



Transport of Hexabromocyclododecane (HBCD) into the soil, water and sediment from a large producer in China



Yueqing Zhang^{a,b}, Yonglong Lu^{a,*}, Pei Wang^a, Qifeng Li^{a,b}, Meng Zhang^{a,b}, Andrew C. Johnson^c

^a State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

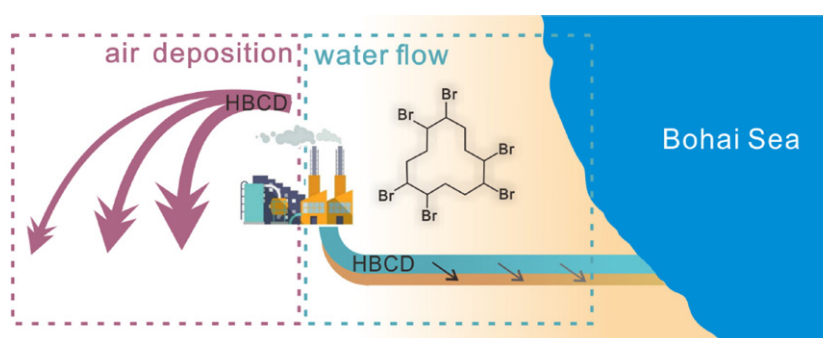
^b University of Chinese Academy of Sciences, Beijing 100049, China

^c Centre for Ecology & Hydrology, Wallingford, Oxfordshire, OX 10 8BB, UK

HIGHLIGHTS

- Emission from the biggest HBCD manufacturing facility in China was explored.
- Greatest world concentrations of HBCD in soil, water and sediment were found.
- Transport of HBCD through air and water decreased dramatically in several kilometers.
- Soil was the main fate of HBCD rather than sediment in the production region.

GRAPHICAL ABSTRACT



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ABSTRACT

Hexabromocyclododecane (HBCD) is used as a flame retardant with extensive industrial applications, which is mainly produced at facilities on the coast of China. Radially distributed soil samples and equidistant paired water and sediment samples were taken around one of the biggest HBCD production enterprises to reflect its environmental behavior via air deposition and wastewater discharge of HBCD diastereoisomers (α -, β - and γ -HBCD). Worldwide high concentrations of HBCD (11,700 ng/g in the soil, 5080 ng/L in the water and 6740 ng/g in the sediment) were detected in these environmental samples. Concentrations dropped by two orders of magnitude over several kilometers distance from the plant. The diastereoisomer pattern varied in the three environmental compartments examined, such that γ -HBCD was the predominant diastereoisomer in the soil and sediment whilst α - and γ -HBCD shared the predominance in the water. The mass inventories of HBCD in the local soil and sediment were estimated to be 5006 kg and 30 kg respectively, suggesting that soil was the major sink of HBCD in the production area. As for the soil, the environmental burdens in the areas with radiuses of 2, 4 and 6 km were 3210, 3770 and 4590 kg respectively.

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1. Introduction

Hexabromocyclododecane (HBCD) is a widely used brominated flame retardant. It is mainly used as an additive in expanded

polystyrene (EPS) and extruded polystyrene (XPS) foam in buildings and vehicles for thermal insulation. In addition, a lower amount of HBCD is used on textiles and electric/electronic equipment (POPRC, 2010). HBCD has become a widespread flame retardant after the phase out of polybrominated diphenyl ethers (PBDEs) (Chen et al., 2011). Commercial HBCD is composed of three diastereoisomers: α -, β -, and γ -HBCD, of which γ -HBCD is the predominant diastereoisomer

* Corresponding author.

E-mail address: yllu@ceees.ac.cn (Y. Lu).

accounting for 70%–89% to the total HBCD (Covaci et al., 2006; Marvin et al., 2011).

The global market demand for HBCD increased from 16,500 tons in 2001 to 31,000 tons in 2011 (POPRC, 2011; POPRC, 2012). HBCD was produced in China, Europe, Japan, and the USA. According to the report of the Persistent Organic Pollutants Review Committee of Stockholm Convention, China is the major producer of HBCD that 18,000 tons of HBCD was produced in China in 2011, which amounted to more than half of the global production (POPRC, 2012). The production of HBCD in China is intensively distributed in Weifang City, Shandong Province, which is a coastal city with abundant bromine resources (Shan et al., 2016). This is the biggest flame retardant production base in China and here there are several enterprises which produce thousands of tons of this product per year. With HBCD being listed in the Stockholm Convention, the Chinese government announced the ban of HBCD production, use, import and export at the end of 2016. However, an exemption was given to the production, use, import and export of HBCD for EPS and XPS from 2016 to 2021 (Ministry of Environmental Protection of China, 2016).

HBCD can be released into the environment during the whole life cycle from production to waste disposal via air and water transport. Because of its hydrophobicity, HBCD has been widely detected in the soil (Gao et al., 2011; Meng et al., 2011; Thanh et al., 2013; Zhang et al., 2016), sediment (Feng et al., 2012; Harrad et al., 2009a; Letcher et al., 2015; Oh et al., 2014; Tang et al., 2015; Wang et al., 2017) and aquatic organisms (Haukas et al., 2009; Roosens et al., 2010; Ueno et al., 2006; Wu et al., 2010) but rarely detected in the water (Harrad et al., 2009a; He et al., 2013; Oh et al., 2014). However, those previous studies were not conducted focusing on areas with an important point source of HBCD that may greatly affect the surrounding environment. The transport of HBCD through different environmental media has seldom been investigated, especially for the direct emission source of HBCD production. Therefore, the objectives of this study were to investigate the occurrence, spatial distribution and diastereoisomer pattern of HBCD in the soil, water and sediment at one of the biggest production enterprises in China to trace the transport of HBCD via air deposition and wastewater discharge. Moreover, mass inventories in the soil and sediment were evaluated to identify the important sinks for HBCD.

2. Materials and methods

2.1. Sample collection

The sampling campaign was conducted around one of the biggest HBCD manufacturing enterprises in China in July 2015. This manufacturing enterprise claimed an annual production capacity of 8000 tons and it was located at the southern coast of the Bohai Sea. The distance between the enterprise and the coastline was about 8 km. Twenty-four soil samples were collected around this enterprise at eight directions with distances of 2 km, 4 km and 6 km. Each sample consisted of five sub-samples within a 50 m × 50 m area and was collected by spade at a depth of 20 cm. Locations and descriptions of each sampling site were summarized in Table S1. At the same time, eleven paired water and sediment samples were collected along the wastewater discharge trench and its connected river, the Mihe River. The waterbody could be divided into two sections: Section 1 from site A to D was the wastewater discharge trench; Section 2 from site F to K was the natural riverbed of the Mihe River receiving the water from other trenches. The wastewater was discharged from the HBCD production facility and the drainage effluent was at Site B. Site E was the furthest downstream at the estuary to the Bohai Sea. The sampling depth 10 cm for the sediment, 10 cm for the water (At site B, the water in the drainage trench was <10 cm-deep, so the water was collected as its depth). Sampling tools were cleaned before the operation at each site (Fig. 1).

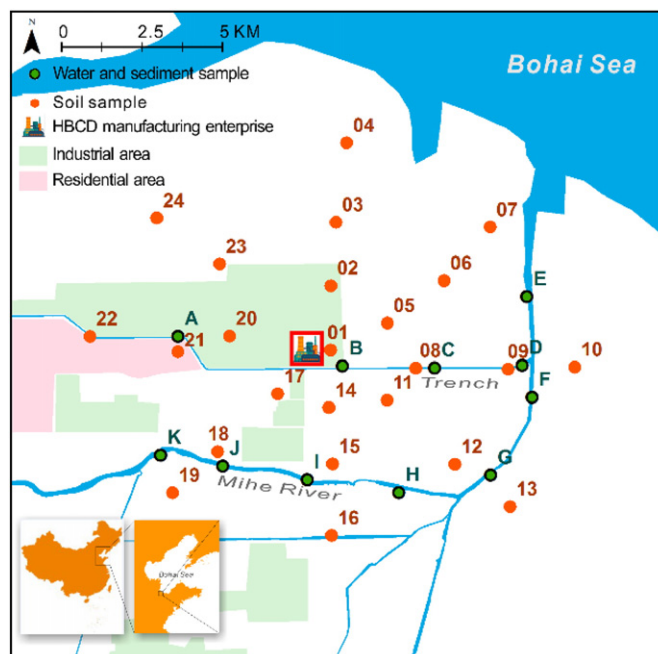


Fig. 1. Research area and sampling sites of this study.

During the campaign, basic water parameters including temperature, dissolved oxygen, pH and conductivity were measured by HACH water quality analyzer. The natural flow of the Mihe River was dammed further the upstream, so the riverbed in the downstream was dry at the time of sampling. Therefore, the river in the study area received the wastewater only from the adjacent industry. Aquatic organisms in the river were searched, but unfortunately, no living organisms were found due to the poor water quality. At the estuary (site E), fishnet had been put in the river for a week, but only one species of young fish (about 10 cm long) were found dead and rotted.

2.2. Reagent and standards

Solvents used in extraction and analysis procedures were HPLC-grade and purchased from Fisher Scientific. Silica gel 60 (63–100 μm) were purchased from Merck. Individual standard stock solutions (α -HBCD, β -HBCD, γ -HBCD, C13- γ -HBCD, d18- γ -HBCD) were obtained from Wellington Laboratories (Canada) with concentration of 50 μg mL⁻¹ in toluene with purity >98%. The Florisil was purchased from Sigma-Aldrich. The other chemicals were purchased from Sinopharm.

2.3. Extraction and cleanup

Soil and sediment samples were freeze-dried. All samples were stored at -20 °C until extraction. Extraction followed Harrad et al. (2009a) and Somoano-Blanco et al. (2016) with modification. For soil and sediment, samples (2 g for soil and 1 g for sediment) were spiked with 10 ng of C13- γ -HBCD as surrogate standard and was extracted using ASE 350 with hexane/dichloromethane (1:9, v/v) at 90 °C and 1500 psi (heating time 5 min, static time 4 min, purge time 90 s, flush volume 50%, static cycle 3). For water, samples (1.5 L) were spiked with 10 ng of C13- γ -HBCD as surrogate standard and were extracted with 20 mL hexane by manual shake for 20 min. The concentrated extract was then loaded onto a multi-layer silica gel column (15 mm I.D.) for purification, filled from bottom to top with 1 g of anhydrous sodium sulfate, 1 g activated silica gel, 2 g of Florisil, 1 g of activated silica gel, 3 g of basic silica gel, 1 g of activated silica gel, 8 g of acid silica gel, 1 g of activated silica gel, 1 g of anhydrous sodium sulfate. The column was wet-filled with hexane. Analytes were eluted with 120 mL

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