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Overview on relative importance of house dust ingestion in human exposure to polybrominated diphenyl ethers (PBDEs): International comparison and Korea as a case

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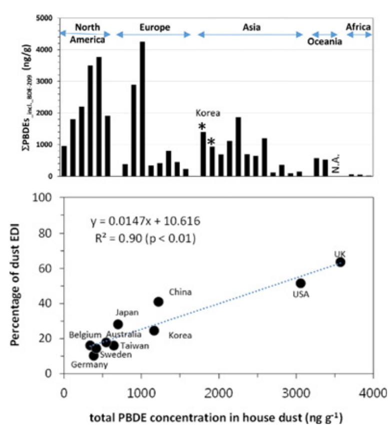
HIGHLIGHTS

- International difference in relative importance of house dust ingestion were observed.
- PBDE levels in house dusts were strongly linked to GDP of the countries.
- House dust ingestion was less important in most countries than diet ingestion.
- House dust can be a good monitoring matrix to screen the effect of regulation.

GRAPHICAL ABSTRACT



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ABSTRACT

Human exposure studies to polybrominated diphenyl ethers (PBDEs) have reached different results about the relative importance of diet intake and house dust ingestion. In the present study, concentrations of PBDEs in Korean house dust ($n = 15$) from geographically different cities were measured, which were in agreement with a previous result, and compared with those for 22 countries of five continents collected from the most recent scientific literature. Compared with other exposure pathways, diet intake was the most important contributor to total PBDEs exposure of Korean adults (i.e., 71% of overall intake). On global comparison, total PBDE levels in house dust differed by two to three orders of magnitude among the countries investigated, with a significant relationship with gross domestic product (GDP). Whereas, dietary daily intakes exhibited a narrow difference within one order of magnitude worldwide and no relationship with GDP. Consequently, the relative importance of major two pathways depended on the contamination extent of PBDEs in house dust, which may be associated with the amount of PBDE products in use. In most countries except for UK and USA, the contribution of house dust ingestion was less important than diet intake in the current and are expected to much more mitigate in the future. However, how fast the effect of regulation will be reflected to house dust and human exposure is necessary to be monitored steadily.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs), one class of the most widely used brominated flame-retardants (BFRs), have been used in a wide array of products, including electronics, furnishings, building materials, motor vehicles, textiles, plastics, paints, polyurethane foams, and textiles since 1970s. They were commercially produced as three technical mixtures as penta-BDE, octa-BDE, and deca-BDE (Alaee et al., 2003). In 2001, the market demands of total PBDEs technical products were 67,390 metric tons in globe and the percentage of America, Europe, and Asia were 95% vs. 2% vs. 3% for penta-BDE, 40% vs. 16% vs. 44% for octa-BDE, and 44% vs. 13% vs. 43% for deca-BDE (BSEF, 2003). Global market demand of deca-BDE was 66,677 metric tons in 2002 and 56,418 metric tons in 2003 (de Wit et al., 2010).

Animal studies have shown their neurotoxicity, developmental and reproductive toxicity, and endocrine disturbance (Kodavanti et al., 2005). In human, PBDE exposure has exhibited the direct association with altered thyroid hormone levels (Herbstman et al., 2008; Chevrier et al., 2010), neurobehavioral development in children (Eskenazi et al., 2013; Chao et al., 2014a), and the fertility reduction (Harley et al., 2010). Because of their toxicity and persistence, penta-BDE and octa-BDE were voluntarily banned in North America and the Europe in 2004 (Bett, 2008), and later were listed as persistent organic pollutants (POPs) by the Stockholm Convention in 2009 (Stockholm Convention secretariat, 2009). The use of deca-BDE, which accounts for ~83% of the total PBDE production worldwide (BSEF, 2009), has been restricted in electrical and electronic applications in the Europe since 2008 and was phased out of all applications in North America (Canada and USA) at the end of 2013 (BSEF, 2012, 2014). In Asia, however there are currently limited regulatory restrictions on the use of deca-BDE (BSEF, 2014).

For example, the BFR market in Korea has increased annually by 13.5% and the consumption of PBDEs, accounting for 25% of the total consumption of BFRs, was estimated to be 12,408 tons in 2002 (Kim et al., 2007). In response to growing concerns, the Korean government began the regulation in 2008 by establishing the PBDE content limit to be <0.1% in the electrical and electronic equipment under the Korea Restriction of Hazardous Substances Directives (RoHS). Furthermore, in 2011 PBDEs were listed in the Persistent Organic Pollutants (POPs) Control Act, which is the guideline for the control of POPs in Korea (Kim and Yoon, 2014). Nevertheless, old products manufactured before the regulation was implemented can still contain significant amounts of PBDEs and then from which more amounts of PBDEs may have entered Korean household environment. For example, the amount of PBDEs introduced into Korean households from newly manufactured TVs and computer monitors, for which obsolete plastics were recycled, were estimated to be 1.87 and 0.28 tons in 2011, respectively (Lee et al., 2015). A chronological trend of PBDEs exposure did not show any reduction in Korean human breast milk, even though a slight decrease in foodstuffs has been observed over time (Kim and Yoon, 2014). This can be evidence for the increasing contribution of non-diet exposure pathways to overall intake. However, most studies for Korean exposure to PBDEs has focused to dietary intake alone (Na et al., 2013; Nguyen et al., 2014) and some studies including inhalation or dust ingestion were also limited to residents in only one city (Lee et al., 2013) or elementary school students (Lim et al., 2014).

The health hazards of these emerging chemicals have attracted increasing scrutiny for human exposure and risk assessment. Globally, geographical differences in internal human exposure of PBDEs have been found with generally much higher levels in North America than in Europe and Asia (Lorber, 2008; Besis and Samara, 2012). These differences may not solely be explained by one exposure pathway. Dietary intake has been suggested to be the predominant route of exposure for PBDEs (Fraser et al., 2009; Fromme et al., 2009; Linares et al., 2015), while other studies reported the significant contribution of non-diet exposure routes including inhalation of indoor air (Allen et al., 2007) and ingestion of house dust (Lorber, 2008). Particularly, house dust ingestion accounted for 82% of the overall intake for USA adults (Lorber, 2008). Levels of PBDEs in house dust were also observed to be strong correlations with those in the hairs of the residents (Kang et al., 2011), human serum (Johnson et al., 2010), both maternal and umbilical cord plasma (Frederiksen et al., 2010), and breast milk (Wu et al., 2007).

Significant amounts of PBDEs, physically mixed into consumer products, may be released into indoor environment via volatilization (for lower brominated congeners) or abrasion (for higher brominated congeners) (Rauert and Harrad, 2015). This can cause markedly higher PBDE concentration in indoor than outdoor environment (Besis and Samara, 2012; Yu et al., 2012). Considering a number of potential PBDEs sources in indoor, the relative importance of indoor exposure to PBDEs is growing, which is different from the cases for legacy POPs such as dioxins, PCBs, and organochlorine pesticides. As shown in previous studies mentioned earlier and Besis and Samara (2012), however there were distinct differences among some countries in the relative contribution of indoor exposure to human exposure of PBDEs but the reason remains still unclear.

In this study, we tried to reveal why international differences in the relative importance of non-dietary exposure (particularly, house dust ingestion) occur and by which such differences can be traced. For that, we compiled PBDEs data measured in house dust of 22 countries of five continents and found a good correlation between house dust-contained PBDEs and its relative contribution to overall intake or economic development level, which is expressed by gross development product (GDP). As a case study, furthermore we determined PBDE levels in Korean house dust samples and dust-ingested PBDE intake. Daily intake of house dust-ingested PBDEs were compared with those of inhalation and dietary ingestion which were compiled from the literature to estimate the overall intake via multiple pathways and the relative contribution of each pathway in Korea.

2. Methods and materials

2.1. Sample collection

Indoor dust samples were collected from fifteen homes of geographically different nine cities ($n = 3$ in Seoul, $n = 3$ in Daegu, $n = 3$ in Ansan, each $n = 1$ in Busan, Jeonju, Kwangju, Cheongju, Jeju and Chungbuk) and one university library (in Seoul) in May 2009, Korea. Those cities represented densely populated cities (Seoul, Busan, and Kwangju; over 1,500,000 capita), cities bearing big industrial complex (Ansan and Daegu), medium-sized cities having population of 600,000 to 850,000 capita (Jeonju and Cheongju), and rural area (Jeju and Chungbuk). In all homes, house dust samples were collected using household vacuum cleaner from the floor of main living areas, including the bedroom and living room. Volunteers collected the house dust for 3 weeks at each home using their own household vacuum cleaner and we gathered the house dust collected in each vacuum cleaner bag. A non-residential indoor dust sample from library was collected on the tops of shelves (an area of 30 × 30 cm) using a pre-cleaned stainless plate. All dust samples obtained were transferred to glass jars pre-cleaned by methylene chloride, acetone and hexane, transported to the laboratory, and stored at −18 °8 until further treatment. Before extraction, each of dust samples was passed through a pre-cleaned stainless sieve (mesh size = 500 μm) to obtain the homogenized dust particles from which other non-dust particles, such as hairs and plastic garbage, were removed.

2.2. Analysis of PBDEs in indoor dust

For PBDEs, sieved dust samples of approximately 1 g were Soxhlet extracted for 18 h using 200 ml of dichloromethane and hexane (3:1, v/v) after homogenized with anhydrous sodium sulfate. Before extraction, 2–10 ng of ^{13}C-labeled PBDE congeners as surrogate standards