BROMINATED FLAME RETARDANTS IN BREAST MILK FROM NORWAY

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Introduction

Breast milk monitoring studies have long been used to follow temporal and spatial trends in the body burden of persistent organic pollutants (POPs) in lactating women, as an indicator of the general environmental quality and for assessing possible effects on health and development of the infant.[1,2] It is assumed that the levels in breast milk reflect the total maternal body burden during pregnancy and thereby provide a dosimeter of prenatal exposure to POPs. Further, through the process of breast-feeding, the mother transfers potentially toxic POPs to the suckling infant.

Restrictions and bans on the use of many polychlorinated POPs, as well as the reduction of their emission into the environment, have led to a decline of these chemicals in breast milk in most Western countries.[3,4] In contrast, a recent investigation has demonstrated an exponential increase of polybrominated diphenylethers (PBDEs) in Swedish breast milk from 1972 to 1997 with a doubling of the concentrations every five years.[3] This is due to the increasing use of this compound class as flame retardants in polymers and textiles used in electronic and electric equipments, construction materials and furnitures. More recent data have shown that the PBDE body burdens in Sweden began to trend downwards presumably due to the phase-out of the penta-BDE flame retardant in the late 1990s.[5]

Since 1986, the World Health Organization's Regional Office for Europe (WHO/EURO) has coordinated three international breast milk monitoring studies on PCDDs/PCDFs and PCBs using a common standardized study protocol.[6] Norway has participated in all three rounds and breast milk samples were collected in 1986, 1993 and 2001from the same three different geographical areas representing a coastal area in the North (Tromsø), a rural inland area (Hamar) and an industrialized area in the South with known dioxin source (Skien/Porsgrunn). In this contribution, we report on the results for brominated flame retardants in breast milk samples from these WHO studies in addition to samples obtained in 2001 in Oslo.

Methods and Materials

Chemicals

The PBDE standards (BDE-28, 47, 85, 99, 100, 119, 138, 153, 154 and 183) as well as hexabromocyclododecane (HBCD) and tetrabromobisphenol-A were obtained from Wellington Laboratories (Guelph, Ontario, Canada) or CIL (Andover, MA). All solvents used were of pesticide grade from sds (Peypin, France).

Breast milk samples
Breast milk from 10-12 primiparaous mothers living for at least 5 years in the particular area was collected according to the WHO protocol.[6] Samples collected in 1993 and 2001 in Tromsø, Hamar and Skien/Porsgrunn were pooled. From the 1986 study only two individual samples from Tromsø were available. The individual samples from Oslo were obtained from mothers living in a district situated in the vicinity of a municipal waste incinerator.

Sample preparation and quantitative determination
Extraction and clean-up of breast milk samples was done on solid-phase extraction columns (OASIS HLB, 500 mg, Waters Corporation, Milford, MA) according to a previously described method.[7] Separation and quantitative determination of the BFRs were performed by capillary gas chromatography coupled to a mass spectrometer operated in the electron capture mode with methane as buffer gas. The brominated compounds were monitored at *m/z* 79;81. Identification was based on retention time and isotope abundance ratio. The total uncertainty of the analytical method was found to be about 20 %.

Results and Discussion
Geographical and temporal trends
The sum of the PBDEs in the different breast milk pools is shown in Figure 1A. In the samples from 2001, BDE-37, 85, 119 and 138 were included in the sum, however these congeners did not constitute more than 3 %. There are no clear geographical differences, though it appears to be a North-South gradient for the samples from 2001. However, since pooled samples were analyzed, it is not clear if the differences are statistically significant. In average, the PBDE content in Norwegian breast milk has increased by 368 % from 1986 to 2001 and by 58 % from 1993 to 2001. The mean PBDE level in 2001 of 3038 pg/g lipids agrees well with the level of 2800 pg/g lipids reported in Swedish breast milk collected in 2000.[5] The Norwegian breast milk pools have also been analyzed with respect to PCDD/PCDFs and PCBs. In contradiction to the PBDEs, the levels of these chlorinated POPs have decreased by about 60 % from 1986 to 2001.[4]

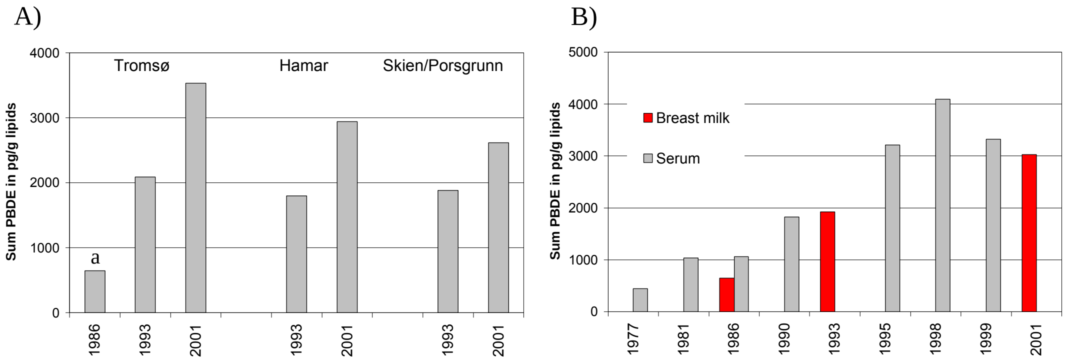


Figure 1. A) The sum of 7 to 11 BDE congeners in pg/g lipids in Norwegian breast milk sampled during the three rounds of the WHO coordinated surveys. **B)** The mean PBDE levels in breast milk compared to previously reported levels in Norwegian serum.

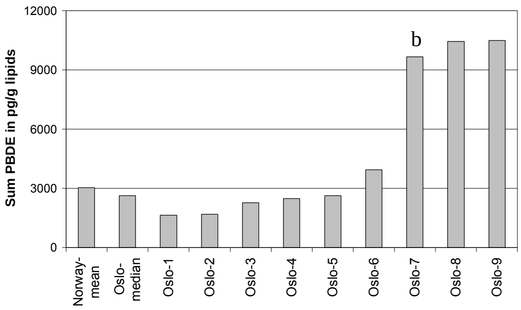
[a]The mean of two individual breast milk samples.

We have previously investigated the PBDE levels in archived serum samples from Norwegian men[8] and these results are presented together with the results from this breast milk study in Figure 1B. As is seen, the levels in serum and breast milk are quite consistent and the PBDE level seems to peak around 1998 which supports the recent trend found in Swedish breast milk.[5]

Individual samples

In 2001, breast milk samples were obtained from mothers living nearby a municipal waste incinerator in Oslo in order to evaluate a possible influence of stack gas emissions on the body burden of PCDDs/PCDFs. The PBDE concentrations in these individual samples are presented in Figure 2A. Three of the nine samples had a total PBDE content exceeding the median by a factor of three, and the relative standard deviation (RSD) was as high as 80 %. The PCDD/PCDF and PCB levels in the same samples showed smaller variation, about 30 % RSD. Accordingly, van Bavel et al.[9] found very high PBDE levels in 5 % of 143 Swedish blood samples. From Figure 2B it is seen that the level of the predominant congener BDE-47 is not correlated with the level of 2,3,4,7,8-PeCDF, the third most abundant PCDD/PCDF congener. This indicates different sources of exposure to these pollutants. Significantly higher blood levels of PBDEs have previously been reported for persons working in electronic dismantling plants,[10,11] at a rubber factory,[12] at a municipal waste incinerator[13] and as computer technicians.[14] None of the persons involved in our study was working at these types of industries and the reason for their high PBDE levels is unknown.

A)



B)

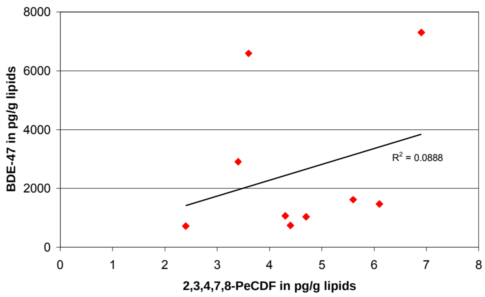


Figure 2. A) The sum of PBDEs in pg/g lipids in individual breast milk samples obtained from mothers living nearby a municipal waste incinerator in Oslo. **B)** The level of BDE-47 compared to the level of 2,3,4,7,8-PeCDF in the samples in 2A.

[b]Low recovery

Other brominated compounds
HBCD, dimethyl-tetrabromobisphenol-A (DiMeTBBP-A) and tribromoanisole (TBA) were determined semi quantitatively in all samples from 2001. HBCD, which is a large production volume flame retardant, was observed in all samples, but at highly varying levels (range ~250-2000 pg/g lipids). DiMeTBBP-A was also detected in all of the samples (range ~10-100 pg/g lipids). This compound has infrequently been used as a flame retardant,[15] but it may also originate from biological methylation of TBBP-A.[16] TBA was found in 6 out of 13 samples (range ~0.5-5 pg/g lipids). It is not known whether the presence of this compound is due TBA occurring naturally in the marine environment or originates from the flame retardant tribromophenol.

Our results demonstrate that it is of great importance to continue surveillance of BFRs in breast milk and to disclose pathways of human exposure that may lead to high body burdens of BFRs in certain individuals.

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