Still relatively high PCDD/PCDF concentrations in human milk of mothers living in a contaminated area in Germany

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Introduction

Eight years ago, the small town Ilsenburg at the Harz (federal land: Saxony-Anhalt, Germany) was identified as an area highly contaminated with PCDDs and PCDFs, resulting from emissions of a copper plant including cable and electronic waste melting. In 1990/91, human milk samples of 9 mothers living in Ilsenburg were analysed for PCDDs/PCDFs (1,2). The mean I-TEq concentration (59 pg I-TEq/g milk fat) was about twice the mean level of 728 samples collected during the same time in Western Germany (31 pg I-TEq/g) (2).
The copper plant in the former GDR was closed in 1990. In order to determine the expected decline of PCDD/PCDF concentrations, 10 human milk samples were collected in 1997 from mothers living directly in Ilsenburg or within a radius of 8 km.

Methods

For PCDD/PCDF analysis, the same methods were used as in 1991 (2). All concentrations were based on fat weight. I-TEq levels were calculated using I-TEFs (3) not including PCBs.

Results and Discussion

For comparison, PCDD/PCDF concentrations measured in 1990/91 (1,2) are presented in Table 1, including basic data with known main influence on PCDD/PCDF body burden (age, years lived near to Ilsenburg , nursing weeks for other children and time of sample collection). Four of the nine mothers had worked at the industrial complex next to the copper plant, but not in the plant itself. Most mothers consumed fruit and vegetable from own garden (as well as those from the 1997 samples).
Concentrations of PCDDs/PCDFs measured in 1997 are compiled in Table 2. Maximum level was 81 pg I-TEq/g. This mother was the only one of the second series who worked at the industrial complex next to the copper plant (from 1976 until now). She is relatively old (36 years), but already breast-fed her first child for 17 weeks. Minimum level was 11 pg I-TEq/g, from a 37-year-old mother with first child, but she moved to the Ilsenburg area only one year ago. Nowadays, this concentration is typical for Western Germany (4) where she lived before.

Table 1 PCDD/PCDF concentrations (pg/g fat) in nine human milk samples collected **1990/91** from mothers living in Ilsenburg or surrounding area. Data from Alder et. al. (1,2), completed by basic data of the mothers.

No.	946	947	948	949	950	951	952	954	955	Mean
Age of mother (years)	24	27	28	21	23	36	29	28	23	27
Years lived										
A) within a radius of 4 km*	24	27	21	5	23	25	19	28	3	20
B) outside A), but within 15 km				16		11			20	5
C) outside a radius of 15 km			7				10			2
Other children breast-fed for (wks)	0	12	17	0	13	4	0	15	0	7
Sample collected (wks after deliv.)	4	4	4	3	4	3	7	4	8	5
2378-T4CDF	0.3	0.3	0.5	0.3	0.3	0.3	0.8	0.3	0.3	0.4
2378-T4CDD	6.6	3.1	6.3	4.9	3.8	7.8	7.0	5.6	6.7	5.8
12378-P5CDF	1.2	1.0	2.2	0.3	0.3	0.3	0.9	1.1	0.6	0.9
23478-P5CDF	57	33	43	50	37	67	84	47	66	54
12378-P5CDD	23	12	18	18	20	25	26	17	26	21
123478-H6CDF	41	25	41	30	24	31	46	29	39	34
123678-H6CDF	40	24	34	31	26	39	53	29	41	35
234678-H6CDF	15	11	30	17	12	27	39	15	11	20
Sum H6CDFs	95	60	105	78	62	97	137	73	92	89
123478-H6CDD	16	12	17	13	8.4	17	27	17	23	17
123678-H6CDD	40	22	34	40	36	39	56	26	64	40
123789-H6CDD	15	6.1	11	11	5.9	9.8	12	7.4	14	10
Sum H6CDDs	71	39	62	64	51	66	95	51	101	67
1234678-H7CDF	22	10	14	9.0	3.6	3.6	15	8.6	14	11
1234678-H7CDD	24	23	21	43	17	26	43	23	33	28
OCDF	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
OCDD	157	104	65	475	147	134	202	182	287	195
I-TEq	**64**	**36**	**54**	**54**	**44**	**71**	**86**	**51**	**73**	**59**

*Ilsenburg and the villages Drübeck, Darlingerode, Veckenstedt

Mean age of mothers was higher in 1997 (30 years vs. 27 years in 1990/91), but the average sum of years the mothers had lived near to the copper plant (radius of 4 km) was lower in 1997 (16 vs. 20 years). Additionally, the 1997 mothers on average had breast-fed other children for a longer time (20 vs. 7 weeks), and the milk samples were collected later (19 vs. 5 weeks after delivery). Nevertheless, mean I-TEq level (41 pg I-TEq/g) of the 1997 samples was still relatively high (even 48 pg I-TEq/g if only the 8 mothers were taken who had lived more than half their life within a radius of 15 km).

Compared to the 1990/91 samples (mean 59 pg I-TEq/g), this decrease might seem low, especially if compared to the general decrease of PCDD/PCDF background concentrations in human milk from Western Germany in recent years: from 31 pg I-TEq/g in 1990 (2) to 14 pg I-TEq/g in 1996 (4). However, this is a result of a continually reduced daily PCDD/PCDF intake (due to reduced emissions) which can be assumed to have started long before 1990 and which leads to a lower increase of PCDD/PCDF concentrations during life-time. On the contrary, we

Table 2 PCDD/PCDF concentrations (pg/g fat) in ten human milk samples collected in 1997 from mothers living in Ilsenburg or surrounding area, and basic data of the mothers.

No.	1073	1074	1075	1076	1077	1078	1079	1080	1081	1082	Mean
Age of mother (years)	31	33	29	30	27	37	27	25	36	28	30
Years lived											
A) within a radius of 4 km*	2**		29	9**	7**	1**	27	25	36	21	16
B) outside A), but within 15 km	19	33			20						7
C) outside a radius of 15 km	10			21		36				7***	7
Other children breast-fed for (wks)	48	0	0	12	22	0	44	31	17	28	20
Sample collected (wks after deliv.)	19	22	49	12	12	8	17	15	18	21	19
2378-T4CDF	<0.5	<0.5	<0.5	<0.5	<0.5	2.1	<0.5	1.4	<0.5	<0.5	
2378-T4CDD	2.4	4.5	7.5	3.4	2.6	0.9	4.0	5.8	8.3	4.4	4.4
12378-P5CDF	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	1.3	<0.5	<0.5	
23478-P5CDF	21	53	55	17	26	10	52	52	93	45	42
12378-P5CDD	8.0	15	19	6.2	7.7	4.2	15	16	25	13	13
123478-H6CDF	5.7	13	60	7.2	10	3.0	23	27	28	14	19
123678-H6CDF	9.4	13	37	6.0	10	2.5	20	28	30	14	17
234678-H6CDF	2.9	4.1	30	2.2	2.8	1.3	11	12	12	3.9	8.2
Sum H6CDFs	18	30	127	15	23	6.8	54	67	69	31	44
123478-H6CDD	4.6	9.4	18	4.5	3.7	2.4	13	14	14	11	9.4
123678-H6CDD	15	34	36	13	13	13	33	31	36	25	25
123789-H6CDD	2.3	4.8	6.4	4.0	2.2	1.4	5.9	8.8	7.3	3.7	4.7
Sum H6CDDs	22	48	61	22	19	17	52	53	57	39	39
1234678-H7CDF	6.7	1.0	5.5	1.0	2.3	2.1	5.1	6.0	4.0	3.0	3.7
1234678-H7CDD	19	15	30	17	12	12	71	40	25	25	27
OCDF	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	1.0	<0.5	0.7	
OCDD	73	57	90	111	78	77	143	137	135	145	105
I-TEq	**22**	**47**	**64**	**19**	**24**	**11**	**49**	**52**	**81**	**41**	**41**

* Ilsenburg and the villages Drübeck, Darlingerode, Veckenstedt
** during recent years; *** 1988-1995

have to assume a high PCDD/PCDF intake mainly from food in Ilsenburg until 1990 (closure of the copper plant; probably in the years before PCDD/PCDF emissions were highest due to melting of electronic waste). Thereafter, the area around Ilsenburg was no longer used for agriculture, common recommendations were published (e.g. no eggs from hens living on the ground), and with the social changes, food was mainly obtained from supermarkets (widespread origin).

Indeed, food monitoring performed during the last years in a few cows' milk and grain samples showed no conspicuous PCDD/PCDF concentrations (personal communication by Dr. Siegl, Veterinary Office, Wernigerode), with the exception of one carp sample from 1990 (444 pg I-TEq/g fish fat). In order to investigate the general PCDD/PCDF intake from food in Ilsenburg, a duplicate study was performed with seven male and seven female volunteers in 1994. The results did not show a higher PCDD/PCDF intake in these persons (0.91 and 0.79 pg I-TEq/kg*day in

the male and female, respectively) (5), compared to the general German population (decreased from about 2 pg I-TEq/kg*day in 1989 to about 1 pg I-TEq/kg*day in 1996) (4).

Assuming an average daily PCDD/PCDF intake of 1 pg I-TEq/kg*day since 1990 in a mother with a concentration in body fat of 60 pg I-TEq/g in 1990, a half-life of 7 years for the main congeners and 20% body fat, a concentration of 39 pg I-TEq/g can be calculated for 1997 as a rough estimate. This is near to the mean level measured. These mothers had an actual nutrition with food mainly obtained from supermarkets. Only mothers No. 1074 and 1075 had eggs from own hens living on the ground which are presently being analysed for their PCDD/PCDF concentrations.

In 1992, PCDD/PCDF blood analyses were performed in men from Ilsenburg who had never worked in the copper plant (6). Highest concentrations (172 pg I-TEq/g blood fat) were found in a 43-year-old man with a high consumption of fruit and vegetable from own garden as well as meat from self-bred pigs and rabbits.
In contrast to the relatively high background contamination in the Ilsenburg area, former workers of the copper plant had only moderately higher PCDD/PCDF blood concentrations (76.7 - 197.6 pg I-TEq/g blood fat, n=15) (7).

Compared to their mothers, infants will reach clearly higher PCDD/PCDF body concentrations after several months of breast-feeding (8). 11-month-old breast-fed children from the Ilsenburg area (as well as children from Berlin) are participating in an ongoing study which looks for changes of sensitive biomarkers possibly attributable to the PCDD/PCDF exposure. Results will be presented next year.

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