DIOXINS AND DIOXIN-LIKE PCBS IN HUMAN MILK FROM GERMANY – TIME TRENDS AND CHANGES BY USING WHO-TEFS FROM 2005

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Introduction:

Due to a variety of measures and regulations being in force in Germany since 20 years to reduce dioxin emissions, the dioxin levels in human milk and in blood samples decreased considerably [1,2,3,4]. A similar decline was observed in other European countries [5]. This distinct decrease of the background body burden corresponds to a similar decrease of dioxin concentrations in food of animal origin, which was reflected by the dietary dioxin intake [1]. At present, the time trend is decelerated and questionable [6, 7, 8]. The aggregation of data from the Federal Laender shall provide the basis for a better understanding of the time course, including the actual situation, of levels in human milk from Germany.
In 2005, WHO introduced revised TEFs for PCDDs/Fs and dioxin-like PCBs (dl-PCBs) [9]. The influence of the revised WHO-TEFs on TEQ-levels is compared to those TEFs introduced in 1998.

Materials and methods:

***Origin of data*:**
On request of concerned mothers, human milk samples, were analysed by the food control laboratories of the Federal Laender Baden-Württemberg, Lower Saxony and North Rhine-Westphalia. The German data of the 3rd WHO Human Milk Field Study conducted in 2000/2002 were included in the survey as well as some results obtained with pooled samples[10]. All data on human milk were collected in the German Data Bank for Residues in Human Milk. Altogether, data on PCDDs/Fs from 646 human milk samples collected between 1998 and 2004, and data on dl-PCBs from 254 samples collected between 2000-2003, were included.

***Data evaluation*:**
The program SPSS was used for statistical evaluation. Pooled samples were included according to their corresponding individual sample number. For concentrations of single congeners below the LOQ values equal to one half of the LOQ were laid down. To compare the influence of different WHO-TEFs, only those samples were included, which provide data on both, PCDDs/Fs and dl-PCBs.

Results and discussion:

Current levels and time trends of PCDD/F background contamination
In Germany, between 1990 and 1997 the mean values of dioxin background concentrations in human milk decreased steeply by about 60% from 35.7 to 13.5 ng WHO-PCDD/F-TEQ/kg fat. The decline of both, the 95th percentile and the maximum value, were in the same order of magnitude [1,4]. For 2004, the mean dioxin concentration was calculated to be 9.2 ng and the 95th percentile to be 15.7 ng WHO-PCDD/F-TEQ/kg fat, respectively. This mean value is consistent with results of the Bavarian breast milk survey from 2005 which are not included in the present study[8]. Data on PCDDs/Fs in breast milk samples collected in the period between 1997 and 2004, are summarised in Table 1.

Depending on the regional provenience within Germany, minor differences in levels of PCDDs/Fs and dl-PCBs in human milk may occur. The same is true for meat and meat products, samples of which were collected representatively in different regions of Germany [12].

Table 1: PCDD/F background concentrations in human milk from Germany (ng WHO-PCDD/F-TEQ/kg fat; WHO 1998 TEF)

Sampling Year	Number of samples	Min	Mean	Median	95 Perc.	Max
1997	126	7.0	13.5	15.7	27.1	33.3
1998	69	5.4	15.0	13.9	26.7	31.9
1999	72	4.5	13.8	14.7	23.8	33.9
2000	86	4.2	14.5	13.8	27.0	43.0
2001	154	1.8	13.2	12.0	27.3	37.1
2002	157	2.9	12.5	12.4	22.2	30.2
2003	51	2.5	10.6	9.3	22.3	30.4
2004	57	3.1	9.2	8.9	15.7	25.6
Decrease 1997-2004			- 32%	- 43%	- 42%	- 23%

As shown in Figure 1, the decline of the background concentrations of PCDDs/Fs seems to continue over the period 1997 to 2004. During those 8 years, the mean values and the 95th percentiles decreased by 30-40%. In comparison to the former period from 1990 to 1997, this decrease is of lower magnitude and slowing down.

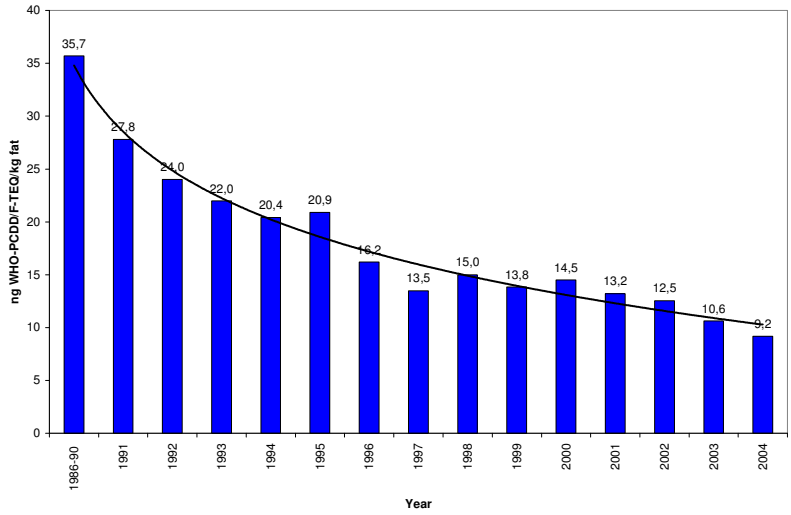

Figure 1: Time trend of the mean levels of WHO-PCDD/F-TEQs in human milk from Germany between 1990 to 2004 (Number of samples >2300; data from Bavaria, analysed between 1990 and 1997, included)

Figure 1 shows mean concentrations in the period 1998–2000 being slightly higher than in 1997. This tendency, even though slight, was found in human milk and in human blood samples at the same time. Presumably, the slightly higher body burden during this period must be associated with the consumption of food produced with feed containing Brazilian citrus pulp as an ingredient with high levels of dioxins [11].

Levels and time trend of the dl-PCB background contamination

Data on dl-PCBs in human milk have been reported for the years 2000 to 2003 from the Federal Laender Baden-Württemberg and North Rhine-Westphalia. The mean values of non-ortho PCBs, mono-ortho PCBs and the total

of WHO-TEQs are summarised in Table 2. They were obtained from samples with concentrations of both, dl-PCBs and PCDDs/Fs.

Table 2: Mean values of non-ortho PCBs, mono-ortho PCBs, and the sum of dl-PCBs and PCDDs/Fs in human milk from Germany (WHO 1998 TEF)

Sampling Year	Number of samples	Non-ortho PCBs [ng WHO-PCB-TEQ/kg fat]	Mono-ortho PCBs [ng WHO-PCB-TEQ/kg fat]	dl-PCBs+PCDDs/Fs [ng WHO-TEQ/kg fat]
2000	11	9.4	10.8	34.6
2001	96	8.0	6.9	28.5
2002	118	7.0	6.2	26.7
2003	29	5.2	5.5	23.3

Although the number of samples per year varies and is low in 2000 and in 2003, a decline of the mean levels of dl-PCBs is likely and calculated to be about 45-50% in this 4 years period. In comparison, during the same period of time the mean level of PCDDs/Fs decreased by 20% in the same data set.

Changes by using the revised WHO 2005 TEFs

To get a more comprehensive data set, samples analyzed for both, PCDDs/Fs and dl-PCBs, and collected between 2001 and 2003, were summarised for the following calculations.

Based on the WHO 1998 TEFs, the contribution of non-ortho PCBs, mono-ortho PCBs, PCDDs and PCDFs to the total of WHO-TEQs is similar and ca. 20-30%, of each, respectively. The most contributing congeners for TEQs are 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF in the group of PCDDs/Fs, and PCB 126 and PCB 156 in the group of dl-PCBs. These congeners account for about 75% of the total of WHO-TEQs..

In 2005, the WHO published revised TEFs [9]. While the TEFs were maintained for about half of the congeners, they were mostly lowered in the other cases. Calculating the WHO-TEQ-concentrations on the basis of the revised WHO-TEFs, the TEQs of PCDDs do not change whereas those of PCDFs and dl-PCBs are lower. WHO-PCDD/F-TEQs are reduced by 16%, and the decline of WHO-PCB-TEQ represents 34%. These changes are associated with a remarkable shift as to the contribution of different groups of congeners (Table 3, Figure 2). Comparable changes have been observed in another human milk study [14], while changes observed in food are slightly different [13, 15].

Table 3: Comparison of WHO-PCDD/F-TEQ, WHO-PCB-TEQ and WHO-TEQ, calculated on the basis of WHO 1998 TEFs and WHO 2005 TEFs [ng WHO-TEQ/kg fat]

	PCDDs	PCDFs	PCDDs/Fs	Non-ortho PCBs	Mono-ortho PCBs	dl-PCBs	Total
WHO-TEQ (TEF 1998)	7.5	6.0	13.5	7.4	6.0	13.4	26.9
Share in Total	28%	22%	50%	27%	22%	49%	
WHO-TEQ (TEF 2005)	7.5	3.9	11.4	8.0	0.8	8.8	20.2
Share in Total	37%	19%	55%	40%	4%	43%	
Changes	0%	-35%	-16%	-9%	-87%	-34%	-25%

A high contribution to the decrease of WHO-PCDD/F-TEQs is due to the relatively high concentrations of 2,3,4,7,8-PeCDF and its decline in TEF from 0.5 to 0.3. Consequently, the share of PCDDs contributing to WHO-PCDD/F-TEQs is growing when TEFs from 2005 are used, whereas PCDFs contribute accordingly lower. The considerable reduction of WHO-PCB-TEQs can be explained by the decline of TEFs for all but one mono-ortho PCBs from 0.0001/0.0003 to 0.00003. In breast milk samples a relatively high concentration of the mono-

ortho PCB 156 was found. As the TEF of this congener was lowered by a factor of ca. 15, WHO-PCB-TEQs decreased distinctly.

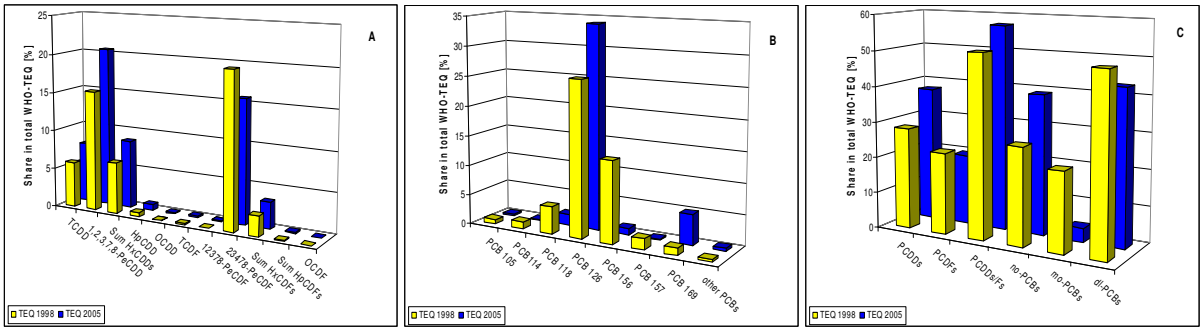

Figure 2: Comparison of the congener pattern calculated with WHO 1998 TEF and WHO 2005 TEF; A: PCDD/F congeners; B: dl-PCB congeners; C: PCDDs, PCDFs, PCDDs/Fs, non-ortho PCBs, mono-ortho PCBs, dl-PCBs

Current intake of PCDDs/Fs and dl-PCBs by breast-feeding

Based on the mean level of the total of WHO-TEQs in human milk from 2003, and using WHO 1998 TEFs the intake of a 4 month old breast-fed infant (mean body weight 6.5 kg) with a consumption of 800 mL (4% fat content) amounts to 115 pg WHO-TEQ/kg bw per day. This value is far higher than the TDI of 1-4 pg WHO-TEQ/kg bw and day derived by WHO in 1998. Nevertheless, WHO recommended breast-feeding for other relevant facts. Obviously, further efforts are mandatory to reduce PCDD/F/PCB body burden in humans, and specifically breast-fed infants.

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