SPATIAL DISTRIBUTION OF POLYBROMINATED DIPHENYL ETHERS IN TROUT FROM NORWEGIAN LAKES

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Introduction

The environmental levels of the brominated flame retardants (BFRs) are increasing and as previously shown for the polychlorinated biphenyls (PCB) the BFRs have now a worldwide distribution. Relatively little is known about the toxicology of the BFRs and the observed increase in the environment is of major concern. The polybrominated diphenyl ethers (PBDE) are used as additive flame retardants and annual demand has now reach approximately 65 000 ton¹ of which the use of the deca mixture constitute of approximately 85%.

The lower brominted diphenyl ethers with a major origin from the so-called penta mixture are shown to be especially prone to bioaccumulation. The levels of this group of compounds are still generally approximately ten times lower than observed for the PCB. However, in some areas the levels are comparable to what is reported for the PCB²⁻⁴. This makes it important to have an extensive monitoring program to reveal potential sources of pollution to avoid future exposure and to make an end of the release.

In this study we have collected big trout (*Salmo trutta*) from five different fresh water lakes from the south-eastern part of Norway. The lakes represent both remote areas with low population density and no industry, and more populated and industrialized areas. The lakes have large populations of trout often used for human consumption.

Methods and Materials

Sample collection

Samples of trout (*Salmo trutta*) were collected from five different lakes from the south-eastern part of Norway in autumn 1999 and 2000 (Fig 1). The size of the collected trout varied between 750g to 12kg. Fillets from the trout were kept frozen at -20°C until analysis.

Extraction and clean up

Fillets from the trout were homogenized in Na_2SO_4 to remove water. Fish representing 0.1-0.25g fat were then spiked with internal standard (BDE-71 and BDE-77) and subjected to cold extraction with cyclohexane and ethylacetate (1:1). The crude solvent extract where cleaned by eluting the extract through a column with acid treated silica with 3% ether in hexane. The purified extracts

with a final volume of 100μ l were then added recovery standard (1234-TCN) and subjected to GC/LRMS-NCI analysis.

Analysis of PBDE by GC/LRMS-NCI

Analysis of the PBDEs (BDE28, -47, -99, -100, -153, -154, -183) where performed by GC/LRMS in the NCI mode. The PBDE were separated by a fused silica capillary column from J&W Scientific (Ultra 2, 25m, 0.2mm id, 0.11 μ m film thickness). The PBDE were detected by negative chemical ionisation monitoring at *m*/*z* 79 and 81, with methane as the chemical ionisation gas.



Fig 1. The trout were collected from five different lakes from the southeastern part of Norway.

Results and Discussion

Big trout were collected from five different lakes located in the south-eastern part of Norway. PBDE were detected in all samples (Table 1 and 2). The study revealed extremely high levels of PBDEs in trout from lake Mjøsa. The levels found are amongst the highest levels of brominated diphenylethers ever measured in fish. The levels were even higher than the levels of PCB previously analysed in trout from the same lake⁵.

PBDE	BDE-28	-47	-99	-100	-153	-154	-183	SUM
Lake								
Mjøsa	0.87	144	126	52.5	14.8	15.7	n.d.	353
Heddalsvatn	0.08	9.6	4.0	2.2	1.1	1.2	0.05	18.2
Eikern	0.05	4.4	3.4	1.9	0.7	1.3	0.03	11.7
Femund	0.04	1.3	1.4	0.4	0.2	0.3	0.01	3.6
Randsfjorden	0.03	2.0	1.3	0.7	0.2	0.3	n.d.	4.6

Table 1: Mean concentration (ng/g wet weight) of seven PBDE congeners in big trout from six Norwegian lakes.

Table 2: Mean concentration $(ng/g \ lipid)$ of seven PBDE congeners in big trout from six Norwegian lakes.

PBDE	BDE-28	-47	-99	-100	-153	-154	-183	SUM
Lake								
Mjøsa	13.3	2197	1763	807	242	260	n.d.	5283
Heddalsvatn	1.9	222	89.5	50.3	24.2	26.0	1.1	415
Eikern	2.7	237	172	100	34.2	68.7	1.9	616
Femund	2.9	99.3	103	32.5	12.2	19.4	0.7	269
Randsfjorden	1.2	69.9	46.2	23.7	8.2	11.5	n.d.	161

Lake Mjøsa is located at a relatively densely populated (a population of approximately 200 000 inhabitants) and industrialized area and one should therefore expect higher levels than in fish collected from more remote areas. However, previously it has only been measured such high levels in fish collected from lakes or rivers near industrialized areas using these brominated compounds^{1,3}. This knowledge and the congener distribution of the PBDE indicates a local release of the penta mixture probably in near history. However, at present there is no certain information regarding the sources of the pollution.

The levels of PBDE in trout from Lake Femund and Lake Randsfjorden were relatively low and on the basis of their location may represent expected background values of PBDE in fish. Lake Femund has a remote location with a very low population living near the lake and no industry around. Lake Randsfjorden has no industry around although it is located closer to more populated areas. Lake Heddalsvatn and especially Lake Eikern are two lakes located longer south near rather highly populated area with some industry. The levels of PBDE in trout from these lakes were higher than in trout from Lake Femund and Lake Heddalsvatn, which indicate a spatial distribution of PBDE towards higher populated areas.

The variation of the results within sample groups was high (Table 3). This is probably due to differences in size and age of the trout. This will be studied further. Previously it has been shown that the levels of environmental contaminant generally are proportional to size and age^{2, 3}. One shall either not exclude the possibility that differences in the food intake between fishes may influence the PBDE levels.

	Sum PBDE (ng/g wet weight)			Sum PBDE (ng/g fat weight)		
Lake	Mean	Median	Range	Mean	Median	Range
Mjøsa	353	161	72-1120	5283	4461	1496-7915
Heddalsvatn	18.2	16.9	5.2-32.4	415	420	99.4-932
Eikern	11.7	9.7	3.1-21.9	616	677	259-828
Femund	3.6	3.6	0.1-8.8	269	254	9.4-572

Table 3 Mean, media and range of SUM PBDEs from trout.

This study confirms the importance of monitoring environmental contaminants in the biota to reveal possible sources of pollution and sources of exposure. Of special concern is the limited knowledge about the toxic effects of chronic exposure to brominated flame retardants, both to human and animals. In Norway trout is a popular fish for consumption and may therefore be an important source of exposure. Furthermore, wildlife animals are periodically vulnerable to starvation and this leads to redistribution of the contaminants in the body from the fat tissue to the blood, liver and brain^{6, 7}. It has been demonstrated that there is a dramatic increase in PCB levels in the brain and in the liver under such conditions^{6, 7}. This phenomenon is of major concern and should be investigated further.

Acknowledgement

The authors which to thank the Norwegian Council for Research for financial support under the PROFO program.

References

- 1. de Wit, C.A. (2002) Chemosphere 46, 583
- Hale, R.C., La Guardia, M.J., Harvey, E.P., Mainor, T.M., Duff, W.H. and Gaylor, M.O. (2001) Environ. Sci. Technol. 35, 4585
- Manchester-Neesvig, J.B., Valters, K. and Sonzogni, W.C. (2001) Environ. Sci. Technol. 35, 1072
- 4. Strandberg, B., Dodder, N.G., Basu, I. and Hites, R.A. (2001) Environ. Sci. Technol 35, 1078
- 5. Fjeld, E., Knutzen, J., Breivik, E. M., Schlabach, M., Skotvold, T., Borgen, A.R. and Wiborg, M., L. (2001) NIVA-rapport 4402-01 (ISBN 82-577-4044-6) [Norwegian].
- 6. Wiemeyer S.N. and Cromartie, E. (1981) Bull. Environ. Contam. Toxicol. 27, 499
- 7. Ingebrigtsen, K., Skaare, J.U., and Teigen, S.W. (1984) J. Toxicol. Environ. Health 14, 813