CONGENER-SPECIFIC DIFFERENCES IN TEMPORAL TRENDS OF PCDD, PCDF AND PBDE IN MOTHER´S MILK FROM UPPSALA COUNTY, SWEDEN

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Abstract
Temporal trends of PCDD, PCDF and PBDE in mother´s milk from primiparous women were studied during the period 1996 to 2004. The concentrations of ΣPCDD/F TEQ declined with 6.8% (adjusted geometric mean) per year. Among individual PCDD/F congeners, 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PeCDF gave the largest contribution to ΣPCDD/F TEQ (median:85%). 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF had the longest half-lives (12 yr) followed by TCDD (9 yr) and 1,2,3,6,7,8-HxCDD (8 yr). The half-life of ΣPCDD TEQs was shorter than the half-life of ΣPCDF TEQs. As a consequence, the contribution of PCDF to the ΣPCDD/F TEQ is increasing in Swedish mother´s milk. Regression analysis did not show any significant trend in ΣPBDE concentrations during the study period. However, concentrations of BDE-47 and -99 declined slowly, whereas BDE-153 concentrations slowly increased. Thus it is important to determine temporal trends of individual PCDD, PCDF and PBDE congeners in order to better understand the observed decline of ΣPCDD/F TEQ and the lack of trend for ΣPBDE in mother´s milk in Sweden.

Introduction
Concentrations of PCDD, PCDF and PBDE in mother´s milk can be used to determine the body burdens of the compounds among young women in child-bearing age. Earlier studies have shown that PCDD/DF concentrations in mother´s milk in Sweden have decreased during the last decades of the 20th century[1]. In contrast, concentrations of PBDE increased dramatically during the same time period[1]. Between 1996 and 2001, however, the increase in PBDE concentrations appeared to level off[2]. Here we present congener specific temporal trends for PCDD, PCDF and PBDE in mother´s milk from primiparous women from Uppsala County, Sweden, during the period 1996 to 2004.

Materials and Methods
Primiparas from the general population in Uppsala County (median age 29 (range 21-41) years) were recruited annually between 1996 and 2002 and year 2004. Mothers sampled milk during the third week after delivery, using a manual breast pump and/or a passive breast milk sampler. Milk was kept frozen in hexane-washed glass bottles. The newly sampled milk was poured on top of the frozen milk.

The PBDEs were analyzed at the NFA[2] and the PCDD/DFs at the National Institute of Public Health and Environment (RIVM), the Netherlands[3]. Statistical analysis was performed using the Mann-Whitney U test and simple or multiple linear regression analysis.

Results and Discussion
Most of the 154 samples had quantifiable concentrations of PCDD (Table 1). Among PCDDs, 2,3,7,8-TCDD and the 1,2,3,7,8- and 1,2,3,6,7,8-substituted congeners contributed most to the ΣPCDD/F TEQ. Among the PCDFs the 2,3,4,7,8-substituted congener gave a significant contribution to the ΣPCDD/F TEQ. Together these four congeners contributed with 88% (median, min-max=76-93%). Five PBDE congeners were analysed and BDE-47 was present at the highest concentrations followed by BDE-99 and BDE-153 (Table 1).

Regression analysis showed that the concentrations of PCDD/F significantly decreased during the study period after adjustment of the results with the most important co-variates 'age of the mother', 'weight increase during

pregnancy' and 'weight loss after delivery' (Table 2). Among the congeners giving the largest contribution to ΣPCDD/F TEQ, TCDD and 1,2,3,6,7,8-HxCDD had shorter half-lives than 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF. As a result, the contribution of PCDF TEQ to the ΣPCDD/F TEQ increased during the study period, illustrated by the decreased (p=0.0002) PCDD TEQ/PCDF TEQ quotient between 1996-97 (median: 2.11, N=46) and 2004 (median:1.74, N=15).

No significant trend in ΣPBDE concentration was observed (Table 2). A closer look at the temporal trends of the single congeners contributing most to the ΣPBDE concentration showed that concentrations of BDE-47 and BDE-99 significantly decreased during the study period (Table 2). At the same time, the concentrations of BDE-153 increased significantly.

The results show that it is important to determine the temporal trends of individual PCDD, PCDF and PBDE congeners in order to understand the reasons behind observed trends of ΣPCDD/F TEQ and ΣPBDE.

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Table 1. Concentrations of PCDD, PCDF and PBDE in mother´s milk[a].

Compound	N	pg/g lipid	pg TEQ/g lipid	% of ΣPCDD/F TEQ	N<LOQ
TCDD	154	0.9 (0.05-2.8)	0.9 (0.05-2.8)	14 (1-27)	6
1,2,3,7,8-PeCDD	154	2.4 (1.1-6.5)	2.4 (1.1-6.5)	35 (25-46)	0
1,2,3,4,7,8-HxCDD	154	1.2 (0.4-2.6)	0.1 (0.04-0.3)	2 (0.8-3)	3
1,2,3,6,7,8-HxCDD	154	8.1 (3.4-21)	0.8 (0.3-2.1)	12 (7-28)	0
1,2,3,7,8,9-HxCDD	154	1.8 (0.6-9.0)	0.2 (0.06-0.9)	3 (1-10)	1
1,2,3,4,6,7,8-HpCDD	154	14 (4-69)	0.1 (0.04-0.7)	2 (0.7-10)	0
OCDD	153	74 (26-316)	0.02 (0.01-0.09)	0.3 (0.1-0.8)	0
ΣPCDD TEQ	153		4.7 (1.9-12.0)		
TCDF	154	0.4 (0.05-1.2)	0.04 (0.01-0.1)	0.6 (0.1-2)	22
1,2,3,7,8-PeCDF	154	0.2 (0.05-0.8)	0.006 (0.002-0.02)	0.1 (0.03-0.9)	41
2,3,4,7,8-PeCDF	154	5.9 (2.3-20)	1.8 (0.7-6.2)	26 (15-38)	0
1,2,3,4,7,8-HxCDF	154	1.4 (0.6-3.1)	0.1 (0.06-0.3)	2 (0.9-4)	1
1,2,3,6,7,8-HxCDF	154	1.2 (0.5-3.1)	0.1 (0.05-0.3)	2 (0.9-3)	2
1,2,3,7,8,9-HxCDF	154	0.05 (0.03-1.0)	0.01 (0.003-0.1)	0.2 (0.04-2)	150
2,3,4,6,7,8-HxCDF	154	0.6 (0.2-2.3)	0.06 (0.02-0.2)	0.9 (0.3-3)	4
1,2,3,4,6,7,8-HpCDF	154	1.7 (0.7-9.9)	0.02 (0.07-0.1)	0.3 (0.06-1)	112
1,2,3,4,7,8,9-HpCDF	154	0.06 (0.04-1.5)	0.001 (0.004-0.02)	0.02 (0.004-0.2)	27
OCDF	154	0.4 (0.05-2.0)	<0.001	<0.02	0
ΣPCDF TEQ	154		2.1 (0.9-7.0)		
ΣPCDD/F TEQ	153		6.7 (3.0-19.0)		
BDE-47[b]	181	1.5 (0.20-16)			1
BDE-99[b]	181	0.30 (0.060-5.2			12
BDE-100[b]	181	0.28 (0.050-5.1)			12
BDE-153[b]	181	0.58 (0.20-4.6)			35
BDE-154[b]	181	0.06 (0.03-0.36)			150
ΣBDE[b]	181	2.9 (0.91-28)			

[a]median (min-max). Values <LOQ are set to 1/2LOQ. TEQ according to [4].
[b]ng/g lipid, ΣBDE includes BDE-47, -99, -100, -153 and -154, .

Table 2. Temporal trends of PCDD, PCDF and PBDE in mother´s milk.

Compound	% change/year[a]	Half-life (year)
TCDD	-7.2±0.9*	-9
1,2,3,7,8-PeCDD	-5.5±0.8*	-12
1,2,3,6,7,8-HxCDD	-9.0±1.0*	-8
2,3,4,7,8-PeCDF	-5.7±1.1*	-12
PCDD TEQ	-7.0±0.7*	-10
PCDF TEQ	-5.4±0.7*	-13
PCDD/F TEQ	-6.8±0.7*	-10
BDE-47	-3.9±1.7*	-18
BDE-99	-8.0±1.8*	-8
BDE-100	-1.7±1.1	
BDE-153	+4.7±1.1*	+15
ΣBDE	-1.5±1.6	

[a]PCDD/F adjusted for mother´s age, weight increase during pregnancy, weight loss after delivery. BDE congeners was adjusted for age, BMI, weight gain during pregnancy and weight loss after delivery.
*$p \leq 0.05$, N=153-181