TIME TRENDS AND CURRENT LEVELS OF DIOXINS AND DIOXIN-LIKE PCBS IN HUMAN MILK FROM GERMANY

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Introduction

Due to a variety of measures and regulations being in force in Germany for almost 30 years to reduce emissions of PCDDs/Fs, the PCDD/F levels in human milk and in blood samples decreased considerably [1, 2, 3, 4]. A similar decline was observed in other European countries [5]. The time trends on PCDDs/Fs in human milk from Germany have been reported until 2004 [1, 6]. During that time, the strong decrease of PCDDs/Fs observed in the 1990s has been decelerated and questionable [7, 8, 9]. In this study data on PCDDs/Fs and also on dioxin-like PCBs (dl-PCBs) in human milk from Germany collected between 2004 and 2009 are reported and the current time trend is discussed.

Materials and methods

Origin of data:
In most cases, human milk samples were analysed on request of the mothers. Between 2004 and 2009, human milk samples were analysed by the food control laboratories of the Federal Laender Bavaria, Lower Saxony and North Rhine-Westphalia. All data on human milk were collected in the German Data Bank for Residues in Human Milk [10]. Altogether, data on PCDDs/Fs from 2779 human milk samples have been analysed between 1986 and 2009, and data on dl-PCBs from 646 samples collected between 2000 and 2009 were included. Because of the high number of samples investigated this data set should be representative and permit reliable statements on temporal trends and the actual situation of levels in human milk in Germany.

Data evaluation:
The calculations are based on the WHO-TEFs derived in 1998. For concentrations of single congeners below LOQ values equal to one half of the LOQ were laid down.

Results and discussion:

Time trends and current levels of PCDD/F background contamination
In Germany, between 1990 and 1997 the mean values of PCDD/F background concentrations in human milk decreased steeply about 60%. The decline of both the 95th percentile and the maximum value were in the same order of magnitude [1, 4]. Between 1997 and 2004 the mean values and the 95th percentiles of WHO-PCDD/F-TEQs decreased by 30-40% only [6]. During those 8 years the decline seemed to decelerate. This slow down was also reported by others in human milk and in blood [7, 8]. Figure 1 shows that the mean concentrations of PCDDs/Fs in the period of 1998 and 2001 were slightly higher than 1997. This tendency was also found in blood samples at that time. Probably, this slightly higher body burden was associated with the consumption of food contaminated with PCDDs/Fs from Brazilian citrus pulp [11].
Since 2004 to 2009 a distinct decline of PCDDs/Fs in human milk from Germany is observed. During this 5 years period the mean values, the 95th percentiles and the maximum values decreased by 32 to 55%. During the last 10 years these values decreased by more than 55%. A consistent reduction of PCDD/F concentrations

between 2005 and 2008 has been found in the Bavarian breast milk study [12]. In 2009 the mean value of 6.3 ng WHO-PCDD/F-TEQ/kg fat is in the same range as the minimum values before 1998. It is also remarkable that the current maximum value of 11.5 ng WHO-PCDD/F-TEQ/kg fat is lower than the mean value 10 years ago. Data on PCDDs/Fs in breast milk samples collected between 1997 and 2009 are summarised in Table 1.
As shown in Figure 1, over the past 20 years the PCDD/F background contamination of human milk from Germany declined by more than 80% to 17% of the content measured 20 years ago. This is the result of regulations and technical measures to minimise environmental emissions and to reduce the entry into the food chain and the human body burden.

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	37.0
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
2005	30	2.1	7.8	7.7	12.7	19.1
2006	47	2.4	7.2	6.6	12.2	16.3
2007	205	1.2	6.2	5.8	11.6	15.1
2008	142	0.8	6.2	5.7	11.8	13.8
2009	46	2.4	6.3	6.0	10.0	11.5
Decrease 1997-2004			-32%	-43%	-42%	-23%
Decrease 2004-2009			-32%	-32%	-36%	-55%
Decrease 2000-2009			-57%	-56%	-63%	-73%

Time trends and current levels of dl-PCB background contamination
Data on dl-PCBs in human milk have been reported between 2000 and 2003 and between 2006-2009. The mean values of non-ortho PCBs, mono-ortho PCBs and total dl-PCBs as well as their share to total WHO-TEQs and the sum of dl-PCBs and PCDDs/Fs are summarised in Table 2. They were obtained from those samples, where concentrations of both, dl-PCBs and PCDDs/Fs are available.
Although in 2000 and in 2006 the number of samples is low and data from 2004 and 2005 data are missing, there is a distinct decline of the dl-PCBs between 2000 and 2009. The mean values of non-ortho and mono-ortho PCBs decreased from 9.4 to 3.8 ng WHO-PCB-TEQ/kg fat (-59%) and from 10.8 to 3.7 ng WHO-PCB-TEQ/kg fat (-66%) respectively. In this 10 years time period the relative contributions of non-ortho PCBs, mono-ortho PCBs and total dl-PCBs to total WHO-TEQs remain relatively constant about 26%, 26% and 52% respectively with slight variations. A comparable share of about dl-PCBs to total WHO-TEQs of about 50% has also been found in Bavaria (data not included in the present study) and in Sweden [9, 13]. As shown in Figure 1, the levels of dl-PCBs and PCDDs/Fs decline in parallel between 2000 and 2009.
At the time the mean total background contamination of dl-PCBs and PCDDs/Fs in human milk from Germany has decreased to 13.8 ng WHO-TEQ/kg fat and the 95th percentile is lower than 22 ng WHO-TEQ/kg fat.

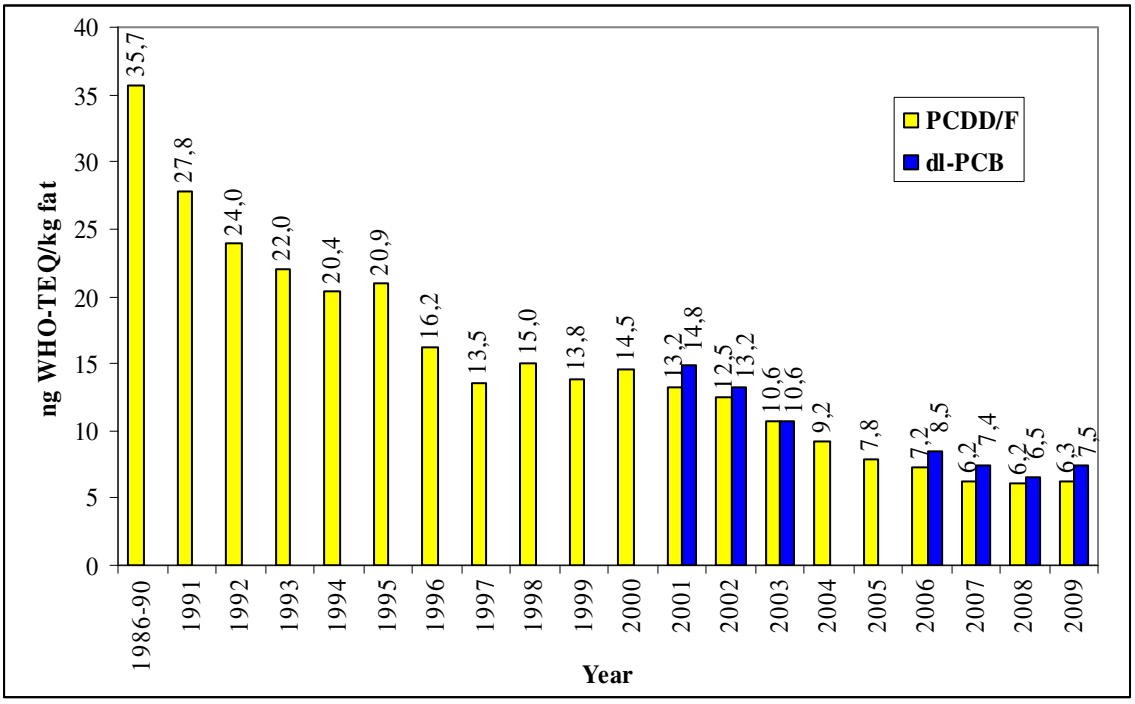

Figure 1: Time trend of the mean levels of WHO-PCDD/F-TEQs (Number of samples 2779) and WHO-PCB-TEQs (Number of samples 646) in human milk from Germany between 1986 to 2009

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

Sampling Year	Number of Samples	ng WHO-PCB-TEQ/kg fat			ng WHO-TEQ/kg fat	
		Non-ortho PCBs	Mono-ortho PCBs	dl-PCBs	dl-PCBs + PCDDs/Fs	
		Mean	Mean	Mean	Mean	95. Perc.
2000	11	9.4 (27%)	10.8 (30%)	20.2 (58%)	34.6	48.2
2001	96	8.0 (27%)	6.9 (24%)	14.8 (51%)	28.5	55.1
2002	118	7.0 (26%)	6.2 (23%)	13.2 (49%)	26.7	46.0
2003	29	5.2 (22%)	5.5 (23%)	10.6 (45%)	23.3	44.2
2006	6	4.1 (26%)	4.3 (28%)	8.5 (54%)	15.7	24.7
2007	200	3.8 (28%)	3.6 (26%)	7.4 (54%)	13.6	24.9
2008	141	3.4 (27%)	3.1 (25%)	6.5 (51%)	12.7	21.8
2009	45	3.8 (28%)	3.7 (27%)	7.5 (55%)	13.8	21.5
Decrease 2000-2009		-59%	-66%	-63%	-60%	-55%

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

According to the current data, an infant of three months (5,6 kg body weight, daily intake of 159 ml breast milk per kg body weight corresponding 31,4 g fat) who is exclusively breast fed, takes in a mean amount of 35 pg

WHO-PCDD/F-TEQ/kg body weight of PCDDs/Fs daily. In comparison, the average PCDD/F intake of an infant in 1990 amounted to 200 pg WHO-PCDD/F-TEQ/kg body weight daily. The average daily intake for the sum of PCDDs/Fs and dl-PCBs today is 77 pg WHO-TEQ/kg b.w. and the high intake (95th percentile) is 120 pg WHO-TEQ/kg b.w.

Although the TDI of 1 to 4 pg WHO-TEQ/kg body weight and day derived by WHO in 1998 is clearly exceeded during the short period of breastfeeding, the WHO working group fully recommends breastfeeding due to its health benefits for the child [14]. The German National Breastfeeding Committee recommends breastfeeding unconditionally, too [15]. Even in former times of elevated concentrations of contaminants in breast milk, the Committee did not see any reason to restrict the recommendation to breastfeeding. Nevertheless, further efforts are mandatory to reduce the background body burden of humans and specifically, breast-fed infants.

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