HUMAN SOURCES AND ACTIVITIES ASSOCIATED WITH DIOXIN-LIKE COMPOUNDS AND POPS IN THE ENVIRONMENT

DIFFUSE AND SECONDARY SOURCES OF ATMOSPHERIC PCDD/Fs: ARE THEY SIGNIFICANT?

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Introduction
Governments and other organisations have spent huge sums of money to identify, quantify and try to reduce/eliminate sources of PCDD/Fs to the environment. This has principally focussed on obvious 'primary' sources (defined here as a 'new or fresh release' of compound, e.g. from key point sources such as municipal waste incinerators). As these sources have been reduced in many countries, it is pertinent to consider which source categories dominate the inputs of PCDD/Fs to the atmosphere and how we can obtain reliable information to help us make that assessment. The focus of our discussion here is:

- Diffusive inputs. This term is taken to mean 'non-point' or 'scattered' sources, such as domestic burning of coal/wood/gas for heating, vehicle emissions, bonfires, 'uncontrolled' fires, fugitive emissions from landfill etc. These source types are generally difficult to control.
- Secondary sources. This term refers to a re-emission of previously emitted/deposited material, such as volatilisation from soil, vegetation or water bodies. This process is known to be important for other POPs.

It is important that we have a clear understanding of how these various sources impact on emissions, so that cost-effective source reduction can be achieved and we can manage future trends in exposure.

How can we get evidence about the importance of different sources?
Broadly, evidence can be gained by the following approaches:

1. Source inventories and estimates. This is, of course, a widely used approach. However, there are some fundamental problems in reliably quantifying certain key sources. For example, the uncertainties over releases from accidental fires are so large that – depending on the emission factors selected – this can be estimated to be the dominant source to air, or just a minor one (1). In short, inventories are only as good as the measured data going into them - and that is often incomplete and imprecise. Sometimes inventory predictions about major sources seem inconsistent with field observations of ambient trends. We would have greater confidence in inventories if they could be related to measurements of PCDD/Fs in air. However, this approach is still in it's infancy (1, 2) and we are hampered by needing to quantify advective/long range *transport inputs/outputs for the region which is the subject of the inventory.*

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2. Modelling. Models can be used, to relate emission estimates to ambient measurements (1). They can also be applied to predict the net direction of inter-compartmental fluxes (3, 4). Modelling approaches can give important clues about the likely significance of secondary sources, but their application to PCDD/Fs is still hampered by uncertainties over: i. key physico-chemical properties and their temperature-dependence; ii. knowledge/measurement of some key processes of inter-media compound transfer; iii. the environmental distribution of PCDD/Fs e.g. their depth distribution in soil.

3. Trends in environmental levels. This approach can yield clues, by relating trends in environmental levels or fluxes to changing sources. Do trends in ambient air, for example, coincide with known reductions in primary sources (5)? There is still a general shortage of good quality long-term (decades – e.g. derived from dated sediment cores or archived samples) (6) and short-term (years – e.g. derived from direct measurement programmes) (7, 8) data sets for ambient air. However, the emerging picture is that PCDD/F levels in Europe peaked in the late 1960s/early 1970s and have been dropping consistently since then. This would appear to be inconsistent with suggestions that MSWI was the dominant source of PCDD/Fs to air in the 1980/90s (5).

4. Laboratory and field measurements Controlled measurements of fluxes (e.g. soil-air) or emission factors (e.g. domestic burning of coal/wood) yield key data, provided they can be understood and related to the 'real world'.

The improvements in analytical sensitivity enable measurements of ambient PCDD/F concentrations to be made on sampling intervals of the order of a day or so – even in remote locations such as the Antarctic (9, 10). It has therefore been possible to relate changing ambient concentrations temporally and spatially to environmental factors and sources (2, 9). Direct measurement yields the best form of evidence, because it relates to the 'real world'. Such studies have shown:

- powerful evidence that seasonally-dependent diffusive, primary sources – the domestic burning of coal and wood – has a major influence on ambient air in the UK (9, 11, 12);
- uncontrolled burning can release substantial amounts of PCDD/Fs and dominate air quality (9, 11, 12);
- secondary (re-cycling) sources are influencing the contemporary ambient levels of lower chlorinated PCDD/Fs in urban areas (13);
- the net air-soil flux is still currently into the soil for the important TEF-rated PCDD/Fs (4);
- numerous environmental factors influence ambient levels and these can confound studies seeking to quantitatively assess the link with sources (9).

5. Chemical tracers. There is considerable interest in the use of chemical tracers for source apportionment work. Often when close to a strong source with a characteristic signature, this approach is effective, but it can be far more challenging after an air mass has become diluted and weathered. The approach is hampered by our continuing uncertainties over atmospheric reactions and loss processes.

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Summary of studies and discussion points

Collectively, our studies on source inventories, laboratory and field measurements and modelling lead us to the following conclusions about PCDD/Fs in the UK atmosphere:

- Current ambient levels of PCDD/Fs are probably controlled by diffusive / secondary sources; these sources will also control ambient levels in the coming decades;
- An important diffusive primary source appears to be the domestic burning of coal and wood for space heating;
- Levels of PCDD/Fs have come down dramatically over several decades, but this is probably more by accident than design. We currently believe this trend is probably a reflection of regional changes in the burning of coal and wood for space heating, rather than a response to the targetted reduction of point sources over the last decade or so;
- Future regional rates of decline in ambient concentrations will be controlled by international source reductions and 'environmental loss processes', although we are not sure which ones dominate (e.g. biodegradation in soils/sediments; incorporation or 'dilution' into soils and sediments; atmospheric reactions with OH radicals);
- Complete elimination of PCDD/Fs is obviously not possible. We have to decide what levels of exposure are 'safe' or 'acceptable' and see whether current emissions 'comply' with those. A modelling approach that links levels in exposed organisms (e.g. humans) to those in foodstuffs and ultimately to those in air is being developed as a management tool (6).

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