THE CONCENTRATIONS OF PBDES IN SERUM FROM A GROUP OF HIGH CONSUMERS OF FISH FROM A PBDE CONTAMINATED LAKE IN NORWAY

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Introduction

Brominated organic compounds such as polybrominated diphenyl ethers (PBDEs) are used as flame retardants to protect a wide variety of products from catching fire. Several PBDEs have been shown to be persistent organic pollutants (POPs) and are found widespread in the environment, in wildlife and in humans[1,2]. PBDEs are both lipophilic and able to biomagnify, thus intake of fatty fish is supposed to be a major source of human exposure. However, only a few studies have found positive associations between fish consumption and body burdens of PBDEs[3,4].

In Lake Mjøsa, the largest lake in Norway located in the south-eastern part of the country, especially high concentrations of PBDEs have been reported in trout[5,6]. High PBDE levels were also found in perch, pike and burbot[6] from this lake. All these fish species are part of the diet for many people living near the lake. The aim of this study was to investigate the blood levels of PBDEs in a group of high consumers of fish from Lake Mjøsa and correlate these levels with information of intake obtained through food frequency questionnaires.

Materials and Methods

Chemicals

The PBDE standards (BDE-18, 28, 37, 47, 51, 77, 85, 99, 100, 103, 119, 138, 153, 154, 181 and 183) were obtained from Wellington Laboratories (Guelph, Ontario, Canada), CIL (Andover, MA) or AccuStandard (New Haven, CT). All solvents used were of pesticide grade from sds (Peypin, France).

Serum samples

This investigation was conducted on 66 serum samples from a study organised by the Norwegian Institute of Public Health, to investigate the body burdens of POPs in high consumers of inland fish caught in Lake Mjøsa. The participants were recruited among local hobby fishermen and women. Participants provided serum and urine samples and filled in detailed questionnaires regarding personal background data and dietary habits concerning their regular diet and intake of fish from the lake. The project was approved by the Regional Committees for Medical Research Ethics.

Sample preparation and quantitative determination

The serum samples were extracted using solid phase extraction (Oasis® HLB, 540 mg/ 3 mL, custom made from Waters Corporation (Milford, MA)). Our previously presented SPE method[7] was modified and adapted to an automated solid phase extractor (ASPEC XL4, Gilson, Middleton, WI)[8]. 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 and 81. Identification was based on retention time and isotope abundance ratio. The total uncertainty of the analytical method was found to be about 20%. Both the sample preparation and quantitative method are described in detail elsewhere[8]. The lipids were determined enzymatically at Haukeland University Hospital (Bergen, Norway) and the total lipid content of the samples calculated according to the method described by Grimvall et al.[9].

Results and Discussion

The participants in this study consisted of a total of 41 men and 25 women, their mean age being 58 and 54.3 years, respectively (median age 58 and 58 years). The PBDE serum concentrations are presented in Table 1. As can be seen, BDE-47, BDE-99, BDE-100, BDE-153 and BDE-154 were found in almost all of the samples,

while BDE-28, BDE-85, BDE-138 and BDE-183 were more occasionally detected. BDE-47 was the PBDE found at highest concentration in all but six samples, where BDE-153 was highest, and contributed to about 41% of the sum of 7 PBDEs (28, 47, 99, 100, 153, 154 and 183). The next most abundant PBDE observed was BDE-100 (24%), followed by BDE-153 (18%) and BDE-99 (11%). There were no differences in the congener pattern observed in serum from men and women. The high contribution of BDE-100 to the sum 7 PBDE is in contradiction to what has been observed in other Norwegian surveys[10,11], where BDE-100 accounts only for about 10% of the sum 7 PBDE. Whether this congener pattern resembles that of the fish in the lake, has yet to be investigated.

Table 1. Concentrations of the PBDEs in ng/g lipids in the 66 samples.

	Men (n=41)					Women (n=25)				
	mean	**median**	**min**	**max**	**No. det.**	**mean**	**median**	**min**	**max**	**No. det.**
BDE-28	0.30	0.29	0.17	0.58	19	0.33	0.37	0.19	0.46	5
BDE-47	10	7.2	0.30	38	41	5.9	3.5	0.68	24	25
BDE-100	6.1	4.5	0.37	26	40	3.5	1.9	0.36	19	25
BDE-99	3.1	1.5	0.13	22	41	1.6	1.1	0.25	5.8	25
BDE-85	0.43	0.31	0.17	1.1	9	0.23	0.23	0.09	0.36	6
BDE-154	1.3	0.60	0.08	13	41	0.69	0.25	0.11	3.3	25
BDE-153	4.7	3.5	0.77	18	37	2.6	1.7	0.56	14	25
BDE-138	0.22	0.23	0.14	0.29	4	0.19	0.19	0.18	0.20	2
BDE-183	0.35	0.35	0.21	0.49	2	0.23	0.3	0.11	0.32	3
Sum 7 PBDE	25	18	0.51	117	41	14	8.6	2.3	66	25

The frequency distribution of the sum of the 7 PBDEs in the 66 samples is shown in Figure 1. As can be seen, several samples in this study have a PBDE content much higher than the median. For comparison, the median sum 7 PBDE has been found to be 4.7 ng/g lipids in a study on 120 samples comprising serum from men and women from Norway[11] and 2.3 ng/g lipids in a study on 151 Norwegian breast milk samples[10]. PBDE concentrations were also determined in serum from 21 persons who had consumed little or no fish from the lake but were living in the Lake Mjøsa region. The serum concentrations for the high consumers of fish were significantly higher compared to this control group, both for men and woman (medians for sum 7 PBDEs: men 18 ng/g lipids vs 4.7 ng/g lipids, women 8.6 ng/g lipids vs 4.7 ng/g lipids).

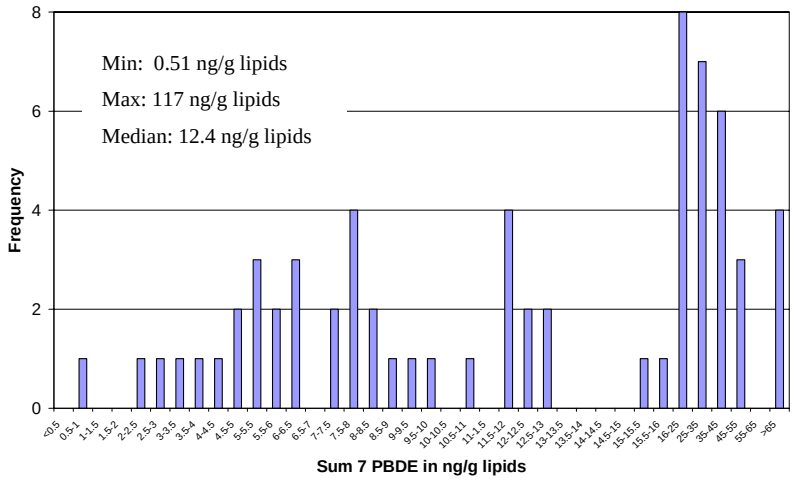

Figure 1. The frequency distribution of the sum of 7 PBDEs in ng/g lipids in the 66 serum samples.

The serum concentrations of BDE-47, BDE-99, BDE-100, BDE-153 and BDE-154 were all strongly correlated, as exemplified in Figure 2 A for BDE-47 and BDE-100. Several other environmental contaminants have been determined in these serum samples. PBDE levels turned out to be strongly correlated with the levels of several PCBs. The relationship between sum 7 PBDE and sum 5 PCB (i.e. 101, 118, 138, 153 and 180) is presented in Figure 2 B.

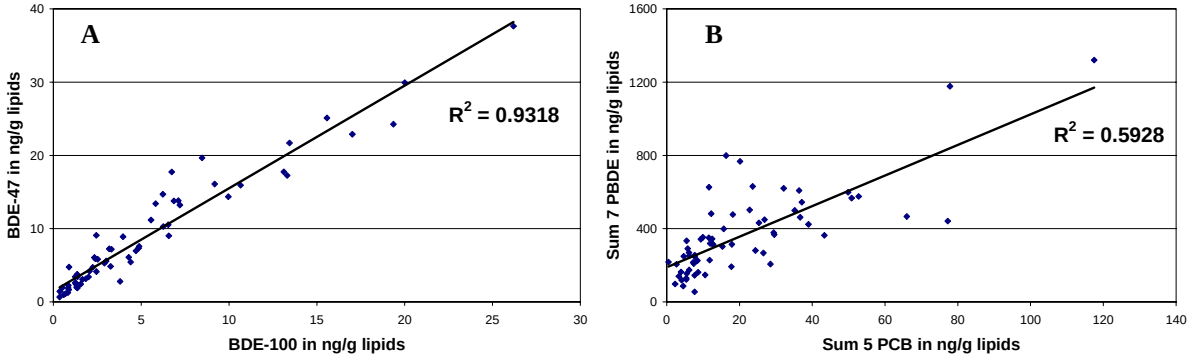

Figure 2. The relationship of the serum concentrations of BDE-47 and BDE-100 (A) and sum 7 PBDE and sum 5 PCB (B) in ng/g lipids in the 66 serum samples.

Fish consumption

Trout was the fish the participants had consumed most frequently, followed by perch and pike. From the questionnaire consumption data, the estimated mean intake of trout was 22.0 g/day (median 14.2 g/day) with quite a wide range (0-101g/day). For comparison, consumption of freshwater fish in Norway is, in general, low and the median intake of those who eat such fish is 4.4 g/day. As can been seen from Figure 3, the participants' blood levels of PBDEs correlated to their reported intake of trout and with their age. The associations were found to be strong and significant when adjusted statistical calculations were performed. For some PBDEs significant differences were also found between men and women. In a recent study by Morland et al.[12] only insignificantly elevated blood levels were observed in urban anglers in the USA, and positive associations between fish consumption and body burdens of PBDEs have so far only been shown in a few studies[3,4]. Due to the contamination of this lake, dietary restrictions have been issued by the Norwegian Food Safety Authority, however more than half of the participants in this study report an intake of fish from the lake far exceeding the advices.

To summarise, this study on 66 hobby fishermen and women shows clear associations between the concentrations of PBDEs in serum and the subjects' age and intake of freshwater fish. BDE-47 was the PBDE found at highest concentration in almost all samples, and concentrations of most of the PBDEs were strongly correlated.

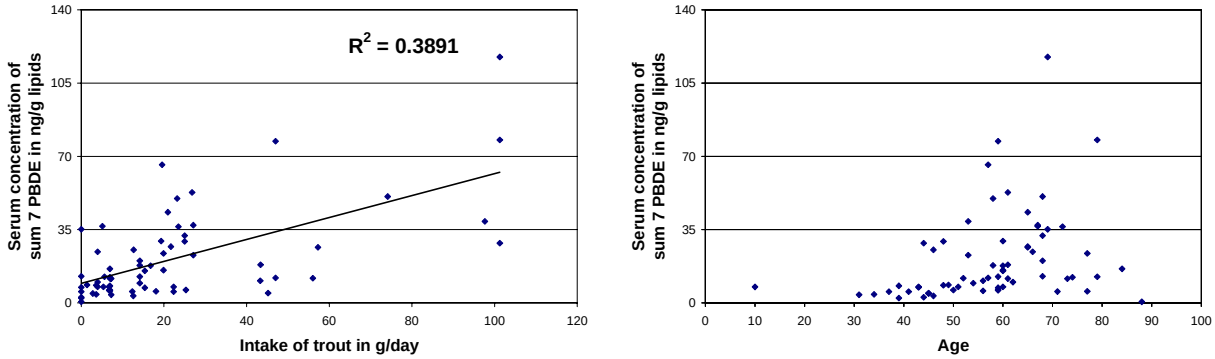

Figure 3. The relationship of the serum concentration of sum 7 PBDE in ng/g lipids (Y-axis) for the 66 study objects and their intake of trout in g/day and age (X-axis).

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