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Comparison of Polychlorinated Biphenyl Concentrations in Indoor and Outdoor Air and the Potential Significance of Inhalation as a Human Exposure Pathway

GIAN MARCO CURRADO AND STUART HARRAD*

Institute of Public & Environmental Health, School of Chemistry, University of Birmingham, Birmingham B15 2TT, United Kingdom

Polychlorinated biphenyl (PCB) concentrations were measured in air from a total of 14 different indoor environments in Birmingham and the West Midlands area of the United Kingdom and compared to ambient outdoor levels. In four instances, spatially and temporally consistent indoor and outdoor samples were taken for comparison. Other indoor samples were compared to 25 outdoor samples from the Birmingham area taken at regular intervals between February 1997 and February 1998. Higher levels were present in indoor air (1.1−69 ng of ∑PCB m^{-3}, mean = 9.0 ng of ∑PCB m^{-3}) than in outdoor air (0.08−1.5 ng of ∑PCB m^{-3}, mean = 0.31 ng of ∑PCB m^{-3}). This limited data set indicates daily mean background U.K. PCB intake via inhalation to be 110 ng of ∑PCB $person^{-1}$ (0.7 pg of ∑i-TE $person^{-1}$), with a range of 23−590 ng of ∑PCB $person^{-1}$ (0.03−2 pg of ∑i-TE $person^{-1}$). Inhalation may thus represent a significant exposure pathway to ∑PCB but not ∑i-TE for some individuals, given that 1992 daily U.K. intake of PCB via diet has been estimated elsewhere to be 340 ng of ∑PCB $person^{-1}$ (54 pg of ∑i-TE $person^{-1}$).

Introduction

Polychlorinated biphenyls (PCBs) are a group of industrial chemicals with a wide range of suspected and proven adverse health effects (*1, 2*). Between 1954 and the late 1970s, ca. 40 × 10^4 t was used in the U.K. However, some items manufactured before the introduction of restrictions on their manufacture remain in use today—inter alia sealants and small capacitors in electrical equipment such as refrigerators and starter motors for fluorescent light switches. As a result, there remains the possibility of PCB contamination of indoor microenvironments where such items are located. Although there have been several reports of elevated levels of PCBs in indoor air (*3*−*7*), to date little consideration appears to been given to the potential impact of these elevated concentrations on human exposure. Indeed, it is generally assumed that nonoccupational exposure to PCBs occurs predominantly via dietary ingestion. To illustrate, one of the most recent exposure estimates for the U.K. estimated that ca. 97% of ∑PCB intake occurs via diet, with inhalation contributing the majority of the remainder (*8*). This paper reports the concentrations of PCBs found in air samples taken from a variety of buildings in Birmingham and the West Midlands area of the U.K., compares them with those present in ambient outdoor air, and assesses the potential significance of inhalation as a pathway of human exposure to PCBs.

* Corresponding author telephone: +44 121 414 7298; fax: +44 121 414 3078; e-mail: S.J.Harrad@bham.ac.uk.

Experimental Methods

Sampling Locations. Two laboratories and five offices at Birmingham University were investigated between December 1996 and February 1998. These were located in four separate buildings on the campus; one built in the late 19th century, two constructed in the 1960s, and one in the 1990s. Both laboratories had been used for PCB analysis for the previous 2 years. Seven homes in Birmingham and the West Midlands area were also sampled between January 1997 and March 1998. These were not preselected and covered a range of different house types and ages. For four indoor locations, spatially and temporally consistent outdoor air samples were taken for direct comparison of indoor and outdoor concentrations. The remaining 10 indoor air samples were compared to the levels detected in 25 samples of ambient outdoor air taken at regular intervals between February 1997 and February 1998 at a campus site and are considered representative of the Birmingham area. For ease of reference, we have assigned a numerical identifier to each indoor and outdoor sample. These are listed in Table 1.

Air Sampling. Air samples were taken at ground level and clear of any building air outfalls using a Graseby-Andersen hi-vol sampler modified to hold a Teflon-coated glass fiber filter (GFF, 0.6 μm pore size) and a precleaned polyurethane foam (PUF) plug. For each indoor microenvironment, two separate samples (of equal volume) were taken one after the other, either on the same day or over a period of 2 days with the windows closed, each for a period ranging from 2 to 24 h at a flow rate of 0.7−0.9 m^3 min^{-1} yielding sample volumes of ca. 80−1300 m^3. Outdoor samples were taken using the same equipment for periods of approximately 24 h (sample volume ca. 1300 m^3). Flow rates were measured directly using a precalibrated Kurtz portable hi-vol calibrator.

Sample Purification and Analysis. All PCB analyses were conducted using well-validated, containment enrichment, GC/MS procedures reported in detail elsewhere (*9*). Briefly, this involved Soxhlet extraction with dichloromethane, acid washing, Florisil chromatography, lipid removal via solvent exchange between DMSO and hexane, and concentration prior to GC/MS analysis on a Fisons' MD-800 instrument, fitted with an HP-5 trace analysis column (60 m × 0.25 mm i.d. × 0.25 μm film thickness). One microliter of sample extract was injected in the splitless mode (injector and transfer line temperatures both 300 °C), and the oven program was 140 °C for 2 min; 5 °C min^{-1} to 200 °C; and 2 °C min^{-1} to 280 °C. Ten ions were monitored in EI selected ion monitoring mode (ionization voltage = 70 eV) for the analysis of the trichlorinated biphenyls through the heptachlorinated biphenyls, two ions for each homologue group. These ions were 255.95, 257.95, 289.95, 291.95, 325.90, 327.90, 359.90, 361.90, 393.85, and 395.85. ∑PCB refers to the sum of all trichlorinated through heptachlorinated PCB congeners detected in a sample, and GFF- and PUF-retained PCB concentrations were determined separately.

Quality Control and Quality Assurance. To evaluate analyte losses during sampling, sampling efficiency standards (SESs, PCB congeners 19 and 147) were added to the GFF prior to sampling. There was a possibility that varying sampling times (3−24 h) would influence the reported

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TABLE 1. Identification Codes for Samples

numerical identifier	sampling location and date	numerical identifier	sampling location and date
1	office 1, 01/16/1998	12	office 4, 04/03/1997
2	outdoor office 1, 01/16/1998	13	office 5, 01/23/1997
3	office 2, 02/21/1998	14	laboratory 1, 01/16/1997
4	outdoor office 2, 02/21/1998	15	laboratory 2, 12/01/1996
5	house 1, 03/24/1998	16	house 3, 03/21/1997
6	outdoor house 1, 03/24/1998	17	house 4, 02/06/1997
7	house 2, 03/10/1998	18	house 5, 02/13/1997
8	outdoor house 2, 03/10/1998	19	house 6, 02/24/1997
9	office 1, 03/15/1997	20	house 7, 03/03/1997
10	house 1, 04/08/1997	21	mean outdoor (25 samples)
11	office 3, 12/06/1996		

concentrations due to variations in sampling efficiency. SES recoveries (which reflect analyte losses due to both sampling *and* analysis) were however found to be similar in all samples. Indoor 3-h sample recovery means of 61% and 79% (congeners 19 and 147), respectively, compared well to both indoor 24-h recovery means (51% and 91%) and outdoor 24-hr recovery means (68% and 66%). Recoveries of internal standards added to check analyte losses during sample analysis alone (PCB congeners 34, 62, 119, 131, and 173) ranged between 47 and 89% for all samples.

The repeatability of sampling and analysis combined was assessed by taking duplicate outdoor air samples at the same site (Birmingham University) over an identical period (24 h), using two identical hi-vol samplers. This exercise was repeated three times. The results (calculated as ∑PCB for the sum of vapor and particulate phase) revealed a mean relative standard deviation of 22% and demonstrated the validity of comparing spatially and temporally consistent indoor/outdoor air sample pairs.

To gauge the effectiveness of using outdoor samples from one site on campus as a comparison for indoor environments located in a variety of locations in the Birmingham area, two temporally consistent 24-h samples were taken at two outdoor sites on campus, located approximately 1 km apart. One of the sites was our usual sampling site and that used for collection of outdoor air samples between February 1997 and February 1998. The mean relative standard deviation (calculated as ∑PCB for the sum of vapor and particulate phase) for these samples was 19% and considered to confirm the validity of our approach.

Results and Discussion

PCB Levels in Indoor Air. Table 2 shows PCB concentrations found in indoor microenvironments sampled in this study. The results are shown for the 37 most abundant congeners (i.e., those whose concentrations >0.05% ∑PCB) and ∑PCB (sum of all congeners detected) in both of the samples taken. The mean indoor ∑PCB concentration of 9.0 ng m^{-3} (range 1.1−69 ng m^{-3}) is lower than that reported elsewhere. MacLeod (*6*) reported mean levels in offices (100 ng m^{-3}), laboratories (210 ng m^{-3}), and homes (310 ng m^{-3}), while other studies (*3*−*5*) reported mean levels varying between 174 and 457 ng m^{-3}. More similar to the levels found in this study are the geometric means of 18 and 10 ng m^{-3} reported by Vorhees et al. (*7*) for indoor environments located in two neighborhoods. Given that the Vorhees et al. study is the most temporally comparable with our work, the difference between the levels reported in our study and in other work, (*3*−*6*) may be evidence of a significant temporal decline in indoor contamination with PCBs following restrictions on their use.

The highest concentrations were in offices 1, 3, and 4 and in laboratories 1 and 2 that were located in two similar buildings built in ca. 1960 on the campus of Birmingham University. As both of the laboratories had been used to prepare environmental samples for PCB analysis or to handle concentrated PCB standard solutions, we originally suspected that these high concentrations were associated with such activities. This possibility was investigated by analyzing a sample from laboratory 1 to which no internal standards were added. Our analytical methodology uses as internal standards nonisotopically labeled PCB congeners that are essentially absent from environmental samples and commercial PCB formulations. Hence, the presence of elevated levels of these congeners in this sample—quantified via the external standard technique—would confirm our experimental activities as the source of the high levels of PCBs in these laboratories. However, when we conducted such an analysis, the concentration of these congeners was found to be very low as compared to other congeners in laboratory 1 (500−3000 times lower) and suggests the existence of another source of PCBs in these rooms. Indeed, the high concentration of PCBs found in offices 1, 3, and 4 indicate that the elevated levels of PCBs detected were not confined to the laboratories alone and may be present throughout these buildings.

PCB levels in microenvironments located in homes and other university buildings were generally lower than those in offices 1, 3, and 4 and the laboratories. Excluding these samples, the range of ∑PCB concentrations in the remaining microenvironments was 1.1−6.3 ng m^{-3} (arithmetic mean 2.9 ng m^{-3}).

Comparison of PCB Levels in Indoor and Outdoor Air. Figure 1 shows mean ∑PCB concentrations in indoor and outdoor samples. The four indoor environments for which spatially and temporally consistent outdoor comparison samples were taken all show indoor ∑PCB concentrations to be higher than the equivalent outdoor concentrations (1.8−180 times higher). There appears to be no correlation between elevated indoor concentrations and outdoor concentrations (e.g., samples 1 and 2, where the highest indoor concentration corresponds to the lowest outdoor value), agreeing with data that showed concentrations in 34 indoor environments were not significantly correlated to outdoor levels (*7*). However, in spring 1998, house 1 had only slightly higher indoor concentrations (3.8 and 1.4 ng of ∑PCB m^{-3}) than outdoor (mean of two samples = 1.4 ng of ∑PCB m^{-3}). This is unsurprising as house 1 was the most recently constructed building (1995) and contained no obvious PCB sources.

Other indoor environments were compared to 25 outdoor samples taken at regular intervals between February 1997 and February 1998 at our campus site. The arithmetic mean of these samples was 0.25 ng of ∑PCB m^{-3} (range = 0.08−1.0 ng of ∑PCB m^{-3}). In all cases, the indoor concentrations were higher than the mean outdoor concentration and exceeded the maximum value recorded outdoors. These data demonstrate that levels of PCBs in air from all of the laboratories, offices, and homes studied are higher than those in outdoor air.

Factors Influencing PCB Levels in Indoor Air. Newer buildings (office 2 constructed in 1992 and house 1 constructed in 1995) along with buildings of Victorian age (office 4 and houses 6 and 7) had the lowest concentrations. It appears likely from these results that the age of the building in which the indoor microenvironment is located is of importance in determining PCB concentrations. However, it was rarely possible to determine the exact age of the buildings sampled nor the dates of any alterations made (e.g., installation of electrical apparatus, or permanently elastic sealant), and thus quantitative correlations between

TABLE 2. PCB Concentrations (pg m^{-3}) in Indoor Air Samples[a]

	sample identifier																															
congener	1a	1b	3a	3b	5a	5b	7a	7b	9a	9b	10a	10b	11a	11b	12a	12b	13a	13b	14a	14b	15a	15b	16a	16b	17a	17b	18a	18b	19a	19b	20a	20b
18	394	408	172	191	100	94	163	145	1138	514	146	65	360	345	773	827	251	293	1178	683	817	507	902	1251	471	412	356	397	181	215	136	137
17	134	133	52	57	30	30	46	48	617	308	35	18	nd (0.08)	nd (0.08)	215	250	85	84	nd (0.08)	216	nd (0.08)	nd (0.08)	209	188	157	110	119	107	36	28	45	44
32/16	267	258	107	115	63	54	33	89	750	372	73	42	159	138	434	465	139	149	347	352	489	306	350	346	248	212	200	219	180	136	91	86
31/28	749	736	214	229	189	132	233	192	2199	1404	226	118	440	379	1005	1072	393	411	1088	938	1966	1130	671	660	583	484	604	614	389	307	274	251
33	329	318	93	94	88	57	102	86	948	774	124	42	173	130	449	483	157	184	419	382	581	490	240	243	240	190	166	201	180	130	102	96
22	175	166	49	42	50	27	54	46	523	265	43	24	94	71	220	244	83	108	210	188	561	315	139	129	137	97	110	121	117	80	51	52
52	5549	5214	209	154	483	206	117	111	14766	5143	83	103	1406	1973	422	425	367	332	2797	2759	1526	1008	580	450	400	331	323	277	219	159	317	129
49	573	533	29	31	59	98	29	20	2757	1035	53	24	267	379	144	152	93	88	861	561	485	258	185	154	109	90	86	82	66	52	64	36
44	1050	945	6	36	119	65	36	32	3161	1263	75	38	365	498	144	149	123	117	775	760	777	484	186	150	125	111	85	80	72	69	79	51
41/64/71/72	574	534	74	48	80	53	36	26	1417	684	79	55	136	191	164	171	123	117	371	275	421	282	303	196	130	115	137	138	96	86	77	67
74	281	271	22	15	55	19	16	14	825	395	28	16	95	142	56	58	46	47	178	213	337	171	65	62	50	40	38	32	33	25	27	24
70/76	1258	1177	58	40	245	55	38	35	4294	1991	115	58	425	611	154	151	161	154	1179	1029	1323	678	239	213	127	109	110	94	106	93	94	70
66	504	465	42	27	99	24	26	23	992	541	49	26	108	164	93	99	55	67	1*	236	401	173	105	97	69	60	54	92	54	43	37	39
95	1045	949	44	32	201	46	22	21	5501	1532	104	47	645	600	165	175	164	158	1456	1588	253	323	242	190	105	91	108	92	87	59	99	66
90/101	1567	1462	56	63	515	102	43	41	5352	3677	153	76	562	591	217	228	204	200	1763	1548	431	509	245	226	134	121	176	130	151	103	281	118
99/113	327	318	13	14	116	21	9	9	1596	1099	50	26	177	170	46	44	55	53	448	484	178	197	91	83	36	29	43	33	43	29	30	24
87	297	277	12	11	117	20	7	7	1470	482	53	30	160	158	57	54	55	58	452	529	160	204	80	75	35	33	40	32	45	31	28	24
110	709	655	27	25	321	51	16	16	3829	1265	139	81	361	348	158	149	138	146	1067	1283	476	573	199	191	90	85	99	74	128	86	70	57
118	339	316	18	17	234	40	10	9	1709	1197	100	67	141	136	100	96	79	81	534	653	369	435	91	95	58	53	62	38	92	65	40	29
105	83	81	5	4	56	11	2	2	525	383	34	23	41	33	32	30	22	25	119	158	116	144	26	28	19	16	21	10	30	19	13	8
148	66	63	4	4	21	18	3	3	527	164	16	9	34	43	37	40	15	16	87	105	79	42	33	27	18	16	13	10	15	10	13	11
149	143	138	10	11	71	18	8	7	1183	380	52	32	133	159	101	111	54	56	379	443	371	204	69	71	46	43	50	32	42	32	30	27
153	136	134	12	14	88	26	9	8	885	276	55	38	61	80	88	97	42	45	264	296	392	168	51	57	40	35	52	30	42	33	25	21
138/164	63	60	5	6	38	12	4	3	1134	321	53	40	70	83	86	87	55	69	370	338	429	218	55	64	46	39	76	42	47	36	2*	74
179	16	16	2	2	13	nd (0.25)	1	1	29	21	3	2	11	12	12	14	5	5	14	14	24	30	4	4	9	8	nd (0.25)	4	4	3	2	3
187/182	17	16	2	2	14	nd (0.25)	2	2	42	25	5	4	10	12	15	18	6	6	17	15	31	36	5	6	10	8	10	4	4	3	3	4
180	20	18	3	3	16	26	2	2	132	57	21	11	17	22	24	29	14	19	29	46	25	14	39	24	17	12	32	22	9	9	1*	62
∑PCB	18489	17486	1592	1529	3745	1362	1136	1109	68608	29708	2478	1440	7556	8692	6447	6836	3546	3657	18492	19142	15859	10787	6278	6104	4113	3484	3770	3520	3038	2386	2384	1901
∑i-TE	0.043	0.041	0.002	0.002	0.029	0.005	0.001	0.001	0.317	0.353	0.037	0.031	0.030	0.040	0.040	0.040	0.020	0.020	0.105	0.109	0.100	0.080	0.034	0.025	0.019	0.017	0.036	0.031	0.035	0.033	0.020	0.014

[a] Sample a is the first sample. Sample b is the second sample. nd, not detected; detection limit in parentheses. *, value reported only for particle fraction due to interference in chromatogram.

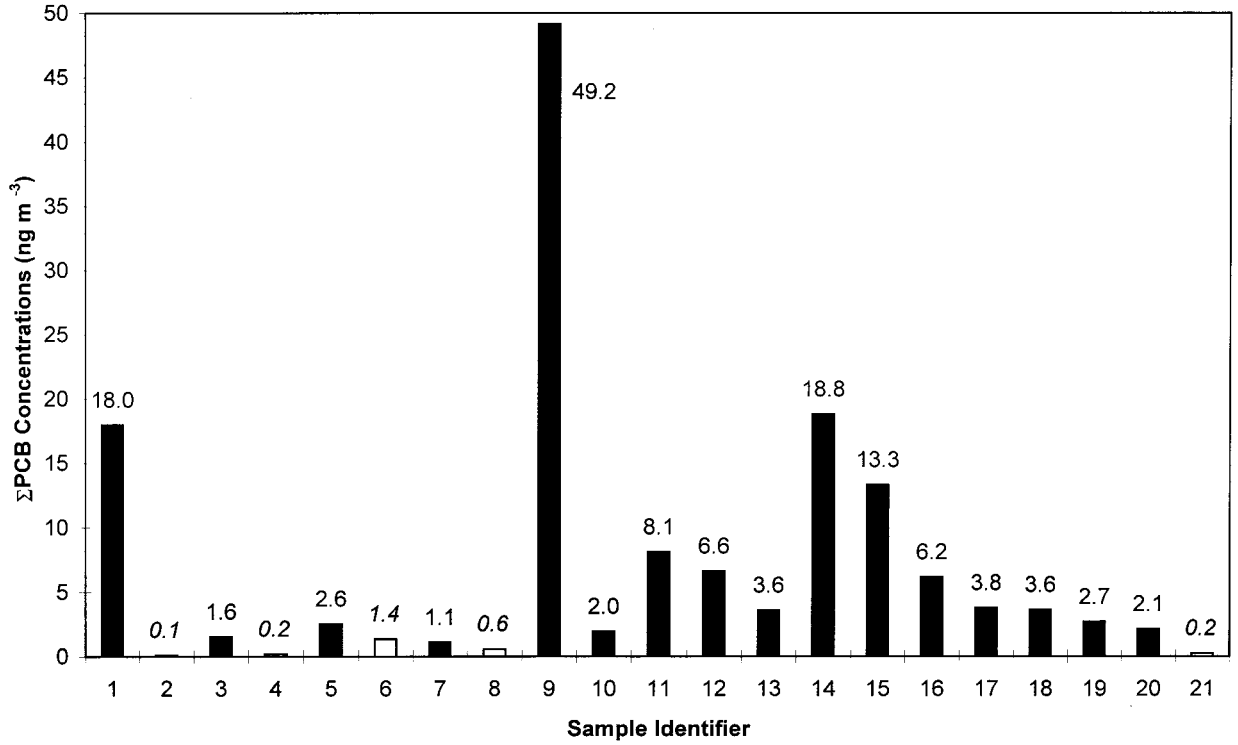

FIGURE 1. ∑PCB concentrations in indoor and outdoor air samples (individual samples and outdoor values).

indoor PCB concentrations and factors such as building age were not possible.

As illustrated by Table 2, for the majority of microenvironments, a lower PCB concentration was found in the second repeat sample. This is considered likely to be due to insufficient time elapsing between the end of the first and the start of the second sampling events, with the result that PCB concentrations were unable to regain equilibrium before the second sample was taken. This hypothesis is supported by the fact that, when the two sampling periods were separated (by ca. 8−12 h; for samples 1a/b, 3a/b, 11a/b, and 14a/b), the concentrations obtained in the second sample were similar if not higher than in the first. If correct, this indicates the existence of indoor sources of PCBs as opposed to infiltration of outdoor air, otherwise little difference would be observed between the first and second samples. Where levels in the first sample exceed those in the second, the mean concentrations given for each microenvironment are likely to be underestimates. It is thus recommended that future work should allow for this potential sampling artifact by allowing sufficient time to elapse between repeat samples in the same microenvironment.

Interestingly, when office 1 was sampled in spring 1997, a mean concentration of 49 ng of ∑PCB m^{-3} was recorded. By winter 1998, the mean concentration in the same microenvironment had declined to 18 ng of ∑PCB m^{-3}. The difference in concentrations clearly exceeds that attributable to combined sampling and analytical variability, and as the diurnal temperature variation in this room does not exceed 4 °C and this is unlikely to exert a significant influence on concentrations, the decline may possibly be due to a change in the room usage pattern between the two sampling events. While in spring 1997 office 1 was used on a limited basis, by winter 1998 it was fully occupied, leading to a possible reduction of PCB levels due to factors such as increased ventilation due to opening doors and windows.

Vapor/Particle Phase Partitioning. Given concerns that particle-bound airborne contaminants deposit more efficiently in the human respiratory system, we determined in all air samples the "operational" fraction of PCBs that was particle-bound (f_p)—i.e., that detected on the glass fiber filter. The mean f_p value for ∑PCB in indoor air samples (0.01) was significantly lower than that for outdoor air (0.08) and could potentially mitigate (to an unknown degree) the human health implications of exposure to the higher indoor PCB concentrations reported. The lower f_p values indoors are unsurprising given that indoor temperatures were always higher than those outdoors and that gravimetrically determined TSP concentrations were similar for both indoor (mean = 29 μg m^{-3}) and outdoor air (mean = 32 μg m^{-3}) in the temporally and spatially consistent samples.

Significance of Indoor Air as a Source of Human Exposure to PCBs. The U.K. Ministry for Agriculture, Fisheries, and Food (MAFF) estimated that in 1992 the U.K. daily mean dietary intake of the sum of 53 PCB congeners was 340 ng $person^{-1}$ (*10*). Using the data reported here, we have estimated the likely range and arithmetic mean of daily human intake of PCBs via inhalation. On the basis of studies on 47 U.K. individuals designed to monitor personal exposure to VOCs, we have assumed that the typical percentage of time spent outdoors is 8.3% (Leung and Harrison, unpublished data). We have also assumed that 26% of time spent indoors is spent in the workplace, with the remainder at home. Minimum, mean, and maximum daily exposures via inhalation (assuming 100% absorption of intake) were calculated using the algorithm below:

$$\Sigma\text{exposure}_i = ([C_w F_w] + [C_h F_h] + [C_o F_o])R_r$$

where $\Sigma\text{exposure}_i$ is the daily adult human exposure through inhalation (ng of ∑PCB $person^{-1}$ day^{-1}), $C_{w/h/o}$ is the ∑PCB concentration in workplace/home/outdoor air, respectively (ng m^{-3}), R_r is the adult respiration rate (20 m^3 d^{-1}), and $F_{w/h/o}$ is the respective fraction of day spent at workplace/home/outdoors.

Using the above algorithm, daily human exposure to ∑PCB falls within the range 23−590 ng $person^{-1}$, with a mean of 110 ng of ∑PCB $person^{-1}$. For a typical U.K. individual receiving 340 ng of ∑PCB day^{-1} from dietary sources, inhalation could thus represent between 6 and 64% of overall

human exposure to ∑PCB (mean 25%). If PCB levels in foodstuffs continue to fall (reported daily human exposure in 1982 was 1 μg of ∑PCB person^{-1}) (*10*), then the significance of inhalation as a human exposure pathway is likely to increase.

The U.K. Department of Health has recently accepted advice from its Committee on the Toxicity of Chemicals that the toxic equivalency concept applied to PCDD/Fs may also be used for PCBs in a limited manner. Hence, daily human exposure to PCBs via inhalation was estimated using the algorithm shown above for congeners that have been assigned i-TEFs (*11*) and found to lie in the range 0.03−2 pg of ∑i-TE person^{-1} (mean 0.7 pg of ∑i-TE person^{-1}). For a typical 60 kg U.K. individual whose PCB exposure from dietary sources expressed as ∑i-TE amounts to 54 pg of ∑i-TE day^{-1} (*12*), inhalation could thus represent between 0.06 and 3.6% (mean = 1.3%) of overall human exposure to PCBs expressed in terms of ∑i-TE. Although the results expressed in terms of ∑i-TE are reassuring in that they show a considerably lower percentage contribution of inhalation to total exposure than for ∑PCB, it must be noted that our estimates assume zero concentrations for those congeners that were below detection limits (e.g., 77, 126, and 169), but are assigned relatively high TEFs. Future measurements should therefore ensure that sufficient sample is acquired to provide measurable quantities of such toxicologically significant congeners.

The results presented in this paper show that levels of PCBs in air sampled within a variety of workplace and domestic indoor environments are significantly higher than those present in outdoor air and are slightly lower but comparable with those recently reported in Massachussets (*7*). Given the recent fall in U.K. human exposure to PCBs via dietary ingestion, inhalation may well constitute a much more significant human exposure pathway than previously assumed, although reassuringly our data indicate that, when expressed as ∑i-TE, exposure via inhalation is a much less significant contributor to total exposure than when ∑PCB is considered. Despite this, our data suggest that it would be prudent to evaluate more closely the efficacy of current strategies that aim to limit exposure via an almost exclusive focus on controlling dietary intake. It is therefore to be hoped that the U.K. government's action plan to phase out and destroy PCBs (*13*) is successful. In the meantime, a rigorous source apportionment exercise coupled to more detailed study of PCB levels in different indoor microenvironments is needed to fully assess both the sources of elevated concentrations of PCBs in indoor air and the significance of its inhalation as a source of human exposure.

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