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Effects of hydrodynamic disturbances and resuspension characteristics on the release of tetrabromobisphenol A from sediment☆

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Tetrabromobisphenol A (TBBPA) exists widely in river and lake sediments; it has raised growing attention in recent years as emerging contaminant due to its possible threats to the aquatic environment and human health. Using a specialized simulator, the relationships between hydrodynamic disturbances and resuspension characteristics were simulated, with an emphasis on microscopic characteristics. Furthermore, TBBPA release from sediment was studied in relation to hydrodynamic disturbances and resuspension characteristics. The results show that stronger water disturbances caused an increase in suspended solids concentration (SSC) and produced different behaviors of particle size distribution (PSD) and media diameter (D_{50}) in the slight and large-scale resuspension situations. As for microscopic resuspension characteristics, the specific surface area (SSA) of suspended particulate matter (SPM) was very different from that of smooth particles. This difference may result from the fractal nature of the SPM. The fractal dimension (FD) of SPM was found to have a significant correlation with turbulent kinetic energy. TBBPA release into overlying water and adsorption onto SPM both increased with hydrodynamic disturbances; but the release into overlying water is more dominant. The TBBPA concentrations in SPM under different hydrodynamic conditions were significant related to SSA, indicating that SSA is a key factor affecting the TBBPA adsorption capacity of SPM. TBBPA concentrations in sediment decreased slightly with the increased hydrodynamic dispersion. Findings from this research show the importance of considering the hydrodynamic disturbance and resuspension characteristics in understanding TBBPA release behavior in aquatic environment.

1. Introduction

Tetrabromobisphenol A (TBBPA) has been widely used in textiles, plastics, and electronic circuit boards as a flame-retardant substance (Alaee et al., 2003). The worldwide market demand for TBBPA was 121,300 tons in 1999 and 170,000 tons in 2004 (de Wit, 2002; Makinen et al., 2009). China is one of the largest producers and users of TBBPA. Annual output of TBBPA in China increased from 8000 tons in 2000 to 18,000 tons in 2007 (Jiang, 2007; Sun et al., 2008), and TBBPA use has been maintaining a fairly high growth rate in China (Shi et al., 2009).

TBBPA is a chemical compound that is characterized by persistence, bioaccumulation, and toxicity (WHO/ICPS, 1995). TBBPA has been shown to be toxic to aquatic organisms, terrestrial animals, and even humans (Darnerud, 2003; Jagnytsch et al., 2006; Shi et al., 2005). In the process of TBBPA production, use, and disposal, TBBPA inevitably flows into aquatic environments through industrial and domestic sewage (de Wit, 2002). Part of the TBBPA is released by dissolution into the overlying water or adsorption on suspended particulate matter (SPM). However, most of the TBBPA is stored in sediment because of its low water solubility (de Wit, 2002). TBBPA

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has reportedly been detected in a number of environmental water systems such as estuaries, rivers, and lakes (Morris et al., 2004; Saint-Louis and Pelletier, 2004; Yang et al., 2012). The sediment from the Lake Chaohu estuary may be the most contaminated in the world; its TBBPA concentration has already reached 518.3 μg/kg (dry weight; d.w.) (Yang et al., 2012). Furthermore, once the contaminated sediment has been disturbed by hydrodynamic forces, the contaminated sediment is likely to enter the overlying water through resuspension processes (Je et al., 2007). In this way, TBBPA in sediment can be released persistently to water and can migrate over long distances (WHO/ICPS, 1995). Moreover, aquatic plants and organisms easily uptake TBBPA from both overlying water and SPM in contaminated aquatic environments, causing bioaccumulation throughout the food chain (Debenest et al., 2010; Morris et al., 2004). Thus, TBBPA can not only pollute water environments, but can also threaten the health of aquatic flora and fauna.

For these reasons, researching the sediment release behavior of TBBPA has direct practical significance. Current studies of water disturbance effects on contaminants have mainly focused on nitrogen, phosphorus, heavy metals, and other organic contaminants (Fan et al., 2001; Wang et al., 2009; Zheng et al., 2013). For example, Fan et al. (2001) reported that phosphorus release was significantly influenced by hydrodynamic disturbance. Recently, Zheng et al. (2013) illustrated that the distribution of metals in overlying water was significantly affected by hydrodynamic disturbance. However, the effects of hydrodynamic disturbance on the remobilization and fate of TBBPA are not fully understood.

Under natural conditions, hydrodynamic disturbance is ubiquity. Sediment resuspension occurs frequently, mainly under the influence of flow (Wang et al., 2015; Eggleton and Thomas, 2004). When hydrodynamic disturbance becomes stronger, sediment resuspension characteristics vary, resulting in contaminant redistribution (Fan et al., 2004a; Tengberg et al., 2003). It is the main reason that hydrodynamic disturbance is an important factor influencing sediment release (Fan et al., 2004b; Kalnejais et al., 2007; Xie et al., 2015), and clarifying resuspension characteristics under hydrodynamic disturbance is an important prerequisite to explaining sediment contaminant release behavior.

Most recent research on sediment release has focused on the basic resuspension characteristics influenced by hydrodynamic disturbance, such as suspended solids concentration (SSC), particle size distribution (PSD), and media diameter (D50) (Kalnejais et al., 2007; Kim et al., 2004; Wu and Hua, 2014). These studies reported that contaminant release increases with SSC and that fine small-diameter particles have a stronger ability to release from SPM than larger particles. In fact, the microscopic characteristics of SPM, such as specific surface area (SSA) and fractal dimension (FD), have a more direct reflection of pollutant adsorption ability (Gašparović et al., 2007; Macht et al., 2011). This may be particularly important for persistent organics, because of these high hydrophobicity and poor water solubility. However, few studies have observed the variation of these microscopic characteristics in SPM influenced by water disturbance in a flume simulation experiment. Furthermore, little is known about the effects of microscopic resuspension characteristics and hydrodynamic disturbance on release of TBBPA from sediment.

The purpose of this research was to investigate the effects of hydrodynamic and resuspension characteristics on TBBPA release by adsorption on SPM, and release into the overlying water. A racetrack-style flume was used to simulate sediment resuspension under a range of typical hydrodynamic conditions in natural aquatic environment. The sediment resuspension characteristics, both basic and microscopic, induced by different hydrodynamic conditions were determined.

2. Materials and methods

2.1. Sampling site and sample collection

The experimental sediment was collected from the western Lake Chaohu near the Nanfei River estuary (China) (31°41′51″N, 117°24′16″E) in October 2015. The sediment at the Nanfei River estuary is affected by water disturbances; and resuspension occurs frequently, which strongly influences TBBPA release from sediment. Fig. 1 shows the five sampling sites at the Nanfei River estuary. A Peterson grab sampler (WHL 15-HL-CN) was used to collect surface sediment samples (0–15 cm), which is considered to be the main active layer taking part in dynamic exchange (Qin et al., 2004). Additional ten sediment samples were acquired at each site to measure the background value of Lake Chaohu. Polyethylene boxes were used to hold the sediment samples and transport them to the laboratory. All the sediment samples were pooled, and mixed fully before being preserved at under 4 °C until the experiments were performed. The tap water was used as the overlying water during the experiment.

2.2. Experimental design

A type of flume with a racetrack-style configuration was used to perform hydrodynamic disturbance and sediment resuspension experiments. The total height of the flume was 0.8 m, and the flow cross section was 0.3 m. A range of hydrodynamic conditions were simulated by adjusting two sets of driving devices, including several axially assembled dishes connected to a variable-frequency motor on top of the flume at counter-rotating positions, as shown in Fig. 2. The driving mechanism used in this study was consistent with natural water conditions (Hua et al., 2012). All pooled and homogenized sediment were placed on the bottom of the flume to a thickness of 15 cm. The overlying water was slowly injected into the flume by a siphon until the water depth was 50 cm. To approximate the field situation, the whole system was left undisturbed for two weeks.

Eight levels of hydrodynamic disturbances and resuspension processes were simultaneously simulated via eight identical flumes and sediment in this study (events 1–8). The hydrodynamic conditions was designed on the basis of actual hydrodynamic disturbances observed at the sampling site and referring to previous studies (Chen et al., 2013; Wang et al., 2011). The flow structure, resuspension characteristics and contaminant distribution can reach a steady state in 12 h (Wu and Hua, 2014); the sample collection and flow measurement were conducted at the steady state. The temperature was controlled at 15 °C during the whole experiment.

The flow characteristics (velocity and turbulent kinetic energy) of the overlying water were ascertained from the water surface to the bottom by 1-cm steps by an Acoustic Doppler Velocimeter (ADV) for each event. The pH levels of the overlying water were measured using a Hach pH meter. Fig. 2 presents the sampling points in the experiment. Sediment samples were collected using a cylindrical sampler (11 cm I.D., 50 cm in length), and water samples were composited and withdrawn using a layered hydrophore located at intervals of 10 cm in this balanced system. Water samples were composited. SPM samples were obtained by centrifuging overlying water (Wang et al., 2015).

2.3. Analytical technique

The flow characteristics, resuspension characteristics, and TBBPA distribution were analyzed for each simulated hydrodynamic condition. Additional sediment samples at each site from the