THE RISE AND FALL OF PCBs: TIME-TREND DATA FROM TEMPERATE INDUSTRIALISED COUNTRIES

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INTRODUCTION

Long-term environmental monitoring data are required to assess the impact of international restrictions on the production and use of PCBs. The late 1960s/early 1970s was the period of maximum usage and production of these compounds, with restrictions introduced in the mid/late 1970s. However, little or no reliable long-term data has been available from that time. Consequently, retrospective sampling techniques have been used to help reconstruct past trends. Environmental samples deposited in discrete or identifiable layers, and well preserved archived samples have both been used.

In this paper congener-specific chronological trends derived for 3 lacustrine sediment cores, 1 ombrotrophic peat core and a set of archived herbage samples are summarised. Time trends from archived soil samples are presented in another contribution to this conference (1).

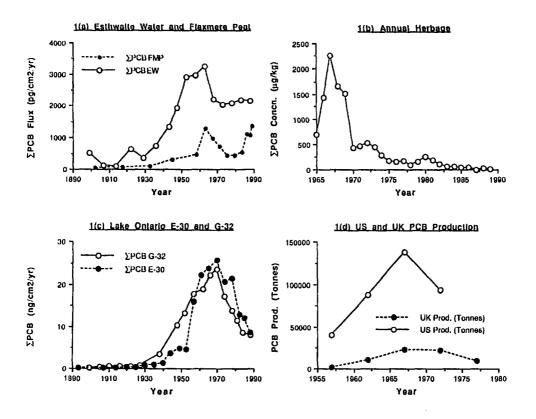
EXPERIMENTAL

Lacustrine sediment cores were obtained from Esthwaite Water (EW) in north west England, and two separate locations from the Rochester Basin of Lake Ontario (E-30 and G-32), using Mackereth mini- and box-coring techniques, respectively (2, 3). A wide-diameter, high-resolution sampler was used to extract a core from Flaxmere peat bog (FMP) in Cheshire, north west England (3). A collection of 25 separate samples (HERB) representing one year intervals from 1965 to 1989 from the long-term agricultural field experiments at the Rothamsted Experimental Station were also obtained (4). Sample preparation, extraction, clean-up techniques, analysis, and quantification are described in detail elsewhere (e.g. 2-4). Sediment dating and accumulation rates were based on 210-Pb, 137-Cs and 134-Cs determinations for the EW core, and on 210-Pb for Lake Ontario cores.

RESULTS AND DISCUSSION

Total (Σ) PCB flux and concentration profiles are presented in Figure 1 (a-c). Figure 1 d presents estimated trends in PCB production for the UK and US (5, 6).

Figure 1: Historical time-trends of PCBs determined in different matrices



Trends, fluxes and concentrations

The different historical monitoring techniques obviously gave different concentrations and fluxes, dependent on location, but generally good agreement on the trends and their dates. This is also summarised in Table 1.

Detection of PCBs in the early samples varied between 1925 to 1932. PCBs were

not manufactured until 1929, so the presence of PCBs in the early samples probably reflects mixing of more recent sediment to depth by benthic organisms and not the presence of 'pre-industrial' PCBs. In each of the sediment cores the apparent onset in PCB levels is related to sedimentation rate, i.e. the lower the accumulation rate, the greater the chronological effect of bioturbation, and therefore the earlier the apparent onset in PCBs.

Table 1: Summary of the sample dates when PCBs were first detected and were present at the maximum concentrations, the ratio of initial to present, and maximum to present fluxes, and the temporal resolution of the different techniques.

Matrix and <u>location</u>	Year o <u>Onset</u>		Ratio of fluxes <u>Pres : onset</u>	<u>Pres : Max</u>	Temporal <u>Resolution (yrs)</u>
<u>UK</u> EW	1929	1963	5.4	0.6	10
FMP	1932	1964	14	1.1	7
HERB	na	1967	na	0.02	1
<u>USA</u> E-30	1925	1969	21	0.34	4
G-32	19 27	1970	10	0.35	4

The period of maximum input showed relatively good agreement for all methods (1963 - 1970). Consequently, input profiles correspond well with production and usage data (Figure 1d). Some of the matrices gave a well defined and chronologically distinct peak input, e.g. FMP and HERB. However, maximum PCB inputs to the EW core seemingly lasted for a decade (1953 to 1963), probably due to either benthic mixing or core bulking (2).

Substantial declines in UK and US PCB levels have occurred over the past 2 decades. PCBs in annually cut herbage have declined by 99% since the 1967 maximum. Despite an enhancement in loadings at the actively growing surface layers, sections of FMP immediately below the living material demonstrate a decrease of 59% from peak inputs. Similarly, surface sections of the EW sediment core showed a reduction of 39%. Inputs to Lake Ontario have also shown a large decrease in loadings since the late-1960s/early-1970s, giving flux

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reductions in both cores since the peak of ~65%. Distinct differences in the mixture of PCBs were apparent between some of the matrices. Sediments and peat are dominated by the higher molecular weight congeners (e.g \geq 5 chlorine atoms), whereas the lower chlorinated species dominated the herbage samples.

Comments on historical monitoring techniques

Each method has certain advantages and disadvantages, and will provide information on different locations and environmental compartments. It is therefore important to collect historical time trend data from different sources to build up an integrated picture of environmental change. Sediment cores, for example, are useful in the retrospective analysis of hydrophobic time-trends and provide information on an environmental PCB 'sink'. However, possible destruction of temporal resolution by mixing, disturbance of stratigraphy during sampling, and costs of dating are drawbacks associated with this method. Losses of low-Cl PCB congeners incurred during settling through the water column tends to bias toward the accumulation of heavier congeners (7). Ombrotrophic peats derive their nutrients solely from atmospheric sources and with organic matter contents in excess of 90%, will bind hydrophobic pollutants strongly. Advection of 210-Pb and 137-Cs can lead to problems with dating (8). Peat can be a difficult matrix to work with analytically and requires extensive preparation and clean-up. Well preserved and detailed archived samples are rare and sitespecific.

Collectively the information summarised here provides good evidence that the concentrations of PCBs in some compartments of the contemporary environment of temperate industrialised countries are declining. The issues still to be resolved are: 1). is this translating into a measureable reduction in top predator species? 2). where are the PCBs moving to - ocean systems or colder polar regions? and 3). when will equilibrium concentrations be reached and what will they be?

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