A CONGENER-SPECIFIC BUDGET & SOURCE INVENTORY FOR POLYCHLORINATED BIPHENYLS (PCBs) IN THE BRITISH ENVIRONMENT

Stuart J. Harrad[1] and Kevin C. Jones[2]

[1]Scientific Analysis Laboratories Ltd.,
Business and Technology Centre, Green Lane, Eccles,
Manchester, M30 0RJ, UK.

[2]Institute of Environmental and Biological Sciences,
Lancaster University, Lancaster, LA1 4YQ, UK.

INTRODUCTION

This paper outlines a budget and source inventory for PCBs in the contemporary British environment. Discussion centres on both ∑PCBs and congeners 28, 52, 101, 138, 153 and 180. Temporal information regarding PCBs in the British environment is also reviewed.

THE PCB BURDEN IN THE CONTEMPORARY BRITISH ENVIRONMENT

The current burden of PCBs in the UK environment has been estimated for the main environmental compartments and the data summarised in Table 1. The bulk of the contemporary UK burden of ∑PCB (400 t) resides in soils (93.1%), whilst the remainder is predominantly associated with marine sediments and seawater (a combined total of 5.6%). The apportionment of individual congeners basically reflects that of ∑PCB, although the relative importance of humans - and by implication, that of biota generally - as a sink for congener 180 is interesting.

SOURCES AND RELEASES OF PCBs

The following sections address UK sales, fluxes and environmental releases of PCBs.

Sales of PCBs in the UK

Based on the data of Bletchley (1983), estimated UK sales of PCBs totalled 39 446 t, consisting of 8 017 t in large capacitors, 5 742 t in transformers, 8 017 t in small capacitors and 17 670 t in miscellaneous other applications. There is no definitive information concerning UK sales of different Aroclor formulations, although Christensen and Lo (1986) reported US EPA Aroclor-specific production figures for the USA for the period 1957-1974. On the basis of these data, we have estimated UK sales of individual congeners by assuming that UK sales of PCBs consisted solely of Aroclors 1242, 1254 and 1260 in the ratio 10:5:2 and by using the weight percent contributions of individual congeners measured by Schulz *et al* (1989) for these formulations (Table 2).

Annual Flux of PCBs to the UK Surface

We have averaged deposition fluxes recorded during the first six months of 1991 in the British cities of Cardiff and Manchester (Clayton *et al*, 1992) and taken these as representative of the flux in urban

Britain. No measurements of rural UK fluxes exist; instead we have estimated the PCB flux to such areas (assumed to constitute 90% of the total UK surface) by assuming that the ratio of rural to urban deposition fluxes will be identical to that between German rural (Wittlinger and Ballschmiter, 1987) and UK urban (Clayton *et al*, 1992) air concentrations. In this way, we have estimated the total flux to the surface of the UK (Table 3).

Contemporary Releases of PCBs to the British Environment

This section considers potential sources of PCBs to the contemporary British environment. Where necessary, congener-specific emission estimates are calculated using the data of Bletchley (1983) and Schulz *et al* (1989), who respectively reported on the distribution of different Aroclor formulations according to use category and the weight distribution of individual congeners in specific Aroclors. We have identified various processes as likely routes via which PCBs may enter the environment and our estimates are summarised in Table 4. Note that although there are minor variations for individual congeners, the relative importance of the various sources is broadly similar for each congener and $\sum$PCBs. The major sources are considered to be:

(a) volatilisation of PCBs from topsoil. This is probably the main contributor to the annual deposition flux - our estimate is that it constitutes $\approx$ 88.1% of the 45 t $\sum$PCB annually released into the UK atmosphere.

(b) leaks from improperly maintained PCB-containing transformers and large capacitors. Our estimate is that such leaks represent $\approx$ 9% of present annual releases to the atmosphere.

(c) emissions arising during the production of refuse-derived fuel, which we estimate could contribute around 2% of the UK's annual flux to the atmosphere of PCBs. By comparison, the limited data available indicate that volatilisation of PCBs from landfilled MSW is negligible and that its contribution to annual atmospheric releases is not significant.

(d) Production of steel from ferrous scrap recycled by fragmentising facilities. Our estimate is that PCB atmospheric emissions from this source amount to $\approx$ 0.5% of the annual UK total. In addition, we consider that overall releases from fragmentising facilities may constitute an even more significant source, given the elevated concentrations detected in fragmentiser deduster sludge.

(e) Volatilisation from sewage sludge applied to land. We estimate that this constitutes $\approx$ 0.2% of annual emissions to the atmosphere of $\sum$PCB in the UK.

TEMPORAL TRENDS

Analyses of dated sediment cores (Sanders *et al*, 1992) and archived soils and vegetation (Alcock *et al*, submitted; Jones *et al*, 1992) from the UK demonstrate that input of PCBs to the UK environment grew steadily following the onset of their commercial production, peaked in the early to mid-1960s, declined significantly following restrictions on their use in the 1970s and have since continued to decrease, albeit less precipitously.

As an illustration of the significance of the dramatic fall in levels of PCBs in UK soils and vegetation, we cite the data of Alcock *et al* (submitted). They reported that mean $\sum$PCB concentrations in soils from four UK sites fell from 360μg/kg in the late 1960s/early 1970s to 20 μg/kg in the late 1980s/early 1990s. Assuming that levels in these soils were/are representative of the UK as a whole, it is clear that the UK burden of PCBs has declined significantly in the last two decades. Whilst conclusive evidence of the fate of the PCBs "lost" from the UK is not available, it is likely that a significant fraction have undergone atmospheric and pelagic transport away from the UK and may be accumulating in polar regions.

In contrast, levels in biota appear to be taking longer to respond to the restrictions on PCB use. Specifically, Duarte-Davidson *et al* (in press) found little evidence of a decline in concentrations of PCBs in human adipose tissue between the 1970s and 1990/91 and Boumphrey *et al* (submitted) detected only a moderate decline in PCB levels in archived UK gannet eggs over the period 1977 to 1991.

Clearly, for some species, the direct transfer of PCBs across generations - via breast-feeding and transplacental transfer during pregnancy for mammals, or via eggs for avian species - is partially responsible for the relatively slow decline in PCB residues in biota. Another influential factor - particularly for humans - is the persistence of PCBs within biotic tissue. This means that current tissue levels will reflect elevated past, as well as lower present exposure and for this reason, the burden of PCBs in humans and other biota close to the top of food chains is unlikely to decrease significantly in the near future.

CONCLUSIONS

Of the estimated ≈ 40000 t ∑PCB sold in the UK, only an estimated 1% (400 t) currently reside in the UK environment. Analysis of congener-specific data on UK production and the current environmental loading suggests that PCB persistence broadly increases with increasing chlorination. The remainder are considered to be still in use, to reside in landfills, to have been destroyed by natural or anthropogenic processes, or to have undergone environmental transport away from the UK. The significance of this latter route is illustrated by the dramatic fall in PCB levels detected in UK archived soils and vegetation between the late 1960s and the present. More recent trends indicate that whilst environmental inputs may still be declining, the rate of decrease has diminished significantly. Given the important contribution of volatilisation from soils, future reductions in fresh environmental input will be small and PCBs are likely to continue to circulate throughout the environment for many years. Furthermore, levels in biota are likely to diminish at a far slower rate than that observed for soils and vegetation.

The primary sinks of the contemporary environmental burden of ∑PCBs in the UK and the surrounding ocean are soils (93.1%), seawater (3.5%) and marine sediments (2.1%). Although individual consideration of congeners 28, 52, 101, 138, 153 and 180 does not reveal any major differences to the relative partitioning calculated for ∑PCBs, the importance of sinks other than soils is greater for individual congeners, especially 138 and 180. Interestingly, humans represent a sink for 2.0% of the total burden of congener 180, implying that biota as a whole may be of some significance as a sink for the more chlorinated congeners.

The contemporary flux of ∑PCBs to the UK surface is estimated at 19 t/yr, whilst estimates of annual fluxes to the atmosphere total 45 t. Congener-specific data reveal similar excesses of estimated releases to the atmosphere over annual deposition fluxes for congeners 28, 52 and 101. Conversely, fluxes in both directions across the soil/air interface correlate closely for congeners 138, 153 and 180, thus implying that these and other more chlorinated congeners are less susceptible to environmental transport and atmospheric degradation. The major contributors to the contemporary flux to the atmosphere of ∑PCBs are thought to be: volatilisation from soils (88.1%), leaks from transformers and large capacitors (9.1%), the production of refuse-derived fuel (RDF) (2.2%), the recovery of contaminated scrap metal (0.5%) and volatilisation from sewage sludge-amended land (0.2%).

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PCB

Compartment	UK Burden (kg) of Congener # in.... 28	52	101	138	153	180	ΣPCB
Soil	21000	14000	22000	12000	19000	8600	370000
Seawater	670	660	1300	1600	780	860	14000
Marine Sediments	1600	820	740	890	1000	600	8200
Freshwater Sediments	180	93	110	93	68	160	2200
Vegetation	580	180	-	<6.5	<9.8	-	1800
Humans	8.3	1.5	1.9	98	110	210	760
Sewage Sludge	15	24	20	13	15	16	480
Air	4.6	4.5	5.3	2.7	3.4	1.3	65
Freshwater	1.4	1.7	1.6	1.0	0.58	-	30
Total Burden	24000	16000	24000	15000	21000	10000	400000

Table 1: Estimated Distribution of UK PCB Burden

UK Sales (t) of Congener # 28	52	101	138	153	180	ΣPCB
1500	1600	1500	780	1200	390	40000

Table 2: Estimated UK PCB Sales for the period 1954-1979

Congener #	Flux ($\mu g/m^2/yr$) Rural	Urban	Annual UK Deposition Flux (kg)
28	2.1	62	2000
52	1.5	22	890
101	2.0	8.2	660
138	3.1	7.6	890
153	3.5	8.2	990
180	1.7	8.9	610
ΣPCB	48	310	19000

Table 3: Deposition Flux of PCBs in the UK

Source	Estimated Atmospheric Releases (kg/yr) of Congener # 28	52	101	138	153	180	ΣPCB
Volatilisation from Soil	5100	4000	2400	840	1000	680	40000
Capacitor Leaks	300	180	41	20	19	1.7	3900
RDF Production	?	?	?	?	?	?	1000
Transformer Leaks	6	8	13	8.5	14	6.5	250
Scrap Metal Recovery	26	11	7.6	3.2	2.0	0.6	240
Volatilisation from land-applied sewage	5.9	12	3.7	1.6	1.2	1.8	85
Volatilisation from landfills			Negligible				
Total	5400	4200	2500	870	1000	690	45000

Table 4: Estimated Annual Releases of PCBs to the UK Atmosphere