PBDEs in Serum from Persons with Varying Consumption of Fish and Game

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Introduction

Food is thought to be the major exposure pathway of polybrominated diphenylethers (PBDEs) in humans, and the dietary intake has recently been assessed[1,2]. However, several studies have reported a wide range in PBDE concentrations in human samples[3-5], indicating that there might be other important sources or exposure routes as well. The Norwegian Institute of Public Health has in collaboration with the Norwegian Food Control Authority conducted three dietary surveys which focus on consumption of fish and game. Different seafood and game are known to contain high levels of environmental contaminants such as cadmium (Cd), mercury (Hg), polychlorinated biphenyls (PCBs) and dioxins, and may thus be important with regard to dietary exposure of such compounds. Based on these surveys, the intake of the aforementioned contaminants has been estimated. Furthermore, blood samples have been obtained from people with expected different consumption of the contaminants, to see if their consumption is reflected in their blood levels. The objective of this study was to determine the levels of PBDEs in these blood samples.

Materials and Methods

The dietary surveys were started in 1999 and the two first parts encompassed a qualitative (part A) and a semi quantitative (part B) food frequency questionnaire, focussing on consumption of seafoods such as crab, flatfish, halibut, crustaceans, perch and pike, and liver and kidney from game. Based on concentrations of different contaminants in the actual foods, a rough estimate of the individual intake for the participants in part B was established. Samples of blood and urine were obtained from persons with an estimated high consumption of foods with high concentrations of PCB/Dioxins, Hg and Cd, together with an in depth questionnaire. In addition, samples were obtained from persons with a normal consumption of fish and game, resulting in 130 samples in total.

Hexabromocyclododecane (HBCD) and the PBDE standards were obtained from Wellington Laboratories (Guelph, Ontario, Canada) or CIL (Andover, MA). All solvents used were of pesticide grade from sds (Peypin, France).
The serum samples were extracted using solid phase extraction (Isolute 101, (200 mg) from International Sorbent Technology, Mid Glamorgan, UK) after minor modifications

of a previously described method[6]. An additional clean-up on sulphuric acid-silica columns was performed. Separation and quantitative determination of the PBDEs 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 and confirmed by controlling the isotope abundance ratio and the retention time. The uncertainty of the analysis was found to be about 20 %.

Results and Discussion

Preliminary statistical data of the total set of serum samples are shown in Table 1. The seven PBDEs presented, were the most abundant congeners. In addition BDE-37, BDE-119, BDE-85 and BDE-138 were occasionally detected, at very low concentrations. HBCD were found in most of the samples, but their determination is considered to be semi quantitative and the serum concentrations are thus not presented here. The median PBDE level is in accordance with concentrations previously reported in human samples in Norway[7,8].

Table 1. Statistical data of the total sample set.

BDE	28	47	99	100	153	154	183	Sum 7
Mean								
ng/g lipid	0.30	2.98	1.00	0.56	1.45	0.58	0.29	6.84
pmol/g lipid	0.74	6.13	1.77	0.98	2.26	0.90	0.40	12.59
Median								
ng/g lipid	0.18	1.68	0.69	0.34	1.11	0.41	0.22	4.71
pmol/g lipid	0.45	3.46	1.22	0.61	1.73	0.64	0.31	8.45
RSD[a]								
%	187	268	119	190	87	82	76	162
Min								
ng/g lipid	0.05	0.25	0.27	0.04	0.02	0.10	0.009	1.40
Max								
ng/g lipid	4.56	87.93	10.60	11.20	9.56	2.91	1.09	121.67
Detected in no. of samples[b]	66	123	123	119	123	122	61	123

[a]Relative standard deviation of the mean (%).
[b]Seven samples have been excluded from the calculations due to a high blank value.

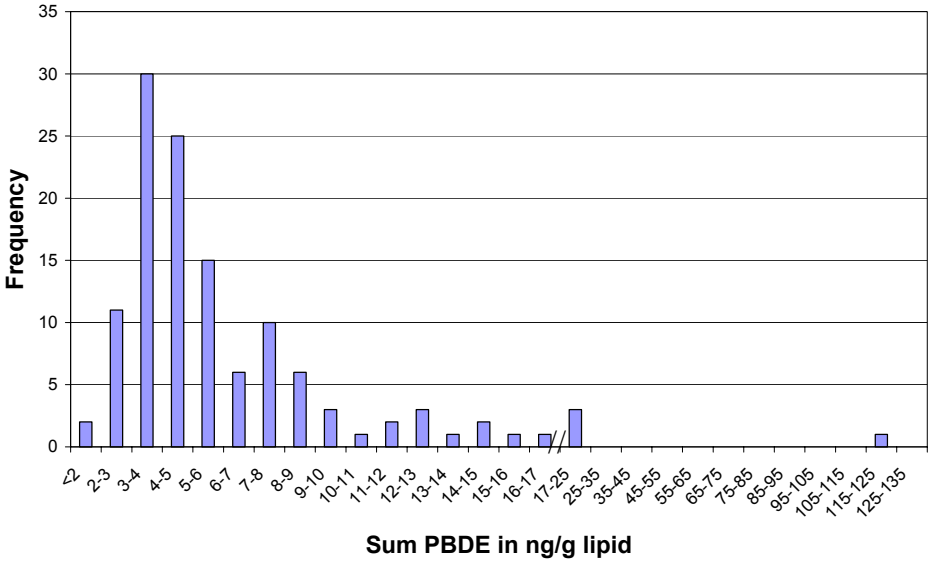

Figure 1. The frequency distribution of the sum of seven PBDEs in ng/g lipid in the 123 serum samples.

As can be seen from Figure 1, the individual serum PBDE concentrations are highly varying. However, group stratification did not identify any increased intake of PBDE for high consumers of seafood. Future statistical analysis of the detailed questionnaire on individual food consumption and life style may reveal causes for the variation in the PBDE levels.

The serum samples have previously been analyzed with respect to PCBs and preliminary results point in the direction of significantly elevated levels in the serum from the persons with an estimated high intake of PCB and dioxins. In Figure 2, serum levels of BDE-47 are compared to the corresponding levels of CB-153. The lack of correlation between these to contaminants indicates other exposure pathways of PBDEs compared to PCBs, and suggests that there are other important sources to PBDE exposure than food.

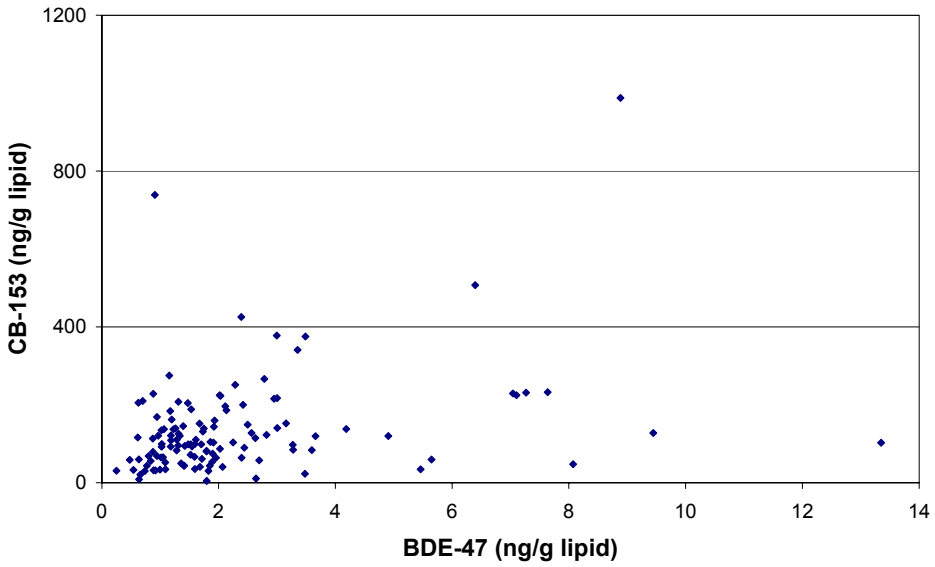

Figure 2. The level of BDE-47 compared to the level of CB-153 in the 123 serum samples.

References

1. Bocio, A., Llobet, J.M., Domingo, J.L., Corbella, J., Teixidó, A., Casas, C., **2003**. *J. Agric. Food Chem.* 51, 3191-3195.
2. de Winter-Sorkina, R., Bakker, M.I., van Donkersgoed, G., van Klaveren, J.D., **2003**. RIVM report 310305001
3. Lind, Y., Darnerud, P.O., Atuma, S., Aune, M., Becker, W., Bjerselius, R., Cnattingius, S., Glynn, A., **2003**. *Environ. Res*. 93, 186-194.
4. Schechter, A., Pavuk, M., Päpke, O., Ryan, J.J., Birnbaum, L., Rosen, R., **2003**. *Environ. Health Perspect*., 111, 1723-1729.
5. van Bavel, B., Hardell, L., Kitti, A., Liljedahl, M., Karlsson, M., Petterson, A., Tysklind, M., Lindström, G., **2002**. *Organohalogen Compounds* 58, 161-164.
6. Thomsen, C., Lundanes, E., Becher, G., **2001**. *J. Sep. Sci*. 24, 282-290.
7. Thomsen, C., Lundanes, E., Becher, G., **2002**. Environ. Sci. Technol. 36, 1414-1418.
8. Thomsen, C., Frøshaug, M., Leknes, H., Becher, G., **2003**. *Organohalogen Compounds* 64, 33-36.