

## **PCB Congeners in Human Milk in Germany from 1984/85 and 1990/91**

S. Georgii,<sup>1</sup> G. Bachour,<sup>1</sup> I. Elmadfa,<sup>2</sup> H. Brunn<sup>1</sup>

<sup>1</sup>State Laboratory of Middle Hesse for Medicine, Food Chemistry and Veterinary Medicine, Marburger Straße 54, D-35396 Giessen, Germany

<sup>2</sup>Institute for Nutrition Science, Lammgasse 8, A-1080 Vienna, Austria

Received: 10 April 1994/Accepted: 29 August 1994

Studies of so-called bioindicators clearly show the impact of environmental legislation on PCB contamination of the environment and of the food chain in the Federal Republic of Germany. This is evident from the results of studies performed in our laboratory on samples of muscle tissue from foxes that were submitted for post-mortem examination in the years 1983, 1987 and 1991. Whereas the concentration of low-chlorinated congeners increased throughout this time period, concentrations of high-chlorinated biphenyls, with the exception of PCB no. 101, decreased (Georgii et al. 1994). This trend may also be responsible for changes in the otherwise stable PCB concentrations in human milk. Consequently, in the study describe here, milk samples from 1990/91 from women in Middle-Hesse, Germany, were examined and the results compared to samples stemming from 1984/85.

### **MATERIALS AND METHODS**

Sample material consisted of milk from first-bearing German women from the area of Middle-Hesse, W. Germany. The ages of the women were 19–30 years (samples from 1984/85, n=69) and 25–30 years (samples from 1990/91, n=68). PCB determination was done by gas chromatography on three capillary columns with different polarities as well as by means of coupled capillary gas chromatography / mass spectroscopy in NCI mode. Calculation of total contamination with PCDD and PCDF was performed by multiplying the concentrations according to Furst et al. (1989) (average values) with the appropriate toxicity equivalent factors (revised TEF according to Safe (1990)).

Statistical analysis: Data management and evaluation were performed using the following systems: an IBM AT personal computer with the statistics software package STATGRAPHICS Vers. 2.6 (STSC Inc. Software Publishing Group, Rockville, Maryland 20852, USA); a CDC Cyber 960 at the computing center of the University of Giessen, using the statistics package BMDP (for details see Dixon 1987).

*Correspondence to:* H. Brunn

For the variables PCB nos. 138 and 153 it was possible by logarithmic transformation to obtain a near-normal distribution of the test values allowing use of a t-test for independent random samples. For the other congeners determined (PCB nos. 28, 49, 52, 101 and 180), the distribution of the values obtained was discrete, making it necessary to use a ranking-test (Wilcoxon-Mann-Whitney-Test).

Abbreviations: PCB polychlorinated biphenyl(s); min minimum value;  $q_1$  first quartile;  $\bar{x}$  median;  $q_3$  third quartile; max maximum value;  $\bar{x}$  arithmetic mean.

## RESULTS AND DISCUSSION

Tables 1 and 2 present a comparison of the polychlorinated biphenyl concentrations in human milk in 1984/85 and 1990/91. These data show that the high-chlorinated congeners nos. 138, 153 and 180 (nomenclature according to Ballschmiter and Zell 1980) remain the chief PCB contaminants in milk fat. It must be noted, however, that the concentrations of PCB nos. 138 and 153 show a statistically significant reduction in 1990/91 over 1984/85. In contrast, contamination with the low chlorinated biphenyls nos. 28, 49 and 52 increased throughout the same time period. Concentrations of PCB no. 101 remained almost unchanged. Pentachlorobiphenyl 101 is a component of both the high-chlorinated technical mixture, Clophen A60®, and Clophen A30® a low-chlorinated mixture. These results indicate a reduction in the release of Clophen A60® into the environment, but an increase in contamination of the general environment and the food chain by the congeners present in Clophen A30®.

The fact that PCB no. 180 is found in higher concentrations than no. 138 can be explained by differences of metabolism of these two congeners. In general, as the release of high-chlorinated biphenyls into the food chain is reduced, a consequent reduction is becoming evident in the concentrations of high-chlorinated biphenyls in human fat tissue and in human milk. It therefore appears that the change in the relationship between the various PCBs in milk in 1990/91 is the result of the fact that hexachlorobiphenyls no. 138 and no. 153 are more rapidly biotransformed, due to their substitution patterns, than is PCB no. 180 (Schulte and Acker 1974). This same development was indicated in experiments in which human fat tissue was examined (Brunn et al. 1990) a few years ago.

The milk samples from 1990/91 were also analyzed to determine concentrations of the monoortho- and diortho-chlor substituted, "dioxin-like" PCB nos. 118, 156 and 170. The concentrations of these congeners as well as those of congener no. 138 were recalculated with the help of preliminary toxicity equivalent factors (Safe 1990) in 2,3,7,8-TCDD toxicity equivalents. Comparison of these results with the sum of the resultant toxicity equivalents from PCDD- and PCDF-contamination (Fürst et al. 1990) show that the 2,3,7,8-TCDD equivalents (TEQ) calculated from

PCB concentration are almost three times greater than PCDD- and PCDF-contamination of milk from German women (Table 3). These results agree with those obtained from human milk as reported by Dewailly et al. (1991) and Böhm et al. (1993).

Table 1. PCB congeners in human milk from Middle-Hesse, Germany 1984/85 (n=69)

PCB congener	min	q <sub>1</sub>	$\tilde{x}$	q <sub>3</sub>	max	$\bar{x}$
[µg/kg milk fat]						
no. 28**	0.5	0.5	5	0.5	40	0.5
no. 49**	0.5	0.5	11	15	70	0.5
no. 52**	0.5	0.5	2	0.5	60	0.5
no.101	0.5	0.5	21	35	90	15
no. 138**	60	190	254	330	660	250
no. 153**	70	250	324	400	750	325
no. 180*	40	110	162	200	400	160

\*, \*\*: significant ( $p < 0.05$ ) or highly significant ( $p < 0.01$ ) differences, respectively, in the PCB concentrations between the years 1984/85 and 1990

Table 2. PCB congeners in human milk from Middle-Hesse, Germany 1990/91 (n=68)

PCB congener	min	q <sub>1</sub>	$\tilde{x}$	q <sub>3</sub>	max	$\bar{x}$
[µg/kg milk fat]						
no. 28**	9	12	18	20	46	17
no. 49**	7	12	22	28	87	18
no. 52**	6	9	15	18	44	13
no.101	0.5	10	17	23	55	14
no. 138**	65	151	184	213	669	168
no. 153**	108	214	264	303	968	240
no. 180*	75	144	194	236	1023	173

Legend see table 1

Table 3. "Dioxin-like" PCB congeners in human milk

Substance	$\bar{x}$ [µg/kg]	TEF (Safe 1990)	TE [µg/kg]
2,3',4,4',5- P <sub>5</sub> CB (no. 118)	44	0.001	0.044
2,2',3,4,4',5'- P <sub>6</sub> CB (no. 138)	168	0.00002	0.00336
2,3,3',4,4',5- P <sub>6</sub> CB (no. 156)	27	0.001	0.027
2,2',3,3',4,4',5- P <sub>7</sub> CB (no. 170)	25	0.00002	0.00005
Total PCB	--	--	0.075
Total PCDD/F	--	--	0.028

n = 68

Calculated on the basis of milk fat

The exposure to polychlorinated biphenyls may be of importance to those children that are breast fed for extended periods. The excretion kinetics of PCB congeners nos. 101,138, 153 and 180 throughout such longer periods (196 compared to 84 days) show that the PCB concentrations in human milk do not change measurably after the 14th day *post partum* (Georgii et al. 1988). As a consequence, it cannot be expected that the exposure of infants to polychlorinated biphenyls will decrease, even after extended lactation periods.

## REFERENCES

- Ballschmiter K, Zell M (1980) Analysis of polychlorinated biphenyls (PCB) by glass capillary gas chromatography. *Fresenius Z Anal Chem* 302: 20-31
- Böhm V, Schulte E, Thier H-P (1993) Polychlorinated biphenyl residues in food and human milk: determination of co-planar and mono-ortho substituted congeners. *Z Lebensm Unters Forsch* 196:435-440
- Brunn H, Georgii S, Prucha J (1990) Polychlorierte Biphenyle (PCB) im menschlichen Fettgewebe. *Z Lebensm Unters Forsch* 190:108-11
- Dewailly E, Weber J-Ph, Gingras S, Laliberte C (1991) Coplanar PCBs in human milk in the Province of Quebec, Canada: Are they more toxic than dioxin for breast fed infants ? *Bull Environ Contam Toxicol* 47:491-498

- Dixon WL (ed.) (1987) BMDP statistical software. University of California Press, Berkeley Los Angeles London
- Fürst P, Krüger C, Meemken HA, Groebel W (1989) PCDD and PCDF levels in human milk - dependence on the period of lactation. *Chemosphere* 18:439-44
- Georgii S, Bachour Gh, Failing K, Eskens U, Elmadfa I, Brunn H (1994) PCB congeners in foxes in Germany from 1983 to 1991. *Arch Environ Contam Toxicol* 26:1-6
- Georgii S, Muskat E, Kleinstein J, Schubring Chr, Brunn H (1988) PCB-Einzelkomponenten und chlororganische Pestizide in Frauenmilch in Abhängigkeit von der Stilldauer. *Ernährungs-Umschau* 35:352-356
- Safe S (1990) Development of toxic equivalency factors (TEFs) for halogenated aromatic hydrocarbons. In: Hutzinger O, Fiedler H, eds. *Organohalogen Compounds, Vol. 1. Bayreuth: Dioxin '90 - EPRI Seminar Toxicology, Environment, Food, Exposure - Risk*, 329-31
- Safe S (1990) Polychlorinated biphenyls (PCBs), dibenzo-p-dioxins (PCDDs), dibenzofuranes (PCDFs), and related compounds: environmental and mechanistic considerations which support the development of toxic equivalency factors (TEFs). *Crit Rev Toxicol* 21:51-58
- Schulte E, Acker L (1974) Identifizierung und Metabolisierbarkeit von polychlorierten Biphenylen. *Naturwiss* 61:79-80