Chemosphere xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Leaching of brominated flame retardants from mixed wastes in lysimeters under conditions simulating landfills in developing countries

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HIGHLIGHTS

- Waste was aged in lysimeters under aerobic, semi-aerobic, and anaerobic conditions.
- Leachate PBDE, TBBPA, TBP, and HBCD concentrations were analyzed for 3.5 years.
- BFRs leached from mixed wastes with high organic matter and moisture content.
- BFRs leached more readily under anaerobic conditions than under aerobic conditions.
- Supplying air to landfills may reduce BFR elution into the environment.

ARTICLE INFO

Article history:
Received 15 October 2013
Received in revised form 10 January 2014
Accepted 11 January 2014
Available online xxxx

Keywords:
Brominated flame retardant
Elution
Landfill
Leachate
Aerobic
Anaerobic

ABSTRACT

In developing countries, wastes are usually not separated before being disposed of in solid-waste landfills, most of which are open dumps without adequate measures to prevent environmental pollution. To understand the leaching behavior of brominated flame retardants (BFRs) from waste consumer products in landfills, we have been conducting a long-term landfill lysimeter experiment since 2006 under conditions designed to mimic three types of landfill conditions in developing countries: aerobic, semi-aerobic, and anaerobic. Pilot-scale lysimeters (60-cm i.d.) were filled with a 400-cm layer of mixed wastes consisting of 35 wt% food, 20 wt% paper, 20 wt% paper pulp, 13 wt% plastic, 10 wt% wood chips, 1 wt% glass, and 1 wt% metals, proportions that are typical of unsorted municipal solid waste in Asian developing countries. In the present study, we determined the concentrations of polybrominated diphenyl ethers, tetrabromobisphenol A, tribromophenols, and hexabromocyclododecanes in leachate samples collected from the lysimeters during the first 3.5 years of the experiment, to evaluate BFR elution behavior in early-stage landfills. Under all three conditions, BFR elution started at the beginning of the experiment. The BFR concentrations in the leachates from the aerobic lysimeter tended to be lower than those from the anaerobic lysimeter, suggesting that the presence of air inside landfills considerably reduces BFR elution to the surrounding environment. During the 3.5-year experiment, BFR outflow from the lysimeters was only 0.001–0.58% of the total BFRs in the loaded waste; that is, most of the BFRs in the waste remained in the lysimeters.

1. Introduction

Brominated flame retardants (BFRs) are synthetic additives widely used in plastics, textile coatings, and electronic appliances to reduce their flammability. However, despite the benefits of BFRs, their persistence, bioaccumulation, and possible adverse effects on wildlife and humans, even in remote areas, are cause for concern (Ikonomou et al., 2002; Birnbaum and Staskal, 2004; Ueno et al., 2004). Among the BFRs available commercially, polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyls are regulated by the Restriction of Hazardous Substances (RoHS) Directive (Directive 2002/95/EC), which entered into force in 2006 in the European Union. In May 2009, tetra- to hepta-BDEs were listed

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http://dx.doi.org/10.1016/j.chemosphere.2014.01.025
0045-6535/

Please cite this article in press as: Kajiwara, N., et al. Leaching of brominated flame retardants from mixed wastes in lysimeters under conditions simulating landfills in developing countries. Chemosphere (2014), http://dx.doi.org/10.1016/j.chemosphere.2014.01.025

as persistent organic pollutants (POPs) under the Stockholm Convention (Annex A), and hexabromocyclododecanes (HBCDs) were listed in May 2013 (Stockholm Convention, 2013). Despite these restrictions, large amounts of consumer products (including upholstered furniture and electrical and electronic equipment) and building materials containing BFRs of concern are still in use and must be disposed at the end of their lifetimes. Disposal of these items creates outdoor reservoirs (e.g., landfills, wastewater treatment plants, electronic waste recycling facilities, and stockpiles of hazardous wastes) for the future dispersal of BFRs to the environment. Moreover, large volumes of these materials are in the global recycling flow and will continue to be used in consumer products for a considerable time. Therefore, even though the use of BFRs of concern have been phased out in new products, special attention should be paid to their potential for emission to the environment by means of material recycling and the disposal of end-of-life products.

Leaching from final disposal sites is thought to be a major pathway for BFR release to the environment. So far, however, the number of studies of BFR leaching behavior in and around actual landfill sites is limited (Osako et al., 2004; Odusanya et al., 2009; Oliaei et al., 2010; Weber et al., 2011; Daso et al., 2013; Kwan et al., 2013). Because reclamation of a landfill site takes several decades, long-term studies of the leachability of BFRs from waste and of the fate and transport of BFRs in landfill sites are required. This is particularly true in developing countries, where garbage is usually not separated, and everything is thrown away together in solid-waste landfill sites, most of which are open dumps without adequate measures to prevent environmental pollution. Leaching tests have shown that although PBDEs have low solubility in water, they nevertheless leach from treated products, and their leachability from flame-retardant polymers is remarkably enhanced when methanol or dissolved humic solution is used instead of distilled water (Kim et al., 2006; Choi et al., 2009). These results indicate that the organic matter content of landfill waste has important consequences for BFR elution.

Landfill structures can be broadly separated into three categories: anaerobic, semi-aerobic, and aerobic, depending on the amount of air introduced into the waste layer (Aerobic landfills include mechanical aeration system). Semi-aerobic landfill systems were developed more than 30 years ago at Fukuoka University and have since been introduced all over Japan (Matsufuji, 2004), as well as in some developing countries (Chong et al., 2005). The main structure of semi-aerobic landfill systems is the leachate collection pipe, which is placed under the bottom layer of the landfill and is surrounded by pebbles. The most important functions of this pipe are to drain leachate from the waste layer and to bring air into the waste layer. The biodegradation of waste organic matter can produce heat and increase the temperature of the waste layer, and as a result, air can enter the landfill body naturally by heat recirculation. This landfill technology, often referred to as the Fukuoka method, is the first method accredited by the United Nations Framework Convention on Climate Change to control methane emissions at landfill sites (UNFCCC, 2013). Many studies suggest that the quality of leachate characteristic is low in anaerobic landfills; these landfills are more like open dumps, and they require more time for waste stabilization than do aerobic and semi-aerobic systems (Matsufuji, 2004; Aziz et al., 2010). However, the leaching behavior of hazardous substances from landfills has been less investigated. There have been lysimeter studies of the leaching of mercury from used batteries (Yanase et al., 2004, 2009) and lead from electronic waste (Spalvins et al., 2008) in landfills, but no study of BFR leaching with a focus on landfill conditions in developing countries has been conducted.

Since 2006, we have been conducting a long-term landfill lysimeter experiment under conditions designed to mimic three types of landfill conditions in developing countries: aerobic, semi-aerobic, and anaerobic. Our goal is to understand the leaching of BFRs from waste consumer products dumped together with waste containing a large amount of organic matter and having a high moisture content. In the present study, we determined the concentrations of PBDEs, tetrabromobisphenol A (TBBPA),

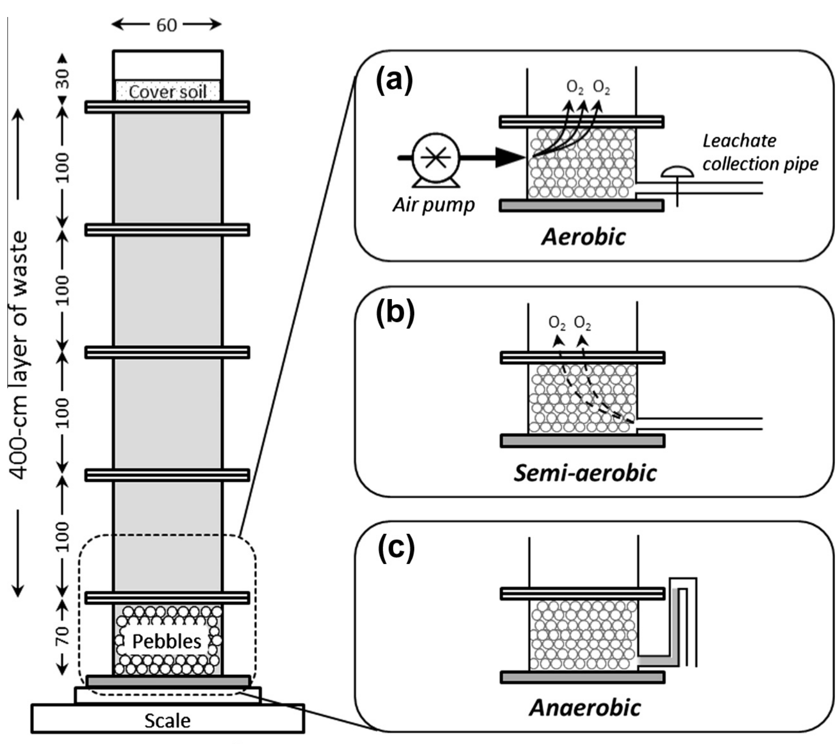

Fig. 1. Schematic of pilot-scale lysimeters used in this study.

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