

POLYBROMINATED FLAMES RETARDANTS

TIME TREND OF POLYBROMINATED DIPHENYL ETHER (PBDE) LEVELS IN BREAST MILK FROM UPPSALA, SWEDEN, 1996-2001

Per Ola Darnerud¹, Marie Aune¹, Samuel Atuma¹, Wulf Becker¹, Rickard Bjerselius², Sven Cnattingius² and Anders Glynn²

¹Swedish National Food Administration, Departments of Research & Development and Nutrition, P.O. Box 622, SE-751 26 Uppsala, Sweden

²Department of Medical Nutrition, Karolinska Institutet, SE-171 77 Stockholm, Sweden

Introduction

Recently, the occurrence of brominated flame-retardants (BFRs) in the environment and in human samples has been revealed^{1,2}. One group of BFRs that has lately been studied rather extensively is the polybrominated diphenyl ethers (PBDEs). These compounds are used as flame retardants in various electronics, plastics and textiles, are spread in the technosphere and occur in environmental samples³. In a Swedish time trend study the breast milk levels of PBDEs were shown to increase markedly between 1972 and 1997, while more recent samples result in somewhat lower levels^{4,5}. In addition, studies on individual milk samples showed that the PBDE levels in milk are rather widely distributed and that neither maternal age nor computer usage could explain the milk PBDE levels⁶. However, one group of occupationally exposed persons that could increase their body burdens of PBDEs due to their work is the personnel at electronics-dismantling plants, as monitored by analyses of blood levels⁷. Compared to European and Japanese data, markedly elevated levels of PBDEs have been found in breast milk from the U.S.A.⁸. However, we do not know if this pooled sample is representative for American breast milk levels in general.

In 1998, we presented preliminary PBDE results from 39 Swedish primiparous women from Uppsala County⁶. The mean level in that study was similar to the most recent pooled sample concurrently presented by Meironyté and co-workers⁴. By adding data from additional and new samples to the existing PBDE data pool we have now extended our study to include samples from totally 124 subjects, sampled 1996-2001.

Materials and methods

The women recruited 1996-1999 were participating as controls in a case-control study on risk factors for early miscarriages. Of the 376 primiparae mothers (i.e. having their first child) that were asked to participate 180 agreed to donate breast milk. Of the 180 samples, 93 were randomly taken out for PBDE analyses. In addition to these women, additionally 31 mothers from Uppsala County, sampled 2000-2001, were recruited in order to improve the basis for time trend observations. The age of the participating women ranged from 20 to 35 years old. The mothers sampled their milk during the third week after delivery, and up to 500 ml was obtained during these seven days. The milk was kept frozen until analysis.

The PBDE analyses were carried out at the Swedish NFA, Uppsala⁹. In short, the milk was thawed and 35 g was taken for extraction (2 x n-hexane/acetone 1:1). After addition of ethanol, the solvent was evaporated. After redissolving the sample in n-hexane, fractionation on silica gel and fortification with an internal standard (BDE 85), the sample was run on a GC with dual capillary columns and dual ECD, and the five main PBDE congeners (BDE 47, 99, 100, 153 and 154) were quantified.

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Table 1. PBDE levels (ng/g fat wt.) in breast milk from primiparae women in Uppsala County, Sweden, 1996-2001 (n=124)

	<i>BDE 47</i>	<i>BDE 99</i>	<i>BDE 100</i>	<i>BDE 153</i>	<i>BDE 154</i>	<i>sumPBDE</i>
Mean	2.24	0.52	0.36	0.61	0.07	3.79
Median	1.77	0.37	0.27	0.51	0.06	3.13
Min	0.20	0.06	0.05	0.20	0.03	0.91
Max	16.1	4.47	5.14	4.32	0.28	28.2

Results and Discussion

Data from the PBDE analysis of the breast milk from 1996 to 2001 are presented in Table 1. The mean and median values for sumPBDE (five congeners) are 3.8 and 3.1 ng/g fat. A high sumPBDE level, 28.2 ng/g fat, is seen in one sample but it does not differ from the general relative congener pattern of PBDE congeners. BDE-47 is the major congener in the milk samples (50-60 % of sumPBDE), and consequently, the BDE 47 levels are strongly correlated to the sumPBDE levels in breast milk (data not shown).

The distribution of sumPBDE concentrations in breast milk from all participating subjects is shown in a frequency diagram (Figure 1). Per interval, most samples (33 individuals) have sumPBDE levels between 2 and 3 ng/g fat. The frequency distribution is skewed which means that a few samples with comparatively high levels are present. However, the single peak value at 28 ng/g is an isolated finding.

The PBDE values from breast milk obtained 1996-2001 have been arranged according to year of sampling in order to follow possible time trends (Figure 2). In this figure the data from the present

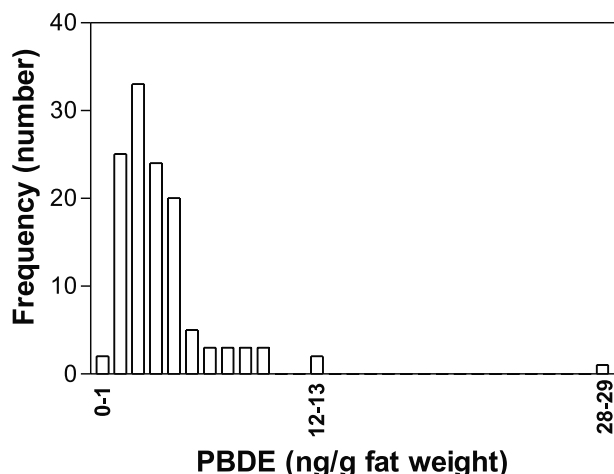


Figure 1. The frequency distribution of sumPBDE values in breast milk from primiparae women in Uppsala County, 1996-2001 (n=124). Note the single high value at above 28 ng/g fat.

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study is compared with a set of data from another Swedish breast milk study, representing pooled banked milk from mothers in the Stockholm region^{4,5}. In our set of data, the result from 1999 has been omitted due to the low number of samples (n=4). Apart from 1999, our data represent 12-40 samples per year.

Our data show an increase in mean sumPBDE levels from 1996 to 1998, after which the levels decrease till the last measured value in 2001. This trend is similar to what has been reported by Meironyté-Guvenius and Norén⁵, in pooled samples from mothers in the Stockholm region. Consequently, we have now two separate study groups, which show a similar picture regarding alterations of PBDEs in Swedish breast milk the last five to six years. If we interpret the results as a peak of PBDE in Swedish human milk at around 1997-1998, this peak comes at a later time point compared to the levels in Swedish environmental samples, e.g. guillemot eggs, where the PBDE peak seems to have occurred around 1990. We should however be careful to draw far-reaching conclusions from these PBDE fluctuations in milk during this relatively short time, especially with respect to the wide distribution of individual samples that make up the mean values.

To conclude, we report on PBDE levels in milk from primiparae women of Uppsala County, Sweden, obtained 1996-2001. Mean and median values for the whole period lie between 3 and 4 ng sumPBDE/g fat (sum of BDE 47, 99, 100, 153 and 154), and a single peak value of above 28 ng/g fat is observed. If the data are separated according to sampling year, a peak in sumPBDE is suggested at around 1998. These results are similar to what has been shown in a study on pooled mother's milk from the Stockholm region. For the future, it is of important to continue the monitoring of PBDEs and other POPs in human fluids in order to estimate future POP body burdens and to warn against possible increasing levels.

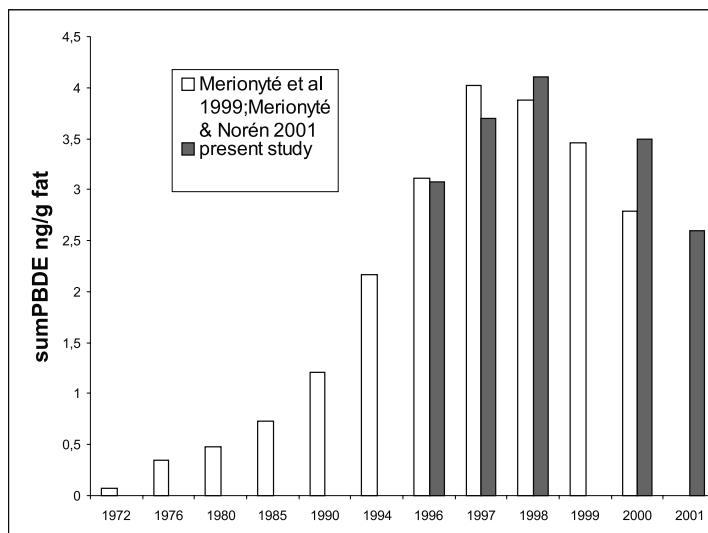


Figure 2. The present data (individual milk samples from Uppsala) arranged according to sampling year, in comparison with pooled breast milk data from mothers from Stockholm region^{4,5}. During the comparable period (1996-2000, the two data sets more or less in concordance).

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