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Flame retardants: Dust – And not food – Might be the risk

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HIGHLIGHTS

- Levels of flame retardants in indoor dust are much higher than in outdoor environmental matrices.
- This is true for many organophosphorus, brominated and chlorinated flame retardants.
- Chlorinated paraffin levels are much higher than outdoor and indoor levels of other flame retardants.
- Flame retardants in dust might be of higher risk to human health than flame retardants in food.

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ABSTRACT

Flame retardants (FRs) are used to delay ignition of materials such as furniture and electric and electronic instruments. Many FRs are persistent and end up in the environment. Environmental studies on flame retardants (FRs) took off in the late 1990s. Polybrominated diphenylethers (PBDEs) appeared to be bioaccumulative and were found in many organisms all over the world. When PBDEs were banned or their production voluntarily terminated, alternatives appeared on the market that often had similar properties or were of more concern due to their toxicity such as halogenated phosphorus-based FRs. Here we show that in spite of the ban on PBDEs more brominated FRs are being produced, an increasing number of other FRs is being applied and FR levels in our homes are much higher than in the outdoor environment. While nowadays we live in better isolated houses and sit in front of the computer or television, on flame retarded upholstery, we are at risk due to the toxic effects of a suite of FRs. The high exposure to these substances indoors calls for better risk assessments that include mixture effects.

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1. Introduction

Environmental studies on flame retardants (FRs) took off in the late 1990s (Norén and Meyronité, 2000; De Boer et al., 1998). Polybrominated diphenylethers (PBDEs) originating from the technical PentaMix appeared to be highly bioaccumulative and were found in many organisms all over the world (De Boer et al., 2000). The fully brominated decaBDE is present in high concentrations in sediments (Ross et al., 2009). In the early 2000's all PBDEs were banned or their production voluntarily terminated. However, alternatives either had similar properties, such as hexabromocyclododecane (bioaccumulative) (Covaci et al., 2006) or decabromodiphenylethane (accumulating in sediments just as DecaBDE) (Ricklund et al., 2010), or were less persistent but of more concern due to their toxicity (halogenated phosphorus-based FRs). In spite of the ban on PBDEs more brominated (B) FRs are being produced, more than 200,000 metric tons per year (Covaci et al., 2011; López et al., 2011; Picó, 2008). This is mainly caused by the steeply increasing demands in Asia and is based on many different types of BFRs, including newer ones such as decabromodiphenylethane, 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE), 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (TBB or EHTBB), bis(2-ethylhexyl)-3,4,5,6-tetrabromophthalate (TBPH or BEHTBP), tetrabromobisphenol A-bis(2,3-dibromopropylether) (TBBPA-DBPE) and hexachlorocyclopenta-dienyldibromo-cyclooctane (HCDBCO). Ballesteros-Gómez recently identified tris(2,4,6-tribromophenoxy) 1,3,5-triazine (TTBP-TAZ) in plastic electronic products and house dust, showing that new BFRs are still entering the market (Ballesteros-Gómez et al., 2014). Abassi et al. (Abbassi et al., 2015) estimated that considering only the first use (no reuse and/or storage) of PBDE-containing products, approximately 60% of the US/Canadian stock of PBDEs in 2014 (i.e. ca. 70,000 tonnes, 95% of which is decaBDE), will still be in use in 2020. This prompted the

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suggestion of the creation of large environmental reservoirs of decaBDE. Leaching of two new FRs, used as alternatives for decaBDE, resorcinol bis(diphenylphosphate) (PBDPP) and bisphenol A-bis(diphenylphosphate) (BPA-BDPP) from consumer products was shown by Kemmlein et al. (Kemmlein et al., 2003). These substances were also reported by Brandsma et al. in dust from various EU countries, as well as in dust collected in cars (Brandsma et al., 2014).

The number of novel phosphorus flame retardants (PFRs), including chlorinated PFRs detected in the environment over a short period of time is overwhelming (Stapleton et al., 2008, 2009). PFRs are present in many different types of equipment. A chlorinated PFR not previously recorded in the environment, namely 2,2-bis(chloromethyl)propane-1,3-diyl-tetrakis(2-chloroethyl)bis (phosphate) and commercially known as 'V6', was identified in polyurethane foam from baby care products, in houses and in cars (Fang et al., 2013).

On top of BFRs and PFRs, also chlorinated (C) FRs are still present in the outdoor environment and indoors. Examples are chlorinated paraffins (CPs), although not exclusively used as FRs, and dechloranes. CPs are also used as plasticizer, as extreme pressure additives in metalworking fluids and as additives in paints (Fiedler, 2010). Fridén et al. reported that adult exposure to CPs was predominantly via inhalation, while dust ingestion was suggested to be more important for toddlers (Fridén et al., 2011). In China an increase of CP concentrations has been reported in the outdoor environment as well (Chen et al., 2011). CPs are being produced in very high volumes, more than 700,000 tons per year in China alone. These ongoing high production volumes – compared e.g. to the total global production of polychlorinated biphenyls (PCBs) ever of between 1 and 1.5 million tons – and their application as FR, and additives in plastics and paints, and their persistence may explain the significant levels found indoors (Fiedler, 2010). Dechloranes have been found in the US and Canadian environments in substantial amounts (Shen et al., 2014), and their accumulation in dolphins in the Southern Atlantic Ocean demonstrated their global distribution (De la Torre et al., 2012). Although CPs and dechloranes are both highly persistent, they have not been listed as official persistent organic pollutants (POPs) under the Stockholm Convention yet. For CPs the delay is due to analytical difficulties, although interlaboratory studies show improvements in the comparability of data from laboratories analyzing CPs (Van Mourik et al., 2015). SCCPs have recently been adopted at a meeting of the POPs Review Committee. This means that SCCPs are in a few years expected to be listed as POPs by the Stockholm Convention.

Finally, most FRs contain impurities or byproducts (Qu et al., 2013; Liu et al., 2015; Ballesteros-Gómez et al., 2015) or degrade in the environment into a variety of other products (Su et al., 2014; Altarawneh and Dlugogorski, 2014). These related compounds that until now have been studied to a lesser extent might be also of concern in relation to human health effects. Obviously, in this way a large, unexpectedly complex cocktail of substances is created in indoor environments. In 2010, Harrad et al. already pointed to the risk for human health due to exposure to BFRs and PCBs from house dust (Harrad et al., 2010). However, the situation indoors is even more complex as not only other FRs are also present but some of them have multiple functions besides flame retardancy; some are also applied as plasticizers or as additives in waxes. This dual functionality increases the number of applications these chemicals are used in, and consequently increases the total indoor exposure of humans. In addition, there are many examples of combined applications of various FRs in the same product, often together with one or more synergists (Waaijers, 2014; Hoffman et al., 2014).

The 'Future Market Insights' industry analysis report 2014–2020 confirms that FR markets will continue to grow, particularly in the Asia–Pacific region, driven by growth in the construction and automotive industries and by fire safety regulations (http://www.futuremarketinsights.com/reports/flame-retardant-chemicals-market). All FRs have their own physical-chemical characteristics and toxicity, which makes their analysis, evaluation and risk assessment complex. The FR concentrations indoors – in close proximity to the products that contain them – appear relatively high compared to outside concentrations (Table 1), even when we are looking at BFRs, CFRs and PFRs in sediment and fish from some of the most contaminated areas of the world (Great Lakes, USA/Canada), Baltic Sea (Scandinavia), Western Scheldt (Netherlands) and some Spanish and Korean rivers and estuaries. Clearly, the relatively high levels in dust stand out for all three FR groups. The low accumulation of the PFRs in fish compared to the BFRs and CFRs is striking. The higher levels of the CPs and Dechlorane Plus are remarkable and require further attention. It is known that phthalate levels in dust can be high. Hwang et al. reported 0.1–7 mg/g di-(2-ethylhexyl)phthalate (DEHP) in dust from Californian apartments (Hwang et al., 2008). Fromme et al. reported levels of various phthalates of 0.010–0.055 mg/g in German apartments with an exception of 0.775 mg/g for DEHP (Fromme et al., 2004). The CP levels reported in Table 1 are at the lower end of the range of those phthalate levels but approximate these.

2. Indoor exposure

FRs are present at high concentrations in close proximity to where we live and work, at our desks, in our computers and phones, in the upholstery we sit on, in cars as well as in the many products and materials in the buildings we spend time in (Covaci et al., 2011; Stapleton et al., 2008, 2009). Although worldwide people do not seem to spend more time indoors (15.7–15.8 h on average in Germany, Canada and the USA) (Brasche and Bischof, 2005) people do live in better insulated buildings, and are more heavily engaged with multiple electronic devices.

FRs can be emitted from the equipment and furniture through evaporation (off gassing) or abrasion (small particles breaking off from foam, textile fibers, etc.). Also, FRs can directly be taken up by particles on the surface of furniture or equipment. Consequently, the discussion of exposure of humans to FRs has suddenly got a different character. Persistent and bioaccumulative compounds in the environment often lead to dietary exposure, e.g., through consumption of fish and dairy products. But the high chemical concentrations in indoor dust and air now suggests that the major human exposure routes are inhalation, dermal contact and, especially for young children, hand-mouth contact. Hoffman et al. showed that many infant products based on polyurethane foam are treated with PFRs, including tris(1,3-dichloro-2-propyl)phosphate (TDCIPP) and TPHP. Infants may have greater exposure to these PFRs due to frequent contacts with these products (Hoffman et al., 2015). The babies studied showed higher PFR levels when more of these infant products were used in their families. The risk of dietary exposure seems still to be the greatest for the legacy BFRs and possibly some CFRs. The less persistent PFRs and halogen-free flame retardants (HFFRs) have a lower ability to bioaccumulate. Some chlorinated PFRs were found in herring from the Western Scheldt, but that area is known to be a hot spot for these chemicals, so the continuous input of new PFRs may supersede metabolism and degradation (Brandsma et al., 2015). Indoors, the situation is entirely different. Both legacy BFRs and many emerging HFFRs, CFRs and PFRs have been identified in indoor air and dust. Abdallah et al. concluded that compared to dietary and inhalation exposures, dust ingestion constitutes an important pathway of exposure to HBCD and TBBP-A for the UK population (Abdallah et al., 2008). PFRs are rapidly metabolized in humans and many cannot easily be

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