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Insight into the local source of polybrominated diphenyl ethers in the developing Tibetan Plateau: The composition and transport around the Lhasa landfill☆

Jun Li [a, b], Guo-Li Yuan [a, b, *], Ping Li [b], Xu-Chuan Duan [b], Hong-Hui Yu [b], Jun-Lang Qiu [c], Gen-Hou Wang [b]

[a] State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Beijing 100083, China
[b] School of the Earth Sciences and Resources, China University of Geosciences, Beijing 100083, China
[c] School of Chemistry, Sun Yat-Sen University, Guangzhou 510275, China

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ABSTRACT

In the background region of the Tibetan Plateau (TP), the rapid urbanization probably results in the massive generation of persistent organic pollutants (POPs), which lacks monitoring and evaluation. Since landfill could serve as an important sink of the locally used POPs, the analysis of POPs in the Tibetan landfill area might help us to understand the source composition and their transport in the TP. In this study, the concentration variations of polybrominated diphenyl ethers (PBDEs) in five soil profiles and seven surficial sediments around the largest Tibetan landfill were investigated. The total concentrations of PBDEs ranged from 128 to 1219 ng/kg in soils, and from 447 to 7295 ng/kg in sediments. The dominance of nona- and deca-BDEs possibly indicated the wide usage of deca-BDE as flame retardant in the TP. The vertical and spatial distribution patterns of PBDEs within soils plausibly revealed their main transport pathways by atmospheric dispersion and leachate seepage from landfill. Based on principal components analysis and multiple linear regression, these two pathways were estimated to account for 61% and 39% of the total concentrations, respectively. Additionally, the spatial and vertical distributions of octa-to deca-BDEs within soils were significantly influenced by soil particle size. Although the PBDEs inventory in the study area was comparatively low, the rapid urbanization in the TP might dramatically accelerate the PBDE emissions in the future. This study firstly introduced the presence of local PBDEs in the TP, and the inventory already influenced the surrounding environment. Once involved in the regional cycle of the TP, the local source of PBDEs from waste might significantly serve to raise background level resulting otherwise primarily from long-range atmospheric transport.

1. Introduction

The Tibetan Plateau (TP), occupying an area of 2.6×10^6 km^2 with an average elevation of over 4000 m, is the largest and the highest plateau in the world, and has long been known as the “Third Pole” (Tao et al., 2011; Wang et al., 2010). This remote region was regarded as the background for persistent toxic substances (PTSs) on the earth since its low population density and less frequent human activities (Tao et al., 2011; Yuan et al., 2012). Among PTS chemicals, persistent organic pollutants (POPs) are an important class of compounds in the environment and have the potential to affect the mountain ecosystems due to long-range atmospheric transport (LRAT) (Bettinetti et al., 2008; Wang et al., 2015). In the TP, POPs were generally considered to be derived from the outside environment through LRAT (Wang et al., 2010, 2012; Yuan et al., 2012). However, in the recent decades, the development in the TP is characterized by rapid urbanization, along with a sustained increasing influx of local population and tourists (Yu et al., 2012). This development pattern is creating an enormous amount of wastes more than the region can handle (Qiu, 2014), which might result in the local source of POPs in the TP (Yuan et al., 2015b; Li et al., 2017). Unfortunately, only limited data on the usage history and current application are available for various POPs in this

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* Corresponding author. State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Beijing 100083, China.
E-mail address: yuangl@cugb.edu.cn (G.-L. Yuan).

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background region.

Landfill is an important disposal pathway for waste consisting of, or containing toxic and carcinogenic organic pollutants (Melnyk et al., 2015). Therefore, the detailed investigation on the Tibetan landfill areas may provide the adequate records for the application situation of local POPs (Huang et al., 2013; Wang et al., 2014). Landfills would massively release pollutants to the adjacent environment through gases and leachates (Eggen et al., 2010; Weinberg et al., 2011). Soils are the major reservoir for POPs due to their strong sorption quality and retention capacity (Cabrerizo et al., 2011; Komprda et al., 2013). Thus, it is significant to study the fate of POPs in the soils surrounding the Tibetan landfills, since it could reflect the influence of urbanized waste on the fragile ecosystem of the TP. At the same time, the analysis of sediments around the landfills might help to reveal the transport of POPs from waste to environmental media because sediments could better archive the initial composition of POPs from local source (Ulrich et al., 2009; Yuan et al., 2012).

In the terrestrial environment, POPs are preferentially adsorbed onto fine particle minerals in soils, such as clay (Harner and Shoeib, 2002). The close association between POPs and soil compositions indicated that these organic pollutants could be vertically and laterally transported with clay minerals within soils (Cheng et al., 2014; Yuan et al., 2012). Similarly, the sorption of soil organic carbon (SOC) might also influence the distribution of POPs in soil compartments (Hassanin et al., 2004). Besides, the waste leachate could directly release pollutants into the deeper soils (Bergknut et al., 2011; Liu et al., 2015). If POPs were actually accumulated in deeper soils, the measurement in surficial layer might not reflect sites' contamination. However, the contamination situation of deeper soils was scarcely reported in the TP. Thus, it is also important to study the concentration profiles of POPs in the deeper soils surrounding the Tibetan landfills and their relationship with soil parameters, for the purpose of better illustrating the POPs' situation. Simultaneously, this investigation might provide the additional information about the transport process of POPs within deeper soils.

Lhasa, as the most urbanized area of the TP, generates 215 million tons of solid waste each year, which was disposed into the Lhasa landfill without appropriate treatment (Dian and Bu, 2012). In this case, the Lhasa landfill is an excellent site for tracing local POPs in the TP, such as polybrominated diphenyl ethers (PBDEs). PBDEs are a group of POPs that are used as additives in electronic products, textiles, plastics and building materials (Li et al., 2016), and could be easily escaped from these products (de Wit, 2002; Rahman et al., 2001). Their amounts released into the environment increased dramatically due to the rapid urbanization and enhanced socioeconomic status of the local population (Li et al., 2016; Wang et al., 2014; Zhao et al., 2016). Due to the bioaccumulative and toxic properties (Ma et al., 2013), the commercial penta- and octa-BDEs were banned globally. However, the commercial deca-BDE remains available for all uses in most regions of China, including Tibet (Liu et al., 2017). In this study, the analysis of PBDEs in the Lhasa landfill area was conducted, attempting to offer an initial and particular insight into the potentially environmental issue faced by the economically developing TP. Our objectives were (1) to investigate the level and source composition of PBDEs in the environment of the TP; (2) to reveal the transport processes of PBDEs and the influencing factors on their distribution within soils; (3) to discuss the current loadings of the local PBDEs and their future trends due to the rapid development.

2. Materials and methods

2.1. Study site and sampling

The targeted landfill is located in a valley of Qushui County and 20 km away from central Lhasa city in southern Tibet, where the average annual precipitation is 452 mm (Du et al., 2008). It is the largest landfill in the Tibet with waste treatment of 680 tons/d and the service began in 2002 (Tibet statistical yearbook, 2016). The waste soil (WS) sample was collected inside the landfill. In the catchment area where the landfill is located, four soil profiles (SP1, SP2, SP3 and SP4) and three river sediments (DN1 and DN2 and DN3) were collected from a 370 m valley-slope gradient (2.2–5.1%) with altitudes from 3604 to 3980 m (Fig. 1). Meanwhile, three sediments (DN4, DN5 and DN6) were collected from the downhill river (Fig. 1). In addition, one soil profile (SP5) and one sediment (DN7) were collected in another catchment, which is separated from the landfill catchment by hills (Fig. 1). Five sites where were selected for soil profile sampling had not been recently disturbed. The waste soil and the five soil profiles were collected by SH-30 drilling rig (diameter: 130 mm), and the river sediments (DN1-DN7) were collected using a grab sampler. In each profile (SP1-SP5), five soil samples were collected from surface to bottom and sectioned onsite into one surficial layers (0–20 cm) and four deeper layers (20–60, 60–100, 100–150 and 150–200 cm). More detailed location information for soils and sediments is outlined in Table S1 and Table S2 in Supplementary material (SM), respectively. All samples were wrapped with aluminum foil, transported to the laboratory, and stored at −5 °C. In the investigated catchment area, the shallow groundwater is located in the silt/silty clay layers at a depth of 0.8–3.1 m, which decreases downslope along the hydrological gradient.

2.2. Chemicals

Target analytes including BDE-28, 47, 99, 100, 153, 154, 183, 197, 206, 207, 208 and 209 were purchased from AccuStandard (New Haven, CT, USA). The ^{13}C-labeled BDE-209 and 2,4,5,6-tetrachloro-mxylene (TCMX) (Supelco, Bellefonte, PA, USA) were used as

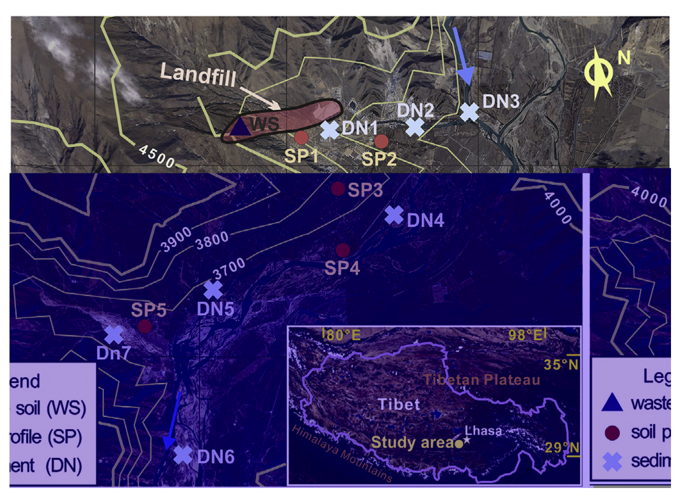

Fig. 1. Map showing the Lhasa landfill with the sampling site. The grid representing 10 km. (Bottom right) Overview map of Tibet showing the location of the study area.