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The presence and partitioning behavior of flame retardants in waste, leachate, and air particles from Norwegian waste-handling facilities

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ABSTRACT

Flame retardants in commercial products eventually make their way into the waste stream. Herein an investigation of the presence of flame retardants in Norwegian landfills, incineration facilities and recycling sorting/defragmenting facilities is presented. These facilities handled waste electrical and electronic equipment (WEEE), vehicles, digestate, glass, combustibles, bottom ash and fly ash. The flame retardants considered included a suite of polybrominated diphenyl ethers ($\sum$BDE-10) as well as dechlorane plus, polybrominated biphenyls, hexabromobenzene, pentabromotoluene and pentabromoethylbenzene (collectively referred to as $\sum$FR-7). Plastic, WEEE and vehicles contained the largest amount of flame retardants ($\sum$BDE-10: 45,000–210,000 μg/kg; $\sum$FR-7: 300–13,000 μg/kg). It was hypothesized leachate and air concentrations from facilities that sort/defragment WEEE and vehicles would be the highest. This was generally supported for total air phase concentrations ($\sum$BDE-10: 9000–195,000 pg/m^3 WEEE/vehicle facilities, 80–900 pg/m^3 in incineration/sorting and landfill sites), but not for water leachate concentrations (*e.g.*, $\sum$BDE-10: 15–3500 ng/L in WEEE/Vehicle facilities and 1–250 ng/L in landfill sites). Results showed that landfill leachate could exhibit similar concentrations as leachate from WEEE/vehicle sorting and defragmenting facilities. To better account for concentrations in leachates at the different facilities, waste-water partitioning coefficients, K_{waste} were measured (for the first time to our knowledge for flame retardants). WEEE and plastic waste had elevated K_{waste} compared to other wastes, likely because flame retardants are directly added to these materials. The results of this study have implications for the development of strategies to reduce exposure and environmental emissions of flame retardants in waste and recycled products through improved waste management practices.

Introduction

Flame retardants include a large range of organic and inorganic chemicals that are added to polymeric materials and textiles to prevent and retard fires (Bergman et al., 2012). For these purposes, flame retardants have been produced in vast quantities. For instance, the total historic production of the group of flame retardants known as polybrominated diphenylethers

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(PBDEs) has been estimated to amount to 1.3–1.5 million tons between 1970 and 2005 (UNEP, 2010). These PBDEs are persistent, bioaccumulative, toxic, and prone to long-range transport in the environment (de Wit et al., 2010; Xiao et al., 2013). This has led to restrictions on their use, including a the ban in the EU on the penta- and octa-diphenylether mixtures (pentaBDE and octaBDE) in products from 2004 (UNEPPOP, 2006, 2007), and restrictions for pentaBDE under the UNEP Stockholm Convention (chm.pops.int).

Waste treatment facilities constitute a large repository for both banned and emerging flame retardants, and can be considered hotspots for their environmental emissions (Abbasi et al., 2014; Earnshaw et al., 2013; Leung et al., 2007; Robinson, 2009; Stubbings and Harrad, 2014). Flame retardants are added in particularly high concentrations to plastics found in electronic products, and therefore waste electrical and electronic equipment (WEEE) recycling facilities can have particularly high concentrations (Sjödin et al., 2001; Wong et al., 2007), unless precautionary actions are taken (Thuresson et al., 2006). It follows that the recycling of WEEE and plastics may result in a reintroduction of banned flame retardants into commercial products. Recently, several types banned flame retardants were identified in black polymeric food-contact products on the European market, with the most likely source being recycled WEEE (Puype et al., 2015; Samsonek and Puype, 2013).

In addition to emissions from WEEE recycling facilities, brominated flame retardants (BFRs) such as PBDE have been reported to be emitted from waste incinerators, landfills, and water treatment plants (WTPs). PBDE along with minor levels of polybrominated biphenyl (PBB) flame retardants have been quantified in flue gases and in the vicinity of a municipal solid waste incinerator (L.-C. Wang et al., 2010; M.-S. Wang et al., 2010b). Several studies have reported that landfills can contain large accumulated amounts of BFRs, which can be emitted via water leachate (Arp et al., 2011; Nyholm et al., 2013; Odusanya et al., 2009; Osako et al., 2004) and through air emissions (Weinberg et al., 2011). In a previous Norwegian study, PBDEs as well as several emerging BFRs, such as hexabromobenzene (HBB), pentabromotoluene (PBT), pentabromoethylbenzene (PBEB), and 1,2-dibromo-4-(1,2-dibromoethyl) cyclohexane (TBECH), were identified near a metal recycling station and a landfill (Arp et al., 2011; Nyholm et al., 2013). Several PBDEs have been quantified in outgoing water from WTPs, with key sources being indoor dust, textiles, human excretion, and industrial discharges (Melymuk et al., 2014; Nyholm et al., 2013; Vogelsang et al., 2006). WTPs, along with tributary runoff were one of the largest sources of PBDE emissions to Lake Ontario from the Toronto area (Melymuk et al., 2014).

In order to obtain a more systematic understanding of the sources and mechanisms regulating emissions of BFRs and other flame retardants at recycling, incineration and landfilling waste-facilities, this study presents a comprehensive field campaign to measure the presence of various flame retardants at 12 different facilities in Norway. The waste categories found at these facilities comprised of plastic, WEEE, vehicle fluff, combustible waste sorted for incineration, glass, bottom ash, fly ash and digestate. Leachate and air samples from these sites were sampled over the summer, autumn and spring seasons.

This study focused on ten PBDEs: 244′-tribromodiphenylether (BDE-28), 22′44′-tetrabromodiphenylether (BDE-47), 22′44′5-pentabromodiphenylether (BDE-99), 22′44′6-pentabromodiphenylether (BDE-100), 22′44′55′ hexabromodiphenylether (BDE-153), 22′22′56′-hexabromodiphenylether (BDE-154), 22′344′5′6-heptabromodiphenylether (BDE-183), 22′33′44′56-octabromodiphenylether (BDE-196), 22′33′44′55′6-nonabromodiphenylether (BDE-206), and 22′33′44′55′66′-decabromodiphenylether (BDE-209), and seven other BFRs: HBB, PBT, PBEB, 22′44′55-hexabromobiphenyl (BB-153), decabromobiphenyl (BB-209), and two isomers of dechlorane plus (DP): DP-syn and DP-anti.

The European Food Safety Authority has indicated HBB as a compound of potential concern due to its bioaccumulation potential (EFSA, 2012) and it has recently been detected in air masses over the North Sea (Moller et al., 2012b). BB-153 was banned for use as flame retardants after the Michigan farm incident in 1973 (Chanda et al., 1982), and production of BB-209 ceased in Europe by 2000 (De Wit, 2002). Both BB-153 and BB-209 have recently been discovered in the blood of indoor cats (Norrgran et al., 2015), have been found to be formed during incineration of municipal solid waste (L.-C. Wang et al., 2010; M.-S. Wang et al., 2010) and are together with PBDEs regulated as BFRs in electronic products by the RoHS directive (ec.europa.eu). In addition, BB-153 is listed under the UNEP Stockholm Convention on POPs (chm.pops.int). DP is still produced in large quantities, and has been shown to be ubiquitous in the environment, with elevated concentrations near WEEE (Guerra et al., 2011; Moller et al., 2012a; Sverko et al., 2011; Tomy et al., 2007; Xiao et al., 2013).

Due to the large amounts of flame retardants added to plastic and WEEE materials (Ortuno et al., 2015; Tamade et al., 2002; Wong et al., 2007), the two central hypotheses of the study were: (1) plastic-containing waste, such as plastic WEEE and vehicle waste fractions, have elevated flame retardant concentrations compared to other types of waste sampled in this study (combustibles, glass, ash, digestate); (2) concentrations in the leachate and air from WEEE and vehicle defragmentation and sorting sites are substantially larger than from landfills and waste incinerators. To better account for the link between solid concentrations and concentrations in leachates, and incidentally the link between these two hypotheses, waste-water partitioning coefficients, K_{waste}, were measured for the sampled waste fractions. Many of the data reported in this study are presented for the first time.

1. Materials and methods

1.1. Chemicals

The flame retardants studied included the PBDEs (BDE-28, -47, -99, -100, -153, -154, -183, -196, -206 and -209), PBT, PBEB, HBB, BB-153, BB-209, and DP-syn and DP-anti. Their physical chemical properties are presented in Appendix A-Table S1. The sum of the BDE compounds are here referred to as $\sum$BDE-10 (all 10 congeners) or $\sum$BDE-6 (congeners 28, 47, 99, 100, 153 and 154). The sum of the other flame retardants are referred to as $\sum$FR-7.

Analytical standards (^{12}C) of the PBDEs and the brominated benzenes (BBs, comprising HBB, PBT and PBEB) were purchased

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