

# PCDD/Fs IN THE UNITED KINGDOM: PAST, PRESENT AND FUTURE PROSPECTS

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## Abstract

Ambient air concentrations of PCDD/Fs have declined in the UK over recent years and decades, although we do not have a clear understanding of why this has occurred. The decline in air concentrations has resulted in a decline in human exposure, through reductions in the transfers of PCDD/Fs (and related compounds) to crop plants and grazing animals in terrestrial foodchains, and to aquatic organisms in marine and freshwater ecosystems. Further declines in human exposure and therefore human tissues are desirable, following revisions of the WHO/ECEH tolerable daily intake (TDI) of the toxicity equivalents of PCDD/Fs and PCBs. However, it is likely to be increasingly difficult for industrialised countries to take further measures to reduce sources and hence human exposures.

## 1. Introduction

The United Kingdom has had a programme to reduce PCDD/F emissions to the environment for some years now. This has principally been aimed at identifying and reducing key sources to the atmosphere and reducing human exposure. This short article briefly describes the situation in the UK, the relationships between emissions and environmental distribution/burden and the transfer of PCDD/Fs to humans. Time trends of PCDD/Fs into the environment are also briefly discussed and future developments considered.

## **2. The UK position**

In 1989 the UK established a policy framework (DoE, 1989) for addressing PCDD/Fs, based on the identification and control of sources, monitoring of the foodchain and human exposure and continuing review of the information on the toxicology of the compounds. In addition, there are various international actions which impinge upon UK activities in this field. For example, changes have been made to the system of toxicity equivalency factors (TEFs) used to determine the toxicity of complex mixtures with the WHO/ECEH recommending a revised scheme. The addition of selected PCB congeners to the standard assessments of exposure has resulted in an increase of exposure to 'TEQ' of dioxin-like compounds of up to 50% when compared with the TEQ of just PCDD/Fs (Alcock *et al.*, 1998a). In 1998 the WHO/ECEH reconsidered the acceptable exposure to dioxin-like compounds and recommended a reduction in the tolerable daily intake (TDI) from 10 pg/kg body weight/day to a range of 1-4 pg/kg/day expressed as TEQ using the latest TEF scheme. As in other countries, a substantial proportion of the UK population is exposed above the lower end of this range. The UK government is therefore reviewing the current situation and considering what further measures, if any, can or should be taken to further reduce emissions and human exposure.

## **3. Brief comments on sources, environmental distributions and burdens**

PCDD/Fs enter the environment from various combustion processes and as impurities from the manufacture and use of various chlorinated compounds (e.g. Duarte-Davidson *et al.*, 1997). Considerable effort has been expended in the UK and elsewhere to try and quantify and rank these sources and the emissions to the environment, principally the atmosphere, so that cost-effective source reduction measures can be taken (e.g. Duarte-Davidson *et al.*, 1997; Alcock and Jones, 1997; Alcock *et al.*, 1999; Jones and Sewart, 1997). However, there is still considerable uncertainty about the key sources of PCDD/Fs (and other TEF-rated substances such as PCBs and PCNs) and their fluxes into the UK environment. This uncertainty will undermine any future efforts at further reductions in human exposure.

The most recent estimates of primary atmospheric emissions of PCDD/Fs to the UK atmosphere are ~220-660 g I-TEQ/year in the mid-1990s (Alcock *et al.*, 1999). However, there are considerable uncertainties over the contribution of so-called 'secondary sources', such as volatilisation from soils (Harner *et al.*, 1995; Cousins *et al.*, 1998), emissions from accidental fires and diffuse sources (see below).

There have also been attempts to link the estimated current and past emissions of PCDD/Fs into the UK environment with the 'environmental burden' of these compounds in soils, sediments and other environmental compartments (see Duarte-Davidson *et al.*, 1997). This kind of 'environmental budgetting' or 'accounting' can be useful and highlights areas of scientific uncertainty and differences between compounds. Major complications can arise, however, because of a lack of information on PCDD/F persistence (e.g. McLachlan *et al.*, 1996), fluxes between environmental compartments (e.g. Cole *et al.*, 1999), uncertainties over the emissions of PCDD/Fs into the environment in the past (see below) and imports/exports of PCDD/Fs in/out of the UK via long-range atmospheric transport.

#### **4. The role of the atmosphere and the link to human exposure**

The focus on identifying and reducing primary emissions to the atmosphere stems from our knowledge that key environmental compartments are intimately linked, transferring PCDD/Fs, PCBs *etc.* present in air (e.g. Duarte-Davidson *et al.*, 1994; Lohmann and Jones, 1998a; 1998b) via dry gaseous-, dry particulate- and wet-deposition (e.g. Halsall *et al.*, 1997; Lee *et al.*, 1998; Lohmann *et al.*, 1998) to vegetation and soil (e.g. Jones and Duarte-Davidson, 1997; Cousins and Jones, 1998; Thomas *et al.*, 1998a; 1998b), to grazing animals and their milk, meat and dairy products (Thomas *et al.*, 1998c; Thomas *et al.*, 1999) and then ultimately to the human diet. This pathway is believed to dominate human exposure to PCDD/Fs and PCBs in the UK and other western countries (Duarte-Davidson and Jones, 1994). However, the pathway air - water bodies - aquatic foodchains - fish - human consumption are also very important and will obviously ultimately be affected by reductions in atmospheric emissions. Important studies are therefore aimed at establishing quantitative models of the transfer factors and dynamics of PCDD/Fs and PCBs between air and humans (e.g. McLachlan, 1996; Douben *et al.*, 1997; Sweetman *et al.*, 1998; Thomas *et al.*, 1998a; 1998b; 1998c; Thomas *et al.*, 1999; Cole *et al.*, 1999) which can be used as predictive tools.

#### **5. The search for evidence of past trends**

It is interesting to note that data from sediment cores and archived samples suggest that the inputs of PCDD/Fs to the environment have been declining for many years, probably having peaked in the late-1960s/early 1970s (see Alcock and Jones, 1996; Kjeller *et al.*, 1991; 1996; Duarte-Davidson *et al.*, 1997; Alcock *et al.*, 1998b). However, the large-scale efforts at primary source reduction did not begin until much later in the UK (and other countries), with efforts to

tackle emissions from municipal solid waste incineration (MSWI) and other important primary sources. This has important implications, because it suggests that we do not adequately understand why PCDD/Fs levels have reduced and which sources/measures were responsible for the observed declines. Indeed, other major uncertainties about PCDD/Fs remain. From our own studies, for example, we have detected the presence of PCDD/Fs in environmental samples collected and stored from the late-1800s (Kjeller *et al.*, 1991; 1996; Alcock *et al.*, 1998b) and believe this is consistent with the emission of trace quantities of PCDD/Fs from combustion of coal/wood and/or metal smelting activities prior to the development of the 'Chlorine Industry' during this century. However, this has been challenged by others (Baker and Hites, 1999; Alcock *et al.*, 1999), who are concerned that what is reported as 'pre-industrial' PCDD/Fs may be contamination of samples since their collection. Clearly this highlights some fundamental uncertainties/questions which urgently need to be resolved.

It is important to acknowledge an important distinction between trends in *concentrations* of PCDD/Fs in the environment and trends in *the environmental burden*. Time trend data for air (e.g. Coleman *et al.*, 1997), vegetation (Kjeller *et al.*, 1991; 1996), cows milk and human tissues (Alcock and Jones, 1996) clearly show a decline in PCDD/F concentrations through the 1980s and 1990s. These media are all affected by emissions to atmosphere and hence air concentrations, as briefly explained above. However, the soil represents the major environmental storage compartment for PCDD/Fs released to the terrestrial environment and becomes a repository for cumulative PCDD/F deposition. Concentrations in the soils of the UK have been increasing through this century (Kjeller *et al.*, 1991; Alcock *et al.*, 1998b) and - because the soil dominates the environmental burden of PCDD/Fs (Duarte-Davidson *et al.*, 1997), the total mass of PCDD/Fs in the environment is still increasing (i.e. the rates of emission exceed the rates of degradation). The role of the soil is therefore of key importance and it is necessary to understand whether the soil has the capacity to bind and ultimately degrade these environmentally recalcitrant compounds, or whether it could potentially hold them in a bioavailable or desorbable form, such that they may ultimately be re-released to other environmental compartments (e.g. the air by volatilisation) or foodchains (Cousins *et al.*, 1998).

## **6. A brief comment about atmospheric concentrations**

Reliable measurements of PCDD/Fs in air require sensitive and precise analytical techniques; air concentrations of 2,3,7,8-TCDD, for example, are <1 fg/cu. m in rural areas, requiring the sampling of many hundreds of cubic metres of air, even when the most sensitive MS instrument is available for quantification. However, much information can be gained to

make inferences about sources and their trends and the role of degradation and loss processes from detailed studies of the factors that influence or control atmospheric concentrations of PCDD/Fs.

Detailed studies are being conducted at our field station site close to Lancaster University on the north west coast of England, relating changing atmospheric concentrations to seasonal, anthropogenic, air mass and meteorological influences (e.g. Lee *et al.*, 1998; Lohmann and Jones, 1998a; 1998b; Lohmann *et al.*, 1998). Some important findings are as follows: i. generally higher levels of PCDD/Fs are associated with air masses that originate and move over land, particularly during periods of low ambient temperature; ii. low PCDD/F concentrations are associated with air masses that arrive from the Atlantic Ocean/Irish Sea to the west and have little or no contact with urban/industrialised areas; iii. concentrations in the autumn months are 2-10 times higher than those found in the summer; iv. concentrations in the autumn rise as ambient temperatures decrease; v. the highest PCDD/F concentrations measured have been associated with a national festival when it is customary to light fireworks and bonfires burning wood, garden refuse and wastes.

## **7. A brief comment about sources**

The observations just made in the previous section have implications for current sources of PCDD/Fs to the UK atmosphere.

The major sources that have been estimated to dominate the UK primary atmospheric emissions inventory will essentially be consistent through the year and not display seasonality (i.e. emissions from MSWI, the iron and steel industry, metal smelting etc). Power stations increase their activities during the darker, colder months of winter, but PCDD/F emissions from these high temperature 'clean combustion' sources are thought to be minor. It is therefore unlikely that any of the major primary sources mentioned above are responsible for the seasonal air concentration differences noted above, even though these 'non-seasonally dependent' emission sources have been estimated to contribute the majority of the national annual primary UK PCDD/F inventory. However, there are clearly other diffuse sources which could make an important contribution to emissions which are seasonally dependent. Foremost amongst these are inputs associated with domestic heating during colder conditions, namely domestic burning of coal, wood and gas. This implies that it may be difficult to bring about substantial further declines in PCDD/F emissions, without fundamentally altering the nature of the techniques used for space heating in the UK. It also implies that source inventory estimates may have

underestimated the contribution that secondary, diffuse emissions are making to the UK environment.

Finally, it is worth commenting that the time trends in UK ambient levels noted above in section 5 (i.e. declining from the 1960/70s) are perhaps more consistent with the declines in the rather inefficient burning of coal and other fuels for domestic space heating (Jones *et al.*, 1989) than the trends in MSWI and other large-scale primary emissions (Alcock and Jones, 1996; Duarte-Davidson *et al.*, 1997).

Further work is required to understand and quantify the nature of PCDD/F formation/release from small, inefficient and diffuse combustion sources.

## **8. Environmental response times to source reduction measures**

Air concentrations of PCDD/Fs are variable spatially and temporally, responding rapidly (i.e. over hours/days) to changing emissions and loss processes. Reductions in the important emissions to atmosphere should therefore quickly become apparent by declining air concentrations. Other environmental compartments will then respond to this change, but the kinetics or 'dynamics' of changes in other compartments will presumably be more complex. For example, vegetation concentrations respond primarily to air concentrations (rather than to soil), but may broadly integrate/reflect deposition or uptake that has taken place over days/weeks/months or possibly years, depending on the plant species (Thomas *et al.*, 1998a; 1998b; Ockenden *et al.*, 1998). As air concentrations decline, it may therefore follow that there is a shift in the net fluxes of PCDD/Fs and PCBs between the air and the soil compartments (Harner *et al.*, 1995; Cousins and Jones, 1998), such that there is net outgassing rather than net deposition. Grazing animals live for many months/years and - depending on their contribution to the human foodchain - may be either at steady or unsteady state with respect to their intake (Sweetman *et al.*, 1998; Thomas *et al.*, 1999). Likewise, human exposure, absorption efficiencies, tissue concentrations and body burdens vary with age, sex, diet and time in a more complex way. This gives rise to a very wide range in exposures within the population at any given time and to a wide range in tissue concentrations/body burdens in any one individual through their lifetime. These processes can be unravelled and better understood by the use of dynamic modelling techniques.

## **9. Speculation about the future**

As noted in the Introduction, further declines in human exposure and therefore human tissue concentrations are desirable, following revisions in the WHO/ECEH tolerable daily

intake (TDI) of the TEQs of PCDD/Fs and PCBs. However, as we have seen, it is likely to be increasingly difficult for industrialised countries to take further measures to reduce sources and hence human exposures of these compounds. The persistence of PCDD/Fs in the environment and in human tissue means that it may take some time for the full effects of emissions reductions to 'cascade through' to the full range of environmental compartments, notably human tissues. It is therefore unclear at the present time whether the marked and steady decline in human tissue concentrations is still the result of emission reductions which occurred many years ago, or reflect a sharper response to primary emission reductions in the late 1980s and early 1990s. Further work is therefore required to unravel the complex dynamics and interactions of these compounds with the environment and top predators which are prone to adverse effects from these persistent, bioaccumulatory and toxic substances.

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