BROMINATED FLAME RETARDANTS IN THE ENVIRONMENT - AN OVERVIEW

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Several brominated flame retardants (BFRs) have been found in environmental samples. Previous emphasis was on polybrominated biphenyls (PBB) after a poisoning accident in the USA (1). Later, several tetra- and pentabrominated diphenyl ethers, 2,2',4,4'-TeBDE (BDE-47), 2,2',4,4',5-PeBDE (BDE-99) and 2,2',4,4',6-PeBDE (BDE-100) were discovered in the environment. Recently attention has also been focussed on decabrominated diphenyl ether (BDE-209), tetrabromobisphenol A (TBBPA) and hexabromocyclododecane (HBCD).

Air: BDE-209 was found in air particulate near plants manufacturing BFRs (2), and in airborne dust from near Osaka, Japan (3). Tri-hexaBDEs were detected in air from Taiwan and Japan near metal recycling plants, with concentrations ranging from 7.1-53 pg/m^3 (4). Swedish air samples contained 1-8 pg/m^3 of sumPBDE (sum of BDE-47, -99 and -100) as well as 5.3-6.1 pg/m^3 HBCD (5). Work environments also contain TBBPA, BDE-47 and -99 in indoor dust and air (6).

Sewage sludge: Digested sludge samples from Gothenburg, Sweden contained sumPBDE concentrations of about 20-30 ng/g dry weight (dw) (7). SumPBDE levels in German sewage sludge were 0.4 - 15 ng/g (8). Sewage sludge from a plant with input from a plastics industry using TBBPA and a plant having no known sources of TBBPA had levels of 56 and 31 ng/g dw, respectively (9). Digested sewage sludge samples collected in 1998 from Stockholm showed the presence of BDE-47 (39-91 ng/g dw), BDE-99 (48-120 ng/g dw), BDE-100 (11-28 ng/g dw) and BDE-209 (140-350 ng/g dw) as well as TBBPA (2.9-76 ng/g dw) (10).

Sediments: Japanese river sediments contained TeBDE, PeBDE, HxBDE and DeBDE (3, 11, 12). Concentrations of TeBDE and PeBDE together were 21-59 ng/g dw. Sediment samples collected up- and downstream of a Swedish plastics industry using TBBPA contained TBBPA and dimethylated TBBPA (MeTA) (9) as well as BDE-47, -99 and -100 (13). TBBPA, MeTA and all three PBDE congeners were found in higher concentrations downstream of the plant than upstream: 270, 1500, 490, 750 and 170 ng/g dw, respectively, in the downstream samples.

Surficial sediment samples from a Swedish river with numerous textile industries had increasing concentrations of BDE-47, -99, -100 and -209 as well as HBCD further downstream as more industries were passed (14). The range of sumPBDE levels were nd-9.6 ng/g dw, BDE-209 were nd-360 ng/g dw and HBCD were nd-1600 ng/g dw.

The upper layer of a sediment core collected in the southern Baltic Sea contained sumPBDE levels of 0.52 ng/g dw (7). In a more recent study, twenty surficial sediment samples taken from numerous sites in the Baltic Sea had sumPBDE levels of nd-1.1 ng/g dw (15).

Several BFRs were analyzed in sediments collected from the mouths of major European rivers (16, 10). BDE-47 and -99 concentrations were highest in two Great Britain rivers (Humber and Mersey) and two in the Netherlands (1.61-13.1 ng/g dw). Highest 2,2',4,4',5,5'-HxBB levels

were found in the Seine (France), three rivers in the Netherlands, the rivers Schelde (Belgium), Forth (Great Britain) and Ems (Germany) (0.013-0.056 ng/g dw). Levels of DeBB were highest in sediment from the Seine (2.4-3.9 ng/g dw). BDE-209 levels were highest in the River Mersey (Great Britain), followed by the Schelde and River Liffey (Ireland) (34-1800 ng/g dw).

Biota: Levels of sumPBDE are low in mammals and birds from the terrestrial ecosystem: 5.7-13 ng/g lipid weight (lw) in starlings, 0.47-1.7 ng/g lw in reindeer and moose (all from Sweden) (17, 13, 18) and 2.5-4.5 ng/g fat in German cow's milk (19). PBDE concentrations found in freshwater fish depend on the species and the collection site. Loganathan et al. (20) found TeBDE to HxBDE in carp from Buffalo River, New York and TeBDE and PeBDE levels were 13-22 ng/g fresh weight (fw). In eels from Dutch rivers and lakes (10 locations), levels of BDE-47 ranged from <20 to 1700 ng/g lw (21). Several species of freshwater fish from North-Rhine Westphalia contained 18-983 ng PBDE/g lw (19). Various fish species studied from background areas in Sweden had sumPBDE concentrations ranging from 26-1200 ng/g lw (13, 17, 18). Along the River Viskan, upstream and downstream of textile industries using BFRs, sumPBDE concentrations in pike ranged from nd-4600 ng/g lipid and HBCD ranged from nd-8000 ng/g lw (14).

TeBDE and PeBDE levels in several Japanese marine fish and shellfish were between 0.1 and 17 ng/g fw samples (12). A mussel sample from Osaka Bay also contained BDE-209. Cod liver collected from three regions of the North Sea had sum levels of BDEs -47 and -99 of 1.9-360 ng/g lw (22, 23). SumPBDE levels in herring from five sites along the Swedish coast ranged from 17-62 ng/g lw (13, 18) and in the Baltic Sea, 3.2 to 32 ng/g lw (24). Levels of 8.4-100 ng/g lw were found in herring from three regions in the North Sea (21, 23). BDE-47 and -99 levels in Baltic salmon were 167-190 and 52 ng/g lw, respectively (24, 25).

Di-TrBDE were identified but not quantified in black skimmer tissues and eggs in the U.S. (26). SumPBDE concentrations in osprey found dead in Sweden were 2100 ng/g lw (13, 17, 18) and levels in white-tailed sea eagle collected from the Baltic Sea were 350 ng/g lw (27). Common guillemots collected in 1979-81 from the Baltic and North Seas contained 370 and 80 ng PBDE/g lw and Brunnich's guillemot from Svalbard (Arctic) contained 130 ng PBDE/g lw (27). Baltic guillemot eggs collected in 1994 contained sumPBDE of 570 ng/g lw (13).

Female ringed seals collected in 1981 from Svalbard contained sumPBDE levels of 40-51 ng/g lw (13, 18, 27). Baltic Sea harbour seal contained 90 ng PBDE/g lw, and harbour seal from the North Sea contained 10 ng PBDE/g lw (13, 27). Female grey seals from the Baltic Sea collected in 1979-1985 contained 730 ng/g lw (13, 17, 18). Blubber from Baltic grey and ringed seals collected between 1981 and 1988 contained 419 and 350 ng/g lw, respectively (24). Harbour seal from the Dutch coast contained 605-6010 ng/g lw (28).

Bottlenose dolphin samples collected during a mass mortality event on the south Atlantic U.S. coast in 1987/88 contained 180-220 ng PBDE/g lw (29). Bottlenose dolphins from the Gulf of Mexico contained up to 8000 ng PBDE/g lw (30). Recently, de Boer et al. (28) found PBDE levels of 187-349 ng/g lw in sperm whale, 869 ng/g lw in minke whale and 7700 ng/g lw in white-beaked dolphin collected along the Dutch coast in 1998. Long-finned pilot whales from the Faroe Islands had PBDE concentrations (19 congeners) ranging from 843-3160 ng/g lw (31).

Humans: Hexa-decaBDE have been found in human adipose tissue samples from the USA (32, 33) with levels of nd-1 ng/g fat for HxBDE, 0.001-2 ng/g fat for HpBDE, and nd-8 ng/g fat for OcBDE. NoBDE levels were estimated to exceed 1 ng/g fat and DeBDE levels were estimated to range between nd-0.7 ng/g fat. BDE-47 concentration in adipose tissue of a Swedish 74-year old

male was 8.8 ng/g lw (24). Human adipose tissue samples from 77 Swedish individuals contained a mean BDE-47 level of 3.8 to 16 ng/g lw (34).

Human plasma samples were found to contain both TBBPA and PBDEs (35) with TBBPA levels in the low ng/g lw. Mean concentrations of 6 PBDE congeners (sum of BDE-28, -47, -66, -99, -100 and -153) were 2.1 ± 1.4 ng/g lw. In a study of workers at a computer disassembly plant, workers in a computerized office and cleaners (control), BDE-47, -153, -154, -183 as well as -209 were found in blood plasma for all 3 groups (36). The median concentrations were highest in the computer disassembly plant workers (26 ng/g lw), next highest in the office workers (4.1 ng/g lw) and lowest in the cleaners (3.3 ng/g lw).

PBDE levels in human breast milk from 25 German mothers ranged from 0.6-11 ng/g fat (19). Norén and Meironyté (37) found PBDE levels (sum of 8 congeners) of 4 ng/g lw in breast milk from Swedish mothers in Stockholm in 1997. Total PBDE levels of 5 congeners (BDE-47, -99, -100, -153, and -154) in breast milk from mothers in Uppsala were 4.4 ng/g fat (38).

Temporal trends: The temporal trends studied all indicate increasing levels of BDE-47, -99 and -100 in the environment since the 1970s. The trends in guillemot indicate that levels of BDEs -47, -99 and -100 have begun to decline in the Baltic Sea in the 1990s (39). The trends in pike from Lake Bolmen, Sweden, indicate a levelling off of PBDE levels in the 1990s. However, the human breast milk trend indicates that levels are increasing exponentially, doubling every five years (38). These differences may reflect differences in exposure routes.

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