



# Pollution characteristics and sources of polybrominated diphenyl ethers in indoor air and dustfall measured in university laboratories in Hangzhou, China

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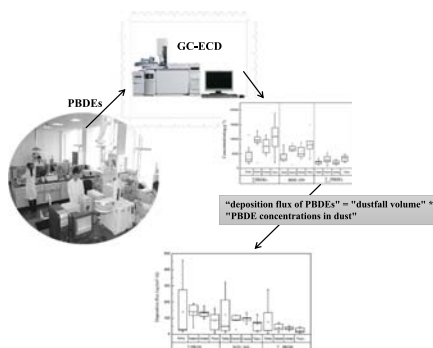
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## HIGHLIGHTS

- PBDE concentrations in air and dust in laboratories in four seasons were observed.
- The highest PBDE concentrations in air and dust were found in the winter.
- BDE-209 is accounted for 70% of the  $\sum$  PBDEs in indoor dustfall, but 50% in indoor air.
- There are correlation between deposition flux of PBDEs with appliances and furniture.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Laboratories at Zhejiang University of Technology in Hangzhou were selected to ascertain the pollution status of polybrominated diphenyl ethers (PBDEs) in indoor city air. Air and dustfall samples were collected during 2015 and analysed using gas chromatography. This study provides important information on the typical indoor pollution of brominated flame retardants in urban areas in China, particularly in university laboratories. The annual geometric mean concentration of PBDEs of passive air samples in the university laboratories was 545.64  $\text{pg}/\text{m}^3$ , corresponding to a medium level of air pollution. The highest level was recorded in winter and the lowest in summer. The average concentration of PBDEs in dustfall was 8198.69  $\text{ng}/\text{g}$ , generally higher than that reported in other studies. The annual geometric mean of deposition fluxes of PBDEs was 121.96  $\text{ng}/(\text{m}^2 \text{d})$ . BDE-209 was the main congener of PBDEs in all areas studied, and BDE-47, BDE-66, and BDE-99 also made a significant contribution to the total concentration of PBDEs. The detection rates were 100% for BDE-17, -28, and -66 in air samples, while the detection rates of BDE-17, -28, -71 and -66 were all 100% in dustfall samples. PBDE concentrations and deposition fluxes were unevenly distributed and showed sampling point differences, mainly attributed to differences in the materials used for indoor furnishings, the function of the room, the ventilation systems and the surrounding environment. Seasonal variations of PBDE concentrations in air samples ranged as winter > autumn > spring > summer, while seasonal variations in dustfall samples were winter > summer > autumn > spring. Two significant positive linear relationships were found between the total quantity of electronic equipment ( $r = 0.722$ ,  $p < 0.05$ ) and the quantity of plywood desktop ( $r = 0.745$ ,  $p < 0.05$ ) with the deposition fluxes of PBDEs.

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## 1. Introduction

Polybrominated diphenyl ethers (PBDEs), a type of brominated flame retardant (BFR), have been widely used due to their low price and good flame retardant effects. At present, there are three main commercial products of PBDEs with wide industrial applications: penta-BDE, octa-BDE and deca-BDE (Toms et al., 2009). PBDEs started being used in the 1960s. In 1981, PBDEs were identified as environmental pollutants, and studies of PBDEs in the environment followed thereafter (Li et al., 2015; Li et al., 2014; Zheng et al., 2011). Scientists in Sweden, Japan and the United States (Currier et al., 2015; Gassmann et al., 2014; Liu et al., 2011) found that PBDEs are not only biologically cumulative but also cause endocrine disruptors. In 2004, the European Union, followed by North America, started a comprehensive ban on the production and use of penta-BDE and octa-BDE. Two of America's largest manufacturers of deca-BDE announced the cessation of production and sales of deca-BDE commercial products by the end of 2012. In 2013, the EPA also announced a comprehensive ban on the use of commercial products containing deca-BDE (Covaci and Dirtu, 2008). Although the production and use of products treated with PBDEs have gradually been banned, the impact of PBDEs on the environment and biology does not cease immediately (Fromme et al., 2016). Products treated with PBDEs that were produced in the past 20 years are still in use. Therefore, PBDE research is still very relevant.

PBDEs can enter the environment in a variety of ways, including emissions during production, use, and recycling of the products treated with PBDEs. Due to the lack of strong chemical bonds between the materials and PBDEs, PBDEs are released easily from the material into the environment, resulting in environmental pollution. Many studies indicate that PBDE pollution is more serious in indoor air than outdoors (Afafe and Martincigh, 2015; Ding et al., 2016; Kefeni and Okonkwo, 2014). PBDE concentration in indoor air is higher than in outdoor air by 1–2 orders of magnitude (Huang et al., 2010), indicating potential interior sources of PBDE pollution. On the one hand, the indoor environment is complex and variable, and on the other hand, the indoor environment is connected by air convection with the outdoor atmosphere, in which many additional potential pollution sources of PBDEs exist. Electrical appliances, furniture, and interior decoration materials (such as curtains, paint, etc.) with added flame retardants may be sources of PBDE pollution indoors, with electronic appliances being reported as the main indoor pollution source of PBDEs (Chen et al., 2008). Indoor concentrations of PBDEs are influenced by both the pollution source (e.g., electrical appliance, furniture) and the environment (e.g., temperature, ventilation). At present, little research has been reported on the identification of PBDE sources. Rauert and others conducted preliminary studies in environmental simulation chambers, which

did not explicitly consider the sources, and processes of transformation and elimination of indoor PBDEs (Rauert et al., 2014). It is important to monitor and control semivolatile organic pollutants such as PBDEs at the source.

University laboratories are important bases of personnel training and scientific research. These laboratories often present a closed, crowded environment, with many pieces of large electronic equipment and regular use of toxic substances during experimental processes, all of which constitute environmental pollution sources. Students and teachers spend an average of 8 h in the laboratory, in which the poor air quality can affect the health and productivity of teachers and students. Extensive research from the bedroom and office has been reported, while little research has been reported from the laboratory. Therefore, for this research laboratories were chosen as sites for investigation.

This paper focuses on the analysis of indoor sources of PBDEs and the mechanisms of PBDE pollution formation and deposition. This study, conducted in university laboratories in Hangzhou, is based on theoretical and empirical analyses and statistical methods. The objectives of this paper were to (i) determine the pollution patterns of PBDEs in air and dustfall from university laboratories; (ii) identify pollution sources; and (iii) comprehensively assess the contributory factors to indoor pollution.

## 2. Materials and methods

### 2.1. Chemicals

A standard mixture of 14 PBDEs (including BDE-17, -28, -47, -66, -71, -85, -99, -100, -138, -153, -154, -183, -190, -209) was purchased from Accustandards, Inc. (New Haven, CT, USA). Dichloromethane (DCM) and *n*-hexane were obtained from the Lingfeng Chemical Reagent Co., Ltd. (Shanghai, China). Acetone was obtained from the Shuanglin Chemical Reagent Factory (Hangzhou, China). *n*-Hexane (chromatographically pure) was obtained from the TJSIELD Co., Ltd. (Tianjing, China). Silica gel of 200–300 mesh (Haiyang Chemical, Qingdao, China) was activated at 180 °C for 2 h. Concentrated sulfuric acid (XiLong Chemical Co., Ltd., Shantou, China) was used to prepare acidic silica gel: 44 g of concentrated sulfuric acid was added to 100 g of the silica gel. Anhydrous sodium sulfate (Lingfeng Chemical Reagent Co., Ltd.) was baked at 400 °C for 4 h. A glass fibre (GF) filter (90 mm), which was purchased from Staplex Air Sampler Division (Brooklyn, NY, USA), was baked at 450 °C for 12 h on a resistance furnace. Polyurethane foam (PUF), which was purchased from Safelab (Beijing, China), was stored for 24 h before Soxhlet extraction with 200 mL of 1:1 (v/v) acetone/hexane.

**Table 1**  
The information of laboratory sampling sites.

Sampling site	Area (m <sup>2</sup> )	Computer		Air-conditioning		Large-scale instrument		Incubator		Total electronic equipment	Plywood desktop
		Quantity	Operational time (h/d)	Quantity	Operational time (h/d)	Quantity	Operational time (h/d)	Quantity	Operational time (h/d)		
L1	40	10 desktop 4 portable	6 8	1 wall-mounted	8	3	5	/	/	18	24
L2	15	3 desktop 1 portable	10 7	1 wall-mounted	8	4	8	/	/	9	11
L3	60	1 desktop	3	1 vertical	6	/	/	13	24	15	35
L4	15	3 desktop	16	1 wall-mounted	8	3	10	/	/	7	15
L5	18	2 desktop 1 portable	24 14	1 wall-mounted	5	3	24	/	/	7	7
L6	25	/	/	1 wall-mounted	3	/	/	6	24	7	3
L7	28	1 desktop	8	1 wall-mounted	8	1	8	3	24	6	4
L8	20	4 desktop	10	1 vertical	15	4	8	/	/	9	13
L9	10	3 desktop	8	1 wall-mounted	12	3	10	/	/	7	6
L10	10	4 desktop	24	1 wall-mounted	12	4	24	/	/	9	8

The operational time of air-conditioning is the average operation time in summer and winter.

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