A RETROSPECTIVE STUDY OF PBDEs IN HUMAN MILK FROM THE FAROE ISLANDS

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Introduction

Polybrominated diphenyl ethers (PBDEs) were first reported in fish from Sweden and Japan [1,2] and in German human milk [3], which all have been followed by a large number of studies over the last few years; in humans, fish, birds and mammals [4,5]. A strong increase in PBDE levels was reported in Swedish human milk from the beginning of the 1970s to 1997 [6], but samples from 1998 to 2000 are indicating decreased levels of PBDEs in the milk [7]. A similar trend has been seen in human milk samples from Japan [8]. In Norway the PBDE levels in human milk are increasing from 1986 to 2001, however the concentrations are in the same range as has been seen in Sweden and Japan [9]. In the United States the PBDE levels reported in human milk are about 4 times higher than what have been seen in Europe and Japan [5,10].

The aim of the present temporal study was to determine PBDE concentrations in human milk from Faroe Island women sampled 1987-1999. The Faroe Islands are located far from the European main land and away from industrial sources for PBDEs.

Material and Methods

Chemicals: Standard reference compounds were synthesized in house [11] and those used for quantifications in the present study are shown in Table 1. All solvents were of the highest available commercial grade.

Instruments: Analysis and quantification were performed by gas chromatography/mass spectrometry (GC/MS) a Finnigan TSQ 700 instrument coupled with a Varian 3400 gas chromatograph. The transfer line temperature was set at 270°C and the ion source temperature was 200°C. Helium was used as carrier gas (head pressure 3 psi) and methane as reagent gas at 5.6 torr. On-column injections were made using a septum equipped programmable injector fitted with high-performance insert. The PBDE analyses were performed on a DB-5HT capillary column (15 m x 0.25 mm i.d., 0.1 µm film thickness, J & W Scientific, Folsom, CA, USA). The injector temperature was programmed as 60°C, 180°C/min, to 320°C. The oven temperature was programmed from 80°C (1 min),

15°C/min, to 300°C (16 min). PBDE congeners were quantified using bromide ions formed by electron capture negative ion chemical ionization (ECNI) with a primary electron energy of 70eV. Selected ion monitoring (SIM) was carried out for both bromide isotopes, m/z 79 and 81 [12]. Methane 4.5 with 5 ppm oxygen was used as the electron thermalization buffer gas.

Samples: Mother-child pair cohorts were generated in 1987, 1994-1995 and 1998-1999, and milk was sampled and analyzed for major PCB congeners. Based on these results, samples were selected, and stored milk was retrieved for analysis. Three pooled milk samples from 1987, 1994-1995 and 1998-1999 and 9 individual samples from 1998-1999 were analyzed for the major PBDE congeners.

Analysis: The extraction and clean-up procedure for the milk samples is a modified version of a method described elsewhere [13]. In the extraction formic acid and diethyl ether is used instead of hydrochloric acid and Methyl *tert*-butyl ether. Surrogate standards, BDE-77 and BDE-138 were added to the samples prior to extraction.

Results and Discussion

This is the first report of PBDEs in human milk from the Faroe Islands. The concentrations of four PBDE congeners and the sum of these four are given in Table 1. The levels reported in this study are in a similar concentration range as has been observed in the United Kingdom and in Germany, with sPBDE levels at 6.6 ng/g l.w. [14,15]. The sPBDE levels reported from Sweden and Japan are about 1-3 times lower than what are seen in this study [6-8].

The PBDE congener profile was shown to be different from what has previously been reported in human milk samples [7,8,10,14]. In the present study, BDE-153 is the dominating congener followed by BDE-47, BDE-100 and BDE-99 (Table 1), at each time point. Usually BDE-47 is the most abundant congener followed by either BDE-99 or BDE-153 [7,8,10,14]. The BDE-153 levels reported from the last cohort (1998-1999) are about 6 times higher than what have been seen in Sweden and in a similar concentration range as has been seen in the United States with median values of 2.1 ng/g l.w. and 2.0 ng/g l.w. respectively [6,7,10].

Table 1. Concentrations (ng/g lipid weight) of PBDE congeners identified in human milk from the Faroe Islands. The samples were pooled with 10 mothers in each pool.

Year	1987	1994-1995	1998-1999
No. of mothers	10	10	10
BDE-47	0.5	1.2	1.7
BDE-99	0.20	0.50	1.0
BDE-100	0.25	0.60	1.0
BDE-153	0.60	1.4	3.6
sPBDE	1.5	3.6	7.2

The sPBDE levels from the pooled samples are compared to the nine individual samples from the last cohort (1998-1999), and a quite wide range is seen between the highest and the lowest concentration (Figure 1A). The reason for these concentration differences may be due to life style and dietary habits.

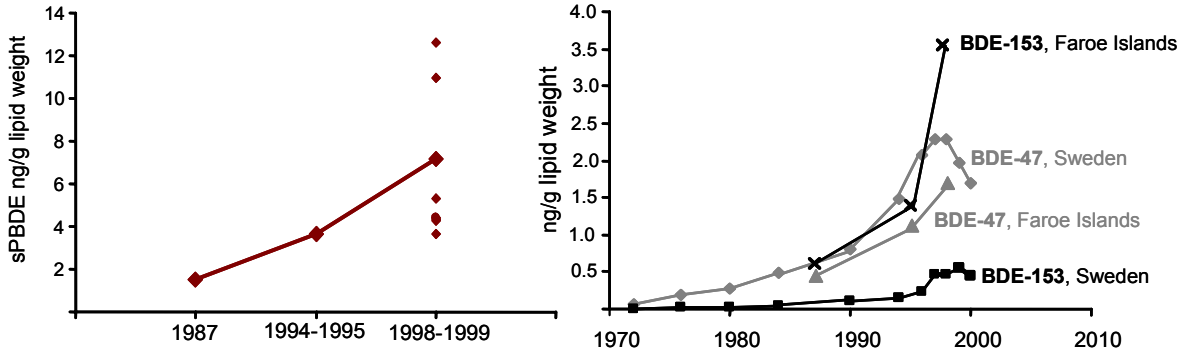

Figure 1. A) sPBDE concentration in ng/g lipid weight (sum of BDE -47, BDE-99, BDE-100 and BDE-153). Three pooled milk samples from mother-child pairs generated in 1987, 1994-1995 and 1998-1999 from the Faroe Islands and 9 individual samples from 1998-1999. **B)** A time trend comparison between Sweden [6,7]and the present study.

All the PBDE congeners and consequently also the sPBDE levels in milk from Faroe women are shown to increase from 1987 to 1999 (Figure 1A and 1B). The BDE-47 trend is in accordance to what have been seen in Sweden c.f. Figure 1B. On the other hand the BDE-153 profile is different to what has previously been reported from Sweden, with considerable higher levels and an increasing trend of the concentration [6,7]. However the PBDE levels for the last years is still unknown at the Faroe Islands calling for additional analyses.

Conclusions

This is the first time PBDEs are detected in humans from the Faroe Islands. An increase of PBDE levels are shown from 1987-1999 in the milk from the Faroese women. The PBDE pattern is different from what have been seen in other studies, with BDE-153 as the dominating congener instead of BDE-47 that is normally the dominating congener [6,7,9].

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