAEI. These emission factors should also be modified in line with any changes in particulate matter emission factors in the earlier part of the time series.

Appendix A3: Blast furnaces

SNAP CODE: **040202**
040203

SOURCE ACTIVITY TITLE: **Blast furnace charging**
Pig iron tapping

NACE CODE: **27100**

PARCOM-ATMOS CODE: **2.1.0**

ACTIVITIES INCLUDED

The charging and operation of blast furnaces, the removal of molten iron from them (pig iron tapping), and refining operations such as desulphurisation.

GENERAL PROCESS DESCRIPTION

Blast furnaces are large vertical cylindrical refractory lined vessels. They are used for the reduction of iron bearing materials with a hot gas to produce liquid iron (known as pig iron). Pig iron is used later as the basic input for steel making processes. Blast furnaces are charged from the top with iron (as ore, pellets and/or sinter). Flux materials (as limestone, dolomite and sinter) are also added, along with coke for fuel. Preheated air is blown into the furnace from below. The charge reacts with the blast air to form molten iron, carbon monoxide and slag. The molten iron and slag collect in the hearth at the base of the furnace. The by-product gas is collected through off-takes located at the top of the furnace and is recovered for use as a fuel. At the top of the furnace there is a mechanism that allows material to be charged into the furnace without the escape of gas. The area around the base of the furnace, including all iron and slag runners, is enclosed by a casthouse.

The high cost of good quality coking coal has led many steel makers to develop systems to reduce coke consumption by direct injection of other fuels such as oil, gas or coal into the furnace.

The production of 1 tonne of iron requires 1.4 tonnes of ore or other iron bearing material; 0.5 to 0.65 tonnes of coke; 0.25 tonnes of limestone or dolomite; and 1.8 to 2.0 tonnes of air. By-products consist of 0.2 to 0.4 tonnes of slag, and 2.5 to 3.5 tonnes of blast furnace gas containing up to 45 kg of dust.

The liquid metallic iron and slag are removed or cast from the furnace periodically. During casting, molten metal flows from a furnace taphole through an iron notch and into runners that lead to transport ladles or torpedo ladles. The slag is directed through separate runners to a slag pit adjacent to the furnace or into slag pots for transport to a remote slag pit.

After tapping the molten iron is desulphurised. This operation is sometimes carried out in the ladle prior to transferring the material to a steel-making furnace. Reactants are injected into the ladle under controlled conditions. These are usually either calcium carbide, magnesium granules coated with salt or combinations of the two. The added chemicals react with the sulphur in the molten iron to form a slag that can be skimmed off the surface of the metal.

The blast furnace by-product gas, which is collected from the furnace top, contains particulate. This blast furnace gas is used as a fuel in the steel plant.

SOURCES OF PARTICULATE EMISSIONS

Particulate material emissions can arise from:

- Raw material handling
- Charging slip
- Casting house operations
- Desulphurisation

Although the top gas leaving the furnace contains up to 15 g/Nm^3 of dust (WS Atkins, 1993), this gas is not vented to atmosphere, but is instead used as a fuel in the sinter strand and in boilers. This gas contains ammonia and other compounds which need to be water scrubbed to remove them from the gas stream before it is used for fuel purposes.

The raw material handling areas, especially the returned fines section, is a major source of dust but the particle size will be quite large given the nature of the material.

Minor emissions may occur during charging due to imperfect seals but occasionally significant particulate release can be produced through “slip”. This is caused by a cavity forming in the blast furnace charge, which causes a collapse of part of the charge above it. The resulting pressure surge in the furnace opens a relief valve to the atmosphere to prevent damage to the furnace and thereby allows dust to “slip” to atmosphere. The composition of the dust is related to the composition of the charge. It is rather coarse with a mean particle size considerably greater than 10 μm.

Casthouse operations account for a significant proportion of blast furnace particulate production. Particulate matter is emitted when the molten iron and slag contact air above their surface. Drilling and plugging the taphole also generates particulate emissions. In addition, the occasional use of an oxygen lance to open a clogged taphole can cause heavy particulate emissions. During the casting operation, iron oxides, magnesium oxide and carbonaceous compounds are generated as particulate. These sources of particulate are controlled by the casthouse extraction system.

Particulate emissions also occur during the hot metal desulphurisation stage when sulphur reacts with reagents and is skimmed off. The dust contains mainly iron and calcium oxides.

SOURCES OF HEAVY METAL EMISSIONS

The sources of metal emissions are as those described above for particulate matter emissions.

ABATEMENT MEASURES AVAILABLE

Air releases occurring during charging are generally not abated and neither are those occasional releases associated with slip. These sources are considered to be relatively minor.

Particulate emissions occurring during casting operations are controlled by a number of techniques. Some facilities are uncontrolled with roof discharges. More commonly, existing facilities are controlled by evacuation through retrofitted capture hoods to a gas cleaner, or by suppression techniques. Emissions controlled by hoods and an evacuation system are usually vented to a bag-house. The basic concept of suppression techniques is to prevent the formation of pollutants by excluding ambient air contact with the molten surfaces. New facilities have been constructed with evacuated runner cover systems and local hooding ducted to a bag-house.

Particulate emissions generated during desulphurisation are normally collected by a hood positioned over the ladle and are vented to a bag-house.

UK SITUATION

UK plants

Table A3.1 lists the 10 large blast furnaces in operation in the UK at the end of 1997. These are all operated by Corus. All smaller facilities have now been taken out of operation.

Table A3.1. Blast furnaces in the UK (Iron and Steel Statistics Bureau, 1998)

Site	**Hearth Diameter**			**Total**
	≥ 8m, < 9m	**≥ 9m, < 10m**	**≥ 10m**	
Redcar			1	1
Scunthorpe	2	2		4
Llanwern		1	1	2
Port Talbot	1	1	1	3
Total	3	4	3	**10**

UK activity statistics

Information on UK pig iron production from blast furnaces are published by the Iron and Steel Statistics Bureau (1998).

Abatement measures used in UK plants

As in other parts of the world, the air releases occurring during charging and those occasional events associated with slip are generally not abated. However observations would indicate that about 10 slips occur per furnace per year and that the lifetime of the event is only about 15 seconds.

Eight out of the nine UK casthouses are tightly controlled regarding particulate air releases. These plant are fitted with local evacuation, which also covers tapholes, troughs and runners. The emissions are ducted to bag-houses where particles are efficiently removed.

In the UK desulphurisation is normally carried out in a dedicated facility, i.e. not in the transfer ladles. The particulate emissions are tightly controlled by the use of bag-houses.

PARTICULATE EMISSION FACTORS

The EIPPCB (2000a) presents a set of emission factors from various operations at blast furnaces (Table A3.2). The "overall" emission factor is based on a selection of four blast furnaces in four EU states (data from 1996).

Table A3.2. Dust emission factors (after abatement) for blast furnaces (EIPPCB, 2000a)

Process	Abatement technology	Emission factor g/t (LS)[a]	Emission factor g/t (pig iron)[a]
Overall	After abatement	10 – 50	10 – 55
Charging zone	After abatement	14 (5 – 38)[b]	15 (5 – 40)[b]
Coal preparation for injection	After abatement	12 (2 – 54)	13 (2 – 57)
BF cast house	After abatement	30 (2 – 79)	32 (2 – 84)
Hot stoves (cowpers)	After abatement	3 – 6	3 – 6

[a] LS = liquid steel (crude steel). Conversion factor (weighted average of all European blast furnaces and basic oxygen steelworks): 0.94 t (pig iron)/t (LS).

[b] Unreasonably low emission factors (< 1 g/t LS) have not been taken into account; nor have data from wet scrubbers, which have much higher emission factors (2 – 20 times higher).

Table A3.3 shows the various total particulate emission factors quoted by US EPA (2000) for blast furnace operations, including desulphurisation. Where quoted, an estimate has been given for the PM_{10} and $PM_{2.5}$ emissions, after taking account of the abatement equipment employed.

The US EPA data gives total particulate emission factors ranging from 300 to 650 g/t of pig iron for the blast furnace operations. PM_{10} emission factors are about 150 g/t, whilst $PM_{2.5}$ ranges from 70 to 100 g/t. These data do not take account of the total particulate factor of 39,500 g/t (slip) quoted for slip releases, as there is no conversion factor available to convert this to g/t (hot metal); however this element is likely to be relatively minor (see below).

For desulphurisation the total particulate factors range from 550 g/t for uncontrolled facilities to only 4 g/t for those employing fabric filters. This equates to PM_{10} factors of 100 and 3 g/t respectively, whilst the factors for $PM_{2.5}$ are 60 and 2 g/t respectively. However, emissions from desulphurisation in the UK are not necessarily comparable with other countries (e.g. the US) as in the UK this activity is performed in a dedicated facility away from the blast furnace.

Table A3.3. Blast furnace particulate emission factors (US EPA, 2000)

Process Stage	**Abatement**	**Emission factors g/t (hot metal)**			**Quality rating**[a]
		Tot	**PM_{10}**	**$PM_{2.5}$**	
Slip	Uncontrolled	39500[b]	n.d.	n.d.	D
Casthouse	Uncontrolled	300	150	70	B
Furnace with local evacuation	Not stated	650	160	100	B
Taphole and trough only (not runners)	Not stated	150	n.d.	n.d.	B
Hot metal desulphurisation	Uncontrolled	550	100	60	D
Hot metal desulphurisation	Baghouse	4.5	3	2	D

[a] Quality factor for total particulate matter. Quality factors for PM_{10} and $PM_{2.5}$ are given in Table A3.5.
[b] Unit: g/t (slip)

Estimates of UK total particulate matter emissions for slip range from 5 to 10 t per year per furnace, with about ten slips occurring for around 15 seconds for each furnace per year. This equates to a total particulate matter factor of about 2 g/t of pig iron produced. There are no PM_{10} or $PM_{2.5}$ data available for this release route, but it is likely that a high proportion of the dust will be greater than 10 μm.

The UK Steel Association (personal communication, 2000) suggest a typical total particulate matter emission factor of about 130 g/t, with a PM_{10} emission factor of about 115 g/t, based on the mean of UK plants.

HEAVY METAL EMISSION FACTORS

Iron and steel works are reported as integrated processes within the Pollution Inventory therefore it is not possible to disaggregate blast furnace emissions from this source. The EIPPCB (2000a) presents a set of emission factors from various operations at blast furnaces (Table A3.4). The "overall emission factor is based on a selection of four blast furnaces in four EU states (data from 1996).

Table A3.4 Heavy Metal emission factors (after abatement) for blast furnaces (EIPPCB, 2000a)

Metal	g/t liquid steel		g/t pig iron	
	Lower	Upper	Lower	Upper
Manganese	<0.01	0.13	<0.01	0.14
Nickel	<0.01	0.02	<0.01	0.02
Lead	<0.01	0.12	<0.01	0.13

Emission factors given for g/t of pig iron assume 940 kg of pig iron is produced per tonne of liquid steel

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from various blast furnace processes. These are reproduced in Table A3.5. None of the profiles appear to take account of control equipment and are therefore not very applicable to most UK sites. Nevertheless, the profile for furnace emissions with local evacuation would seem most appropriate.

Table A3.5 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	**Abatement technology**	**Particle size (µm)**	**Cumulative mass % < stated size**	**Cumulative mass EF (g/t)**	**Quality rating**
Slip	Uncontrolled	Total	100	5,560	D
Castshouse emissions	Uncontrolled — roof monitor	0.5	4	10	C
		1	15	50	
		2.5	23	70	
		5	35	110	
		10	51	150	
		15	61	180	
			100	300	
Furnace	Local evacuation	0.5	7	040	C
		1	9	060	
		2.5	15	100	
		5	20	130	
		10	24	160	
		15	26	170	
			100	650	
Hot metal desulphurisation	Uncontrolled	0.5	0		E
		1	2	10	
		2.5	11	60	
		5	19	100	
		10	19	100	
		15	21	120	
			100	550	
Hot metal desulphurisation	Baghouse	0.5	8	0.4	D
		1	18	0.9	
		2.5	42	1.9	
		5	62	2.8	
		10	74	3.3	
		15	78	3.5	
			100	4.5	

RECOMMENDED EMISSION FACTORS

Particulate emissions

The EIPPCB emission factors are significantly lower than the US EPA ones. This is likely to be largely because the US EPA figures are older, and therefore reflect old plant and superseded regimes. The EIPPCB figures are considered to be more appropriate for most current UK plant with the US EPA factors more applicable to the uncontrolled plant. However the EIPPCB figures do not include fugitive emissions from casthouses.

The NAEI uses a PM_{10} emission factor of 195 g/t (pig iron). This is derived from the US EPA emission factors (see Tables 3.3 and 3.5). It combines the US EPA emission factors for blast furnace with local evacuation (156 g/t), taphole and trough only (36 g/t - PM_{10} is assumed to constitute 24% of the total particulate emission), and hot metal desulphurisation controlled by baghouse (3 g/t).

The Pollution Inventory does not list emissions from blast furnaces separately from other parts of the integrated steelworks. Data in the Pollution Inventory is too aggregated to use for specific iron and steel processes.

It is recommended that a lower figure should be used in the NAEI. The figures quoted by UK Steel of 130 g/t for particulate matter and 115 g/t for PM_{10}, seem reasonable.

Heavy metal emissions

The NAEI currently uses emission factors taken either from Entec (1996) or from a German report dating from 1991 (Jockel & Hartje, 1991). These data are shown in Table A3.6.

Table A3.6: Heavy metal emission factors reported in the 1999 NAEI (g/t pig iron)

Metal	Emission factor
Arsenic	0.08
Cadmium	0.016
Chromium	0.08
Copper	0.4
Lead	0.07
Mercury	0.00675
Nickel	0.12
Selenium	0.013
Vanadium	0.4
Zinc	4.0

No alternative factors are available, except those given by EIPPCB (2000a) for nickel and lead, and the current NAEI factors are close to the midpoint of the range suggested by EIPPCB. It is therefore recommended that the current NAEI methodology is retained.

Appendix A4: Basic oxygen furnaces

SNAP CODE: **040206**

SOURCE ACTIVITY TITLE: **Basic oxygen furnace steel plant**

NACE CODE: **2.1.4**

PARCOM-ATMOS CODE: **27100**

ACTIVITIES INCLUDED

The operation of basic oxygen furnaces.

GENERAL PROCESS DESCRIPTION

Pig iron contains dissolved carbon (up to 4.5% by weight) that comes from the coke used to reduce iron ore in the blast furnace. Iron containing this much carbon is weak and brittle. The basic oxygen process removes some of the carbon to produce steel that is strong and ductile.

The process involves injecting pure oxygen into the molten iron. The oxygen reacts with dissolved carbon producing carbon monoxide that escapes from the metal. The reactions take place in a large refractory lined pear-shaped vessel called a converter. These may be up to 400 t in capacity.

The refining process can be enhanced, where necessary, by “bottom stirring” with argon gas injected through porous bricks in the bottom lining in certain phases of the process. This produces a more intensive circulation of the molten steel and an improved reaction between the gas and the molten metal. The input material is typically 75 percent blast furnace iron and up to 25 percent scrap metal.

There are two major variations of the process. Conventional furnaces have oxygen blown into the top of the furnace through a water-cooled lance. In the newer, Quelle Basic Oxygen process (Q-BOP), oxygen is injected through tuyeres located in the bottom of the furnace.

A typical basic oxygen cycle consists of the scrap charge, hot metal charge, oxygen blow (refining) period, testing for temperature and chemical composition of the steel, alloy additions and reblows (if necessary), tapping, and slagging. The full furnace cycle typically ranges from 25 to 45 minutes. In a modern steelworks, 300 tonnes of steel are produced in a 30 minute cycle.

At the end of the refining process the molten steel is conveyed to the continuous casting machine. Continuous casting, in which billets or slabs are cast direct from molten metal, largely replaces the traditional method of pouring molten steel into moulds to produce ingots which, when solidified, are reheated and rolled into slabs or billets. Continuous casting not only saves time and energy, but also improves the quality of the steel and increases the yield. However, ingot casting still has to be used for some products.

SOURCES OF PARTICULATE EMISSIONS

The most significant particulate emissions from the basic oxygen furnace process occur during the oxygen blow period. The predominant compounds emitted are iron oxides, although heavy metals and fluorides are usually present.

Charging emissions will vary with the quality and quantity of scrap metal charged to the furnace and with the pour rate.

Tapping emissions include iron oxides, sulphur oxides (if sulphur is being added), and other metallic oxides, depending on the grade of scrap used. Hot metal transfer emissions are mostly iron oxides.

SOURCES OF HEAVY METAL EMISSIONS

The sources of metal emissions are as those described above for particulate matter emissions.

ABATEMENT MEASURES AVAILABLE

Basic oxygen furnaces are equipped with a primary hood capture system located directly over the open mouth of the furnaces to control emissions during oxygen blow periods. Two types of capture systems are used to collect exhaust gas as it leaves the furnace mouth: closed hood (also known as an off gas, or OG, system) or open, combustion type hood. A closed hood fits snugly against the furnace mouth, ducting all off gas to a wet scrubber gas cleaner. The open hood design allows dilution air to be drawn into the hood (this is to improve carbon monoxide abatement).

Charging and tapping emissions are controlled by a variety of evacuation systems and operating practices. Charging hoods, tapside enclosures, and full furnace enclosures are used in the industry to capture these emissions and send them to either the primary hood gas cleaner or a second gas cleaner.

UK SITUATION

UK plants

Table A4.1 lists the UK steelworks that have basic oxygen furnaces.

Table A4.1 Oxygen converters at UK steelworks at the end of 1997

Site	Number	Average capacity per vessel (tonnes)
Teesside	3	261
Scunthorpe	3	300
Llanwern	3	189
Port Talbot	2	340

UK activity statistics

Information on UK steel production from basic oxygen furnaces is published annually by the Iron and Steel Statistics Bureau.

Abatement measures used in UK plants

In the UK, as in most countries, primary gas cleaning is undertaken using wet scrubbers. Secondary fume extraction systems operate at low level near to the converters and at roof level to capture those emissions it was not possible to capture at or near to source; these use either bag filters, wet scrubbers or electrostatic precipitators. All plants in the UK have primary and secondary extraction.

PARTICULATE EMISSION FACTORS

The EIPPCB (2000a) presents a set of emission factors from various operations at basic oxygen steel-making plants (Table A4.2). The "overall" emission factor is based on a selection of four plants in four EU states (data from 1996).

Table A4.2 Dust emission factors (after abatement) for basic oxygen furnaces (EIPPCB, 2000a)

Process	Abatement technology	Emission factor g/t (steel)
Overall	After abatement	15 – 80
Furnace operation	Full combustion	66 (10 – 200)
Furnace operation	Suppressed combustion without fuel gas recovery	74 (15 – 190)
Furnace operation	Suppressed combustion with fuel gas recovery	8 (1.5 – 16)
Hot metal desulphurisation	After abatement	1 – 7
Hot metal handling (reladling)	After abatement	1 – 7
BOF charging, tapping, deslagging, and blowing	After abatement	1 – 30
Secondary metallurgy	After abatement	0.1 – 10
Continuous casting	After abatement	0.5 – 4
Sum of "other than BOF emissions"	After abatement	20 – 80

Table A4.3 shows total particulate emission factor data quoted by US EPA (2000) for both controlled and uncontrolled stages of the basic oxygen process. For the top blown furnace the total particulate emission factor for uncontrolled operation is around 15,000 g/t (steel produced) but this is not relevant to modern plant. If all the process stages are controlled the particulate emission factor is significantly affected by the type of hood vent arrangement. When open hoods are used the total particulate factor is around 60 g/t but much lower results are obtained from closed hood arrangements. The Quelle furnace seems to exhibit lower particulate emission characteristics. Use of a baghouse reduces emissions from charging and tapping to very low levels.

Table A4.3 Basic oxygen furnace particulate emission factors (US EPA 2000)

Process Stage	**Abatement**	**Emission factors g/t (steel)**			**Quality rating**[a]
		Tot	**PM_{10}**	**$PM_{2.5}$**	
Top blown furnace melting and refining	Uncontrolled	14,250	n.d.	n.d.	B
Top blown furnace melting and refining	Open hood vented to ESP	65	n.d.	n.d.	A
Top blown furnace melting and refining	Open hood vented to scrubber	45	n.d.	n.d.	B
Top blown furnace melting and refining	Closed hood vented to scrubber	3.4	2.3	2.2	A
BOF charging	At source	300[b]	140[b]	70[b]	D
BOF charging	At building monitor	71[b]	n.d.	n.d.	B
BOF charging	Baghouse	0.3[b]	0.1[b]	0.066[b]	B
BOF tapping	At source	460	210	170	D
BOF tapping	At building monitor	145	n.d.	n.d.	B
BOF tapping	Baghouse	1.3	0.4	0.2	B
Hot metal transfer	At source	95	n.d.	n.d.	A
Hot metal transfer	At building monitor	28	n.d.	n.d.	B
BOF monitor (all sources)	–	250	n.d.	n.d.	B
Q-BOF melting and refining	Scrubber	28	19	16	B

[a] Quality rating for total particulate matter. Quality rating for PM10 and PM2.5 are given in Table A4.6.
[b] Unit: g/t (hot metal)

Corus (2000) suggest a typical total particulate emission factor of about 200 g/t, with a PM_{10} emission factor of about 170 g/t, based on the mean of UK plants. They further suggest that the EIPPCB figures above do not include sufficient allowance for fugitive emissions from the roof.

Data in the Pollution Inventory are too aggregated to use for specific iron and steel processes.

HEAVY METAL EMISSION FACTORS

EMEP/CORINAIR provide a range of heavy metal emission factors for basic oxygen furnaces. These data are presented in Table A4.4

Table A4.4 Heavy metal emission factors in g/t (EMEP/CORINAIR, 1999)

Compound	Germany	Netherlands		France	Sweden	Poland	proposed
abatement	partially abated	wet scrubbers fabric filters	partially abated	unknown	wet scrubbers fabric filters	abated	unknown
Arsenic	0.0040	0.02	0.0001	0.02	- -		0.015
Cadmium	0.031	0.003	0.024	0.002-0.05	0.04		0.025
Chromium	0.50	0.04	0.011	0.07	- 0.026	0.04-0.07	0.1
Copper	0.13	0.04	0.010	0.25	- 0.066	0.01-0.04	0.1
Mercury	-	0.004	0.002	-	0.001		0.003
Nickel	0.09	-	-	0.05	- 0.024		0.05[1])
Lead	1.30	2.3	1.08	0.9	4 4.6	0.08-0.14	1.5
Selenium	-	-	-	-	- -		0.003
Zinc	3.3	4.1	2.7	4.1	6 6.4	0.4-0.8	4
Dust	-	100	53	-	- -		-

The EIPPCB (2000a) provides a set of emission factors based on a selection of four plants in four EU states (data from 1996). The factors used by Corus are based on the BREF Note figures and these are adopted as the default factors for reporting to the Pollution Inventory. These data are presented for comparison with German (UBA) and NAEI data in Table A4.5

Table A4.5 Heavy metal emission factors for basic oxygen furnaces (g/t LS)

Metal	EIPPCB (2000a)		UBA (1990)	NAEI	Corus (2000)
	Lower	Upper			
Arsenic			0.0016	0.015	
Cadmium			0.015	0.03	
Chromium	0.01	0.36	0.23	0.1	0.05
Copper	0.01	0.04	0.059	0.1	0.02
Lead	0.13	0.9	0.55	0.89	0.2
Mercury			0	0.003	
Manganese	<0.01	1.2	0.9		0.5
Nickel			0.039	0.05	
Zinc			1.3	4	
Selenium			0	0.003	
Vanadium				0.025	

The NAEI emission factors are taken from the EMEP/CORINAIR Guidebook, with the exception of vanadium which is taken from a German report from 1991 (Jockel & Hartje, 1991), and, in the case of cadmium and lead, from data supplied by the British Iron & Steel Producers Association (1997). The values given by UBA and Corus, and those used in the NAEI generally fall within the ranges suggested in the BREF note. However, most of the UBA figures and all those suggested by Corus are lower than the NAEI figures.

Comparison of all data presented in Table A7 with the BREF note figures demonstrates good agreement. However, the current NAEI factor for lead is at the upper range of the BREF note factors and is more than four times greater than the factor used for the Pollution inventory. Accordingly consideration should be given to the revision of the NAEI factor for consistency with the Pollution Inventory.

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from various basic oxygen furnace processes. These are reproduced in Table A4.6. Because no information is available on the control techniques in use in the UK, the applicability of the various profiles is not known. However, the fraction of PM_{10} is fairly consistent, averaging 50%, and this could be used as a rough figure. The fraction of $PM_{2.5}$ is more variable between processes and therefore harder to determine an overall value for.

Table A4.6 Particle size distributions and size-specific emission factors (US EPA 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Top blown furnace melting and refining	Closed hood vented to scrubber	0.5	34	1.2	C
		1	55	1.9	
		2.5	65	2.2	
		5	66	2.2	
		10	67	2.3	
		15	72	2.4	
			100	3.4	
Basic oxygen furnace charging	At source	0.5	8	20	E
		1	12	40	
		2.5	22	70	
		5	35	100	
		10	46	140	
		15	56	170	
			100	300	
Basic oxygen furnace charging	Baghouse	0.5	3	0.009	D
		1	10	0.03	
		2.5	22	0.066	
		5	31	0.093	
		10	45	0.1	
		15	60	0.2	
			100	0.3	
Basic oxygen furnace tapping	At source	0.5	0	0	E
		1	11	50	
		2.5	37	170	
		5	43	200	
		10	45	210	
		15	50	230	
			100	460	

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Basic oxygen furnace tapping	Baghouse	0.5	4	0.052	D
		1	7	0.1	
		2.5	16	0.2	
		5	22	0.3	
		10	30	0.4	
		15	40	0.5	
			100	1.3	
Q-BOF melting and refining	Scrubber	0.5	45	13	D
		1	52	15	
		2.5	56	16	
		5	58	16	
		10	68	19	
		15	85	24	
			100	28	

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI uses a PM_{10} emission factor of 30.7 g/t (steel). This is derived from the US EPA emission factors (see Tables 4.3 and 4.6). It combines the US EPA emission factors for top blown furnace melting and refining controlled by open hood vented to scrubber (for which PM_{10} is assumed to constitute 67% of the total particulate), basic oxygen furnace charging controlled by baghouse, and basic oxygen furnace tapping controlled by baghouse. Emissions from hot metal transfer are not included.

The industry's own estimates are significantly higher than the NAEI estimate; this is likely to be because they take better account of fugitive emissions and other sources. It is therefore recommended that the industry figures (200 g/tonne (steel) for particulate matter and 170 g/t for PM_{10} (steel)) are used. We have found only one factor which is described as relating to uncontrolled emissions, and this is unlikely to be representative of UK processes even in the early part of the period covered by the NAEI. The US EPA do give a number of emission factors 'at source' and this might be taken to mean before abatement of the emissions. The total emission would be about 850 g/tonne (steel) for particulate matter and about 400 g/tonne (steel) for PM_{10} assuming that emissions from hot metal transfer are about 50% (see size distribution for tapping from basic oxygen furnaces). These factors could be used for the earlier part of the NAEI time series, prior to the Environmental Protection Act.

Heavy metal emissions

We recommend that the Corus figures should be used for copper, chromium, lead and manganese. The existing NAEI emission factor for lead, based on data supplied by the UK steel industry, may still be relevant for the earlier part of the NAEI time series. The NAEI figures for the remaining pollutants are unlikely to be any less reliable than the UBA data which are old and in any case relate to German plant only. We recommend that the existing NAEI factors for these metals are retained, unless further factors can be obtained from Corus or from the Pollution Inventory.

Appendix A5: Electric arc furnaces

SNAP CODE	**040207**
SOURCE ACTIVITY TITLE	**Electric furnace steel plant**
NACE CODE	**27100**
PARCOM-ATMOS CODE	**2.1.6**

ACTIVITIES INCLUDED

The use of electric arc furnaces as part of iron and steel production, including the use of electric arc furnaces to produce iron and steel in iron or steel foundries.

GENERAL PROCESS DESCRIPTION

Electric arc furnaces (EAFs) are used to produce carbon and alloy steels. The feedstock is mainly scrap steel from steel product manufacturers (e.g. vehicle builders) and from consumers (e.g. end of life products) and waste pig iron from steel works. EAFs are cylindrical refractory lined vessels with (usually) three carbon electrodes that can be raised or lowered through a removable furnace roof. Furnace capacities range from 5 to 350 tonnes.

With the electrodes retracted the roof can be swung to the side to permit charging directly into the furnace using an overhead crane. In some cases the scrap is preheated in a shaft or on a conveyor.

The scrap may be contaminated with paint, oil or grease and is supplied graded according to levels of contamination. Scrap is usually stored outside in large uncovered compounds on unpaved ground. Some scrap sorting is carried out to reduce the risk of including hazardous contaminants.

Other raw materials can include flux, coke or coal and alloying agents. These materials can be introduced through doors on the side of the furnace.

With the roof replaced the electrodes are lowered and energised by 3-phase alternating current. This creates arcs between the electrodes that melt the metallic charge. Oxy-fuel burners are widely employed to assist melting. Such burners are located in the furnace side walls or sometimes in the roof. As the charge melts its volume decreases, consequently another charge is added and melting continues. Once the melting cycle is complete, the electrodes are raised and the roof is removed. The vessel can then be tilted to pour the molten steel. Slag is also produced.

Appendix A5: Electric arc furnaces

The production of steel in an EAF is a batch process. Cycles, or "heats", range from about 1.5 to 5 hours to produce carbon steel and from 5 to 10 hours or more to produce alloy steel. For the production of carbon steel and low-alloyed steels, the main operations are:

- raw material handling and storage
- furnace charging (possibly with scrap pre-heating)
- melting and refining (including oxygen blowing)
- steel and slag tapping
- ladle furnace treatments (secondary metallurgy) for quality adjustment
- slag handling
- continuous casting

For high-alloyed and special steels, additional secondary metallurgy is necessary depending on the exact product requirements. Operations include:

- desulphurisation
- degassing, the removal of dissolved gases such as nitrogen and hydrogen
- decarburisation

SOURCES OF PARTICULATE EMISSIONS

The operations that can generate particulate emissions are charging the scrap, melting and refining, tapping steel, casting operations, and dumping slag. The particulate matter is predominantly iron oxide for most stages of the process but calcium oxide emissions predominate during the refining period.

SOURCES OF HEAVY METAL EMISSIONS

The sources of heavy metal emissions are as those described above for particles.
Since the major feedstock for the EAF is scrap and because polluted scrap is used, the emitted dust contains heavy metals such as lead and zinc. In addition other metals including copper, chromium, nickel, arsenic, cadmium, and mercury are present.

ABATEMENT MEASURES AVAILABLE

Particulate emission control techniques involve an emission capture system and a gas cleaning device. A range of capture systems can be used including direct shell evacuation, side draft hood, combination hood, canopy hood and furnace enclosures.

In direct shell evacuation a "fourth hole" in the roof of the furnace is attached by ductwork to the gas cleaning system. However, this device only works when the furnace is upright and the roof is in position. Side draft hoods collect furnace off gases around the electrode holes and the work doors after the gases leave the furnace. The combination hood has elements of both the direct shell and side hood systems. The canopy hood is the least efficient of the four ventilation systems but it does capture emissions during charging and tapping. The full furnace enclosure or doghouse completely surrounds the furnace and evacuates furnace emissions through hooding in the top of the enclosure. If secondary metallurgy is performed in the same building, these emissions can also be collected.

In all cases the captured gases are normally passed to a fabric filter or ESP to remove particulate matter before the exhaust is discharged to air. Fabric filters are normal in the UK, ESPs are widely used in Europe. Some emissions escape as fugitive releases through the roof vents and these discharges are unabated.

UK SITUATION

UK plants

Several UK secondary steel-making facilities use electric arc furnaces. Details of their locations and capacities are given in Table A5.1 below, based on data published by the Iron & Steel Statistics Bureau (1998). Most plants are of the classical design whereby the roof is moved aside to allow charging and tapping. However, there is at least one large furnace installed that loads around 40% of the scrap charge through a fixed roof section. In this case the furnace is on rails and it is moved sideways to permit loading with the remainder of the charge.

Table A5.1 Electric arc furnaces at steelworks: Plants in existence in UK at the end of 1997

Operator	**Location**	**Capacity (tonnes)**			**Total**
		20 - 39	**40 - 99**	**100 +**	
Avesta	Sheffield			1	1
Corus Engineering Steels	Rotherham			2	2
Corus Engineering Steels	Stocksbridge, Sheffield			1	1
Sheffield Forge masters Ltd	Sheffield	1	1		2
Unsco Steels Ltd	Sheffield	1			1
ASW Sheerness Steel Ltd	Sheerness		2		2
Allied Steel & Wire Ltd	Cardiff			2	2
Alphasteel Ltd	Newport			4	4
Avesta	Pontypool		1		1
Total		**2**	**5**	**10**	**17**

In addition, electric arc furnaces are also in use at the following foundries:

Hopkinsons Ltd	Huddersfield
Wm Cook Hi Tec Integrity Ltd	Penistone
Wm Cook Lloyds Burton	Burton-on-Trent
ANI Bradken Ltd	Bathgate
British Rollmakers	Coatbridge
Glencast	Leven
Ervin Armasteel Ltd	Tipton
Weardale Steel (Wolsingham) Ltd	Wolsingham
Sheffield Forgemasters Rolls Limited	Crewe
London & Scandinavian Metallurgical Co Limited	Rotherham
GKN Sheepbridge Stokes Ltd	Chesterfield

UK activity statistics

Information on UK steel production from electric arc furnaces is published annually by the Iron and Steel Statistics Bureau.

Abatement measures used in UK plants

All furnaces have fourth hole (direct shell) emission control whereby fumes are extracted directly from the furnaces and fabric filtration is used to abate the particulate discharges. They also all employ extraction hoods or canopies to collect particulate emissions associated with charging, tapping and slagging. In the UK, these secondary collection systems may be connected to a separate fabric filter bag house, or may share a baghouse with the primary system. Some plants also have a tertiary collection and control system whereby the furnace is totally enclosed in a doghouse. Any fugitive emissions released during the melting and tapping periods are collected from the top of the doghouse and passed to a fabric filter.

PARTICULATE EMISSION FACTORS

The EIPPCB (2000a) presents overall dust emission factors from electric arc furnaces based on a selection of 38 EU plants (data from 1996). The mean and standard deviation are 124 ± 166 g/t (steel), with a range of 1 – 780 g/t (steel). This extremely wide range indicates large differences in collection and abatement efficiency. The reference notes that plants with well-designed bag filters can achieve dust emission factors of < 20 g/t (steel).

These emission factors combine both primary and secondary emission factors since they are normally treated in the same equipment, though this is not always the case in the UK. (Primary emissions are those collected directly from the EAF; secondary emissions are from scrap handling and charging, steel tapping, secondary metallurgy with tapping operations and continuous casting.) However, primary emissions are estimated to account for 95 percent of the total, and information on secondary emissions is limited.

Table A5.2 lists US EPA total particulate emission factors for electric arc furnaces. In some cases additional particle size information is available and this is shown in Table A5.5. According to these data, at controlled facilities a total particulate emission factor of around 150 g/t is appropriate for those units employing building evacuation. A factor of around 20 g/t is more appropriate for those plants using direct shell evacuation plus charging hood and a common baghouse, although this would vary according to the type of steel (and the quality of the scrap).

Table A5.2 Electric arc furnace particulate emission factors (US EPA, 2000)

Process Stage	**Abatement**	**Emission factors g/t (steel)**			**Quality rating***
		Tot	**PM10**	**PM2.5**	
Melting and refining (carbon steel)	Uncontrolled	19,000	11,020	8170	C

Appendix A5: Electric arc furnaces

Process Stage	Abatement	Emission factors g/t (steel)			Quality rating*
		Tot	PM10	PM2.5	
Charging, tapping and slagging	Uncontrolled emissions escaping monitor	700	n.d.	n.d.	C
Melting, refining, charging, tapping and slagging (alloy steel)	Uncontrolled	5650	n.d.	n.d.	A
Melting, refining, charging, tapping and slagging (carbon steel)	Uncontrolled	25,000	n.d.	n.d.	C
Melting, refining, charging, tapping and slagging (alloy steel)	Building evacuation to baghouse	150	n.d.	n.d.	A
Melting, refining, charging, tapping and slagging (carbon steel)	Direct shell evacuation (plus charging hood) to baghouse	21.5	16.3	15.9	E

[a] Quality rating for total particulate matter. Quality ratings for PM_{10} and $PM_{2.5}$ are given in Table A5.5.

The UK Steel Association (personal communication, 2000) suggests that a factor of around 75 g/t is appropriate for plants using direct shell evacuation plus charging hood and a common bag house, with good capture of charging and tapping fumes. With less effective capture the figure would be higher.

The Pollution Inventory includes emissions data for most UK processes involving the use of electric arc furnaces (those in Scotland are obviously excluded and emissions are not reported by a few of the other processes). These emissions data can be combined with NETCEN estimates of steel production to give the following emission factors:

Table A5.3 Summary of emissions data reported in the Pollution Inventory and derived emission factors for electric arc furnaces

	1998	1999
Reported emissions of PM	1188.4 tonnes	572.6 tonnes
Reported emissions of PM_{10}	1019.5 tonnes	473.0 tonnes
% PM_{10}	86%	83%
Production from reporting sites	2448 ktonnes	2326 ktonnes (PM_{10}) 2631 ktonnes (PM)
Total production	3779 ktonnes	3590 ktonnes
Calculated PM emission factor	485 g/t (steel)	218 g/t (steel)
Calculated PM_{10} emission factor	416 g/t (steel)	203 g/t (steel)

The production estimates are rather uncertain and should be improved where possible.

HEAVY METAL EMISSION FACTORS

Heavy metal emission factors for electric arc furnaces are provided by the EMEP/CORINAIR Guidebook, the EIPPCB (2000a) and the NAEI. Table A5.4 provides a comparison of the emission factors from these references. The NAEI emission factors have been calculated from Pollution Inventory data using the same approach as for particulate matter.

Table A5.4 Heavy metal emission factors for electric arc furnaces (g/t liquid steel)

Species	**EIPPCB (2000)**		**CORINAIR**	**NAEI**	
	Low	**High**		**1998**	**1999**
Arsenic			0.1	0.095	0.073
Cadmium	<0.001	0.072	0.25	0.12	0.055
Chromium	0.008	2.5	1	5.3	3.5
Copper	<0.001	0.46	0.8	0.80	0.65
Mercury	0.006	4.47	0.15	0.050	0.059
Manganese					
Nickel	0.001	1.4	0.25	3.9	2.1
Lead	0.016	3.6	14	6.7	6.1
Selenium			0.05	0.010	0.010
Vanadium				0.96	0.13
Zinc	0.28	45.6	50	75	30

In a number of cases, NAEI factors are higher than the factors suggested in the literature – in particular, the factors for chromium and nickel exceed those given elsewhere. Factors for the remaining pollutants are within the range of values given by EIPPCB or EMEP/CORINAIR, although often at the high end.

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from certain electric arc furnace processes. These are reproduced in Table A5.5. For plants with direct shell evacuation, charging hood and baghouse, a quarter of the total particulate is greater than 10 µm and virtually all the rest is smaller than 1 µm.

Table A5.5 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	**Abatement technology**	**Particle size (µm)**	**Cumulative mass % < stated size**	**Cumulative mass EF (g/t)**	**Quality rating**
Melting and refining (carbon steel)	Uncontrolled	0.5	8	1520	D
		1	23	4370	
		2.5	43	8170	
		5	53	10,070	
		10	58	11,020	
		15	61	11,590	
			100	19,000	

Process	**Abatement technology**	**Particle size (µm)**	**Cumulative mass % < stated size**	**Cumulative mass EF (g/t)**	**Quality rating**
Melting, refining, charging, tapping and slagging (carbon steel)	Direct shell evacuation plus charging hood vented to common baghouse	0.5	74	15.9	E
		1	74	15.9	
		2.5	74	15.9	
		5	74	15.9	
		10	76	16.3	
		15	80	17.2	
			100	21.5	

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI uses particulate matter and PM_{10} emission factors based on the analysis of Pollution Inventory data given above As noted above, some improvements to the calculation of the emission factor is required but we recommend that the approach of using pollution inventory data is continued given that the emission factors calculated this way are higher than the figures given as typical in other sources.

Heavy metal emissions

The current NAEI factors are based on data reported in the Pollution Inventory. It is recommended that this approach is maintained, especially given that the emission factors calculated this way are either higher than or similar to the figures given as typical in other sources.

Appendix A6: Iron and steel foundries

SNAP CODE	**030303**
SOURCE ACTIVITY TITLE	**Grey iron foundries**
NACE CODE	**275**
PARCOM-ATMOS CODE	**2.1.8**

ACTIVITIES INCLUDED

The operation of iron and steel foundries, including the melting and refining of iron and steel, the preparation of moulds, and the production of castings. This chapter does not cover the melting of scrap steel in electric arc furnaces for iron or steel production in iron or steel foundries.

GENERAL PROCESS DESCRIPTION

The major process operations in iron and steel foundries are:

- raw material handling and preparation,
- metal melting and refining
- desulphurisation of molten iron
- slag removal
- mould and core production
- casting and finishing.

Raw material handling and preparation

Metals used as raw material for iron and steel foundries include pig iron, iron and steel scrap, foundry returns, and metal turnings. Fluxes include carbonates (limestone, dolomite), fluoride (fluorspar), and carbide compounds (calcium carbide). Fuels include coal, oil, natural gas, and coke. Foundries try to avoid melting painted scrap, but some will melt oily or greasy scrap. Pre-treatment of scrap is normally carried out by the steel-scrap dealer.

Metal melting and refining

The three most common furnaces used in the iron foundry industry are cupolas, electric arc furnaces, and electric induction furnaces. Electric arc furnaces are only used in a small number of foundries and emissions from these furnaces is included in the previous appendix. Typically, scrap is charged into a furnace and sometimes pre-heated using furnace off-gases before the melting proper begins.

Cupolas

The cupola is a refractory lined cylindrical steel vessel. A solid charge of metal, flux and fuel is fed into the top of the furnace. Heat generated by the combustion of the fuel melts the metal. Air, usually enriched with oxygen and sometimes pre-heated, is fed in at the base of the furnace. Molten metal and slag are tapped from the bottom of the furnace.

Electric induction furnaces

Induction furnaces are used in the foundry industry both to melt and hold liquid metal. There are two types, coreless crucible furnaces and induction channel furnaces. In induction furnaces, heat is generated in the metal charge by current induced by an electrical coil surrounding part of the furnace. The crucible furnace consists of a cylindrical ceramic crucible surrounded by a water-cooled copper coil. They range in size from a few tens of kg to over 100 tonnes. They are operated in batch mode.

Induction channel furnaces are used principally to hold and pour molten metal and may have capacities of several hundred tonnes. The coils do not surround the entire furnace but instead surround a small side arm leading off from the main vessel.

Refining

Refining is the adjustment of chemical composition to meet product specifications. There are two refining operations commonly used. These are decarburisation, which is done by oxygen injection in the furnace itself using an oxygen lance, and nodularisation, which involves the addition of magnesium to the melt and is usually done in the ladle after tapping. Oxygen decarburisation also removes other unwanted elements such as phosphorus, silicon and sulphur. The purpose of nodularisation is to produce ductile iron. When metal with a high carbon content solidifies, carbon crystallises out in the form of flakes. These carbon flakes render the iron brittle. Iron containing carbon flakes is known as flake iron or grey iron. The nodularisation process converts the flakes into spheroidal particles and this causes the iron to be much less brittle. Iron that has been nodularised is called ductile iron. Adding magnesium metal to the molten iron effects the nodularisation process.

Two widely used nodularisation processes are the plunge method and the pour-over method. In plunging, magnesium or a magnesium alloy is loaded into a graphite “bell” which is plunged into a ladle of molten iron. A turbulent reaction takes place as the magnesium boils under the heat of the molten iron. As much as 65 percent of the magnesium may be evaporated. The magnesium vapour ignites in air, creating large amounts of smoke.

In the pour-over method, magnesium alloy is placed in the bottom of a vessel and molten iron is poured over it. Although this method produces more emissions and is less efficient than plunging, it requires no capital equipment other than air pollution control equipment.

Steel and slag tapping

Steel and slag are removed from the furnace through a tapping hole in the door. Since slag is lighter than molten iron, it remains on top of the molten iron and can be raked or poured out.

After slag has been removed, the iron is cast into moulds. Sometimes alloying and other additions are made to the ladle before or during tapping.

Desulphurisation of molten iron

Molten metal from the furnace can be desulphurised using calcium carbide which reacts with sulphur to produce a slag which can be separated from the metal. A number of different methods can be used. In the “shaking” process a barrel-shaped ladle containing molten iron and desulphurising agents is shaken to promote mixing. In the “divided ladle” process the iron is tapped continuously though a slag of lime and calcium carbide into the first compartment of the divided ladle. The compartment is stirred by injecting nitrogen or argon into the ladle through a porous plug. Iron that reaches the bottom of the compartment will be desulphurised and passes under the dividing wall to the second compartment. From here it flows into a transfer ladle.

Mould and core production

Moulds are forms used to shape the exterior of castings. Cores are moulded sand shapes used to make internal voids in castings. Moulds are prepared from wet sand, clay, and organic additives, and are usually dried with hot air. Cores are made by mixing sand with organic binders or organic polymers, moulding the sand into a core, and baking the core in an oven. Used sand from castings shakeout is recycled and cleaned to remove any clay or carbonaceous build-up. The sand is screened and reused to make new moulds.

Casting and finishing

Molten metal is tapped into a ladle or directly into moulds. In larger, more mechanised foundries, filled moulds are conveyed automatically through a cooling tunnel. The moulds are then placed on a vibrating grid to shake the mould sand and core sand loose from the casting.

In the cleaning and finishing process, burrs, risers, and gates are broken or ground off to match the contour of the casting. Afterward, the castings can be shot-blasted to remove remaining mould sand and scale.

SOURCES OF PARTICULATE EMISSIONS

Raw material handling and preparation

Fugitive particulate emissions are generated from the receiving, unloading, and conveying of raw materials.

Metal melting and refining

Melting furnaces emit particulate matter as well as other pollutants. These are generated from incomplete combustion of carbon additives, flux additions and dirt and scale on the scrap charge. The furnaces also emit fine particulate fumes as a result of the condensation of volatilised metal and metal oxides. The highest concentrations of furnace emissions occur when furnace doors are open during charging, backcharging, alloying, slag removal, and tapping operations. These emissions can escape into the furnace building or can be collected and vented through roof openings.

Coke burned in cupola furnaces produces emissions of several pollutants including particulate matter. Electric induction furnaces using clean steel scrap produce particulate emissions comprised largely of iron oxides. When contaminated charges are used, higher emission rates result. Particulate emissions from electric induction furnaces depend on the charge material composition, the melting method (cold charge or continuous), and the melting rate of the materials used. The highest emissions occur during a cold charge.

Refining

Particulate emissions are generated during the refining of molten iron before pouring.

During decarburisation, oxygen is injected into the molten metal in the furnace through a lance to remove unwanted elements such as phosphorus, silicon and sulphur. This results in a marked increase in fume generation from the furnace containing large quantities of extremely fine iron oxide and other oxidation products.

The addition of magnesium to molten metal to produce ductile iron causes a violent reaction between the magnesium and molten iron, with emissions of magnesium oxide fume.

Emissions from pouring include, amongst other pollutants, metal fume from the melt.

Steel and slag tapping

Tapping the molten metal and slag into a ladle generates particulate fume. Sometimes alloying and other additions are made to the ladle before or during tapping. This can lead to an increase in the amount of fume generated during tapping.

Desulphurisation of molten iron

This operation produces particulate emissions.

Mould and core production

The major pollutant emitted in mould and core production operations is particulate from sand reclaiming, sand preparation, sand mixing with binders and additives, and mould and core forming.

Casting and finishing

Emissions during pouring include decomposition products of resins, other organic compounds, and particulate matter. Emissions are related to mould size, composition, sand to metal ratio, pouring temperature, and pouring rate. As the mould cools, emissions continue. A significant quantity of particulate emissions is generated during the casting shakeout operation.

Emissions from finishing operations consist of particulate matter resulting from the removal of burrs, risers, and gates and during shot blasting.

SOURCES OF HEAVY METAL EMISSIONS

The emissions of heavy metals will be predominantly associated with particles. Accordingly the principal sources of heavy metal emissions will be those as described above for particulate matter.

ABATEMENT MEASURES AVAILABLE

Raw material handling and preparation

Fugitive particulate emissions generated from the receiving, unloading, storing, and conveying of raw materials are controlled by enclosing the major emission points and routing the air from the enclosures through fabric filters or wet collectors.

Emissions from the storage and use of powdered materials can be minimised by storage in sealed silos and movement by pneumatic conveyor. Alternatively, they can be kept and handled in sealed bags.

Metal melting and refining

Emission controls for melting and refining operations involve venting furnace gases and fumes directly to a control device. Canopy hoods or special hoods near furnace doors and tapping points capture emissions and route them to emission control systems.

High-energy scrubbers and fabric filters are used to control particulate emissions from cupolas and electric arc furnaces. A cupola furnace typically has an afterburner as well which is located in the furnace stack to oxidise carbon monoxide and burn organic fumes, tars, and oils. Reducing these contaminants protects the particulate control device from possible plugging and explosion.

Because induction furnaces emit relatively little particulate and negligible amounts of other pollutants, they have typically been uncontrolled, except during charging and pouring operations. Extraction is increasingly being fitted and most new large installations would be fitted with bag filters.

Steel and slag tapping

Emissions are collected by hoods or local fume enclosures and extracted to a fabric filter.

Desulphurisation of molten iron

The usual abatement option is to desulphurise using a technique such as the “divided ladle” method that can be fully enclosed with extraction to a dry filter system.

Mould and core production

Fabric filters and high-energy scrubbers generally are used to control particulate from mould and core production.

Casting and finishing

Emissions are controlled by cyclone separators and fabric filters.

UK SITUATION

UK plants

The UK ferrous foundry industry produces products ranging in size from small castings of a few kg to large roll castings of many tonnes. The Part A (IPC controlled) part of the industry consists of foundries which use electric arc furnaces and these are covered in the previous appendix. The Part B (LAPC controlled) part of the industry in England and Wales includes about 70 processes authorised to operate hot or cold blast cupolas and about 170 processes authorised to operate electric induction, rotary or reverberatory furnaces. The number of processes in Scotland and Northern Ireland is not known. The UK iron foundry capacity is approximately 1,000,000 tonnes per year and the steel sector is approximately 80,000 – 100,000 tonnes per year (Castings Development Centre, 2000).

The Iron and Steel Statistics Bureau (1998) lists steel furnaces at steel foundries in the UK. According to them in 1997 there were 145 electric induction furnaces operating in UK steel foundries, excluding those also operating electric arc furnaces. No data were available on furnaces in iron foundries.

UK activity statistics

Information on UK iron and steel production is published annually by the Iron and Steel Statistics Bureau. The data include production of steel castings for the period up until 1997 and production of iron castings for the period up until 1992.

Abatement measures used in UK plants

No information on the abatement measures used in UK plants is available.

PARTICULATE EMISSION FACTORS

Relatively little information on emission factors from foundries is available. Tables 6.1 and 6.2 list US EPA emission factors for a range of processes and control technologies in iron and steel foundries respectively.

Table A6.1 Iron foundry particulate emission factors (US EPA, 2000)

Process Stage	Abatement	Emission factors g/t (metal produced)			Quality rating[a]
		Tot	PM_{10}	$PM_{2.5}$	
Cupola	Uncontrolled	6900	6200	5800	E
Cupola	Scrubber	1600			C
Cupola	Venturi scrubber	1500	1170	1170	C
Cupola	Electrostatic precipitator	700			E
Cupola	Baghouse	300[b]	380	380	E
Cupola	Single wet cap	4000			E
Cupola	Impingement scrubber	2500			E
Cupola	High-energy scrubber	400			E
Electric induction furnace	Uncontrolled	500			E
Electric induction furnace	Baghouse	100			E
Reverberatory	Uncontrolled	1100			E
Reverberatory	Baghouse	100			E
Scrap and charge handling, heating	Uncontrolled	300			E
Scrap and charge handling, heating	To atmosphere	100			E
Magnesium treatment	Uncontrolled	900			E
Magnesium treatment	To atmosphere	200			E
Refining	Uncontrolled	1500–2500			E
Pouring, cooling	Uncontrolled	2100	1030	500	E
Shakeout	Uncontrolled	1600	1120	670	E
Cleaning, finishing	Uncontrolled	8500			E
Cleaning, finishing	To atmosphere	50			E
Sand handling	Uncontrolled	800[c]			E
Sand handling	Scrubber	23[c]			D
Sand handling	Baghouse	100[c]			E
Core making, baking	Uncontrolled	600			E
Core making, baking	To atmosphere	600			E

[a] Quality rating for total particulate matter. Quality ratings for PM_{10} and $PM_{2.5}$ are given in Table A6.6

[b] Note that Table A6.6 has a total particulate emission factor for this process of 400 g/t. This is an inconsistency in the source.

[c] Unit: g/t (sand handled)

Table A6.2 Steel foundry particulate emission factors (US EPA, 2000)

Process Stage	Abatement	Emission factors [a] g/t (metal processed) Tot	PM$_{10}$	Quality rating
Melting: open hearth	See note [b]	5,500 (1,000–10,000)		E
Melting: open hearth oxygen lanced	See note [c]	5,000 (4,000–5,500)		E
Melting: electric induction	Usually uncontrolled	50	45	E
Sand grinding/ handling in mould and core making	Not specified		270 3,000[d]	E
Core ovens	Not specified		1,110 450[d]	E
Pouring and casting	Not specified		1,400	E
Casting cleaning	Not specified		850	E
Charge handling	Not specified		180	E
Casting cooling	Not specified		700	E

[a] If the scrap metal is very dirty or oily, or if increased oxygen lancing is employed, the emission factor should be chosen from the high side of the factor range.

[b] Electrostatic precipitator, 95 to 98% control efficiency; baghouse, 99.9% control efficiency; venturi scrubber, 96 to 99% control efficiency.

[c] Electrostatic precipitator, 95 to 98% control efficiency; baghouse, 99% control efficiency; venturi scrubber, 95 to 98% control efficiency.

[d] Unit: g/t (sand handled)

HEAVY METAL EMISSION FACTORS

There are very few heavy metal emission factor data for this sector. However, the US EPA (1992) have published some data from the analysis of dust emissions from open cupolas. These data are reproduced in Table A6.3. The data shows that the emissions from an open cupola contain significant amounts of zinc, lead and copper.

Table A6.3 Content of heavy metals for cupola dust, mg/kg

As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
130	60	380	1900	0	670	3850	18650

The results of experimental studies in the former USSR countries has been published by MSC-East under the EMEP programme (Kakareka *et al*, 1999). These data are reproduced in Table A6.4 and also show high concentrations of lead, zinc and copper in the particulate emissions. However, the emissions are circa an order of magnitude lower for all measured species than those reported in the US data.

Table A6.4 Content of heavy metals in dust for grey iron foundries, mg/kg

Sample	As	Cd	Cr	Cu	Ni	Pb	Zn	Hg
1	10.3	<	59.6	345	75.4	451	2180	no data
2	26.5	<	47.4	148	56.5	305	1800	no data
3	<	3.12	76	425	30.7	240	1950	no data
4	<	5.18	152	232	65.8	344	1990	<
5	<	0.84	46.3	70.3	31.1	59.6	209	0.057
6	<	2.4	47.5	223	76.6	187	2330	<

In addition, emission factors are provided by EMEP/CORINAIR (1999). These data are presented in Table A6.5 and date from the 1980's.

Table A6.5 Emission factors for foundries (g/t product)

Substance	Emission factor	Range
Arsenic	0.3	0.02 - 3.6
Cadmium	0.14	0.006- 0.45
Chromium	1.1	0.09 - 3.0
Lead	7.2	0.24 -15
Nickel	0.5	0.01 - 1.3
Zinc	5.0	2.4 - 7.2

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from various foundry processes. These are reproduced in Table A6.6.

Table A6.6 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Cupola furnace	Uncontrolled	0.5	44.3	3100	C
		1	69.1	4800	
		2	79.6	5500	
		2.5	84.0	5800	
		5	90.1	6200	
		10	90.1	6200	
		15	90.6	6300	
			100	6900	

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Cupola furnace	Baghouse	0.5	83.4	330	E
		1	91.5	370	
		2	94.2	380	
		2.5	94.9	380	
		5	94.9	380	
		10	94.9	380	
		15	95	380	
			100	400	
Cupola furnace	Venturi scrubber	0.5	56	840	C
		1	70.2	1050	
		2	77.4	1160	
		2.5	77.7	1170	
		5	77.7	1170	
		10	77.7	1170	
		15	77.7	1170	
			100	1500	
Pouring, cooling	Uncontrolled	0.5	0	0	D
		1	19	400	
		2	20	420	
		2.5	24	500	
		5	34	710	
		10	49	1030	
		15	72	1510	
			100	2100	
Shakeout	Uncontrolled	0.5	23	370	E
		1	37	590	
		2	41	660	
		2.5	42	670	
		5	44	700	
		10	70	1120	
		15	99.9	1600	
			100	1600	

RECOMMENDED EMISSION FACTORS

Particulate emissions

Currently, emissions from iron & steel foundries are estimated using an emission factor of 608 kg per site for processes covered by PG 2/3 (electrical and rotary furnaces) and PG 2/5 (hot and cold blast cupolas). The number of sites used to calculate UK emissions is taken from a survey of local authority regulated processes carried out by DETR, AQ4(97); the original survey covered England and Wales but the results have been extended to the UK. It should be noted that this emission estimate will include emissions from some non-ferrous foundries as well.

The recommended approach for calculating emissions from foundries would be to use an emission factor expressed in terms of metal processed which could be combined with activity statistics available from the Iron and Steel Statistics Bureau and elsewhere. However, the emission factor for iron and steel foundries seems to depend strongly on the types of abatement technologies used and especially on whether any sources are uncontrolled. No information on the UK situation in this regard has been found, so it is not possible to make recommendations about UK emission factors. Given good abatement, factors are likely to be in the middle hundreds g/t. Individual uncontrolled processes could increase this significantly. Therefore, the nature of abatement systems used by the foundry industry should be identified so that appropriate emission factors can be chosen.

In the interim, a particulate matter emission factor of 1500 g/tonne (metal) could be used for cupolas and a figure of 100 g/tonne for other furnace types, based on the assumption that wet scrubbers and bag filters would be fitted in order to comply with LAPC (PG 2/3 & PG2/5). The PM_{10} emission factors would be 1170 g/tonne (metal) and about 80 g/tonne, assuming a similar size distribution to emissions from cupolas. For the earlier part of the time series, it would be more appropriate to use emission factors for uncontrolled systems e.g. 6900 g/t (metal) for cupolas and 500 g/tonne for particulate matter from induction furnaces with PM_{10} factors of 6200 g/t (metal) and perhaps 450 g/tonne for induction furnaces, assuming the same size distribution as for cupolas.

Heavy metal emissions

Current NAEI emission factors for lead, arsenic, chromium, nickel and zinc are based on those given by EMEP/CORINAIR (1999). However, since the EMEP/CORINAIR factors are based on a dust concentration in waste gases of 300 mg/m^3, whereas current UK regulation should result in dust concentrations of no more than 20 mg/m^3, the factors have been reduced accordingly (i.e. divided by 15). For cadmium and mercury, emission factors are based on emission estimates given by Entec (1996). The factors for copper, vanadium and selenium are based on emission factors for electric arc furnaces given by Jockel & Hartje (1991) in the case of vanadium or EMEP/CORINAIR (1999) in the other two cases. The factors used for 1999 are shown in Table A6.7.

Table A6.7 Heavy metal emission factors used in the 1999 NAEI (expressed as g/tonne iron castings produced)

Metal	Emission factor
Arsenic	0.02
Cadmium	0.0567
Chromium	0.073
Copper	0.5
Lead	0.48
Mercury	0.15
Nickel	0.033
Selenium	0.05
Vanadium	0.05
Zinc	0.33

Given the lack of emission factors from other sources, we recommend that the current approach be continued for most metals. However, in the case of copper, we recommend that consideration be given to using the dust composition data presented in Tables 6.3 and 6.4 to generate a more appropriate factor for this sector than the current factor which is based on a factor for electric arc furnaces.

Appendix A7: Primary aluminium

SNAP CODE	**040301** **030322**
SOURCE ACTIVITY TITLE	**Aluminium production** **Alumina production**
NACE CODE	**27420**
PARCOM-ATMOS CODE	**2.2.4**

ACTIVITIES INCLUDED

This chapter covers the production of alumina from bauxite and the production of aluminium from alumina.

GENERAL PROCESS DESCRIPTION

Alumina is produced by the Bayer process which involves drying and grinding of bauxite ore followed by leaching using sodium hydroxide at elevated temperature and pressure. This leaching stage produces sodium aluminate which is purified and then cooled. Alumina precipitates and can be separated, dried and then calcined.

Aluminium is produced from electrolytic reduction of alumina using the Hall-Heroult process. The electrolysis cells comprise a steel shell lined with refractory material, equipped with carbon cathodes and carbon anodes. The cells contain molten alumina dissolved in cryolite ($3NaF.AlF_3$). Aluminium is deposited at the cathode while oxygen is produced at the anodes and combines with the carbon to form carbon dioxide. Fluoride compounds are added to lower the melting point of the alumina.

A number of different variations on the electrolytic cell exist. The main option is the use of either Soderberg self-baking anodes or pre-baked anodes. Soderberg anodes consist of carbon paste that is baked by the heat of the molten alumina. The anode paste is a mixture of pitch and coke. Use of Soderberg anodes mean that the process is continuous since, as carbon in the anode is consumed, fresh paste can descend to take its place. As their name suggests pre-baked anodes are baked in a separate process. As these anodes are consumed, they are lowered into the alumina bath until about 75% of the anode has been consumed. The anodes are then replaced.

The molten aluminium deposited at the cathode is periodically tapped and transferred to holding furnaces ready for casting into slabs, billets and other products. Some further purification of the aluminium may be carried out including degassing using nitrogen, chlorine or argon. Alloying metals may also be added.

SOURCES OF PARTICULATE EMISSIONS

Alumina production results in emissions of particulate matter from handling and processing of the bauxite ore and from drying and calcining of the alumina product.

Emissions during aluminium production can occur from the baking of pre-baked carbon anodes, from the electrolytic process, from tapping and from casting of the aluminium product.

SOURCES OF HEAVY METAL EMISSIONS

Metals occur in trace concentrations in alumina and aluminium fluoride and can therefore be emitted during electrolysis. A limited amount of data is available on the environmental impact but trace metals from this source are not considered to be a significant by CORINAIR/UNECE (1999) and the EIPPCB (2000b).

ABATEMENT MEASURES AVAILABLE

The electrolytic processes are hooded and gases extracted to gas treatment plant.

UK SITUATION

UK plants

The UK currently has three aluminium smelters, although a fourth smelter was in operation until closure in mid 2000. This plant, at Kinlochleven, used vertical-stud Soderberg (VSS) technology, while the other three (at Holyhead, Lynemouth and Lochaber) use the pre-baked anode process. Anodes are baked at two of the sites (Holyhead and Lynemouth).

There is one UK plant at Burntisland in Scotland which produces alumina from bauxite.

UK activity statistics

The United Kingdom Minerals Yearbook, published annually by the British Geological survey gives production of alumina and primary aluminium.

Abatement measures used in UK plants

No information is available on the control measures used at the Burntisland plant.

Dry scrubbing using alumina in bag filters is used at three aluminium smelters to remove gaseous fluorides but also removes particulate matter. Bag filters are also used for anode baking plants with alumina dry scrubbing again being used. The fourth plant uses wet scrubbing with water sprays to control fluorides after which gases pass to an electrostatic precipitator.

PARTICULATE MATTER EMISSION FACTORS

The Environment Agency's Pollution Inventory gives particulate matter emission estimates for two aluminium plants — those located at Lynemouth and Holyhead. Both these plants use pre-baked anodes that are produced on-site. The Scottish Environment Protection Agency have provided data on emissions from the Scottish sites during 1999. The data are presented in Table A7.1.

Table A7.1 Emissions from UK primary aluminium plants during 1999

Plant	**Total particulate t**	**Production t**	**Emission factor g (TPM)/t (aluminium)**
Lynemouth:			
smelting	202		
anode baking	26		
Holyhead:			
smelting	229.4		
anode baking	142.5		
Lochaber	55.3		
Kinlochleven	183.4		
Total	**838.6**	**272,200**	**3080**
smelting	**670.2**	**-**	**2460**
anode baking	**168.5**	**-**	**620**

TPM = total particulate matter

Emissions of PM_{10} are only given for smelting at Lynemouth in 1999 – the figure of 4 tonnes suggests a PM_{10} ratio of 2%, which seems low.

The EIPPCB (2000b) observes that total dust generation from electrolysis depends on the process applied and the type of alumina, but ranges from 600 to 7,000 g/t (aluminium) for pre-bake plants and 1,500 to 10,000 g/t (aluminium) for vertical Soderberg plants. Emissions from primary smelter cast houses are in the range 20 – 300 g/t (aluminium), but it is not clear whether this is controlled or uncontrolled.

US EPA (2000) emission factors for alumina and aluminium production are shown in Table A7.2. These date from the early 1960s. Taking into account the emission control devices in place in the UK would suggest total particulate emission factors for the UK aluminium plants of 3,430 g/t (aluminium) for the smelters using pre-baked anodes and 9,850 g/t (aluminium) for the plant using vertical-stud Soderberg anodes. In both cases, around two-thirds of the emissions are fugitives. No information is available on control methods at the alumina plant, but it is unlikely to be uncontrolled. The total emission factor for the various control options range from 30.5 kg/t (aluminium) to 2.06 kg/t (aluminium).

Information on PM_{10} is more limited (see Table A7.3) but for pre-baked anode plants, the PM_{10} emission factor will be no more than 2,380 g/t. Assuming a 50% PM_{10} fraction for particulate from the dry alumina scrubber would imply a factor of 1,930 g/t.

Table A7.2 Alumina and aluminium production particulate emission factors (US EPA, 2000)

Process Stage	Abatement	Emission factors g/t (aluminium) Tot	PM_{10}	$PM_{2.5}$	Quality rating*
Bauxite grinding	Uncontrolled	3000			A
	Spray towers	500			A
	Floating bed scrubber	850			A
	Quench tower	500			A
	ESP	60			A
Alumina calcining	Uncontrolled	100000			A
	Spray towers	30000			A
	Floating bed scrubber	28000			A
	Quench tower	17000			A
	ESP	2000			A
Anode baking	Uncontrolled	1,500			A
	ESP	375			A
	Floating bed scrubber	375			A
	Dry alumina tower	30			A
Prebake cells	Uncontrolled	47,000			A
	Fugitive	2,500	1450	700	A
	Collected	44,500			A
	Crossflow packed bed	13,150			A
	Multiple cyclones	9,800			A
	Spray tower	8,900			A
	Dry ESP plus spray tower	2,250			A
	Floating bed scrubber	8,900			A
	Dry alumina scrubber	900			A
	Coated bag filter dry scrubber	900			A
	Dry plus secondary scrubber	350			A

Process Stage	**Abatement**	**Emission factors g/t (aluminium)**			**Quality rating***
		Tot	**PM_{10}**	**$PM_{2.5}$**	
Vertical stud Soderberg	Uncontrolled	39,000			A
	Fugitive	6,000			A
	Collected	33,000			A
	Multiple cyclones	16,500			A
	Spray tower	8,250			A
	Venturi scrubber	1,300			A
	Dry alumina scrubber	650			A
	Scrubber plus ESP plus spray screen and scrubber	3,850			A
Horizontal stud Soderberg	Uncontrolled	49,000			A
	Fugitive	5,000	1550	850	A
	Collected	44,000			A
	Spray tower	11,000			A
	Floating bed scrubber	9,700			A
	Scrubber plus wet ESP	900			A
	Wet ESP	900			A
	Dry alumina scrubber	900			A

Quality rating for total particulate matter. Quality rating for PM_{10} and $PM_{2.5}$ are given in Table A7.3.

HEAVY METAL EMISSION FACTORS

The production of alumina and aluminium are not expected to give rise to significant emissions of heavy metals and no emission factors have been found.

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from aluminium production using pre-bake and horizontal stud Soderberg cells. These are reproduced in Table A7.3. and relate to emissions from fugitive sources only. In the absence of any data on contained emissions from pre-bake and Soderberg cells, we recommend that the fugitive emission size distribution be used In the case of emissions from anode baking, the US EPA suggests that a default profile for ‘condensation, hydration, absorption, prilling and distillation’, which gives a figure of 94% less than 10 microns. Overall, this gives the following PM_{10} figures:

Pre-baked process, smelting	58%
Pre-baked process, anode baking	94%

VSS process 31%

Table A7.3 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Pre-bake cells	Uncontrolled	0.625	13	330	C
		1.25	18	460	
		2.5	28	700	
		5	43	1080	
		10	58	1450	
		15	65	1620	
			100	2500	
Horizontal stud Soderberg cells	Uncontrolled	0.625	8	400	D
		1.25	13	650	
		2.5	17	850	
		5	23	1150	
		10	31	1550	
		15	39	1950	
			100	5000	

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI uses a PM_{10} emission factor of 2,660 g/t (aluminium product). This is derived from the emissions data provided in the Pollution Inventory for 1998, although there is a transposition error in the original calculations. However, the NAEI also includes a separate figure for anode baking, whereas the emission factor derived from the Pollution Inventory includes both anode baking and aluminium production. The NAEI therefore double-counts emissions from anode baking and this must be eliminated in the next NAEI. The NAEI also needs to be updated to take into account of data held in the 1999 Pollution Inventory and data available from SEPA.

The Pollution Inventory data yields a particulate matter emission factor of 2,460 g/t (aluminium product) for smelting at UK plant and a figure of 619 g/t (aluminium) for anode baking. The combined figure is within the range suggested by the EIPPCB, although below the mid-point. EPA factors are also generally higher, however the US data are old and the level of control current in the UK might be expected to be higher than that assumed in that reference. It is therefore recommended that the Pollution Inventory continue to be used as the source of emission estimates for the two sites involved in anode baking and smelting and that data from SEPA be used for the Scottish plants. Emissions of PM_{10} should be calculated from the particulate matter emission estimates using the size distribution figures suggested in the previous section.

Emission factors for alumina production cover a wide range and are dependent upon the abatement systems in place. In the current day, we would expect the level of abatement to be high and recommend the use of an emission factor of 2.06 kg/t. However, further investigation should be carried out so that a more certain assessment of emissions can be made. For the purpose of deriving a PM_{10} emission estimate, it should be assumed that the proportion of sub 10 micron material is 85% in the case of bauxite grinding and 53% in the case of alumina calcining. These figures are based on the US EPA's default emission profiles for mechanically generated dusts and for dusts from calcining and other heat reaction processes. This gives a PM_{10} emission factor of 1.11 kg/t (aluminium). In summary, it is recommended that the current NAEI figure for aluminium production be corrected and that further investigation be made to obtain better particulate matter emission estimates for the Soderberg process and for the production of alumina from bauxite.

Heavy metal emissions

Heavy metal emissions from this source are expected to be trivial. No recommendations are made for emission factors.

Appendix A8: Secondary aluminium

SNAP CODE	**030310** **040300**
SOURCE ACTIVITY TITLE	**Secondary aluminium production**
NACE CODE	**3.0**
PARCOM-ATMOS CODE	**27.42**

ACTIVITIES INCLUDED

This section covers the production of aluminium metal from scrap materials, but not the use of aluminium or aluminium alloys in the production of fabricated goods.

GENERAL PROCESS DESCRIPTION

Secondary aluminium production is the process of converting aluminium-containing scrap into saleable metal. It involves many steps but they all fall into one of two main categories; pre-treatment and smelting/refining. Pre-treatment includes sorting, processing, and cleaning scrap. Smelting/refining includes melting, refining, alloying, and pouring. Some or all of the above steps may be undertaken at any one facility and some may be combined or reordered, depending on the quality of the scrap.

The secondary aluminium industry processes both “new” and “old” scrap. New scrap is material discarded from manufacturing processes and includes extrusion discards, sheet edge trim, turnings, millings and drosses. Old scrap is material that has been used by final consumers and discarded for recycling. This includes, for example, used beverage cans, car cylinder heads, window frames and electrical conductor cable.

Pre-treatment

The pre-treatment of the scrap involves sorting and processing to remove contaminants and prepare the material for smelting. It can include any of the following mechanical, pyrometallurgical or hydrometallurgical operations.

Mechanical cleaning

This includes the physical separation of aluminium from other scrap, with hammer mills, crushers and other machines to break the scrap into smaller pieces. It improves the efficiency of downstream recovery of iron by magnetic separation. Other recovery processes include vibrating screens and air classifiers. Dry milling is a particular example of mechanical cleaning used for processing cold aluminium-laden dross and other residues. These materials are milled and screened to obtain a fine powder product containing at least 60 to 70 percent aluminium.

Pyrometallurgical treatment: roasting, sweating and hot dross processing

Roasting. The roasting process involves heating aluminium scrap that contains organic contaminants in rotary dryers to temperatures hot enough to vaporise or carbonise organic contaminants but not hot enough to melt aluminium (660 °C).

Sweating. The sweating process involves heating aluminium scrap containing other metals in a sweat furnace (usually a sloped hearth furnace) to temperatures above the melting temperature of aluminium, but below that of the other metal. Aluminium and other low melting constituents melt and trickle down the sloped hearth into air-cooled moulds or collecting pots. The higher-melting materials, including iron, brass, and the oxidation products formed during the sweating process, are periodically removed from the furnace.

Hot dross processing. Dross is a layer of impurities and semisolid flux that has been skimmed from the surface of molten aluminium. Aluminium may be recovered from dross by batch fluxing with a salt/cryolite mixture in a mechanically rotated, refractory-lined barrel furnace. Cryolite acts as a catalyst that decreases aluminium surface tension and therefore increases recovery rates. Aluminium is tapped periodically through a hole in the base of the furnace.

Hydrometallurgical cleaning

This involves leaching and separation. It doesn’t lead to significant particulate emissions and so is not discussed further.

Smelting/refining

After pre-treatment, the scrap is smelted and refined. Melting and impurity removal (refining) in secondary aluminium recovery usually takes place in a reverberatory furnace which is brick-lined with a curved roof. It is so called because heat rising from the flame is reflected (reverberated) back down from the curved furnace roof and onto the melted charge. A typical furnace has an enclosed melt area where the flame operates directly above the molten aluminium. A reverberatory furnace is normally linked to two or more smaller holding furnaces, where alloying and final adjustments can be more easily made prior to pouring. Smaller scale smelting operations can also take place in a crucible furnace.

Smelting and refining operations can involve the following steps:
- charging
- melting
- fluxing
- demagging
- degassing
- alloying
- skimming
- pouring.

Charging, melting and fluxing. These are a combined operation. The scrap, mixed with flux material, is placed into the furnace charging well where it melts. Flux materials combine with contaminants and float to the surface of the aluminium trapping impurities and providing a barrier (up to 15 cm thick) that reduces oxidation of the molten aluminium.

Demagging. This is the removal of magnesium contamination from the molten aluminium. It normally involves the addition of chlorinating agents or halide fluxes and reduces the magnesium content from approximately 0.5% to about 0.1% by releasing the magnesium as a halide fume or vapour. Aluminium halide vapour or fume can also be released. Several different demagging technologies are used. These are:

- Liquid chlorine, injected under pressure. The pressurised chlorine is released through carbon lances directed under the surface. This operation results in high chlorine emissions as well as significant releases of magnesium chloride and the technique is no longer used in the US or EU.
- Chlorine gas, metered into the turbulent molten metal flow in the circulation pump discharge pipe. This significantly reduces chlorine usage and release of the free gas.
- Other chlorinating agents or fluxes such as aluminium chloride can be added to the molten metal to release magnesium as magnesium chloride. Fluxes containing fluorides can also be used for demagging. Chlorinated organic compounds can also be employed but the use of many of these compounds has been discontinued in the US and EU.

Degassing. This involves the removal of entrained gases in the molten metal. High-pressure inert gases are released below the molten surface to violently agitate the melt.

Alloying. As its name suggests, alloying is the production of alloys of aluminium by adding other metals in the required proportions.

Skimming. This operation removes semi-solid material (dross, slag, or skimmings) that float on the surface of the molten aluminium by ladling them from the surface of the melt.

SOURCES OF PARTICULATE EMISSIONS

Pre-treatment operations

The following pre-treatment operations can lead to emission of particulate matter:

Mechanical cleaning. The crushing, shredding, and screening steps involved in cleaning and sorting the raw scrap all produce metallic and non-metallic particulate emissions. Dry milling in particular generates large amounts of dust. The emissions are likely to be coarse in size and easily controllable.

Roasting. Burning and drying operations emit particulate matter. Oxidised aluminium fines are blown out of the rotary kiln by the combustion gases. In addition, particulate can arise from the charring of carbonaceous materials (ash).

Sweating. Sweating furnaces may emit smoke particles from the incomplete combustion of organic contaminants (e.g., rubber, oil and grease, plastics, paint, cardboard, paper). Metallic fumes can also be generated from the oxidation of magnesium and zinc contaminants and from fluxes in recovered dross and skims. These emissions vary with the feed scrap composition.

Hot dross processing. Rotating barrel dross furnaces emit mechanically generated dust and the furnace off-gases also contain fluxing salt fume. Fugitive particulate emissions may also be generated.

Hydrometallurgical operations. As noted above, these only produce a small amount of particulate emissions during drying.

Smelting/refining operations

The following smelting/refining operations can lead to emissions of particulate matter:

Smelting using reverberatory furnaces. The use of reverberatory furnaces for smelting aluminium account for a significant fraction of the total particulate emissions associated with the secondary aluminium industry. Emissions from the charging well consist of organic and inorganic particulate material.

Smelting with crucible furnaces. Crucible furnaces also produce particulate emissions but at a lower level than reverberatory units.

Fluxing. These emissions include the entrainment of particulate from the common fluxing salts (e.g. sodium chloride, potassium chloride, and cryolite). Aluminium and magnesium chloride fume may also be generated from the fluxing materials being added to the melt. Studies have suggested that fluxing particulate emission are typically less than 1 μm in diameter.

Demagging. Chlorine demagging results in the formation of magnesium chloride that can be emitted as a fine fume. Excess chlorine combines with aluminium to form aluminium chloride. This is a vapour at furnace temperatures but condenses into sub-micron particles as it cools. Emissions from fluoride demagging are highly corrosive as they contain hydrogen fluoride, but this method additionally generates aluminium fluoride and magnesium fluoride fumes.

SOURCES OF HEAVY METAL EMISSIONS

There are potential emissions to air of metal compounds from the melting and treatment furnaces. The type and quality of scrap material used will strongly influence the significance of releases. The emissions of heavy metals are associated with particulate matter and therefore the sources of release will be similar to those described above for particulate matter.

ABATEMENT MEASURES AVAILABLE

Hot dross processing

Furnace off-gas emissions, containing mainly fluxing salt fume, can be controlled by a venturi scrubber but the use of the fabric filter has become widespread in the US and in the EU.

Roasting and sweating

Wet venturi scrubbers have been used in the US and EU to control particulate emissions from these process stages. However, the use of fabric filters is much more common.

Demagging

Two approaches have been employed for some time for optimising releases from demagging. The Derham process uses proprietary fluxes and claims more than 97% magnesium-chlorine efficiency for the chlorination stage at magnesium levels of less than 0.1%. The Alcoa fumeless process depends on effecting a stoichiometric chlorination of magnesium in a multi-stage enclosed settler-reactor tank after melting and prior to casting. Efficient gas-liquid contact gives a selective magnesium chlorination reaction which is 99% efficient. Although such techniques can significantly reduce the amount of aluminium chloride particulate matter generated, they are normally backed by wet scrubbers or, more commonly, fabric filters.

UK SITUATION

UK plants

A review of pollution control for the secondary aluminium industry carried out by McLelland & Partners (1993) estimated that 25 companies were engaged in secondary smelting of aluminium, 26 were involved in manufacture of aluminium semis, and 3 manufactured aluminium powders and/or pastes.

The current number of UK plants is not known with certainty. However, at least 30 processes, regulated by the Environment Agency in England and Wales, are thought to be involved predominantly in the recovery of aluminium from scrap materials. As well as a smaller number regulated by SEPA under IPC, there are probably about 200 processes regulated by local authorities as ‘aluminium processes’ or by SEPA under APC. Many of these processes will be foundries using small quantities of aluminium or aluminium alloys and are excluded from the scope of this chapter.

UK activity statistics

The Aluminium Federation publishes aluminium production and trade statistics on its web site and some information is given in the UK Minerals Yearbook, published annually by the British Geological Survey. Secondary production of aluminium, predominantly from old scrap is around 240,000 t per annum while production of wrought remelt predominantly from new scrap is around 560,000 t per annum.

Abatement measures used in UK plants

In the UK, the fabric filter, or in some cases the ceramic filter, has been widely adopted at Part A facilities as the best available technique (BAT) for the control of particulate releases from the secondary aluminium sector. Therefore, in this sector all the process steps mentioned above as potential sources of particulate release are tightly controlled.

Part A processes cover the majority of secondary aluminium production in the UK, but there is significant production associated with Part B processes. Although the fabric filter is widely used in Part B processes to control particulate releases its adoption is by no means complete. For example, some older facilities producing aluminium from scrap, or which are involved in the thermal decontamination of oily / greasy scrap, are not yet fitted with any particulate control equipment other than afterburners to destroy condensing products of incomplete combustion.

Cyclones are mainly used in the aluminium industry for the collection of the relatively coarse particulate matter associated with mechanical treatments but even here it is mostly used as a pre-collector upstream of a more efficient particle removal system (i.e. the fabric filter).

Wet collection devices such as the venturi scrubber are now rarely used for particulate control in the secondary aluminium production sector, although a few still remain in Part B processes.

PARTICULATE EMISSION FACTORS

Although the EIPPCB (2000b) states that particulate emissions from secondary aluminium processing are up to 50 mg/Nm^3, no conversion factor is given to allow this to be converted to g/t (product).

Table A8.1 shows the total particulate emission factors quoted by US EPA (2000) for both uncontrolled and controlled stages of secondary aluminium production. Estimates have been given for the PM_{10} and $PM_{2.5}$ emissions for the uncontrolled process stages only as no data were given for the corresponding abated process stages. Total particulate emission factors for sweating operations controlled by fabric filters are given as 1650 g/t of metal processed. This seems rather high for this air pollution control technology and a figure of about half this value would be more reasonable. Although there are no data available for roasting operations using rotary furnaces it is likely that the total particulate emission factor would be similar if a fabric filter was used. The factor for smelting and refining using fabric filter or electrostatic precipitators is the same at 650 g/t.

The US EPA gives factors for chlorine demagging in terms of grams emitted per tonne of chlorine used. If the magnesium content of the melt is around the normal level of 0.5%, then these figures may be converted to g/t (aluminium) by multiplying by 0.005 t (chlorine)/t (aluminium). Table A8.1 gives figures in g/t (aluminium) using this conversion factor, as well as the original US EPA figures in g/t (chlorine).

Table A8.1 Secondary aluminium production, particulate emission factors (US EPA, 2000)

Process stage	Abatement	Emission factors (g/t aluminium)			Quality rating
		Tot	PM_{10}	$PM_{2.5}$	
Sweating furnace	Uncontrolled	7,250			E

Process stage	**Abatement**	**Emission factors (g/t aluminium)**			**Quality rating**
		Tot	**PM_{10}**	**$PM_{2.5}$**	
Sweating furnace	Baghouse	1,650			E
Crucible	Uncontrolled	950			E
Reverberatory	Uncontrolled	2,150	1,300	1,080	E
Reverberatory	Baghouse	650			E
Reverberatory	ESP	650			E
Demagging	Uncontrolled	500,000[a]	266,000[a]	99,500[a]	E
Demagging	Uncontrolled	2,500[b]	1330[b]	500[b]	E
Demagging	Baghouse	25,000[a]			E
Demagging	Baghouse	125[b]			E

a – units are grams emitted per tonne of chlorine used; b – units are grams emitted per tonne of aluminium produced, assuming a conversion factor of 0.005 t (chlorine)/t (aluminium).

Emissions data from the Pollution Inventory for 1998 and 1999 are shown in Table A8.2. A problem with using Pollution Inventory data for this sector is that, for many of the processes within the sector, emissions of particulate matter are reported to be below the *de minimis* limit for reporting. This is set at 10 tonnes for particulate matter, but because there are a large number of sites which are below the limit, the emissions from these sites could be significant. Therefore we have estimated emissions from these sites either by extrapolating from data reported for earlier years when *de minimis* limits were not used or, where no emissions are reported for any year, the emission is assumed to be 5 tonnes (i.e. midway between the *de minimis* limit and zero). Both the original reported data and the revised estimate are shown in Table A8.2.

Table A8.2 Emissions of particulate matter reported in the Pollution Inventory for secondary aluminium processes.

	Total particulate t		**Production kt**		**Emission factor g (TPM)/t (aluminium)**	
	1998	**1999**	**1998**	**1999**	**1998**	**1999**
Original data	110.6	150.1	274.8	285.3	402	526
Revised estimate	242.7	204.6	274.8	285.3	883	717

TPM = total particulate matter

Only, a very small number of sites report emissions of both particulate matter and PM_{10}. From these very limited data, an average PM_{10} to particulate matter ratio of 0.65:1 can be calculated. If applied to the emission factors based on our revised estimates for particulate matter emissions, this would give PM_{10} emission factors of 574 g / t (aluminium) and 466 g/ t (aluminium) in 1998 and 1999 respectively.

HEAVY METAL EMISSION FACTORS

Some emissions data are available in the Pollution Inventory however, as with particulate matter, most sites do not report emissions because their emissions are below *de minimis* limits. Emission factors have therefore been calculated by examining the metal to particulate ratio for cases where emissions of both have been reported. This analysis suggests ‘typical’ heavy metal contents of particulate matter from secondary aluminium works as shown in Table A8.3. Mercury is treated in the same way for convenience, although most of the mercury emitted would be expected to be in the gaseous form.

Table A8.3 Calculated heavy metal contents of particulate matter from secondary aluminium processes based on Pollution Inventory data.

Metal	% of TPM[a]
Arsenic	0.15
Cadmium	0.45
Chromium	0.14
Copper	0.15
Mercury	0.56
Manganese	0.47
Nickel	0.14
Lead[b]	1.17
Vanadium	0.29
Zinc	0.48

a TPM is total particulate matter

b – the calculated figure for lead excludes data for one site for one year where lead was 78% of the particulate matter emission. This figure seemed far too high for the process, which recovers aluminium from general aluminium scrap.

The above figures could be used to calculate heavy metal emission factors for the secondary aluminium industry, however they are based on very limited data.

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from chlorine demagging and refining for uncontrolled reverberatory furnace operations. These are reproduced in Table A8.4

Table A8.4 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Refining	Uncontrolled	2.5	50	1080	E
		6	53.4	1150	
		10	60	1300	
			100	2150	
Chlorine demagging	Uncontrolled	2.5	19.8	99,500*	E
		6	36.9	184,500*	
		10	53.2	266,000*	
			100	500,000*	

Units: grams emitted per tonne of chlorine used.

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI currently includes the secondary aluminium industry under the “other non-ferrous metals” source sector. It does not have a separate emission factor for this category, but notes that the total emission for “other non-ferrous metals” was approximately 190 t in 1999.

The available US EPA emission factors do not appear to cover all of the stages of secondary aluminium production that might be expected to produce emissions of particulate matter, so it is not possible to estimate an overall emission factor for the sector. However, an estimate covering major process stages can be made by combining US EPA factors for sweating, smelting and demagging. US EPA factors for reverberatory furnaces, together with factors for sweating and demagging, all three processes being controlled using baghouses, would suggest a particulate emission factor of 2425 g/t or around 1600 g/t accepting a correction to the sweating furnace factor as mentioned above. This emission factor would still exclude any contribution from mechanical pre-treatments, degassing, and cast-house operations. However, the US EPA figures are some thirty years old and may well be inappropriate for current technologies.

A more reliable estimate is to use emission factors based on data reported to the Pollution Inventory and we recommend that the emission factors calculated from our revised estimates of emissions be used as a starting point. A number of factors, including the number of sites which do not report emissions, and the absence of data for Scottish, Northern Irish and Part B processes in England and Wales, make these emission factors rather uncertain and so opportunities should be sought for collecting further information, with which to improve the numbers.

Heavy metal emissions

As a starting point, we recommend that emission factors be calculated by applying the estimates of heavy metal contents of particulate matter, shown in Table A8.3, to the emission factors for particulate matter calculated above from our revised estimates of emissions.

Appendix A9: Primary lead/zinc

SNAP CODE: 030305

SOURCE ACTIVITY TITLE: Primary zinc production

NACE CODE: 2743

PARCOM-ATMOS CODE: 2.2.3

ACTIVITIES INCLUDED

Primary lead/zinc production covers the production of lead and zinc metal and alloys from ore concentrates.

GENERAL PROCESS DESCRIPTION

Lead and zinc are often found in association with other metals, especially copper, and therefore primary lead or zinc production is often carried out in conjunction with the production of other metals.

The main zinc ore is sphalerite (ZnS) which is treated at the majority of primary zinc processes. There are four main processes for zinc production:
- Electrothermic process;
- Roast-leach-electrowin process;
- Pressure leach-electrowin process;
- Imperial smelting process

The electrothermic process is only used at two plants, one in the USA and one in Japan, and will not be described further. The most common process is the roast-leach-electrowin process. The first stage of this process involves roasting the sulphide ore which converts the zinc sulphide to zinc oxide and the sulphur is removed as sulphur dioxide gas. Dust control is necessary due to the fine particle size of the roasted concentrate (calcine). The calcine is then leached by any of a number of techniques; generally these involve removal of zinc as zinc sulphate in solution. The solid residue will generally contain other values and will undergo further treatment. The solution is purified by addition of reagents to precipitate metallic impurities that are filtered out. The purified solution is then electrolysed with zinc being deposited on the cathodes. Finally, the zinc coating is melted and cast.

The pressure leach-electrowin process involves the leaching of zinc concentrates in an autoclave. Oxygen is injected and converts the sulphide direct to sulphate. The zinc sulphate solution can then be treated as described above to recover zinc.

Appendix A9: Primary lead/zinc

The Imperial smelting process (ISP) involves the production of a sinter from lead/zinc concentrates. The sintering process agglomerates and desulphurises the concentrates. The sinter is then crushed to the correct size for the smelter and mixed with coke. The mixture is then smelted in a blast furnace, the lead and zinc being reduced to the metallic elements. Liquid slag and molten lead collect at the base of the blast furnace and are tapped. Zinc is vaporised and passes out of the smelter in the off-gases. The zinc is then recovered by passing the gases through a spray of liquid lead that is subsequently cooled to solidify the zinc. The lead and zinc metals undergo various refining operations before final casting.

The major lead ore is galena (PbS) which is treated at the majority of primary lead processes. The main process for lead production involves sintering followed by smelting in a blast furnace. The sintering process requires a high proportion of recycled material in order to limit sulphur contents. The sinter product is screened and crushed. Sinter and coke is added to a blast furnace where the lead oxide is reduced to metallic lead. Molten lead and slag are tapped from the bottom of the blast furnace. Generally, zinc oxide present in the feed is not reduced and remains in the slag. It can be recovered later from the slag by further processing. A variant of this traditional smelting process is the Imperial Smelting Process (described above), which was developed for lead/zinc concentrates. Here, more highly reducing conditions are used so that zinc oxide is also reduced to zinc metal. At the operating temperatures of the blast furnace the zinc is volatilised and passes out of the top of the furnace.

Other processes include the Kivcet process where galena is converted to lead in a multi-compartment furnace. In one compartment of the furnace, oxygen is added to convert the sulphide to oxide and sulphate, while in another the oxide and sulphate is converted to lead metal and sulphur dioxide. In a final zone, lead oxide in the slag is reduced to the metal with carbon monoxide being evolved. Finally, lead smelting in an electric arc furnace is carried out at one Scandinavian plant — oxygen is injected into the furnace to oxidise the sulphide concentrate. These oxides react with further sulphide to form lead metal and sulphur dioxide. Carry-out of oxide fume is very high with this process and this has to be captured in a gas cleaning system and returned to the furnace.

The above smelting methods produce lead bullion, which will usually be contaminated with any or all of the following: copper, arsenic, antimony, silver and bismuth. This crude lead has to be refined in order to remove most of this contamination. Most lead is refined pyrometallurgically, the remainder being treated electrolytically.

Pyrometallurgical refining involves the manipulation of the temperature of the molten lead plus the addition of various reagents with the aim of precipitating the impurities. Copper is removed first, as the molten lead cools. It is removed as sulphide dross; if sulphur levels are insufficiently high then this must be added. Antimony and arsenic are removed by oxidation, either by addition of caustic soda and sodium nitrate or by injection of oxygen. Silver is removed by addition of zinc with which it forms an alloy. After cooling, the zinc-silver forms a solid crust while the still molten lead is pumped out. Finally, bismuth is removed by addition of a calcium-magnesium mixture.

Electrolytic refining makes use of hydrofluorosilicic acid as an electrolyte. Lead bullion makes up the cathodes while thin sheets of refined lead are the anodes. On application of an electric potential, lead dissolves at the cathode and deposits at the anode. The impurities remain at the cathode where they form a sludge that has to be removed periodically.

SOURCES OF PARTICULATE EMISSIONS

All of the processes used may give rise to particulate matter emissions. Handling of fine concentrates and other materials can cause dust emissions. The roasting of concentrates in the roast-leach-electrowin process and the sintering and smelting of concentrates in the Imperial smelting process are likely to be major sources of particulate matter. In comparison, hydrometallurgical processes such as leaching and electrolysis are unlikely to have significant emissions. Refining of molten metals and casting operations are likely to emit particulate matter, for instance during tapping.

SOURCES OF HEAVY METAL EMISSIONS

Lead and zinc ores typically may contain cadmium, copper, silver and iron in addition to lead and zinc, and therefore these metals may be emitted during the production process. The emissions of heavy metals are closely associated with particulate emissions and therefore the sources of emissions will be similar to those described above for particles.

ABATEMENT MEASURES AVAILABLE

Dust arrestment devices are in common use on primary zinc plant. Cyclones, electrostatic precipitators, fabric filters and scrubbers are all employed. Roasters generally employ cyclones, ESPs and venturi scrubbers. Sinter plant for the Imperial smelting process also use ESPs or venturi scrubbers. A wet scrubber has to be used for the smelting process. Other parts of the ISP usually use fabric filters.

UK SITUATION

UK plants

In the UK, the only primary lead/zinc production plant is located at Avonmouth, where the Imperial smelting process is used to produce lead and zinc from lead/zinc concentrates.

Most of the feedstocks are imported and are delivered from the local docks on a long conveyor belt. Most but not all of the belt is enclosed and transfer points are sealed. The concentrates are stored in a building although some other feed materials may be stored in the open.

Concentrate is mixed with recycled fines from the sinter process before sintering. Approximately 250 tonnes of material is sintered each hour. The sinter machine is enclosed, and off gases are first cleaned in a venturi scrubber for solids removal and then sent for gas cleaning. After cooling, the gas is treated in two ESPs.

The sinter is then crushed and sized with the undersize materials being further crushed, cooled by quenching and then being returned to the sinter machine. All parts of the sintering process where dry materials are handled are enclosed and vented through scrubbers.

Hot sinter is charged to the blast furnace with pre-heated coke. Preheated air reacts with the coke, generating enough heat to form slag. The carbon monoxide produced reduces the metal oxides to lead and zinc. Lead and molten slag accumulate in the bottom of the furnace and are tapped out at intervals. Fume is extracted and sent for gas treatment. The molten lead and slag collect in the forehearth, separate by gravity, and are collected separately. The slag is cooled and granulated using a water spray, with gas extraction and scrubbing being used to clean the waste gas. Zinc is volatilised in the furnace and passes out of the furnace with the off-gases. These are then quenched in a splash condenser using a stream of molten lead. Metals in the furnace off-gas (including amounts of cadmium and lead, as well as zinc) are absorbed into the molten lead which is then cooled, causing the zinc and cadmium to come out of solution. The zinc floats to the top of the mixture and is removed.

The gases from the lead splash condenser are treated using two stages of water washing with the collected sludge being thickened and returned to the sinter plant. The cleaned gas is used to preheat coke for the blast furnace.

The lead collected from the smelter is transferred to a kettle and allowed to cool until copper is precipitated. Sawdust is added and the mixture stirred causing copper dross to form. This is removed and the lead transferred to another kettle, reheated and cast.

Zinc is refined by reflux distillation in two columns with a number of refractory trays. In the first column, zinc and cadmium are separated from the lead by distillation. In the second column, which is operated at a lower temperature, the cadmium is distilled leaving the zinc. The distilled cadmium (including significant levels of zinc) is then refined further to produce cadmium. The run-off from the second distillation column is 99.99% pure. Run-off from the first column is cooled to separate the lead and then the arsenic and antimony are removed by adding sodium. Zinc refined in the latter way is less pure than that produced from the second distillation column. Both grades of zinc are cast into ingots.

UK activity statistics

The UK Minerals Yearbook, includes data on annual production of primary lead and zinc.

Abatement measures used in UK plants

Emission controls in place at the UK's only primary lead/zinc producer may be summarised as follows:

- Enclosure of most of the conveyor belt carrying concentrates from the docks to the smelter;
- Storage of concentrates in a building;
- Washing of road ways and use of wheel washes;
- Ducting of storage bins in the proportioning plant to gas cleaning for particulate matter removal;

- Ducting of off-gases from the sinter plant to gas cleaning for particulate matter removal;
- Ducting of off-gases from the smelter to gas cleaning for particulate matter removal;
- Hooding of tapping points, slag granulation launder and drossing pots with ducting to gas cleaning equipment;
- Ducting of off-gases from the zinc refinery to gas cleaning for particulate matter removal;
- Transportation of drosses in covered containers.

Most gas cleaning is done using wet scrubbing using venturi and impactor scrubbers, although some fabric filters are used, for example on the reflux columns.

PARTICULATE MATTER EMISSION FACTORS

The EIPPCB (2000b) suggests that emissions from the Imperial Smelting Furnace and sinter plant are 30 – 90 g (zinc)/t (metal) and 5 – 40 g (lead)/t (metal). The report also notes that zinc constitutes approximately 50% of the dusts emitted. This implies a dust emission factor in the range of 60 – 180 g (dust)/t (zinc plus lead product).

Details of emission factors published by the US EPA (2000) for primary zinc production are summarised in Table A9.1. Although the processes used in the US are different to those currently used in the UK, some of them give an indication of the potential levels of particulate material that might be released. The high levels of uncertainty must be noted and this is particularly true regarding the releases from the sinter plant. Based on the data in Table A9.1, it seems possible that emission factors up to several thousand g/t are possible.

Table A9.1 Primary zinc production particulate emission factors (US EPA, 2000)

Process Stage	**Abatement**	**Emission factors g/t (zinc)**	**Quality rating**
Sinter plant	Cyclone	24,100	E
Sinter plant	Cyclone and ESP	8,250	E
Fugitive emissions (slab smelting):			
Sinter plant – wind box	Uncontrolled	120 – 550	
Sinter plant – discharge screens	Uncontrolled	280 – 1,220	E
Casting	Uncontrolled	1,260	E

Table A9.2 lists some emission factors quoted by the US EPA (2000) for lead production. Whilst again not specific to the ISP technique, they have been selected to give an indication of probable particulate emission factors.

Table A9.2 Primary lead production total particulate emissions (US EPA, 2000)

Process Stage	Abatement	Emission factor	Unit	Quality rating
Blast furnace operation	baghouse	430[a]	g/t (bullion processed)	E
Sinter machine leakage	ESP / scrubber	58	g/t (sinter produced)	E

a this factor is actually for PM_{10}, however the source gives a particulate matter emission factor of 210 g/t which suggests that one or both figures are erroneous

As stated above, these US emission factors do not relate to the Imperial Smelting Process, but might give a crude indication of a factor for the UK process. Because of the differences in processes however this would not be a recommended approach.

Emissions of total particulate from the UK site, as reported in the Chemical Release Inventory (1996 and 1997) and Pollution Inventory (1998 and 1999) are given in Table A9.3

Table A9.3 Emissions from UK primary zinc/lead smelting

Year	Total particulate emission (t)	Slab zinc production (kt)	Lead bullion production (kt)	Emission factor (g/t lead & zinc)
1996	191.8	96.867	41.991	1381
1997	184.2	107.704	38.000	1264
1998	129.5	99.6	37.927	942
1999	82.0	132.8	40.635	473

The EMEP/CORINAIR (1999) Guidebook does not include any emission factors for particulate matter and, in any case, does not consider the Imperial Smelting Process.

HEAVY METAL EMISSION FACTORS

Emission factors given by the US EPA are shown in Tables 9.4 and relate to emissions of lead from lead smelting.

Table A9.4 Emission factors for lead from primary lead production (US EPA, 2000)

Process Stage	Abatement	Emission factor	Unit	Quality rating
Blast furnace operation	baghouse	34	g/t (bullion processed)	E
Sinter machine off-gas	ESP / scrubber	9	g/t (sinter produced)	E
Sinter machine fugitives	uncontrolled	16	g/t (sinter)	E

No detailed data are given by the EIPPCB but the EMEP/CORINAIR Guidebook suggests the emission factors shown in Table A9.5 for primary lead and primary zinc production.

Table A9.5 Heavy metal emission factors for primary lead and primary zinc production (EMEP/CORINAIR, 1999) – figures in g/tonne of product

Metal	Primary lead	Primary zinc
Arsenic	0.5-10	
Cadmium	1-15	
Copper	5-10	
Lead	200-2000	500
Mercury	3	20
Zinc	20-100	10000

As with other sources, the EMEP/CORINAIR guidebook does not specifically cover the Imperial Smelting Process. As a result, the most appropriate data would seem to be those in the Pollution Inventory. These are shown in Table A9.6.

Table A9.6 Emissions of heavy metals reported in the Pollution Inventory (figures in kg)

Metal	1996	1997	1998	1999
Arsenic	661	719	736	610
Cadmium	1683	2297	979	1252
Chromium			73	31
Copper			633	303
Lead	19540	20450	12378	10867
Mercury			1352	294
Nickel			36	24
Zinc	49690	47880	31687	23152

These data can be used with the activity data given in Table A9.3 to calculate emission factors as shown in Table A9.7.

Table A9.7 Emission factors for heavy metals based on Pollution Inventory data (figures in g/tonne lead bullion & slab zinc)

Metal	1996	1997	1998	1999
Arsenic	4.76	4.94	5.35	3.52
Cadmium	12.1	15.8	7.12	7.22
Chromium			0.528	0.179
Copper			4.60	1.75
Lead	141	140	90.0	62.7
Mercury			9.83	1.70
Nickel			0.265	0.138
Zinc	358	329	230	133

The above data do not take account of fugitive sources, but a report by Entec (1996) suggested that fugitive emissions might be 2 to 4 times process emissions in the case of lead, and 2 times process emissions in the case of cadmium. However, Britannia Zinc (Nash, 2001) have suggested that, since they have not any data on which to make any estimate of fugitive emissions, that Entec's figures should not be used. The company plan to make measurements in the next year and so it is recommended that no attempt be made to estimate emissions until more data become available.

The Entec report quoted above also gives the following data for actual emissions of lead, cadmium and mercury from process vents at the plant during 1994/95:

Lead	26 tonnes	189 g/tonne (lead bullion & slab zinc)
Cadmium	2.4 tonnes	17.4 g/tonne (lead bullion & slab zinc)
Mercury	2.9 tonnes	21.0 g/tonne (lead bullion & slab zinc)

These figures have been converted to emission factors using activity data for 1994 taken from the UK Minerals Yearbook for 1999 (British Geological Survey, 1999).

On comparing these emissions data with those reported in the Pollution Inventory for 1996 onwards, it seems probable that those in the Pollution Inventory are for emissions from process vents only and exclude fugitive emissions. This will presumably be true for particulate matter emissions as well.

SIZE DISTRIBUTION

A generalised size distribution has been given by the US EPA (2000) for the uncontrolled smelting, refining, and melting of metals (excluding aluminium). This is reproduced in Table A9.8

Table A9.8 Mineralised particle size distribution for the smelting, refining, and melting of metals (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size		
			Best	Min	Max
Smelting, refining, and melting of metals (excluding aluminium)	Uncontrolled	1	72*	–	–
		2.5	82	63	99
		6	89	75	99
		10	92	80	99
			100	100	100

* Extrapolated

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI currently uses particulate matter emission factors based on the Pollution Inventory data given above, although the value for 1999 needs to be updated – the existing number was calculated using 1998 activity data since the 1999 data were not available at the time. Emission factors for PM_{10} are then calculated by assuming that 92% of particulate matter is sub 10 microns, based on the US EPA default size profile for smelting, refining and melting of metals (excluding aluminium). This gives a PM_{10} emission factor of 867 g/t (lead plus zinc) in 1998 and 435 g/t (lead plus zinc) in 1999, which are both significantly lower than the US EPA figure for sinter plant. However, the US EPA data are very old (published in 1977), have the lowest data confidence rating, and do not inspire confidence. The Pollution Inventory figures are significantly higher than those given by EIPPCB, but it seems sensible to use the higher figures given that these are reported by the operator. It is recommended therefore that the NAEI continues to use the Pollution Inventory figures although consideration should be given to collecting more information on emissions in the years prior to reporting to the Pollution Inventory.

One area of concern with the Pollution Inventory data is that it these data relate to emissions from process vents only. However, no measurements have been made which would allow an estimate of fugitive emissions to be made (although measurements are planned by the operator).

Heavy metal emissions

The NAEI uses emission factors based on Pollution Inventory data, and, in the absence of detailed data from other sources, it is recommended that this approach is continued. As with particulate matter emissions, it is also recommended that some effort be made to collect data for those years prior to the reporting of data to the Pollution Inventory and also that, as a matter of urgency, the issue of whether the Pollution Inventory data covers fugitive emissions be resolved. In the absence of any definite information either way, it would seem sensible to modify the Pollution Inventory data by assuming that fugitive emissions of heavy metals are 3 times the emissions given in the Pollution Inventory, with the exception of cadmium where a figure of 2 times the Pollution Inventory data is recommended.

Appendix A10: Secondary lead

SNAP CODE: 030307 040300

SOURCE ACTIVITY TITLE: Secondary lead production

NACE CODE: 2743

PARCOM-ATMOS CODE: 3.0

ACTIVITIES INCLUDED

The secondary lead industry is concerned with the production of lead and lead products from primary metals or scrap. Important sectors include the recycling of batteries and recycling of lead piping and sheet. This sector also includes the refining of crude lead to produce saleable grades. This chapter does not include the use of lead, lead alloys or lead based materials to produce fabricated metal products or batteries.

GENERAL PROCESS DESCRIPTION

Battery recycling involves principally the recovery of lead from car batteries. These are approximately 75% lead by weight. The remainder is acid, rubber and plastic (polypropylene and PVC). Batteries are broken up in hammer mills, with the fragments being flushed from the mill using water. Screens are used to separate lead paste from the remainder and classifiers are used to separate the screen oversize on the basis of density. Typically a polypropylene fraction as well as a lead fraction will be recovered from the remaining materials.

The lead paste is dewatered, and sent to the smelter together with the lead fraction from the classifiers. The smelting process melts the charge, reduces any metal oxides to the metal, and allows separation of metallic and non-metallic components. Smelting may be carried out in various forms of furnace including rotary, reverberatory, blast or Isasmelt furnaces. Crude lead product is tapped from the smelting furnace and then may be either sent to refining or cast into blocks for subsequent refining.

Refining is generally done in kettle furnaces. The crude lead is heated and when molten, reagents are added to remove impurities as drosses. Air blowing can also be used to remove the impurity antimony. Drosses are removed by automatic means or using perforated ladles. Alloying metals may be added before the purified lead is cast.

Other scrap products such as piping and sheets are melted and refined in kettles in a similar manner to battery lead.

Other secondary lead processes involve the manufacture of lead products. Although no detailed descriptions of these processes are given, the main stages likely to give rise to particulate matter emissions are melting and casting operations. Production of lead oxide by the oxidation of molten lead has the potential to give rise to significant levels of particulate matter.

SOURCES OF PARTICULATE EMISSIONS

In the case of battery recycling, emissions of particulate could occur during battery breaking and during smelting and refining. The following activities are likely to give rise to the most significant emissions:

- battery breaking;
- smelter charging;
- smelter tapping;
- refining operations (particularly air blowing);
- dross removal.

Emissions from other secondary lead processes are likely to occur only as a result of melting of lead. In the case of lead oxide manufacture, significant particulate matter emissions could occur from the oxidation of molten lead.

ABATEMENT MEASURES AVAILABLE

Waste gases from battery breaking are typically treated in wet scrubbers for both particulate and acid removal. Dusty feed materials for smelting may be wetted to reduce fugitive emissions. Waste gases from smelters and refining kettles can be captured using hoods and evacuated to gas cleaning equipment, typically fabric filters.

In other secondary lead processes, melting furnaces can be enclosed and waste gases vented to fabric filters.

UK SITUATION

UK plants

During 1999, there seems to have been about 8 processes regulated by the Environment Agency in England and Wales which fit into the category ‘secondary lead production’. Two UK processes recycle batteries, four recycle lead sheet and pipe, one processes electrical cables and switchgear, and one is involved in the production of lead from drosses. No secondary lead producers are known in Scotland or Northern Ireland. The processor of electrical cables and switchgear is now closed.

One battery recycling process uses an Isasmelt plant to recover lead. Oil and air are injected into the Isasmelt furnace to provide heat, and a flux is used to promote slag formation. Following smelting, the crude lead bullion is refined to remove antimony and copper. The second battery recycling process uses rotary furnaces to smelt lead pastes using coke breeze, cast iron borings and sodium carbonate as additional feedstocks. This plant has gas fired refining kettles and also

has a lead shot production facility involving spraying of molten lead at just above its freezing point.
The processes which recover lead from other scrap and drosses, generally involve melting of the scrap in gas or oil fired furnaces followed by refining to remove impurities such as tin and antimony and then casting of ingots etc., which may be further processed to produce sheet or other products.

In addition, in the UK there is one lead bullion refinery. This consists of two refinery processes, one treating imported bullion and the other treating bullion from the UK lead/zinc smelter. Key features of this plant are as follows.

- The imported lead contains high levels of silver. Refining is carried out in a series of kettles. Lead is melted, and silver removed by addition of zinc in a two-stage process. After removal of silver, residual zinc must be removed by heating the lead while maintaining a vacuum above the surface of the metal. Zinc is boiled off and condenses on the lid, which is water-cooled. The final refining stage involves removal of antimony using caustic soda and sodium nitrate, oxidising the antimony, which then forms dross. Finally, the lead is cast either as pure metal or alloy.

- The second refinery is more complicated since it treats lead with higher impurity levels. After melting, copper is removed as dross by addition of sulphur. Excess sulphur is removed by addition of caustic soda. Zinc is added to remove silver in a similar way as for the other refinery. The lead is then treated with caustic soda and sodium nitrate to remove antimony. Bismuth is removed through addition of metallic calcium and magnesium. In a final refining step, sodium hydroxide and sodium nitrate are added to remove excess calcium and magnesium. Finally, the lead is cast either as pure metal or alloy.

- Other processes on site include the silver plant, which treats silver/zinc drosses from each refinery. First, the material is melted, separating into two liquid phases; a silver/zinc alloy floats on top of a lead layer. The lead layer is siphoned off and the silver/zinc tapped. Zinc is removed from this alloy by means of vacuum distillation. This leaves high silver content (70%) lead/silver bullion. The lead is removed by oxidation using oxygen injection.

- The by-product plant treats various materials from the other processes on site. The material is pelletised and smelted using coke in a cupola. After decopperising the molten lead by dropping its temperature, it is cast and returned to the refinery.

UK activity statistics

Statistics on secondary lead production are available from the British Geological Survey's Minerals Yearbook, which is published annually. Production of lead or lead alloys from batteries is about 100,000 t per year (Hatch Associates Ltd, 1993).

Abatement measures used in UK plants

Almost all of the UK's secondary lead processes use bag filters to abate emissions from furnaces. The battery recycling processes also have wet scrubbers for abating emissions from battery breaking mills. It is not known to what extent fugitive emissions are controlled or prevented in these plants.

EMISSION FACTORS

The EIPPCB (2000b) offers emissions of lead from various battery recycling operations (see Table A10.3). No figures for total particulate matter are given, but they may be estimated to be roughly double the lead emissions, or up to about 50 g (TPM)/t (lead).

Various particulate matter emission factors quoted by the US EPA (2000) are listed in Table A10.1. Most of the figures in this table are for uncontrolled processes, so that it might be expected that emission factors for controlled processes could be around two orders of magnitude less. Consequently, particulate matter emission factors around 1,000 g/t might seem appropriate for secondary lead production.

Table A10.1 Secondary lead production total particulate emission factors (US EPA, 2000)

Process Stage	Abatement	Emission factor	Unit	Quality rating
Reverberatory furnace	Uncontrolled	162,000	g/t (metal produced)	–
Reverberatory furnace	Miscellaneous	500	g/t (metal produced)	–
Rotary sweating furnace	Uncontrolled	14,286	g/t (metal charged)	–
Kettle refining or casting	Uncontrolled	20	g/t (metal cast)	–

The Pollution Inventory includes emissions for all of the UK secondary lead processes mentioned earlier in this appendix. Table A10.2 summarises the available data.

Table A10.2 Emissions from UK secondary lead processes

Year	Total particulate emission (t)	Secondary lead production (kt)	Emission factor (g/t lead)
1996	34.24	174.7	196
1997	37.22	170.9	218
1998	31.38	163.5	192
1999	31.35	162.6	193

It should be noted that some of the processes do not report emissions in 1998 and 1999 other than to record that emissions are below the *de minimis* limit for reporting. An estimate of the actual quantity emitted is therefore assumed to be the same as reported for earlier years. No attempt has been made to obtain emissions data for the two processes which have not reported emissions in any year since these processes are believed to be very small.

PM_{10} emissions are only reported by one process operator. The data suggest that PM_{10} emissions comprise 87.5% of particulate matter emissions.

HEAVY METAL EMISSION FACTORS

The EIPPCB quote lead emission factors for various battery recycling processes; these data are shown in Table A10.3.

Table A10.3 Mass release of lead from some European processes (EIPPCB, 2000)

Process stage	Abatement	Emission factors (g lead/t metal)
Battery — whole	Controlled	< 15
Battery — desulphurised paste	Controlled	10
Battery — desulphurised paste	Controlled	5 – 25
Battery — oxide paste sold	Controlled	5 – 25
Batteries + extra paste	Controlled	5 – 25
Battery — MA process	Controlled	20

EMEP/CORINAIR (1999) quotes emission factors for lead of 100 – 770 g (lead)/t (lead product) for secondary lead processes. The Pollution Inventory also includes some reported emissions for heavy metals, although the data are very patchy. This can be used to calculate the following 'typical' metal to particulate matter emission ratios. Mercury is treated in the same way for convenience although most of the emission would be expected to be in the gaseous form. Table A10.4 shows the resulting factors, which are calculated for each year between 1996 and 1999, although a weighted average is also given.

Table A10.4 Typical ratio of metal emissions to particulate matter emissions based on Pollution Inventory data (expressed as percentages)

Metal	1996	1997	1998	1999	Average
Arsenic	1.14	0.125	0.103	0.129	0.346
Cadmium	1.36	0.392	0.676	0.484	0.664
Copper	1.06	0.0900	0.0859	2.19	0.401
Lead	14.4	9.94	13.5	11.7	12.2
Mercury	0.0166	0.708	0.169	0.312	0.192
Selenium	0.0321	0.0550	0.0381	0.159	0.0476
Zinc	16.1	2.14	4.05	2.73	4.03

In theory, these data can be used to generate emission factors by combining them with the data given in Table A10.2. Some screening of data to remove suspect values might however be necessary since the values in Table A10.4 are surprisingly variable from year to year.

SIZE DISTRIBUTION

A generalised size distribution has been given by the US EPA (2000) for the uncontrolled smelting, refining, and melting of metals (excluding aluminium). This has been given in Appendix 9 as Table A9.8.

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI currently uses emission factors based on the Pollution Inventory data. These factors are somewhat higher than the EIPPCB figure but appreciably lower than the EMEP/CORINAIR and US EPA figures. However, there is considerable uncertainty associated with all the data on secondary lead emissions, and it is therefore recommended that the use of estimated factors based on Pollution Inventory data is continued in the absence of better data.

Heavy metal emissions

As with particulate matter, NAEI factors are based on Pollution Inventory data. This general approach should be continued, although some revisions to the calculation of emission factors might be adopted in order to eliminate the use of less reliable data if this can be identified.

Appendix A11: Secondary copper

SNAP CODE:	**030309** **040300**
SOURCE ACTIVITY TITLE:	**Secondary Copper Production**
NACE CODE:	**2743**
PARCOM-ATMOS CODE:	**3.0**

ACTIVITIES INCLUDED

This chapter includes the manufacture of high-grade copper metal and alloys from low-grade copper and copper scrap.

GENERAL PROCESS DESCRIPTION

The secondary copper sector consists of production of high-grade copper from various low-grade sources such as blister copper, copper drosses and residues and low and high-grade scrap. The sector may also be defined so as to include the casting of high-grade copper into semi-finished items such as wire and tubes. However, this chapter will not cover these processes.

Scrap may undergo various pre-treatment operations such as sorting, cleaning and shredding. Burning may be used to remove the insulation from copper wire.

The secondary production of copper can be via a number of routes. One method begins with the smelting of low-grade scrap with coke and fluxes in a blast furnace. Black copper is produced together with slag and fume. The fume, containing lead, zinc and tin, is collected in a gas treatment plant. The black copper, which contains 70–80% copper, has to be refined further. Medium-grade scrap is smelted in a reverberatory furnace with fluxes and fuel. The products are copper, ready for the anode furnace, and slag. The black copper from the blast furnace contains impurities such as iron, lead, nickel, tin and zinc and must be treated in a converter. This is a furnace in which the black copper is melted and air blown through the molten metal to oxidise impurities, which are then removed in the slag. The products are ‘rough copper’ and slag and are discharged from the mouth of the converter. The rough copper is transferred to the anode furnace where further air, or oxygen, are blown through the metal to oxidise remaining impurities. Following removal of impurities, excess oxygen is removed by reduction using natural gas. The copper is then cast into moulds or continuous strip.

An alternative to the blast furnace, reverberatory furnace and converter is the top blown rotary converter (TBRC) process which combines the functions of all three. TBRCs are housed inside enclosures so capture of emissions is good.

Electrolysis is used to refine the copper. Copper anodes are suspended in a bath of acidified copper sulphate. The cathodes can be made from copper or, in a variant of the process, from stainless steel. Copper is dissolved from the anodes and deposits on the cathodes. Impurities form a sludge on the base of the electrolysis cell or dissolve in the electrolyte.

SOURCES OF PARTICULATE EMISSIONS

Particulate emissions could occur from the following stages:

- Pre-treatments, especially involving burning of insulation and other material, and shredding operations;
- Smelting operations, especially waste gases from blast furnaces;
- Transfer operations, including charging of furnaces and tapping of products and slag.

SOURCES OF HEAVY METAL EMISSIONS

Zinc, lead, cadmium and arsenic can be present in both the vapour phase and in dusts released during the process stages.

ABATEMENT MEASURES AVAILABLE

Sources of particulate matter are generally extracted to end-of-pipe equipment where the material can be recovered. Bag filters are most commonly used in the industry due to the fine nature of the particulate matter evolved.

UK SITUATION

UK plants

There was one UK refinery, using medium and high-grade scrap. The refinery contained a blast furnace, reverberatory furnace and converters, but closed down during 2001.

UK activity statistics

The United Kingdom Minerals Yearbook, published annually by the British Geological Survey, gives annual production of secondary copper.

Abatement measures used in UK plants

The off-gases from the blast furnace, reverberatory furnace and converters are ducted to bag filters.

PARTICULATE EMISSION FACTORS

The EIPPCB (2000b) gives the range of dust emission factors from secondary copper production to be 100 – 1000 g (dust)/t (product metal). This is based in part on data for the UK plant.

The US EPA (2000) has published emission factors for a number of secondary copper processes; the most relevant of these are shown in Table A11.1. These emission factors are not entirely relevant to the UK situation but suggest that the emission factor for secondary copper production could be as high as 5,100 g/t. In the UK, fabric filtration is used rather than ESPs, and these would be expected to be more efficient. In addition, fugitive emissions from the UK process might be expected to be reduced through efficient capture of emissions.

Table A11.1 Secondary copper production particulate emission factors (US EPA, 2000)

Process	**Feedstock**	**Abatement**	**Emission factors g/t (ore processed)**		**Quality rating***
			Total	**PM_{10}**	
Cupola	Scrap copper and brass	Uncontrolled	35,000	32,100	B
Cupola	Scrap copper and brass	ESP	1,200		B
Cupola fugitive emission	–	Uncontrolled		1,100	E
Reverberatory furnace	Copper	Uncontrolled	2,600	2,500	B
Reverberatory furnace	Copper	Baghouse	200		B
Reverberatory furnace	Brass and bronze	Uncontrolled	18,000	10,800	B
Reverberatory furnace	Brass and bronze	Baghouse	1,300		B
Reverberatory furnace fugitive emission	–	Uncontrolled		1,500	E

* Quality rating for total particulate matter. Quality ratings for PM_{10} are E.

The Pollution Inventory gives data for 1995, 1996, 1998 and 1999. The emissions reported for the first year are very much lower than subsequent years and should probably be ignored. The data for 1996 is also somewhat lower than later emissions data and may be less complete than the 1998 and 1999 data. Alternatively, the process may have changed leading to an increase in emissions. The Pollution Inventory data are shown in Table A11.2.

Table A11.2 Emissions from the UK secondary copper process

Year	Total particulate emission (t)	Secondary copper production (kt)	Emission factor (g/t copper)
1996	47.72	56.6	843
1998	61.30	54.0	1135
1999	62.65	50.0	1253

HEAVY METAL EMISSION FACTORS

EMEP/CORINAIR (1999) provides emission estimates for secondary copper production, assuming that ESPs with 99% efficiency are used for waste gas treatment. These estimates are provided in Table A11.3

Table A11.3 Compilation of emission factors for secondary copper production (g/tonne of copper produced)

Metal	Emission factor
Arsenic	2
Antimony	3
Cadmium	2-4
Copper	20-150
Lead	50-130
Nickel	1
Zinc	250-500

The Pollution Inventory includes data on heavy metal emissions from the UK secondary copper process and these data have been used to generate the emission factors shown in Table A11.4

Table A11.4 Heavy metal emission factors for the UK secondary copper process taken from the Pollution Inventory (g/t of copper produced)

Metal	1996	1998	1999
Arsenic	4.93	3.89	3.60
Cadmium	1.11	2.22	1.52
Copper	118	126	125
Nickel	0.247	2.44	6.90
Lead	81.3	67.6	94.8
Zinc	228	285	261

As with the particulate matter emissions data, the data reported for 1996 give lower emission factors than do the data for 1998, especially in the case of cadmium, nickel and zinc. This is unexpected, but may be due to a change in the process. No data are available for 1997.

SIZE DISTRIBUTION

The size of fume from secondary copper processes has been reported to be very fine, generally sub-micron. However no size distribution has been found. A generalised size distribution has been given by the US EPA (2000) for the uncontrolled smelting, refining, and melting of metals (excluding aluminium). This has been given in Appendix 9 as Table A9.8. This would give a figure of 92% of particulate matter in the sub10 micron size fraction.

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI uses a PM_{10} emission factor derived from Pollution Inventory data, assuming that 92% of the emissions of particulate matter reported are sub-10 microns. This figure is appreciably below the US EPA figure of 5,100 g/t, but this is roughly as expected. It is at the high end of the EIPPCB's range but is nonetheless recommended as the best available figure. Given the difference between the Pollution Inventory derived figure and the US EPA data, it might be desirable to investigate whether emissions might have been higher in the years prior to reporting to the Pollution Inventory, when the level of control might have been less good.

Heavy metal emissions

The emission factors derived from Pollution Inventory data are recommended as the best available figures. However, some further investigation would be warranted to discover whether the lower emission factors for 1996 are realistic. As with particulate matter, it would be desirable to investigate further whether emissions might have been higher in the years prior to reporting to the Pollution Inventory.

Appendix A12: Cement production

SNAP CODE: 030311

SOURCE ACTIVITY TITLE: Cement

NACE CODE: 26500

PARCOM-ATMOS CODE: 2.8.1

ACTIVITIES INCLUDED

This chapter covers particle emissions released from rotary kiln exhaust and other operations in cement manufacture such as milling, storage and on-site transportation. It does not include the production of any raw materials, for example by quarrying.

GENERAL PROCESS DESCRIPTION

The vast majority of cement production is of Portland cement but small amounts of other types, including blast furnace cement, are produced for specialised applications.

Portland cement manufacturing can be divided into the following primary process stages:

- Raw material handling;
- Fuel grinding if solid fuel is used;
- Kiln feed preparation;
- Pyroprocessing in a rotary kiln to form clinker;
- Finished cement grinding and blending.

The raw materials (limestone, sand, shale, clay and iron oxide) are first brought to site, and some of the material will normally be conveyed from nearby quarries or open pits. The materials are then mixed and crushed in carefully specified proportions. Further grinding of the mixture is carried out to produce a raw mix (raw meal) of the correct particle size and chemical properties.

The raw meal is converted into cement clinker by pyroprocessing in large rotary kilns which consist of a refractory lined cylindrical steel shell slightly inclined to the horizontal and rotating at 1 – 3 rpm. Raw material is fed in at the upper end and gradually moves downward towards the lower end where there is a burner providing counter current heating.

Most cement kilns now use the dry process, in which raw mill material is fed into the rotary kiln dry. Before passing into the kiln the material may be preheated in a vertically arrayed multi-cyclonic preheater, in which the rising hot gases exiting the kiln contact the downward flowing raw materials. Some dry processes also employ a precalciner stage beneath the preheater, just before the raw material enters the kiln. Preheaters and precalciners often have an alkali bypass between the feed end of the rotary kiln and the preheater to remove undesirable volatile components.

The use of the wet process, where the ground meal is mixed with water and fed into the kiln as a slurry, is now less common. The wet process uses about 40% more energy than the dry process.

Cement kilns are highly energy intensive and use large quantities of fuel. The fuel price has a critical effect on profitability and this has led to a drive to find cheaper fuels. In the past coal, oil and natural gas were used but since the late eighties most plants have switched to coal or mixtures of coal and waste fuels, such as tyres and secondary liquid fuels (mainly waste solvents and other chemicals which would otherwise be sent to specialist chemical incinerators for disposal). This practice has led to concerns about emissions of toxic substances into the air.

Irrespective of the type of process used, the last stage involves cooling the clinker. As the hot clinker comes off the end of the lower end of the kiln it is rapidly cooled by ambient air in a clinker cooler. There are many different designs of cooler, the most common of which is a travelling grate with under-grate fans that blow cool air through the clinker. Some of this air can be used for combustion, but some is vented to atmosphere or used for drying solid fuels and raw materials.

Finally, the cooled clinker is then mixed with 4 to 6% gypsum and ground to a fine homogeneous powder to produce the final product, which is then stored in silos prior to bulk transportation or bagging.

SOURCES OF PARTICULATE EMISSIONS

The potential sources of particulate matter at cement plants include the following process stages:

- Dry raw material storage;
- Grinding and blending of dry raw materials;
- Preheating and precalcining of raw materials and clinker production (rotary kiln);
- Clinker cooling;
- Clinker grinding and blending;
- Storage, bulk loading, packaging of final product;

The main source of particulate matter is from the kiln and clinker cooler exhaust stacks. Often, some of the cement kiln dust is recycled into the process to produce more clinker but this is limited by the alkali content of the product.

Fugitive emissions of particulate matter can arise from materials handling and transfer operations, as well as from raw milling dry process facilities, and finish milling operations.

SOURCES OF HEAVY METAL EMISSIONS

Raw materials and fuel contain metal elements and their concentrations can vary widely. Non-volatile heavy metal compounds (arsenic, chromium, copper, nickel and vanadium) remain within the process and exit the kiln as part of the clinker. Semi-volatile metals (cadmium, lead, selenium and zinc) are partly volatilised in the hotter parts of the kiln and condense on raw material in the cooler parts of the kiln. This leads to a cyclic effect within the kiln which eventually results in an equilibrium between input and output via cement clinker. Mercury is readily volatilised and may react with feedstock or precipitate out in cooler parts of the process. All the metals can exit the process via the kiln stack or as fugitive dust emissions arising from the storage and handling of raw and finished materials and solid fuels.

ABATEMENT MEASURES AVAILABLE

Particulate matter emissions from rotary cement kilns are normally controlled by fabric filters and electrostatic precipitators. Electrostatic precipitators in this sector can achieve dust concentrations of 30 to 40 mg/m^3 while fabric filters in this sector are commonly delivering values between 20 and 50 mg/m^3. Clinker cooler discharges are frequently controlled with fabric filters or electrostatic precipitators. A few gravel bed filters have also been used to control clinker cooler emissions. Typical outlet particulate loadings are identical to those reported for kilns.

Process fugitive emission sources are normally captured by a ventilation system and the dust is collected by fabric filters.

UK SITUATION

UK plants

There are 23 cement plants with kilns in the UK, and one cement plant with mills only (EIPPCB, 2000c). Less than 20% of UK plants use the wet process.

UK activity statistics

Annual production of clinker is available from the British Cement Association or given in the UK Minerals Yearbook.

Abatement measures used in UK plants

A mixture of fabric filters and electrostatic precipitators are used to control the particulate emissions associated with coal, cement and raw meal crushing.

Electrostatic precipitators are used almost exclusively in the UK to control the particulate emissions associated with kiln operations, although some fabric filters are beginning to be introduced at new plants. The same situation applies for the control of clinker cooler discharges.

PARTICULATE EMISSION FACTORS

The EIPPCB (2000c) gives a dust emission factor range from cement kilns of 10 – 400 g/t (clinker). This is presumably after abatement as installed at European plants. No data are given for other stages of cement production.

EMEP/CORINAIR (1999) give emission factors for a number of processes in cement manufacture (Table A12.1). No information is given on what abatement, if any, is included in these figures. The totals for Portland cement and blast furnace cement are 250 g/t and 115 g/t respectively.

Table A12.1 Particulate matter emission factors for cement processing and manufacture (EMEP/CORINAIR, 1999)

Process	**Abatement**	**Emission factor g/t (product)**	**Quality rating**
Clinker production	Not specified	212	E
Cement production	Not specified	29 – 46	E
Portland cement, dry process, fuel preparation	Not specified	20	E
Portland cement, dry process, clinker firing	Not specified	100	E
Portland cement, dry process, cement milling	Not specified	100	E
Portland cement, dry process, cement shipping	Not specified	30	E
Blast furnace cement, raw materials transport	Not specified	55	E
Blast furnace cement, cement drying, milling, shipping	Not specified	60	E

Table A12.2 summarises total particulate matter emission factors quoted by US EPA (2000) for the various cement manufacturing and processing operations. Where quoted, an estimate has been given for the PM_{10} and $PM_{2.5}$ emissions, after taking account of the abatement equipment employed.

The US EPA data gives around 60 g of total particulate matter per tonne of clinker from crushing, screening and handling of raw materials but no PM_{10} and $PM_{2.5}$ data is given. Between 150 and 500 g of total particulate matter per tonne of clinker could be emitted from wet kiln and cooler operations, whilst between 70 and 500 g/t (clinker) could be released for the various dry kiln and clinker cooler operations. The lower emission factor assumes that the best available technique has been employed for each process stage whilst the second assumes that the least effective control technology has been employed.

Table A12.2 Cement processing and manufacture particulate emission factors (US EPA, 2000)

Process	Abatement	Emission factors g/t (clinker)			Quality rating*
		Total	PM_{10}	$PM_{2.5}$	
Raw material grinding	Fabric filters	34			E
Clinker grinding	Fabric filters	24			E
Limestone crushing	Fabric filters	1			E
Wet process kiln	Uncontrolled	65,000	16,000		D
Wet process kiln	ESP	380	330	240	D
Wet process kiln	Fabric filter	230			E
Wet process kiln	Multicyclone + ESP	100			E
Dry process kiln	ESP	500	420	220	D
Dry process kiln	Fabric filter	100	84		D
Preheater kiln	ESP	130			D
Preheater kiln	Fabric filter	130			C
Preheater/precalciner kiln	ESP	24			D
Preheater/precalciner kiln	Fabric filter	100			D
Clinker cooler	ESP	48			D
Clinker cooler	Fabric filter	68			D
Clinker cooler	Gravel bed filter	110	84	44	D

The Pollution Inventory gives emission estimates for most UK cement works and, when combined with estimates of production, these yield the following emission factors shown in Table A12.3

Table A12.3 Emissions and emission factors for UK cement processes based on Pollution Inventory data

Year	Total particulate emission (t)	PM_{10} emission (t)	Clinker production[a] (Mt)	TPM emission factor (g/t clinker)	PM_{10} emission factor (g/t clinker)
1994	3303	-	11.1	298	-
1995	2699	-	11.1	243	-
1996	2370	-	8.63	274	-
1997	2058	-	8.17	252	-
1998	2950	2205	9.35 (TPM) 9.39 (PM_{10})	315	235
1999	4223	2708	10.46 (TPM) 11.04 (PM_{10})	404	245

a - reporting sites only, thus different figures are given for 1998 and 1999, since data were reported for different sets of sites.

PM_{10} emission factors can be calculated for the years 1994 – 1997 by assuming that the ratio of TPM to PM_{10} at each site is the same in these years as in 1998 or 1999 when both TPM and PM_{10} are reported. This gives the following PM_{10} emission factors:

1994	242 g/t clinker
1995	179 g/t clinker
1996	222 g/t clinker
1997	207 g/t clinker

The reason why the emission factors are highest in 1999, when it might be expected that improvements in emission control should have led to smaller emission factors, is not known. The most likely explanation is that the emissions data for 1999 is more accurate and complete than the emission estimates for earlier years.

HEAVY METAL EMISSION FACTORS

The emission factors for heavy metals are determined by the composition of the raw material and the type of fuels used for firing. An illustration of the range of emission factors to be expected is given in Table A12.4, which consists of emission factors for 5 plants, taken from EMEP/CORINAIR (1999). The data in this table dates from 1992 or before and no information is given on the abatement level for these emission levels.

Table A12.4 Emission factors for heavy metals from cement production in g/tonne cement (EMEP/CORINAIR, 1999)

Metal	Coal & oil fired	Proportion of waste oil	Fuel unknown	Fuel unknown	Fuel unknown
Arsenic	-	-	-	0.012	-
Cadmium	-	-	0.04	0.008	< 0.001
Chromium	0.006-0.02	0.02-0.3	-	0.105	-
Lead	0.006	0.012-0.2	1.1	0.216	<0.033
Mercury	-	-	-	0.275	-
Nickel	-	-	-	0.111	-
Selenium	-	-	-	0.002	-
Zinc	-	-	-	0.293	0.003-0.47

The EIPPCB (2000c) gives a range of heavy metal emission factors from European cement kilns. This is presumably after abatement as installed at European plants. No data are given for other stages of cement production but it is not clear whether these factors apply to stack emissions only or both stack and fugitive emissions. The data are presented in Table A12.5.

Table A12.5 Emission factor ranges for European cement kilns (EIPPCB, 2000c)

Metal	kg/tonne clinker
Hg, Cd, Tl	0.01-0.3 (mainly Hg)
As, Co, Ni, Se, Te	0.001-0.1
Sb, Pb, Cr, Cu, Mn, V, Sn, Zn	0.005-0.3

The Pollution Inventory also includes data on heavy metal emissions, from which emission factors can be derived in a similar fashion to those presented in Table A12.3 for particulate matter. The heavy metal factors are shown in Table A12.6.

Table A12.6 Heavy metal emission factors for UK cement processes based on Pollution Inventory data (g/t of cement produced)

Metal	1998	1999
Arsenic	0.141	0.0697
Cadmium	0.0114	
Chromium	0.0745	0.0511
Copper	0.0312	0.121
Mercury	0.0353	0.0202
Manganese		0.277
Nickel	0.119	0.0406
Lead	0.268	0.380
Selenium	0.0297	0.0127
Vanadium	0.190	0.0761
Zinc	0.194	0.286

Surprisingly, the emission factors calculated for 1998 are often lower than those calculated from 1999 data. This may be due to less accurate or complete reporting in 1998, and thus it might be prudent to disregard the 1998 figures.

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from various cement production processes. These are reproduced in Table A12.6. The profiles for controlled dry process kilns and controlled clinker coolers seem most appropriate for the UK and suggest that particulate matter emissions comprises about 80% PM_{10}.

Table A12.6 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size
Kilns, wet process	Uncontrolled	2.5 5 10 15 20	7 20 24 35 57 100
Kilns, wet process	ESP	2.5 5 10 15 20	64 83 58 91 98 100

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size
Kilns, dry process	Uncontrolled	2.5	18
		5	n.d.
		10	42
		15	44
		20	n.d.
			100
Kilns, dry process	Fabric filter	2.5	45
		5	77
		10	84
		15	89
		20	100
			100
Clinker coolers	Uncontrolled	2.5	0.54
		5	1.5
		10	8.6
		15	21
		20	34
			100
Clinker coolers	Gravel bed filter	2.5	40
		5	64
		10	76
		15	84
		20	89
			100

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI uses PM_{10} emission factors based on Pollution Inventory data These emission factors are in reasonable agreement with the EMEP/CORINAIR figure and close to the middle of the range given by the US EPA. They are however somewhat above the middle of the range given by the EIPPCB (although, as already noted, the EIPPCB figures only include emissions from the kiln). Overall, the NAEI emission factors do not appear to be unreasonable apart from the fact that the emission factor for 1999 is significantly higher than the figures for earlier years when the opposite would be expected. It is recommended that the 1999 emission factor be used for all years, unless it can be confirmed that the entire series of estimates in the Pollution Inventory are all consistently accurate.

Heavy metal emissions

As with particulate matter emission factors, it is recommended that those factors developed from Pollution Inventory data are retained in the NAEI, although with the same proviso for factors for the period before 1999 as mentioned above for particulate matter.

Appendix 13: Lime production

SNAP CODE: 030312

SOURCE ACTIVITY TITLE: Lime production

NACE CODE: 26.52

PARCOM-ATMOS CODE: N/A

ACTIVITIES INCLUDED

The production of lime (calcium oxide) from limestone.

GENERAL PROCESS DESCRIPTION

The lime making process involves heating limestone (calcium carbonate, $CaCO_3$) to temperatures between 900 and 1200°C. Carbon dioxide is driven off leaving lime (calcium oxide, CaO), often referred to as 'burnt lime':

$CaCO_3 \rightarrow CaO + CO_2$

Limestone contains between 97% and 98% calcium carbonate on a dry basis. The rest includes magnesium carbonate, aluminium oxide, iron oxide and silica. However, some limestones contain as much as 35% to 45% magnesium carbonate and are classified as dolomite.

Limestone is a cheap, abundant material and is obtained by quarrying. It is used for a variety of other purposes besides lime production, mainly as a construction material. This chapter focuses on emissions from the lime burning plant.

Lime works are usually located adjacent to, or sometimes within, limestone quarries to minimise transportation costs. The rock is crushed in the quarry and delivered to the lime burning plant in the form of aggregate, sized in the range 10 mm to 50 mm for rotary kilns or 50 mm to 300 mm for vertical kilns. Emissions from the blasting and crushing are classified as part of quarry operations and not as lime production emissions.

The three most common types of kiln are the rotary, vertical shaft and moving grate. Vertical kilns, because of the larger size of charge material, lower air velocities, and less agitation emit smaller amounts of particles. However, in recent years there have been important developments in the design and use of rotary kilns.

Crushed or ground burnt lime can be 'slaked' by reacting it with water, producing mainly calcium hydroxide ($Ca(OH)_2$).

SOURCES OF PARTICULATE EMISSIONS

The kiln is the most important source of particle emissions, followed by the hydrator. Fugitive emissions can occur from almost any part of the process.

SOURCES OF HEAVY METAL EMISSIONS

The sources of metal emissions will predominantly be as those described for particulate emissions.

ABATEMENT MEASURES AVAILABLE

Modern lime works are equipped with electrostatic precipitators that remove at least 98% of the particulate matter from exhaust gases. Other control devices are also used including multiple cyclones, wet scrubbers, and baghouses.

UK SITUATION

UK plants

There are nine lime-producing plants in the UK, with a total of 26 lime kilns (excluding captive lime kilns). Of these, eight are rotary, seven are regenerative shaft, ten are other shaft and one is “other” (EIPPCB, 2000c). Captive lime kilns are operated at nine factories involved in sugar beet processing and two sites as part of a chemical manufacturing process.

UK activity statistics

Statistics on UK lime production are given in the UK Minerals Yearbook.

Abatement measures used in UK plants

Some information on abatement systems during the early 1990s is available from IPC authorisation documents. Most UK plants used either scrubbers or ESPs for abatement of kiln emissions. Hydrators were either fitted with cyclones or were vented directly to atmosphere. Fugitive emissions are generally controlled through enclosure of dusty transfer operations and storage of dusty materials in enclosed areas. Some processes had bag filters to treat dust laden air from local extraction vents. Crushing operations are enclosed but not always fitted with arrestment devices.

No information is available on current levels of control.

PARTICULATE EMISSION FACTORS

Because of the variety and number of kiln and plant designs, emissions and abatement technologies vary widely. The EIPPCB (2000c) gives a number of emission factors for some of the different stages of lime production. These are reproduced in Table A13.1 and suggest a particulate matter emission factor of between 145 and 1035 g/tonne (lime) for ‘typical’ modern plant. Estimates for subsidiary operations such as crushing, screening, conveying, slaking, storage and discharge are not given, but it notes that fugitives from, for example, stock piles of raw materials and solid fuels "can cause problems".

Table A13.1 Lime manufacture dust emission factors (EIPPCB, 2000c)

Process	**Abatement**	**Dust emission factors g/t (lime)**
Calcining of limestone	Uncontrolled	2,000 – 20,000
Calcining of limestone	Typical abatement	100 – 800
Lime hydrating	Uncontrolled	1,600
Lime hydrating	Wet scrubbers, Bag filters	16 – 160
Lime grinding	Bag filters as part of process	30 – 75

Emission factors EMEP/CORINAIR (1999) are presented in Table A13.2. Figures are presented for all the main sources, apparently including fugitive emissions. The figures indicate that for the total factor ranges from 800 g/t for the best techniques to some 55,000 g/t for the worst controlled techniques.

Table A13.2 Lime manufacture particulate emission factors (EMEP/CORINAIR, 1999)

Process	**Abatement**	**TPM g/t (lime produced)**
Coal storage	Open piles	500
	Semi-enclosed piles	250
	Compartments	100
	Silos	100
Coal crushing and screening	Uncontrolled	180
	Fabric filter	2
Coal grinding	(Semi) direct fired system	0
	Indirect fired system	n.d.
	Uncontrolled	10,000
	Fabric filter	100
Raw material storage	–	160
Crushing and screening	Uncontrolled	1,500
	Fabric filter	< 1
Crushed material storage	Open piles	1,000
	Semi-enclosed piles	500
	Compartments	200
	Silos	200
Raw material conveying	Uncontrolled	1,200
	Fabric filter	10

Process	**Abatement**	**TPM g/t (lime produced)**
Raw material calcining:		
Vertical shaft kiln	Uncontrolled	3,000
	Cyclone	1,000
	Multicyclones	750
Vertical double inclined kilns	Uncontrolled	10,500
	Cyclone	3,600
	Multicyclones	2,600
Parallel flow/counterflow regenerative kilns	Uncontrolled	8,000
	Cyclone	2,800
	Multicyclones	2,000
Annular kilns	Uncontrolled	12,000
	Cyclone	4,200
	Multicyclones	3,000
Rotary short kiln/air suspension preheater	Uncontrolled	40,000
	Cyclone	14,000
	Multicyclones	9,000
	ESP	600
	Fabric filter	200
Rotary long kiln	Uncontrolled	140,000
	Cyclone	49,000
	Multicyclones	35,000
	ESP	2,000
	Fabric filter	400
Calcimatic kiln	Uncontrolled	25,000
	Cyclone	8,700
	Multicyclones	6,200
Lime cooling	Grate cooler	
	Uncontrolled	20,000
	Cyclone	4,000
	Multicyclones	2,000
	Fabric filters	100
	Planetary, rotary, or vertical shaft coolers	0
Lime packaging/shipping	–	120
Lime hydration	Uncontrolled	35,000
	Scrubber	40

The US EPA (2000) offers the emission factors reproduced in Table A13.3. No figures are given for controlled product cooling or controlled product transfer and conveying; excluding these processes, totals range from about 400 g/t for the best technologies to over 60,000 g/t for the worst control technologies.

Table A13.3 Lime production particulate emission factors (US EPA, 2000)

Process	Abatement	Emission factors g/t (ore processed)		Quality rating
		Total	PM10	
Coal-fired rotary kiln	Uncontrolled	180,000	22,000	D
Coal-fired rotary kiln	Cyclone	60,000		D
Coal-fired rotary kiln	Fabric filter	140	77	D
Coal-fired rotary kiln	ESP	4,300	2,200	D
Coal-fired rotary kiln	Venturi scrubber	720		D
Gas-fired rotary kiln	ESP	86		E
Gas-fired rotary kiln	Gravel bed filter	510		E
Coal- and gas-fired rotary kiln	Uncontrolled	40,000		E
Coal- and gas-fired rotary kiln	Venturi scrubber	440		D
Coal- and coke-fired rotary kiln	Venturi scrubber	830		D
Coal-fired rotary preheater kiln	Multiclone	42,000		E
Coal-fired rotary preheater kiln	Gravel bed filter	590		E
Coal-fired rotary preheater kiln	Multiclone, water spray and fabric filter	560		E
Gas-fired calcimatic kiln	Uncontrolled	48,000		E
Gas-fired parallel-flow regenerative kiln	Fabric filter	51		D
Atmospheric hydrator	Wet scrubber	33		D
Product cooler	Uncontrolled	3,400		E
Primary crusher	Uncontrolled	8.3*		E
Scalping screen and hammer mill (secondary crusher)	Uncontrolled	310*		E
Primary crusher	Fabric filter	0.21*		D
Primary screen	Fabric filter	3*		D
Crushed material conveyor transfer	Fabric filter	0.044*		D
Secondary and tertiary screen	Fabric filter	0.065*		D
Product transfer and conveying	Uncontrolled	1,100*		E
Product loading	Enclosed truck	310*		D
Product loading	Open truck	750*		D

Units: g/t (material processed/handled)

The Pollution Inventory does not give a complete picture of particulate matter emissions from lime production since emissions from only a few processes are reported (only 5 processes reported emissions of PM_{10} in 1999).

HEAVY METAL EMISSION FACTORS

The BREF Note for cement and lime kilns states that there are little data available for metal emissions from lime kilns and further states that due to the high purity of the raw materials generally used in these kilns, metal emissions will be low. There are no heavy metal emission factor data given for this sector by EIPPCB (2000c), by EMEP/CORINAIR (1999) or by US EPA (2000).

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from various lime production processes. These are reproduced in Table A13.4

Table A13.4. Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Quality rating
Rotary kiln	Uncontrolled	2.5 5 10 15	1.4 2.9 12 31 100	–
Rotary kiln	Multicyclone	2.5 5 10 15 20	6.1 6.8 16 23 31 100	–
Rotary kiln	ESP	2.5 10 15	14 50 62 100	–
Rotary kiln	Fabric filter	2.5 10 15	27 55 73 100	–

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI uses a particulate matter emission factor of 611 g/t (lime) and a PM_{10} emission factor of 336 g/t (lime). This is based on the means of the ranges given by EIPPCB, plus the emission factor for packaging from EMEP/CORINAIR (1999) as follows:

Calcining of limestone	450 g/t	Applies to 100% of production
Slaking of lime	88 g/t	Applies to 50% of production (estimated)
Grinding of lime	52.5 g/t	Applies to 100% of production
Packaging	120 g/t	Applies to 54% of production (estimated)

This gives an overall factor of 611 g/t (lime produced) which is converted to a PM_{10} emission factor by assuming that 55% of the particulate matter is sub 10 micron (based on the US EPA profile for rotary kiln emissions controlled by bag filters – see Table A13.4).

This NAEI factor is largely based on EIPPCB data for ‘typical’ modern plant, and is assumed to be reasonable. Ideally, more information is needed on the current level of abatement in UK lime processes so that a better judgement can be made as to whether UK plants are ‘typical’. It is recommended that further investigation be carried out to determine whether the EIPPCB factors are applicable, and also to review forthcoming Pollution Inventory data for 2000, to determine whether sufficient information is available to develop a UK specific emission factor. In the meantime, the existing NAEI factor is probably the best available.

The development of an emission factor to represent emissions prior to use of modern abatement systems should also be considered since the available data shows that lime processes can emit considerable quantities of dust if not controlled effectively.

Heavy metal emissions

Currently, the NAEI does not include any emission estimates for heavy metal emissions from the lime production process (although it does include emissions of metals from fuel combustion by lime producers). This is not unreasonable, since lime processes are not believed to be significant sources of metal emissions, and no emission factors or emission estimates have been found.

Appendix A14: Glass

SNAP CODE: **030314**
030315
030316
030317
030318
040613

SOURCE ACTIVITY TITLE: **Glass production**

NACE CODE: **26000**

PARCOM-ATMOS CODE: **2.8.2**

ACTIVITIES INCLUDED

This chapter includes the manufacture of all products made from glass including glass wool and glass frit.

GENERAL PROCESS DESCRIPTION

The term glass includes materials covering a wide range of compositions, but a large proportion of glass products use soda-lime glasses. Soda-lime glasses are produced by melting silicon oxide, sodium oxide and calcium oxide, together with small quantities of other additives. The main raw materials are sand, soda ash and limestone. Other types of glass include lead glass, where lead oxide replaces much of the calcium oxide used in soda-lime glass. A large number of metals are used to impart colour to glass including chrome, copper, manganese, iron, cobalt, nickel, vanadium, titanium, neodymium, selenium and praseodymium. As an alternative to the use of virgin raw materials, recycled glass (cullet) can be used and is, in fact, an important source of glass.

The main categories of glass products are:
- container glass
- flat glass
- domestic glass
- glass fibre
- special glasses
- mineral wool
- ceramic fibre
- frits.

Appendix A14: Glass

In Europe, container glass is the most significant tonnage use of glass, producing bottles, jars and other containers for the food and drink industry. Soda-lime glasses are used. The glasses are melted in large furnaces fired with fossil fuels or (more rarely) electricity and the containers are then formed in automatic machines.

Flat glass is the second most important tonnage use of glass. Flat glasses are used in construction and in the automotive industry. As with containers, soda-lime glasses are used. The bulk of flat glass is produced using the float glass process.

Other product types are produced in much smaller quantities. Glass fibres are produced mainly for use as reinforcement in various building materials. Domestic glass products include tableware and cookware and covers manufacture of lead glass items. Manufacturing processes range from fully automated processes to manufacture by hand. Special glasses cover a wide range of products including television screens, light bulbs, laboratory glassware and high temperature cookware. Glass wool and mineral wool are used for their insulating and fire retardant properties and are supplied mainly to the construction industry. Ceramic fibres and frits are produced in relatively small quantities. Ceramic fibres are used as high performance insulating materials and frits are used in the manufacture of enamels and glazes.

Although there are differences between the processes used in each of the eight sectors of the glass industry, there are a number of common steps, for example raw material handling and melting.

Raw materials are generally inorganic and most are powders or granules. Depending upon the usage of materials they may be held in storage silos or supplied in drums or other containers. A wide range of materials handling equipment is in use in the glass industry including both open and completely closed systems.

Melting occurs in three stages: an initial stage where the raw materials melt and react; a fining and mixing stage where the molten glass is degassed and mixed; and conditioning where the glass is cooled and the remaining gases in the melt allowed to adsorb or be released.

Melting furnaces may be fired by gas, oil, or electricity. Various designs of furnace are in use, ranging from large continuous furnaces with regenerative heat recovery systems to small pot furnaces used for melting batched materials.

Following melting, processing steps depend upon the type of glass product manufactured. Containers are manufactured in a two stage forming process utilising blowing of and in some cases the application of pressure to molten glass held in moulds.

Flat glass is produced by one of two methods, the float glass process or rolling. The float glass process involves pouring molten glass onto molten tin. The glass forms a thin layer on top of the tin with parallel upper and lower surfaces. The glass cools as it travels along the bath and at the exit to the bath it is further cooled before cutting. As the name suggests, rolling of flat glass consists of pressing molten glass between steel rollers that can be smooth or patterned. The rollers are water-cooled thus cooling the glass as well as forming the glass sheet.

Glass fibres are formed by passing molten glass through a perforated steel plate. The strands of glass so formed are attenuated using a winding mechanism into continuous fibres. These are cooled, coated with a water-based binder and then formed into various products. Processing steps can include drying and chopping.

Domestic glass products can be manufactured using automatic or manual processes. Automatic processes are similar to those used in the container glass industry although additional methods such as pressing in moulds (without blowing) and spinning of products in moulds can be used. Items can be decorated using cutting.

Special glass products are formed using a wide variety of processes similar to those used in other sectors such as blowing, pressing, floating and rolling. Glass wool is prepared by pouring molten glass through an orifice and then spinning the glass to produce fibres. The fibres are coated with a resin, formed into masses, dried in an oven, and then cut. Ceramic fibres are formed in a similar fashion.

Frits are formed by melting special coloured, or opaque, glasses and then quenching the glass with water so that the glass shatters as it cools rapidly. The frit is then ground using ball mills. The grinding may be carried out dry.

SOURCES OF PARTICULATE EMISSIONS

Many of the raw materials for the glass industry are dusty and their handling is a potential source of particulate emissions.

During melting, large quantities of gases are evolved and these can contain particulate matter. The main source of this material is the volatilisation and subsequent condensation of volatile batch materials. Fine particles in the raw materials can also be carried out of the melting furnace. Some particulate matter will also arise from the combustion of fossil fuels.

Downstream of melting there are various sources of particulate matter depending upon the sector. Coating processes can give rise to particulate matter emissions but, as a result of the low tonnage used, are unlikely to be significant. Emissions from the molten tin bath used in the float glass process are also low due to the low vapour pressure of the tin at the temperatures employed and the use of inert atmospheres to prevent oxidation of the tin. Cutting and grinding of domestic glass and special glass products can give rise to dusts but the quantities are small. The forming, coating and curing of glass wool will also give rise to emissions of particulate matter

SOURCES OF HEAVY METAL EMISSIONS

The most important source of atmospheric emissions is the hot furnace. Heavy metals within the raw materials or the fuel partly vaporise in the hot furnace. The heavy metals which are emitted to air are primarily arsenic, cadmium, chromium, lead, tin, and selenium. If fuel oil is used in the furnace then nickel and vanadium may also be found. The majority of heavy metals emitted from the process will be closely associated with emissions of particulate matter.

ABATEMENT MEASURES AVAILABLE

Most large glass works have filtration systems fitted to silos and batch mixing plant. Melting furnaces may or may not be fitted with dust arrestment. Systems employed include electrostatic precipitators and bag filters, while wet scrubbing techniques are not generally favoured. Primary measures can help to reduce particulate matter emissions. Examples include:

- ensuring sufficient moisture content in the feed materials to suppress carry over of fine material;
- modifying the feed materials, for example lowering the sodium chloride content of soda ash used in the batch can reduce emissions;
- lowering the surface temperature of the melt by a variety of means including use of cullet, use of electric heating, or improvements to the thermal efficiency of the furnace;
- modifying the burner position;
- using gas as fuel rather than oil, although this can be offset by increased emissions of other pollutants.

UK SITUATION

UK plants

Estimates of UK plant numbers are given in Table A14.1

Table A14.1 UK glass making plants [a] (EIPPCB, 2000d)

Sector	Number of plants	UK share of EU market (%)	Annual EU production, Mt	Calculated annual UK production, Mt
Container	14	11	17.3	1.9
Flat	3	10	6.9	0.69
Fibre	2	17 [b]	0.475	0.081
Domestic	16	11	1	0.11
Special	3 [c]	19 [c]	1.46	0.28
Glass wool	6	10	2	0.20
Ceramic fibres	2	33 [b]	0.042	0.014
Frits	4	8 [b]	1-1.25	0.090

[a] Glass wool figures for 1996, others for 1997.
[b] Based on proportion of EU processes
[c] Cathode ray tubes and lighting glass only

Most UK processes are regulated by local authorities under the Local Authority Air Pollution Control regime. These will include those plants manufacturing container, flat, domestic and special glasses. The manufacture of glass and mineral wool, frits and continuous filament glass fibres are regulated under Integrated Pollution Control by the Environment Agency and the Scottish Environment Protection Agency.

UK activity statistics

The UK production of glass is not given in either the UK Abstract of Statistics or the British Minerals Yearbook. The EIPPCB (2000d) gives UK percentage share of the EU market and the annual EU production, from which the UK production can be calculated (see Table A14.1 above). The total UK production is estimated to have been 3.37 Mtonnes in 1997.

The NAEI includes estimates of annual production of glass using data from a variety of sources, including industry. The figure for 1997, given in the current version of the NAEI, is 2.94 Mtonnes, which is 0.43 Mtonnes less than the figure calculated in Table A14.1. The difference may be due to certain types of glass, such as domestic glass, glass & mineral wool, and frits being excluded from the current NAEI estimates. This should be checked and the more accurate figure used in the NAEI in future.

Abatement measures used in UK plants

No information is available on the control techniques used in the glass industry in the UK. However, the level of control used by the industry are likely to be based on the guidance given in Secretary of State's Guidance (PG 3/3 and PG 3/4). The particulate matter emission factors given for glass melting furnaces are as follows:

All glass (except lead glass, glass frit, and enamel frit)
- by October 1996 250 mg/m^3 (where mass emission greater than 0.5 kg/hr)
- by October 2001 100 mg/m^3 (where mass emission greater than 0.5 kg/hr)

Lead glass, glass frit, and enamel frit
- by October 2001 100 mg/m^3 (where mass emission greater than 0.5 kg/hr)

The emission factor of 100 mg/m^3 would possibly mean that abatement plant were needed, for example electrostatic precipitators or bag filters. The interim standard could probably be achieved by most or all plants using primary measures.

PARTICULATE EMISSION FACTORS

Emission factors for glass manufacturing are shown in Tables 14.2, Table A14.3 and Table A14.4, compiled from US EPA (2000), EMEP/CORINAIR (1999) and EIPPCB (2000d) respectively. The US EPA factors tend to be somewhat higher than the EMEP/CORINAIR and EIPPCB ones, however these date from the mid 1980s and often refer to uncontrolled melting operations.

Table A14.2 Emission factors for particulate matter emissions from the glass industry (US EPA, 2000), expressed in g/t (glass product).

Process	**Abatement**	**Emission factor**	**Data Quality**
Raw materials handling:			
Glass fibre/Glass wool	Uncontrolled	1900	B
Container, flat & domestic	-	Neg	B
Melting:			
Glass wool (electric furnace)	Uncontrolled	250	B
Glass wool (regenerative gas)	Uncontrolled	11000	B
Glass wool (recuperative gas)	Uncontrolled	14000	B
Glass fibre (regenerative gas)	Uncontrolled	1000	B
Glass fibre (recuperative gas)	Uncontrolled	8000	B
Container glass	Uncontrolled	700	B
Container glass	Low-energy scrubber	400	B
Container glass	Venturi	<100	B
Container glass	Baghouse	Neg	B
Container glass	ESP	Neg	B
Flat glass	Uncontrolled	1,000	B
Flat glass	Low-energy scrubber	500	B
Flat glass	Venturi	Neg	B
Flat glass	Baghouse	Neg	B
Flat glass	ESP	Neg	B
Pressed and blown	Uncontrolled	8,400	B
Pressed and blown	Low-energy scrubber	4,200	B
Pressed and blown	Venturi	500	B
Pressed and blown	Baghouse	100	B
Pressed and blown	ESP	100	B
Frits	Uncontrolled	16000	E
Frits	Venturi scrubber	1800	E
Frits	Fabric filter	20	E
Forming and finishing:			
Container	–	Neg	B
Flat	–	Neg	B
Pressed and blown	–	Neg	B
Glass wool	Uncontrolled	1000	B
Glass fibre	Uncontrolled	500	B
Curing			
Glass wool	Uncontrolled	3000	B
Glass fibre	Uncontrolled	600	B

Table A14.3 Emission factors for particulate matter emissions from the glass industry (EMEP/CROINAIR, 1999), expressed in g/t (glass product).

Process	**Abatement**	**Emission factor**	
Handling:			
Container glass	–	90[a]	
Flat glass	–	150	
Glass wool	–	90	
Melting:			
Container glass	–	300	
Flat glass	–	370	
Glass wool	Controlled	40	
All processes:			
General	–	400	

a - mean of range given

Table A14.4 Emission factors for particulate matter emissions from the glass industry (EIPPCB, 2000d), expressed in g/t (glass melted).

Process	**Abatement**	**Emission factor**
Melting		
Container glass	Uncontrolled	400[a]
Container glass	Secondary abatement	26[a]
Flat glass	Uncontrolled	400[a]
Flat glass	Secondary abatement	60[a]
Glass fibre	Uncontrolled	1,750[a]
Glass fibre	Secondary abatement	140[a]
Domestic glass (soda-lime)	Typical furnace	400
Domestic glass (lead)	Typical furnace	20
Special glass		84[b]
Glass wool	Typical furnace	270[c]
Frits	Typical furnace	4,550[a]
Forming and finishing:		
Forming, fiberising and curing of glass wool	Typical operation	1,400[a]
Cooling & finishing	Typical operation	220[a]
All processes:		
Glass fibre	Uncontrolled	2,700[a]
Glass fibre	Secondary abatement	130[a]

a Mean of range given.
b Mean of three case studies given in the reference
c Mean of the middle 80% of emissions for glass wool melting operations

To convert g/t (glass melted) to g/t (product), the EIPPCB (2000d) gives the “pack to melt” ratio for container glass as 91 percent, and for domestic soda-lime glass as 85 percent and domestic lead crystal glass as 75 percent. Conversion factors for other types of glass are not given.

The available factors show a wide variation. This is likely to be due partly to differences in the level of control, for example the highest factors are for uncontrolled melting and date from the mid 1980s. There is also a variation depending upon the type of glass product being made with generally higher emission factors for glass fibre, glass wool and frits.

HEAVY METAL EMISSION FACTORS

Emission factors for glass manufacturing are shown in Table A14.5. These are compiled from the EIPPCB (2000d). These data relate to 1996-1997 emission figures and generally do not provide disaggregated emission factors for individual metal compounds.

Table A14.5 Heavy metal (V, Ni, Cr, Se, Pb, Co, Sb, As, Cd) emission factors for glass manufacturing (1996-1997), expressed as g/t (glass melted).

Product	**Abatement**	**Emission factor**
Container glass[a]	With or without primary measures	≤16
Container glass[a]	Secondary abatement	≤2
Flat glass[a]	Unabated	<1-15
Flat glass[a]	Secondary abatement	<1
Special Glass		
Glass Fibre [a]	Primary	<25
Glass Fibre [a]	Secondary	<25
Lead crystal [a]	unknown	10
TV funnel glass [a]	unknown	1400 (lead)
TV funnel glass [a]	unknown	140 (other metals)
TV panel glass [a]	unknown	3300 (Sb)
Mineral wool[a,b]	unknown	0.3-6
Frits [a]	unknown	<10-200

[a] emissions from melting activities only
[b] middle 80% of emissions from all Cupola Furnaces for stone wool in the EU

The US EPA only recommend an emission factor for lead from lead glass manufacturing which is 2.5 kg/t (presumably this is per tonne of molten glass). EMEP/CORINAIR (1999) provides German emission factor data (from circa 1990) for general, lead crystal, and crystal glasses, presented in Table A14.6.

Table A14.6 Emission factors for heavy metals for glass production (EMEP/CORINAIR, 1999), expressed as g/tonne glass.

Metal	General glass[a]	lead crystal glass[a]	Crystal glass
Arsenic	0.12 (0.10-0.25)	140 (22-310)	96
Cadmium	0.15 (0.06-0.24)		
Chromium	2.4 (0.5-5)		
Copper	0.6 (0.4-1.1)		
Lead	12 (2-24)	2700 (2200-3200)	480
Mercury	0.05 (0.036-0.072)		
Nickel	1.9 (1.2-2.6)		
Selenium	18 (2.4-24)		
Zinc	11 (4.8-24)		

[a] The range is given with the mean in brackets

These German data probably date from the mid 1980s and may not therefore be particularly appropriate for current UK practice.

The Pollution Inventory provides mass emissions data for production of frits, continuous filament glass fibre and glass wool production. These data can be converted to emission factors using estimates of UK production taken from Table A14.1. The resulting emission factors are shown in Table A14.7.

Table A14.7 Emission factors for heavy metals from glass processes based on Pollution Inventory data for 1999, expressed as g/tonne (glass).

Metal	Continuous filament glass fibre	Glass wool	Frits
Arsenic	0.247	0.0100	
Cadmium		0.0131	0.0167
Chromium	38.1	0.105	
Copper		0.0500	
Lead		0.0500	0.178
Mercury		0.0100	
Nickel		0.0500	
Selenium		0.0180	
Zinc	22.2	0.515	

The data in the Pollution Inventory is very patchy and many processes do not report emissions of all heavy metals, since these are below *de minimis* limits. If emission factors are recalculated assuming that emissions in these cases are 50% of the *de minimis* limit, then the emission factors shown in Table A14.8 are obtained.

Table A14.8 Revised emission factors for heavy metals from glass processes based on Pollution Inventory data for 1999, including sites below the reporting threshold, expressed as g/tonne (glass).

Metal	Continuous filament glass fibre	Glass wool	Frits
Arsenic	0.256	0.0180	0.00556
Cadmium		0.0141	0.0172
Chromium	38.1	0.159	0.0556
Copper	0.0617	0.127	0.111
Lead		0.127	0.233
Manganese		1.00	1.11
Mercury		0.0113	
Nickel		0.102	0.111
Selenium		0.0180	0.00556
Vanadium		1.00	0.556
Zinc	22.2	1.266	0.556

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from uncontrolled and controlled melting furnaces in glass manufacturing. These are reproduced in Table A14.9.

Table A14.9 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size	Cumulative mass EF (g/t)	Quality rating
Melting furnace	Uncontrolled	2.5	91	640	E
		6	93	650	
		10	95	660	
			100	700	
Melting furnace	ESP	2.5	53	Negligible	E
		6	66	Negligible	
		10	75	Negligible	
			100	Negligible	

RECOMMENDED EMISSION FACTORS

Particulate emissions

The above data show that emission factors vary considerably for different types of glass and levels of abatement. The abatement techniques used in the UK are uncertain but the application of secondary measures is believed not to be universal. However, more information is needed on the level of emissions control by the glass industry. For the moment, a figure intermediate between uncontrolled and controlled values would be appropriate. With these points in mind, an overall emission factor for the UK industry may be taken to be about 400 g/t (product).

The NAEI currently uses an emission factor based on this type of approach. Emission factors are taken from the EIPPPCB report and a mean of uncontrolled and controlled factors is used for each sector of the glass industry. This approach gives a production-weighted particulate matter emission factor of 415 g/t (glass). This is converted to PM_{10} by assuming that 85% of emissions are less than 10 microns (from mean of the US EPA profiles for controlled and uncontrolled melting furnaces – see above) giving a PM_{10} factor of 353 g/t (glass). Emission factors for uncontrolled processes are also taken from the EIPPCB to give emission factors for uncontrolled processes for use in the NAEI for years prior to 1993 (which is taken as the first year of improvements to meet the requirements of the Environmental Protection Act) These factors are 658 g/t (glass) for particulate matter and 559 g/t (glass) for PM_{10}.

The existing NAEI factors are probably reasonable given a lack of detailed information on the UK glass industry. It is recommended, however, that further attention be given to obtaining better data on the UK glass industry, particularly on the level of abatement in place and the level of emissions from processes, so that the most appropriate emission factors can be selected from those available in the literature. In particular, those suggested by EIPPCB (2000d) appear to be most compelling, seemingly being based on data from between 6 and 11 examples of each type of glass production process. In the case of glass fibre, frit, and glass wool producers, data reported to the Pollution Inventory may be suitable for use in the NAEI.

Heavy metal emissions

The NAEI uses German data to generate emission factors for heavy metals, which is also the basis of the emission factors given by EMEP/CORINAIR (1999). The German data relate to emissions of metals from the German glass industry in the mid 1980s and are probably not very relevant for modern UK practice. The EMEP/CORINAIR factors could, however, be used in the NAEI for historical emissions at least (say to the early 1990s). For later years it is recommended that emissions from glass fibre, frit, and glass wool producers are based on the revised Pollution Inventory data given in Table A14.8, and that for other glass processes a revised set of emission factors, based on those given in EMEP/CORINAIR (1999) be used, but assuming modern levels of control. These new factors could be extrapolated from the emission factors for uncontrolled and controlled particulate matter emissions given by EIPPCB (2000). In the longer term, more investigation is needed to generate accurate sector-specific emission factors for heavy metals.

Appendix A15: Bricks and ceramics

SNAP CODE:	**030319** **030320**
SOURCE ACTIVITY TITLE:	**Bricks and tiles** **Fine ceramics**
NACE CODE:	**26400**
PARCOM-ATMOS CODE:	**2.8.4**

ACTIVITIES INCLUDED

This chapter covers the manufacture of bricks, tiles and other ceramic goods manufactured by the firing of clays and other minerals.

GENERAL PROCESS DESCRIPTION

The manufacture of bricks and related products involves the preparation of the raw materials, followed by the forming, cutting or shaping, drying or curing, and firing of the final product.

To start the forming process, the raw materials (clay, water and additives) are mixed, and the products are formed into the shape of the final product. The products are then heated.

Three stages of heating are almost invariably involved.
- The initial drying period, in which appreciable volumes of hot air must be used in order to remove moisture until the ware is completely dry.
- The oxidation preheating period, in which chemically combined water is removed and oxidation of any carbonaceous matter in the product is completed.
- The finishing period, during which the required final temperature of 950 – 1100 ºC is attained.

Two types of brick-making can be identified in the UK – Fletton and non-Fletton. Fletton bricks are manufactured from the Lower Oxford Clay by one UK company. Lower Oxford Clay contains high levels of carbonaceous material which is burnt when the bricks are fired, thus lowering the requirement for fuel. Lower Oxford Clay also contains high levels of sulphur. Non-Fletton bricks are manufactured using other clay types such as Keuper Marl, Etruria Marl, and Weald Clay which generally do not contain significant quantities of carbonaceous material, although they may contain significant levels of other components which lead to emissions of pollutants.

SOURCES OF PARTICULATE EMISSIONS

Pollution from the brick-making industry is predominantly confined to stack emissions of kiln exhaust gases. The pollutants in the exhaust gas originate mainly from impurities within the clay, although firing with coal or heavy fuel oil will make a significant contribution to the overall emissions to atmosphere.

SOURCES OF HEAVY METAL EMISSIONS

Emissions to air of heavy metals from the brick making industry will be predominantly associated with emissions of particulate matter and occur due to the presence of heavy metals in the fuel and heavy metals in the clays.

ABATEMENT MEASURES AVAILABLE

Most flue gas cleaning systems currently in operation within the brick industry are dry absorption based processes. Two systems are employed, packed bed filters and cloth filters.

- Packed bed filter systems: the flue gas passes through a filter bed of granular limestone. Gaseous pollutants are absorbed on the filter media which also allows for dust deposition, thereby avoiding the need for a separate dust filter. The efficiency of these units is generally high, with typical particulate matter levels in the treated gas quoted as being below 50 mg/m^3.

- Cloth filter systems: lime or hydrated lime is injected into the gas stream to absorb the gaseous compounds. The resulting fluorspar and gypsum are then removed from the gas stream using cloth filters. The particulate matter removal efficiencies for such a system are of the same order as the packed bed system.

UK SITUATION

UK plants

At least thirty companies manufacture bricks and tiles in the UK. Non-Fletton brickworks are Part B processes, but the four remaining Fletton brickworks are regulated under Part A of the Environmental Protection Act 1990.

Fletton brickworks involve the firing of bricks in Hoffman kilns. Green (unfired) bricks are stacked inside a chamber in the Hoffman kiln then the kiln is closed. Hoffman kilns have multiple chambers, connected by flues, and the flow of gases between these chambers can be controlled by a series of dampers. The green bricks in a chamber are first dried using hot gases from chambers where bricks are cooling. Next, the temperature in the chamber is increased rapidly by allowing the spread of fire from another chamber which is undergoing firing. The chamber is held in a reducing atmosphere during this stage of the process, but once the temperature in the chamber has reached the required level, air is allowed into the chamber together with supplementary fuel to maintain the temperature at about 900^{o}C for 36 hours. Finally, the bricks are allowed to cool.

Clay grinding and the loading, mixing, discharge and application of facing materials are maintained under local extraction and venting systems (LEVs). The extracted air is passed through fabric filters before discharge.

Non-Fletton brickworks generally employ tunnel kilns. In these kilns, green bricks are placed on a rail car which travels through warming, firing and cooling zones of the kiln. Air is moved counter-currently through the kiln. These kilns are generally fired with gas. Some smaller, intermittent kilns are also used, mainly for the production of small numbers of specialist bricks. These are operated batchwise, and have higher energy consumption than continuous kilns since heat released as bricks cool cannot be used.

Other ceramic products are manufactured in similar ways to bricks, with the same basic stages of raw material preparation, product forming and firing. Refractories are materials that can withstand high temperatures without being deformed or destroyed. They are based on oxides of aluminium, silicon, magnesium, calcium, and chromium. The manufacture of refractories can involve the use of kilns for production of raw materials as well as for firing of refractory bricks and shapes. Rotary kilns are commonly used for raw material preparation, while continuous tunnel kilns or intermittent kilns are used for brick firing. Natural gas is likely to be the most common fuel used. Pottery products include china and earthenware tableware, sanitaryware, electrical ware, and glazed and unglazed tiles, all manufactured from clays. While electrical ware, sanitaryware and unglazed tiles are only fired once, glazed tiles and tableware may be fired many times, as various layers of decoration are applied to the product. Continuous tunnel kilns and intermittent kilns are used for product firing and natural gas is likely to be the most commonly used fuel.

UK activity statistics

UK brick production statistics are given in the Annual Abstract of Statistics. This production refers to Fletton and non-Fletton combined. 53% of the number of bricks produced in 1980 were Fletton bricks (ETSU, 1984). By 1990, about one quarter of production was at Fletton brickworks (Ove Arup, 1991), and by 1995, 20% of production was of Fletton bricks (Blythe, 1995).

Production data for ceramics, refractories, and bricks are given in the PRODCOM series of publications from the Office of National Statistics.

Abatement measures used in UK plants

No specific information on abatement measures used in UK Part B plants is available. The abatement systems at Fletton brickworks in the first half of the 1990s can be determined from IPC authorisation documents. They were fitted with bag filters on local extraction and venting systems, which captured emissions from clay grinding and the loading, mixing, discharge and application of facing materials. No abatement was fitted to the main kiln stacks.

PARTICULATE EMISSION FACTORS

EMEP/CORINAIR (1999) quotes a single dust emission factor of 50 g/t (product) for bricks, although, since this figure is based on Dutch data, it will not be applicable to the UK-specific Fletton brickworks. Dutch data is also used to generate an emission factor range for ‘fine ceramics’ of 35-80 g/t (product).

The US EPA (2000) gives emission factors for a variety of processes in brick manufacture (Table A15.1). Most UK kilns are believed to be gas-fired, but Fletton brickworks were traditionally fired with coal, and some will still use coal.

Table A15.1 Particulate emission factors from brick manufacture (US EPA, 2000)

Process Stage	**Abatement**	**Emission factors g/t (fired bricks)**			**Quality rating**
		Tot	**PM_{10}**	**$PM_{2.5}$**	
Primary crusher	Fabric filter		0.295		
Grinding and screening operations - wet material	Uncontrolled	12.5	1.15		E
Grinding and screening operations - dry material	Uncontrolled	4250	265		E
Grinding and screening operations - dry material	Fabric filter	3.1	1.6		E
Extrusion line	Fabric filter		1.8		E
Brick dryer	Uncontrolled	38.5			E
Natural gas-fired kiln	Uncontrolled	480	435		D
Coal-fired kiln	Uncontrolled	900	700	435	C
Coal-fired kiln	Fabric filter	315			E
Sawdust-fired kiln	Uncontrolled	465	425	375	D
Sawdust-fired kiln and sawdust dryer	Uncontrolled	700	155		E
Natural gas-fired kiln firing structural clay tile	Uncontrolled	500			E

These US EPA factors are much higher than that proposed by EMEP/CORINAIR, although many of the factors are for uncontrolled processes. No information has been found on the crushing and grinding operations in the UK, but it is assumed that the factor for uncontrolled wet grinding is most appropriate. US EPA does not give an estimate for controlled emissions from gas-fired kilns, but, given that uncontrolled emissions are 480 g/t (product), then controlled emissions might be estimated to be in the range 50 – 200 g/t for total particulate matter. Taken with the factors for grinding and brick drying, a tentative emission factor of 175 g/t (brick) is suggested for controlled non-Fletton brickworks. The factors for coal fired kilns might be appropriate for Fletton brickworks, suggesting an overall emission factor of 912.5 g/t (bricks) for uncontrolled and 327.5 g/t (bricks) for controlled works (Fletton bricks are not dried before firing).

The Pollution Inventory gives emissions data for Fletton brickworks, shown in Table A15.2.

Table A15.2 Emissions of particulate matter reported for Fletton brickworks in the Pollution Inventory, expressed in tonnes

Pollutant	1993	1994	1995	1996	1997	1998	1999
Particulate matter	371	723	621	592	683	532	406
PM_{10}						399	304

These data can be converted to emission factors, based on our own estimates of Fletton brick production. These are shown in Table A15.3.

Table A15.3 Emission factors for particulate matter from Fletton brickworks based on the Pollution Inventory, expressed in g/t (bricks)

Pollutant	1993	1994	1995	1996	1997	1998	1999
Particulate matter	332	573	494	504	591	460	358
PM_{10}						345	268

The PM_{10} emissions are both equal to 75% of the particulate matter emission for the same year.

The US EPA (2000) also gives a range of emission factors for refractories manufacture (Table A15.4) and ceramics manufacture (Table A15.5).

Table A15.4 Particulate emission factors for refractory manufacture (US EPA, 2000)

Process Stage	Abatement	Emission factors g/t (fired material)		Quality rating
		Tot	PM_{10}	
Fire clay:				
Rotary dryer	Uncontrolled	33,000	8100	D
Rotary dryer	Cyclone	5600	2600	D
Rotary dryer	Cyclone & wet scrubber	52	-	D
Rotary calciner	Uncontrolled	62,000	14,000	D
Rotary calciner	Multicyclone	31,000	-	D
Rotary calciner	Multicyclone & wet scrubber	150	31[a]	D
Chromite-magnesite ore				
Rotary dryer	Uncontrolled	830	200	D
Rotary dryer	Cyclone & fabric filter	150	-	D
Tunnel kiln	Uncontrolled	410	340	D

a – this figure is not consistent with the size distribution given by US EPA and is from a different source to the particulate matter emission factor. We would expect a higher PM_{10} content than 20% for a controlled source and so this value is disregarded here.

Table A15.5 Particulate emission factors for ceramic products manufacture (US EPA, 2000)

Process Stage	Abatement	Emission factors g/t (fired material)	Quality rating
Raw material crushing & screening	Fabric filter	60	D
Dryer	Uncontrolled	1150	E
Cooler	Uncontrolled	55	E
Granulation – natural gas fired spray dryer	Fabric filter	30	E
Granulation – natural gas fired spray dryer	Venturi scrubber	95	D
Firing - gas fired kiln	Uncontrolled	245	D
Refiring – gas fired kiln	Uncontrolled	34	E
Ceramic glaze spray booth	Uncontrolled	9500	E
Ceramic glaze spray booth	Wet scrubber	900	D

The US EPA data cover a wide range, depending upon the level of control. Total particulate matter emission factors for the highest level of control are about 200 g/t (fired material) for fire clay products, and 560 g/t (fired material) for chromite-magnesite ore products (although this assumes no controls on the kiln). Figures for ceramic products would be about 335 g/t (fired material) for unglazed materials and perhaps 1270 g/t (fired material) for glazed wares, assuming that each item is re-fired once.

HEAVY METAL EMISSION FACTORS

The US EPA (2000) gives emission factors for a variety of processes in brick manufacture, shown in Table A15.6. Metals are emitted both due to the presence of metals in the fired material and due to the burning of fuel. The factors given for gas-fired brick kilns may therefore be the most reliable guide to emissions of metals from the clay alone. In any case, most UK kilns are gas fired.

Table A15.6 Heavy metal emission factors for brick kilns (US EPA, 2000)

Source	Element	g/tonne of brick produced	Factor Rating
Kiln[a]	Antimony	0.0135	D
	Cadmium	0.0075	D
	Chromium	0.0255	D
	Cobalt	0.00105	E
	Lead	0.075	D
	Nickel	0.036	D
	Selenium	0.115	D
Coal-fired kiln	Arsenic	0.065	E
	Beryllium	0.008	E
	Manganese	0.145	D
	Mercury	0.048	E

Natural gas fired kiln	Arsenic	0.0155	D
	Beryllium	0.0002	D
	Manganese	0.195	D
	Mercury	0.00375	D

[a] Coal, natural gas, or saw dust fired tunnel kiln.

Heavy metal emission factors for refractories and ceramic products are shown in Table A15.7.

Table A15.7 Heavy metal emission factors for production of refractories and ceramic products (US EPA, 2000)

Source	Element	g/tonne of material processed	Factor Rating
Chromite-magnesite based refractories			
Rotary dryer	Chromium	35	D
Rotary dryer	Hexavalent chromium	0.038	D
Rotary dryer – cyclone & fabric filter	Chromium	64	D
Rotary dryer – cyclone & fabric filter	Hexavalent chromium	0.019	D
Tunnel kiln	Chromium	130	D
Tunnel kiln	Hexavalent chromium	8.7	D
Ceramic products manufacture			
Ceramic glaze spray booth - uncontrolled	Lead	1500[a]	E

a Emission factor is in terms of g/tonne of glaze used. The emission factor is based on use of a glaze containing 24% lead.

The Pollution Inventory does not include any reported emissions of heavy metals from Fletton brickworks.

SIZE DISTRIBUTION

US EPA (2000) gives size distributions for particulate matter emitted from brick kilns, and manufacture of refractories. Selected profiles are reproduced in Table A15.8.

Table A15.8 Particle size distributions and size-specific emission factors (US EPA, 2000)

Process	Abatement technology	Particle size (µm)	Cumulative mass % < stated size
Bricks: sawdust-fired kiln	–	1 2.5 10	44 48 75
Bricks: coal-fired kiln	–	1 2.5 10	10 23 63

Process	**Abatement technology**	**Particle size (µm)**	**Cumulative mass % < stated size**
Refractories - fire clay: rotary dryers	Uncontrolled	2.5 6 10	3 10 24
Refractories - fire clay: rotary dryers	Cyclone	2.5 6 10	14 31 46
Refractories - fire clay: rotary calciners	Uncontrolled	1 2.5 6 10	3 7 17 34
Refractories - fire clay: rotary calciners	Cyclone & scrubber	1 2.5 6 10	31 46 55 69
Refractories – chromite-magnesite: rotary dryers	Uncontrolled	1 2 10	1 13 24
Refractories – chromite-magnesite: tunnel kiln	Uncontrolled	1 2 10	71 78 84

RECOMMENDED EMISSION FACTORS

Particulate emissions

Currently, the NAEI does not include separate emission estimates for bricks and ceramics.

It is recommended that emission estimates for Fletton brickworks be based on the data given in the Pollution Inventory. The factors calculated from these data are lower than the US EPA factor for uncontrolled coal fired kilns (900 g/t (brick), however these Fletton brickworks may now have some dust arrestment measures for their kiln waste gases. It should also be noted that the US EPA factor will not be for a Fletton brick process.

In the case of non-Fletton brickworks it is recommended that the NAEI adopts the US EPA particulate matter factor for uncontrolled gas fired kilns (480 g/t (brick)). A PM_{10} emission factor can be calculated by assuming that the PM_{10} component is 75% as for the UK Fletton brickworks, giving 360 g/t (brick). Further research is needed in order to determine whether particulate matter emissions are controlled at UK non-Fletton brickworks. If this is the case, then an emission factor of 175 g/t (product) for particulate matter could be used. Again, assuming a figure of 75% of particulate matter below 10 microns gives a PM_{10} factor of 131 g/t (bricks).

For refractories and ceramic goods, US EPA factors are recommended. No information is available on the level of control, but, given the very high emission factors for uncontrolled processes, it seems very likely that some controls will be in place. As a starting point it is recommended that emission factors of 200 g/t (fired material) for fire clay products, 560 g/t (fired material) for chromite-magnesite ore products, 335 g/t (fired material) for unglazed materials and 270 g/t (fired material) for glazed ceramic wares are used. Further investigation is needed to provide better UK-specific data.

In the case of refractories, PM_{10} emission factors can be generated from the particulate matter emission factors suggested above by applying appropriate US EPA size distributions. This approach gives PM_{10} emission factors of 127 g/t (fired material) for fire clay products and 413 g/t (fired material) for chromite-magnesite ore based products.

In the case of ceramics manufacture, no data are available and so a figure of 70% is suggested, since this is approximately the average for Fletton bricks and refractory materials.

In all cases it will be necessary for the NAEI to distinguish between the particulate matter generated by fuel and that generated from the bricks. This is because the NAEI will already take account of particulate matter emissions from brick and ceramics manufacture in its estimates for industrial fuel use.

Heavy metal emissions

The only emission factors available are those from the US EPA. These could be used, however it will be necessary to distinguish between metal emissions generated by fuel and those generated from the bricks. This is because the NAEI will already take account of metal emissions from brick manufacture in its estimates for industrial fuel use. It is possible that practically all of the metal emissions will be generated from fuel combustion in which case there is no need to calculate heavy metal emissions separately for this source.

Appendix 16: Chromium Chemicals

SNAP CODE: 040400

SOURCE ACTIVITY TITLE: Inorganic chemical processes

NACE CODE:

PARCOM-ATMOS CODE:

ACTIVITIES INCLUDED

The manufacture of chromium-based chemicals by a specialist chemicals manufacturer.

GENERAL PROCESS DESCRIPTION

This appendix covers a single process, located in the UK and so no general description is needed.

SOURCES OF PARTICULATE EMISSIONS

Sources of particulate matter are covered in the section on UK plants (see below)

SOURCES OF HEAVY METAL EMISSIONS

Sources of heavy metals are covered in the section on UK plants (see below)

ABATEMENT MEASURES AVAILABLE

Abatement measures are described in the section on UK plants (see below)

UK SITUATION

UK plants

The UK has one major producer of chromium based chemicals. This site may be divided into five areas:

- Primary processing, effluent treatment, & residue disposal plant: chromite ore is dried and milled, then mixed with sodium carbonate and process residues. The mixture is then calcined, oxidising insoluble trivalent chromium to soluble hexavalent sodium chromate. After quenching, the sodium chromate is separated and purified, then acidified to form sodium dichromate liquor and a sodium sulphate by-product. The sodium dichromate is evaporated to form either concentrated liquors for use elsewhere on site or for sale, or crystalline sodium dichromate for sale.

- Manufacture of chromic sulphate: chromic sulphate is produced by the reduction of sodium dichromate solution with sulphur dioxide, produced by the combustion of molten sulphur in a furnace. Excess sulphur dioxide in the outlet from the reactor is absorbed in fresh sodium dichromate and a candle filter is used to remove sulphur trioxide droplets. The chromic sulphate is then dried in a spray drier as is or used as a feedstock to produce further products.

- Manufacture of potassium and ammonium dichromate: potassium dichromate is produced by the reaction of chromic acid produced on site with potassium hydroxide solution. The potassium dichromate product is dried in a thermal Venturi drier after various treatments to remove impurities. Ammonium dichromate is also produced on the same plant by reacting chromic acid and ammonia.

- Manufacture of chromic oxides: the manufacture of chromic oxides consists of two stages, the first being the between ammonium sulphate and sodium dichromate dihydrate solution. The resulting slurry of ammonium dichromate and sodium sulphate is thermally decomposed in a rotary kiln to form chromic oxide. This product can either be quenched, filtered and dried for sale or can be further processed by calcining in a rotary kiln before quenching, filtering and drying.

- Manufacture of chromic acid: chromic acid is produced by reacting sodium dichromate with sulphuric acid. The resulting slurry is centrifuged, washed and dried in a gas fired drier. The final powder is compacted, granulated and sieved to remove fines before being packaged for sale.

Emissions of both particulate matter and chromium can occur from various stages of the processes but particularly from drying, handling and packaging of feedstocks and products.

UK activity statistics

No activity statistics are available.

Abatement measures used in UK plants

Waste gases from driers are generally fitted with wet scrubbers, while reverse-jet fabric filters are used on product handling duties and the kilns for the chromic oxide process are fitted with 2 field ESPs. Waste gases from the quenching operations on the chromic oxide process are treated using wet scrubbers.

PARTICULATE EMISSION FACTORS

The only data available are from the Pollution Inventory and are shown below in Table A16.1.

Table A16.1 Emissions of particulate matter and PM_{10} reported in the Pollution Inventory for chromium chemicals manufacture, expressed in tonnes

Year	Particulate matter	PM_{10}
1995	65.7	7.9
1996	108.0	11.6
1997	58.4	6.8
1998	53.0	6.0
1999	43.0	5.0

HEAVY METAL EMISSION FACTORS

Data are available from the Pollution Inventory and are shown below in Table A16.2

Table A16.2 Emissions of chromium reported in the Pollution Inventory for chromium chemicals manufacture.

Year	Chromium (tonnes)
1995	21.0
1996	20.5
1997	23.4
1998	25.1
1999	17.9

The bulk of emissions occur from sodium dichromate, chromic oxides and chromic sulphate plants. The following speciated data, shown in Table A16.3 has also been provided by the process operator (Darrie, 2001).

Table A16.3 Emissions of chromium by species for chromium chemicals manufacture, expressed in kg.

Year	Hexavalent	Soluble trivalent	Insoluble trivalent	Total
1988	13630	2880	19880	36390
1989	13300	2790	19490	35580
1990	10590	3190	15520	29300
1991	7780	3370	15350	26500
1992	10300	3300	15100	28700
1993	12160	4300	13300	29760
1994	8490	4890	15150	28530
1995	5261	4880	15355	25496
1996	3668	3782	12288	19738
1997	5547	2960	14994	23501
1998	4733	2265	17630	24628
1999	3454	2254	12098	17806
2000	3749	1471	15935	21155

SIZE DISTRIBUTION

The Pollution Inventory data for particulate matter and PM_{10} suggests that approximately 12% of the particulate matter emitted from the processes are PM_{10}. This doesn't seem unreasonable, given that most of the emissions will be from driers and processes involving the handling of dusty materials (for example US EPA profiles for driers for some chemical products give sub 10 micron fractions of around 10%).

RECOMMENDED EMISSION FACTORS

Particulate emissions

The NAEI currently uses data from the Pollution Inventory. These data are the only available, and their use in the NAEI should continue.

Heavy metal emissions

The NAEI currently uses data from the Pollution Inventory. We recommend that this continues, but the NAEI should also incorporate additional data provided by the process operator.

Appendix A17: Alkyl lead manufacture

SNAP CODE: 040500

SOURCE ACTIVITY TITLE: Organic chemical processes

NACE CODE:

PARCOM-ATMOS CODE:

ACTIVITIES INCLUDED

The manufacture of tetraethyl lead and tetramethyl lead.

GENERAL PROCESS DESCRIPTION

This appendix covers a single process, located in the UK and so no general description is needed.

SOURCES OF PARTICULATE EMISSIONS

Sources of particulate matter are covered in the section on UK plants (see below)

SOURCES OF HEAVY METAL EMISSIONS

Sources of heavy metals are covered in the section on UK plants (see below)

ABATEMENT MEASURES AVAILABLE

Abatement measures are described in the section on UK plants (see below)

UK SITUATION

UK plants

The UK has one plant manufacturing tetraethyl lead (TEL) and tetramethyl lead (TML). Total site capacity for organo-lead compounds is 100,000 tonnes per year. The process involves the reaction between a sodium/lead alloy (containing 10% sodium) and either methyl chloride or ethyl chloride to give the product. Aluminium chloride is also required as a catalyst for TML manufacture. The reaction takes place in a autoclave, and after completion of the reaction, distillation is used to obtain the pure product.

About one quarter of the lead present in the lead/sodium alloy is consumed in the reactions to produce TML or TEL. The remainder is left as a watery sludge which is recycled in reverberatory furnaces.

The lead recovery process involves feeding the lead sludge to a drier to remove water (sent to effluent treatment) and volatile organometallic compounds (condensed and recycled). The dried sludge is fed to a oxygen/gas heated reverberatory furnace where the lead is melted and recovered. Slag, consisting of lead salts and oxides is skimmed from the surface of the molten metal and sent off-site for recovery.

The molten lead is combined with molten sodium to form the lead/sodium alloy, which is cooled on a drum flaker to produce flakes of alloy.

Sodium is produced on site by electrolysis of molten sodium chloride using ‘Downs’ cells. The salt is first dried in a gas fired rotary drier then fed to the cells. Calcium chloride and barium chloride are also added periodically. During electrolysis, sodium is produced at the mild steel anode and chlorine is produced at the graphite cathode. A steel mesh prevents recombination of these products. The chlorine is liquefied and used in the manufacture of alkyl chlorides or sold. The sodium is filtered and stored ready for use in alloy production or sold.

UK activity statistics

The capacity of the UK plant in the early 1990s is thought to have been about 100,000 tonnes of organometallic compounds per year. However, production levels are down more than 90% since 1990 (Associated Octel, 2000) and so production is likely to be 10,000 tonnes or less.

The PRODCOM survey, produced by the Office of National Statistics, gives data on UK manufacturers’ sales of anti-knock preparations based on lead in PRQ29. The figure for 1998 was 62,800 tonnes, but no figure is available for 1999.

Abatement measures used in UK plants

Excess alkyl chlorides and organic by-products are vented from the autoclaves at the end of the reactions and recovered using condensation. Emergency vents from the autoclaves vent to containment vessels.

The material for lead recycle is contaminated with TML and TEL. This contamination is extracted and sent for recovery by absorption or carbon adsorption. The product handling streams and the TML distillation vents also pass to scrubbers. Air from the lead recovery furnaces is treated in a venturi scrubber followed by a wet ESP to remove any residual particulate.

PARTICULATE EMISSION FACTORS

Emissions of particulate matter from this process are not considered to be particularly significant, and therefore these are estimated as part of the sectors ‘chemical industry’ and ‘non-ferrous metals (other)’

HEAVY METAL EMISSION FACTORS

The Pollution Inventory contains the following data, shown in Table A17.1 for the manufacture of alkyl lead compounds.

Table A17.1 Emissions of lead from manufacture of alkyl lead compounds from the Pollution Inventory (expressed in tonnes)

Year	TEL/TML manufacture	Lead recovery
1994	91	-
1995	77.7	-
1996	66.1	-
1997	105.5	2.67
1998	59.18	25.0
1999	18.6	7.01

It is not known why the emissions from lead recovery in 1997 were significantly lower than in 1998, or whether the 1997 level of emissions was similar to emissions in 1996 and before. Some effort should be made to check that these data are consistently accurate. It would also be worthwhile to investigate whether additional data might be available for the years before data were reported in the Pollution Inventory.

The Pollution Inventory figures for 1998 are equivalent to an emission factor of 1.34 kg/t (sales of lead anti-knock preparations). The US EPA give lead emission factors for four sources – process vents on TML production, process vents on TEL production, sludge pits and lead recovery furnace. The total emission factor is 111 kg/tonne. This is exceptionally high, and so the factors must be for uncontrolled processes and therefore irrelevant to the current UK situation.

SIZE DISTRIBUTION

No data are available on size distributions of particulate emissions from this source.

RECOMMENDED EMISSION FACTORS

Particulate emissions

Emissions of particulate matter are relatively small from this sector and we do not recommend any detailed estimates are included for this source. Emissions given in the Pollution Inventory should instead be included with other miscellaneous non-ferrous metal and chemical processes.

Heavy metal emissions

The NAEI calculates lead emissions based on the data available in the Pollution Inventory. We recommend that this approach continues, although effort should be made to collect additional data for earlier years with which to improve the time series.

Appendix A18: Chloralkali process

SNAP CODE: 040413

SOURCE ACTIVITY TITLE: Chlorine production

NACE CODE:

PARCOM-ATMOS CODE:

ACTIVITIES INCLUDED

This section concerns the production of chlorine and sodium hydroxide by electrolysis of brine in mercury cells.

GENERAL PROCESS DESCRIPTION

The electrolysis of brine is the most common method for production of chlorine. The electrolysis produces sodium hydroxide at the cathode and chlorine gas at the anode. Three types of electrolysis cell technology can be used: the mercury cell, diaphragm cell, and the membrane cell; the difference being the method used to prevent recombination of the chlorine and sodium. Most European production uses mercury cell technology. The mercury cell is the only technology which is covered here since it is the only one with significant emissions of metals (mercury). None of the process types emits significant quantities of particulate matter.

In the mercury cell, the brine flows on top of a thin layer of co-currently flowing mercury which functions as the cathode. Titanium anodes are suspended in the brine. Sodium, liberated at the mercury cathode, immediately forms an amalgam with it, while chlorine is liberated at the anode and can be removed for purification. The mercury-sodium amalgam passes to a hydrolyser where it is reacted with water in the presence of a graphite catalyst. Sodium hydroxide and hydrogen gas are formed leaving sodium-free mercury to return to the electrolysis. Spent brine is treated to remove chlorine gas, a portion or all of it is resaturated with sodium chloride and returned to electrolysis.

SOURCES OF PARTICULATE EMISSIONS

There are no significant sources of particulate matter from chloralkali processes.

SOURCES OF HEAVY METAL EMISSIONS

The main source of mercury is that emitted as vapour into the cell rooms which is then vented to atmosphere in cell-room ventilation air. Ventilation of the cell room is necessary to control temperature, and although mercury levels in the air are very low, the quantity of air exhausted to atmosphere is very large (typically 20,000 – 120,000 Nm^3/tonne chlorine produced, according to EIPPCB, 2000e).

Mercury can also be emitted from various process exhausts, by losses from the brine in the brine circuit, losses from caustic soda products, and by losses in the hydrogen produced in the hydrolysers (strong hydrogen) and during washing of mercury prior to addition to the cells (weak hydrogen). Finally, mercury can be emitted from storage vessels and also during recovery of mercury from wastes.

ABATEMENT MEASURES AVAILABLE

Emissions in ventilation air are minimised by good cell design and good practice, such as good housekeeping and good maintenance routines. Losses from the brine circuit are minimised by careful control of the brine characteristics to prevent metallic mercury being formed. Emissions in process exhausts, and in strong and weak hydrogen can be controlled by processes such as condensation, scrubbers, and carbon adsorption.

UK SITUATION

UK plants

The UK has three plants as shown in Table A18.1.

Table A18.1 Mercury cell plants in the UK (EIPPCB, 2000e)

Company	Site	Capacity (t Cl_2)
ICI	Runcorn	738,000
Hays	Sandbach	89,000
Rhodia	Staveley	29,000
Total		**856,000**

The Runcorn process involves electrolysis of both sodium and potassium chloride brines.

UK activity statistics

The PRODCOM survey gives data on chlorine production in PRQ16. This will include production at sites which use either membrane cell or diaphragm cell technology. Total UK chlorine capacity in 1999 was 1216 ktonnes (EIPPCB, 2000e), so mercury cell plant represent 70% of the UK total. A similar figure could be assumed for production in the absence of better data.

Abatement measures used in UK plants

UK plants have adopted good practice measures to reduce mercury emissions in ventilation air. Hydrogen streams are treated to remove mercury using adsorption or absorption. Activated carbon filters are used to remove mercury from the sodium hydroxide stream. Arrestment for other mercury-containing streams include condensation.

PARTICULATE EMISSION FACTORS

There are no significant emissions of particulate emissions from chloralkali processes.

HEAVY METAL EMISSION FACTORS

The EIPPCB report (2000e) gives the range 0.2 – 2.1 g Hg per tonne of chlorine produced (data for 1998). This report also gives emission factors of 27 g Hg per tonne of chlorine in 1977 and 8 g in 1987.

The US EPA gives emission factors for controlled hydrogen vents and end boxes of 0.6 and 5 g/t (chlorine) respectively. These seem high, but the factors are given a low (E) data quality and are of unknown age.

The Pollution Inventory gives the following data on mercury emissions, shown in Table A18.2

Table A18.2 Mercury emissions from chloralkali plants in the UK (in grammes)

Process	**Capacity (t Cl_2)**	**1996**	**1997**	**1998**	**1999**
ICI, Runcorn	738,000	848	770	1050	1277
Hays, Sandbach	89,000	156	212	106	118
Rhodia, Staveley	29,000	119	57	14	14
Total	**856,000**	**1115**	**1039**	**1170**	**1409**

This yields the following emission factors, shown in Table A18.3

Table A18.3 Mercury emission factors for chloralkali plants in the UK, expressed in g/t (chlorine)

Process	**1996**	**1997**	**1998**	**1999**
ICI, Runcorn	1.15	1.04	1.42	1.73
Hays, Sandbach	1.75	2.38	1.19	1.33
Rhodia, Staveley	4.10	1.97	0.48	0.48
Total	1.30	1.21	1.37	1.65

These emission factors are within the range of values reported by EIPPCB and seem to be reasonable. The only cause for concern is that the emissions and therefore the factors for ICI, Runcorn increase over the period 1996-1999 when emission controls might be expected to cause the opposite effect. Therefore it is recommended that the operator be consulted to determine whether the PI data are consistently accurate.

SIZE DISTRIBUTION

A size distribution for particulate matter is not relevant for this sector.

RECOMMENDED EMISSION FACTORS

Particulate emissions

No emission factors are required.

Heavy metal emissions

The emission factors from the Pollution Inventory are currently used by the NAEI and it is recommended that this approach is continued. However, consideration should be given to using the emission factors for 1977 and 1987, which are given by EIPPCB (2000e), for earlier periods in the time series, as these should better reflect the, generally, lower level of control at these times. Efforts should also be made to verify the data in the Pollution Inventory for the Runcorn chloralkali plant, since emissions reported for 1996-1998 are lower than those given for 1999, whereas the opposite might be expected.

Appendix 19: Other processes

SNAP CODE:

SOURCE ACTIVITY TITLE:

NACE CODE:

PARCOM-ATMOS CODE:

ACTIVITIES INCLUDED

This chapter covers various UK chemical and metal industry processes which are not covered by any of the preceding sections. Specifically, the following industrial processes are covered:

Nickel production
Lead battery production
Copper alloys, rod, wire and semis production
Zinc oxide production
Fertiliser production
Detergent production
Titanium dioxide production
Glucose & starch production
Magnesia production
Cadmium pigment production
Soda ash production
Catalyst manufacture
Sodium tripolyphosphate production
Zinc alloy production
Carbon black manufacture

In addition, miscellaneous non-ferrous metal and chemical processes are included.

GENERAL PROCESS DESCRIPTION

A brief description of the processes in included in UK plants (see below).

SOURCES OF PARTICULATE EMISSIONS

Sources of particulate matter are covered in the section on UK plants (see below).

SOURCES OF HEAVY METAL EMISSIONS

Sources of heavy metals are covered in the section on UK plants (see below).

ABATEMENT MEASURES AVAILABLE

Abatement measures are described in the section below on abatement measures at UK plants.

UK SITUATION

UK plants

This appendix covers the following UK plants

1. Nickel refinery. The UK's only nickel refinery uses the nickel carbonyl process to produce nickel pellets and powder from imported nickel oxide. The process involves reducing the nickel oxide to crude nickel matte using hydrogen, activating the matte using sulphur bearing gas and hydrogen, and then reacting the nickel matte with carbon monoxide to form gaseous nickel carbonyl. This is thermally decomposed to form pure nickel onto the surface of nickel pellets and powders.

2. Lead battery production. A number of plants produce lead batteries. Processes involve the melting of lead followed by casting of grids and/or lead pellets. The pellets are milled to produce lead oxide powder which is captured in cyclones and/or filters. The lead oxide powder is mixed with sulphuric acid, water and additives to form a paste which is applied to the lead grid. Finally the grids are cured, generally in electric ovens.

3. Copper alloys, rod, wire and semis production. Production of various products involving melting, casting and forming of copper.

4. Zinc oxide production. These processes involve boiling of zinc in retorts. Zinc vapour oxidises in air and the resulting fine fume of zinc oxide can be recovered in fabric filters.

5. Ammonium nitrate fertiliser production. These processes involve manufacture of solid or liquid fertilisers based on ammonium nitrate. Ammonium nitrate is produced by reaction of nitric acid and ammonia. Particulate emissions will occur from dehydration of the ammonium nitrate product, and from prilling or fluid-bed granulation processes. Prilling involves cooling of hot, molten ammonium nitrate to form a solid granular product. Fluid bed granulation involves spraying concentrated ammonium nitrate into a fluidised bed of ammonium nitrate seed particles where the solution evaporates from the surface of the seed particles.

6. Titanium dioxide production. Titanium dioxide is produced from crude titanium dioxide ores by one of two methods – chlorination or sulphonation. Chlorination involves reacting the crude ore with chlorine to form gaseous titanium tetrachloride, which is purified by means of cooling to condense first various metal chloride impurities, followed by the titanium tetrachloride which then undergoes a final purification process using distillation. The titanium tetrachloride is then oxidised to titanium dioxide. The sulphonation process involves reacting the crude ore with sulphuric acid, removing iron and other impurities by a series of filtration and precipitation stages before precipitating the pure titanium dioxide which is then calcined. Both types of process involve milling of the titanium dioxide product followed by filtration and drying.

7. Glucose production. One UK process wet processes maize in order to produce various animal feeds which are dried, and starch which is further processed to glucose. Glucose is

produced by addition of chemicals (including hydrochloric acid) and enzymes. Most glucose is dehydrated to 80% before sale, but some is dried to a powder in a spray drier.

8. Magnesia production. One UK plant produces magnesia from sea water. The process includes precipitation of magnesium hydroxide from sea water using hydrated lime, dolime, or caustic soda. The precipitate is dewatered and treated in kilns. Milling and pelletisation process are also carried out.

9. Cadmium pigment production. One UK plant manufactures cadmium based pigments. Processes include evaporation of cadmium followed by oxidation to the oxide, reaction of cadmium oxide with sulphuric acid to produce cadmium sulphate, and precipitation of various cadmium based pigments.

10. Catalyst manufacture. This refers to a UK process for the manufacture of catalysts based on metals including chromium, copper, nickel, palladium, rare earth metals, and zinc. Metals are dissolved in nitric acid, precipitated by addition of various chemicals and then heat treated to form metal oxides which are used in catalyst fabrication.

11. Phosphate fertiliser production. Two UK plants manufacture single superphosphate by reacting ground phosphate rock with sulphuric acid. Emissions can occur from grinding and handling of phosphate rock and from handling and granulation of the single superphosphate product.

12. Detergent production. This category covers the manufacture of solid surfactants and detergents at one site. A range of chemical processes occur, involving reaction of oleum with either alkyl benzene, fatty acids or toluene to produce a surfactant.

13. Soda ash production. The production of soda ash by the Solvay process is carried out at two locations. Ammonia is dissolved in brine, allowing carbon dioxide to be dissolved as well. This causes the precipitation of sodium bicarbonate which is removed. Addition of calcium hydroxide produced by hydration of lime liberates ammonia which is recycled. The sodium bicarbonate is calcined to release carbon dioxide leaving sodium carbonate.

14. Zinc alloy production. Production of alloys of zinc, mainly for use in die-casting and galvanising. This sector includes some recovery of zinc from scrap as well as production of zinc alloys from refined metals.

15. Carbon black manufacture. Carbon black is produced by incomplete combustion of oils to leave a fine particulate carbon product which is removed from the waste gases using fabric filters.

16. Sodium tripolyphosphate production. Production of sodium tripolyphosphate at a single plant by reaction of phosphoric acid and sodium carbonate. The sodium tripolyphosphate is spray dried and calcined.

17. Other non-ferrous metal processes. These will include a wide range of processes involving recovery or use of metals such as tin, bismuth, antimony, precious metals, as

well as alloys such as solder. Other chemical processes will include a wide range of chemicals involving the use of or the production of dusty materials.

UK activity statistics

Activity statistics for nickel production, magnesia production, and copper consumption are given in the British Geological Survey's Annual Report. The PRODCOM survey gives data on the following:

PRQ 15	titanium oxides, zinc oxides, cadmium pigments
PRQ 16	sodium carbonate, sodium tripolyphosphate
PRQ 18	ammonium nitrates, superphosphates
PRQ 24	detergents
PRA 6	glucose
PRA 34	zinc dusts, zinc alloys, copper alloys, copper wire, copper tubes
PRA 64	batteries

Abatement measures used in UK plants

Abatement options for the [remaining] processes are shown in Table A19.1

Table A19.1 Abatement equipment in use by UK processes

Sector	Cyclones	Bag filters	Scrubbers	ESPs	Comments
Nickel refining	✓	✓	✓	✓	
Lead batteries	✓	✓			Minor sources probably not controlled
Copper alloys	✓	✓	✓		Possibly not all controlled to same level
Zinc oxide	✓	✓			Minor sources probably not controlled
Ammonium fertilisers		✓	✓		
Titanium dioxide		✓	✓		
Glucose refining		✓	✓		
Magnesia		✓	✓		
Cadmium pigments		✓			
Catalyst manufacture		✓	✓		
Phosphate fertiliser	✓	✓	✓		
Detergent manufacture	✓				
Soda ash		✓	✓		
Zinc alloys		✓			Possibly not all controlled to this level
Sodium tripolyphosphate	✓		✓		
Carbon black	✓	✓			

PARTICULATE EMISSION FACTORS

Table A19.2 shows particulate matter emission factors from the US EPA and Table A19.3 gives data taken from EIPPCB (2000b).

Table A19.2. Particulate emission factors for various processes (US EPA, 2000)

Process Stage	**Abatement**	**Emission factors g/t (product)**		**Quality rating** [a]
		Tot	**PM_{10}**	
Detergent manufacture: spray drying	Cyclones	7000		E
Detergent manufacture: spray drying	Bag filter or wet scrubber	540		E
Ammonium nitrate fertiliser: prilling tower	Controlled	260-600		A
Ammonium nitrate fertiliser: concentrators	Uncontrolled	260		A
Ammonium nitrate fertiliser: rotary drum granulator cooler	Controlled	80		A
Phosphate fertiliser: rock unloading	fabric filter	280	150	E
Phosphate fertiliser: rock feeding	fabric filter	60	30	E
Phosphate fertiliser: mixing & den	wet scrubber	260	220	E
Phosphate fertiliser: curing building	uncontrolled	3600	3000	E
Maize processing: grain receiving	fabric filter	16		E
Maize processing: grain handling	uncontrolled	430		E
Maize processing: grain cleaning	uncontrolled	820		E
Maize processing: grain cleaning	cyclone	86		E
Maize processing: gluten drying	cyclone	130-250		E
Lead battery production: grid casting	uncontrolled	80-1420		B
Lead battery production: paste mixing	uncontrolled	1000-1960		B
Lead battery production: PbO mill	baghouse	50-100		C
Lead battery production: lead reclaim furnace	uncontrolled	700-3030		B
Lead battery production: 3-process operation	uncontrolled	13200-42000		B
Lead battery production: dry formation	uncontrolled	14000-14700		B

Process Stage	Abatement	Emission factors g/t (product)		Quality rating [a]
		Tot	PM_{10}	
Lead battery production: small parts casing	uncontrolled	90		C
Carbon black – main process vent	uncontrolled	3270		C
Carbon black – main process vent	flare	1350		C
Carbon black – main process vent	CO boiler & incinerator	1040		C
Carbon black - dryer	Bag filter	120		C

For carbon black the most appropriate factors would be to use that for CO boiler & incinerator to treat process emissions and bag filter to control emissions from the dryer, giving an emission factor of 1160 g/t (product). In other cases, emission factors can be selected taking into account the abatement technologies given in Table A19.1.

Table A19.3 Dust emission factors for various processes (EIPPCB, 2000b)

Process	Abatement technology	Emission factor g/t (product)
Copper wire/rod production	Not stated (probably refers to range of processes)	20-500
Nickel carbonyl process	Not stated (factor is probably for UK process)	280

The Pollution Inventory does not include reported emissions for the UK nickel production site, but the EIPPCB figure seems to be derived from data on the UK process. Pollution Inventory data for other sources are given in Table A19.4.

Table A19.4 Emissions of particulate matter reported in the Pollution Inventory for various processes, expressed in tonnes

Type	Pollutant	1995	1996	1997	1998	1999
Cadmium pigments	TPM	3.72	0.198	2.75		
Chemical industry (ammonium based fertilisers)	TPM	413	482	446	609	669
Chemical industry (ammonium based fertilisers)	PM_{10}				50.0	80.0
Chemical industry (carbon black)	TPM	49.2	54.8	47.8	88	78
Chemical industry (carbon black)	PM_{10}					31.0
Chemical industry (detergents)	TPM	16.8	20.8		20.3	36.0
Chemical industry (detergents)	PM_{10}				18.3	15.3
Chemical industry (magnesia)	TPM	38.0	32.8	20.4		18.7
Chemical industry (phosphate based fertilisers)	TPM	16.7	17.3	20.6		0.184
Chemical industry (titanium dioxide)	TPM	76.2	53.4	80.8	144	165
Chemical industry (titanium dioxide)	PM_{10}				127	154
Glucose refining	TPM				137	89.1

Non-ferrous metals (copper alloys & semis)	TPM	2.8	20.3	13.6	57.8	26.2
Non-ferrous metals (zinc alloys & products)	TPM		5.28	3.10		
Non-ferrous metals (zinc alloys & products)	PM_{10}					1.00
Soda ash	TPM	10.8	23.0	95.9	16.2	15.0
Zinc oxide production	TPM	44.0	6.52	1.00	1.08	1.79
Zinc oxide production	PM_{10}				3.40	3.75

These data may be converted into emission factors using activity data or chemical plant capacity data. The resulting emission factors are shown in Table A19.5

Table A19.5 Emission factors for particulate matter based on Pollution Inventory data for various processes, expressed in g/t (product)

Sector	Pollutant	1997	1998	1999
Cadmium pigments & stabilisers	TPM	2160		
Chemical industry (ammonium based fertilisers)	TPM		341	309
Chemical industry (ammonium based fertilisers)	PM_{10}		28.0	37.0
Chemical industry (carbon black)	TPM		436	386
Chemical industry (carbon black)	PM_{10}			153
Chemical industry (detergents)	TPM		43.0	79.5
Chemical industry (detergents)	PM_{10}		38.7	33.7
Chemical industry (magnesia)	TPM			187
Chemical industry (phosphate based fertilisers)	TPM			3.52
Chemical industry (titanium dioxide)	TPM		553	632
Chemical industry (titanium dioxide)	PM_{10}		488	591
Glucose refining	TPM		455	297
Non-ferrous metals (copper alloys & semis)	TPM		127	67.9
Non-ferrous metals (zinc alloys & products)	TPM	40.2		
Non-ferrous metals (zinc alloys & products)	PM_{10}			16.3
Soda ash	TPM		16.2	15
Zinc oxide production	TPM		41.7	77.6
Zinc oxide production	PM_{10}		131	162
Sodium tripolyphosphate	TPM			276

HEAVY METAL EMISSION FACTORS

Table A19.6 shows particulate matter emission factors from the US EPA and Table A19.7 gives data taken from EIPPCB (2000b).

Table A19.6 Heavy metal emission factors for various processes (US EPA, 2000)

Process Stage	**Abatement**	**Emission factors g/t (product) Pb**	**Quality rating** [a]
Lead battery production: grid casting	uncontrolled	350-400	E
Lead battery production: paste mixing	uncontrolled	500-1130	B
Lead battery production: PbO mill	baghouse	50	C

Process Stage	**Abatement**	**Emission factors g/t (product) Pb**	**Quality rating [a]**
Lead battery production: lead reclaim furnace	uncontrolled	350-630	B
Lead battery production: 3-process operation	uncontrolled	4790-6600	B
Lead battery production: small parts casing	uncontrolled	50	C

Table A19.7 Heavy metal emission factors for various processes (EIPPCB, 2000b)

Process	**Pollutant**	**Abatement technology**	**Emission factor g/t (product)**
Copper semis production	Cu	Not stated (probably refers to range of processes)	1-3.5
Copper semis production	Pb	Not stated (probably refers to range of processes)	0.1-1
Copper semis production	As	Not stated (probably refers to range of processes)	0.01-0.2
Copper wire/rod production	Cu	Not stated (probably refers to range of processes)	12-260
Nickel carbonyl process	Ni	Not stated (factor is probably for UK process)	7

The Pollution Inventory gives a nickel emission of 1.374 tonnes in 1999. Using production data from the British Geological Survey (2000), an emission factor of 34.8 g/t (nickel) can be derived. Data from the Pollution Inventory for other processes are shown in Table A19.8.

Table A19.8 Emissions of heavy metals from various process types reported in the Pollution Inventory, expressed in kg.

Sector	Metal	1995	1996	1997	1998	1999
Cadmium pigments & stabilisers	Cadmium	11.3	6	1	3	5
Cadmium pigments & stabilisers	Lead				60	14
Cadmium pigments & stabilisers	Selenium				2	3
Chemical industry (magnesia)	Chromium				243	690
Lead batteries	Lead	1340	1420	833	2530	2540
Non-ferrous metals (copper alloys & semis)	Arsenic				38	44
Non-ferrous metals (copper alloys & semis)	Cadmium	0.67	50		12	44
Non-ferrous metals (copper alloys & semis)	Copper	2.74	2390	1560	1980	4080
Non-ferrous metals (copper alloys & semis)	Nickel	21.0	84		47	317
Non-ferrous metals (copper alloys & semis)	Lead	108	628	40	845	975
Non-ferrous metals (copper alloys & semis)	Zinc	228	41	311	4640	12000
Non-ferrous metals (zinc alloys & products)	Cadmium					1
Non-ferrous metals (zinc alloys & products)	Zinc	2240	1450	1480	4090	12600
Zinc oxide production	Zinc	114	175	182	5980	5750

These are converted into emission factors using suitable activity data as shown in Table A19.9

Table A19.9 Emission factors for heavy metals based on Pollution Inventory data for various processes, expressed in g/t (product)

Sector	**Metal**	**1998**	**1999**
Cadmium pigments & stabilisers	Cadmium	2.35	1.65
Cadmium pigments & stabilisers	Lead	47.1	4.61
Cadmium pigments & stabilisers	Selenium	1.57	0.988
Chemical industry (magnesia)	Chromium	2.43	6.90
Lead batteries	Lead	24.1	24.2
Non-ferrous metals (copper alloys & semis)	Arsenic	0.0834	0.114
Non-ferrous metals (copper alloys & semis)	Cadmium	0.0263	0.114
Non-ferrous metals (copper alloys & semis)	Copper	4.35	10.5
Non-ferrous metals (copper alloys & semis)	Nickel	0.103	0.820
Non-ferrous metals (copper alloys & semis)	Lead	1.85	2.52
Non-ferrous metals (copper alloys & semis)	Zinc	10.2	31.0
Non-ferrous metals (zinc alloys & products)	Cadmium		0.0163
Non-ferrous metals (zinc alloys & products)	Zinc	53.1	205
Zinc oxide production	Zinc	231	249

SIZE DISTRIBUTION

No size distributions are available.

RECOMMENDED EMISSION FACTORS

Particulate emissions

It is recommended that the emission factor given by EIPPCB (2000b) is used for the UK nickel process, and that emissions data taken from the Pollution Inventory are used as the source of emission estimates and emission factors for other processes.

Heavy metal emissions

It is recommended that the Pollution Inventory is used as a source of emission factors for these processes.

Appendix B1 – Chromium speciation

INTRODUCTION

The sources of chromium emissions in 1999 are shown in Table B1.1. This shows that the most significant emissions are from relatively few sources.

Table B1.1 Summary of emissions of chromium in 1999

Sector	% of total emissions
Chromium chemicals	26.9
Coal-fired power stations	25.7
Electric arc furnaces	19.0
Industrial/commercial coal combustion	5.2
Glass production	4.7
Domestic combustion of smokeless fuels	3.7
Coke production	3.3
Domestic combustion of coal	2.4
Basic oxygen furnaces	1.9
Sinter plants	1.6
Blast furnaces	1.5
Other sources	4.2

Species profiles are required for the sources listed in Table B1.1, but the lowest priority has been given to those numerous sources grouped together as 'other sources' as these form less than 5% of the total UK emission.

Chromium (Cr) occurs ubiquitously in nature. It can occur in two main oxidation states: trivalent (Cr^{3+}) and hexavalent (Cr^{6+}). Almost all the hexavalent chromium is produced from human activity. Hexavalent chromium is derived from the industrial oxidation of mined chromium deposits and can be derived from the combustion of fuels. In this oxidation state, chromium is relatively stable in air and pure water. Hexavalent chromium is quickly reduced to the trivalent form when it comes into contact with organic matter in air, soil and water. The Cr^{3+} and Cr^{6+} oxidation states have very different biological properties, and should be considered separately.

TOXICOLOGY

The inhalation of chromium-containing aerosols is an important route of exposure to chromium compounds because the bronchial tree is the major target organ for the carcinogenic effects of Cr^{6+}. Certain trivalent chromium compounds are essential to man (Mertz, 1967; Anderson, 1989) and are required for normal sugar and fat metabolism.

Chromium is a good example of where the valence state of an element profoundly affects its toxicity. Chromium(III) is probably an essential element needed for glucose metabolism, but Chromium(VI) is genotoxic and carcinogenic (Mertz, 1967; Katz and Salem, 1994).

Chromium(VI) does not bind to DNA, but is intracellularly reduced to Cr^{3+} which does (Whitmer *et al.*, 1989). The binding of Cr^{3+} is insufficient to damage DNA *in vitro* and it is likely that that the damage arises from the intermediaries in the reduction process, perhaps intermediate valence states of chromium itself (Aiyar *et al.*, 1991).

Toxicological effects

The WHO have summarised the toxicological effects of Cr. These include chrome ulcers, corrosive reaction on the nasal septum, acute irratative dermatitis and allergic eczematous dermatitis.

Carcinogenic effects

Epidemiological studies of workers exposed to chromate have an excess of lung cancers that ranged in magnitude from a slight excess to a 50-fold excess (Langard, 1983). There are also several other studies that link occupational exposure to chromate to lung cancer. IARC (1990) have classified hexavalent chromium compounds as Group 1 (carcinogenic to humans) and trivalent chromium and metallic chromium as Group 3 (not classifiable as to their carcinogenisity to humans).

WHO guideline for human exposure to air

Hexavalent chromium is carcinogenic, and as there is no know safe threshold, the WHO has not recommended a safe level for exposure to hexavalent chromium (WHO, 1987). With an air concentration of 1 $\mu g\ m^{-3}$, the lifetime risk is estimated to be $4x10^{-2}$.

Requirements for speciation

Species profiles need to distinguish between chromium present in the hexavalent state from other forms of chrmoium, although at present no further details on speciation are required.

SPECIES IN EMISSIONS

Chromium chemicals

Data have been provided by the process operator (see Table 16.3) which should be used in the NAEI. These data are shown in Table B1.2, expressed as percentages of total chromium emissions from the plant.

Table B1.2 Percentage of chromium emission by species from UK chromium chemicals manufacture

Year	Hexavalent	Soluble trivalent	Insoluble trivalent	Trivalent total
1988	37%	8%	55%	63%
1989	37%	8%	55%	63%
1990	36%	11%	53%	64%
1991	29%	13%	58%	71%
1992	36%	11%	53%	64%
1993	41%	14%	45%	59%
1994	30%	17%	53%	70%
1995	21%	19%	60%	79%
1996	19%	19%	62%	81%
1997	24%	13%	64%	76%
1998	19%	9%	72%	81%
1999	19%	13%	68%	81%
2000	18%	7%	75%	82%

The emissions from the plant in the last three years have been 19% hexavalent and 81% trivalent (mostly insoluble). Data for earlier years suggests that the proportion of hexavalent chromium was higher then (e.g. 41% in 1993).

Combustion

Chromium is emitted into the air by all combustion processes. The oxidation state of chromium in these emissions is not well defined, but it might be assumed that the heat of combustion will oxidise a proportion of the element to the hexavalent state. While suspended in air, this state is probably stable, until it comes into contact with organic matter which will eventually reduce it to the trivalent form (WHO, 1988).

Laboratory studies of fly ash particles carried out by Rai and Szelmeczka (1990) have shown that chromium is present in these particles as Cr(III). The predicted primary species is expected to be Cr_2O_3. Eary *et al.* (1990) quote investigations where chromium substitutes into the structure of iron minerals in solid solution with Fe(III). Chromium is likely to be mainly found on the surface of particles produced during combustion ('Group 2' behaviour element, Clarke and Sloss, 1992).

A report by the US EPA (Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units, Interim Final Report Volume 1, EPA-453/R-96-013a, 1996) is quoted by Wright (2001), in which the following data are given:

coal-fired power plant	Cr^{6+} is 11% of emitted chromium
oil-fired power plant	Cr^{6+} is 18% of emitted chromium

On the basis of the above data, we will assume that most of the chromium in emissions from combustion processes is present in the trivalent state and that emissions of hexavalent chromium are below 20% of the total chromium emissions. For power stations burning fuel oil and coal, the US EPA data above should be used, while for other combustion processes we propose that emissions from combustion processes burning liquid fuels be assumed to be 20% hexavalent and 80% trivalent and that emissions from combustion processes burning solid fuels be assumed to be 10% hexavalent and 90% trivalent.

Iron & steel industry

No literature data have been identified on the form of chromium released from coke ovens, electric arc furnaces, blast furnaces, sinter plant or basic oxygen furnaces. Some general information on emissions from integrated steelworks has been provided by Brooks (2001), who states that emissions of chromium are in the form of oxides, probably as complex (mixed) oxide phases or mineralogical phases such as spinels, rather than as simple oxide species. In the case of emissions from sinter plant, emissions may also be in the form of chlorides. Brooks also estimates that chromium is present as Cr^{3+}, and states that there is little evidence that Cr^{6+} is present in measurable quantities. On the basis of this information, we propose that the emissions from iron and steel industry processes are assumed to be Cr^{3+} only.

Glass manufacture

Chromium is known to be used generally within the glass industry as a colourant and also as a coupling agent to link the surface of continuous filament glass fibres to the applied polymer coating. An important colourant is iron chromite ($Fe_2O_3.Cr_2O_3$), although both Cr^{3+} and Cr^{6+} are used, to impart green and yellow colour respectively. Iron chromite is used to produce green container glass and, since this is a major product of the glass industry, it might be expected that most chromium is emitted in the trivalent state. Yellow glass is relatively uncommon and so the use of hexavalent chromium might be expected to be relatively small. It is possible that some chromium present in the trivalent state could be oxidised to the hexavalent form in the high temperature burners used to melt the glass. No measurement data has been identified on the form of chromium released from glass processes, however we will tentatively propose a profile of 10% hexavalent and 90% trivalent chromium.

PROPOSED SPECIES PROFILES

Initial estimates of the proportion of hexavalent and trivalent chromium species emitted from major sources in the UK are shown in Table B1.3 below. Note that the estimates refer to emissions at the point of release, and not to the proportion of species that may be found in ambient air.

Table B1.3 Proposed species profiles for emissions of chromium

Sector	Hexavalent	Trivalent
Chromium chemicals	19%[a]	81%[a]
Coal-fired power stations	11%	89%
Electric arc furnaces	0%	100%
Industrial/commercial coal combustion	10%	90%
Glass production	10%	90%
Domestic combustion of smokeless fuels	10%	90%
Coke production	0%	100%
Domestic combustion of coal	10%	90%
Basic oxygen furnaces	0%	100%
Sinter plants	0%	100%
Blast furnaces	0%	100%
Other sources	-[b]	-[b]

a – These values are averages for the period 1998-2000, and are given to illustrate the typical split between haxavalent and trivalent chromium. However, the operator has provided detailed data and this should be used in the NAEI.

b- No profiles are proposed, however combustion related sources may be assumed to be 20% hexavalent and 80% trivalent in the case of liquid fuels and 10% hexavalent and 90% trivalent in the case of solid fuels.

The profiles are generally subject to great uncertainty and the collection of further data through desk-based and/or measurement work is a high priority.

Appendix B2 Mercury speciation

INTRODUCTION

The sources of mercury emissions in 1999 are shown in Table B2.1. This shows that the most significant emissions are from relatively few sources, although a larger number of sources might be considered significant than is the case with chromium (see Table B1.1).

Table B2.1 Summary of emissions of mercury in 1999

Sector	% of total emissions
Coal-fired power stations	18.2
Chloralkali processes	16.5
Crematoria	15.7
Industrial/commercial coal combustion	11.5
Domestic combustion of smokeless fuels	4.8
Disposal of waste containing mercury	4.6
Clinical waste incineration	4.3
Primary lead/zinc production	3.4
Cement production	2.8
Electric arc furnaces	2.5
Foundries	2.4
Domestic combustion of coal	2.2
Industrial combustion of wood	1.5
Sewage sludge incineration	1.4
MSW incineration	1.3
Sinter plants	1.0
Blast furnaces	1.0
Other sources	4.9

Species profiles are required for the sources listed in Table B2.1, but the lowest priority has been given to those sources grouped together as ‘other sources’ as these form less than 5% of the total UK emission.

Mercury may exist in three oxidation states: Hg^0, Hg^{1+} and Hg^{2+}. There is more information about the environmental chemistry and more measurements of speciation of Hg than for chromium and nickel. Divalent mercury can be present in the atmosphere in either organic or inorganic forms and may also be attached to particles, often given the notation $Hg^{II}_{(p)}$.

TOXICOLOGY

The toxic effects of mercury and its compounds depend on the chemical form of mercury. Methyl mercury is converted to inorganic mercury in humans (WHO, 1990). The duration of exposure and the time after the exposure ceased determine the fraction of Hg^{2+} present in tissues. There is a complex interplay between inorganic mercury and some other elements, including selenium. The formation of a selenium complex may be responsible for the long half-life of a fraction of the inorganic mercury in humans, although the biological half-life of most absorbed Hg is days or weeks (WHO, 1991b).

Mercury vapour

Mercury vapour largely damages the nervous system, but effects can be seen at higher doses in the oral mucosa and the kidneys (US EPA, 1984). Most effects of mercury vapour usually disappear within a few months after exposure has ceased. There is no evidence that mercury vapour is a human carcinogen (WHO, 1987).

The sole source of human exposure to mercury vapour is the atmosphere. Inhaled mercury vapour is oxidised to divalent ionic Hg in the human body by the hydrogen-peroxide catalase pathway (Hursch *et al.*, 1988). IARC (1993) have classified inorganic mercury compounds as Group 3 (not classifiable as to their carcinogenisity to humans).

Inorganic divalent mercury compounds

These are corrosive poisons and acute single doses can cause death by kidney failure and systemic shock (WHO, 1987). There is little information on the effects of chronic exposure of inorganic mercury compounds in humans.

Occupational exposure to mercuric oxide has been shown to damage the peripheral nervous system. Exposure to inorganic divalent mercury has been known to produce 'pink disease' in susceptible children. The effects of chronic exposure to mercuric oxide and divalent mercury are normally reversible. IARC (1993) have classified inorganic mercury compounds as Group 3 (not classifiable as to their carcinogenisity to humans).

Methyl mercury

This compound almost exclusively damages the nervous system. In human adults, the damage is selective and affects certain areas of the brain concerned with sensory and co-ordination functions. Some of these effects are not reversible. The diet is the dominant source of methyl mercury compounds. IARC (1993) have classified methyl mercury as Group 2B (possibly carcinogenic to humans).

WHO guideline for human exposure to air

There are no data to indicate that Hg or Hg compounds human carcinogens, and the WHO has set no risk estimate.

Adequate protection from indoor air pollution by mercury is provided by a guideline value of exposure of approximately 1 μg Hg m^{-3}, as an annual average, irrespective of the form of mercury.

Requirement for speciation

Species profiles should include metallic mercury vapour, inorganic divalent mercury, particulate mercury, and methyl mercury if possible.

SPECIES IN EMISSIONS

Combustion

At combustion temperatures, in an oxidising environment, elemental Hg will evaporate. In combustion systems, most chemicals forms of Hg (e.g. HgS) are expected to leave the *combustion zone* as gaseous, elemental Hg (Hall *et al.*, 1990).

Mercury volatilised during combustion can leave *a stack* as a mixture of the elemental form (Hg^0) or in oxidised forms such as HgO, $HgCl_2$, and CH_3HgCl (Bloom, 1991). Unlike other trace elements, very little Hg is emitted in particulate from.

Most combustion processes appear to emit the Hg(II) oxidised species, rather than organo-Hg or other oxidised species (Clarke and Sloss, 1992).

The major source of data on species of mercury in emissions are Pacyna *et al*, 2001 who have proposed profiles for various sources and Lee *et al*, 2000 who have presented a review of the literature on mercury speciation, Relevant figures are shown in Table B2.2.

Table B2.2 Species present in mercury emissions from combustion plant

Source	Hg^0	Hg^{2+}	Hg(p)	MeHg	Source[a]
Coal combustion with ESP	62%	38%			1
Coal fired stations with ESP & FGD	58%	42%			1
Coal combustion	50%	40%	11%		2
Coal fired power plants	50%	40%	10%		3
Oil fired power plants	77%	23%			1
Oil combustion	51%	39%	10%		2
Oil combustion	50%	40%	10%		3
Coal combustion in residential plant	50%	40%	10%		3

a - 1 = Data quoted in Lee et al, 2000, 2 = Profile suggested by Lee et al, 2000, 3 = Pacyna et al, 2001

Both Lee and Pacyna are in close agreement in the profiles they recommend. We propose therefore to use a profile of 50% Hg^0, 40% Hg^{2+}, and 10% Hg(p) based on these two profiles for all large-scale combustion. In the case of domestic combustion, however, we consider that the level of mercury on particulate matter will be higher, given the significantly higher emissions of particulate and so we propose a modified profile of 40% Hg^0, 40% Hg^{2+}, and 20% Hg(p).

Chlor-alkali processes

Pacyna *et al* 2001 suggest that the predominant species is Hg^0 at 70% with the remaining 30% of mercury present as Hg^{2+}. This is supported by Lee *et al*, 2000, who proposes 29% for Hg^{2+} and 71% for Hg^0. We propose to use a profile of 70% Hg^0 and 30% Hg^{2+}.

Waste incineration and cremation

Pacyna *et al* 2001 suggest that 60% of mercury is in the form Hg^{2+}, with 20% present as Hg^0 and as Hg(p). Lee *et al*, 2000, suggest 22% for Hg^0 and Hg(p), with 56% as Hg^{2+}. The percentage suggested for particulate-bound mercury in both these sources seem very high given the stringent particulate matter controls at modern incineration plant. In the case of crematoria, particulate matter abatement is less good and so higher levels of Hg(p) might be expected. We would also suggest that the proportion of Hg^{2+} might be higher than the 60% given by Pacyna *et al*, 2000, due to the highly oxidising conditions with excess air. We therefore propose to use the following profiles:

Crematoria	95% Hg^{2+}, 4% Hg(p), 1% Hg^0
Other incineration	95% Hg^{2+}, 1% Hg(p), 4% Hg^0

Disposal of mercury containing waste

Emissions of mercury from the disposal of mercury-containing waste such as measurement and control equipment is assumed to be 100% Hg^0.

Cement manufacture and ferrous and non-ferrous metal processes

Pacyna *et al*, 2001 suggest that 80% of mercury from this source is emitted as Hg^0 with 15% as Hg^{2+}, the remaining 5% being Hg (p). Lee *et al*, 2000 suggest the same split for cement and very similar splits for ferrous and non-ferrous processes and therefore this profile is proposed for the NAEI.

PROPOSED SPECIES PROFILES

Based on the data given above, the following species profiles, shown in Table B2.3 are proposed for use in the NAEI.

Table B2.3 Proposed species profiles for emissions of mercury

Sector	Hg^0	Hg^{2+}	Hg (p)	MeHg
Coal-fired power stations	50%	40%	10%	-
Chloralkali processes	70%	30%	-	-
Crematoria	1%	95%	4%	-
Industrial/commercial coal combustion	50%	40%	10%	-
Domestic combustion of smokeless fuels	40%	40%	20%	-
Disposal of waste containing mercury	100%	-	-	-
Clinical waste incineration	4%	95%	1%	-
Primary lead/zinc production	80%	15%	5%	-
Cement production	80%	15%	5%	-
Electric arc furnaces	80%	15%	5%	-
Foundries	80%	15%	5%	-
Domestic combustion of coal	40%	40%	20%	-
Industrial combustion of wood	50%	40%	10%	-
Sewage sludge incineration	4%	95%	1%	-
MSW incineration	4%	95%	1%	-
Sinter plants	80%	15%	5%	-
Blast furnaces	80%	15%	5%	-
Other sources	80%	15%	5%	-

Currently, the available data only allows the identification of major components. The absence of data on methyl mercury reflects the fact that it is likely to be present in trace concentrations, but the collection of more data on this species is a priority due to concerns about its toxicological properties. The profiles are generally subject to great uncertainty and the collection of further data through desk-based and/or measurement work is a high priority.

Appendix B3 - Nickel speciation

INTRODUCTION

The sources of nickel emissions in 1999 are shown in Table B3.1. This shows that the most significant emissions are from relatively few sources.

Table B3.1 Summary of emissions of nickel in 1999

Sector	% of total emissions
Industrial/commercial fuel oil combustion	40.1
Industrial/commercial coal combustion	13.4
Industrial combustion of petroleum coke	8.6
Domestic combustion of coal	6.9
Domestic combustion of smokeless fuels	6.6
Coal-fired power stations	6.1
Electric arc furnaces	5.2
Primary aluminium production	1.8
Glass production	1.5
Fuel oil fired power stations	1.2
Sinter plants	1.2
Blast furnaces	1.0
Industrial combustion of coke	0.9
Nickel refining	0.9
Other sources	4.6

Species profiles are required for the sources listed in Table B3.1, but the lowest priority has been given to those numerous sources grouped together as ‘other sources’ since these contribute less than 5% of total UK emissions.

Nickel forms compounds in several oxidation sates, but only the divalent ion is important for both inorganic and organic species. Nickel compounds that are almost insoluble in water include (WHO, 1987):

- carbonate
- sulphides (main forms are nickel sulphide, NiS, and nickel subsulphide, Ni_3S_2)
- oxides (NiO and Ni_2O_3)

Nickel carbonyl $Ni(CO)_4$ is probably the only gaseous Ni compound of environmental importance (WHO, 1991) and is a suspected human carcinogen. However, it has a short atmospheric lifetime. Nickel carbonyl is unstable in air and decomposes to form nickel carbonate. Stedman & Hikade (1980) have estimated the atmospheric lifetime of nickel carbonyl. At 25°C, the lifetime of ng m^{-3} concentrations is around one minute, and increases by one minute for each mg m^{-3} of CO_2 present. Nickel can be produced from nickel carbonyl by thermal decomposition, and this is the basis of the process operated at the UK nickel refinery.

TOXICOLOGY

Humans are exposed to the majority of nickel though consuming food; typical daily intakes are below 300 µg (Clemente *et al.*, 1980). Daily intakes from urban ambient air typically will be below 0.4 µg, but as cigarette smoke contains nickel, daily intakes of smokers are higher and typically are below 15 µg (NAS, 1975).

Pulmonary absorption of nickel carbonyl is rapid and extensive, and this compound will pass through the alveolar wall intact (Sunderman and Selin, 1968). Few data exist on the pulmonary absorption of nickel from particulate matter deposited in the lungs.

Nickel seems to have quite a short biological half-life of tens of hours (Onkelinx and Sunderman, 1980). Absorbed nickel is cleared in urine and unabsorbed nickel in the faeces.

Toxicological effects

In man, acute intoxication with nickel carbonyl, allegy dermatitis (most common in women), asthma (in nickel workers) and mucosal irritations are reported (WHO, 1987).

Carcinogenic effects

Studies linking uptake from the environment and cancer incidence in the general population are not available (WHO, 1987). In the past, workers in the nickel industry were at significantly higher risk for cancer of the lungs and the nasal cavity. IARC (1990) have classified nickel compounds (sulphides, oxides, sulphates etc) as Group 1 (carcinogenic to humans) and metallic nickel as Group 2B (possibly carcinogenic to humans).

WHO guideline for human exposure to air

It is likely that nickel compounds are carcinogenic for humans. Therefore, the WHO sets no safe level for nickel exposure. For an air nickel dust concentration of 1 μg^{-3}, a conservative estimate of the lifetime risk is 4×10^{-4}.

Requirements for speciation

As a starting point, species profiles should include nickel carbonyl, nickel sulphides & nickel oxides as well as other forms such as metallic nickel and soluble nickel salts (sulphates etc.)

SPECIES IN EMISSIONS

Combustion

The major source of airborne nickel is from fossil fuels containing trace amounts of the metal. Petroleum products, in particular fuel oils are the main sources of nickel. The main nickel species from fossil fuel combustion are soluble salts of Ni^{2+}, typically sulphates, and often smaller quantities of insoluble oxides (Goldstein, 1991). Small quantities of complex metal oxides containing nickel may also be emitted (spinels, possibly trevorite - $NiFe_2O_4$)

(Bodog *et al.*, 1994; Galbreath *et al.*, 2000). A report by CONCAWE (1999) also mentions sulphides and nitrates emitted from fuel oil combustion.

A report by the US EPA (Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units, Interim Final Report Volume 1, EPA-453/R-96-013a, 1996) is quoted by Wright (2001), in which the following data are given for oil fired power stations:

Soluble nickel (sulphates, chloride etc.)	58%
Nickel sulphides (Ni_2S_3, NiS, NiS_2)	3%
Nickel oxides	39%

Formation of nickel carbonyl from coal combustion is suggested by Wright (2000) to be as no more than trace quantities, otherwise there would be a significant discrepancy between stack emission (solid) analytes and fuel composition. This is supported by CONCAWE (1999) which quotes measurements which either did not find nickel carbonyl or where it was detected at levels below 1%.

Recent measurements in German urban background ambient air (Füchtjohann *et al.*, 2000) point to the possibility that upto 8% of the nickel compounds belong to the sulphidic fraction and this also suggests that there is a contribution from combustion sources.

Diesel oil may contain upto 2 mg/litre of nickel (2 ppm) (Fishbein, 1981) and the vapour phase of diesel engine exhaust may contain nickel carbonyl. Measurements of urban air near a busy road junction (Filkova and Jager, 1986) have shown nickel carbonyl concentrations in the range from below the LOD upto 14 ng m^{-3}.

As a starting point, it is proposed that emissions from combustion processes are speciated using a modified version of the EPA data:

Nickel sulphides	3%
Nickel carbonyl	0.5%
Nickel oxides	39%
Soluble nickel	57.5%

Iron and steel industry processes

No literature data has been identified to data on the form of nickel released from this source, however Brooks (2001) states that nickel is emitted in the divalent form, as complex (mixed) oxide phases or mineralogical phases (such as spinels). Based on this information, it is proposed that emissions from this source are assumed to be nickel oxides.

Primary aluminium production

No data has been identified to data on the form of nickel released from this source.

Glass manufacture

Nickel in the divalent state is known to be used generally within the glass industry as a colourant, imparting various colours to glass, depending on the composition of the glass. No measurement data has been identified on the form of nickel released from glass processes, but we propose to assume that emissions are nickel oxides and other species such as sulphates.

Nickel refining

The nickel refining process involves the reduction of nickel oxide to crude nickel matte which is then reacted with carbon monoxide to produce nickel carbonyl ($NI(CO)_4$). The nickel carbonyl is allowed to decompose to pure nickel on the surface of nickel pellets or powders. The produced nickel powders are recovered in bag filters. No information has been found on the species emitted from the plant, however it is possible that all of the forms present in the process could be emitted i.e. nickel oxide, nickel carbonyl and metallic nickel. We will assume that nickel carbonyl, which is an intermediate formed and then consumed in the process, is much less likely to be emitted than the fine particulate nickel oxide and nickel powders which are the feedstock and product respectively. We therefore propose that emissions be assumed to be a trace of nickel carbonyl (say 2%) and the rest is assumed to be nickel oxide and metallic nickel (say 49% of each).

PROPOSED SPECIES PROFILES

Proposed species profiles for nickel emissions are given in Table B3.2. The profiles are subject to great uncertainty and the collection of further data through desk-based and/or measurement work is a high priority.

Table B3.2 Proposed species profiles for emissions of nickel

Sector	MN	ON	SO	NC	SU
Industrial/commercial fuel oil combustion		39%	57.5%	0.5%	3%
Industrial/commercial coal combustion		39%	57.5%	0.5%	3%
Industrial combustion of petroleum coke		39%	57.5%	0.5%	3%
Domestic combustion of coal		39%	57.5%	0.5%	3%
Domestic combustion of smokeless fuels		39%	57.5%	0.5%	3%
Coal-fired power stations		39%	57.5%	0.5%	3%
Electric arc furnaces		100%			
Primary aluminium production					
Glass production		50%	50%		
Fuel oil fired power stations		39%	57.5%	0.5%	3%
Sinter plants		100%			
Blast furnaces		100%			
Industrial combustion of coke		39%	57.5%	0.5%	3%
Nickel refining	49%	49%		2%	
Coke production		100%			
Other sources[a]	-	-		-	-

MN=metallic nickel, ON=oxidic nickel, SO=soluble nickel salts, NC=nickel carbonyl, SU=sulfidic nickel,
a – Emissions from combustion sources can be assumed to be 39% ON, 57.5% SO, 0.5% NC and 3% SU.

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