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Factors Influencing Atmospheric Concentrations of Polychlorinated Biphenyls in Birmingham, U.K.

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Atmospheric concentrations of PCBs were measured in Birmingham, U.K., between 7/13/97 and 7/27/98. A significant positive relationship (>95%) was detected between the cosine of the wind direction and ln *P* for ΣPCB and tri- and tetrachlorinated PCBs. This is consistent with input from northerly winds passing over Birmingham city center, 3 km from our site. Plots of the enthalpy of surface-air exchange (ΔH_{SA}) versus laboratory-measured heats of vaporization (ΔH_v) for all congeners revealed a highly significant (>99.9%) positive linear relationship. According to Simcik et al. (*1*), this indicates local sources to be the predominant influence. However, separate plots of ΔH_{SA} versus ΔH_v for (a) the tri- and tetrachlorinated PCBs and (b) the penta-, hexa-, and heptachlorinated congeners revealed a highly significant linear relationship for the latter group but ΔH_{SA} to be essentially constant for the former. Strict interpretation of Simcik et al.'s hypothesis (*1*) suggests long-range transport as the principal source of tri- and tetrachlorinated PCBs but not of the higher chlorinated congeners. This is not thought credible given the nearby influence of the city center on concentrations of the lower chlorinated congeners, and we suggest correlation between ΔH_{SA} and ΔH_v will only be observed when the principal source is both close to the sampling site *and* governed by equilibrium surface-air partitioning. We therefore believe that at our site, the principal source of tri- and tetrachlorinated PCBs is nonequilibrium short-range transport, while for penta- through heptachlorinated PCBs, equilibrium short-range transport is the predominant source. Indoor air ventilation is a short-range source governed by factors such as the building ventilation rate, rather than surface-air partitioning. We therefore hypothesize that it constitutes the principal source of tri- and tetrachlorinated PCBs at our site.

Introduction

Despite stringent restrictions on the use and manufacture of polychlorinated biphenyls (PCBs) since the late 1970s, continuing leaks from PCB-filled equipment remaining in use, along with the persistence of these chemicals, means that they still maintain a significant environmental presence. Although direct human exposure via inhalation of outdoor air is not a significant exposure pathway, the atmosphere represents the primary vector by which PCBs enter the grass−cattle−human food chain, which is responsible for a significant proportion of human exposure (*2*). As a consequence, it is important to monitor ad improve understanding of the factors that influence atmospheric PCB concentrations.

This study reports levels of individual PCB congeners in samples of air collected at a background urban site over 41 separate 24 h periods between 7/13/97 and 7/27/98. These levels are placed in context with those reported both in other U.K. locations and elsewhere (*3*−*17*). The influence on atmospheric PCB concentrations of rainfall, air temperature and humidity, wind speed, and direction is examined. Plots of the natural logarithm of PCB partial pressure versus reciprocal temperature are interpreted to provide clues as to the principal sources of atmospheric PCB at this location.

Experimental Section

Sampling Location. Samples were taken every 1−2 weeks on the campus of Birmingham University, about 3 km southwest of the city center of Birmingham, U.K. Birmingham is the major city within the West Midlands conurbation, a major urban center with a population of ca. 2.5 million. Sampling equipment was located at ground level ca. 20 m from the nearest building, well clear of any building air outfalls.

Air Sampling. Our sampling procedures for determining PCBs in air have been reported previously (*18*). In summary, samples were taken using a Graseby-Andersen Hi-Vol sampler modified to hold a glass-fiber filter (GFF, 0.6 μm pore size) and a precleaned polyurethane foam (PUF) plug. Sampling was conducted for 24 h at a flow-rate of 0.7 $m^3\ min^{-1}$ yielding sample volumes of ca. 1000 m^3.

Sample Purification and Analysis. Although data are reported here as the sum of vapor and particulate phases, GFFs and PUFs were analyzed separately. Analyses were conducted using well-validated, containment enrichment, GC/MS procedures reported elsewhere (*18, 19*). Mean recoveries of quantitation standards added to check analyte losses during both sampling (sampling efficiency standards (SESs)—PCB congeners 19 and 147) and analysis (PCB congeners 34, 62, 119, 131, and 173) ranged between 67 and 92% for all samples. Concentrations were not corrected for SES recoveries. The limits of detection for individual congeners (typically 0.1 pg m^{-3}) were essentially defined by the levels detected in method blanks. In all, 15 method blanks were conducted, with mean PCB concentrations in blanks typically 6−7% of those in samples. All samples were corrected for the mean blank concentrations, and a sample concentration only recorded if it exceeded twice the mean blank level. Note that ΣPCB refers to the sum of all trichlorinated through heptachlorinated PCB congeners detected in a sample.

Determination of Meteorological Parameters. Automatic monitoring of air temperature (*T*), relative humidity (RH), rainfall (RF), wind direction (WD), and wind speed (WS) was conducted during all sampling events. Until 5/6/98, these data were collected from a weather station located ca. 400 m from our air sampling site. After this date, meteorological data was measured by a station colocated with our sampling equipment. At both stations, hourly averages were taken of measurements made every 20 s for *T* and RH and every 15 min for RF, WD, and WS.

Results and Discussion

Comparison of Levels with Other Studies. Table 1 compares ΣPCB levels detected in this study with those reported elsewhere. While levels in Birmingham exceed the majority of data reported for rural locations in the U.K. and North

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TABLE 1. Atmospheric Concentrations of ΣPCB at Various Locations (Sum of Vapor and Particle Phases)

study	arithmetic mean and range (pg m^{-3})	sampling details
this study	290 (76–1000)	Birmingham, U.K. (urban) 97–98
3	422	Denver, U.S. (urban) 85–86
4	40–20 800	Bloomington, U.S. (urban) 86–88
5	65–2530	Bloomington, U.S. (urban) 93
6	670	Ulm, Germany (urban) 87
7	75–850	Kiel, Germany (urban, during rainfall only)
8	55–823 (monthly means)	Egbert, Canada (rural) 88–89
9	1350 (413–3850)[a]	London, U.K. (urban) 91–92
9	370 (141–1840)[a]	Stevenage, U.K. (urban) 91–92
9	1490 (415–3710)	Cardiff, U.K. (urban) 91–92
9	1160 (223–2360)	Manchester, U.K. (urban) 91–92
10	1184 (1090–1450)[b] (annual means)	London, U.K. (urban) 91–95
10	400 (340–460)[b] (annual means)	Manchester, U.K. (urban) 91–95
11	128	Great Lakes, U.S. (rural) 91–95
11	160	Great Lakes, U.S. (rural) 91–95
11	315	Great Lakes, U.S. (20 km from Buffalo)
12	2330 (270–14 200)	Chicago, U.S. (urban) 94–95
12	480 (230–780)[c]	U.S. (rural) 94
13	100–8200	New Bedford, U.S. (urban) 94–95
14	350 (270–910)	Chicago, U.S. (urban) 95
15	58–288	Northwest U.K. (rural) 96
16	150 (99–199)	Eastern U.K. (rural) 96–97
17	46 (17–78)	Northwest U.K. (rural) 95–96
17	141 (48–268)	Northwest U.K. (rural) 95–96
17	471 (156–1155)	Northwest U.K. (rural) 95
17	217 (55–2253)	Northwest U.K. (rural) 96

[a] Sum of congeners 28, 52, 77, 101, 118, 138, 153, and 180 only. [b] Sum of congeners 28, 52, 101, 118, 138, 153, and 180 only. [c] Vapor phase only.

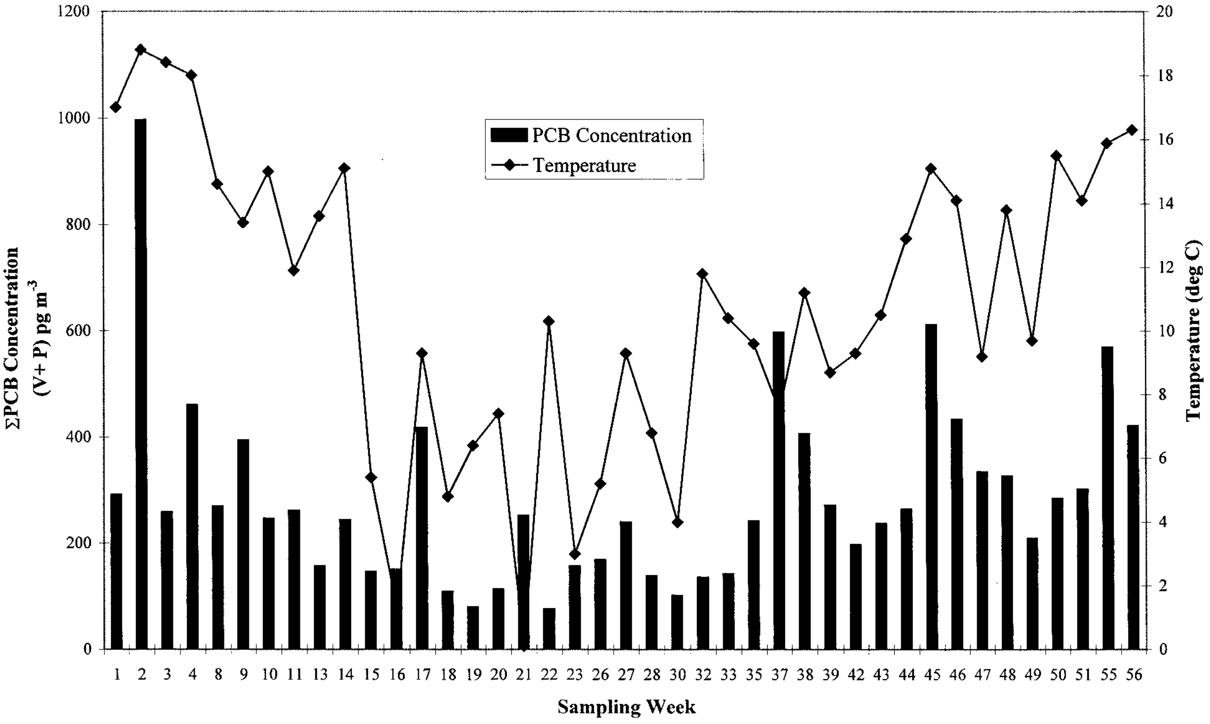

FIGURE 1. Variation of ΣPCB with air temperature.

America, they are at the lower end of those reported for urban areas. This may be due to the noncity center location of our sampling site.

Seasonal Variation. Figure 1 plots the variation of ΣPCB with mean air temperature for each individual sampling event. PCB concentrations in April to September inclusive

TABLE 2. Regression Parameters for $\ln P = m_1(1/T) + b_1$ Plots for Individual PCBs

congener	m_1	b	ΔH_{SA} (kJ mol^{-1})	congener	m_1	b	ΔH_{SA} (kJ mol^{-1})
18	−6898	−10	57 ± 13	**83**	−6102	−17	51 ± 18
32/16	−5338	−16	44 ± 12	**97**	−7906	−9	66 ± 13
31/28	−6239	−12	52 ± 12	**87**	−7801	−9	65 ± 13
33	−5720	−14	48 ± 12	**111**	−5673	−17	47 ± 11
22	−5945	−14	49 ± 14	**110**	−8100	−7	67 ± 13
53	−5089	−19	42 ± 14	**82**	−6417	−15	53 ± 16
51	−4347	−22	36 ± 13	**118**	−8698	−5	72 ± 16
45	−5552	−17	46 ± 15	**105**	−7361	−11	61 ± 18
52	−5566	−14	46 ± 12	**148**	−8352	−7	69 ± 13
49	−4421	−19	37 ± 14	**151**	−9679	−3	80 ± 12
47	−5429	−17	45 ± 15	**135/144**	−8987	−6	75 ± 14
44	−4466	−19	37 ± 12	**149**	−9229	−3	77 ± 13
42	−3684	−23	31 ± 14	**153**	−10794	2	90 ± 21
41/64	−5325	−16	44 ± 11	**132**	−9151	−5	76 ± 17
40	−3248	−26	27 ± 16	**141**	−8735	−7	73 ± 19
74	−5757	−16	48 ± 11	**138/164**	−7022	−11	58 ± 15
70/76	−6152	−13	51 ± 11	**179**	−11259	2	94 ± 12
66	−5753	−15	48 ± 13	**176**	−10617	−1	88 ± 14
56	−5051	−18	42 ± 11	**187/182**	−12724	7	106 ± 18
95	−6061	−14	50 ± 15	**183**	−11908	4	99 ± 15
91	−6892	−13	57 ± 13	**174**	−11257	2	94 ± 15
84/92	−6370	−14	53 ± 12	**177**	−8838	−7	73 ± 20
90/101	−7659	−8	64 ± 12	**180**	−13475	10	112 ± 18
99/113	−7908	−8	66 ± 12	**ΣPCB**	−6323	9.5	53 ± 11

(mean concentration = 380 pg ΣPCB m^{-3}; mean temperature = 13.9 °C) clearly exceed those in October to March inclusive (mean concentration = 210 pg ΣPCB m^{-3}; mean temperature = 7.6 °C); with a two-tailed *t*-test showing the difference between April to September and October to March mean ΣPCB concentrations, to be significant at the 99.7% level. This "summer peak" in PCB concentrations is consistent with other studies (*4, 5, 9−11, 20*).

Influence of Temperature on Atmospheric PCB Concentrations. Thermodynamically, the vapor-phase behavior of PCBs can be described in terms of the Clausius−Clapeyron equation

$$\ln P = \left(\frac{-\Delta H_v}{R}\right)\left(\frac{1}{T}\right) + \text{const} \qquad (1)$$

where P is the partial pressure (atm), T is the temperature (K), ΔH_v the heat of vaporization (kJ mol^{-1}), and R the gas constant.

Hence, regression of $\ln P$ against $1/T$ should be linear with negative slope m_1 and intercept b_1.

$$\ln P = m_1\left(\frac{1}{T}\right) + b_1 \qquad (2)$$

Partial pressures of each individual PCB listed in Table 2 were calculated for each sample from gas-phase concentrations using the ideal gas law (for ΣPCB, an average molecular mass of 326.4 was assumed). Natural logarithms of these partial pressures were plotted against reciprocal mean temperature for each sampling event. Figure 2 shows the plot obtained for ΣPCB, while Table 2 summarizes the relevant data obtained from such plots for individual chromatographic peaks. For all PCBs studied, the temperature-dependence of vapor-phase concentrations was significant at at least the 90% level, with the significance level exceeding 99% for 44 out of 47 chromatographic peaks. For ΣPCB, the temperature dependence is highly significant ($R^2 = 0.37$; 99.9% significance), and the m_1 value (−6323 ± 1328 K) is within the range typically reported elsewhere (−4500 to −7500 K) (*20*).

Influence of Atmospheric Turbulence on Temperature Dependence of Atmospheric PCB Concentrations. For ΣPCB, the R^2 value indicates 37% of the variation in vapor-phase levels to be due to temperature variation. A previously noted effect is the influence of wind speed on this temperature dependence, whereby periods of atmospheric turbulence lead to a collapse of the relationship between air concentration and temperature (*21*). We therefore examined our data for evidence of such an effect, by dividing them into three groups: (i) those where the mean wind speed for the sampling period < 2.2 m s^{-1} ($n = 11$); (ii) those where 2.2 > WS < 4.4 m s^{-1} ($n = 16$); and (iii) those where WS > 4.4 m s^{-1} ($n = 14$). Clausius−Clapeyron plots were recalculated for samples in each category for ΣPCB and individual homologue groups.

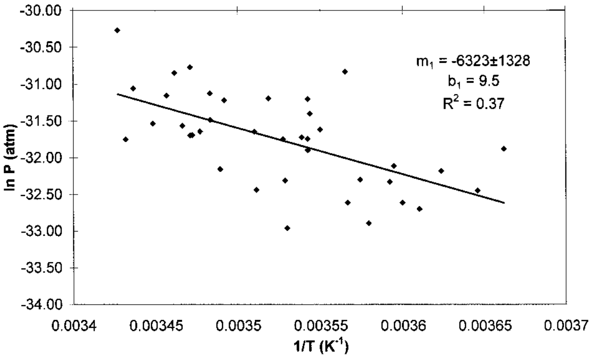

FIGURE 2. Clausius-Clapeyron plot for ΣPCB.

A clear decrease in the magnitude of m_1 with increased wind speed was observed for ΣPCB and all homologue groups, confirming the observations of other workers (*20*). When WS > 4.4 m s^{-1}, temperature dependence was lost for ΣPCB, and all homologue groups bar the heptachlorinated group. Furthermore, for samples where WS < 2.2 m s^{-1}, the R^2 value indicates that 78% of the variation in PCB vapor-phase levels was attributable to temperature variation. Evidently, wind speed is an important factor in determining the temperature dependence of atmospheric PCB levels at this site.

Influence of Other Meteorological Parameters on Atmospheric PCB Concentrations. In addition to temperature dependence, the potential influence on atmospheric PCB levels of wind speed and direction, air humidity, and rainfall has also been indicated (11). To assess the influence of these parameters on PCB concentrations at our sampling site, we introduced each into the Clausius–Clapeyron equation to yield four new equations (11):

$$\ln P = m_1\left(\frac{1}{T}\right) + m_2 \ln WS + b \qquad (3)$$

$$\ln P = m_1\left(\frac{1}{T}\right) + m_3 \sin WD + m_4 \cos WD + b \qquad (4)$$

$$\ln P = m_1\left(\frac{1}{T}\right) + m_5\, RH + b \qquad (5)$$

$$\ln P = m_1\left(\frac{1}{T}\right) + m_6\, RF + b \qquad (6)$$

Wind direction is expressed in degrees relative to true north (0°), and mean WD for a given sampling event was determined via use of trigonometric relations to calculate the direction of the sum of individual wind vectors.

Multiple regression analysis of the relationship between each of these parameters and ln P of both individual homologue groups and ΣPCB was conducted. No significant relationships exist between ln P and any of the variables studied, with the exception of the cosine of the wind direction. While there was no significant relationship (<95% significance) between ln P and the cosine of the wind direction for penta-, hexa-, and heptachlorinated PCBs, significant positive relationships were observed for ΣPCB ($m_4 = 0.23 \pm 45\%$; 96% significance) and the tri- ($m_4 = 0.27 \pm 41\%$; 98% significance) and tetrachlorinated ($m_4 = 0.22 \pm 46\%$; 96% significance) homologue groups. Positive cosine values are associated with wind directions of 270°–0°–90°, implying a statistically significant increase in atmospheric concentrations of ΣPCB, tri-, and tetrachlorinated congeners at our sampling site when winds come from the north, passing over the center of Birmingham. This bifurcation of behavior between the tri- and tetrachlorinated and the penta- through heptachlorinated homologue groups suggests the possible existence of two distinct sources of the lower and higher chlorinated PCBs at our site. The following sections examine this issue in more detail.

Enthalpies of Surface-Air Exchange as a Source Apportionment Tool. The slope m_1 of the Clausius–Clapeyron plots may be used to estimate the heat of vaporization (ΔH_v) for PCBs using eq 1. Under environmental conditions, Simcik et al. (1) point out that this term is really the energy required to cause the phase transition from surfaces such as soil, vegetation, and water to the atmosphere. They therefore referred to this energy term as ΔH_{SA}, the enthalpy of surface-air exchange. Table 2 gives ΔH_{SA} values obtained for individual PCBs from our data. The range of 27–112 kJ mol^{-1} (arithmetic mean = 61 ± 20 kJ mol^{-1}) is comparable to those derived from other field observations (5, 8, 11, 17, 22) and—in line with these studies—is lower than estimates derived from laboratory-based determinations (23).

ΔH_{SA} values can yield important information regarding the relative influence of short- versus long-range transport on atmospheric concentrations of PCBs (1, 20). Wania et al. (20) hypothesized that lower ΔH_{SA} values indicated that long-range transport controlled atmospheric levels, while higher ΔH_{SA} values were indicative of volatilization from local sources being the dominant factor. Simcik et al. presented an alternative hypothesis (1). They monitored atmospheric PCB concentrations at a number of North American locations and plotted ΔH_{SA} versus ΔH_v for each PCB congener. In essence, they argued that for a given site: (a) if such plots were linear, then short-range transport (i.e. local sources) was the predominant influence; and (b) if they were nonlinear, then long-range transport would predominate. To account for the seemingly contradictory evidence presented by Wania et al.—who detected a linear relationship between m_1 (ΔH_{SA}) and the number of chlorines (essentially reflecting ΔH_v) for three rural/remote sites but not urban sites—Simcik et al. suggested that there was no direct link between the remoteness of a site and the relative influence of short- or long-range transport to that site. Simcik et al. then reported that while the relationship between ΔH_{SA} and ΔH_v for three remote sites in the Great Lakes was nonlinear (ΔH_{SA} approached a constant value at each site), a significant positive linear relationship existed at an urban location in Chicago. They interpreted this as demonstrating the dominant influence of long-range transport at the remote locations and short-range transport at the Chicago site.

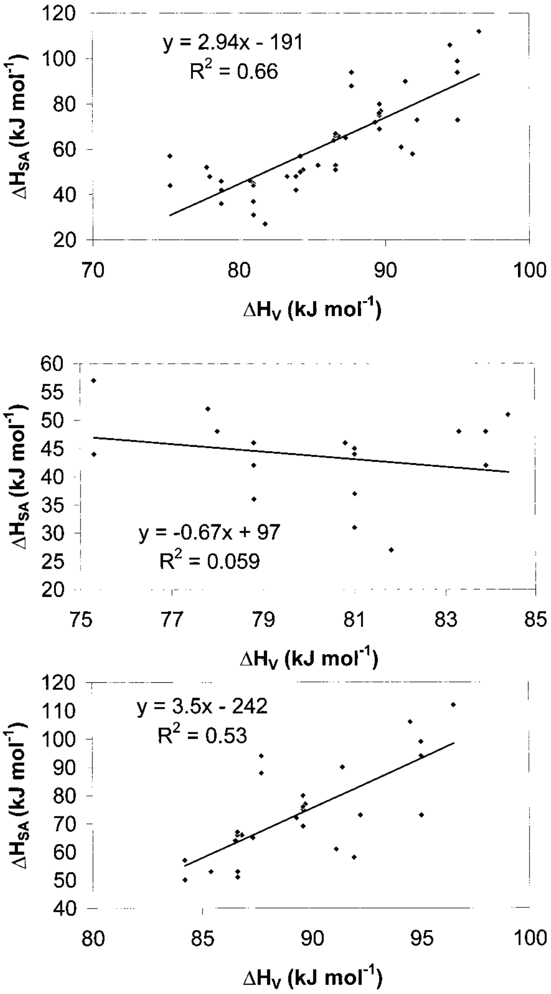

FIGURE 3. (a) ΔH_{SA} versus ΔH_v for all congeners, (b) ΔH_{SA} versus ΔH_v for tri- and tetrachlorinated PCBs, and (c) ΔH_{SA} versus ΔH_v for penta-, hexa-, and heptachlorinated PCBs.

To test these hypotheses, we plotted ΔH_{SA} versus ΔH_v (from ref 23) for our data. The resultant plot (Figure 3a) shows a highly significant (>99.9%) positive linear correlation, which according to Simcik et al. (1) is evidence that short-range transport is the predominant influence at our site. However, when we conducted separate plots of ΔH_{SA} versus ΔH_v for (a) the tri- and tetrachlorinated congeners (Figure 3b) and (b) the penta-, hexa-, and heptachlorinated congeners (Figure 3c), we observed a distinct dichotomy in behavior. While ΔH_{SA} was essentially constant for the tri- and tetrachlorinated congeners, a highly significant (>99.9%) linear relationship was evident for the higher chlorinated PCBs. Interestingly, while no such dichotomy in behavior is evident for samples arriving over land at Simcik et al.'s Chicago site, a similar effect is detectable for the over water samples. If the hypothesis of Simcik et al. is correct, and the linearity (or otherwise) of the relationship between ΔH_{SA} and ΔH_v is indicative of the relative influence of short- and long-range transport at a given location, then we believe this dichotomy of behavior to be consistent with the existence at our site, of separate sources of the tri- and tetrachlorinated congeners and the penta-, hexa-, and heptachlorinated species, respectively. Given that Simcik et al. suggested that a linear ΔH_{SA} versus ΔH_v plot for a given site is indicative of short-range transport being the predominant influence, a strict interpretation of this hypothesis, would conclude that at our site long-range transport is the principal source of the tri- and tetrachlorinated PCBs but not of the penta- through heptachlorinated congeners. We do not believe this to be a credible explanation, not least because we observed higher concentrations of the tri- and tetrachlorinated PCBs associated with winds arriving at our site having passed over the city center, only 3 km distant. Instead, we believe that a caveat must be added to the hypothesis put forward by Simcik et al. (1). In short, we believe that ΔH_{SA} will be a linear function of ΔH_v, when the PCB source is both close to the sampling site, and is governed by equilibrium partitioning between surfaces and the overlying atmosphere. Further, we contend that ΔH_{SA} will not be a linear function of ΔH_v for individual congeners, when the PCB source is close to the sampling site but is not governed by equilibrium partitioning between surfaces and the overlying atmosphere. One possible such source is the ventilation of indoor air, which Halsall et al. (9) indicated to be a potentially important source of outdoor air concentrations of PCB 52 in Manchester, U.K., and which is known to contain mean ΣPCB concentrations ca. 30 times higher than those in outdoor air in Birmingham (18). While PCB concentrations in indoor air will be governed at least partially by surface-air partitioning, the contribution of indoor air ventilation to PCB concentrations in outdoor air is controlled by the building ventilation rate. It is the latter that is important in this context. We therefore suggest that at our site, indoor air ventilation is an appreciable source of outdoor air concentrations of the tri- and tetrachlorinated PCBs, which are the dominant congeners in indoor air in our conurbation—comprising ca. 70% of the geometric mean of the sum of individual congeners (18). Conversely, we believe that interpretation of the data presented here shows the principal source of penta- through heptachlorinated PCBs is equilibrium partitioning with local surfaces such as soils. Interestingly, when we plot ΔH_{SA} versus ΔH_v for samples for which the predominant wind direction was from Birmingham city center (i.e. positive cosine of wind direction), there is no correlation for either: (a) all congeners; (b) tri- and tetrachlorinated PCBs; and (c) penta-, hexa-, and heptachlorinated congeners. This implies that ventilation of indoor air from buildings constitutes the principal source of all congeners at our site when the wind comes from the city center. For samples taken when the wind was from the south, the situation was similar to that observed for the dataset as a whole—i.e., correlation between ΔH_{SA} and ΔH_v for both all congeners and penta-, hexa-, and heptachlorinated PCBs but no correlation for tri- and tetrachlorinated congeners. For such samples, indoor air ventilation only appears significant for the tri- and tetrachlorinated PCBs. This does not imply that for such samples indoor air ventilation is not a source of penta- through heptachlorinated PCBs, rather that its contribution is less significant than that from other sources such as volatilization from surfaces. Clearly, if our suggested caveat to the hypothesis of Simcik et al. (1) is correct—and further work at other locations is needed to confirm or disprove it—then we believe that plots of ΔH_{SA} versus ΔH_v provide a powerful source apportionment tool.

This study reveals ΣPCB concentrations in Birmingham, U.K., to be at the low end of values reported for other major conurbations, although this is likely a reflection of the location of the sampling site, 3 km from the city center. Air temperature was found to exert a significant effect on PCB levels, with low wind speeds (<2.2 m s^{-1}) causing a significant increase in this temperature dependence. Evidence is presented that suggests the principal source of tri- and tetrachlorinated PCBs at our site to be ventilation of indoor air, while volatilization from surfaces such as soil and vegetation constitutes the main source of the penta-, hexa-, and heptachlorinated PCBs.

Acknowledgments

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Supporting Information Available

A table giving the range, arithmetic mean, standard deviation, and geometric mean of concentrations of ΣPCB and 57 individual congeners (47 chromatographic peaks) detected in >70% of samples recorded in this study. This material is available free of charge via the Internet at http://pubs.acs.org.

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