PCDD/Fs IN THE BRITISH ENVIRONMENT: SINKS, SOURCES AND TEMPORAL TRENDS.

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ABSTRACT

This paper estimates the present UK environmental burden of total tetra- through octa-PCDD/Fs (ΣPCDD/Fs) in the environmental compartments of soil, air, vegetation, freshwater and freshwater sediments. Greater than 99% of the estimated total UK ΣPCDD/F loading of 5.6 t is present in surface soils. Annual emissions from known primary sources of PCDD/Fs are estimated. The major sources include MSWIs and the industrial and domestic combustion of coal. Whilst such primary sources are relatively easy to define, secondary releases - such as those from the production and use of organochlorine chemicals like pentachlorophenol (PCP) - are more difficult to quantify, yet may be amongst the most important sources of ΣPCDD/Fs in quantitative terms.

Whilst there is broad agreement between the estimated contemporary annual flux to the UK surface and environmental loading of ΣPCDD/Fs, there is a significant discrepancy between the sum of the annual emissions from primary sources and the annual deposition flux. Although the existence of an as yet unidentified source (or sources) cannot be excluded, it is proposed that much of this discrepancy may be accounted for by secondary releases from the use and disposal of chlorophenols and the long-range transport, continual re-mobilisation and subsequent re-deposition of PCDD/Fs already present in the environment.

Examination of temporal trends via retrospective analysis of archived soils reveals that UK PCDD/F levels increased significantly following the onset of widespread organochlorine chemical use and waste incineration in the late 1920's through to the mid-1970's. Since then, there is limited evidence to suggest that levels have peaked and may even be decreasing, although the rate of decline is likely to be slow, owing to the continued recirculation of the existing PCDD/F burden.

INTRODUCTION

This paper uses some of the available information to produce a budget and source inventory for PCDD/Fs in the contemporary UK environment. This is seen as an important prelude to the identification of research priorities and measures to reduce the environmental burden of these compounds. Temporal trends are also reviewed, as knowledge of past behaviour can assist in projections of future trends.

ENVIRONMENTAL LEVELS

Recently, several comprehensive surveys of PCDD/Fs in British soils[1,2] and herbage[3] have been undertaken. These show that PCDD/Fs are ubiquitous in the UK - as in other industrialised nations - although levels in urban areas are on average four times greater than in rural locations. Using British data where available, the current UK burden of ΣPCDD/Fs is estimated at ≈ 5.6 t, apportioned thus: 5560 kg in topsoil (0-5 cm), 2.9 kg to vegetation, 0.85 kg to air, 0.44 kg to freshwater and 25 kg to freshwater sediments[4].

Discussion centres on levels and sources of ΣPCDD/Fs, as opposed to just 2,3,7,8-T_4CDD or TEQs. The value of the TEQ approach is acknowledged, however, presently available data is inadequate to produce a sufficiently detailed budget. Similarly, confining discussion to 2,3,7,8-T_4CDD could severely underestimate the environmental hazard posed by PCDD/Fs. We believe that consideration of ΣPCDD/Fs is a satisfactory compromise.

SOURCES OF PCDD/Fs TO THE UK ENVIRONMENT

Whilst natural sources of PCDD/Fs such as forest fires and volcanic eruptions exist, such phenomena are unlikely to be significant contributors to the UK environmental loading. Instead, anthropogenic sources are universally accepted as the predominant vector for the release of PCDD/Fs into the environment. Two main categories of anthropogenic sources are recognised. Firstly, the production and use of organochlorine chemicals that are contaminated with PCDD/Fs. Secondly, the combustion of chemical, clinical and household waste, as well as fuels like coal and leaded petrol. Table 1 shows the estimated annual releases of ΣPCDD/Fs from major sources - detailed derivations of these estimates are given elsewhere[4]. Anthropogenic combustion activities and pathways for the secondary release of dioxins in organochlorine biocides - *e.g.* the disposal to land of contaminated sewage sludge - represent approximately equal sources of ΣPCDD/Fs in Britain. Consideration of homologue-specific emissions indicates that combustion processes represent a more significant source of tetra- and penta- CDD/Fs than does the production and use of chlorophenols, which in turn constitutes a greater source of the hepta- and octachlorinated congeners. Direct emissions into the atmosphere from combustion processes facilitates their atmospheric transport to remote areas. This, coupled with the diffuse nature of processes like fossil fuel combustion, means that PCDD/F contamination originating from anthrogenic combustion is more widespread than that resulting from the use and disposal of chlorophenols. Contamination from chlorophenols is more localised, as direct atmospheric release pathways for this source are insignificant. Extrapolating from US data[5] the annual flux (or environmental input) of ΣPCDD/Fs to the surface of the UK is calculated to be 250 kg, of which only 11% (26.2 kg) may be accounted for by those primary sources for which estimates are possible. However, even when emissions from other identified primary sources and secondary release mechanisms are quantified there is still a significant disparity. This is thought to be due to either secondary deposition or presently unidentified sources.

Table 1. Estimated PCDD/F Emissions from Major Primary Sources in the UK.

Source.	Estimated Annual Emissions.
Municipal Waste Incinerators	10.9 kg.
Chemical Waste Incinerators	Minimal - "a few grammes of T_4CDDs"[6].
Clinical Waste Incinerators	1.7 kg.
Domestic Coal Combustion	5.1 kg.
Industrial Coal Combustion	7.7 kg.
Coal Combustion for Power Generation Purposes	No precise figures. DoE[6] estimates British emissions to be about 5 % of total emissions for all UK combustion sources - 1.3 kg.
Leaded Petrol Combustion	0.7 kg.
Volatilisation from Chlorophenol treated substrates.	1.7 kg.
Secondary releases from Chlorophenol treated substrates.	Very difficult to estimate. Potentially substantial contribution.

TEMPORAL TRENDS

Analysis of archived British soils[7] shows that present PCDD/F levels substantially exceed that of a century ago. Significant increases occurred between the late 1920's and the mid-1970's, in line with the onset of large -scale organochlorine production and waste incineration. Current indications are that levels of PCDD/Fs are no longer increasing and indeed, may even be starting to fall[7,8]. The future rate of decline in the total PCDD/F burden of the UK is likely to be modest however, owing to the persistence of PCDD/Fs and the recirculation of the PCDD/Fs already present in the environment.

CONCLUSIONS

There remain key areas where further research is required to facilitate the formulation of policies designed to mitigate the potentially damaging effects of PCDD/Fs on the British environment.

One area involves the identification of presently unknown sources and the quantification of emissions from those that have been identified, but not as yet fully quantified, in particular, the application of contaminated sewage sludge to agricultural land (which probably constitutes a important vector for the release of PCDD/Fs in organochlorine chemicals) and the combustion of coal for power generation purposes. Such work is essential if the present disparity between the estimated annual flux and emissions from known sources is to be fully accounted for. Other research designed to account for this discrepancy include: measurements of the PCDD/F flux to the surface of the UK and investigations into the significance of secondary deposition. Temporal trends should also be monitored, in order that the effects of measures aimed at curbing PCDD/F contamination may be evaluated. Also of interest are field determinations of PCDD/F half-lives, as uncertainty still exists regarding their persistence.

In summary, levels of PCDD/Fs in the British environment essentially reflect those in other Western nations. The major terrestrial sink is soil and the principal contemporary sources are waste incineration, coal combustion and secondary releases from the manufacture and use of organochlorine chemicals. Encouragingly, there are indications that past measures aimed at curbing emissions are exerting a beneficial effect and that fresh PCDD/F inputs are now declining.

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