

# TEMPORAL TRENDS OF PBDEs IN MOTHER'S MILK FROM UPPSALA COUNTY, SWEDEN

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## Introduction

In order to estimate the body burdens of persistent organic pollutants (POPs) among pregnant and nursing women, and to estimate the intake of these compounds by breast-feeding infants, the Swedish National Food Administration (NFA) has made recurrent measurements of concentrations of POPs in mother's milk. Earlier studies have shown that concentrations of many POPs, e.g. DDE, PCBs, PCDDs and PCDFs, in mother's milk in Sweden have decreased during the last decades of the 20<sup>th</sup> century<sup>1</sup>. In contrast, levels of polybrominated diphenyl ethers (PBDEs), ubiquitously used as flame retardants, increased dramatically during the same period<sup>1</sup>. Between 1996 and 2001, however, the increase in PBDE concentrations appeared to level off<sup>2</sup>. Here we present levels and temporal trends of PBDEs and hexabromocyclododecane (HBCD) in mother's milk from primiparous women from Uppsala County, Sweden during the period 1996 to 2006.

## Materials and Methods

Primiparas (N=325) from the general population in Uppsala County were recruited annually between 1996 and 2002 and also during 2004 and 2006. The median age of the participating women was 29 years (range 19-41). Participating mothers sampled milk at home during the third week after delivery, using a manual milk pump and/or a passive breast milk sampler. The milk samples were kept frozen in acetone-washed bottles during the sampling week. Newly sampled milk was poured on top of the frozen milk.

PBDEs (BDE-28, -47, -66, -99, -100, -138, -153, -154 and -183) and HBCD were analysed at the NFA using a method described by Atuma et al.<sup>3</sup>, 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.

Lipid adjusted mother's milk concentrations of PBDEs were used in the statistical analysis. Multiple linear regression analysis was used to analyse associations between concentrations of PBDEs in mother's milk and sampling year. Independent variables that have been shown to influence body burdens of other POPs (e.g. PCB, DDT)<sup>4, 5, 6</sup> were included as explanatory variables in the model. Variables considered were 'age of the mother' (years), 'pre-pregnancy BMI' (Body Mass Index, kg/m<sup>2</sup>), 'weight increase during pregnancy' (%/week) and 'weight loss after delivery' (%). In the case of concentrations below the limit of quantification (LOQ), half of LOQ was taken as an estimated value in the regression analyses. The distributions of the PBDE analytical results closely followed a log-normal distribution. Therefore all regression analyses were performed on log transformed data. As a consequence of the logarithmic transformation, the associations between sampling year and PBDE concentrations are presented as percent change per year, and not as change in absolute levels.

## Results and Discussion

BDE-47 showed the highest median concentration in mother's milk, followed by BDE-153, BDE-99 and BDE-100 (Table 1). The levels of BDE-28, BDE-66, BDE-138, BDE-154, BDE-183 and HBCD were below LOQ in most samples, and temporal trends were not established for these congeners. There was no similar trend for all PBDE congeners during the time period (Table 2, Figure 1). The concentrations of BDE-47 and BDE-99 decreased significantly, while the concentrations of BDE-153 increased. No significant trend was shown for BDE-100. The concentrations of sumPBDE (sum of BDE-47, -99, -100, -153 and 154) showed a slow but statistically significant decline. The uncertainty of this result is however large since the estimated time needed for the adjusted mean concentration to be halved in mother's milk were much longer than the duration of the study period.

The only significant associations between the concentrations of BDE-47, BDE-99, BDE-100 and sumPBDE in mother's milk and the explanatory variables included in the regression model were negative associations between weight gain during pregnancy and the levels of BDE-99 and sumPBDE. The whole regression model only explained 2-13 % of the variation in concentrations (Table 2). BDE-153 deviated from the other PBDEs in this aspect, and the levels of BDE-153 were significant associated with age (positive), BMI (negative), weight gain during pregnancy (negative) and weight loss after delivery (positive). The regression model explained 31 % of the variation in BDE-153 levels (Table 2). BDE-153 is in this aspect more like other POPs, such as PCBs and chlorinated pesticides (e.g. DDE), whose concentrations in serum similarly have been shown to be associated to age, pre-pregnancy BMI and weight gain during pregnancy<sup>4</sup>.

Similarly to our results, a Swedish study based on PBDE concentrations in pooled milk samples from women in the Stockholm region<sup>7</sup> indicate that the concentrations of lower brominated PBDE congeners (e.g. BDE-47, BDE-99, BDE-100) have decreased from the middle of the 1990's while the concentrations of BDE-153 have increased (Figure 2). The explanation to these findings may be a reduced use of lower brominated PBDEs and/or a higher persistence of BDE-153 compared to BDE-47. By this time, it is not possible to establish any solid temporal trend for the PBDE congeners studied in mother's milk from Uppsala. This makes it important to continue to sample mother's milk for PBDE analysis in the future.

### Acknowledgements

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**Table 1. Concentrations (ng/g lipid) of PBDEs and HBCD in mother's milk from primipara women in Uppsala County, Sweden, collected between 1996 and 2006.**

Compound	N	Mean <sup>a</sup>	Median	SD <sup>a</sup>	Min	Max	% <LOQ <sup>b</sup>
BDE-28	196	0.12	0.11	0.07	<0.06	0.60	76
BDE-47	276	1.9	1.5	1.7	<0.40	16	1
BDE-66	196	0.09	0.09	0.05	<0.06	0.32	98
BDE-99	276	0.45	0.32	0.51	<0.12	5.2	16
BDE-100	276	0.36	0.29	0.40	<0.10	5.1	9
BDE-138	196	0.09	0.09	0.06	<0.06	0.55	100
BDE-153	276	0.64	0.57	0.45	<0.23	4.6	2
BDE-154	276	0.09	0.07	0.04	<0.06	0.94	88
BDE-183	167	0.10	0.10	0.05	<0.06	0.32	98
sumPBDE <sup>c</sup>	276	3.5	2.9	2.7	0.91	28	-
HBCD	177	0.39	0.25	0.70	<0.20	7.8	66

<sup>a</sup>levels below the limit of quantification (LOQ) were set to ½ LOQ in the calculations of mean and SD

<sup>b</sup>% of the samples with concentrations below LOQ

<sup>c</sup>sum of BDE-47, -99, -100, -153 and 154

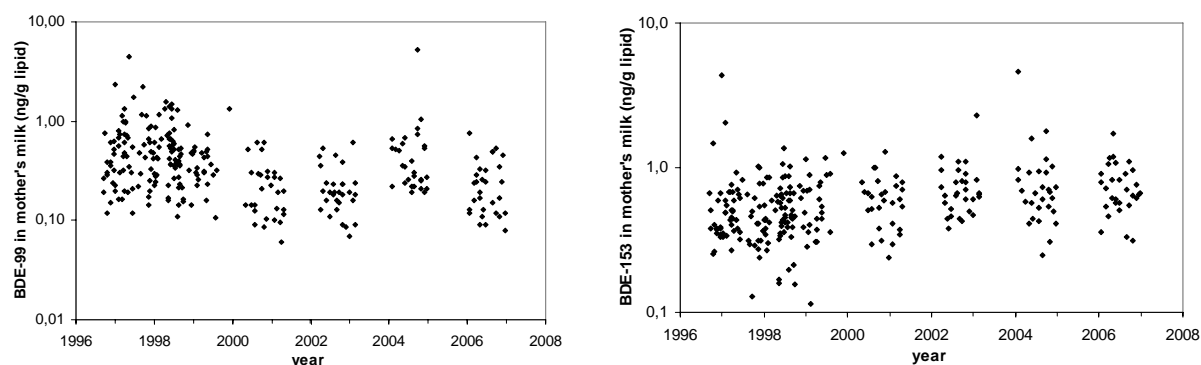
**Table 2. Percent change in concentrations of PBDEs per year in mother's milk from primiparae women living in Uppsala County, Sweden 1996-2006. Temporal trends were adjusted for age, pre-pregnancy BMI, weight gain during pregnancy and weight loss after delivery.**

Compound	Change per year (%)		R <sup>2</sup> (%) <sup>a</sup>	"half- time" <sup>b</sup> (years)	P
	Mean	SE			
BDE-47	-4.3	1.1	7	16	<0.001
BDE-99	-7.5	1.2	13	9	<0.001
BDE-100	-1.2	1.2	2	-	0.324
BDE-153	+4.9	0.7	31	-	<0.001
sumPBDE <sup>c</sup>	-2.0	0.9	3	34	0.03

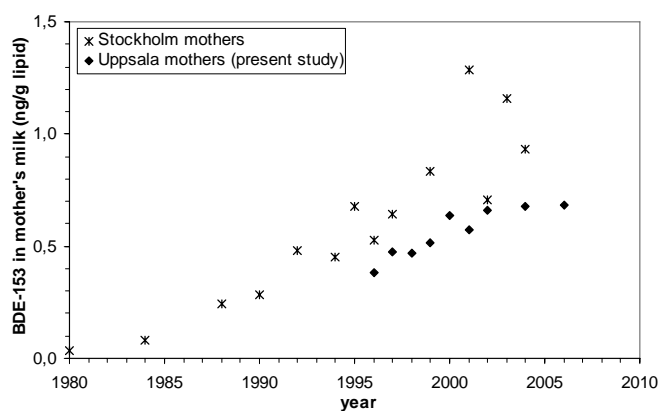
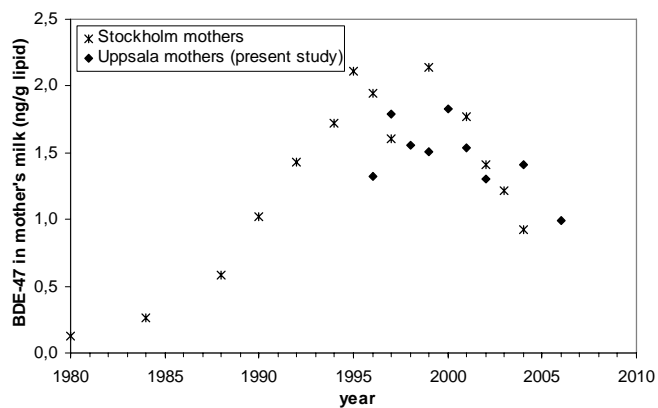
<sup>a</sup>coefficient of determination for the whole regression model

<sup>b</sup>the estimated time it takes for the concentrations to be halved in the population

<sup>c</sup>including BDE-47, -99, -100, -153 and 154



**Figure 1. Concentrations of BDE-99 and BDE-153 in mother's milk from primipara mothers living in Uppsala County, Sweden (1996-2006, N=276). Each point corresponds to the PBDE level in a milk sample from an individual woman. 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 2.** BDE-47 and BDE-153 concentrations in human milk samples in the present study (yearly median concentrations) and in a previous study on pooled milk samples from mothers in the Stockholm region<sup>7</sup>.