Report

Summary Report for Project EPG 1/3/195 "Emission Factors for Air Pollutants", Year 1

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

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Executive Summary

This report has been prepared for the Department for the Environment, Food and Rural Affairs; the National Assembly of Wales: the Scottish Executive; and the Department of Environment in Northern Ireland by **netcen** (an operating division of AEA Technology) under the contract EPG 1/3/195, "Emission factors for air pollutants".

The Department for Environment Food and Rural Affairs (Defra) Air and Environment Quality (AEQ) Division is responsible for maintaining the UK National Atmospheric Emissions Inventory (NAEI). The NAEI is maintained by **netcen** on behalf of Defra. As part of the ongoing quality control of the NAEI the quantitative uncertainty in the national emission total of each component pollutant in NAEI is reviewed annually. Based on the findings of this review project EPG 1/3/195 aims to characterise and minimise uncertainty in the emission factors used in the compilation of the NAEI and by association those of other UK inventories. The project has objectives (Tasks) that are set and reviewed annually; these comprise data collection and evaluation via literature review, personal contact with industrial representatives, direct source measurement and other means as appropriate.

This summary report presents a review of Year One tasks, outlines the principal findings and recommendations, and describes the nature of the improvement to the NAEI. The formal reports for each task are available from the Defra research report web site at <u>http://www.airquality.co.uk/archive/reports/list.php</u>

The emission factor improvement tasks for the first year were prioritised according to a ranking procedure based on the magnitude of the current uncertainty, policy relevance, and the cost-effectiveness of resource allocation. Year One tasks comprised the following:-

- Characterisation of new persistent organic compounds
- Ad-hoc data collection at a domestic and international level
- Fuel characterisation
- Combustion processes
- Iron and steel production
- Cement production
- Pesticide composition

Through the completion of these tasks, an estimated **100** emission factor and other inventory changes have been recommended for the NAEI and the overall quality of the emission database has improved as follows: -

- There is a more comprehensive body of information available for compiling the NAEI, and the database is more complete and self consistent as a result.
- The data collected has enabled more accurate estimate of time series.
- Knowledge of the uncertainty of key pollutants has been improved.

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1 Introduction

1.1 PROJECT BACKGROUND

This report has been prepared for the Department for the Environment, Food and Rural Affairs; the National Assembly of Wales: the Scottish Executive; and the Department of Environment in Northern Ireland by **netcen** (an operating division of AEA Technology) under the contract EPG 1/3/195 - Emission factors for air pollutants.

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This report summarises the individual tasks, listing findings and noting the improvements to the inventory.

1.2 DEVELOPMENT OF YEAR ONE WORK PROGRAMME

The tasks included in this project include reappraisal of the adequacy of emission factors used in the NAEI and updating the priority list of sources and pollutants in order to improve the overall quality of the NAEI and other inventories that feed from it. This focusing of the work programme was achieved using: the output of NAEI work on uncertainties, the material in Annex A of the contract technical specification, and an associated ranking system developed for this (EPG 1/3/195) programme (Appendix 1). A short discussion document, "Prioritisation for Year One", was prepared for ranking sub projects and determining success criteria (an extract is included in Appendix 2). Subsequent discussion with the Department led to the decision to concentrate work in Year One on the following: -

- 1. Simple desk-based studies
- 2. Fuel characteristics study
- 3. Characterisation of emissions of new POPs (PBDE, SCCP etc.)
- 4. Iron & Steel/Cement emissions study
- 5. Pesticide analyses
- 6. International collaboration
- 7. Emission factors for combustion

Wherever possible a numerical expression of uncertainty has been used. The principles of such an approach are described in Appendix 3.

1.3 PROGRAMME METHODOLOGY

Project EPG 1/3/195 aims to improve inventory quality by characterising and minimising the uncertainty in the emission factors used in the compilation of the NAEI. The factors that influence an inventory's general quality are its: transparency, completeness, comparability, consistency and accuracy – TCCCA factors. Any of these factors, singly or in combination, can improve or degrade inventory quality. EPG 1/3/195 objectives are directed to systematically examining and then working to strengthen these influences for the better. The work will, as a result, improve the confidence that an inventory user may place, at any one time, in the estimate of the mean emission factor - even if the mathematical expression of its uncertainty goes either up or down.

For the purposes of the emission factors work: -

- Improved *transparency* requires that the assumptions and methodologies used for deriving emission factors should be clearly explained so that users can replicate and assess the data suitability for their particular user requirement.
- **Completeness** requires that an emission source (and its identifier, code, description etc.) covers all known contributions to the emission that could reasonably be expected to be included. Completeness also covers those data aspects relating to speciation, geographical coverage, and temporal resolution.
- **Consistency** requires that any data used is internally consistent in all their elements with data from other years. Emission data are consistent if the same methodologies are used for all years and if consistent data sets are used to estimate emissions.
- *Comparability* requires that estimates of emissions are comparable with similar data reported by others. Ideally, data providers and reporters should use standardised, harmonised, methodologies and formats.
- Accuracy is a relative measure of the exactness of an emission estimate, and requires that essential data are systematically neither over nor under true emissions, as far as can be judged, and that actions have been taken to quantify and reduce uncertainties as far as practicable.

TCCCA influences change and evolve in the course of time as a result of circumstantial changes. The present work seeks to provide: -

- generic descriptive material to enable a user to decide whether inventory suitability criteria have changed to the point where action needs to be taken.
- > specific information to feed into the annual review of NAEI uncertainty review.

2 Simple Desk-based Studies

2.1 BACKGROUND

This task comprised small, simple, pieces of research or information purchases in order to improve generally minor parts of the NAEI. The task included the purchase of publications and the gathering of information from industry via telephone contacts with trade associations and industrial process operators.

A number of areas where research could be done was identified and inventory improvements have been proposed in the following areas: -

- Flat glass production (Cd, Cr, Pb, Se, Zn)
- Primary lead/zinc production (Cd, Pb, Hg, Zn)
- Carbon Tetrachloride manufacture (HCB)
- Car refuelling using unleaded petroleum (VOC)
- Nickel refining (Ni)
- Lime production (Ca)
- Manufacture of chromium based chemicals (Cr)
- Process emissions from crude oil refineries (Benzene)
- Non-aerosol car care products (VOC)
- Maturation of whisky (VOC)
- Glass fibre manufacture (Cr)
- Use of trichloroethylene as a cleaning solvent (VOC)

These items were all investigated except for the zinc production. Some further items of research have also been included when opportunities arose for improvements to the inventory to be made through this type of simple research.

The research has covered the following additional areas: -

- adhesives use (VOC)
- landfill gas combustion (PM₁₀, Benzene, NO_x, N₂O, CO, CH₄, VOC)
- sewage gas combustion (PM_{10} , Benzene, NO_x , N_2O , CO, CH_4 , VOC)
- pesticide use (POPs)
- manufacture of pressure sensitive tapes (VOC)
- tyre manufacture (VOC)
- chlorinated solvent manufacture (HCB)
- malting (VOC)
- creosote use (VOC)
- wood products manufacture (PM₁₀)
- alumina production

In addition, a briefing note on fireworks emissions was prepared. Further minor but worthwhile tasks have included obtaining copies of BREF reports, other publications such as company environmental reports, and miscellaneous background data (e.g. for identification and positioning of point sources) via the internet.

2.2 FINDINGS AND RECOMMENDATIONS

This task has brought about significant improvements to the completeness of NAEI and also the speciation and temporal resolution of sources. The work further enabled our ability to respond to 'ad hoc' enquiries from Defra. The number of work items undertaken under this task was greater than provided for in the original work programme. It is recommended that this task is continued for years 2 & 3 of the project.

2.3 IMPACT ON INVENTORY QUALITY

Table 1 summarises the changes in emission estimates made between the 2000 and the 2001 versions of the NAEI as a result of research carried out as part of this task. Table 2 shows the net change in national totals for selected pollutants as a result of these changes.

Source	Pollutant	Units	Emission	
			2001	2000
Glass (flat)	Zinc	tonnes		13.8
Glass (flat)	Nickel	tonnes		2.4
Glass (flat)	Mercury	tonnes		0.06
Glass (flat)	Chromium	tonnes		3.0
Glass (flat)	Copper	tonnes		0.75
Glass (flat)	Cadmium	tonnes		0.19
Glass (flat)	Arsenic	tonnes		0.15
Glass (flat)	Lead	tonnes		15.1
Glass (flat)	Selenium	tonnes	1.0	22.6
Glass (glass fibre)	Chromium	tonnes	3.2	2.9
Automotive products	VOC	ktonnes	26.8	31.9
Industrial adhesives	VOC	ktonnes	29.9	29.7
Incineration (chemical waste)	Arsenic	tonnes	0.17	
Incineration (chemical waste)	Carbon	ktonnes	311	
Incineration (chemical waste)	Cadmium	tonnes	0.01	
Incineration (chemical waste)	Carbon	ktonnes	0.099	
	monoxide			
Incineration (chemical waste)	Chromium	tonnes	0.09	
Incineration (chemical waste)	Copper	tonnes	0.12	
Incineration (chemical waste)	Hydrogen	ktonnes	0.03	
	chloride			
Incineration (chemical waste)	Mercury	tonnes	0.02	
Incineration (chemical waste)	Manganese	tonnes	0.11	
Incineration (chemical waste)	Nickel	tonnes	0.22	
Incineration (chemical waste)	Oxides of	ktonnes	0.33	
	nitrogen			
Incineration (chemical waste)	Lead	tonnes	0.38	
Incineration (chemical waste)	PM ₁₀	ktonnes	0.08	
Incineration (chemical waste)	Tin	tonnes	0.06	
Incineration (chemical waste)	Sulphur dioxide		0.32	
Incineration (chemical waste)	VOC	ktonnes	0.25	
Incineration (chemical waste)	Zinc	tonnes	0.007	
Incineration (animal carcass)	Carbon	ktonnes	3.3	
	monoxide			

Table 1. Changes in emission estimates resulting from Task 1.

Source	Pollutant	Units	Emis	sion
			2001	2000
Incineration (animal carcass)	Hydrogen	ktonnes	0.001	
	chloride			
Incineration (animal carcass)	Oxides of	ktonnes	0.25	
	nitrogen			
Incineration (animal carcass)	PM ₁₀	ktonnes	0.31	
Incineration (animal carcass)	Sulphur dioxide	ktonnes	0.17	
Incineration (animal carcass)	VOC	ktonnes	2.0	
Petrol stations (petrol delivery)	VOC	ktonnes	6.3	7.3
Petrol stations (spillages)	VOC	ktonnes	2.4	3.6
Petrol stations (storage tanks)	VOC	ktonnes	3.6	5.4
Petrol stations (vehicle refuelling)	VOC	ktonnes	39.5	56.0
Petrol terminals (storage)	VOC	ktonnes	3.7	4.2
Petrol terminals (tanker loading)	VOC	ktonnes	1.3	1.5
Petrol stations (petrol delivery)	Benzene	tonnes	22	38
Petrol stations (spillages)	Benzene	tonnes	12	19
Petrol stations (storage tanks)	Benzene	tonnes	18	28
Petrol stations (vehicle refuelling)	Benzene	tonnes	190	294
Petrol terminals (storage)	Benzene	tonnes	19	22
Petrol terminals (tanker loading)	Benzene	tonnes	6	8
Refineries (road/rail loading)	Benzene	tonnes	4	6
Petrol stations (petrol delivery)	1,3-butadiene	tonnes	0.8	1.7
Petrol stations (spillages)	1,3-butadiene	tonnes	0.5	0.8
Petrol stations (storage tanks)	1,3-butadiene	tonnes	0.7	1.3
Petrol stations (vehicle refuelling)	1,3-butadiene	tonnes	6.9	13.3
Petrol terminals (storage)	1,3-butadiene	tonnes	0.7	1.0
Petrol terminals (tanker loading)	1,3-butadiene	tonnes	0.2	0.4
Refineries (road/rail loading)	1,3-butadiene	tonnes	0.2	0.3
Chemical industry	VOC	ktonnes	70.8	71.1
Chemical industry (titanium dioxide)	Carbon	ktonnes	55.9	-
	monoxide			
Chemical industry	Benzene	tonnes	527	506
Chemical industry	1,3-butadiene	tonnes	264	270
Industrial adhesives (pressure sensitive tapes)	VOC	ktonnes	1.23	1.37
Tyre manufacture	VOC	ktonnes	1.75	1.71
Non-ferrous metals (nickel production)	Nickel	tonnes	5.3	5.1
Chemical industry (Trichloroethylene Prod)	НСВ	tonnes	23.9	54.1
Chemical industry (Tetrachloroethylene Prod)	НСВ	tonnes	20.7	32.6
Chemical industry (Prod Carbon Tetrachloride)	НСВ	tonnes	0	144
Wood products manufacture	PM ₁₀	ktonnes	1.41	-

Pollutant	% change	Pollutant	% change
Selenium	75%	Cadmium	2%
Lead	8%	VOC	2%
НСВ	7%	Nickel	2%
Benzene	4%	Carbon monoxide	1%
chromium	3%	Copper	1%
Zinc	3%	PM ₁₀	1%

Table 2. Change in national totals as a result of Task 1

Table 2 shows that the work carried out under task 1 has sometimes had considerable impact on the NAEI. It is worth noting that the research carried out under Task 1 consisted of a series of simple actions such as contacting industrial process operators. Given that each of these actions generally took only a matter of minutes to carry out, even the small changes in the carbon monoxide, copper and PM_{10} inventories are significant. Generally, Task 1 has led to increased accuracy of the inventory (e.g. changes in metal emissions from flat glass including removal of emissions of certain metals), and improved the completeness of the inventory (e.g. by adding emissions for chemical waste incineration and animal carcass incineration).

3 Fuel Characteristics Study

3.1 BACKGROUND

An in-depth survey of the metal and sulphur contents of solid and liquid fuels is on-going. A number of groups, including fuel suppliers, have been contacted to collect available data. Where fuel composition data are unavailable but representative samples have been obtained, the study has commissioned laboratory analysis of these fuel samples.

It is intended that such analysis data will be used to facilitate discussion with key stakeholders in order to agree suitable estimates of the mean level of specific components in the fuel, associated probability distributions and the range of sources to which the estimates may be applied.

The determinands being considered include S, Na, Ca, Mn, K, Cl, As, Cu, Cd, Cr, Hg, Ni, Mn, Be, Sn, V, Se, Zn, Pb; fuels include woods, coals, petroleum coke, petroleum products (principally fuel oil, gas oil, burning oil, DERV, petrol), SSF, coke, waste oils.

A significant amount of analytical information regarding metals and trace contaminants has been sourced for a variety of fuels, both domestic and imported. This has not been fully written up yet as it is not clear how best to use the information. A very limited set of fuel descriptors is currently used in the NAEI and a decision is needed on whether it is desirable to expand that set or whether to develop aggregate factors.

A review of the most immediate data needs of the NAEI suggest that the solid-fuel information can be most readily used to estimate residential or small-scale combustion emissions in the absence of measured emission data (larger combustion sources have abatement of varying types which make use of emission factors based on fuel data far more uncertain) and this is where effort is being focussed.

Some of the emission issues for small-scale combustion have been addressed within Task 7 ("Review of combustion emission factors"), but several issues remain for pollutants (especially heavy metals) from residential sources. Analysis of liquid and other fuels will provide further benefits to the NAEI and representative samples are currently being sought.

This task has undergone a major change of focus since inception and requires formative input from the NAEI before final reporting can commence. However, it is likely that reporting can be finalised by the end of September 2003.

3.2 IMPACT ON INVENTORY QUALITY

The work carried out under Task 2 is not yet complete, however, the potential for change in NAEI estimates can be seen from Table 3 which shows the contribution to national emission totals from burning of liquid and solid fuels which are the subject of this research. Changes in emission factors for solid and liquid fuels resulting from Task 2 would clearly have a major impact on national emissions of many pollutants.

Pollutant	% of total	Pollutant	% of total
Beryllium	99%	Selenium	80%
Tin	98%	Arsenic	72%
Hydrogen chloride	98%	Copper	59%
Vanadium	93%	Lead	41%
Sulphur dioxide	91%	Cadmium	38%
Manganese	90%	Chromium	37%
Hydrogen fluoride	88%	Mercury	36%
Nickel	87%	Zinc	25%

Table 3. Contribution to national totals of combustion of liquid and solid fuels

4 Characterisation of Emissions of New POPs

4.1 BACKGROUND

The purpose of this study was to improve the National Atmospheric Emissions Inventory (NAEI) estimates of selected Persistent Organic Pollutants (POPs) currently (or likely to become) subject of international agreements. The Persistent Organic Pollutants addressed in this study are;

- Polychlorinated Terphenyls (PCTs)
- Ugilec
- Hexachlorobutadiene
- Pentabromodiphenyl ether (PentaBDE)
- Pentachlorobenzene
- Short Chain Chlorinated Paraffins (SCCPs)

A range of organisations with experience of the use or production of these pollutants was contacted. Data on consumption and emissions of these Persistent Organic Pollutants was gathered from contacts in industry, Trade Associations, producers and information collated from existing research.

4.2 FINDINGS AND RECOMMENDATIONS

Currently the following pollutants are not included in the National Atmospheric Emissions Inventory:

- Polychlorinated Terphenyls (PCTs)
- Ugilec
- Hexachlorobutadiene
- Pentabromodiphenyl ether (PentaBDE)
- Pentachlorobenzene

Short Chain Chlorinated Paraffins (SCCPs) are included within the NAEI emissions estimates. The 2000 inventory (1970 - 1999) included estimates for this pollutant class but there is a high degree of uncertainty associated with the estimates.

Based on the Task 1 study, the NAEI POPs estimates have been reviewed and the following recommendations prepared for the 2001 inventory (1970 - 2000):

Polychlorinated Terphenyls, Ugilec, and pentachlorobenzene : are no longer produced or used in the UK and there are regulations in place to control their identification and destruction. Retrospective inclusion of these species in the NAEI for prior to 2000 is not recommended as it would require substantial effort to obtain data and such data is likely to have a high uncertainty.

Hexachlorobutadiene: While hexachlorobutadiene is no longer deliberately produced or used in the UK it does exist in certain landfills and may in the future be found to be a fugitive emission. It is recommended that the situation is reviewed in a year's time.

Pentabromodiphenyl ether and short chain chlorinated paraffin_emissions have been re-evaluated and SCCP emissions have been updated in the NAEI. Information on pentabromodiphenyl ether is poor and inclusion in the NAEI at the present time is not recommended. However a watching brief should be kept pending new regulation and improved production/use information becoming available.

The information obtained in the study has improved the completeness of the NAEI. The more detailed and accurate information on production and use collected on several of these POPs has made it possible to exclude them from the inventory as they have never been, or are no longer, used/allowed in the UK. The data has enabled a more consistent time series covered by the NAEI for SCCPs. A review of Pentabromodiphenyl ether emission data is recommended once proposed legislation covering use of this material is introduced.

As this is a continuing area of interest, with extensions to the Convention on Long Range Transport of Air Pollutants (CLRTAP) Protocol on POPs currently planned, it is recommended that this task continue into years 2 & 3.

4.3 IMPACT ON INVENTORY QUALITY

Research carried out under Task 3has had the following impacts on the quality of the NAEI:

- emission estimates for short-chain chlorinated paraffins have been reduced for all years covered by the NAEI. For 2001, the new estimate is 0.6 kg which is less than 1% of the estimate (62kg) that would have been included had this research not been undertaken. The accuracy of the NAEI has been substantially improved as a result.
- the research has confirmed that polychlorinated terphenyls, ugilec, and pentachlorobenzene are no longer produced or used in the UK. While this will not result in a change in the NAEI, nonetheless, the NAEI can be said to be more complete and accurate in the sense that emissions of these pollutants have been shown to be zero.
- the research has confirmed that, although it is no longer produced, there is a
 potential for emissions of hexachlorobenzene and that further work should be carried
 out. The work has improved the quantity of information available to the NAEI and
 has helped to prioritise further work.
- the research has produced an estimate of emissions of pentabromodiphenyl ether of 24 kg per year, which compares with an emission estimate of 13.8 tonnes for all polybrominated diphenyl ethers in the NAEI. The estimate for pentabromodiphenyl ether is subject to high uncertainty and has not been included in the 2001 version of the NAEI. Nevertheless, the research has gone some way towards improving the completeness and accuracy of this aspect of the NAEI.

5 Review of Pollution Inventory Data

5.1 ENVIRONMENT AGENCY POLLUTION INVENTORY

The NAEI data for UK industry covered by IPC and IPPC are largely drawn from annual emission reports provided by site operators in England and Wales to the Environment Agency Pollution Inventory (PI). Consequently the continuing comparability of the PI and the NAEI is critical to the quality of the NAEI. This task comprises a review the Pollution Inventory data for the cement and iron & steel industry sectors.

5.2 CEMENT EMISSIONS STUDY

5.2.1 Background

The report highlights potential issues in the PI annual summaries and the industry view. Conclusions are provided on the use of PI data within the NAEI for the cement sector and recommendations for further investigation are provided.

In addition, default emission factors have been derived which may be used in the NAEI (and other inventories) to assess the data provided for certain key pollutants released to the atmosphere from cement installations. Default factors also allow provision of data where source specific data are not available.

5.2.2 Summary of Findings and Recommendations

The accurate mapping of PI sites into the appropriate part of the NAEI contributes to the completeness and transparency of the NAEI. This review suggests that industry sectors used by the PI and NAEI are not consistent and indicates that there is a need for the NAEI project to review either the NAEI categories and/or the mapping between PI (and other inventories) and the NAEI.

Sites outside England & Wales do not report to the PI and it is recommended that the operator and regulatory authorities should be contacted through a task in the emission factor project to obtain source specific data for these sites for incorporation into the NAEI.

A number of incidences of missing or variable data were noted in the PI but this is likely to diminish as operators develop robust reporting procedures and can probably be addressed by the NAEI in an 'ad hoc' fashion, as required.

Development of formal procedures in the NAEI for assessing anomalies in the PI data is recommended.

Reporting of the PM_{10} emission to the PI is inconsistent and, until the PI reports for cement manufacture are reported on a consistent basis, it may be appropriate for the NAEI to adopt a default emission factor for PM_{10} of 100% of particulate emission.

A mass balance approach is considered to be the most reliable method for the operator to estimate CO_2 emissions from cement manufacture and is consistent with the approach adopted for other estimates for CO_2 within the NAEI.

Dioxins emission returns appear to be largely based on measured data however, the results have a high uncertainty and there will be some inconsistency between sites in the way dioxins data are aggregated to estimate annual emissions.

Comparison of reported benzene emissions against estimated production suggests a default emission factor of 0.012 kg benzene per tonne clinker, with an estimated uncertainty of $\pm 100\%$. However, some sites have relatively high emissions and the need for improved source specific data is indicated.

It is recommended that further work be undertaken under this programme with the cement companies to assess more thoroughly the metal emission data, especially mercury, cadmium and copper.

PAH emissions are a relatively new reporting requirement for the cement sector and some of the PI inconsistencies are believed to be temporary. It is recommended that further work be undertaken under this programme to assess the available measured data and derive default PAH emission factors.

5.3 IRON & STEEL

5.3.1 Background

In the NAEI, the iron and steel sector is a significant source of metals and persistent organic compounds. Most of the iron and steel works are regulated by the Environment Agency and report their annual emissions to the Pollution Inventory. The NAEI relies heavily on the PI for this information and it is important to understand the nature of the industry reporting in order to appreciate the effect of any uncertainty on the overall quality of the NAEI.

A thorough analysis was undertaken of past NAEI communications with the industry and the PI returns. Meetings were held with sector representatives to aid our understanding of estimation methodologies and review the NAEI's current assumptions.

5.3.2 Summary of Findings and Recommendations

The final report is awaiting clarification from industry on several key areas of emission estimates and is not yet complete. However similar issues were determined as for the cement sector.

Revised default emission factors are recommended for the following: -

- > Zinc, lead, chromium and vanadium for several process stages
- PCB emissions from electric arc furnaces are believed to be over-stated and a revised figure is proposed

Emissions of dioxins and PAH were confirmed.

The emission data sets of the industry are being improved as the result of considerable work by industry and it is recommended that, under this programme, the sector be revisited as a 'simple desk study' in the near future.

5.4 IMPACT ON INVENTORY QUALITY

Work carried out under Task 4 has had a number of impacts on the quality of the NAEI. These are:

 it has provided an opportunity for contacts to be made with operators in two important industrial sectors, allowing NAEI data to be reviewed by industry representatives;

new data and, in particular, background information on the sectors, has been obtained which will lead to improvements in the completeness and accuracy of the NAEI.

6 Pesticide Analyses

This study will estimate the levels of POPs in certain pesticides used in the UK and collect current data on pesticide use; some analytical work is anticipated.

A pesticide-usage database has been obtained for the period since 1990 from the Pesticide Usage Survey and this will allow improved activity statistic information to be provided in the 2002 NAEI of POPs. The database is particularly useful as it provides some indication of usage trends.

However, progress elsewhere in this task has been less encouraging. Sourcing of pesticide samples has proved difficult. To resolve this, a consultant with particular expertise in the pesticides sector is being employed to review the pesticide production mechanisms to determine likely POPs occurrence. The output of this review will then inform the selection process for samples of pesticide and, the analysis required.

7 International Collaboration

A bilateral meeting has been held with the Dutch Ministry of Environment (VROM) inventory team and the UK/EA PI team to exchange information and co-ordinate inventory development programmes. The aim is to pool resources and information to prevent duplication of effort and promote data quality. A number of areas of immediate co-operation were identified, these included exchange of information on firework, metal coating, and pesticide contamination. An agreement was made to share information on shipping emissions as information becomes available from an on-going Dutch study.

Information from past and present emission factors programmes is being fed into the UNECE TFEIP C&I Expert Panel and also into the WG on Strategies and Review Expert Group on Techno-economic Issues (Egtei). Both these groups have information exchange activities that benefit the current programme. Such informal exchanges have led to the provision of data on the composition of fuels and emission factors from small combustion plant for Tasks 3 and 4.

Within the context of the TFEIP C&I panel and CAFÉ the EU's Joint Research Centre has established an Emissions Unit. Information from the current programme has been used to assist the JRC focus it's own research work. This is expected to lead to substantially improved emission factors for PM, POPs and heavy metals from small combustion within the lifetime of this contract.

8 Combustion Emission Factors

8.1 SMALL COMBUSTION SOURCES

Combustion sources, particularly large sources such as power stations and waste incinerators, have received considerable attention in the past but review is now necessary. Smaller combustion sources (units of less than 20 MW net thermal input) such as domestic fires, domestic boilers, commercial heating appliances and CHP units, are characterised by a very limited set of source descriptors and a correspondingly small number of emission factors. Consequently, the transparency of the calculations from this emission source is poor and it is difficult to assess the quality of the estimation procedure used.

Research into emissions from smaller combustion sources is sparse and significant variability is evident in the emissions data that are available. In addition, the application of combustion control and emission abatement technologies is not uniform across the sector due to economic and practical considerations. Domestic fireplaces, stoves and poorly-maintained central heating units may burn inefficiently. Across this sector, therefore, significant emissions of pollutants such as PM_{10} , carbon monoxide (CO) and organic species such as dioxins and furans and polycyclic aromatic hydrocarbons (PAH) may be produced.

A desk-based study was conducted to collate and review data and background information in two broad areas: -

- 1) Emissions of key pollutants from a range of different types of small-scale combustion device, incorporating inputs for different fuels where possible, to include: coal, coke, smokeless solid fuels, wood, oil & gas.
- 2) The distribution of individual types of small-scale combustion device by fuel type within the UK.

This task aimed to achieve the following steps towards improvement of the NAEI data for the small-scale combustion sector: -

- Identify the range of commonly used combustion appliances across the different subsectors of the UK small-scale combustion market.
- Improvement of the estimation of fuel use allocation across the different combustion appliances identified, to enable more accurate distribution of DUKES annual UK fuel use data within the sector.
- Identify the fuel & appliance combinations, by pollutant, which most influence the uncertainty of the 'aggregate' emission factors used in the inventory and focus the study in these areas.
- Examine the adequacy of available data for each of the different sub-sectors of the UK small-scale combustion market.

8.2 SUMMARY OF FINDINGS AND RECOMMENDATIONS

Revised emission factors are recommended for: -

• PM₁₀ and NO_x factors for domestic coal combustion

- PM₁₀ factors for coke combustion
- PM₁₀ and CO emissions for domestic combustion of natural gas
- Benzo(a)pyrene emissions for domestic wood combustion

The report further recommends focusing on the improvement of emission factors for: domestic firing of natural gas, burning oil and coal and also commercial and agricultural use of gas oil. These sources are the components which form the bulk of the composite emission factors used in the NAEI; they also contribute most to the uncertainty budget. The review of published information on emission factors has been used to identify the values most suitable for the UK in the absence of any UK sourced information.

The inventory contains a limited subset of emission factors and two approaches can be taken to improving small combustion plant emission estimation in the NAEI:

- 1. expand the set of NAEI emission factors allowing a more representative mix of plant and fuels to be represented.
- 2. adopt a 'calculator' based approach whereby a spreadsheet-based programme calculates, on the basis of fuel use, knowledge of the equipment 'pool', and the most up-to-date emission factor information the current most representative 'emission factor for use in the NAEI.

8.3 IMPACT ON INVENTORY QUALITY

The research carried out under Task 7 should be considered as the first stage of a process of improvement of a major source of air pollution - small combustion plant. At this stage it is not possible to quantify exactly how much emission estimates could change as a result of this work since much remains to be done, either extending the level of detail in the NAEI or producing an 'emissions caclulator'. The approaches recommended in this report could also be extended to medium sized combustion plant i.e. plant upto 50MWth in size leading to further, as yet, unquantified improvements in inventory quality. This would also help to improve estimates for large combustion plant since, if these plant were treated separately in the NAEI, then emissions data for these plant, reported by the Environment Agency, Scottish Environment Protection Agency and DOE (Northern Ireland) could be used as the basis of estimates for these sites. The work started with this report could have far-reaching and significant impacts on the quality of the NAEI.

For the moment, some indication of the scale of change in the NAEI that might occur as a result of this process, can be derived by examining the change in emission totals that would occur if emissions from small combustion were estimated using emission factors recommended in this report rather than current NAEI emission factors.

Table 4. Toternal changes in twice estimates add to rask 7 research			
Pollutant	Change	National total	% change
Carbon	+ 1303 ktonnes	3758 ktonnes	+35%
monoxide			
Oxides of	+17 ktonnes	1681 ktonnes	+1%
nitrogen			
VOC	- 3 ktonnes	1514 ktonnes	0%
PM ₁₀	- 12 ktonnes	191 ktonnes	-6%
B[a]P	+ 0.02 tonnes	10.47 tonnes	0%
Dioxins	- 9 grammes	357 grammes	-2%

Table 4. Potential changes in NAEI estimates due to Task 7 research

The changes shown in this table are based on a simple substitution of one set of emission factors for another, rather than the radical overhaul of the estimation methodology used in the NAEI for combustion sources that has been recommended here. The figures given can therefore only be a guide but suggest that, for certain pollutants such as carbon monoxide and PM_{10} and, to a lesser extent NOx and dioxins, this research could lead to major changes in national emission totals.

9 Summary of Project Reports

Details of the project reports and other deliverables are summarised below.

Item	Task	Details
Review of NAEI for project	Identified the project tasks	Attached at Appendix 2
research areas		
Task Report	Simple desk-based studies	AEAT/ENV/R/1421
Task Report	Emissions of new POPs	AEAT/ENV/R /1363
Task Report	Review of Pollution Inventory :	
	Iron and Steel	In preparation
	Cement	AEAT/ENV/R/1425
Task Report	Emission factors for combustion	AEAT/ENV/R/1407
Annual summary report	Annual report	AEAT/ENV/R/1436

Reports for the fuel characterisation and pesticides study tasks will be prepared on completion of the tasks.

Appendices

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- Appendix 1 Prioritisation Methodology
- Appendix 2 Prioritisation for Year One
- Appendix 3 Expressing Inventory Uncertainty

Appendix 1 Prioritisation Methodology

Numerous activities could be undertaken that would result in some improvement of the emission factor data used in the NAEI. The decision mechanism adopted to evaluate each improvement option scored alternatives on each of four parameters: -

Technical

Each option was scored depending upon the perceived uncertainty in the existing emission factor. For each pollutant we propose that a target uncertainty, i.e. an acceptable level of uncertainty, be defined in consultation with the NO. For example, a target of +/- 20% might be set for the NO_x inventory, in which case an emission factor which is believed to be accurate to +/- 50% would score highly, while one that is +/-25% might only be given the lowest score.

Feasibility

Each option was scored depending upon an assessment of the likelihood that research could significantly reduce uncertainty in the emission factor. For example, a low score might be given to a measurement campaign where the technical difficulties in gathering reliable data are high, whereas a high score might be given to a desk-based exercise which involved the collection and analysis of data held by public bodies.

Administrative

Each option was scored depending upon an assessment of the extent to which improvement of the emission factor could aid Defra in the development of regulations and legislation. For example, improvement of an emission factor for a source that is the subject of new legislation would score highly, whereas research relating to sources which are not covered by current discussions or negotiations would score low.

Policy

Each option was scored depending upon an assessment of the importance of improvements to the emission factor in informing Government response to public concerns or large or immediate environmental impacts. For example, recent work to monitor emissions from animal pyres would have scored highly, whereas work to address uncertainty in an area where environmental concerns were considered to have been fully addressed by existing policies. The total score can then be modified depending upon the estimated cost of the option, with expensive options being marked down and cheap options being assigned extra points. In this way, options which score high on the first stage of prioritisation can still be rejected if too expensive, whereas lower scoring, but cheap, options can be recommended.



DECISION/NECHANISM - mark 3 for high, 2 for medium, 1 for low importance

Appendix 2 Prioritisation for Year One

Introduction

This note gives details of options for research into emission factors and other inventory-related data to be funded under the Defra research programme 'Emission factors and other data for inventory development', reference EPG 1/3/195.

Prioritisation of Research Tasks

Prioritisation of research tasks has been done according to the method described in Appendix 1. Some modifications have been made to the input data. In particular, the ratings for each pollutant have been revised as shown in Table 1.

Table 1: Pollutant ratings used in the prioritisation

Pollutants	Rating
VOC, benzene, 1,3-butadiene, NH ₃ , CO, PM ₁₀ , As, Cd, Hg, Ni,	High
Pb, benzo[a]pyrene, HCB, HCH, dioxins, PCP, SCCP, PBDE	
NOx, SO ₂ , Cu, Se, V, Zn, PCN, PCB, Sn, Be, Mn, Ca, K, Mg, Na	Medium
HCI, Cr	Low

The prioritised research tasks have then been grouped into 'projects'. Grouping has been done where two or more tasks are more sensibly carried out in parallel by the same researcher e.g. where two or more tasks involved contacting the same trade association.

Research Options

The research options are described below in <u>descending</u> order of cost effectiveness.

- 1. Simple desk-based studies. A group of tasks involving simple requests for data from industrial contacts.
- 2. Survey of fuel characteristics. An in-depth survey of fuel composition covering the metal and sulphur contents of solid and liquid fuels. The

study may include some analysis of fuels should representative samples be obtained.

- 3. Use & emissions of short chain chlorinated paraffins, pentabromo diphenylether and other persistent organic pollutants. A study involving the collection of data from industry contacts regarding the consumption and emissions of these pollutants.
- 4. Review of iron & steel sector emissions. Emissions from metallurgical coke production, integrated steelworks and electric arc furnaces are largely based on Pollution Inventory data. This study will obtain information on the methods used to calculate these data, derive uncertainty estimates for them, and will provide data to fill gaps in the existing data.
- 5. Analysis of pesticides & pesticide use survey. This study will involve analytical work to measure the levels of persistent organic pollutants in certain pesticides used in the UK and will also collect new data on the levels of pesticide use.
- 6. Co-operative work. This project will involve the dissemination of results from Defra-funded inventory work and the collection of data funded by others. In particular, this study will continue existing links with inventory specialists in other countries.
- 7. Collection and review of stationary combustion emission factors. Emission factors for combustion processes are very important since stationary combustion processes are an important source of almost all pollutants in the NAEI. The factors used in the NAEI have been reviewed periodically in the past, however further work is needed as technology changes and ever greater numbers of measurements are made. This study will collect emission factor data for VOC, CO, NOx, PM₁₀, persistent organic pollutants, and NH₃ and will analyse the available data in order to make recommendations about emission factors for the NAEI and input data for uncertainty analysis.
- 8. Measurement of emission rates from impregnated wood. This project, which would probably be best sub-contracted out, would involve the measurement of emissions of persistent organic pollutants would impregnated wood.
- 9. Burning of treated wood. This study would aim to collect more data on the quantity of wood treated with preservatives which are burnt either as fuel or as a means of waste disposal.
- 10. PCBs in capacitors. This study would involve the collection of data on the quantities of PCBs and other persistent organic pollutants present in electrical equipment such as capacitors.
- 11. Review of mercury emissions from crematoria. This study will involve consultation with crematoria operators and other groups and will make recommendations for any revisions necessary to the mercury emission factor in the light of available data.

12. Sources of benzene, 1,3-butadiene and metals in the chemical and nonferrous metal sectors. This study involves the collection of data from the Environment Agency, SEPA and process operators in order to improve estimates from these sectors.

Table 2: Research projects Ranked in order of cost effectiveness

No.	Title
1	Simple desk-based studies
2	Survey of fuel characteristics
3	Use & emissions of SCCPs, PBDE & other new POPs
4	Iron & steel sector emissions
5	Analysis of pesticides
6	Co-operative work
7	Review of combustion emission factors
~	

- 8 Measurements impregnated wood
- 9 Burning of treated wood
- 10 PCBs in capacitors
- 11 Review of mercury emissions from crematoria
- 12 Benzene, 1,3-butadiene & metals from the
 - chemical & non-ferrous metals sectors

Table 3: Estimated impact of proposed research projects on inventoryuncertainty

No.	Title	
1	Simple desk-based studies	Se (18%), Cd (7%), Cr (4%), HCB, Pb (2%), Hg, Zn, VOC (1%)
2	Survey of fuel characteristics	Be, Mn, Sn (25%), V (19%), Ni (16%), SO2, HCI (10%), As (6%), Mg (5%), Cu (4%), NOx (3%), Na, Cr, Hg (2%), Zn, Pb (1%)
3	Use & emissions of SCCPs, PBDE & other new POPs	PBDE, SCCP (20%)
4	Iron & steel sector emissions	Zn (10%), Na (7%), Pb (6%), Cu (4%), Cr (3%), Cd, K, CO (1%)
5	Analysis of pesticides	HCB (18%)
6	Co-operative work	All pollutants (2%)
7	Review of combustion emission factors	All pollutants (2%)
8	Measurements - impregnated wood	PCP (20%), HCH (19%)
9	Burning of treated wood	As (13%)
10	PCBs in capacitors	PCB (19%)
11	Review of mercury emissions from crematoria	Hg (7%)
12	Benzene, 1,3-butadiene & metals from the chemical & non-ferrous metals sectors	Cu (5%), Hg (3%), 13BD (2%), Cr (1%)

Appendix 3 Expressing Inventory Uncertainty

The annual NAEI reports includes quantitative estimates of uncertainty in the national emission total for each pollutant. At first sight there is very little change in the assessment of uncertainty between the 1996 NAEI report and the 1999 NAEI report, this gives the false impression that the NAEI has not improved despite the considerable research into emission factors carried out in the meantime. In reality the inventory is more complete, detailed and accurate now than in 1996; it is more useful and national emission totals are more likely to be 'right'. The problem is presentational, the current expression of uncertainty lacks rigour in its use of quasi-statistical terminology and so it is not capable of reflecting improvement. A more complete expression of uncertainty is needed – a short overview of the statistical issues is given in Annex 1 below.

As part of the programme of work to maintain the NAEI a more detailed assessment is being made of the uncertainty in the NAEI using software better able to manipulate and display statistical information. By virtue of Defra funded work it is becoming possible to generate the data to determine the statistical uncertainty in emission factors and activity data at the detail required for the analysis described in Annex 1. This detailed approach will provide a more quantitative measure of uncertainty which, if repeated each year, is suitable for the description and monitoring of certain types of improvement. An improved expression of uncertainty would be a presentation of the mean value of uncertainty at a prescribed confidence level together, possibly, with a measure of its dispersion, the standard deviation:

> Total emission = ZZ +/- zzat the 95% confidence level with a standard deviation of zz

This more rigorous expression of 'Uncertainty' differs from that used in the NAEI reports. In the past we have had to use educated guesses of the maximum uncertainty based on conventional wisdom and a limited data set. In many cases we are only just becoming able to describe the distribution of possible values and consequently it has not been possible, until recently, to determine dispersion (σ) and consequently to assess the uncertainty (u) at a specified confidence level.

The comments above all concern the expression of numerical uncertainty. Further, different types, of analysis and indicators may be needed to examine the uncertainty in other parts of the NAEI output e.g. speciation, mapping, and also to deal with issues such as completeness, consistency and comparability of the inventory.

Notwithstanding the important advances being made in our knowledge of the statistical nature of the uncertainty in the NAEI we should not necessarily expect dramatic changes from current estimates of the 'headline' uncertainty figures - a trained eye can make a good guess. This can be illustrated using NMVOC; a considerable improvement of the inventory has occurred since 1996, and the assessment of the uncertainty has changed from +/-50% to +/-30%. A large amount of further research could be needed before we might be able to **demonstrate** a further reduced uncertainty. This is illustrated in the table below:

Contribution of source	Number of sources in	Total contribution of
to UK total emission	category	sources in category
0-0.1%	163	4%
0.1-0.5%	61	16%
0.5-1%	17	12%
1%-2%	19	34%
2%-3%	6	24%
5-10%	0	0%
> 10%	1	10%
All sources		100%

Table 1: Analysis of the 1999 NMVOC inventory by size of source

Most of the estimates in the NMVOC inventory are probably accurate to between +/- 10% and +/- 50%. Few single sources, however, contribute more than 2% of UK emissions. Focussed research to improve emissions for a single sector will have a limited impact on uncertainty in the inventory. Broad research on the large number of sources comprising the larger proportion of the inventory is equally, if not more, important. For example, research on the largest 43 sources would bring some improvement to 80% of the inventory. But in most cases the estimates for these larger sources have already been the subject of research and there is little more to gain. It is now the smaller sources which are now the most uncertain. The situation for other pollutants is less extreme but similar – the few large sources have been the subject of most research while emission estimates for the more numerous small sources are most uncertain. AEAT suggestions on how to optimise research work are included in proposals under consideration by the Department.

It was mentioned above that the current practice of quoting uncertainty does not reflect all the improvement work done. Some examples of these difficult areas are given below:

• Maintaining a steady rate of improvement: New emission factors must often be developed *in order to maintain the level of controlled uncertainty in the NAEI*. Emission factors used for previous years may no longer appropriate when, for instance, the level of emission control has increased.

An example of this is the collection of data on NMVOC emissions from film coating processes for the 1999 NAEI. This research allowed us to update the emission factors used for this source, taking account of abatement systems installed by process operators in the previous couple of years. Our current best guess that the emission estimate for this particular sector is +/-10% is probably no different from what would have been guessed two years ago. Had we not done the research we would, however, have estimated the uncertainty to be significantly higher, perhaps as much as +/-30%.

- Improving levels of detail: Improvements are made to the speciation of emissions, to the mapping, and to the temporal disaggregation of emissions. These are very important outputs from the NAEI, and the emission factor research programme has included a considerable amount of work to improve them, however the current assessments of uncertainty do not address them.
- Improving completeness: Previously omitted sources are included, or estimates are made at a greater level of detail. In the first case, the uncertainty due to the omission of the source in previous years may not have been taken into account.
- **Extension:** New pollutants are added, necessitating research to generate emission factors.

A confounding factor is that research to improve inventory confidence also increases the effort needed to maintain current levels of uncertainty. Such work leads to the identification of sources of uncertainty, previously excluded from assessments, as the result of improvements in knowledge and the acquisition of further data. Some examples of this type of issue are given below:

- In some instances where measurement work has been carried out in the past with the expectation that, by increasing the sample population, uncertainty would diminish, the opposite has been true since the new data are outside the range of values found previously.
- New sources have been identified and emission estimates included in the NAEI leading to an increase in uncertainty if the estimate for the new source is very uncertain. An example of this might be the inclusion of emissions of dioxins from small-scale waste burning in the 2000 NAEI. Previously omitted from the NAEI and from previous assessments of uncertainty, the new estimate of emissions from this source is likely to be very uncertain and will increase uncertainty in the inventory.
- Sometimes, research indicates that assumptions used in previous years are erroneous or provides evidence of problems in methods used to estimate emissions. For example, discussions with the British Rubber Manufacturers Association (BRMA) in recent months highlighted both a double-count in NMVOC emissions in the sources 'textile coating' and 'other rubber goods' and an omitted source (tyre retreading). Both sources of error are likely to be very small but despite research effort, have not been resolved due to lack of data and thus increase uncertainty in the NMVOC inventory.

Annex 1- The expression of Uncertainty and Levels of Confidence

Uncertainty

The ISO Guide to the Expression of Uncertainty in Measurement (GUM) defines 'uncertainty' as: 'the parameter, associated with the result of a measurement, that characterizes the dispersion that would reasonably be attributed to the measurand'. This concept of statistical dispersion is also useful when considering a distribution which is the result of an aggregation of parameters which are themselves 'distributions', such as emissions from a family of sources (sectors), emission factors and, in some cases, activity statistics.

Confidence

The usual method of determining the level of uncertainty of a given measurement/estimation is to determine its combined standard uncertainty, u_c, by combining all the individual, component, standard uncertainties. It is then necessary to provide a measure of this aggregated uncertainty that defines an interval about the measurement/estimation result that is expected to encompass a large fraction of the distribution of values that could reasonably be attributed to the measurement. This is illustrated diagrammatically in Figure 1. This measure of uncertainty is termed the expanded uncertainty, denoted by U, which is obtained by multiplying the standard uncertainty by a coverage factor k;

 $U = k u_c$.

The result of the measurement is then traditionally expressed as;

 $Y = y \pm U$,

This is interpreted to mean that the best estimate of the value attributed to Y is y, and also that y-U to y+U is an interval that may be expected to encompass a large fraction of the distribution of the Y value.

More specifically, U may be interpreted as defining the interval about the measurement/estimation result that encompasses a large fraction, p, of the probability characterized by that result and its associated uncertainty. This fraction, p, is then the 'level of confidence' of that interval. If, for example, the level of confidence, p, is defined as 95% - then 95% of all the individual measurement parameters lie within \pm U of the best estimated value, y, and only 5% lie outside of this interval.

The level of confidence (at whatever level chosen) must be stated together with its expanded level of uncertainty. It should also be noted, however, that the coverage factor, k, which is used as a multiplicand with u_c to obtain an appropriate level of confidence depends on the number of independent (uncorrelated) determinations of that measurand (ie the number of degrees of freedom), which make up the probability distribution (as described in the GUM). Therefore k=2 can be used only when there are a sufficiently large number of such degrees of freedom.

The relationship between Confidence Interval and Uncertainty

The confidence interval of a statistical distribution of measurement results is illustrated in figure 1, where:

$T_1 = \theta - 1.96 \sigma$	lower confidence limit for a large number of degrees of freedom - expressed in this example at a level of confidence of 95%.
$T_2 = \theta + 1.96 \sigma$	upper confidence limit for a large number of degrees of freedom - expressed in this example at a level of confidence of 95%.

and σ is the normally defined standard deviation (uncertainty) associated with the statistical population distribution.





The length of the confidence interval, I, defined in the GUM, for a large number of degrees of freedom expressed at a level of confidence of 95%, is then:

 $I = T_2 - T_1 = 2 \times 1.96 \times \sigma$

Thus the standard deviation (uncertainty) associated with this confidence interval, I, is then:

$$\sigma = \frac{1}{2 \times 1.96}$$

This then enables the expanded uncertainty, U, associated with the confidence interval, I, to be expressed as:

U = 1.96 x σ = 0.5 x I (for a large number of degrees of freedom, expressed at a level of confidence of 95%).

Assessing and describing the 'uncertainty' of an inventories

The bulk of an emission inventory is compiled by collecting activity data and appropriate emission factors according to

 $Emission_{pollutant} = \sum_{activitiess} Activity \ rate_{activity} \times Emission \ factor_{activity, pollutant}$ (1)

Although for some sectors the equation to be used to estimate emissions is more complicated the calculation is not essentially different.

For both variables and parameters quantitative uncertainty ranges are needed to enable a quantitative uncertainty analysis. The means of assessing these will differ depending on whether Measurement, expert judgement, or literature references have been used to collect data.

The TFEIP Guidebook describes the statistical quantification of inventories, the aggregation of uncertainty and the expression of inventory in much more detail.