



## Bioturbation/bioirrigation effect on thallium released from reservoir sediment by different organism types



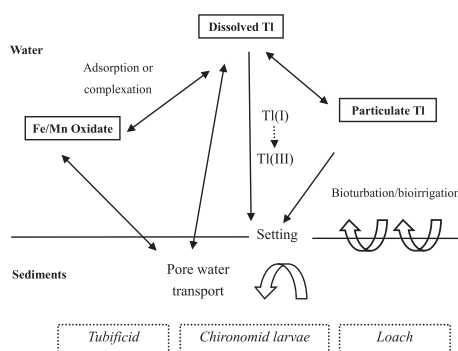
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### HIGHLIGHTS

- The total and particulate Tl concentrations had good correlations with turbidity.
- The bioturbation process suppressed the release of dissolved Tl from the sediment to water after 14 days.
- Loach has the most significant impact on Tl mobilization from sediment.
- Iron and manganese hydrous oxides may sorb or coprecipitate with Tl.
- Tl(I) may be oxidized to be Tl(III) by planktonic bacteria.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Bioturbation can remobilize heavy metal in the sediments and may pose a risk for aquatic biota. The effects of bioturbation/bioirrigation by three different riverine organism types (Tubificid, Chironomid larvae, and Loach) on thallium release from contaminated sediment ( $10.0 \pm 1.1$  mg Tl/kg sediment, dry wt.) were evaluated in this study. The bioturbation by the epibenthos clearly caused an increased turbidity in the overlying water, and the effect was in the order of Loach > Chironomid larvae > Tubificid. A significant release of Tl into the water column via the resuspended sediment particles was observed, especially for Loach. During the first few days, the leaching of dissolved Tl from sediment into water was fast, and the dissolved Tl under bioturbation/bioirrigation was much higher than the control group. However, after 14 days, the bioturbation/bioirrigation process seemed to suppress the release of Tl from the sediment particles to water, especially for sediment with Loach. This may partly be due to the sorption or coprecipitation of Tl simultaneous with the formation of iron and manganese hydrous oxides with increased pH values as a consequence of phytoplankton growth. Linear regression analysis confirmed that both the total and particulate Tl concentrations had good correlations with particulate Fe and Mn concentrations as well as turbidity in the overlying water. Additionally, planktonic bacteria may oxidize the Tl(I) to Tl(III), resulting in a reduced solubility of Tl by which Tl(OH)<sub>3</sub> becomes the predominant form of Tl.

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### 1. Introduction

Thallium is a highly toxic element and is listed as a priority pollutant by the U.S. EPA (Liu et al., 2011; Mulkey and Oehme, 1993; Smith and Carson, 1977). It is a rare element in the natural environments usually with very low abundance (Peter and Viraraghavan, 2005).

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Thallium concentrations in the background soil of China ranged from 0.29–1.17 mg/kg (Qi et al., 1992); and Tl concentrations in Chinese typical background sediments were in the range of 0.30–0.49 mg/kg (Zhao and Yan, 1994). However, Tl concentrations are elevated in the areas impacted by human activities and urban development (Lin et al., 2001; Lis et al., 2003). In China, an incidental Tl pollution (0.18–1.03 Tl  $\mu\text{g/L}$ ) from the wastewater discharge of a lead/zinc smelting plant on drinking water source of the northern branch of Pearl River was reported in 2010 (Agency, 2010), which further aroused public concerns on Tl pollution in China. Therefore, the study of the behaviors and the fate of Tl is significant and necessary.

Once introduced into the aquatic environment, Tl is redistributed throughout the water column, deposited and accumulated in sediments as a result of complex physical, chemical and biological processes (Larsson, 1985). Contaminants accumulate on organic (Sridhar et al., 2008), inorganic (Lin, 1997) sediment particles and their associated microorganism community (D'Souza et al., 2006). So sediments are an important medium to study the Tl behavior which may pose a risk for the ecological quality of surface waters (Duan et al., 2012). Some studies have shown that the disturbance by benthic organisms was an important process affecting secondary pollutants released, because of the behavior as buried hole, ingestion, breathing, movement and discharge. These behaviors changed the porosity, compaction, pH and redox potential distribution of sediments to affect the mutual exchange of pollutants among the sediment, sediment pore water and the overlying water (Banta and Andersen, 2003; Granberg et al., 2008; Hunting et al., 2013; Josefsson et al., 2010; Lagauzere et al., 2009; Nogaro et al., 2013; Thibodeaux and Bierman, 2003). Bioturbation brought the contaminated sediment particles to the interface of sediment and water, that these particle resuspension to provide a contaminated habitat for the benthic organisms lived in the surface sediment (Gang et al., 2006; Mermillod-Blondin et al., 2008; Rhoads, 1974; Schaller, 2014; Simpson et al., 1998; Tengberg et al., 2003; Thibodeaux and Bierman, 2003; Widdows et al., 1998). To date, however, it is not clear which are the effects of bioturbation/bioirrigation on Tl fluxes at the sediment–water interface.

Invertebrates and fish may mix sediment particles in a variety of ways. For example, Tubificid worms are important “conveyor-belt” feeders because dense populations can rapidly rework bottom deposits through selective ingestion of silt and clay (Dafoe et al., 2011). Jiang et al. (2010) compared bioturbation efficiency in *Chironomus plumosus* and *Tanytarsus chinensis* both of arthropoda; the authors demonstrated that the two organism types differ greatly in their feeding and burrowing activities, and certain Chironomid species may play important roles in driving nutrient cycling. Other researchers (Cardoso et al., 2008; Michaud et al., 2006) found that the gallery-diffusers (e.g., *Hediste diversicolor*) have greater effects than bioturbators (e.g., bivalves) on the biochemical processes in marine sediment. Gallery-diffuser animals produce diffusion in the layer with very dense gallery systems and a bio-transport at the end of the burrows. The fauna burrows usually have significant effects on chemical fluxes and microbial activity. As *H. diversicolor* burrows deeper into the sediment, it irrigates a great volume of sediment, having a great influence on pore water chemistry, ammonium release and active bacteria. Additionally, these different activities of different organisms may have different effects on the heavy metal release, but there was little research on the effects before, especially for the riverine and reservoir conditions.

The aim of the present study was to investigate the effect of bioturbation/bioirrigation on the Tl remobilization from sediments to the water using Tl contaminated sediments in batch culture with three different types of organisms included Tubificid (Annelida), Chironomid larvae (arthropod) and Loach (vertebrate) as bioturbator. Furthermore, the accumulation of the Tl within the organisms was investigated.

## 2. Materials and methods

### 2.1. Preparation of contaminated sediment and overlying water

The sediment used as the original matrix was collected from uncontaminated surface sediment (0–30 cm) in the Ming Tombs Reservoir in north China in May 2013. Removed the gravel, plants and other impurities of the sediment and homogenized by mechanical mixing. The original component of the sediment was measured by a wavelength dispersive X-ray fluorescence spectrometer (WDXRF) (Rigaku ZSX Primus II) (Table 1). The water used as the overlying water was collected in the same place of the reservoir before collecting the sediment to avoid turbidity. The two were stored at 4 °C in the dark before use. Before being added to the experimental units, the sediment was spiked with  $\text{TlNO}_3$  (AR, Sinopharm Chemical Reagent Co., Ltd. China) and the nominal Tl concentration being  $10.0 \pm 1.1$  mg/kg (dry weight) in the sediment. The artificially contaminated sediment was aged at room temperature for two months and stirred to allow uniform contamination.

### 2.2. Organism

Disturbed biologically selected Tubificid was collected from the pool in Fangshan southwest of Beijing, Chironomid larvae (*C. plumosus* larvae) were collected from the uncontaminated Yangliuqing River in the outer suburbs of Tianjin, and Loach were from the Miyun reservoir in the northeast of Beijing. The three organisms were cultured in clean water for no less than 48 h by cultivating them in laboratory conditions before use.

### 2.3. Experimental design

This experiment simulated the natural aquatic environment, carried out in an artificial climate chamber (RXZ intelligent, Ningbo Jiangnan Instrument Factory). The temperature was maintained at 23 °C, humidity remained 50% and the daily period of light at 16:24-hour throughout the entire experiment.

#### 2.3.1. Bioturbation/bioirrigation effects on Tl released from the sediment

The basic structure of the experimental systems consisted of a glass beaker (2 L,  $\Phi 13 \times H 19$  cm), lined with a high density polyethylene film. Sediment mesocosms were established by transferring 10 cm depth of spiked and free fauna sediment to each glass container. Then, 600 mL of filtered (0.45  $\mu\text{m}$ ) reservoir water was carefully added to each glass container in order to avoid disturbance at the sediment surface. The glass containers were distributed in room temperature for a 24 h period of stabilization before introducing the organisms. After the 24 h stabilization equilibrium period, 2.0 gram organism ( $7.5 \times 10^5$  Tubificid per square meter,  $9.4 \times 10^4$  Chironomid larvae per square meter,  $1.5 \times 10^2$  Loach per square meter) was introduced into each glass container. The system with no organism added was used as a bioturbation/bioirrigation experiment blank. Four replicates were done for each condition. The complete experimental design consisted of 16 microcosms set up simultaneously. At the exposure time 1, 2, 3, 5, 7, 9, 14, 21 and 28 days, 10 mL of unfiltered water was sampled from each experiment beaker with a polypropylene syringe, then microwave digestion (MARS, China Everbest Machinery Industry Co., Ltd.) with aqua regia, stored at 4 °C and analyzed for total heavy metals within 3 days. 10 mL of filtered water was also sampled from each experiment beaker, filtered through a 0.45  $\mu\text{m}$  mixed cellulose ester membrane filter (0.45  $\mu\text{m}$ ,  $d = 25$  mm, Millipore, USA), then acidified with nitric acid and analyzed for dissolved heavy metals. The overlying water was added after each sampling and aeration for 5 min. The pH and turbidity of the overlying water were measured for each unit during the exposure time of 1, 2, 3, 5, 7, 9, 11, 14, 17, 21, 24, and 28 days. Thallium was determined by inductively coupled plasma mass spectrometry (ICP-MS) (7500a, Agilent Technologies Co. Ltd., America).

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