Supporting Information

Inhalation and Dietary Exposure to PCBs in Urban and Rural Cohorts via Congener-Specific Measurements

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SUPPLEMENTAL METHODS

Passive Air Sampling

Passive air sampling (PAS) was used to determine airborne concentrations for each PCB congener ($C_{air,j}$) by dividing the mass of PCBs on each sampler ($M_{PUF,j}$) by the volume of air sampled in each deployment:

$$C_{air,j} = \frac{M_{PUF,ji}}{R_{s,j} \times t}$$

where $R_{S,j}$ is the sampling rate of PCB j on each polyurethane foam disk (PUF) ($m^3\ d^{-1}$) and t is the deployment period (d). Sampling rate, $R_{S,j}$, can vary by an order of magnitude over short time periods and ranges from 2-10 $m^3\ d^{-1}$.[1,2] We predict $R_{S,j}$ for each congener, location, and deployment period as described in Petrich *et al.*,[3] using a function of meteorological parameters (wind speed, ambient temperature, humidity, pressure).

Parameters for outdoor environments were taken from weather observations made at the nearest airports: the Gary/Chicago International Airport was used for EC outdoor samples and is located 4-8 km southeast of EC houses and schools. The Washington, Iowa Municipal Airport was used for CJ outdoor samples and is located about 20-30 km west of CJ houses and schools. These hourly meteorological datasets were accessed through the Quality Controlled Local Climatological Data repository.[4]

Indoor sampling rates have less variability than outdoor sampling rates, due to controlled climates. Here we use a set of congener-specific indoor sampling rates that assume typical indoor air conditions: temperature of 22.5 °C, pressure equal to the outdoor ambient air pressure plus 50 Pa, air speed of 0.3 $m\ s^{-1}$, and relative humidity of 50%. The modeled indoor flowrates (2.5 to 3.4 $m^3 d^{-1}$) match those derived by depuration compounds for indoor environments (i.e. 2.6 $m^3 d^{-1}$),[5] as adjusted for each congener based on Log K_{OA} values.

Nonlinear sampling may occur during PAS measurements, increasing uncertainty in sampling rates. We expect mono- to trichlorobiphenyls to reach non-linear uptake and potentially saturate during long (90-day) outdoor deployments in summer, and for 90-day indoor samples.[3] Any under-sampled outdoor PCBs would impact a minimal amount of participant's inhalation exposure (< 5%). Possible under-sampling for indoor lower-MW congeners would provide a conservative estimate of inhalation exposure, but would not influence comparisons between EC and CJ, as PUFs from these locations had the same sampling period. Comparisons between school and home concentrations are supported by increased mass on school PUFs relative to home PUFs, despite a shorter deployment period for the former.

Sample Preparation, Extraction, and Analysis

Indoor PAS units were placed in microenvironments receiving airflow but removed from areas of high activity. Outdoor PAS units were hung from nearby trees, posts, or other structures, 2-10 m from each house or school and at a height about 2 m above the ground surface. To prevent uptake of PCBs during transit and storage, each PUF disk was refrigerated (at -20°C) and wrapped in aluminum foil before and after deployment. Deployment, collection, and transport of all PUFs was documented through chain of custody sheets and sample labels, which accompanied each PUF and were taped in lab notebooks upon analysis.

Quality Assurance and Quality Control

To correct for loss of PCBs during extraction, air concentrations were adjusted by surrogate standards' % recoveries. Any samples with recoveries below 40% or above 150% for any surrogate standard were re-analyzed before use in this data set. Laboratory blanks were measured in each batch for verification that PCBs were not introduced during handling. ∑PCB mass for lab and field blanks averaged 10.4 (8.0, 12.8) ng and 15.3 (11.2, 19.4) ng respectively.

Inhalation Exposure

Activity questionnaires were used to determine subjects' time spent in modeled locations and their corresponding inhalation rates. For each season, participants estimated how much time they spent in the four modeled locations (home, school, outdoors, and other). Their estimates were used to determine T_i of eq 1 in the main text. Completed surveys were verified by field staff to cover 24 hours for weekend days and weekdays, and 168 hours for each week. Additional survey data was used to determine weight-adjusted inhalation rates, specific to 5 activity levels (sleeping, passive/sedentary, light-intensity, moderate-intensity, and heavy-intensity activity) and 8 age brackets.[6] Age gender and weight were determined from questionnaires, while the corresponding inhalation rates were derived from US Environmental Protection Agency's Exposure Factors Handbook.[6] Fractional time spent at activity levels per location (e.g. homes, schools) were selected following USEPA (Table S3).[7] Bilingual, trained field staff administered these questionnaires in the subjects' preferred language (English or Spanish).

Dietary Exposure

Food samples were collected from 4 supermarkets in each city, prepared as for consumption, composited, and stored at -20 °C as described in Newsome *et al*.[8] Each composite TDS sample was spiked with internal standards before being extracted with acetone: hexane (2:1 v/v). Lipids were removed from extracts via acid digestion and/or gel permeation chromotography, and extracts were further cleaned using adsorption chromatography with Florisil. Extracts were analyzed using a Fisons 8000 series gas chromatograph, coupled to a Fisons Instrument Auto Spec Ultima mass spectrometer or a Hewlett Packard 5890 Series II coupled to a VG Auto Spec Q. Positive electron ionization (EI) in selected ion monitoring (SIM) served as the mode of quantification, achieving a resolution of $\geq$2,000 counts. This method allowed detection of 40 congeners, with a method quantification limit below 25 pg g^{-1} sample for most congeners.

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TABLES

Table S1 Field blank-derived limits of quantification (LOQ)

Homolog Group (BZ#)	Min. LOQ (ng)	Median LOQ (ng)	Max. LOQ (ng)
Mono-chlorinated (1-3)	1.26×10^{-2}	1.59×10^{-1}	5.67×10^{-1}
Di-chlorinated (4-15)	5.56×10^{-3}	2.78×10^{-2}	1.59×10^{-1}
Tri-chlorinated (16-39)	6.81×10^{-4}	2.21×10^{-2}	2.74×10^{-1}
Tetra-chlorinated (40-81)	1.71×10^{-4}	4.54×10^{-3}	2.33×10^{0}
Penta-chlorinated (82-127)	7.14×10^{-5}	4.92×10^{-2}	3.42×10^{0}
Hexa-chlorinated (128-169)	9.29×10^{-5}	1.90×10^{-3}	1.34×10^{0}
Hepta-chlorinated (170-192)	2.91×10^{-4}	1.13×10^{-3}	4.68×10^{-1}
Octa-chlorinated (193-205)	1.20×10^{-3}	5.03×10^{-3}	1.59×10^{-2}
Nona-chlorinated (206-208)	1.63×10^{-3}	2.25×10^{-3}	7.37×10^{-3}
Deca-chlorinated (209)	---	5.36×10^{-2}	---

BZ# – Ballschmiter and Zell (1980) numbering system for PCBs
LOQ – limits of quantification, calculated as 95% confidence interval for field blanks

Table S2 Fraction of time spent at each activity level, after USEPA (1985)

Activity Level	Indoor Locations	Outdoor Locations
Sleep and Passive	0.480	0.387
Light-intensity	0.480	0.387
Moderate-intensity	0.035	0.198
High-intensity	0.005	0.034

Table S3 Congener-specific inhalation and dietary exposure by cohort for the 93 most abundant congeners (diet + inhalation) excluding outliers

	EC Mother		EC Children			CJ Mother		CJ Children		
Congenerǂ	**Inhalation (SE)**	**Diet**	**Inhalation (SE)**	**♀ Diet**	**♂ Diet**	**Inhalation (SE)**	**Diet**	**Inhalation (SE)**	**♀ Diet**	**♂ Diet**
SUM	7.12(1.20)	83.7	12.0(1.18)	71.9	109	2.43(0.38)	75.1	8.88(0.30)	72.4	96.1
1	0.02(0.00)	---	0.09(0.00)	---	---	0.03(0.00)	---	0.08(0.00)	---	---
3	0.04(0.01)	---	0.11(0.01)	---	---	0.03(0.01)	---	0.08(0.00)	---	---
4	0.07(0.01)	---	0.32(0.01)	---	---	0.06(0.01)	---	0.15(0.01)	---	---
5	0.00(0.01)	---	0.00(0.02)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
6	0.03(0.01)	---	0.10(0.00)	---	---	0.03(0.00)	---	0.06(0.00)	---	---
8	0.19(0.02)	---	0.50(0.02)	---	---	0.10(0.01)	---	0.26(0.01)	---	---
11	0.44(0.05)	---	0.51(0.04)	---	---	0.31(0.03)	---	0.46(0.02)	---	---
15	0.06(0.01)	---	0.12(0.01)	---	---	0.03(0.00)	---	0.05(0.00)	---	---
16	0.09(0.02)	---	0.21(0.02)	---	---	0.05(0.01)	---	0.11(0.00)	---	---
17	0.11(0.02)	---	0.22(0.01)	---	---	0.06(0.01)	---	0.12(0.00)	---	---
18/30	0.24(0.04)	---	0.46(0.03)	---	---	0.12(0.01)	---	0.26(0.01)	---	---
19	0.02(0.00)	---	0.06(0.00)	---	---	0.01(0.00)	---	0.03(0.00)	---	---
20/28 (28)	0.24(0.04)	5.31	0.39(0.03)	5.00	7.05	0.11(0.01)	3.91	0.21(0.01)	3.87	5.27
21/33 (33)	0.13(0.02)	2.73	0.23(0.01)	2.57	3.82	0.06(0.01)	1.96	0.12(0.00)	2.03	2.66
31	0.24(0.05)	---	0.39(0.03)	---	---	0.10(0.01)	---	0.22(0.01)	---	---
32	0.06(0.01)	---	0.11(0.01)	---	---	0.03(0.00)	---	0.06(0.00)	---	---
37	0.03(0.01)	1.32	0.04(0.00)	1.26	1.79	0.01(0.00)	0.96	0.02(0.00)	1.07	1.45
40/41/71 (40)	0.42(0.07)	0.59	0.80(0.07)	0.56	0.77	0.02(0.01)	0.35	0.08(0.00)	0.36	0.49
(41)		2.55		2.18	3.39		2.30		2.28	3.14
44/47/65 (44)	0.50(0.10)	1.98	0.74(0.11)	1.65	2.65	0.08(0.06)	1.88	0.45(0.03)	1.82	2.53
49/69 (49)	0.12(0.02)	1.89	0.21(0.02)	1.66	2.48	0.04(0.01)	1.40	0.23(0.01)	1.29	1.84
52	0.43(0.11)	2.77	0.80(0.11)	2.18	3.60	0.14(0.05)	2.34	1.15(0.04)	2.03	2.98
60	0.02(0.00)	1.88	0.03(0.00)	1.62	2.54	0.00(0.00)	2.23	0.02(0.00)	2.27	3.28
61/70/74/76 (74)	0.28(0.07)	2.78	0.52(0.06)	2.50	3.77	0.13(0.02)	2.34	0.46(0.02)	2.32	3.43
64	0.08(0.01)	---	0.13(0.01)	---	---	0.02(0.01)	---	0.11(0.00)	---	---
66	0.09(0.02)	3.16	0.15(0.02)	2.66	4.11	0.02(0.01)	2.76	0.11(0.00)	2.76	4.07
73	0.06(0.07)	---	0.05(0.06)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
77	0.00(0.00)	---	0.00(0.00)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
81	0.00(0.00)	---	0.00(0.00)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
83/99 (99)	0.06(0.04)	4.44	0.17(0.03)	3.91	5.90	0.02(0.01)	3.55	0.22(0.01)	3.13	4.60
86/87/97/109/119/125 (87)	0.02(0.01)	2.2	0.05(0.01)	1.89	2.81	0.01(0.01)	2.13	0.05(0.00)	1.92	2.78
90/101/113 (90)	0.24(0.09)	3.49	0.51(0.08)	2.58	4.24	0.07(0.02)	3.75	0.55(0.02)	3.26	4.82
95	0.26(0.09)	---	0.55(0.08)	---	---	0.06(0.02)	---	0.63(0.02)	---	---
105 (105)	0.02(0.02)	2.26	0.05(0.01)	2	3.07	0.00(0.00)	2.07	0.04(0.00)	1.86	2.75
108/124	0.00(0.00)	---	0.22(0.01)	---	---	0.00(0.00)	---	0.30(0.01)	---	---
110/115 (110)	0.2(0.10)	8.8	0.16(0.08)	7.15	11.2	0.06(0.01)	8.44	0.05(0.01)	7.27	11
112	0.01(0.03)	---	0.01(0.02)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
114	0.00(0.00)	---	0.11(0.00)	---	---	0.00(0.00)	---	0.15(0.01)	---	---
118 (118)	0.11(0.06)	6.54	0.08(0.04)	5.84	8.99	0.02(0.01)	5.89	0.02(0.00)	5.41	7.92
(128)		1.78		1.63	2.31		1.08		0.90	1.29
129/138/163 (129)	0.06(0.03)	0.37	0.05(0.02)	0.37	0.45	0.01(0.00)	0.14	0.01(0.00)	0.13	0.17
(138)		6.83		5.75	8.84		6.47		5.30	7.72
135/151	0.03(0.01)	0.68	0.06(0.01)	0.45	0.73	0.01(0.00)	0.84	0.05(0.00)	0.56	0.88
136	0.02(0.01)	0.44	0.02(0.01)	0.40	0.54	0.00(0.00)	0.27	0.01(0.00)	0.22	0.32
137	0.00(0.00)	0.4	0.00(0.00)	0.37	0.54	0.00(0.00)	0.3	0.00(0.00)	0.27	0.39
141	0.01(0.01)	0.78	0.01(0.00)	0.69	0.94	0.00(0.00)	0.53	0.00(0.00)	0.41	0.60
147/149	0.09(0.03)	---	0.07(0.02)	---	---	0.02(0.00)	---	0.02(0.00)	---	---
153/168	0.07(0.02)	7.34	0.05(0.02)	6.1	9.38	0.02(0.00)	7.11	0.02(0.00)	5.75	8.41
156/157 (156)	0.00(0.00)	0.68	0.01(0.00)	0.61	0.91	0.00(0.00)	0.65	0.01(0.00)	5.89	0.84
(157)		0.23		0.23	0.30		0.09		0.08	0.11
167	0.00(0.00)	---	0.00(0.00)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
169	0.00(0.00)	---	0.00(0.00)	---	---	0.00(0.00)	---	0.00(0.00)	---	---
170	0.00(0.00)	1.23	0.00(0.00)	1.08	1.67	0.00(0.00)	1.05	0.00(0.00)	0.91	1.27
180/193 (180)	0.01(0.01)	3.07	0.01(0.01)	2.63	4.04	0.00(0.00)	2.69	0.01(0.00)	2.26	3.18
(193)		0.18		0.16	0.23		0.13		0.09	0.12
183	0.00(0.00)	0.85	0.00(0.00)	0.72	1.12	0.00(0.00)	0.76	0.00(0.00)	0.60	0.87
185	0.00(0.00)	0.19	0.00(0.00)	0.19	0.24	0.00(0.00)	0.09	0.00(0.00)	0.07	0.09
187	0.01(0.00)	1.13	0.01(0.00)	0.76	1.22	0.00(0.00)	1.53	0.00(0.00)	1.02	1.54
189	0.00(0.00)	0.04	0.00(0.00)	0.03	0.04	0.00(0.00)	0.02	0.00(0.00)	0.02	0.02
191	0.00(0.00)	0.93	0.00(0.00)	0.81	1.19	0.00(0.00)	1.00	0.00(0.00)	1.52	1.06
194	0.00(0.00)	0.41	0.00(0.00)	0.37	0.54	0.00(0.00)	0.35	0.00(0.00)	0.28	0.41
201	0.00(0.00)	0.53	0.00(0.00)	0.42	0.63	0.00(0.00)	0.69	0.00(0.00)	0.45	0.69
203	0.00(0.00)	0.52	0.00(0.00)	0.47	0.70	0.00(0.00)	0.52	0.00(0.00)	0.41	0.60
206	0.00(0.00)	0.29	0.00(0.00)	0.27	0.39	0.00(0.00)	0.30	0.00(0.00)	0.20	0.29

[a]Where elutions for inhalation and dietary data do not match, the latter are reported in parentheses. [b]Dashed lines indicate the elution was not measured.