PCB congener dynamics in a heavily industrialized river catchment

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Abstract

PCB congener concentrations in the water column of a highly industrialized river catchment, the Aire/Calder, in N.E. England were determined weekly on a routine basis, and 2 hourly through selected high flow (flood) events. Bed, suspended and floodplain sediment PCB congener concentrations were also determined along transects of the rivers investigated. Weekly monitoring revealed that the sum of 11 quantified (Σ11) PCBs rose in concentration by two orders of magnitude during late summer compared to their winter minimum values. This rise was concurrent with sustained periods of low flow. ΣPCB concentrations were rapidly diluted during high flow (flood) events. Suspended sediment was, on average, 13 times more contaminated with PCBs than bed sediment, with means of 4.0 and 53.8 ng/g, respectively, while floodplain samples had an intermediate concentration of 29.8 ng/g. Principle components analysis (PCA) of congener profiles showed that all three sediment types were similar, but that congener profiles differed considerably between sediment and whole-water samples. There was no change in the percentage contribution of individual PCB congeners apparent from weekly whole-water monitoring. However, the congener pattern in whole-waters changed systematically during high flow events. PCA showed that whole-water samples collected during high flow events had progressively more sediment characteristics, and then returned to whole-water characteristics on cessation of the event. The PCA evidence, dilution of PCB concentrations during events, and suspended sediments more contaminated than bed sediments, indicate that the major sources of PCBs in this catchment are current inputs from sewage treatment works, rather than remobilization of bed sediments.

Keywords: Aire; Calder; Humber; Sediments; Pate events; PCBs; Yorkshire
1. Introduction

Urban catchments are frequently contaminated with PCBs (Chevreuil and Granier, 1991; Chevreuil et al., 1987, 1990; Durell and Lizotte, 1998; Loganathan et al., 1997; Pham and Proulx, 1997; Quemarais et al., 1994; Teil et al., 1998). Major current sources to urban rivers include sewage treatment works (STWs) and combined sanitary overflows (CSOs) (Chevreuil et al., 1990; Durell and Lizotte, 1998; Loganathan et al., 1997; Pham and Proulx, 1997), although re-suspension of contaminated bed sediments and direct urban runoff may also be major sources (Loganathan et al., 1997). To enable water quality management decisions to be made at the catchment level, it is essential to identify the relative contribution of particular sources to river water column concentrations. Whilst there have been detailed studies of point source release of PCBs from STWs and CSOs (Chevreuil et al., 1990; Durell and Lizotte, 1998; Loganathan et al., 1997; Pham and Proulx, 1997; McIntyre et al., 1981), the contribution of such point sources to overall PCB catchment scale dynamics is less well understood.

In this study, we report the PCB dynamics of a highly industrialized catchment in N.E. England, drained by the Rivers Aire and Calder (Meharg et al., 1999). Fine scale (2 hourly) sampling and coarse scale (weekly) monitoring of PCB fluxes of 11 PCB congeners were undertaken. Sediment samples (suspended, bed and floodplain) were collected along transects of the catchments and also had these 11 PCBs quantified. The PCB congener profiles were analyzed by multivariate techniques to determine PCB water column concentration dynamics under high flow and low flow conditions.

2. Materials and methods

2.1. Sampling

Whole-water samples (water and suspended sediment from the water column) were collected from the Rivers Aire and Calder at weekly intervals between February 1995 and 1997. Additional samples were collected during high river flow events. The Aire was sampled from a fixed position in its lower reaches, upstream of the tidal limit (site 1, Fig. 1), but as close to this limit as practically possible given the constraints imposed by access and by sampling protocols. The Calder was sampled just above its confluence with the Aire (site 12, Fig. 1). Water samples, were from mid-river in well mixed reaches of the rivers. Samples were collected in 500-ml glass bottles with airtight PTFE lined tops which were pre-rinsed with HPLC grade acetone/hexane and dried with a stream of electron capture (ECD) grade nitrogen (Distillers MG, UK) prior to use. Replicate river water samples were also collected for sediment assessment.

An integrated event monitoring system was designed, using commercially available components to provide a system capable of in situ monitoring of turbidity as well as collecting whole-water samples throughout hydrological events using an automatic sampler (Epic 1011 wastewater sampler, described in detail by Wass et al. (1997). This was deployed on the Calder at site 12 (Fig. 1).

Bed, suspended and floodplain sediments were collected during 6 sampling campaigns from January to July 1998. Sampling locations are outlined in Fig. 1. Bed sediment samples were collected during two visits in April and July 1999. Suspended sediment samples were collected between March and June 1998 during high flow conditions (when suspended sediment was typically between 100 and 1000 mg/l). Samples were collected in 20–25 l containers from the center of the channel using a submersible pump. Upon return to the laboratory, the sediment was separated from the bulk water by continuous-flow centrifugation. The resultant slurry was concentrated by standard centrifugation. Overbank floodplain sediment samples were collected during January and March 1998. Samples we collected by deploying AstroTurf mats prior to flooding events. After the floodwaters had receded, the mats were collected and the sediment collected on the mats removed.

2.2. Sample preparation for PCB determinations

For whole-water samples, the contents of the sample bottle were mixed by shaking and inversion
and a 500 ml sample was transferred to a 500-ml volumetric flask without filtering. The contents of the flask were then sequentially extracted with $4 \times 10$ ml of hexane (HPLC grade, Rathburns, Scotland) and the hexane extracts were transferred to a Kuderna-Danish flask via a funnel containing anhydrous sodium sulphate. The sodium sulphate was rinsed with 10 ml of hexane, which was also collected in the Kuderna-Danish flask. The extract was reduced to approximately 1.5 ml at 80°C and then further reduced to exactly 1 ml using a stream of nitrogen (ECD grade).

Sediment samples were Soxhletted using chromatography grade 1:1 mixture of acetone:hexane for 8 h and the resulting extract cleaned up and concentrated in a similar manner to whole-water extracts.

Extracts from both whole-waters and sediments were cleaned up using alumina column chromatography. The alumina column consisted of an 8 mm i.d. glass column plugged with silanised glass wool and packed with 0.8 g of alumina. The alumina (Aluminium Oxide, neutral, Brockman grade 1, Merck, UK) was prepared by heating in a furnace at 700°C for 4 h, followed by deactivation with 5% (w/w) water. The whole extract was placed on the alumina column and eluted with hexane. The first 5 ml of the eluant was collected and
reduced to exactly 0.5 ml under a stream of ECD grade nitrogen. An internal standard (2,6-dichlorobenzonitrile) was added to the cleaned up extract.

For whole-water sampling, a blank sample bottle (field blank) was included with the test sample bottles (1 blank to 4 samples) during the sampling procedure to take account of contamination during transport and from exposure to the atmosphere whilst sampling. In the laboratory, the blank bottle was rinsed with 5×10 ml aliquots of hexane. This hexane extract was then treated in the same way as those for the samples. Recovery tests were performed for every 3 batches of river waters. Distilled water (500 ml) was spiked with 1 ml of hexane containing all of the analytes and shaken overnight before extraction. A portion of the spiking solution was kept for analysis with the recovery sample. Limits of detection (l.o.d.) were determined for each batch, and were calculated as 4 times the standard deviation of the blank noise at the point in the chromatograph where the compound eluted. Recoveries for whole-waters and sediments are reported in Table 1. Samples were not corrected for recoveries.

2.3. PCB analysis

The GC-ECD model was a Varian 3400 gas chromatograph interfaced with a Varian 8200CX autosampler, split/splitless injector and electron capture detector. The column was a 50 m×0.22 mm i.d. column (SGE [UK] Ltd) connected with glass press-fit connector to a 5 m×0.22 mm i.d. non-polar deactivated retention gap (SGE [UK] Ltd). A single 5 μl injection was used per run. The injector was held at a temperature of 200 °C and operated in split/splitless mode with the split valve opening after 2 min. The detector temperature was 250 °C, and the detector make-up gas was ECD grade nitrogen at 25 ml/min. The carrier gas was hydrogen with an average linear velocity of 450 mm/s. The identity of a chromatographic peak was found by comparison of its relative retention time with the relative retention times of peaks in the PCB standard. Eleven PCBs (Table 1) were quantified.

Quantification was by the internal standard method. Samples were run in batches of eight. Standards were run before and after the samples and the response factor used was an average of the two. Blanks were run before and after standards to check for carryover.

2.4. Measurement of instantaneous flow

Flow data recorded at 15 min intervals were available from the sites where water samples were collected for the weekly monitoring sites. The flow measurements were from the ultrasonic flow gauges on the Calder and a broad-crested weir velocity area gauging station on the Aire. Suspended solid concentrations were determined, as outlined by Leeks et al. (1997), to investigate if HCH dynamics could be accounted for by sediment loadings.

2.5. Statistics

General linear modelling (GLM) and principle components analysis (PCA) were used to analyze the spatial and temporal patterns in the data (MINITAB v.11, MiniInc., PA). To enable investigation of the temporal patterns in routine weekly whole-water monitoring, data were classed by month and sampling year, with on average 3.5 measurements per determinant for each month.

3. Results

3.1. ΣPCB concentrations

ΣPCB concentrations in whole-waters showed strong seasonal cycling, tending to increase by ∼2
orders of magnitude on the Calder during late summer/early autumn rising to almost 10 ng/l, concurrent with sustained periods of low flow and high water temperatures (Fig. 2). The seasonal pattern is less pronounced on the Aire, but still apparent. The Calder is a major tributary of the Aire (Fig. 1), contributing over half of the Aire’s water flow (Table 2). The GLM analysis of ΣPCB concentrations showed that ΣPCB dynamic was significantly explained by water temperature ($P = 0.002$), river (Aire and Calder) ($P = 0.033$), month ($P = 0.048$) and year ($P = 0.002$); with only the suspended sediment and river flow terms not being significant.

The flux of PCBs was greater during the second monitoring year (Table 2). This increase in flux may be potentially due to the unusually low river flows observed during the winter of 95/96, which may have had consequences for PCB residence times in the catchment (Fig. 2). The flux of PCBs measured on the Calder near its confluence with the Aire was equivalent to that determined at the mouth of the Aire, emphasising the considerable contribution that the Calder makes to the Aire's PCB flux (Table 2).

Fluctuations in ΣPCB concentrations and flux during a representative (4 other events were monitored) hydrological event on the Calder, showed

![Graph](image_url)

Fig. 2. Σ11 PCB concentrations (histogram bars) and flux (triangles), instantaneous flow (solid line), suspended sediment (dashed line) and temperature (grey line with crosses) for monthly averages over the 2 year of monitoring on the Aire and Calder Rivers.

<table>
<thead>
<tr>
<th>River</th>
<th>Year</th>
<th>Instant flow (m$^3$/s)</th>
<th>Suspended sediment (mg/l)</th>
<th>PCB concentration (ng/l)</th>
<th>PCB flux (g/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aire</td>
<td>1</td>
<td>26.2</td>
<td>13.2</td>
<td>0.53</td>
<td>14.7</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>28.2</td>
<td>15.0</td>
<td>1.45</td>
<td>25.2</td>
</tr>
<tr>
<td>Calder</td>
<td>1</td>
<td>13.6</td>
<td>12.2</td>
<td>1.1</td>
<td>12.2</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>20.13</td>
<td>22.2</td>
<td>3.18</td>
<td>22.2</td>
</tr>
</tbody>
</table>

Table 2
Mean annual (calculated using monthly averages) instantaneous flow, ΣPCB concentration, suspended sediment concentrations and ΣPCB flux for the Aire and Calder Rivers for each of the 2 sampling years.
a rapid decline in $\Sigma$PCB concentrations and flux as flow increased (Fig. 3). There is evidence that $\Sigma$PCB flux and concentration increased slightly after the initial decline, concurrent with the rise to maximum suspended sediment concentration. $\Sigma$PCB concentrations and flux did not return to the initial starting values when instantaneous flow returned to its baseline level.

A survey of bed, suspended and floodplain sediments was carried out along longitudinal transects of the Aire and Calder, from rural upstream stretches of the rivers through to the heavily industrialized reaches (Fig. 4). GLM analysis of log$_{10}(y+1)$ transformed data showed that there was no significant trend in $\Sigma$PCB with distance from the lowest sampling point. Site and site×sediment type interaction not significant the 5% level, but the sediment type term was significant ($P=0.011$). Based on the results of this GLM, a one-way analysis of variance was performed on the data using sediment type as the comparison and this showed that suspended sediment had a
significantly higher ($P=0.004$) level of $\Sigma$PCB than bed sediment, with a geometric mean of 53.8 ng/g ($n=18$) and 4.0 ng/g ($n=11$), respectively. Floodplain sediment had an average $\Sigma$PCB level of 29.8 ng/g, and was not significantly different from bed or suspended sediment. Suspended sediment concentrations accounted for $\sim 50\%$ of whole-water $\Sigma$PCB concentrations (calculated from Table 2).

3.2. PCB congener patterns

Congener data (expressed as a percentage of $\Sigma$PCB) from routine whole-water sampling showed no trends or clustering with time, river or season when analyzed by PCA (data not shown). Similarly, bed, suspended and floodplain congener profiles showed no pattern based on site of origin or sediment type. When the 2 data sets were analyzed together (Fig. 5), the PCA distinguished the sediments from the whole-water samples, with limited overlap. Interestingly, the only three sediment samples (suspended and floodplain) on the right hand axis of the PCA1 vs. PCA2 plot were samples from upstream, rural locations. Only one water sample had a PCB profile characteristic of sediment. This was a sample collected from the Calder in November of the second year of sampling.

On the rationale that PCA showed no clustering within whole-water or sediment samples, congener profiles for all whole-water samples were averaged, as were those for sediment samples, to compare gross differences in congener patterns between rivers and sediments (Fig. 6). PCB 77, 118, 138, 153 and 159 were roughly equivalent in contribution to $\Sigma$PCB in both sediments and waters, while PCB 101 and 126 had higher percentage contribution in whole-waters, and PCB 128, 149, 170 and 180 had higher contributions in sediments. The ratio of individual congener averages for percentage contribution to $\Sigma$PCB in the whole-water/sediment were plotted against $K_{oc}$ and solubility (data not shown). There were no clear trends with these properties, indicating that sediment and whole-water sample differences in PCB profile was not simply explained by partitioning between the aqueous and sediment phase.

Plots of percentage contribution of individual PCBs during a hydrological event showed that...
congeners exhibited several distinct patterns of a number of behaviour through the event (Fig. 7). PCBs 77, 128 and 180 increased in their contribution as instantaneous flow increased to its maximum and then decreased when instantaneous flow decreased (Fig. 7). The percentage increase in contribution of these congeners was substantial, with PCB 128, for example, increasing from <5% \( \Sigma \text{PCB} \) at the start of the event to >30% at the height of the event. Congeners 128 and 180 are highly diagnostic of sediment (Fig. 6), and the rise and increase in these congeners suggest a greater contribution of sediment to PCB loadings at the height of the event. Congeners 118 and 153 (and perhaps 138) increase rapidly in contribution to the peak of the instantaneous flow, and then remain elevated until the end of monitoring. These congeners 118 and 153 (and 138) have approximately equal weighting in terms of their contribution to \( \Sigma \text{PCB} \) for sediments and whole-waters (Fig. 6). PCB 169 increases in contribution following attainment of maximum instantaneous flow and stays elevated to the end of monitoring. Again, this congener has an equal percentage contribution to \( \Sigma \text{PCB} \) in sediment and whole-waters. Congener 126, which is diagnostic of whole-water samples was below the l.o.d. for all samples during this event.

The PCA presented in Fig. 5 shows the same event presented in Fig. 6 plotted over time in relation to data for whole-waters and sediment samples. The sample at time 0 h is on the divide between sediment and whole-water character. As instantaneous flow increases the event samples move progressively towards having sediment character, where they stay while instantaneous flow plateaus. Towards the end of this plateau the PCB congener composition returns to the boundary between sediment and whole-water character, completing a full cycle, and the PCB profile stabilizes for the remainder of the event as the high instantaneous flow rates decrease to the baseline level.

4. Discussion

There is a strong seasonal dependence of \( \Sigma \text{PCBs} \), with highest concentrations attained in both rivers after periods of extended low flow, when sediment loadings of whole-waters are low. Dramatic dilution of \( \Sigma \text{PCB} \) concentrations occurs during storm events, when sediment loadings of whole-waters increase considerably. Therefore,
sediments mobilized during high flow events are not the dominant source of PCBs in this catchment. Also, $\Sigma$PCB concentrations in suspended sediment are, on average, 13 times higher than bed sediment along the entire length of the river surveyed. Suspended sediment has a lower grain size compared to bed sediment, and therefore, the higher surface area may account for the higher concentrations of PCBs associated with suspended sediment. All the evidence points towards STWs being the dominant source of PCBs in this catchment. The Rivers Aire and Calder drain catchments dominated by the highly industrialized urban centres of Bradford and Leeds (Meharg et al., 1999) Fig. 1. Industrial effluent in these catchments is discharged directly into STWs without pre-treatment (Meharg et al., 1999), and the STWs also receive urban run-off and human waste. All of these sources will contribute to PCB loadings of STWs. Studies of dieldrin (Meharg et al., 2000a) and HCH isomers (Meharg et al., 1999) dynamics within the Aire/Calder catchment show similar
seasonal fluctuations as PCBs, and it is known that STWs are the primary source of these chemicals in this catchment (Meharg et al., 1999, 2000a). In contrast to what can be considered as contaminants associated with historic usage (dieldrin, HCHs and PCBs), chlorobenzenes which still have large volume industrial and domestic usage, have very different dynamics in this catchment, showing no seasonal cycling in concentrations (Meharg et al., 2000b).

Most studies of PCB temporal dynamics in both industrialized and rural rivers have shown a strong seasonal cycling. Bremle and Larsson (1997) studying the dynamics of PCBs in a rural Scandinavian catchment reported $\Sigma$PCB in whole-water samples rising in the summer and decreasing during winter. They explained this cycling by dilution to precipitation levels during high flows and internal sediment recycling during low flows. This hypothesis was backed up by PCA analysis of congener patterns which showed that water samples had rain water characteristics during high flows and sediment characteristics during low flows. No such seasonal/water flow regulation of congener profiles was observed in the present study. Larsson et al. (1990) investigated PCB fluxes on a remote Swedish river and lake system contaminated by a paper mill. $\Sigma$PCB concentrations increased at low flows, indicating that partitioning between contaminated sediments and the aqueous phase had a major influence on PCB burdens. Furthermore, during periods of high discharge, sediment content increased but release of PCBs from sediments did not. There was limited remobilization of contaminated bed sediments during events. PCB release was not directly temperature dependent, even though concentrations rose during summer. Elevation of $\Sigma$PCB levels was attributed to increases due to bio-turbation and gas convection, which were dependent on temperature. Crane and Sonzogni (1992) found that for a contaminated Wisconsin lake the lowest PCB concentrations occurred during ice-cover conditions (3.5 ng/l) while highest occurred during mid summer (140 ng/l). When their data were normalized for suspended particulate matter, these seasonal differences disappeared. Also, there were no seasonal differences in contribution of individual congeners to $\Sigma$PCB, a finding of this present study.

In contrast, total and Arachlor matched PCBs in the Seine rose sharply during winter and then decreased considerably from June onward (Chevreuil and Granier, 1991). Quemerais et al. (1994) investigated PCB dynamics on the contaminated reaches of the St. Lawrence River (and its tributaries). Major increases in $\Sigma$13 PCBs occurred in early spring associated with the dissolved phase, followed by a decrease in concentrations during summer and a slight increase in autumn associated with the particulate phase.

From the studies summarized here, PCB contaminant pools (both historic contamination of bed sediments plus fresh input from STWs etc.) interact with the hydrology of the catchment and season, resulting in PCB dynamics differing on a catchment by catchment basis.

As mentioned above, STWs have been identified as a major source of PCBs in the Aire/Calder catchment. A study by Pham and Proulx (1997) investigated the fluxes of PCBs into and out of the municipal sewage system (MSS) that served the urban island of Montreal. Two thirds of $\Sigma$13 PCBs were removed by the MSS, resulting in a net release of 1 kg/y of PCBs from the catchments which drain a population of 1.8 million people. This contrasts with the present study where the $\Sigma$PCB flux from the Aire was 5 and 9 kg/y for the 2 sampling seasons, respectively, yet the population inhabiting this catchment was 2.1 million, similar to that of Montreal. At the other extreme, the 26 water pollution control plants (WPCPs) on the New York/New Jersey Harbor Estuary released 88 kg/y to the estuary (Durell and Lizotte, 1998).

In the present study, whole-water samples decreased dramatically during storm events. Durell and Lizotte showed that storm events contributed little to PCB inputs into the New York/New Jersey harbor Estuary, and accounted for only approximately 3% of PCB export to the estuary (Durell and Lizotte, 1998). WPCPs influent $\Sigma$PCB concentrations also only rose slightly during storms. PCB congener patterns differed between plants, and were fairly consistent between sampling at low flow conditions, but during high flow (storm)
events influent samples had slightly different PCB profiles suggesting additional sources during such events. Influent and effluent profiles were generally very similar. Chevreuil and Granier (1991) working on the Seine showed that storm flow caused very large increases in PCB concentration and flux in storm drains, which they attributed to runoff from impermeable hard surfaces, where build-up of PCBs had occurred due to dry atmospheric fallout. Loganathan et al. (1997), demonstrated that CSOs congener profiles reflected the congener profile of street dusts, indicating that these were a major PCB source to CSOs during rain events. In their study, the PCB congener pattern of sanitary flow differed greatly from CSOs. Σ35 PCB congeners was 3- to 9-fold higher in CSO compared to sanitary flows, with CSOs dominated by heavier congeners, as was street dust. The catchment that was investigated in the present study showed considerable dilution of ΣPCBs during high flow events in the open river, with the whole-water samples having congener profiles akin to sediments during such events. Even if CSOs are an additive source of PCBs to the catchment during spate events, their contribution to overall PCB flux is limited.

The fact that suspended sediment and floodplain deposits were characterized by higher PCB concentrations than bed sediment on the Aire and Calder indicates (a) that suspended sediment and floodplain deposits are not dominated by re-suspension of bed sediments and (b) that extensive sediment deposition from the sediment phase does not occur over the stretches of the Aire and Calder investigated. The only other explanation for the low levels in bed sediments is high rates of loss of PCBs through biotic and abiotic processing, but this seems unlikely given the long environmental half lives of the PCBs. All the evidence points towards terrestrial sources (through STWs, CSOs and direct run-off) dominating contribution to suspended sediment and floodplain deposits. As PCB congener profiles are similar on both the Aire and Calder, and in all three sediment types investigated, this suggests that the similar sources dominant PCB contamination of these catchments.

5. Conclusions

We show here that PCB concentrations in whole-water and suspended sediment samples collected from an industrialized catchment are dominated by inputs into the river from terrestrial sources, rather than re-suspension of bed sediments. As PCB concentrations in suspended bed sediment were much higher than those in bed sediment along the entire reaches of both rivers, inputs of sediments from terrestrial sources must have short residence times over the reaches investigated. As there is no major remobilization of PCBs during spate events, the normal flux from STWs is again responsible for the vast majority of PCB contamination of this catchment. If water quality with respect to PCB contamination is to be improved in such industrialized regions, the contamination cause by STWs must be addressed.

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