

## **Background Levels of PCBs in Residents of British Columbia, Canada**

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Regulations adopted in 1977 and 1980 by Environment Canada under the Environmental Contaminants Act prohibited the sale of new equipment containing polychlorinated biphenyls (PCBs), and restricted existing PCB uses to electrical equipment sufficiently enclosed to prevent a threat to either the environment or human health (Garrett 1985). In 1985, further regulations were promulgated to control the release of PCBs into the environment and to further restrict the sale of existing PCB equipment. At that time, the major users of PCBs in the western Canadian province of British Columbia (BC) were the electrical utilities (28%), the forest industry (43%, mainly in pulp and paper mills), and the mining industry (15%) (Rottluff et al 1990).

The purpose of this study was to measure the concentrations of individual PCB congeners in British Columbia residents five to ten years after restrictions on PCB use had been put into place. In this paper, we report the levels of PCBs found in the adipose tissue of a sample of British Columbians with an age and sex distribution similar to that of the population. The measured PCB levels are compared to those reported in other population groups worldwide, and are examined to determine whether they were related to characteristics of the sampled group.

### **MATERIALS AND METHODS**

Subject selection was designed to reflect the distribution of the BC population by sex and by age in 5-year groups from the age of 15 to 84. Every patient scheduled for elective abdominal surgery between September 1990 and March 1991 in an age-sex category not previously filled was asked to participate in the survey. Consenting patients were interviewed about their place of birth; height; weight; occupational history; residential history; frequency of consumption of fish, meat,

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dairy products, and vegetables; water supply; surgical procedure; and the number of children breast fed (female subjects only).

Samples of omental adipose tissue collected during surgery were frozen at -20 °C and shipped frozen to the analytical laboratory. PCB analysis was done for total PCBs based on chromatogram peaks for Arochlor 1254/1260, and for 43 congener peaks representing 57 of the 209 individual congeners, including all levels of chlorination except mono. Individual PCB congeners are referred to by number according to the IUPAC system proposed by Ballschmiter and Zell in 1980.

The tissue samples were mixed with powdered anhydrous sodium sulphate and allowed to stand until the mixture was ground to a free-flowing powder. It was then loaded into a glass column, and eluted with 1:1 dichloromethane:hexane. The extract was further eluted on a 60-g Biobeads SX-3 column using the same solvent. The final extracts for analysis were prepared on a Florisil column (8 grams, 1% deactivated) using hexane (fraction 1), followed by 6% diethyl ether in hexane (fraction 2). Each fraction was concentrated to 1 mL for analysis. All PCB congeners except the coplanars were analyzed based on the method of Mullen et al (1984) using a Hewlett Packard 5890 gas chromatograph, with a Ni-63 electron-capture detector (GC/ECD), and a 60-m, 0.25-mm id, 0.25-micron-film, Durabond fused-silica capillary column. For all GC/ECD quantification, peaks were assigned to specific compounds using reference standards for 51 PCB congeners.

Coplanar analysis was based on the method of Stalling et al (1979). The two fractions were recombined and eluted in the following clean-up steps: carbon column elution with 1:1 cyclohexane:dichloromethane (discarded), 1:1 benzene:ethyl acetate (discarded), and toluene (retained); then basic alumina column elution with 3% dichloromethane in hexane (discarded), and 1:1 dichloromethane: hexane (retained). Coplanar PCBs in this final fraction were determined using low resolution analysis in the multiple ion detection mode on a Finnigan INCOS 50 mass spectrometer equipped with a Varian 3400 gas chromatograph with a 60-m DB-5 column (GC/MS).

Twelve sample replicates, eleven reagent blanks, and nine replicates of a reference sample of cod oil (#1588, US National Institute of Standards and Technology (NIST)) were analyzed for quality assurance. To measure recovery, samples were spiked with 40 or 128 ng of carbon-13-labeled PCB 204. All concentrations quantified by GC/ECD were corrected for recovery of this compound. For the GC/MS-quantified coplanars, carbon-13-labeled PCBs 77, 126, and 169 were added to each sample analyzed; concentrations of the coplanars were corrected using recoveries for their surrogate.

Detection limits (DL) were based on the instrument noise in each sample analysis, except where reagent blanks had concentrations greater than these limits. The maximum detection limit for the PCBs was less than 2.5 ng/g for every congener except the dichlorinated PCB 15 (maximum DL = 11 ng/g). For all calculations and analyses, measurements below detection limits were recorded as missing values. In an analysis of polychlorinated biphenyl congeners in human tissue samples, Fait et al (1989) found no difference in median values whether levels below the detection limit were entered as missing data, or estimates were substituted based on a mathematical model of the distribution below the detection limit.

All data analyses were performed using SPSS-X. Frequency histograms and measures of skewness and kurtosis of PCB concentrations indicated that they were approximately log-normally distributed, therefore all analyses were done with these data log-transformed ( $\log_{10}$ ). All analyses were done on concentrations of each PCB congener separately. Regression analyses were done initially on each diet, residential history, and weight-related variable individually, adjusted for age and sex. In order to eliminate variables likely entered in the screening analyses by chance, those significantly related to levels of more than two PCB peaks were included in a final step-wise multiple regression for the PCBs. Analysis of covariance was used to determine whether there were differences in PCB levels according to type of surgery, after adjusting for age and sex. Inferential analyses were restricted to those congeners with more than one-half of the measurements above the detection limits.

## RESULTS AND DISCUSSION

Adipose tissue samples from 18 men and 23 women were analyzed, taken from a patient base of 89. Only 10% of patients refused to participate, but surgeons did not take samples or samples were unusable in 48% of those who agreed to participate (reasons included insufficient omental adipose tissue, problems during surgery, and failure to send the sample to the laboratory for freezing). For the 41 subjects whose tissue samples were analyzed, the mean age was 45 years (range 18-77), the mean weight was 96 kg (range 50-193), and the mean height was 1.7 m (range 1.5-1.9). There were no statistically significant differences (t-test,  $p < 0.05$ ) in age, height, or sex between the patients who agreed to participate and those who had samples analyzed. However the weight of those whose samples were analyzed was significantly higher ( $p = 0.028$ , 15 kg difference in mean weight) and a similar difference in body mass index ( $\text{BMI} = \text{weight}/\text{height}^2$ ) approached statistical significance ( $p = 0.092$ ). The mean BMI of the

analyzed group (33.8 kg/m<sup>2</sup>) was also greater than that of the Canadian population (24.6 kg/m<sup>2</sup>) (Health Promotion Directorate, 1988).

The procedures undergone by the patients were mainly cholecystectomy and/or gastroplasty (N=27), but also included gastroplasty reversal, hernia repair, bowel surgery, Nissen fundoplication, biopsy, liver resection, pelvic pouch procedure, appendectomy, abdominal plasty, femoral cross repair, and laparotomy. The patients had lived in a total of 46 different places in British Columbia, 24 in the rest of Canada, and 12 in other countries. They had worked in a total of 46 industries, from the primary resource sector and manufacturing, as well as service, sales, education, and administration.

Table 1 summarizes the concentrations measured for each PCB congener and for total PCBs as Arochlor 1254/1260. Each congener is listed separately except where chromatogram peaks could not be resolved; for these peaks all possible congeners are listed, though it is impossible to distinguish which congeners in the group were actually detected in a sample. The following congeners peaks were detected in all samples: PCBs 31, 138, 153, 170, 180, 183, 201, and 203/196. The adipose tissue of 85% of the study population also contained congeners 28, 66/80/95, 105, 118, 194, 206 and 209. The following congener peaks were not detected in any sample: PCBs 15, 54, 121, 128, 143, 185, and 40/103. The geometric mean total PCB level was 490 ng/g, with a range of 65.2 to 1,974 ng/g. The hexa-, hepta-, and octachlorinated PCB congeners contributed the greatest amounts to the overall PCB fat burden in the BC residents.

The source of exposure to PCBs among BC residents is suggested by comparing the profiles of six congeners (PCBs 44, 138, 170, 191, 194, 206) measured both in the BC study group and in Washington DC transformer repair workers whose type of PCB exposure was known (Fait et al 1989). The relative levels of the congeners in the BC sample were very similar to those found in the transformer workers, implying exposures to the same PCB mixtures, mainly Arochlor 1260.

As reported in other studies (Kannan et al 1988), the more toxic coplanar PCB congeners were often below detection limits, and when detected, the levels were low compared to other congeners, usually less than 0.1 ng/g. It is interesting to note that the coplanars were still detected at higher concentrations than most polychlorinated dibenzo-p-dioxins and dibenzofurans in an analysis of the latter compounds in the same adipose tissue samples (Teschke et al 1992). Safe (1990) has proposed Equivalency Factors relating the toxicity of the coplanar PCBs to that of 2,3,7,8-tetrachlorodibenzo-p-dioxin. Applying these factors to the results for the coplanar and mono-ortho coplanar PCBs only, the

**Table 1.** PCB concentrations in BC residents, by congener (ng/g lipid)

Congener	Minimum*	Maximum	Mean	GM**	GSD¥	N†
Total PCBs	65.2	1974	626	490	2.09	41
All TriCBs	1.98	44.4	9.56	8.24	1.72	41
PCB 18	0.60	3.03	1.41	1.15	2.02	5
PCB 28	0.81	35.9	4.97	3.85	1.94	39
PCB 31	0.64	10.0	4.65	4.14	1.70	41
All TetraCBs	0.57	38.3	3.96	2.70	2.55	36
PCB 44	0.15	3.92	0.76	0.53	2.19	17
PCB 49	0.32	28.1	4.11	1.78	3.18	18
PCB 52	0.20	6.34	1.86	1.41	2.11	27
PCB 60	0.15	3.96	1.50	1.21	2.12	17
PCB coplanar 77	0.0051	0.041	0.013	0.010	1.95	10
All PentaCBs	2.76	77.2	28.0	23.2	2.03	40
PCB 87	0.35	1.63	0.97	0.82	1.93	8
PCB 90/101	0.25	3.08	1.48	1.31	1.73	32
PCB 105	0.22	11.5	3.85	2.96	2.23	39
PCB 114	0.35	22.9	4.12	3.16	2.10	33
PCB 118	1.94	57.4	20.7	17.0	1.97	39
PCB coplanar 126	0.0095	0.116	0.046	0.035	2.19	18
All HexaCBs	10.4	343	126	102	2.05	41
PCB 138	4.12	145	54.1	43.6	2.07	41
PCB 141	0.11	1.14	0.50	0.38	2.28	13
PCB 151	0.19	3.12	0.88	0.75	1.77	31
PCB 153	6.00	198	71.0	57.5	2.05	41
PCB coplanar 169	0.0058	0.067	0.032	0.026	2.00	12
All HeptaCBs	9.05	441	107	77.3	2.37	41
PCB 170	2.47	100	28.0	20.5	2.35	41
PCB 180	5.47	303	65.9	46.8	2.42	41
PCB 183	0.82	31.6	11.0	8.24	2.28	41
PCB 189	0.11	3.55	1.32	1.03	2.27	25
PCB 191	0.08	4.18	1.31	0.98	2.41	25
All OctaCBs	4.94	351	61.5	38.5	2.76	41
PCB 194	1.40	113	18.5	11.2	2.92	37
PCB 201	2.06	145	25.6	16.1	2.74	41
PCB 203/196	1.47	93.4	18.9	12.6	2.57	41
PCB 205	0.13	2.61	0.92	0.73	2.19	13
All NonaCBs	1.17	33.9	8.47	6.78	2.34	37
PCB 206	1.06	31.2	8.75	6.50	2.25	35
PCB 207	0.11	4.08	1.58	1.21	2.37	26
DecaCB (PCB 209)	0.54	21.3	7.49	5.90	2.15	35
PCB 61/94/74	0.58	19.5	4.06	2.54	2.54	19
PCB 66/80/95	0.45	10.5	2.64	2.17	1.86	38
PCB 77/154	0.25	21.9	2.70	1.09	3.03	12
PCB 126/129	1.05	23.7	7.24	5.24	2.40	15
PCB 187/182	1.53	81.6	18.9	13.8	2.31	40
PCB 173/200	0.67	5.00	2.46	2.12	1.77	25
PCB 202/171/156	1.74	23.5	11.1	9.04	2.09	18
PCB 208/95	0.55	19.7	5.60	3.81	2.55	15

Footnotes for Table 1.

\* Minimum concentration in samples with detectable concentrations

\*\* Geometric mean

¥ Geometric standard deviation, unitless

† Number of samples with concentrations greater than the detection limit

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Toxicity Equivalents (TEQs) for the PCBs in the BC sample ranged from 2.6 to 98.6 pg/g (mean = 34.8), compared to a range of 8.4 to 56.4 pg/g (mean = 29.1) for the dioxins and furans.

Table 2 summarizes data from 11 reports which listed total PCB concentrations in human adipose tissue on a per gram lipid basis (Camps et al 1989; Fait et al 1989; Frank et al 1988; Fujiwara 1975; Hattula et al 1976; Holt et al 1986; Fukano and Doguchi 1977; Jensen and Clausen 1979; Kraul and Karlog 1976; Mori et al 1983; Teufel et al 1990). It should be borne in mind that these studies used different standards to calculate total PCBs, including comparisons with chromatogram peaks for Kanechlor, Chlophen A, and Arochlor mixtures, as well as sums of individual congeners levels. The BC sample had the lowest fat concentrations of total PCBs, with a mean level of 626 ng/g; the other groups all had mean concentrations greater than 1,000 ng/g. Two of these studies also reported results for individual PCB congeners (Fait et al 1989; Teufel et al 1990). The BC data consistently had the lowest levels for every congener detected in common with these two study groups. There are several possible explanations for the lower levels detected in the BC sample. Population-based samples have had decreasing levels of PCBs in adipose tissue over the last 10 years (Robinson et al 1990), and the BC sample is the most recent. In addition, many samples of sediment, sewage discharges, and biota taken in BC have shown only non-detectable to low levels of contamination (Garrett 1985). Finally, the laboratory quality control analysis of the NIST standard reference sample showed a consistent underestimation of the concentrations of PCBs in the sample, by 10% on average. However, the differences in levels between the BC population and the populations studied worldwide are great enough that even if the levels were adjusted upwards to account for this underestimation, the BC results would remain low.

The following characteristics were available to be examined as predictors of PCB levels in the BC sample: age; sex; weight; number of children breast fed; occupation; area of residence; and diet. Of these, two were not included in the regression analyses because they varied little in the study sample: residence in an urban or rural setting (40 of the 41 subjects had lived in an urban setting for the last 20 years); and employment in occupations with potential exposure to sources of PCBs (none of the subjects had jobs judged likely to have had exposures

**Table 2.** Total PCB levels in populations reported worldwide (ng/g lipid)

Year sample taken	Population location	Mean level	GM* level	Range	N	Reference
1972-3	Denmark	5,100	4,100	1,000-49,000	78	Kraul, 1976 <sup>a</sup>
1974	Japan	1,040	-	380-2,500	30	Fukano, 1977
<1975	Japan, Kyoto	4,700	-	1,900-13,300	-	Fujiwara, 1975 <sup>b</sup>
<1976	Finland	2,800	-	-	73	Hattula, 1976 <sup>a</sup>
<1979	Denmark	-	3,600	-	17	Jensen, 1979 <sup>a</sup>
<1979	Greenland	-	5,800	-	33	Jensen, 1979 <sup>b</sup>
1980-4	USA, Louisiana	1,145	-	380-2,330	18	Holt, 1986 <sup>b</sup>
1981	Japan	3,020	-	-	92	Mori, 1983 <sup>b</sup>
1983-4	Canada, Ontario	2,100	-	-	209	Frank, 1988 <sup>a</sup>
1985-7	Spain	1,680	-	-	14	Camps, 1989 <sup>a</sup>
1985-8	Germany	1,614	1,345	414-7,523	183	Teufel, 1990 <sup>c</sup>
<1989	USA, Wash DC	1,500	888	0.8-20,000	52	Fait, 1989 <sup>d</sup>
<1989	USA, Wash DC	5,400	3,180	30-58,000	33	Fait, 1989 <sup>e</sup>
1990-1	Canada, BC	626	490	65 - 1,974	41	b

\* Geometric mean or median

- Not reported

a Autopsy samples of adults of both sexes

b Biopsy samples of adults of both sexes (hospital patients)

c Biopsy samples of children of both sexes to age 16 (hospital patients)

d Biopsy samples from unexposed male operating engineers

e Biopsy samples from exposed male transformer repair workers

to PCBs). Based on the screening analyses, the following independent variables were not included in the final multiple regression for the PCBs: change in weight in the last year; number of children breast fed; municipal or well water sources; and frequency of consumption of poultry, lamb, pork, organ meats, milk, butter, yogurt, cheese, fruit, fish, fat, and vegetables. All the remaining variables were run in a step-wise multiple regression; the results are reported in Table 3. The total proportions of variance explained ranged from 0 to 0.53, with the highest for PCB 208/95 and certain hexa and heptachlorinated PCBs. None of the variables were explanatory for PCBs 31, 66/80/95, 90/101, and 151.

Age was the most consistent determinant of PCB concentrations; it was statistically significant for almost all PCB congeners and total PCBs. PCB concentrations increased with age, as was found in the NHATS sample of the US population (Robinson et al, 1990). The US population also showed a slight tendency for the PCBs to have higher concentrations in males than females, and this trend was confirmed in our data. Sex was significant for nine PCB congeners, with males

**Table 3.** Additional proportions of variance explained (adjusted R<sup>2</sup>) and regression coefficients (β) for variables found to have a statistically significant influence on PCB concentrations in step-wise multiple regression

Congener		Age (years)	Sex*	BMI (kg/m <sup>2</sup> )	Wild Meat†	Cottage Cheese†	Potatoes†
Total PCBs	R <sup>2</sup>	0.24	-	0.10	-	-	-
	β	.0081		-.0097			
PCB 28	R <sup>2</sup>	0.10	0.17	-	-	-	-
	β	.0056	.30				
PCB 52	R <sup>2</sup>	0.19	-	-	-	-	-
	β	.0081					
PCB 105	R <sup>2</sup>	0.12	-	-	-	-	0.12
	β	.0085					-.00086
PCB 114	R <sup>2</sup>	0.24	-	-	-	0.10	-
	β	0.010				-.0010	
PCB 118	R <sup>2</sup>	0.20	-	-	-	-	0.17
	β	.0090					-.00084
PCB 138	R <sup>2</sup>	0.06	0.10	0.19	-	-	0.10
	β	.0064	-.25	-.0055			-.00076
PCB 153	R <sup>2</sup>	0.09	0.10	0.24	-	-	0.07
	β	.0066	-.24	-.0070			-.00068
PCB 170	R <sup>2</sup>	0.25	0.11	-	-	-	0.09
	β	.011	-.30				-.00081
PCB 180	R <sup>2</sup>	0.29	0.11	-	-	-	0.09
	β	.012	-.32				-.00081
PCB 183	R <sup>2</sup>	0.07	0.12	-	-	-	0.10
	β	.0070	-.26				-.00080
PCB 189	R <sup>2</sup>	-	0.12	-	-	-	0.18
	β		-.27				-.0010
PCB 191	R <sup>2</sup>	-	0.12	-	-	-	0.23
	β		-.28				-.0012
PCB 194	R <sup>2</sup>	0.35	-	-	-	0.07	-
	β	.018				-.0013	
PCB 201	R <sup>2</sup>	0.35	-	-	-	-	-
	β	.015					
PCB 203/196	R <sup>2</sup>	0.30	0.06	-	-	-	0.05
	β	.013	-.28				-.00073
PCB 206	R <sup>2</sup>	0.26	-	-	-	0.10	-
	β	.015				-.0012	
PCB 207	R <sup>2</sup>	-	-	-	0.15	-	-
	β				-.0033		
PCB 209	R <sup>2</sup>	-	-	0.16	-	-	-
	β			-.012			
PCB 187/182	R <sup>2</sup>	0.25	-	-	-	-	-
	β	.011					
PCB 173/200	R <sup>2</sup>	-	-	-	0.12	-	-
	β				-.0020		
PCB 208/95	R <sup>2</sup>	-	-	0.53	-	-	-
	β			-.033			

- Variable was not entered in the step-wise regression (p > 0.05)

\* Sex coded as 0 = male and 1 = female;

† Food consumption = number of servings/year



showing higher levels in all congeners except PCB 28. Body mass index was significant for 5 congeners, with concentration increasing with decreasing BMI, suggesting that people with less adipose tissue had PCBs concentrated in a smaller volume of tissue. This may present another reason for the low PCB levels in the BC sample, since the BMI for many of the patients was high. The impact of BMI was assessed by modeling PCB concentrations using the population mean BMI; this indicated potential concentration increases of 11% to 50%, depending on the regression coefficient for BMI. Many of the comparison studies also sampled hospital patients (Fujiwara 1975; Holt et al 1986; Jensen and Clausen 1979; Mori et al 1983; Teufel et al 1990); it is possible that these samples had similar weight distributions, though data on size were not reported.

Residence in a pulp mill town and eating sour cream were never significant in the multiple regression. Consumption of either wild meat, cottage cheese, or potatoes was significant for two or more congeners. The regression coefficients for all the included food variables suggest that as consumption increased, PCB concentrations decreased. An explanation for this relationship might be related to the effect of BMI. Patients who eat more food may have more body fat in which to store their organochlorine burden. The results of the diet analyses need to be interpreted with caution, however, since food consumption is difficult to measure and varies significantly within an individual over time. It is likely that larger sample sizes would be needed to demonstrate consistent and plausible relationships. Type of surgery was not found to be a statistically significant determinant of concentration for any congener.

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