

POLYBROMINATED FLAME RETARDANTS - A THREAT?

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Background

A major threat to human life and to our property is fire. The costs of fires are tremendously high and have always been. It is thus not really surprising that flame retardants were used for more than 2000 years ago. Pieces of tapestry was protected by ammonium salts from catching fire in the castles of the French King, Ludwig XIV¹, in the second half of the 17th century. The needs for fire resistant chemicals grow with the industrial revolution and PCB may be taken as an example of a chemical product that came on the market very much depending on its low flammability. The intense uses of polymeric materials today are a major reason for development and use of flame retardants (FRs).

Resistance to fire development may be met by use of fire proof materials or by using a range of different types of chemicals, inorganics and organics. Halogenated (brominated and chlorinated) compounds, phosphorus containing and to a lesser extent nitrogen based chemicals are used either as additives to polymers or covalently bound (reactive FRs) to the material to be flame retarded. Antimony bromide is often used together with brominated flame retardants (BFRs) to further improve the flame retardant property of the chemical. Generally speaking the flame retardants act mainly through three mechanisms that may be summarised by; *i*: cooling, *ii*: coating or *iii*: dilution of the combustible material (polymer).

The aim of this presentation is to review the state of the science on BFRs and to discuss their potential threats to humans and wildlife. The presentation is also aimed to address the issue on what basis we should assess environmental pollutants in general, BFRs in particular, and what the responsibilities are for us as scientists active in environmental research.

Brominated flame retardants

Forty percent of the annual world production of bromine is consumed in production of BFRs². A limited number of BFR manufacturers produce 250-300 tons of 75 brominated flame retardants per year. The literature on BFRs have increased exponentially and some major sources for information on environmental and health issues of BFRs are several IPCS documents^{1,3-5}, reviews by de Boer⁶, de Wit⁷, Bergman *et al.*⁸, one hundred abstracts from BFR 2001⁹ and an additional number from the present symposium.

Three types of BFRs make up the major proportion of BFRs manufactured. These are tetrabromobisphenol A (TBBPA) and its derivatives (120.000 tons/year), polybrominated diphenyl ethers (PBDEs) that should be divided into the commercial products: PentaBDE, OctaBDE and DecaBDE, produced world-wide in volumes of approximately 8.500, 3800 and 55.000 tons annually, respectively, and hexabromocyclododecane (HBCDD or HBCD) with an annual world production volume of approx. 16.000 tons¹⁰. Only based on the production volumes of these BFRs, they should be regarded to be of environmental concern. For other BFRs produced at lower volumes, such as 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), decabromodiphenylethane (DBDPE), tetrabromophthalic acid diesters, 2,4,6-tribromophenol and pentabromophenol, more

information are necessary for assessing their environmental transport, distribution, fate, exposures to human and wildlife and effects.

Tetrabromobisphenol A and derivatives: TBBPA is mainly a reactive flame retardant but since polymerisation rarely result in total reactions some unreacted monomeric TBBPA will be present in the polymer. This compound will to some extent leak out of the polymer during the lifetime of the product causing some environmental contamination. Handling large volumes of TBBPA will also serve as a source for the environmental contamination. It is confirmed by several studies that TBBPA is distributed to sediments and sewage sludge^{7,11,12}. Interestingly it has been shown that TBBPA may be photochemically transformed to dibromo- and tribromophenols, thus serving as a source for e.g. 2,4,6-tribromophenol^{13,14}. TBBPA is a ubiquitous contaminant in air at e.g. electronics dismantling plants as shown both by air measurements^{15,16} and elevated levels in personnel at such plants¹⁷⁻²⁰. TBBPA has, to my knowledge, not hitherto been reported as a bioaccumulating pollutant or as a chemical with strong affinity to other compartments *in vivo*. This is supported by the short half-life indicated of this compound in humans²¹ and shown in rats²². In the latter study several conjugates of the compound were identified in bile leading to excretion of the parent compound after deconjugation in the intestinal tract.

TBBPA has a low toxicity in general⁵ but *in vitro* experiments have shown a strong affinity for the tyroxine (T₄) binding site on transthyretin, a conservative T₄ transporting protein²³. TBBPA has some cytotoxicity and it has been indicated to inhibit some Cytocrome specific reactions²⁴. The TBBPA toxicity to aquatic species seem to be low⁵.

Summarising the present data on TBBPA does not make this compound to a major BFR threat. However, it is too early to tell and particularly the derivatives of TBBPA, the diallyl, diglycidyl and bis(2,3-dibromopropyl) ethers of TBBPA, are poorly studied. This is intriguing since these compounds all carry functional groups that may be regarded as reactive, particularly the latter two. In depth studies are requested to determine the toxicity profile of TBBPA derivatives. Also the potential exposures to these TBBPA derivatives need to be assessed.

Polybrominated diphenyl ethers - PentaBDE: The present extensive data set on environmental parameters for one of the commercial PBDE products, PentaBDE, clearly show that this product contain several PBDE congeners fulfilling the criteria of persistent organic pollutants²⁵. PentaBDE congeners, 2,2',4,4'-tetraBDE and 2,2',4,4',5-pentaBDE being the most abundant compounds in these products, are transported over long distances and are in fact present in Arctic air masses²⁶. The PentaBDE congeners are found accumulated in wildlife and in humans. PBDE congeners with up to at least six bromine atoms are easily identified and quantified by comparison to authentic reference compounds^{27,28}. The levels have hitherto in general been lower than for the most abundant other environmental contaminants, DDE and CB-153 (2,2',4,4',5,5'-hexaCB). However, a few reports on similar levels of BDE-47 and CB-153 in salmonoid species from the Great lakes show that there is not always a significant difference between the levels of these two compounds^{29,30}. The rapidly increasing trend of PBDEs in herring gull eggs³¹ from the same area and in Beluga whales from St Lawrence estuary³² are other very relevant data showing the general contamination of our environment by PentaBDE. Most recently human mothers milk from the U.S. is reported by Pöpke et al at this symposium³³ to be up in the high ppb range, not significantly different from common PCB levels in mothers milk, at least from Europe. On the other hand, the dominating PentaBDE congener concentrations have been shown to decrease in the Baltic Sea environment as reported in guillemot egg³⁴, herring³⁵, human mothers milk³⁶ or at least to level off as observed in fresh water pike from lake Bolmen in the southern part of Sweden³⁷.

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PentaBDE congeners seem to have low toxicities in general³⁸ but with specific properties such as the potency of inducing behavioural effects as reported for BDE-99³⁹. The lower brominated BDE congeners may undergo metabolism^{23,40,41} to form endocrine active metabolites⁴².

PentaBDE consists of bioavailable, bioaccumulating and toxic compounds that are a threat to the environment.

Polybrominated diphenyl ethers - OctaBDE and DecaBDE: While there is a large data base for PentaBDE congeners there is much less known about the higher brominated compounds. These compounds are in general not yet available as individual standards prohibiting some congener specific work. Only limited data on exposure assessments and other environmentally relevant data have so far been produced, mainly due to technical analytical difficulties, something that is particularly relevant for analysis of decaBDE. The complexity of commercial OctaBDE products have been shown by Sjödin⁴³ while DecaBDE mainly consist of decabromodiphenyl ether (BDE-209). The major constituents of OctaBDE, 2,2',3,4,4',5',6-heptaBDE (BDE-187) and 2,2',4,4',5,5'-hexaBDE (BDE-153), may be used as indicator compounds for exposure via electronics as well as BDE-209^{17,18}. Unfortunately analyses of PBDEs were rarely including reports on hepta- to decaBDE congeners. However, it is evident that also the high mass compound BDE-209 is bioavailable and in fact present in both the general US population and in Swedish citizens^{17,44}. BDE-209 was recently reported at surprisingly high concentrations in Peregrine falcons from Sweden⁴⁵ and it has been indicated in e.g. Cormorant liver (Athanasiadou, unpublished).

DecaBDE, BDE-209, is an interesting compound with properties different from the other PBDE congeners. It has a very low solubility in almost all solvents and hence difficult to study from a toxicological point of view. Still a low uptake of this compound has led to the conclusion in a NTP study that decaBDE showed "some evidence of carcinogenicity" indicating this compound to be potentially carcinogenic⁴⁶. This is reasonable to believe since BDE-209 is more reactive via NAS than possibly expected⁴⁷ and by a short half-life in humans⁴⁸. A recent metabolism study of BDE-209 also support a rapid turnover of the compound in rats⁴⁹. It is also clear that BDE-209 is labile to UV light⁵⁰⁻⁵² leading to the formation of a range of lower brominated diphenyl ethers, and at least under experimental conditions also to form polybrominated dibenzofurans, that will all be more easily absorbed by biota and more readily bioaccumulated than the parent compound.

Toxicologically the most interesting observation is that OctaBDE have shown reproductive toxicity (EU Risk Assessment of OctaBDE) and that BDE-209 induces behavioural effects in mice⁵³.

Taken together the both OctaBDE and DecaBDE congeners are potential threats to human and wildlife but additional information on exposure, dose response relations and on BDE-209 we need information on its potency to induce cancer among other things.

Hexabromocyclododecane: This is a brominated cyclic compound with similarities to hexachlorocyclohexanes (HCHs). Depending on the addition of bromine to the cyclododecatriene precursor for HBCDD three isomers are formed; α , β and γ . It is obvious from this structure that the compound may undergo dehydrobromination to form unsaturated products but due to the ring structure of HBCDD nucleophilic displacement of one or several bromines shall also be expected. In fact, HBCDD does react with the direct alkylating agent nitrobenzylpyridine indicating the potential of the compound to be a direct alkylating agent (unpublished). Still HBCDD is known as a persistent compound that may be present in biota at levels similar to BDE-47. This is true for

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HBCDD in herring³⁵, guillemot⁵⁴ and Peregrine falcons⁴⁵. The present time trend data also indicate an increasing trends of HBCDD in guillemot eggs samples at Stora Karlsö in the Baltic Sea, east of the Swedish main land³⁷ while additional data are requested from herring before any conclusions can be drawn³⁵. HBCDD has been indicated in several other species but the data set is not impressive. Humans are exposed to HBCDD, particularly when working with Styrofoam that is flame retarded with HBCDD as determined by air sampling in a workshop for theatre sidescenes (unpublished). No further work on the personnel was undertaken.

Toxicologically it is not yet obvious what the main effects are even though the liver is a potential target tissue. HBCDD was reported to induce intragenic recombination at an endogenous locus in mammalian cells⁵⁵.

HBCDD is a potential threat to the environment since it is shown to be persistent and bioaccumulating.

Miscellaneous BFRs: A few of all other brominated flame retardants are briefly discussed below to address their potential environmental threat. Since there are a tremendous gap in knowledge of these compounds only some general statements can be made, based on their chemicals structures and structural similarities to other more well known compounds.

1,2-Bis(2,4,6-tribromophenoxy)ethane (BTBPE) has been detected in indoor air at a plant dismantling electronics¹⁵. BTBPE has not yet been detected in the personnel working at the plant. Very poor absorption was reported of BTBPE in rats dosed with radiolabelled BTBPE⁵⁶ indicating this compound to be less bioavailable than other BFRs. Metabolites were indicated in the rat bile. Further studies are necessary to reveal the environmental relevance this compound may have.

Decabromodiphenylethane (DBDPE) is a high mass molecule that may undergo nucleophilic reactions, may have a rather short *in vivo* half-live and in general behave similarly to BDE-209. The compound is a potential environmental threat.

Tetrabromophthalic acid diesters should most likely hydrolyse easier than other non-brominated phthalic acids esters due to the inductive effects of the bromine. The compound should be readily hydrolysed *in vivo*. However, no experimental data have been available to us to further describe the environmental fate of this compound. Speculating, it seems reasonable to regard this compound as a semi-persistent compound with no obvious environmental threat unless it has some specific toxicity.

2,4,6-Tribromophenol and pentabromophenol are weak acidic compounds with a potency to bind to a thyroxine transporting protein (transthyretin) present in the blood of humans and in wildlife⁵⁷. The compounds will not bioaccumulate similarly to other neutral compounds but may exert endocrine related effects through their binding to transthyretin. The potential importance of pollutants bound to blood proteins may have been overlooked and further studies are necessary in this field.

Concluding remarks

Brominated flame retardants are some very different types of compounds that need to be assessed on basis of their individual structures. No general statements on their potency to pose environmental threats can be given. Still there are several BFRs that are of great environmental concern. Those BFRs that are persistent and bioaccumulating compounds will become a problem if manufactured and used in high enough quantities. Toxicological data are required for those BFRs that are semi-persistent to fill the extensive data gaps that we see today on BFRs.

Persisency and bioaccumulation of chemicals must play a more significant role in the future for selection of more environmentally friendly alternatives.

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Finally, it is our responsibility to improve the knowledge to make relevant and better risk assessments in the future of BFRs as well as other chemicals. It is also our responsibility as scientists to share our view and suspicions on potential threat of chemicals used by us. Industry and the society must be able to act faster on emerging issues of environmental concern. As scientists we should supply the tools for faster actions.

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