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Distribution, release and removal behaviors of tetrabromobisphenol A in water-sediment systems under prolonged hydrodynamic disturbances

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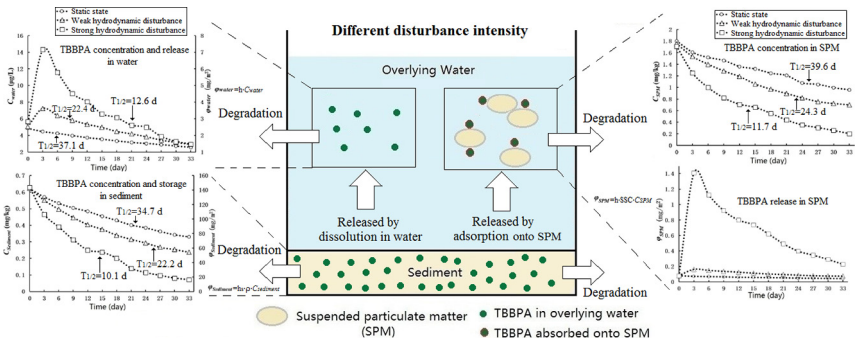
HIGHLIGHTS

- TBBPA distribution, release and removal in water-sediment system were investigated using a racetrack-style flume.
- Prolonged hydrodynamic disturbance had significant indirect or direct influence on TBBPA removal in water, SPM and sediment.
- The half-lives in water, SPM and sediment were shorter in strong hydrodynamic condition, comparing to the static control.
- Enhanced water disturbance led to an increase of TBBPA distribution and release in water and SPM.

GRAPHICAL ABSTRACT



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ABSTRACT

Since tetrabromobisphenol A (TBBPA) has been increasingly used and found widely in the aquatic environment, it has attracted much attention due to its high toxicity to aquatic organisms. However, less work has been carried out for the TBBPA environmental fate in water-sediment systems. In this paper, the distribution, release and removal of TBBPA in different forms of the water-sediment system were investigated under three typical hydrodynamic conditions using a specialized racetrack-style flume. Three water-sediment systems which are water, suspended particulate matter (SPM) and sediment were taken into account in this study. The results of 34 days experiments showed that the equilibrium of physicochemical parameters was reached under different disturbance conditions within a relatively short period. The distribution ratio of TBBPA in three water-sediment systems will also reach a relatively equilibrium state over time under different disturbance conditions. The fluctuation range in each form was <1.26%. The TBBPA released to the water and SPM, increased remarkably with hydrodynamic enhancement due to adsorption and resuspension processes. Removal efficiency of TBBPA in each form was found to be observably accelerated with hydrodynamic disturbances. The half-lives ($T_{1/2}$) in water, SPM and sediment ranged from 10.1 to 12.6 days in strong hydrodynamic condition, comparing to the static control, it ranges from 34.7 to 37.1 days. This phenomenon may result from the increase of dissolved oxygen (DO) and nutrient concentrations in overlying water affected by hydrodynamic force. The results of the experiment demonstrate that hydrodynamic disturbance may be an important driving factor which will influence the TBBPA environmental fate in aquatic environment.

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1. Introduction

Tetrabromobisphenol A (TBBPA) is a preferred flame-retardant; and it is widely used as an additive in commercial and industrial applications, such as textiles, plastics, and electronic circuit boards (Alaee et al., 2003). The yearly TBBPA world market increased from 120,000 tons in 2001 to 200,000 tons in 2013 (Li et al., 2015a; Mäkinen et al., 2009). As a result of its widespread use, TBBPA inevitably goes into aquatic environments such as rivers, lakes and estuaries through anthropogenic effluents (Matsukami et al., 2015; WHO/ICPS, 1995). It is reported that TBBPA exists in various aquatic environmental media, including water, suspended particulate matter (SPM), sediment and aquatic biota (De Wit et al., 2010; Gu et al., 2017; Harrad et al., 2009).

Compared to the other aquatic environmental media, TBBPA tends to adsorb onto particulate matter (SPM and sediment) and accumulates in sediment through sewage discharge. This is due to its low water solubility (4.16 mg/L at 25 °C), high lipophilicity ($\log K_{ow} = 4.50–6.53$) and low volatility (7.0×10^{-11} atm·m^3/mol) (De Wit, 2002; Nyholm et al., 2013). Subsequently, TBBPA in contaminated sediment can potentially migrate into the water column persistently even in a static situation, including dissolved and particulate fractions, through dissolution, diffusion and resuspension processes (Cheng et al., 2013; Malkoske et al., 2016). Then, aquatic biota can easily uptake TBBPA from overlying water (Debenest et al., 2010). However, TBBPA has been proved to be toxic to aquatic biota, and the acute 96 h LC50 values for three species of fish were about 0.5 mg/L (bluegill sunfish, rainbow trout and fathead minnow) (Darnerud, 2003). The evidence increasingly highlights its negative physiological effects (Lyche et al., 2015; Waaijers et al., 2013). Thus, investigating the TBBPA environmental distribution and fate in water-sediment system has important theoretical significance for evaluating TBBPA exposure level.

Once it has entered into water-sediment system, the distribution and fate of TBBPA in environmental media were shown to be influenced by a number of environmental processes, including adsorption in particulate phase (SPM and sediment), sediment resuspension and degradation (Malkoske et al., 2016). Meanwhile, under natural conditions, hydrodynamic force is ubiquity. It is one of the most important physical forces affecting contaminant adsorption and resuspension processes (Eggleton and Thomas, 2004). Large shear stress near the interface can affect the diffusive boundary layer and cause contaminant remobilization (Kalnejais et al., 2007). Previous studies have shown that adsorption and resuspension processes of phosphorus and phenanthrene are closely related to disturbing intensity (Wang et al., 2009a; Wang et al., 2009b). Cheng and Hua (2016) explored the relationship between hydrodynamic disturbances and TBBPA release in a relatively short experimental period without considering the potential of TBBPA removal. However, water disturbance is long-standing in aquatic environments. The information on prolonged hydrodynamic disturbance effects on environmental processes of contaminants, especially organic contaminants, is limited and incomprehensive.

Over time, TBBPA may be transformed by biodegradation, photooxidation and chemical oxidation. Biological processes generally are the major way of TBBPA degradation in aquatic environments (Truu et al., 2009). In water-sediment systems, hydrodynamics are considered as the major agents of physical force for driving the environmental parameters, such as dissolved oxygen (DO), nutrients, turbidity, and dynamics of bacterial community (Besemer et al., 2009; Li et al., 2015b; Peraltamaraver et al., 2017). Nevertheless, variations of these environmental parameters may initiate modifications for removal efficiency of organic contaminants. It has been proven that TBBPA degradation could be accelerated in oxic conditions (Liu et al., 2013). Peng and Jia (2013) indicated that the addition of carbon and nitrogen sources may increase TBBPA degradation efficiency. Dynamics of bacterial communities could also influence the degradation process of TBBPA (Emilie et al., 2016; Li et al., 2016). Hence, it is hypothesized that hydrodynamic disturbances may play an important indirect influence on the removal of TBBPA. Removal behavior may be the main factor affecting TBBPA distribution and release over time. So far, the effects of hydrodynamic disturbances on TBBPA removal have not been taken into account in most studies.

This paper attempts to investigate the effects of water disturbances on distribution, release and removal behaviors of TBBPA in the water-sediment system. Three typical hydrodynamic conditions in an aquatic environment were simulated by using a racetrack-style flume. The amounts of TBBPA in each form, which are water, SPM and sediment, were recorded for different periods. The distribution, release and removal behaviors of TBBPA were analyzed in the presence of hydrodynamic disturbances, and the corresponding environmental processes were also discussed.

2. Materials and methods

2.1. Location of sampling site and experimental materials

The experimental sediment were taken from surface sediment (0–15 cm) located in the Nanfei River estuary in the west of Lake Chaohu by a Peterson grab sampler in October 2015 (Cheng and Hua, 2016; Zheng et al., 2013). Its TBBPA concentration of this estuary was reported at a very high level in the world (518.3 μg/kg) (Yang et al., 2012). Meanwhile, the environmental behaviors of TBBPA in this estuary are strongly affected by water disturbances. The sampling sites are shown in Fig. 1. Additional ten sediment samples were acquired at each site to measure the background value of this area in Lake Chaohu. After being transported to the laboratory immediately, the sediment samples were preserved below −20 °C until the experiment was conducted. Dechlorinated tap water was used as the overlying water throughout the experiment.

2.2. Experimental design

This experiment was conducted by using three parallel racetrack-style flumes to simulate TBBPA environmental behaviors in a water-sediment system under different hydrodynamic conditions. The total height and flow cross section of the flume are 0.8 m and 0.3 m, respectively, as shown in Fig. 2. This experimental flume has proven very effective in simulating contaminant remobilization under hydrodynamic disturbance (Cheng and Hua, 2016; Hua et al., 2013). The sediment samples, which had been pooled, mixed and homogenized fully, were placed on the bottom of the flume. Then, dechlorinated tap water was injected into the flume by a siphon. According to the hydrodynamic characteristics provided by other researchers and the flume configuration, the experimental sediment thickness was designed to be 0.15 m, and the water depth was 0.50 m (Hua et al., 2013; Qin et al., 2003). The whole system was left undisturbed for one week to approach the field situation.

Water-sediment system would show two different resuspension states under the influence of hydrodynamic disturbance. They are slight-scale and large-scale resuspension situations. Based on previous studies, a weak hydrodynamic event and a strong hydrodynamic event were simultaneously simulated corresponding to slight-scale and large-scale resuspension situations respectively (Chen et al., 2013; Cheng and Hua, 2016). Meanwhile, the two representative hydrodynamic conditions were observed *in situ* frequently in the Nanfei River estuary (Chen et al., 2013). A static state was set as a control. The duration of the experiment was designed to 34 days, following the TBBPA removal process. The flow structure was conducted at a steady state during each event. The room temperature was constant at 15 °C using air-conditioning.

At each event (0–34 day), water and sediment samples were collected every three days (0, 3, 6 … 30, and 33 d) at 10 am. Ten parallel water samples were withdrawn using a layered hydrophore located at depths of 0 cm, 10 cm, 20 cm, 30 cm and 40 cm each time. Six parallel