Polybrominated Diphenyl Ethers (PBDE) in the Swedish Environment - a Summary

Ulla Sellström[1], Amelie Kierkegaard[1], Cynthia de Wit[1], Bo Jansson[1], Lillemor Asplund[1], Linda Bergander[1], Anders Bignert[2], Tjelvar Odsjö[2], Mats Olsson[2]
[1]Institute of Applied Environmental Research, ITM, Laboratory for Analytical Environmental Chemistry, Stockholm University, S- 106 91 Stockholm, Sweden
[2]Swedish Museum of Natural History, Contaminant Research Group, Box 50007, S- 104 05 Stockholm, Sweden

INTRODUCTION

Polybrominated diphenyl ethers, PBDE, are used as additive flame retardants in polymeric materials. The use of flame retardants has increased due to stricter fire regulations in many countries and an increased use of plastic materials and synthetic fibres. In 1992, 600,000 tonnes of flame retardants were used worldwide according to OECD (1). 150,000 tonnes were brominated compounds and 40,000 tonnes, were PBDE.

One of the first reports of PBDE in the environment came in 1981 (2). TeBDE to HxBDE were found in fish and eel from the Swedish River Viskan, along which textile and plastics industries are situated. In 1987 PBDE were detected in fish predators (seal, guillemot and sea eagle) from the Baltic Sea, the North Sea and the Arctic Ocean (3). PBDE have also been detected in environmental samples from Japan (4-7), the Netherlands (8,9) and the USA (10,11).

MATERIAL and METHODS

Sediment samples, sewage sludge (Table 1) and biological samples from terrestrial, freshwater and marine ecosystems (rabbit, moose, reindeer, young starlings, whitefish, Arctic char, bream, perch, pike, trout, herring, grey seal, harbour seal, ringed seal, guillemot) were included in a Swedish survey. Fish and fish predators were included in order to study possible bio magnification (herring, grey seal and guillemot). Samples were collected near possible point sources as well as from remote areas where atmospheric deposition is the most probable source. Other samples came from industrialised and more populated areas without known point sources. Geographical differences were investigated (fall-caught herring from five different locations along the Swedish coast, guillemot, ringed seal and harbour seal from the Arctic Ocean, the North Sea and the Baltic Sea) as well as retrospective time trends (a laminated sediment core, guillemot eggs and pike). High volume air samples (48 hours) were collected from two locations without industrial activities.

Three PBDE congeners dominate in environmental samples and were therefore analysed: 2,2',4,4'-teBDE (= TeBDE), an unknown PeBDE congener (= Pe1BDE) and 2,2',4,4',5-peBDE (= Pe2BDE). Sample extraction and clean-up are described elsewhere (3,12-14). Analyses were made using GC/MS-ECNI (electron capture negative ionisation).

RESULTS and DISCUSSION

PBDE levels in sewage sludge and sediments are given in Table 1. The levels were similar in the

two sewage sludge samples, indicating that the primary PBDE sources to this matrix are household and industrial effluents and not washout from the atmosphere. Levels were low in the top 5 mm of the sediment core (13) but were high in the surficial sediment taken downstream from the plastics industry (15,16).

Table 1. Concentrations of PBDE in sediment and sewage sludge, (ng/g dry weight)

Sample	Year	TeBDE	Pe1BDE	Pe2BDE	Sum of 3 congeners
Sediment, Bornholm deep, southern Baltic Sea	1987	0.29	0.056	0.18	0.52
Sediment, upstream industry	1988	2.5	1.1	5.9	9.5
Sediment, downstream industry	1988	490	170	770	1400
Sewage sludge, dry period (1), Gothenburg	1988	10	2.5	13	25
Sewage sludge, rainy period (2), Gothenburg	1988	8.6	1.9	11	21

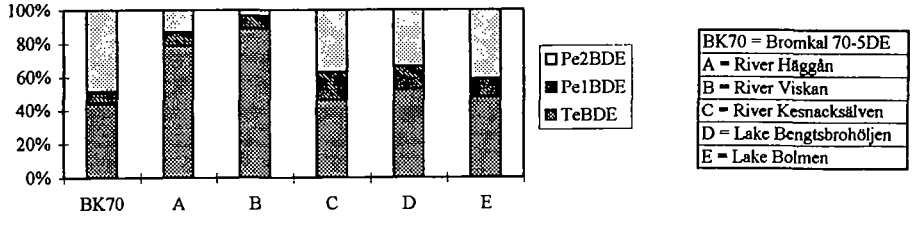
All terrestrial species came from areas without local sources and had low PBDE concentrations. The levels in rabbit were below detection limits and the concentrations in moose, reindeer and starlings were all between 0.47 and 13 ng/g lipid weight (12,14). The PBDE congener patterns were quite similar to that of Bromkal 70-5DE and the sediments and sewage sludge samples.

The results from the freshwater fish studies indicate that southern Sweden is more contaminated with PBDE than northern Sweden (Table 2). Fish from River Viskan (where plastics and textile industries are situated) had the highest concentrations. This area has previously been shown to be polluted with PBDE (2).

Table 2. PBDE levels in fresh water fish (ng/g lipid weight)

Species	Location	Type of area	Year	TeBDE	Pe1BDE	Pe2BDE	Sum of 3 PBDE
Whitefish (Coregonus sp.)	Lake Storvindeln	pristine mountain lake	1986	15	3.9	7.2	26
Arctic char (Salvelinus alpinus)	Lake Vättern	populated area	1987	400	51	64	520
Bream (Abramis brama)	River Viskan, 1	industrial activities	1987	250	11	2.3	260
Bream (Abramis brama)	River Viskan, 1	industrial activities	1987	750	37	2.4	790
Perch (Perca fluviatilis)	River Häggån	industrial activities	1987	2200	230	380	2800
Pike (Esox lucius)	River Häggån	industrial activities	1987	6500	640	1100	8200
Perch (Perca fluviatilis)	River Viskan, 2	industrial activities	1987	24000	3500	9400	37000
Pike (Esox lucius)	River Viskan, 2	industrial activities	1987	2000	170	78	2200
Pike (Esox lucius)	River Kesnacksälven	populated area	1988	98	36	79	210
Trout (Salmo trutta)	River Kesnacksälven	populated area	1988	140	33	130	300
Pike (Esox lucius)	Lake Bengtsbrohöljen	populated area	1988	94	25	60	180
Trout (Salmo trutta)	Lake Bengtsbrohöljen	populated area	1988	250	68	220	540
Trout (Salmo trutta)	Skifors	populated area	1988	190	37	64	290
Pike (Esox lucius)	Lake Bolmen	populated area	1987	47	12	44	103

Figure 1. Relative amounts of PBDE in BK70 and pike from different locations



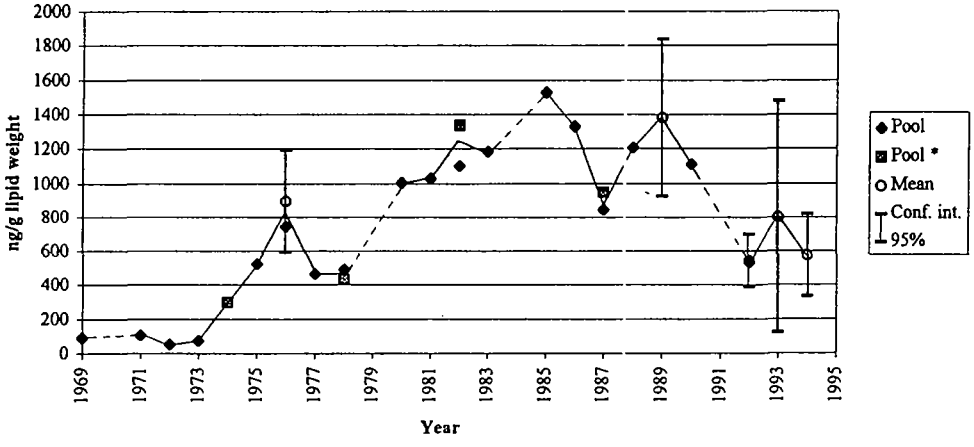
When comparing relative amounts of the three PBDE in pike from different locations, the pattern is different (Figure 1). This indicates that the PBDE source to the River Viskan is different from the others.

The spatial trend of PBDE in herring caught at five different locations along the Swedish coast is almost identical to that previously found for PCB and the DDTs (17). The lowest concentrations are found on the west coast and the highest concentrations in the southern part of the Baltic Sea. The concentrations then decrease from south to north up to Bothnian Bay (14). Also guillemot and seal from the Baltic Sea contain higher PBDE concentrations than guillemot and seal from the North Sea and the Arctic Ocean (3,12,14).

The PBDE concentrations are higher in guillemot egg (7.1 to 19 times for the three PBDE) and grey seal (4.3 to 19 times for the three PBDE) than in their major food item herring, from the same area in the Baltic Sea, indicating biomagnification.

All three time trend studies show the same picture, indicating that the levels of PBDE have increased in the Swedish environment since the 1970s. In Figure 2 the time trend for guillemot eggs is shown.

Figure 2. Time trend study of PBDE in guillemot eggs from the island of Stora Karlsö, sum of 3 congeners



TeBDE and PeBDE were found in both air samples, on both filters and PUFs, indicating that one of the reasons for the widespread distribution of PBDE in Sweden and the Arctic is long range transport by air.

CONCLUSIONS

PBDE seem to be present everywhere in the Swedish environment, in samples from different ecosystems and from different places. Concentrations are highest in samples collected in the vicinity of industrial (textile and polymer) activities. Samples from heavily populated areas contain higher levels of PBDE than samples from more pristine areas. Generally the concentrations are higher in aquatic organisms than in terrestrial organisms.

The spatial trend for PBDE along the Swedish coast is almost identical to that previously found for PCB and the DDTs.

Only long range transport in air can explain the contamination in whitefish in the pristine mountain lake Storvindeln and PBDE have been found in an air sample collected near this lake as well as in another air sample collected in southern Sweden.

Higher levels of PBDE in fish consumers compared to the fish they eat indicate biomagnification of these substances and possibly that TeBDE biomagnify to a higher extent than PeBDE.

Time trend studies of sediment, guillemot eggs and pike indicate that the levels of PBDE in the environment have increased since the 1970s. Analysis of guillemot eggs from the last years indicate however that the levels may have started to decrease.

PBDE have a similar environmental behaviour as PCB and DDT.

REFERENCES

1. OECD & Environment directorate., Selected brominated flame retardants. Risk reduction monograph, 3, 1994.
2. Andersson, Ö. & Blomkvist, G., Polybrominated aromatic pollutants found in fish in Sweden. Chemosphere, 10, 1051-1060, 1981.
3. Jansson, B., Asplund, L., & Olsson, M., Brominated flame retardants - ubiquitous environmental pollutants?. Chemosphere, 16, 2343-2349, 1987.
4. Watanabe, I., Kawano, M., & Tatsukawa, R., Polybrominated and mixed polybromo/chlorinated dibenzo-p-dioxins and -dibenzofurans in the Japanese environment. Proceedings Int Symp Dioxin '95, Edmonton, Canada, 24, 337-340, 1995.
5. Watanabe, I., Kawano, M., Wang, Y., Chen, Y., & Tatsukawa, R., Polybrominated dibenzo-p-dioxins (PBDDs) and -dibenzofurans (PBDFs) in atmospheric air in Taiwan and Japan. Proceedings Int Symp Dioxin '92, Tampere, Finland, 9, 309-312, 1992.
6. Watanabe, I., Kashimoto, T., & Tatsukawa, R., Confirmation of the presence of the flame retardant decabromobiphenyl ether in river sediment from Osaka, Japan. Bull Environ Contam Toxicol, 36, 839-842, 1986.
7. Watanabe, I., Kashimoto, T., & Tatsukawa, R., Polybrominated biphenyl ethers in marine fish, shellfish and river and marine sediments in Japan. Chemosphere, 16, 2389-2396, 1987.
8. de Boer, J., Organochlorine compounds and bromodiphenylethers in livers of atlantic cod (gadus morhua) from the north sea, 1977-1987. Chemosphere, 18, 2131-2140, 1989.
9. de Boer, J., Brominated diphenyl ethers in dutch freshwater and marine fish. Organohalogen Compd., 2, 315-318, 1990.
10. Kuehl, D.W., Haebler, R., & Potter, C., Chemical residues in dolphins from the U.S. Atlantic coast including Atlantic bottlenose obtained during the 1987/88 mass mortality. Chemosphere, 22, 1071-1084, 1991.
11. Loganathan, B.G., Kannan, K., Watanabe, I., Kawano, M., Irvine, K., Kumar, S., & Sikka, H.C., Isomer-specific determination and toxic evaluation of polychlorinated biphenyls, polychlorinated/brominated dibenzo-p-dioxins and dibenzofurans, polybrominated biphenyl ethers, and extractable organic halogen in carp from the Buffalo River, New York. Environ Sci Technol, 29, 1832-1838, 1995.
12. Jansson, B., Andersson, R., Asplund, L., Litzén, K., Nylund, K., Sellström, U., Uvemo, U.-B., Wahlberg, C., Wideqvist, U., Odsjö, T., & Olsson, M., Chlorinated and brominated persistent organic compounds in biological samples from the environment. Environ. Toxicol. Chem., 12, 1163-1174, 1993.
13. Nylund, K., Asplund, L., Jansson, B., Jonsson, P., Litzén, K., & Sellström, U., Analysis of some polyhalogenated organic pollutants in sediment and sewage sludge. Chemosphere, 24, 1721-1730, 1992.
14. Sellström, U., Jansson, B., Kierkegaard, A., de Wit, C., Odsjö, T., & Olsson, M., Polybrominated diphenyl ethers (PBDE) in biological samples from the Swedish environment. Chemosphere, 26, 1703-1718, 1993.
15. Sellström, U., Jansson, B., Jonsson, P., Nylund, K., Odsjö, T., & Olsson, M., Anthropogenic brominated aromatics in the Swedish environment. Organohalogen Compd., 2, 357-360, 1990.
16. Sellström, U., Polybrominated diphenyl ethers in the Swedish environment. ITM-report, ITM Solna, Stockholm University, S-106 91 Stockholm, Sweden, 45, 1996.
17. Swedish Environmental Protection Agency., Sweden's marine environment - ecosystems under pressure. Organochlorine compounds, Monitor , 172-196, 1988.