Sakrapport till Naturvårdsverkets Miljöövervakning:

Tidstrend 1996-2006: Polyklorerade dibenzo-*p*-dioxiner (PCDD) och dibenzofuraner (PCDF), polyklorerade bifenyler (PCBer), klorerade bekämpningsmedel och bromerade flamskyddsmedel i modersmjölk från förstföderskor i Uppsala.

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	Dioxiner, furaner, polyklorerade bifenyler, klorerade bekämpningsmedel och bromerade flamskyddsmedel i modersmjölk – tidstrend för Uppsala mellan 1996 och 2006.

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SAMMANFATTNING

Under perioden 1996 till 2006 har Livsmedelsverket samlat in modersmjölk från förstföderskor i Uppsala län. Ett av syftena med studierna är att undersöka hur halterna av vissa persistenta organiska miljögifter (POP) förändras med tiden. I denna rapport utvärderas eventuella tidstrender för polyklorerade dibensodioxiner (PCDD), polyklorerade dibensofuraner (PCDF), polyklorerade bifenyler (PCB), hexaklorbensen, beta-hexaklorcyklohexan, *trans*-nonaklor, oxyklordan, samt polybromerade difenyletrar (PBDE).

Resultaten visar att halterna av PCBer, PCDD, PCDF och klorerade pesticider i modersmjölk har minskat med i medeltal 3-10 % per år under tidsperioden 1996 till 2006. Halterna minskar långsammast för vissa PCB kongener (CB 28, CB 105 and CB 169), medan minskningen är snabbast för DDT och β–hexaklorcyklohexan.

Inga generella trender observerades beträffande PBDEer i modersmjölk. Halterna av BDE 47 och BDE 99 har långsamt sjunkit (5 % och 7 % per år), medan halterna av BDE 153 har ökat med i medeltal 4 % per år under tio-års perioden. En långsamt nedåtgående trend av summaPBDE koncentrationer (BDE 47, BDE 99, BDE 100 och BDE 153) observerades. Denna nedåtgående trend är dock osäker eftersom den tid det beräknas ta för halterna att halveras är mer än dubbelt så lång som den tid studien sträcker sig över.

Halterna av CB 52, CB 101, CB 114, CB 157, CB 77, CB 81, α-HCH, γ-HCH, *p,p'*-DDD, *o,p'*-DDE, *o,p'*-DDT, BDE 28, BDE 66, BDE 154, BDE 138, BDE 183 och HBCD var under kvantifieringsgränsen i över 50% av proverna och för dessa föreningar kunde tidstrender inte studeras.

Report to the Swedish Environmental Protection Agency, 2007-10-31

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Polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), chlorinated pesticides and brominated flame retardants in mother´s milk from primiparae women in Uppsala County, Sweden – Levels and trends 1996-2006

INTRODUCTION

Among the Swedish human population, food is the major source of exposure to persistent organic pollutants (POPs), such as dioxins (PCDD/DFs), polychlorinated biphenyls (PCBs), DDT-compounds and polybrominated diphenylethers (PBDEs). These compounds are lipophilic and accumulate in the lipid compartment of the human body. The POP levels in body lipids therefore reflect the long-term exposure of the individual. Exposure estimation is an important part of risk assessment of POPs in food. Due to the relatively high lipid content, mother´s milk is a good human matrix for analysis of POP body burdens at the time of pregnancy and nursing.

In order to estimate the body burdens of POPs among pregnant and nursing women, and to estimate the intake of the compounds by breast-feeding infants, the Swedish National Food Administration (NFA) has made recurrent measurements of concentrations of POPs in mother´s milk. This project also establish if there are temporal trends of POP concentrations in mother´s milk. Temporal trends of POPs between 1996 and 2004 have been reported earlier (Lignell et al. 2006). The temporal trends are now revised with data from 2006.

MATERIAL AND METHODS

Recruitment of primiparas

Mother´s milk was exclusively sampled from primipara women in order to minimise variation. The recruitment in 1996-2004 (N=305) is described in Lignell et al. (2006). In addition to the mothers recruited in 1996-2004, another 50 mothers were recruited among primiparas who delivered at Uppsala University Hospital from January to December 2006. Women who delivered during the first week in every month, and on randomly selected days during this week, were asked to participate in the mother´s milk study. Two to three primiparas were recruited every month. As a result, a total of 355 women were recruited from 1996 to 2006. Mothers who were born in non-Nordic countries (N=10) were excluded before the statistical analysis. After this exclusion, a total of 345 women were included in the data set.

Data on age, weight, lifestyle, medical history, dietary habits, etc of the mothers were obtained from questionnaires (Table 1).

Table 1. Characteristics of the participating mothers.

Variable		**N**	**Mean**	**Median**	**Min**	**Max**
Age of the mothers (yr)		345	28.9	28.8	19.3	41.4
Pre-pregnancy body mass index (BMI, kg/m^2)		339	22.8	22.0	16.2	37.7
Weight gain during pregnancy (% of initial wt/week)		338	0.63	0.60	0.03	1.54
Weight reduction from delivery to sampling (%)[a]		330	9.6	9.4	-1.7	21
		N	**%**			
Education	max 3-4 yr high school	134	39			
	1-3 yr higher education	71	21			
	>3 yr higher education	137	40			
Smoking during pregnancy[b]	Non-smoker	254	74			
	Former smoker	48	14			
	Smoker	41	12			

[a]Weight reduction minus birth weight of the child in % of weight just before delivery.
[b]Women who stopped smoking before pregnancy are considered to be former smokers, and women who stopped smoking during the first or second month of pregnancy are considered to be smokers.

Mother´s milk sampling

The mothers sampled milk at home during the third week after delivery (day 14-21 post partum). Milk was sampled during nursing using a manual mother´s milk pump and/or a passive mother´s milk sampler. The women were instructed to sample milk both at the beginning and at the end of the breast-feeding sessions. The goal was to sample 500 mL from each mother during 7 days of sampling. During the sampling week, the mother´s milk was stored in the home freezer in acetone-washed bottles. Newly sampled milk was poured on top of the frozen milk. At the end of the sampling week, a nurse visited the mother to collect the bottles.

Analysis

The specific compounds (congeners/metabolites) that were analysed in the mother´s milk samples are presented in Table 2.

PCBs (with a few exceptions, see below) and chlorinated pesticides were analysed at the NFA using previously described methods (Atuma and Aune 1999; Aune et al. 1999). Brominated flame retardants (polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD)) were also analysed at the NFA using a method described in Atuma et al. (2000), with a few modifications. All samples were fortified with internal standards prior to extraction to correct for analytical losses and to ensure quality control. A number of control samples were analysed together with the samples to verify the accuracy and precision of the measurements. The laboratory is accredited for analysis of PCBs and chlorinated pesticides in human milk.

Table 2. Summary of compounds that were analysed in the mother´s milk samples.

Compound	**Congeners, metabolites etc.**
PCBs	
PCBs	28, 52, 101, 105, 114, 118, 138, 153, 156, 157, 167, 170, 180
non-*ortho* PCBs	77, 126, 169, 81
Chlorinated pesticides	
Hexachlorobenzene (HCB)	
Hexachlorocyclohexane	A-HCH, β-HCH, γ-HCH
Chlordane	oxychlordane, *trans*-nonachlor
DDT	*p,p'*-DDE, *p,p'*-DDT, *o,p'*-DDT, *p,p'*-DDD, *o,p'*-DDE
Dioxins and furans	
PCDDs	7 congeners
PCDFs	10 congeners
Brominated flame retardants	
PBDE	28, 47, 66, 99, 100, 138, 153, 154, 183
HBCD	

Polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) were analysed at the National Institute of Public Health and the Environment (RIVM), the Netherlands, from 1996 to 2004, using methods described in Glynn et al. (2001). In 2006 the analyses were performed by the Department of Chemistry, Environmental Chemistry, Umeå University. Non-*ortho* PCBs were analysed at RIVM (1996-1999), the NFA (2000-2004), or at Umeå University (2006).

During year 2000 a calibration study was performed, in which 26 samples were analysed at both the NFA and RIVM in order to calibrate the results of the two laboratories (Lignell et al 2004). CB 126 is the non-*ortho* PCB congener that gives by far the largest contribution to the concentrations of toxic equivalents (TEQs). A comparison of the CB 126 results of the two laboratories showed no systematic errors (Wilcoxon signed rank test, P=0.798).

During 2006, a calibration study was performed, comparing the PCDD/F, CB 126 and CB 169 results in 10 samples analyzed both at RIVM and Umeå University (Table 3). Results of non-*ortho* analyses performed at the NFA and Umeå University were also compared.

Table 3. Comparisons of PCDD/F and non-*ortho* PCB analyses of 10 samples of mother´s milk performed by two different laboratories (median, min-max).

	RIVM (pg/g lipid)	Umeå (pg/g lipid)	RIVM/Umeå
2,3,7,8-TCDD	0.77 (0.47-1.3)	0.80 (0.44-1.6)	0.97 (0.57-1.3)
1,2,3,7,8-PeCDD	1.7 (1.1-3.9)	2.0 (1.1-3.7)	1.0 (0.78-1.3)
1,2,3,4,7,8-HxCDD	0.75 (0.54-1.6)	0.70 (0.56-1.4)	0.99 (0.92-1.6)
1,2,3,6,7,8-HxCDD	5.4 (3.3-12)	5.9 (4.0-13)	0.89 (0.73-1.0)
1,2,3,7,8,9-HxCDD	1.1 (0.58-2.8)	1.2 (0.79-3.2)	0.92 (0.70-1.2)
1,2,3,4,6,7,8-HpCDD	8.1 (5.0-28)	8.9 (4.7-25)	0.98 (0.85-1.2)
OCDD	48 (26-119)	48 (22-110)	1.1 (0.94-1.3)
2,3,7,8-TCDF	0.46 (0.1-0.69)	0.52 (0.29-0.75)	0.91 (0.26-1.3)
1,2,3,7,8-PeCDF	0.20 (0.09-0.40)	0.26 (0.18-0.39)	0.73 (0.33-1.5)
2,3,4,7,8-PeCDF	4.7 (2.3-9.8)	5.2 (2.4-10)	0.96 (0.84-1.0)
1,2,3,4,7,8-HxCDF	1.1 (0.69-1.8)	1.3 (0.84-2.0)	0.86 (0.77-1.0)
1,2,3,6,7,8-HxCDF	1.0 (0.58-1.4)	1.3 (0.71-1.8)	0.84 (0.63-1.0)
2,3,4,6,7,8-HxCDF	0.54 (0.20-0.87)	0.72 (0.32-0.95)	0.79 (0.61-0.96)
PCDD TEQ	3.4 (2.2-6.6)	3.6 (2.2-7.3)	0.99 (0.76-1.0)
PCDF TEQ	1.8 (0.89-3.4)	2.0 (0.99-3.6)	0.91 (0.78-1.0)
PCDD/F TEQ	5.1 (3.3-9.8)	5.5 (3.4-11)	0.97 (0.77-1.0)
PCB 126	37 (21-61)[a]	42 (24-62)	0.90 (0.84-1.1)
PCB 169	18 (10-40)[a]	19 (10-36)	1.0 (0.78-1.3)

[a]Analyses performed at the NFA.

The comparison showed that Umeå University in most cases reported somewhat higher median concentrations than RIVM (and the NFA) (Table 3). In the trend analyses the concentrations reported by Umeå University for the 2006 sampling were adjusted with the median RIVM/Umeå and NFA/Umeå quotients of concentrations obtained in the comparison study (Table 3).

Calculations and statistics

Lipid adjusted mother´s milk POP concentrations were used in the statistical analysis since lipid-adjusted concentrations give a better estimate of the body burden than non-adjusted concentrations (see Lignell et al. 2004). In the case of concentrations below the limit of quantification (LOQ), half of LOQ was taken as an estimated value in the calculations.

Mother´s milk levels of CB 52, CB 101, CB 114, CB 157, CB 77, CB 81, α-HCH, γ-HCH, *p,p'*-DDD, *o,p'*-DDE, *o,p'*-DDT, BDE 28, BDE 66, BDE 154, BDE 138, BDE 183 and HBCD were low (>50 % of the samples below LOQ), and these substances were therefore omitted from the statistical analysis. Temporal trend was not established for PCB 170 since this compound only was analysed in samples from the most recent years.

The distributions of the organochlorine analytical results closely followed a log-normal distribution, therefore all statistical analyses were performed on log transformed data.

Statistical analysis was performed in MINITAB® for Windows 14. Multiple linear regression was used to analyse associations between POP concentrations and sampling year. Independent variables (life-style factors) that have been shown to influence POP levels in mother´s milk (Lignell et al. 2004) were included as explanatory variables in the model. The variables considered were age of the mother (years), pre-pregnancy BMI (Body Mass Index, kg/m^2) and body weight change during pregnancy as well as after delivery (Table 1). In the multiple regressions, observations with a standard residual ≥3 were excluded due to their large influence on the results. As a consequence of the logaritmic transformation, the associations between sampling year and POP concentrations are presented as percent change of concentrations per year, and not as change in absolute levels.

Table 4. Concentrations of persistent halogenated organic pollutants in mother´s milk 1996-2006.

Compound	**N**	**Mean**	**Median**	**Min**[a]	**Max**
PCBs (ng/g lipid)					
PCB 28	325	2.8	1.8	0.25	31
PCB 105	325	1.3	1.0	0.15	15
PCB 118	325	11	9.5	2.9	64
PCB 138	325	29	26	7.8	94
PCB 153	325	58	52	12	186
PCB 156	325	4.5	3.9	0.45	24
PCB 167	325	1.3	1.2	0.18	5.7
PCB 180	325	28	25	5.0	84
mono-*ortho* TEQ 98 (pg/g lipid)[b]	325	3.5	3.1	0.56	18
mono-*ortho* TEQ 05 (pg/g lipid)[c]	325	0.55	0.48	0.12	2.7
Non-*ortho* PCBs (pg/g lipid)					
PCB 126	220	43	39	7.5	125
PCB 169	220	22	20	6.2	65
non-*ortho* TEQ 1998[d]	220	4.6	4.1	0.97	13
non-*ortho* TEQ 2005[e]	220	5.0	4.6	1.3	14
PCDD/F (pg/g lipid)					
TCDD	184	0.94	0.86	0.05	2,8
1,2,3,7,8-PeCDD	184	2.5	2.3	0.66	6.5
1,2,3,6,7,8-HxCDD	184	8.2	7.4	1.9	21
2,3,4,7,8-PeCDD	184	6.1	5.5	1.9	21
PCDD TEQ 1998[f]	184	4.7	4.3	1.3	12
PCDD TEQ 2005[g]	184	4.7	4.3	1.3	12
PCDF TEQ 1998[h]	184	3.5	3.1	1.1	12
PCDF TEQ 2005[i]	184	2.2	2.0	0.70	7.0
PCDD/F TEQ 1998	184	8.1	7.4	2.6	23
PCDD/F TEQ 2005	184	6.9	6.4	2.2	19
Total TEQ 1998	183	16	15	5.2	39
Total TEQ 2006	183	13	12	3.9	31
Chlorinated pesticides					
HCB	325	14	13	3.9	28
β-HCH	325	12	10	2.7	88
Oxychlordane	325	4.0	3.6	0.78	11
trans-nonachlor	325	7.0	6.4	1.1	27
p,p'-DDT	325	8.2	5.6	0.84	240
p,p'-DDE	325	108	89	19	649
Brominated flame retardants					
BDE-47	211	2.0	1.5	0.20	16
BDE-99	211	0.45	0.29	0.06	5.2
BDE-100	211	0.37	0.27	0.05	5.1
BDE-153	211	0.68	0.60	0.20	4.6
sumPBDE[j]	211	3.8	2.9	0.91	28

[a]Concentrations below LOQ were set to 1/2 LOQ.
[b]Including CB 105, 118, 156, and 167 TEQs based on 1998 WHO TEFs (Van den Berg et al. 1998).
[c]Including CB 105, 118, 156, and 167 TEQs based on 2005 WHO TEFs (Van den Berg et al. 2006).
[d]Including CB 126 and 169 TEQs based on 1998 WHO TEFs (Van den Berg et al. 1998).
[e]Including CB 126 and 169 TEQs based on 2005 WHO TEFs (Van den Berg et al. 2005).
[f]TEQ concentrations of 7 PCDD congeners based on 1998 WHO TEFs (Van den Berg et al. 1998).
[g]TEQ concentrations of 7 PCDD congeners based on 2005 WHO TEFs (Van den Berg et al. 2006).
[h]TEQ concentrations of 10 PCDF congeners based on 1998 WHO TEFs (Van den Berg et al. 1998).
[i]TEQ concentrations of 10 PCDF congeners based on 2005 WHO TEFs (Van den Berg et al. 2006).
[j]Including BDE-47, -99, -100, -153 and 154.

RESULTS

POP concentrations in mother´s milk

Among the PCB congeners (excluding the non-*ortho* congeners), the di-*ortho* congener PCB 153 showed the highest median concentration followed by CB 138 and CB 180 (Table 4). Among the mono-*ortho* congeners, CB 118 showed the highest median concentration. PCB 28 had the largest variation in levels (more than 100-fold). The mono-*ortho* TEQ concentrations were calculated using both 1998 and 2005 TEFs (Van den Berg et al. 1998; 2006). The median TEQ concentrations using the 2006 TEFs were 6-fold lower than the median TEQ concentration obtained using the 1998 TEFs (Table 4).

Among the non-*ortho* PCBs, PCB 126 showed the highest median concentration. The TEQ concentrations using the 2005 TEFs were 12 % higher than when the 1998 TEFs were used (Table 4).

p,p'-DDE was the compound with the overall highest median concentration. Median concentrations of the other chlorinated pesticides were ≥8-fold lower (Table 4).

Among the PCDD/Fs, 2,3,7,8-tetrachloro dibenzo-*p*-dioxin (TCDD), 1,2,3,7,8-PeCDD, 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PeCDD contributed most to the PCDD/F TEQ concentrations (Table 4). Similarly to mono-*ortho* PCBs, the PCDD/F TEQ concentrations were lower when using 2005 TEFs than when 1998 TEFs were used.

Among the PBDEs, BDE-47 showed the highest median concentration.

Table 5. Percent change in concentrations of POPs per year in mother´s milk from primiparae women living in Uppsala County 1996-2006. Adjusted for age, pre-pregnancy BMI, weight gain during pregnancy and weight loss after delivery.

Compound	**Change/yr (%)**[a]		**R^{2b}**	**"half-time"**[c]	**P**
	Mean	SE	(%)	Years	
PCB 28	-4.0	1.4	8	17	0.004
PCB 105	-4.3	1.2	26	16	<0.001
PCB 118	-8.6	0.6	56	8	<0.001
PCB 138	-6.9	0.5	55	10	<0.001
PCB 153	-8.0	0.5	68	8	<0.001
PCB 156	-5.8	0.5	62	12	<0.001
PCB 167	-5.4	0.9	42	12	<0.001
PCB 180	-7.3	0.5	76	9	<0.001
mono-*ortho* TEQ 1998[d]	-6.2	0.6	66	11	<0.001
mono-*ortho* TEQ 2005[e]	-7.4	0.5	60	9	<0.001
PCB 126	-7.7	0.7	49	9	<0.001
PCB 169	-3.4	0.7	54	20	<0.001
non-*ortho* TEQ 1998[f]	-7.7	0.6	49	9	<0.001
non-*ortho* TEQ 2005[g]	-7.3	0.6	52	9	<0.001
PCDD TEQ 1998[h]	-6.9	0.5	66	10	<0.001
PCDD TEQ 2005[i]	-6.9	0.5	66	10	<0.001
PCDF TEQ 1998[j]	-5.7	0.6	52	12	<0.001
PCDF TEQ 2005[k]	-5.7	0.6	52	12	<0.001
PCDD/DF TEQ 1998	-5.4	0.4	64	12	<0.001
PCDD/DF TEQ 2005	-6.4	0.5	66	10	<0.001
Total TEQ 1998	-6.7	0.5	66	10	<0.001
Total TEQ 2005	-7.0	0.5	64	10	<0.001
HCB	-8.2	0.4	52	8	<0.001
β-HCH	-10	0.5	63	6	<0.001
Oxychlordane	-6.9	0.5	58	10	<0.001
trans-nonachlor	-6.3	0.6	53	11	<0.001
p,p'-DDT	-9.4	0.8	32	7	<0.001
p,p'-DDE	-8.5	0.8	41	8	<0.001
Brominated flame retardants					
BDE-47	-5.3	1.4	11	13	<0.001
BDE-99	-7.4	1.4	15	9	<0.001
BDE-100	-0.54	1.5	6		0.703
BDE-153	+4.3	0.7	34		<0.001
sumPBDE[l]	-2.9	1.0	8	24	0.009

[a]Percent change (decrease (-) or increase (+)) of the concentrations per year
[b]Coefficient of determination for the regression model
[c]The estimated time it takes for the concentrations to be halved.
[d]Including CB 105, 118, 156, and 167 TEQs based on 1998 WHO TEFs (Van den Berg et al. 1998).
[e]Including CB 105, 118, 156, and 167 TEQs based on 2005 WHO TEFs (Van den Berg et al. 2006).
[f]Including CB 126 and 169 TEQs based on 1998 WHO TEFs (Van den Berg et al. 1998).
[g]Including CB 126 and 169 TEQs based on 2005 WHO TEFs (Van den Berg et al. 2006).
[h]TEQ concentrations of 7 PCDD congeners based on 1998 WHO TEFs (Van den Berg et al. 1998).
[i]TEQ concentrations of 7 PCDD congeners based on 2005 WHO TEFs (Van den Berg et al. 2006).
[j]TEQ concentrations of 10 PCDF congeners based on 1998 WHO TEFs (Van den Berg et al. 1998).
[k]TEQ concentrations of 10 PCDF congeners based on 2005 WHO TEFs (Van den Berg et al. 2006).
[l]Including BDE-47, -99, -100, -153 and 154.

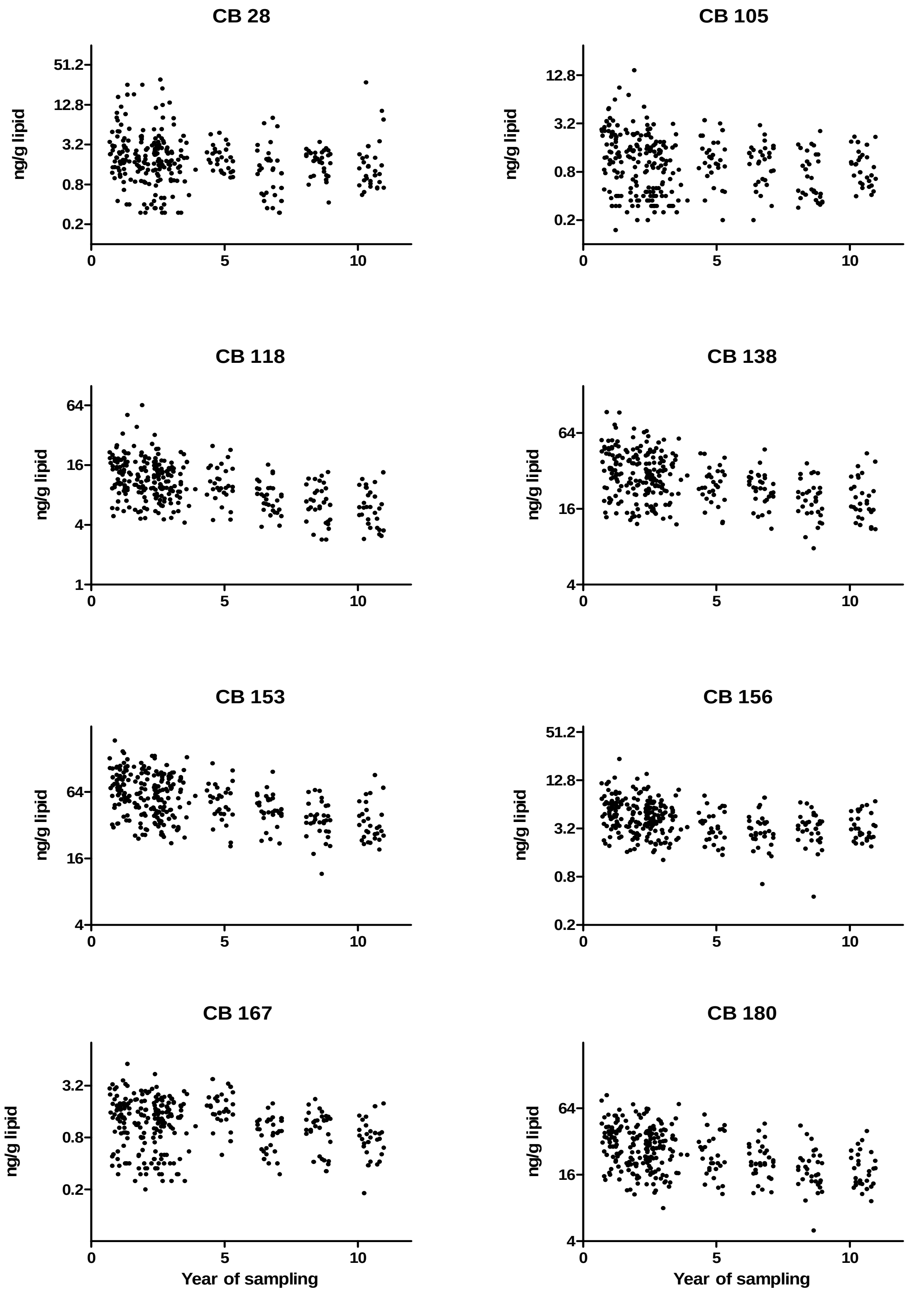

Figure 1. PCB concentrations in mother´s milk from primiparous mothers living in Uppsala County, Sweden (N=325), starting Jan 1 1996 (year 0) and ending December 31 2006. Note that the plots are based on raw data that have not been adjusted for life-style factors and that the y-axis has a log scale.

Temporal trends

PCBs

Multiple linear regression showed that the adjusted mean decrease in mono- and di-*ortho* PCB concentrations varied between 4.0 and 7.3 % per year (Table 5, Figure 1). The fastest decline was shown for PCB 118, 153 and 180, while PCB 28 and 167 declined more slowly during the study period. The regression model explained 7.2-76 % of the variation in PCB-levels, with the lowest degree of explanation for PCB 28 and the highest for PCB 180.

The adjusted mean decrease in concentrations of PCB 126 and PCB 169 (non-*ortho* PCBs) was 7.7 and 3.4 % per year respectively (Table 5, Figure 2). The regression model explained 49-52 % of the variation. There were only small differences in the temporal trend of non-*ortho* PCB TEQs based on the two different WHO TEF systems (Table 5).

PCDD, PCDF and total TEQ

The concentrations of 2,3,7,8-tetrachloro dibenzo-*p*-dioxin (TCDD), 1,2,3,7,8-PeCDD, 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PeCDD declined significantly during the study period (Figure 2). Multiple linear regression showed that the concentrations of PCDD, PCDF, PCDD/DF TEQ and total TEQs declined on average 5.4 % to 7.0 % per year (Table 5, Figure 3). The regression model explained over 50 % of the variation in concentrations (Table 5). Only minor differences in temporal trends were observed when the two different TEF systems were used (Table 5).

Chlorinated pesticides

Multiple linear regression showed that the adjusted mean decrease in levels of chlorinated pesticides varied between 6.3 and 10 % per year. β-HCH and *p,p'*-DDT showed the fastest decline and oxychlordane and *trans*-nonachlor the slowest (Table 5, Figure 4). The regression model explained 32-63 % of the variation in pesticide concentration, with the lowest degree of explanation for *p,p'*-DDT and *p,p'*-DDE and the highest for β-HCH.

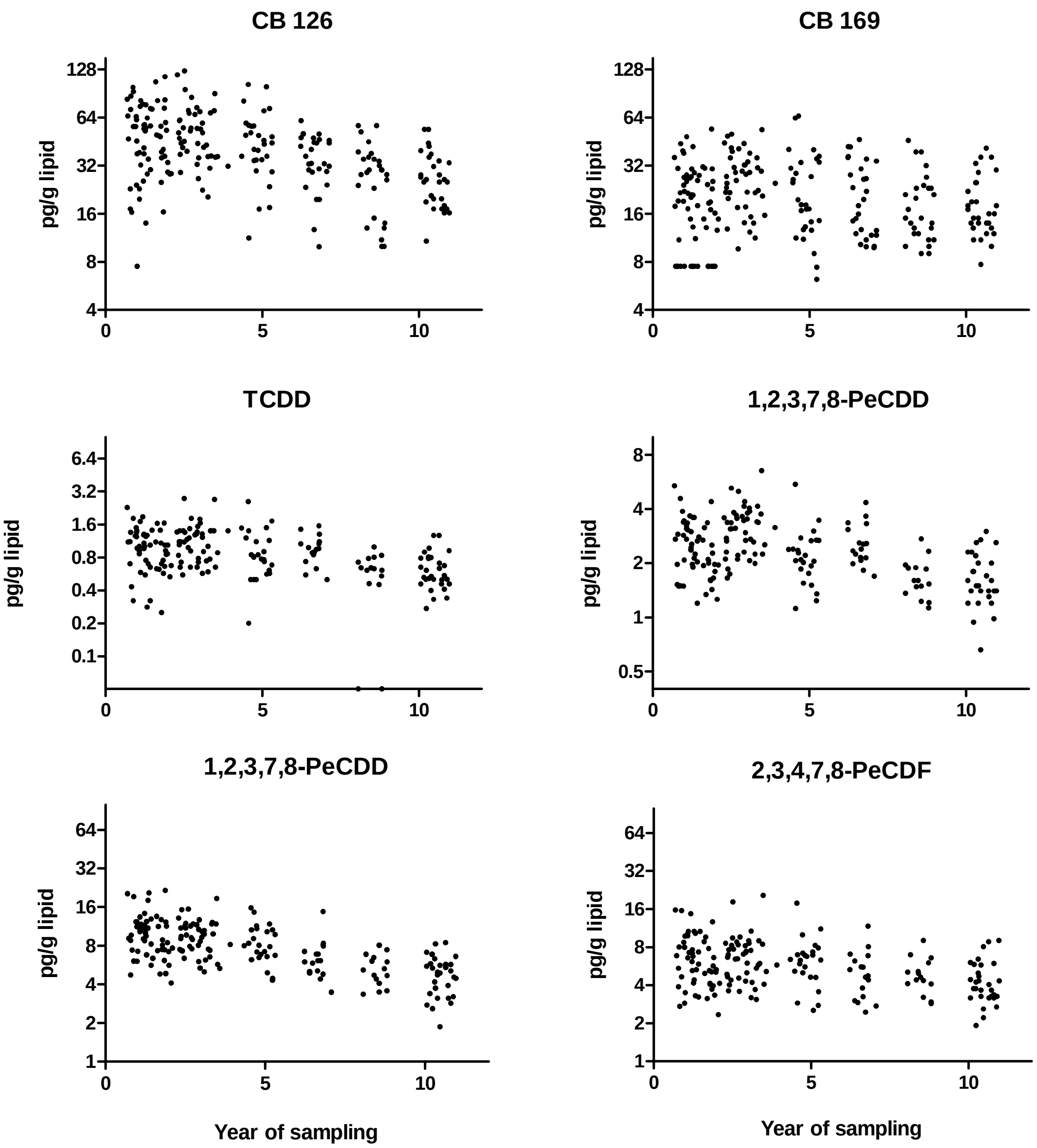

Figure 2. Concentrations of PCB 126, PCB 169, and selected PCDD/Fs (ln transformed data) in mother´s milk from primiparous mothers living in Uppsala County, Sweden (N=184-220, see Table 3), starting Jan 1 1996 (year 0) and ending December 31 2006. Note that the plots are based on raw data that have not been adjusted for life-style factors and that the y-axis has a log scale.

Brominated flame retardants

Regarding the PBDE congeners, there were no general trends during the time period (Table 5, Figure 5). Multiple linear regression showed that the concentrations of BDE-47 and BDE-99 decreased significantly, while the concentrations of BDE-153 increased. No significant trends were shown for BDE-100. The concentrations of sumPBDE showed a slow but statistically significant decline (Table 5, Figure 5). In contrast to the other POPs, there were only few significant associations between PBDE-levels and the explanatory variables included in the regression model. The regression model only explained 0.4-10 % of the variation in levels of BDE-47, BDE-99, BDE-100 and sumPBDE. However, BDE-153 deviated from the other PBDEs in this aspect, and the BDE-153 levels were significant associated to age, BMI, and weight gain during pregnancy (the regression model explained 32 % of the variation in BDE-153 levels).

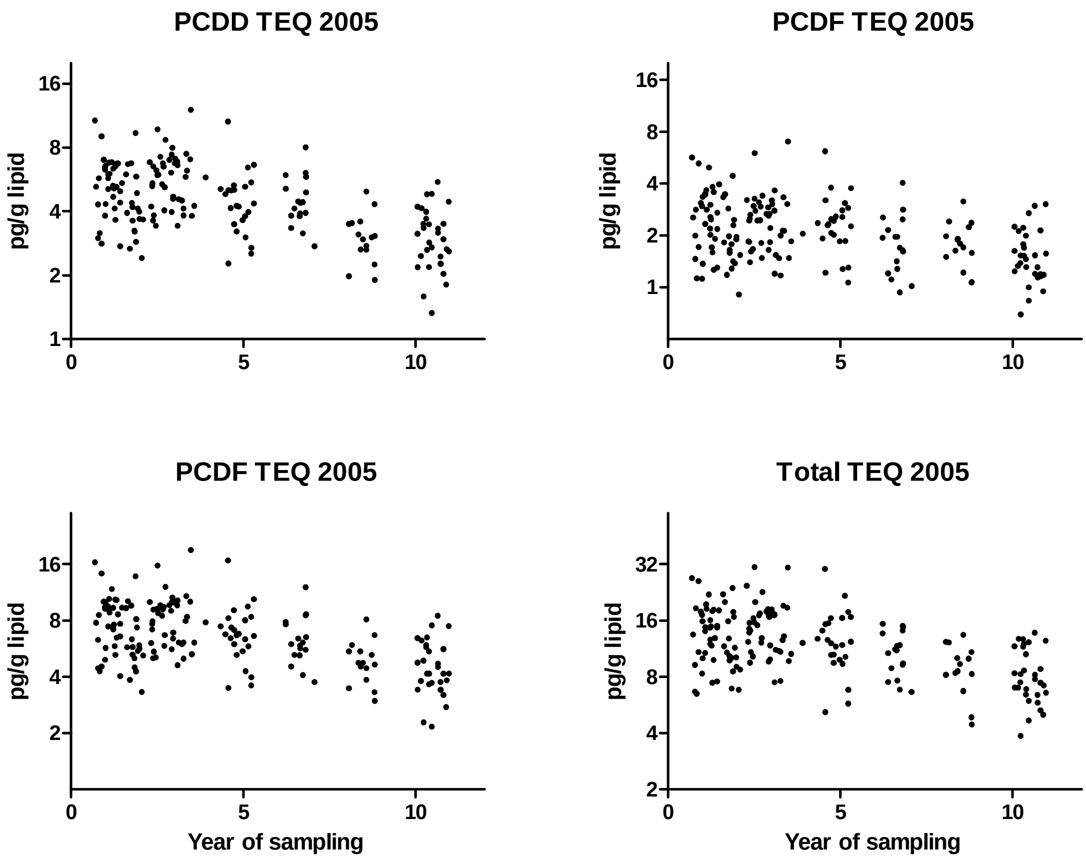

Figure 3. PCDD/F TEQ and total TEQ concentrations (ln transformed data) in mother´s milk from primiparous mothers living in Uppsala County, Sweden (N=295), starting Jan 1 1996 (year 0) and ending December 31 2006. Note that the plots are based on raw data that have not been adjusted for life-style factors and that the y-axis has a log scale.

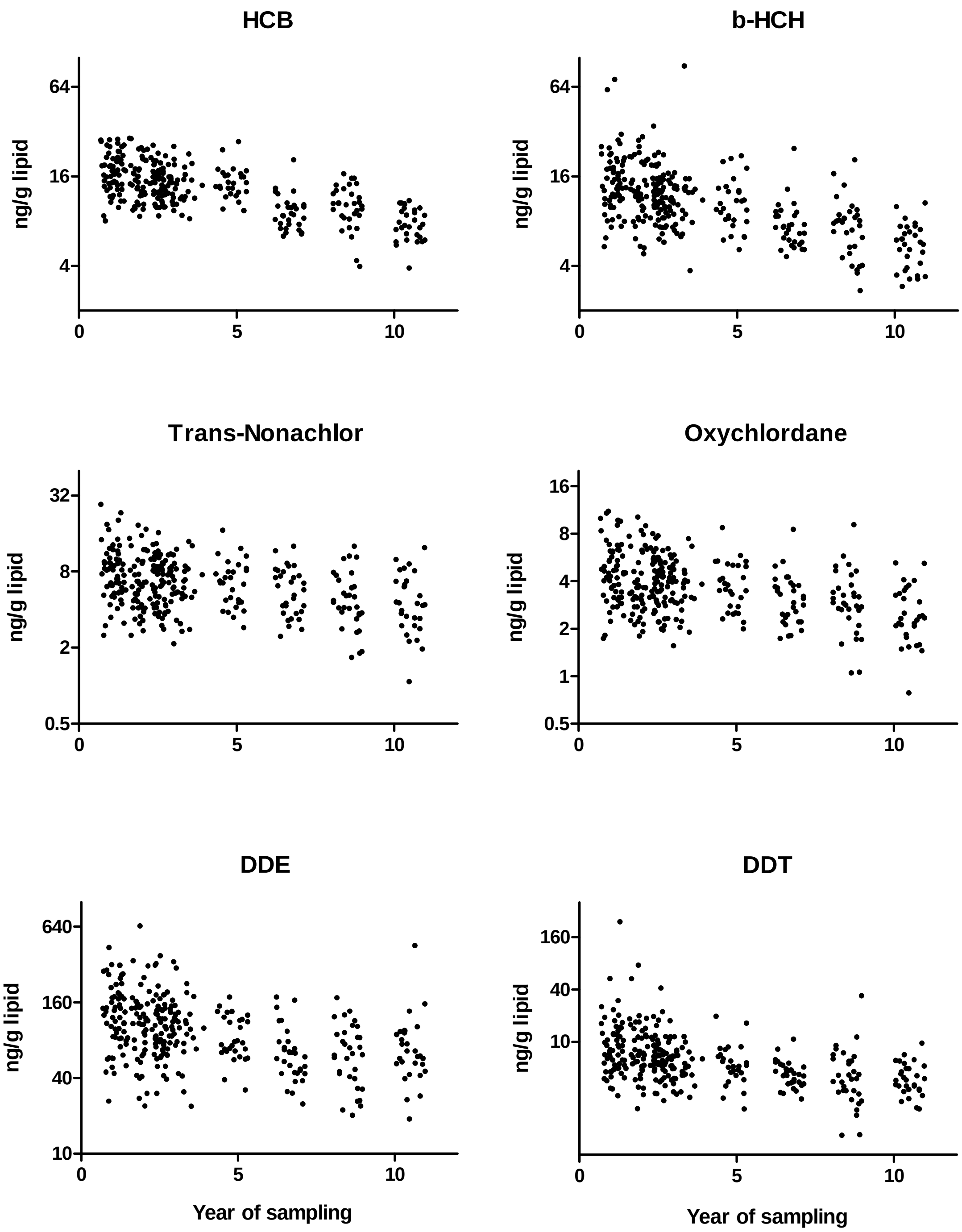

Figure 4. Concentrations of chlorinated pesticides in mother´s milk from primiparous mothers living in Uppsala County, Sweden (N=325), starting Jan 1 1996 (year 0) and ending December 31 2006. Note that the plots are based on raw data that have not been adjusted for life-style factors and that the y-axis has a log scale.

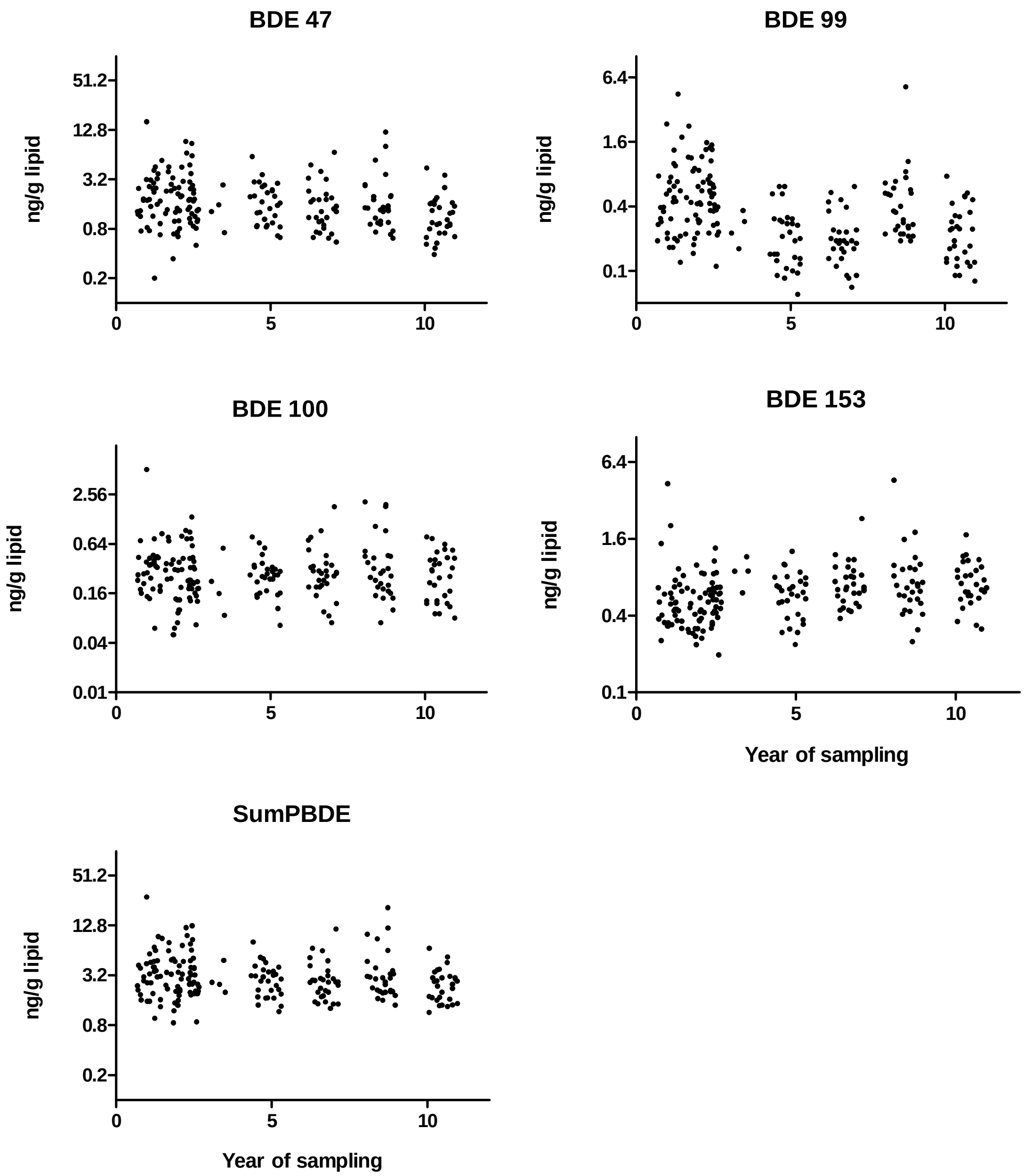

Figure 5. PBDE concentrations in mother´s milk from primiparous mothers living in Uppsala County, Sweden (N=211), starting Jan 1 1996 (year 0) and ending December 31 2006. Note that the plots are based on raw data that have not been adjusted for life-style factors and that the y-axis has a log scale.

DISCUSSION

Our results show that the concentrations of most of the studied POPs (PCBs, dioxins/furans, chlorinated pesticides) in mother´s milk from primiparous women living in Uppsala County, Sweden, have decreased from 1996 to 2006. This is probably a consequence of reduced levels of many POPs in the environment (and in foods) since the 1970s, i.e. a 30-year-old woman who had her first child in 1996 had been exposed to higher life-time cumulative levels of POPs before pregnancy than a 30-year-old woman who had her first child in 2006.

The temporal trends of PBDEs between 1996 and 2006 showed that the concentrations of BDE 47 and 99 decreased significantly and the concentrations of BDE 153 increased. As a consequence a slow but statistically significant decline in sum PBDE concentrations was observed. The uncertainty of this result is however large since the estimated years needed for the adjusted mean concentration to be halved in mothers milk from primiparous women were much longer than the duration of the study period. Similarly to our results, a study by Fängström et al. (2005), based on PBDEs analyses of pooled milk samples from mothers in the Stockholm region in 1980-2004, indicate that that the concentrations of lower brominated PBDE congeners (e.g. BDE-47, BDE-99) have decreased from the middle of the 1990's while the concentrations of BDE-153 have increased.

As mentioned earlier, the levels of many POPs in the environment have decreased since the 1970s. Consequently, the older women in our study have been exposed to higher levels than the younger women. It is however important to consider other life-style/medical factors, such as pre-pregnancy BMI, weight increase during pregnancy and weight loss after delivery, when temporal trends of most POPs in mother´s milk are studied. However, in most cases more than 50% of the variation in POP concentrations was not explained by the regression model used. This shows that there still are unknown factors causing variation in POP concentrations in mother´s milk.

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