A RETROSPECTIVE TIME TREND STUDY OF PBDEs AND PCBs IN HUMAN MILK FROM THE FAROE ISLANDS

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Introduction

The Faroe Islands are located quite far from the European continent and from industrial sources of polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs). However, the Faroese population may be exposed to these substances through contaminated food, via goods and products in their homes, and in their work environment. High trophic level marine species, including pilot whale and seabirds, such as fulmars, have been shown to accumulate high concentrations of organohalogen substances (OHS) like PCBs and PBDEs [1-3]. Possibly due to dietary differences, wide differences exist in regard to PCB exposures among the Faroese. In a birth cohort from 1987, milk pools contained relatively high PCB concentrations between 1.9-2.5 µg/g lipid weight (l.w.) [4]. In another cohort from 1994, serum from pregnant Faroese women was analyzed for PCB and OH-PCBs, with results ranging from 0.15 to 22 µg/g l.w. and 0.02 to 1.8 µg/g l.w., respectively [5].

In a time trend study for PBDEs and PCBs in human milk from Sweden from the early 1970s to 1997, the PBDE concentrations showed a significant increase [6] while the PCB levels showed a decrease[7]. Human milk samples from 1997 to 2000 indicate a decrease for the PBDEs, mainly due to reduced concentrations of BDE-47 [8]. A similar trend has been seen in human milk from Japan [9]. In Norway, PBDE in human milk increased from 1986 to 2001, with similar concentration levels as reported in Sweden and Japan [10]. In the United States the PBDE levels reported in human milk are about 4 times higher than those seen in Europe and Japan [11,12].

The aim of the present study was to determine PBDE and PCB concentrations in a temporal trend study with samples from 1987-1999 in human milk samples from the Faroe Islands.

Material and Methods

Samples: Mother-child pair cohorts were generated in 1987, 1994-1995 and 1998-1999, and milk was sampled consecutively during a short period at Landssjukrahusið in Tórshavn, Faroe Islands for analysis for major PCB congeners. Based on these results, samples were selected to represent the full range of PCB exposures and stored milk was retrieved for analysis. Pooled samples

containing equal amounts of milk from each mother and each pool consisted of 10 mothers. Three pooled milk samples from 1987, 1994-1995 and 1998-1999 and 9 individual samples from 1998-1999 were analyzed for the major PBDE and PCB congeners.

*Chemicals***:** The individual PBDE congeners were synthesized in house [13] and the PCB congeners (numbered according to Ballschmiter et al. [14]) were purchased from Larodan Fine Chemicals AB, Malmö, Sweden. The reference compounds used for quantifications in the present study are shown in Table 1. All solvents were of the highest available commercial grade.

Instruments: The PBDE analysis was performed by gas chromatography/mass spectrometry (GC/MS), a Finnigan TSQ 700 instrument coupled with a Varian 3400 gas chromatograph and 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 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 temperature programmable injector programmed from 60°C, 180°C/min, to 320°C. The oven temperature was programmed from 80°C (1 min) with 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 [15].

The PCB analysis was performed on a Varian 3400 gas chromatograph, equipped with a Varian 8200 autosampler , an electron capture detector (ECD) and a split-splittless injector operated in the splitless mode. Hydrogen was used as carrier gas and nitrogen as make-up gas. A DB-5 column (30m x 0.25 mm internal diameter and 0.25 µm film thickness J & W Scientific, Folsom, CA, USA) was used. The column temperature was 80°C (2 min), 10°C/min, to 300°C (10 min). The injector temperature was 250°C and the detector temperature 360°C. The data were collected using a PC-based ELDS Pro v2.0 system (Chromatograhic Data System AB, Stockhom, Sweden).

Analysis: The extraction and clean-up procedure for the milk samples is a modified version of a method described elsewhere [16]. In the extraction formic acid and diethyl ether is used instead of hydrochloric acid and methyl *tert*-butyl ether. Surrogate standards, BDE-77, BDE-138 and CB-200 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 main PBDE congeners and the sum of these four are given in Table 1. The range of concentrations reported in this study are similar to those observed in the United Kingdom and in Germany, with average sPBDE concentrations of about 6-7 ng/g l.w. [17,18]. The sPBDE results from Sweden and Japan are generally lower (down to about one-third) when compared to this study [8,9].

The concentrations of the three main PCB congeners as well as the sPCB (CB-118, CB-153, CB-105, CB-138, CB-187, CB-183, CB-128, CB-156, CB-180 and CB-170) of the pooled samples are given in Table 1. The PCB levels reported are considerably higher than these reported from other studies of human milk. For example, Swedish and Belgian concentrations in human milk are a factor of about 3-7 below the results presented here [7,19]. The time trend observed in Sweden showed

a steady decrease of PCB in human milk, with the PCB concentration in 1997 reaching about 30 % of the one observed in 1972 [7]. At the Faroe Islands, the PCB concentration seems rather constant over the years, although a slightly declining trend may be present, although not conclusive based upon this study alone, c.f. Figure 1A and Table 1.

Table 1. Concentrations (ng/g lipid weight) of PBDE and PCB congeners identified in human milk from the Faroe Islands. The samples were pooled with 10 mothers in each pool. sPBDE is the sum of BDE -47, BDE-99, BDE-100 and BDE-153 and sPCB is the sum of CB-118, CB-153, CB-105, CB-138, CB-187, CB-183, CB-128, CB-156, CB-180 and CB-170

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**
PCB-153	590	420	470
PCB-138	500	370	420
PCB-180	390	240	270
sPCB	**2300**	**1600**	**1800**

The PBDE congener profile in the Faroes is clearly different from results previously reported for human milk samples [8,9,12,17]. In the present study, BDE-153 is the dominant congener, followed by BDE-47, BDE-100 and BDE-99 at all three points of time (Table 1). Elsewhere, BDE-47 is the most abundant congener followed by either BDE-99 or BDE-153 [8,9,12,17]. The BDE-153 concentrations reported from the most recent Faroese samples (1998-1999) are about 6 times higher than those seen in Sweden and in a range similar to those reported from the United States [6,8,12].

All PBDE congeners and consequently also the sPBDE show increasing concentrations in milk from Faroese women from 1987 to 1999 c.f. Figure 1B. The variability of sPBDE and sPCB concentrations is apparent from analyses of nine individual samples from the last cohort (1998-1999). The wide range between the highest and the lowest concentration (Figure 1A and 1B) could conceivably be due to differences in life style and dietary habits.

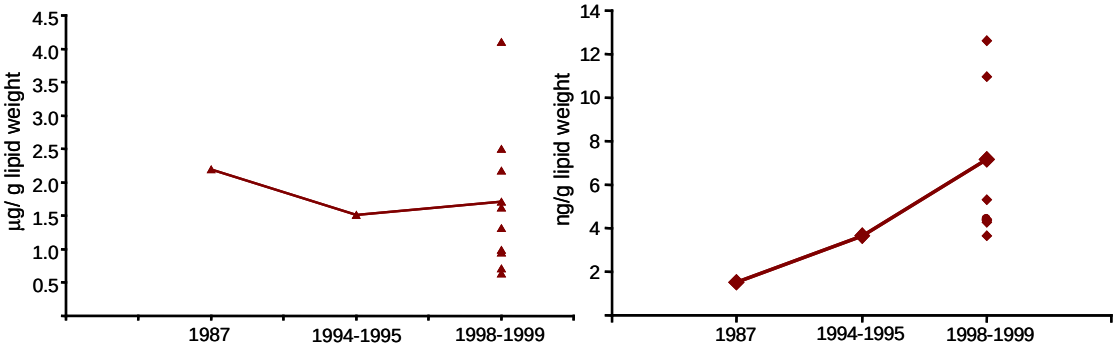

Figure 1. Three pooled milk samples from mother-child pairs generated in 1987, 1994-1995 and 1998-1999 from the Faroe Islands and 9 individual samples from 1998-1999. **A)** sPCB concentration in µg/g lipid weight. (sum of CB-118, CB-153, CB-105, CB-138, CB-187, CB-183, CB-128, CB-156, CB-180 and CB-170). **B)** sPBDE concentration in ng/g lipid weight (sum of BDE -47, BDE-99, BDE-100 and BDE-153).

The BDE-47 trend is in accordance with findings from Sweden c.f. Figure 2. On the other hand, the BDE-153 profile is different from the reports from Sweden, with considerably higher concentrations and a steeply increasing trend of the concentration [6,8]. However, the development in PBDE concentrations during the most recent years is unknown, thus calling for additional analyses from this location.

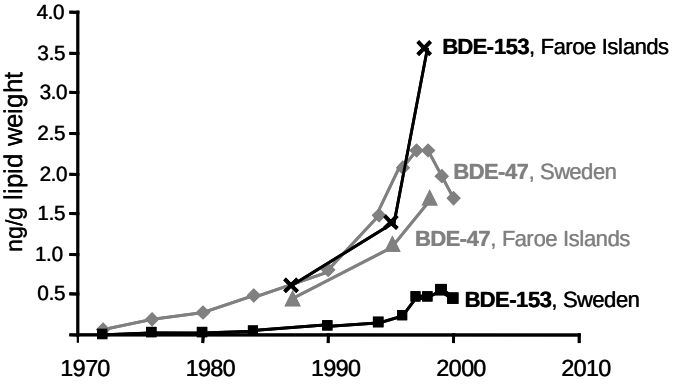

Figure 2. A time trend comparison between Sweden [8,20] and the present study. Three pooled milk samples from mother-child pairs generated in 1987, 1994-1995 and 1998-1999 from the Faroe Islands and 9 individual samples from 1998-1999.

Conclusions

This study reports for the first time PBDEs in human milk from the Faroe Islands. A steep increase of PBDE concentrations is shown from 1987-1999. The PBDE pattern is different from the one reported from studies elsewhere, with BDE-153 as the dominant congener, rather than BDE-47 otherwise being the most prevalent congener [6,8,10].The PBDE sources at this remote location are unknown, and the steep increase calls for further studies.

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