

Distribution of Polychlorinated Biphenyl Congeners in Human Milk and Blood During Lactation

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The polychlorinated biphenyl (PCB) congener distribution in human milk has been reported by several investigators (YOSHIDA et al. 1977; YAKUSHIJI et al. 1979; MES et al. 1980). Yoshida et al. (1977) found less pentachloro-, but more hexachlorobiphenyl in Swedish than Japanese breast milk. Mes et al. (1980) reported both less penta- and hexachlorobiphenyls, but considerable more hepta-chlorobiphenyl in Canadian than Japanese human milk. Although these variations in congener distribution of human milk could be ascribed to differences in environmental contaminants or even extraction techniques (MES et al. 1980), the possibility exists that congener distribution changes during lactation. This possibility was investigated by determining both breast milk and maternal blood PCB congeners of a nursing mother, whose blood and milk PCB levels were earlier reported (MES et al. in press).

MATERIALS AND METHODS

All solvents were glass distilled and tested for interfering substances by gas liquid chromatography (GC). Glassware was heat treated and rinsed with solvents as earlier reported (MES 1981).

The milk and blood samples were extracted and the extracts cleaned up and fractionated according to MES et al. (in press). Essentially milk samples were centrifuged, the resultant water phase removed and the remaining fat extracted with acetone:benzene (19:1, v/v). Blood samples were extracted with benzene and centrifuged.

Extracts were cleaned up on 2% water deactivated Florisil and the PCBs collected in two fractions: fraction I (35 ml hexane) and fraction II (20% dichloromethane in hexane). The PCB content of the two fractions was earlier determined by capillary GC and reported by MES et al. (in press).

The fractions were concentrated to approximately 50 pg estimated PCBs/ μ l hexane for mass spectral (MS) analysis. The GC/MS analysis was carried out by using a Varian 3700 Series gas chromatograph with splitless injector and a chemically bonded SE-54 fused silica capillary column (30 m x 0.25 mm). The column temperature was programmed in two stages: after initially being held for 1 min at 70°C, heated to 170°C within 3 min, followed by subsequent heating to 260°C at 4°C/min. Multiple Ion Detection (MID) was carried out on a VG Analytical ZAB-2F mass spectrometer with an ion source temperature of 250°C, an electron energy of 70 eV and a resolution (10% valley definition) of 3000. Dwell time for each ion was 200 msec. Table 1 lists the respective isomers used for

quantitation, with their corresponding ions and isotope ratios used for the determination of each congener. Only those peaks with isotope ratios within + 20% of their theoretical values and within the expected retention time window were considered to be PCB peaks and used for quantitation.

GC/MS analysis of PCB standards was carried out using 1-50 pg/ μ l, depending on the number of chlorine atoms present.

Statistical evaluation was carried out using correlation and linear regression analysis.

Table 1. Isomers and corresponding ions used in the MS determination of PCB congeners

Number of Chlorine atoms	Substitution pattern of PCB standard	Ions m/z (%) ^a
1	4	188
2	3,3'	222
3	2',3,4	256
4	2,2',5,5'	290 (77), 292 (100), 294 (49)
5	2,3,4,4',5	324 (62), 326 (100), 328 (65)
6	2,2',4,4',5,5'	360
7	2,2',3,4,5,5',6	392 (44), 394 (100), 396 (97)
8	2,2',3,3',5,5',6,6'	428 (88), 430 (100), 432 (65)
9	2,2',3,3',4,4',5,5',6	462 (77), 464 (100), 466 (76)
10	2,2',3,3',4,4',5,5',6,6'	496 (68), 498 (100), 500 (87)

a (%) = Theoretical isotope ratio

RESULTS AND DISCUSSION

The results in Table 2 show the percentage PCB congener distribution in the breast milk of a woman during 98 days of her lactation period.

Table 2. Percent PCB congener distribution in human milk during lactation

Number of Chlorine atoms ^a	Days following parturition							
	7	14	28	42	56	70	84	98
1	0.03	0.02	0.05	0.02	0.03	0.02	0.04	0.01
2	0.45	0.42	0.72	0.43	0.46	0.44	0.63	0.43
3	1.08	1.50	2.06	2.16	2.05	1.76	2.69	0.78
4	8.44	9.37	9.92	12.70	12.55	10.88	8.20	9.84
5	20.11	21.68	18.77	22.29	21.87	17.14	22.32	15.43
6	16.19	22.67	25.55	31.01	34.96	28.82	30.56	30.91
7	44.48	37.46	36.53	23.75	20.60	35.74	31.12	35.45
8	7.29	5.85	5.66	6.33	6.39	4.03	3.38	6.12
9	1.47	1.03	0.52	1.14	0.94	0.96	0.86	0.75
10	0.46	0.20	0.23	0.17	0.16	0.21	0.20	0.27

In biphenyl molecule

Although some fluctuation occurs, the PCB congener content remains relatively constant in the breast milk during lactation, except for those congeners with 6 and 7 chlorine atoms in their biphenyl molecule.

A statistically significant increase was observed in the hexachlorobiphenyl content of the breastmilk during lactation ($r = 0.743$ with 95% confidence level). During the 98 days of lactation no statistically significant decrease in the heptachlorobiphenyl content of the breastmilk could be established. A significant decrease, however, was observed for the first 56 days of lactation.

The variation in the percent congener distribution of breastmilk from different countries, as earlier mentioned, is particularly noticeable for the hexa- and heptachlorobiphenyls. These variations in congener distribution could possibly be attributed to the differences in time at which samples were collected during lactation. The data in Table 2 seem to corroborate this possibility at least for the first 56 days following parturition. After 56 days the ratio of hexa- and heptachlorobiphenyls remains relatively constant.

Table 3. Percent PCB congener distribution in maternal blood during lactation

Number of Chlorine atoms ^a	Days following parturition							
	7	14	28	42	56	70	84	98
1	N.D. ^b	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
2	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
3	3.96	5.29	2.11	1.69	1.99	0.57	3.88	2.55
4	4.89	4.70	1.81	2.55	3.51	3.42	3.37	14.49
5	5.36	9.35	3.61	4.31	4.02	2.79	3.88	3.33
6	81.25	77.66	88.69	84.38	81.28	85.84	77.72	72.28
7	3.54	2.18	2.83	2.63	3.98	3.87	3.27	4.57
8	0.64	0.82	0.96	0.78	1.21	1.03	1.12	0.85
9	0.36	N.D.	N.D.	0.47	0.66	0.68	1.02	N.D.
10	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

a In biphenyl molecule; b N.D. = not detected

At least 10% of all PCB's in the breastmilk of this donor were mono-, di- and trichlorobiphenyls.

The results in Table 3 show the percentage congener distribution in the maternal blood of the same donor, who supplied the breastmilk samples. Breastmilk and blood samples were collected on the same day.

A small, but statistically significant increase of the heptachlorobiphenyl congeners ($r = 0.689$ with 90% confidence level) in maternal blood was observed during lactation, together with a significant decrease in the pentachlorobiphenyl congener ($r = -0.639$ with 90% confidence level). A similar significant decrease in the hexachlorobiphenyl congener ($r = -0.873$

with 99% confidence level) could only be found after the 28th day following parturition.

Table 4. A comparison of the quantitation of PCB's by capillary GC and combined capillary GC/MS

Days following parturition	ng PCB/g substrate			
	Maternal blood		Breastmilk	
	GC ^a	GC/MS	GC ^a	GC/MS
7	2.1	3.1	11.6	7.3
14	2.5	3.3	9.2	7.4
28	2.1	3.0	11.3	6.8
42	2.0	3.4	12.8	9.2
56	1.5	2.9	13.9	11.3
70	1.0	2.1	9.5	7.0
84	3.1	1.7	10.0	7.2
98	2.0	2.5	12.4	9.1
Average (C.V.) ^b	2.0 (29)	2.8 (20)	11.3 (14)	8.2 (18)

^a Unpublished data from a previous study by MES et al. (in press).

^b C.V. = coefficient of variation.

Although the hexachlorobiphenyl content decreased in the blood and increased in the breastmilk after 28 days of lactation while the heptachlorobiphenyl content decreased in the milk, but increased in the blood during the first 56 days following parturition, no statistically significant relationship was observed between these congeners in maternal blood and breastmilk.

The determination of PCB's in maternal blood and breastmilk by both capillary GC and capillary GC/MS is given in Table 4. The average PCB levels obtained by the two methods differ by <30% for each substrate. Although this difference may be expected given the low residue levels and the fact that PCBs are only approximated by either method, there is no ready explanation why the GC/MS values are higher in blood, but lower in breastmilk compared to the respective GC results.

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REFERENCES

- MES, J., DAVIES, D.J., and LAU, P.-Y. Chemosphere, 9, 763 (1980).
 MES, J. Int. J. Environ. Anal. Chem., 9, 283 (1981).
 MES, J., DOYLE, J.A., ADAMS, B.R., DAVIES, D.J., and TURTON, D. (in press).

YOSHIDA, S., and NAKAMURA, A. Proceedings of the Osaka Prefecture, Int. Publ. Health, 8, 67 (1977).

YAKUSHIJI, T., WATANABE, I., KUWABARA, K., YOSHIDA, S., KOYAMA, K., and KUNITA, N. Int. Arch. Occup. Environ. Health, 43, 1 (1979).

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