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Occurrence of polybrominated diphenyl ethers in soil from the central Loess Plateau, China: Role of regional range atmospheric transport

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ABSTRACT

Very few studies were conducted in highland and depositional areas in studying the transport and behavior of polybrominated diphenyl ethers (PBDEs). In this study, surface soils were collected from Huan County to investigate the level, profile, and potential influence of PBDEs via regional range atmospheric transport in the central part of the Loess Plateau (CLP) of China, one of the most extensive areas of loess deposition in the world. PBDEs were ubiquitous and log-normally distributed in soils from the CLP with mean concentrations of 0.91 and 0.54 ng g^{-1} for Σ PBDEs (sum of PBDE congeners except for BDE-209) and BDE-209, respectively. BDE-209 was predominated congener (43.5%), followed by BDE-47 (15.7%), 99 (10.7%), and 153 (7.5%). Further principal component analysis on congener profiles showed that PBDEs in the CLP originated from similar source(s). Additionally, significant differences in the ratios of BDE-47 to 99 and BDE-153 to 154 were found between soil samples and commercial products, indicating that they have undergone fractionation during the process of regional range atmospheric transport. The deposition of PBDEs in the CLP could be influenced by the sources from surrounding regions. For example, Xi'an may have potential influence to the CLP based on geographical analysis and concentrations comparison of PBDEs in this region.

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1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a class of additive brominated flame retardants that are added to plastics, polyurethane foam, textiles, and electronic equipment to reduce the material flammability (Hites, 2004). Despite their benefits, PBDEs is qualified as persistent, bioaccumulative, and toxic substances and is widely concerned by environmental researchers and policymakers (Birnbaum and Staskal, 2004; Hites, 2004). Currently, the Penta- and Octa-formulations have been voluntarily withdrawn from the United States (USA) marketplace since the end of 2004. Deca-BDEs have also been banned in some countries from the Europe Union (EU) and North America. In contrast, in China, the domestic demand of brominated diphenyl ethers (including PBDEs) has increased annually at a rate of \sim 8% (Mai et al., 2005). In addition, China is the world's largest importer and recycler of waste electrical and electronic equipments (Ni and Zeng, 2009). During the electronic waste (e-waste) dismantled with crude methods, PBDEs was released into the environment (Leung et al., 2007; Meng et al., 2008; Luo et al., 2009).

Persistent organic pollutants (POPs) can be diffused from point source area to surrounding and remote areas undergo long/

* Corresponding author. Tel./fax: +86 21 65984261. *E-mail address:* chenling@tongji.edu.cn (L. Chen). regional range atmospheric transport (Wania and Dugani, 2003; Zhao et al., 2009). Therefore, remote regions are beginning to receive increased attention in studying the transport and behavior of POPs (Liu et al., 2010). Currently, to the best of our knowledge, no study on POPs was conducted in the Loess Plateau (LP) of China.

The LP, also known as the Huangtu Plateau, is one of the most extensive areas of loess deposition in the world. Historically, the LP was created by the deposition of wind-blown dust and by glacial till, called loess. Present eolian dust is mainly transported by low level winds in step-wise way to deposit in this area (Sun et al., 2001). The LP (between 33-40°N and 98-115°E) is highland area in north-central China, covering much of Shanxi, northern Henan, Shaanxi, eastern Gansu provinces, and the middle part of the Yellow River basin (Fig. 1). The average elevation is about 1200 m and the area is about 400,000 km². Precipitation totals from 500 mm per year in the east to 250 mm in the northwest. Huan County, located at the central part of the LP (CLP; between 36°01'-37°09'N and 106°21'-107°44'E), has the area of 9236 km² and the population of 351,000, of which 93% are farmers. The annual precipitation is about 300 mm and the elevation ranged from 1136 to 2089 m. The objectives of present study are to investigate the levels and compositions of PBDEs in surface soil from the CLP and to examine the potential influence of regional range atmospheric transport for PBDEs deposition in this area.



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Fig. 1. Map of the study area and sampling sites.

2. Materials and methods

2.1. Soil sampling

Between July and August 2009, soil sampling was conducted 20 locations in Huan County, including of Tianshui, Shancheng, Qintuanzhuang, Nanjiu, Luoshanchuan, Gengwan, Siheyuan, Xiaonangou, Hongde, Maojing, Hudong, Huancheng, Fanjiachuan, Bazhu, Lujiawan, Hedao, Mubo, Quzi, Tianchi, and Yanwu. Prior to soil collection, any overlying vegetation was removed. Composite surface agricultural soil samples (0–10 cm depth) were collected using the cleaned handheld corer from 45 sites in Huan County, the CLP (Fig. 1 and Table S1, "S" designates Supplemental Information here and thereafter). The first two cores were discarded, then the following three cores (taken over an area of several m²) were combined as one sample. The samples were wrapped in aluminum foil and sealed in polyethylene bag, and then transported to the laboratory. Before extraction, the samples were freeze-dried, mixed thoroughly, sieved to 100 mesh and kept at -18 °C.

2.2. Standard materials

Our targets, including BDE-17, 28, 33, 47, 49, 66, 99, 100, 138, 153, 154, 183, 190, 196, 203, 206, 207, 208, and 209, were purchased from AccuStandards (New Haven, CT, USA). Surrogates (BDE-50 and 172) and internal standards (BDE-118 and 128) were also purchased from AccuStandards (New Haven, CT, USA).

2.3. Analytical procedure

The detailed analytical procedure was presented elsewhere (Mai et al., 2005; Duan et al., 2010). Briefly, spiked with BDE-50 and 172, about 10 g soil was Soxhlet extracted with a mixture of acetone and hexane (1:1 in volume) for 48 h. Activated copper

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