A REVIEW OF POTENTIAL PCDD AND PCDF EMISSION SOURCES IN THE UK

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1. *INTRODUCTION*

Estimates of annual PCDD and PCDF emissions to atmosphere from industrial and non-industrial sources in the UK have been presented in the past[1,2,3]. However, the coverage of potential sources has been very limited, and the emissions have been reported in a variety of units which are not readily translated to either a common format, or to the internationally recognised NATO/CCMS toxic equivalent scheme. The UK Government therefore commissioned a study to update previous estimates of PCDD/F emissions to atmosphere as part of an ongoing programme to characterise and regulate emissions from industrial processes. The objectives of the study were the following:

- To systematically screen the range of industrial processes operating in the UK for the potential to release PCDD/Fs to atmosphere;
- To identify those industrial processes which could potentially release PCDD/Fs to atmosphere and to quantify annual emissions from these sources;
- To also estimate annual PCDD/F emissions from a selection of non-industrial thermal sources;
- To assess future trends in PCDD/F emissions in the UK.

This paper presents the outcome of the study, which incorporates data available up to the end of February 1995. In addition to the inventory, the paper also discusses the limitations and data gaps which contribute to the uncertainty in the annual emission estimates.

2. *APPROACH*

Industrial processes were divided into two categories:

- *Chemical processes*, in which reactions are generally in the liquid phase and at temperatures below 250°C. Generally, the product is retained in the reaction vessel and PCDD/Fs, if formed, have a greater potential to be released to water and land than to atmosphere.

- *Thermal processes,* in which temperatures are typical of combustion-related processes (800°C and above) but with the potential for some parts of the process train to be maintained at lower temperatures.

Applying criteria developed to screen processes for their potential to form and emit PCDDs and PCDFs to atmosphere[4,5] 23 industrial process categories were shortlisted for further consideration as potential sources of PCDD/F emissions to atmosphere. These processes are listed in Table 1. All of the non-industrial sources considered in this study fell under the definition of *thermal processes* and satisfied the criteria for potential release of PCDD/Fs to atmosphere.

For each shortlisted process, emission factors were identified, expressed in terms of the quantity of PCDD/Fs emitted per tonne of material produced or treated. In the case of crematoria and traffic, the emission factors were expressed in terms of the quantity of PCDD/F emitted per cremation and per kilometre travelled respectively. In the majority of cases emission factors were obtained from the literature. In others, an emission factor was derived from the concentration of PCDD/Fs in the gases released to atmosphere, making assumptions as to the volume of gases released per tonne of material produced or treated. Wherever possible, data from the UK was used to derive an appropriate emission factor. In the absence of UK data, information from other countries was used for this purpose. Emission factors were combined with the quantity of material treated or produced by each of the shortlisted processes, to generate an estimate of the annual quantity of PCDD/Fs released to atmosphere.

All of the emission estimates relate to sources on a UK-wide basis (ie England, Wales, Scotland and Northern Ireland) save for traffic, which relates to Great Britain (ie England, Wales and Scotland). In the case of the latter source, the total number of vehicles in Northern Ireland (0.56 million) relative to the total number of vehicles in Great Britain (25 million) suggests that the contribution from Northern Ireland to the estimate for great Britain is unlikely to be significant. In the case of chemical waste incinerators, an estimate has been made of the quantity of waste incinerated in on-site incinerators that covers all of the UK.

3. *UK INVENTORY OF PCDD/F EMISSIONS TO ATMOSPHERE*

The resulting emissions inventory is presented in Table 1. The total emissions to atmosphere range from 630 to 3400 g I-TEQ a^{-1}. Industrial processes account for approximately 90% of the total inventory. Emissions from sinter plants and the incineration of MSW are the dominant sources, contributing an average of 63% and 27% respectively to the total emissions from industrial sources. However, as discussed below, PCDD/F emissions from these sources will be subject to increasingly stringent control from 1995/96, and as a result the contribution from these sources will reduce significantly.

The emission sources in the second tier of importance are the following:

- Combustion of coal
- Emissions from iron and steel plants
- Emissions from non-ferrous metals operations
- Combustion of clinical waste

Table 1: UK Inventory of PCDD/F Emissions to Atmosphere

Process	Emissions to Atmosphere (g I-TEQ a^{-1})		Estimate Quality [2]
	Present Estimate	Future Projected Estimate	
Coke production	2	2 [3]	H/M
Coal combustion (industrial)	5 - 67	5 - 67	H/M
Waste oil combustion	0.8 - 2.4	0.8 - 2.4 [3]	M/N
Wood combustion (industrial)	1.4 - 2.9	1.4 - 2.9 [3]	M/L
Straw combustion	3.4 - 10	3.4 - 10 [3]	L/L
Tyres combustion	1.7	1.7 [3]	H/H
Landfill gas combustion	1.6 - 5.5	1.6 - 5.5 [3]	M/N
Sinter plants	90 - 2200	47	M/L
Iron and steel	3 - 41	14	M/L
Non-ferrous metals	5 - 35	10	M/L
Cement manufacture	0.2 - 11	0.2 - 11 [3]	H/M
Lime manufacture	0.04 - 2.2	0.04 - 2.2 [3]	H/M
Glass manufacture	0.005 - 0.01	0.005 - 0.01 [3]	H/M
Ceramic production	0.02 - 0.06	0.02 - 0.06 [3]	H/M
Halogenated chemicals	0.02	0.02 [3]	M/M
Pesticide production	0.1 - 0.3	0.1 - 0.3 [3]	L/M
MSW combustion	460 - 580	15	H/M
Chemical waste combustion	1.5 - 8.7	0.3 [3]	M/M
Clinical waste combustion	18 - 88	5	H/M
Sewage sludge combustion	0.7 - 6	0.9	H/H
Carbon regeneration	0.006	0.006 [3]	H/M
Asphalt mixing	1.6	1.6 [3]	H/M
PCP in timber processes	0.8	0.8 [3]	L/L
Crematoria	1 - 35	1 - 35 [3]	H/L
Domestic wood combustion	2 - 18	2 - 18 [3]	L/L
Domestic coal combustion	20 - 34	20 - 34 [3]	L/L
Traffic	1 - 200	1 - 202 [3]	H/L
Natural fires	0.4 - 12 [1]	0.4 - 12 [3]	L/L
Total	**630 - 3400**	**130 - 500**	

Notes:
[1] Based on open fires data. Soot-based approach not used.
[2] The values are assigned a rating for estimate quality. The first letter for quality of data relates to UK production and the second to the emissions data:
H = High, M = Medium and L = Low.
[3] No reduction has been assumed. This is not necessarily valid (for example with Cremation and Traffic).
[4] Assumed to comply with EU Draft Directive COM(92)9 Final-SYN 406
(0.1 ng I-TEQ Nm^{-3}).

Collectively, these four emission sources contribute an average of 7% to total industrial emissions in roughly equal proportions. The remaining industrial sources account for approximately 3% of the total UK inventory (all sources).

The non-industrial sources assessed in this study contribute emissions of 24 - 300 g I-TEQ a^{-1}, approximately 10% of the total inventory. 50% - 76% of the non-industrial emissions to atmosphere are accounted for by traffic, followed by domestic coal combustion, which contributes 8% - 43%. Emissions from traffic and domestic coal combustion contribute an average of 8% and 1% to the total UK inventory respectively (all sources). The remaining non-industrial sources account for approximately 2% of the total UK inventory (all sources).

4. LIKELY FUTURE CHANGES

The present study has been conducted during a period of rapid change, with industrial practices adapting to UK legislation such as Integrated Pollution Control. In this period of change, the study has encountered industrial processes which operate under a range of conditions, and hence emit widely varying quantities of PCDDs and PCDFs. As a consequence, the uncertainty in the emission estimates is greatly increased. However, emissions to atmosphere from some processes, in particular thermal waste disposal processes and thermal metallurgical processes, will undergo significant reduction over the next few years, as a result of the implementation in the UK of Integrated Pollution Control (IPC). All new plant are to achieve an emission concentration of 1 ng I-TEQ m^{-3}, and old plant are required to meet this emission limit over various timescales, extending to the year 2000. It is therefore of interest to examine a scenario in which these industrial sources conform to the required PCDD/F release concentration, a process which is already under way through the implementation of improvement programmes attached to IPC Authorisations. Table 1 presents the revised emissions data for industrial processes, based on the assumptions that industry size and plant throughput remains unchanged, and that PCDD/F emissions from other industrial and non-industrial remain unchanged.

The net effect is a reduction of annual emissions, from 630 - 3400 g I-TEQ a^{-1} to 130 - 500 g I-TEQ a^{-1}. The largest reduction is effected through the imposition of the 1 ng I-TEQ m^{-3} emission limit on metallurgical processes and MSW incinerators: the contribution from these sources falls from its present range of 80-84% to approximately 10-40%. Other emission sources such as coal combustion assume greater importance, jointly contributing 50-80% to total emissions. The present emission estimates depict industry in a state of flux, while the projected future estimates represent a more settled regulated state which, in terms of future emissions to atmosphere, are of greater interest.

5. LIMITATIONS

There are several caveats that must be noted in relation to the estimates provided in this study:

- There is a scarcity of PCDD/F emissions data in the UK in almost all of the industrial and non-industrial source categories examined. Many of the emission factors have been adopted or derived from studies conducted in countries other than the UK.

- For many of the industrial processes examined, there are a wide range of equipment designs in use, operating with varying inputs and under a range of conditions. In many instances it is not

possible to reliably predict the emissions from different plant configurations and operational conditions.

- For non-industrial sources there is considerable difficulty in estimating emissions due to the lack of data, the widely varying conditions under which processes such as domestic combustion and accidental fires may emit PCDD/Fs, and the uncontrolled, unregulated nature of these emissions. Obtaining representative samples from such processes is extremely difficult.

- At the present time, several industrial processes are subject to significant changes in emissions due to changes in industry practice, the addition of gas cleaning equipment and the expansion or contraction of industry sectors. Many of the technological changes have been in response to the introduction of Integrated Pollution Control, in every case leading to a reduction in the magnitude of the emission factor in the future.

These limitations are shared by all other national inventories. Explicit regulation of PCDD/F emissions, the application of consistent standards across industry sectors (thereby reducing the range of emission possibilities), extension of PCDD/F regulation to an increasing number of processes, more formal sampling requirements and improved reporting procedures will all lead to improved reliability in emission estimates. All of these measures are in the process of being introduced or implemented in the UK through Integrated Pollution Control. There remains the issue of unregulated emission sources such as domestic wood and coal combustion.

6. DATA GAPS

In terms of sources not considered in this study, comparison with other national inventories[6,7] suggests that all industrial point sources presently considered as potentially significant PCDD/F emitters to atmosphere have been included, albeit with the limitations discussed above. However, the following sources have not been considered:

- Diffuse/fugitive emissions from industrial processes.

- Introduction of PCDD/F into the atmosphere through the distribution and use of chemicals, as opposed to emissions during the manufacturing process.

- Unregulated sources such as accidental fires.

At present it is not possible to estimate the magnitude of emissions from the above sources, nor is it possible to assess whether they might be potentially significant contributors to the national budget. Further work is required to improve our understanding of these emission sources.

A final source contribution not accounted for in the present study is that of sinks such as contaminated soil. Some national inventories have noted a shortfall between the estimates of PCDD/F emissions to atmosphere, and measurements of aerial deposition to ground[8]. Suspension of soil particles and resuspension of deposited particulates, both containing PCDDs and PCDFs, has been postulated as a possible source to make up the shortfall. However, other studies have suggested that long range transport of particulate matter into and out of the country and the large uncertainties associated with the characterisation of PCDD/F emissions from known sources may well account for the discrepancy between the atmospheric burden and deposition rates[9]. Thus, while sinks such as contaminated soils

will undoubtedly contribute to the overall national PCDD/F budget in the atmosphere, their significance is as yet unclear. Further clarification of the physicochemical processes governing the fate and transport of PCDDs and PCDFs in soils and in air is required before this source can be adequately assessed.

7. REFERENCES

1) Department of the Environment (1989). *Dioxins in the Environment*. Pollution Paper No. 27. UK Central Directorate of Environmental Protection. Her Majesty's Stationary Office, London, United Kingdom.

2) Eduljee G H (1988). Dioxins in the environment. *Chem Britain*, **24**: 1223-1226.

3) Harrad S J and Jones K C (1992). A source inventory and budget for chlorinated dioxins and furans in the United Kingdom environment. *Sci Total Environ*, **126**: 89-107.

4) Heindl A and Hutzinger O (1986). Search for industrial sources of PCDD/PCDF I: Approaches in the Federal Republic of Germany. *Chemosphere*, **15**: 2001-2002.

5) Bremmer H J and Hesseling W F M (1991). Inventarisatie van processen waarbij dioxines kunnen ontstaan. Report No. 730501010, RIVM, Bilthoven, Netherlands.

6) Bremmer H J, Troost L M, Kuipers G, de Koning J and Sein A (1994). Emissions of Dioxins in the Netherlands. Report No. 770501018, RIVM, Bilthoven, Netherlands.

7) Fiedler H (1993). Formation and sources of PCDD/PCDF. *Organohalogen Compounds*, **11**: 221-228.

8) Hagenmaier H (1994). Contributions of diesel-powered vehicles and wood burning to overall PCDD/PCDF emissions. *Organohalogen Compounds*, **20**: 267-270.

9) van Jaarsveld J A and Schutter M A (1993). Modelling the long range transport and deposition of dioxins; first results for NW Europe. *Chemosphere*, **27**: 131-139.

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