LEVELS OF ORGANOCHLOR PESTICIDES, PCB AND PCDD/PCDF IN HUMAN BREAST MILK – RESULTS OF THE BAVARIAN BREAST MILK MONITORING

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Introduction

The use of human breast milk for monitoring persistent environmental contaminants has a history of over half a century. In the past the Bavarian Health and Food Safety Authority performed exclusively breast milk analyses of persistent pesticides and selected organohalogen compounds. To implement further chemicals, especially those of toxicologically interest and public concern, the first survey of the Bavarian breast milk monitoring (BAMBI) was conducted in 2007/2008 comprising a comprehensive analysis including organochlorine pesticides (OCP), nitro musk compounds, polychlorinated biphenyls (PCB), polychlorinated dibenzo-p-dioxins (PCDD), dibenzofurans (PCDF), phthalates and perfluorinated compounds (PFC). The objectives were to get an overview of the background levels in human breast milk and to evaluate the daily intake of these compounds for exclusively breast-fed infants. Data about phthalates and PFC are published elsewhere [1, 2].

Materials and methods

The study population was recruited by midwives in Bavaria in the southern part of Germany. A total of 525 milk samples, 281 from urban areas and 244 from rural areas, were donated. The participating mothers obtained detailed information about the procedures regarding the sample collection together with manual suction pumps and cups to collect and store the milk sample. The samples were collected within the second month after delivery and stored in the freezer. Subsequently the frozen samples were dispatched to the Bavarian Health and Food Safety Authority and kept at -20 °C until analysis. All participants completed a detailed questionnaire on sociodemographic and anthropometric factors, smoking, nutrition and possible exposure through occupational contact. The ethic committee of the Bavarian Chamber of Physicians approved the study and written informed consent of all participants was obtained.
Sample preparation and measurement have been described elsewhere [3]. Briefly, after a clean-up procedure by gel permeation chromatography organochlorine pesticides and indicator PCB (ndl-PCB) were analysed by dual column gas-chromatography using an electron capture detector. Total ndl-PCB was calculated as the sum of PCB 153, PCB 138 and PCB 180 multiplied by 1.64. Total DDT was calculated as the sum of p,p'-DDE and p,p'-DDT multiplied by 1.11. PCDD/PCDF and dl-PCB were measured by HRGC/HRMS after fat preparation with various column chromatography applications based on the US EPA-Methods 1613 rev B and 1668 rev A [4].

Results and discussion

Study population
The subjects (59 % primiparas, 41 % multiparas) were between 15 and 43 years of age (median 31 years), with a median pre-pregnancy BMI ranging from 16 to 44 kg m^{-2} (median 23 kg m^{-2}) and weight reduction from delivery to sampling from 4 – 25 kg (median 10.7 kg). 82 % of the participating mothers were born in Germany, 9 % came from other European countries and 9 % were born outside of Europe. 4.3 % of the mothers smoked after birth and 7 % were daily exposed to passive smoking during pregnancy and after birth. 95 % of the recruited women were on mixed diet, 5 % of the mothers were vegetarians.

Organochlorine compounds nitro musks concentrations in breast milk

Median concentrations of organochlorine compounds and nitro musks in breast milk are presented in Table 1. HCB, p,p´-DDE and the three indicator PCB 138, PCB 153 and PCB 180 could be measured in all samples. The two nitro musk compounds, musk xylene and musk ketone were detectable in 8% and 1% of the samples, respectively. The median concentration of HCB, total DDT and total ndl-PCB were 0.016 mg kg^{-1} lipid (range 0.003 - 0.14), 0.07 mg kg^{-1} lipid (range 0.01 - 2.98) and 0.15 mg kg^{-1} lipid (range 0.003 - 1.9), respectively. Compared to the results of Bavarian breast milk samples collected in 2005 the levels have slightly decreased [3]. Strikingly, 5 % of the samples showed DDT levels above the reference value of 0.5 mg kg^{-1} lipid for human breast milk [5]. All samples with elevated DDT levels derived from mothers born in Eastern Europe, in countries of the former Soviet Union or South America.

Table 1: Concentration of organochlorine compounds and nitro musks in human breast milk (N= 516; mg kg^{-1} lipid, values below LOQ were calculated as half of the LOQ)

Compound	N>LOQ	Median	Min-max	95th Percentile
Organochloride pesticides				
HCB	516	0.016	0.003 - 0.140	0.036
α-HCH	3	—	0.002 - 0.012	0.002
β-HCH	474	0.006	0.002 - 1.520	0.059
Lindan	11	—	0.002 - 0.034	0.002
Dieldrin	21	—	0.002 - 0.016	0.002
Oxychlordane	69	0.002	0.002 - 0.006	0.004
HCEP	86	0.002	0.002 - 0.008	0.004
p.p`-DDE	516	0.063	0.008 - 2.330	0.460
p.p`-DDT	80	0.002	0.002 - 0.300	0.007
Total DDT	516	0.070	0.009 - 2.980	0.510
Nitro musk compounds				
Musk ketone	5	—	0.003 -0.013	0.003
Musk xylene	44	—	0.002 -0.048	0.004
ndl-PCB				
PCB 138	516	0.028	0.005 - 0.320	0.062
PCB 153	516	0.041	0.006 - 0.470	0.093
PCB 180	516	0.020	0.001 - 0.380	0.046
Total ndl-PCB	516	0.150	0.003 - 1.900	0.340

LOQ: limit of quantification

PCDD, PCDF and dl-PCB concentration in breast milk

The results of the dioxin and dl-PCB concentrations found in the breast milk samples are given in Table 2. Using WHO-TEFs of 1998, median values of non-ortho-PCB, mono-ortho-PCB, PCDD and PCDF were 3.2 pg WHO-TEQ g^{-1} lipid (range 0.2 – 10.0), 3.1 pg WHO-TEQ g^{-1} lipid (range 0.5 - 10.8), 3.2 pg WHO-TEQ g^{-1} lipid (range 0.2 - 11.1) and 2.6 pg WHO-TEQ g^{-1} lipid (range 0.6 - 6.6), respectively. Calculating the WHO-TEQ with the TEFs of 2005 the WHO-TEQ values were about 29 % lower as compared to the values calculated with WHO-TEFs of 1998. Compared to our data of breast milk from 2005 the total WHO-TEQ concentration in the present study is about 38 % lower [3].

The percentage of PCDD, PCDF, non-ortho- and mono-ortho-PCB to total PCDD/F/dl-PCB concentration applying different TEFs is shown in Figure 1. Considering WHO-TEF of 2005 the mean share of PCDD accounts for 36 %, PCDF for 19 %, non-ortho-PCB for 40 % and mono-ortho-PCB for 4 % on the total WHO-TEQ. Using WHO-1998-TEFs PCDD and PCDF account for 26 % and 21 %, respectively, whilst non-ortho-PCB and mono-ortho-PCB come to a share of 26% each.

We found the typical age-dependent increase of the dioxin concentration in breast milk. A significant association between the mother's smoking habit and the level of PCDD and dl-PCB have been demonstrated; non-smoking seemed to lead to higher levels in breast milk. A reduction of the body burden in the mothers who had breastfed more than one child was not observed.

Table 2: Concentration of PCDD, PCDF and dl-PCB in human breast milk (N=273, pg g^{-1} lipid)

Congener	Median	Min-max	95th Percentile
TEQ (WHO_{1998}-TEF)			
PCDD	3.2	0.2 - 11.1	6.4
PCDF	2.6	0.6 - 6.6	5.1
PCDDs/PCDF	5.7	0.8 - 15.1	11.4
Non-ortho-PCB	3.2	0.2 - 10.0	7.0
Mono-ortho-PCB	3.1	0.5 -10.8	6.6
dl-PCB	6.4	1.5 - 18.9	13.3
PCDD/PCDF/dl-PCB	12.3	2.3 - 31.4	23.7
TEQ (WHO_{2005}-TEF)			
PCDD	3.2	0.2 - 11.2	6.4
PCDF	1.7	0.4 - 4.3	3.3
PCDDs/PCDF	4.8	0.6 - 12.9	9.6
Non-ortho-PCB	3.6	0.6 - 10.9	7.5
Mono-ortho-PCB	0.4	0.1 - 1.5	0.9
dl-PCB	4.0	1.1 - 11.8	8.3
PCDD/PCDF/dl-PCB	8.8	1.8 - 22.0	17.2

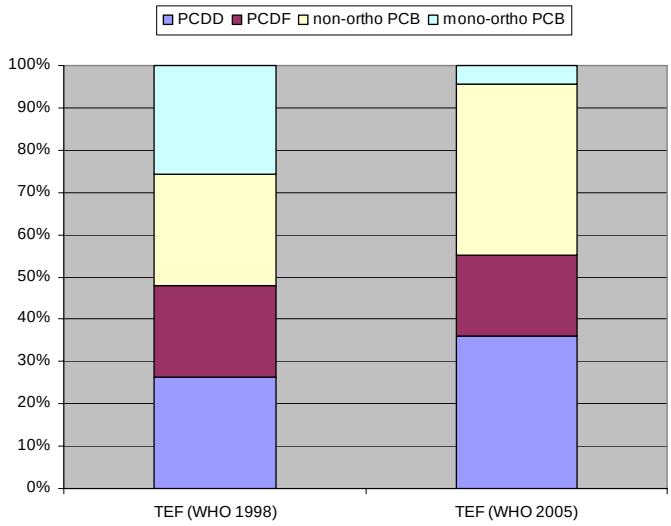

Figure 1: Comparison of the mean shares of PCDD, PCDF, non-ortho- und mono-ortho-PCB to total WHO 1998 TEQ and WHO 2005 TEQ

Daily PCDD/F/dl-PCB intake of infants

Based on our median and 95th-percentile PCDD/F/dl-PCB-concentrations the estimated daily intake for an exclusively breast-fed infant resulted in an average intake of 69 pg WHO-TEQ kg^{-1} bw per day and a high intake of 133 pg WHO-TEQ kg^{-1} bw per day. Thus the recommended level of tolerable daily intake of 1-4 pg TEQ kg^{-1} bw is markedly exceeded during the lactation period [6]. Even though the dioxin background levels in breast milk

strongly decreased in the past 20 years as shown in Figure 2, environmental release of PCDD, PCDF and dl-PCB should be limited to minimize the exposure in the population in order to reduce the elevated intake via breast milk during early phase of the infant's development.

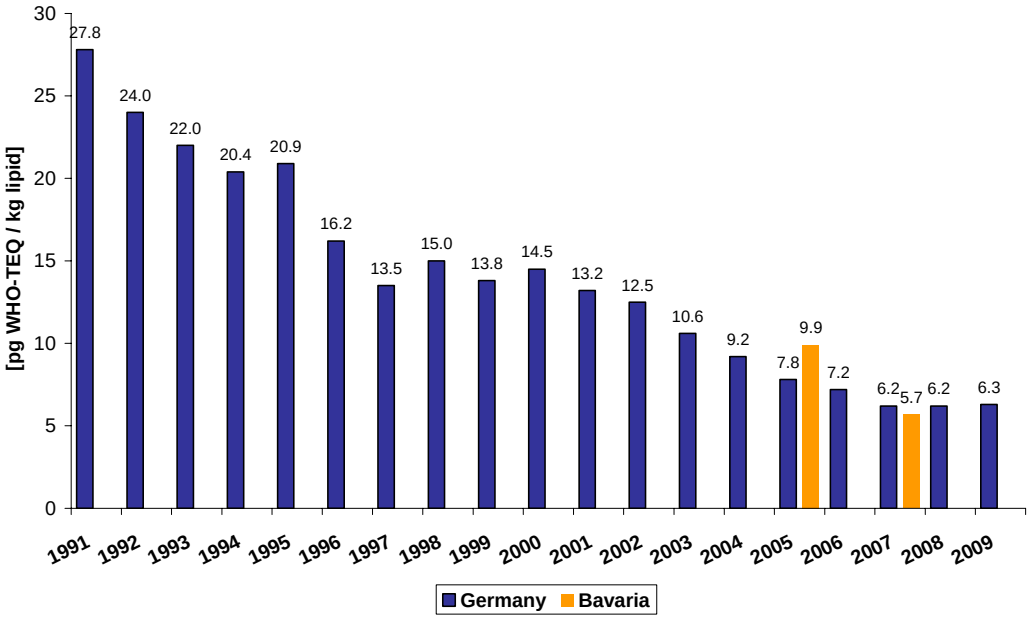

Figure 2: Time trend of the mean levels of PCDD/PCDF in human breast milk analysed in Germany (data source: Federal Environment Agency and Federal Office of Consumer Protection and Food Safety, Germany) and Bavaria.

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References:

1. Fromme H, Gruber L, Seckin E, Raab U, Zimmermann S, Kiranoglu M, Schlummer M, Schwegler U, Smolic S, Völkel W. (2011), Environ Int. 37(4):715-722
2. Fromme H, Mosch C, Morovitz M, Alba-Alejandre I, Boehmer S, Kiranoglu M, Faber F, Hannibal I, Genzel-Boroviczény O, Koletzko B, Völkel W. (2010) Environ Sci Technol. 44(18):7123-9.
3. Raab U, Preiss U, Albrecht M, Shahin N, Parlar H, Fromme H. (2008) Chemosphere 72(1):87-94
4. US-EPA-Methods 1613 rev B and 1668 rev A
5. Bundesgesundheitsbl - Gesundheitsforsch -Gesundheitsschutz (2008), 51:1239–1242
6. WHO, 1998. WHO Experts re-evaluate health risks from dioxins. Press Release WHO/45, 3 June 1998.