



Brominated flame retardant emissions from the open burning of five plastic wastes and implications for environmental exposure in China[☆]



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ABSTRACT

Based on the most widely used plastics in China, five plastic wastes were selected for investigation of brominated flame retardant (BFR) emission behaviors during open burning. Considerable variations were observed in the emission factors (EF) of polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) from the combustion of different plastic wastes. Distribution of BFR output mass showed that Σ PBDE was emitted mainly by the airborne particle (51%), followed by residual ash (44%) and the gas phase (5.1%); these values for Σ HBCD were 62%, 24%, and 14%, respectively. A lack of mass balance after the burning of the plastic wastes for some congeners (output/input mass ratios > 1) suggested that formation and survival exceeded PBDE decomposition during the burns. However, that was not the case for HBCD. A comparison with literature data showed that the open burning of plastic waste is major source of PBDE compared to regulated combustion activities. Even for state-of-the-art waste incinerators equipped with sophisticated complex air pollution control technologies, BFRs are released on a small scale to the environment. According to our estimate, Σ PBDE release to the air and land from municipal solid waste (MSW) incineration plants in China in 2015 were 105 kg/year and 7124 kg/year. These data for Σ HBCD were 25.5 and 71.7 kg/year, respectively. Considering the fact that a growing number of cities in China are switching to incineration as the preferred method for MSW treatment, our estimate is especially important. This study provides the first data on the environmental exposure of BFRs emitted from MSW incineration in China.

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1. Introduction

Due to the widespread use of plastics, the amount of plastic waste generated globally has grown dramatically in recent years (Thompson et al., 2009). As a consequence, substantial quantities of plastic waste have accumulated in the natural environment. For example, approximately 32, 25 and 15 million tons of plastic waste were generated in the United States (US) in 2012, the European Union (EU) in 2012, and China in 2011, respectively (US EPA, 2014; Velis, 2014). Incineration for energy recovery is a traditional and valuable means of plastic waste disposal with the advantage of being highly effective in reducing the volume of plastic waste (Li et al., 2001). Moreover, incineration of municipal solid waste (MSW) is addressed globally under the framework of the

Stockholm and Basel Conventions for reduction of the release of produced persistent organic pollutants (Van Caneghem et al., 2010). Unfortunately, not all the wastes are disposed with state-of-the-art incinerators, and there is still significant combustion of domestic waste in open piles, barrels, fireplaces, household heating stoves, or primitive incinerators, even in developed countries (Zhang et al., 2011). A large number of studies on the emissions of dioxin and dioxin-like persistent organic pollutants from the incineration of MSW have been published (UNEP, 2013). By contrast, investigation on the emission behavior of brominated flame retardants (BFRs) from MSW combustion is rather limited to date. With limited technologies presently able to feasibly recycle plastic material while removing toxic BFRs, destruction by means of burning is a major recourse (Buekens and Yang, 2014).

BFRs are a class of synthetic organic substances and have been used extensively in many plastic consumer products (Tang et al., 2014). Among them, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) are of very high concern because of their persistent, bioaccumulative and toxic properties

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(Birnbau and Staskal, 2004; Johnson-Restrepo et al., 2005; Naert et al., 2006; Zhou et al., 2002). As additives, PBDEs and HBCDs are physically mixed into the products rather than being chemically bound. Therefore, they can be released into the ambient environment from the products. Although some BFRs have been or are being phased out of plastic production, the massive quantities of plastics containing these compounds already in circulation around the world will continue to be a problem for the foreseeable future (Wyrzykowska-Ceradini et al., 2011). There are many publications on BFRs derived from plastic products in the environment, but these studies mainly focused on occurrences of BFRs in the ambient environment of plastic waste recycling sites (Song and Li, 2014; Sun et al., 2012; Zhang et al., 2012; Zheng et al., 2012). Few studies have published results for PBDE emission from the burning of domestic waste (BFR-laden or not) in incinerators (Borgnes and Rikheim, 2004; Van Caneghem et al., 2010; Wyrzykowska-Ceradini et al., 2011) or in open fires (Gullett et al., 2010). However, BFRs emission behavior during open burning of domestic waste, especially for plastic waste has not been studied adequately.

The plastic waste issue is even more prominent in China because China receives 56% (by weight) of the global imports of waste plastics (Velis, 2014). From 2006 to 2012 alone, the plastic waste imports to China increased from 5.9 to 8.9 million tons, which worsened an already grave situation. Thus, the plastic waste problem in China may be a microcosm of the global situation. The present study aims to characterize the emission behavior of BFRs during the open burning of plastic wastes and to assess mass emission of BFRs from MSW incineration, usually containing approximately 10% plastic waste by weight in China (Yang et al., 2012). The data presented in this study are the first measurements of BFRs emitted from the open burning of five types of plastic waste in three emission forms (gas phase, airborne particle, and residual ash). The present study provides information on the survival, transformation, and destruction of PBDEs and also on the formation of PBDEs during the burning of plastic waste. This study also provides the first data on the environmental exposure to BFR emitted from MSW incineration in China by release to air via the gas and airborne particle and to land via residual ash.

2. Materials and methods

2.1. Experimental setup

Forty-five test combustions of plastic waste in open fires were conducted. The experimental setup, shown schematically in Fig. S1 in the Supplementary data (SD), consisted of a 60-mm diameter, 500-mm length, tubular quartz combustion chamber that contains an asbestos gasket bed for the plastic samples. The reactor was heated with an alcohol blast burner. The majority of airborne particle samples were gathered by a fiberglass membrane, and the gas phase samples were collected by the polyurethane foam after cooling with a water jacket. The residual ash samples were left on the asbestos gasket bed. The purging system can generate negative pressure to promote gas phase and airborne particle flow upwards easily.

2.2. Plastic wastes

The uses of BFRs differ by polymer type, depending on how that specific polymer product is intended to be used. Unfortunately, it is impossible to measure thousands of different plastic products being used in daily life. To understand the emission behavior of BFRs from the burning of plastic waste, five types of plastic used most frequently in consumer products were chosen to conduct this preliminary study. The five types of plastic wastes, identified

mainly according to the product type, were collected at two plastic waste recycling sites in Shenzhen, China. At the same time, burn tests were also used to identify plastic materials in the present study (<http://www.boedeker.com/burntest.htm>). Plastic bottles, corrugated pipe, and children's toys were collected as polyethylene (PE) wastes; either rigid or foam disposable cups and frozen packing materials were made from polystyrene (PS). Acrylonitrile butadiene styrene (ABS) waste originated from electronic products, such as housing and casting for telephones, flat screen televisions, and household appliances; polypropylene (PP) wastes were from machine cover shells, lunch boxes, and plastic bailers, etc. The polyvinylchloride (PVC) waste consisted mainly of cable sheath, wire jackets, and pieces of pipe. More information on the plastic waste samples is provided in SD Table S1.

2.3. Combustion test

Prior to the combustion tests, large pieces of plastic waste were cut into pieces a few centimeters in size. Plastics used in more than one test were divided in advance to ensure a fairly uniform composition. The wastes to be combusted were placed loosely in the bed to allow air to flow through it. The plastic waste was set on fire with an alcohol blast burner to avoid open fires, which burn at relatively low temperatures leading to incomplete combustion. All the tests were performed in May 2014 with ambient temperatures of 25–35 °C. The inner wall of the apparatus was cleaned between burns with acetone. To further minimize contamination of equipment from previous combustion tests, the tests were generally performed in order of expected increasing emission generation, i.e., from PP wastes (lunch boxes or cups containing less BFRs) to domestic waste (bags or corrugated pipe) to plastic in electronic waste. For each plastic waste, three types of consumer products were chosen as samples with three repetitions. The flame temperature of the alcohol blast burner was approximately 850 °C–1000 °C. A preliminary experiment indicated that 5 g of plastic waste ensures detectable levels of target compounds in combustion products. The combustion time was set at 3–5 min for each 5 g of plastic waste to ensure complete combustion. A total of 135 samples (three categories: gas phase, airborne particle and residual ash) were collected after combustion of all the plastic wastes. All samples were stored in aluminum foil at –4 °C until further treatment.

2.4. Analysis

Plastic waste samples were cut in pieces and combustion product samples were Soxhlet extracted with a mixture of 200 mL dichloromethane and n-hexane (3:1, v:v) for 24 h. Prior to sample extraction, a known amount of recovery surrogate standards was added (see SD for details). The extracts were concentrated to 2 mL with a rotary evaporator. The extracts were purified and fractionated through a glass column packed with silica gel/alumina. From the bottom to top, the packed silica/alumina column consists of neutral alumina (6 cm, 3% deactivated, w:w), neutral silica gel (2 cm, 3% deactivated, w:w), 33% sodium hydroxide silica (5 cm), neutral silica gel (2 cm, 3% deactivated, w:w), 44% sulfuric acid silica (6 cm) and anhydrous sodium sulfate (1 cm). The extracts were eluted with 70 mL of hexane: dichloromethane (1:1, v:v), and this eluent was concentrated to a final volume of 500 µL by rotary evaporation and a gentle N₂ stream. Prior to instrumental analysis, an internal standard (provided in the SD) was added to the extracts. Before analysis for HBCD, extracts were exchanged into methanol for instrumental analysis.

Fourteen PBDE congeners (BDE28, 47, 49, 85, 99, 100, 138, 153, 154, 183, 196, 206, 208, and 209) and three HBCD congeners (α -, β -,

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