



Research papers

Effect of recurrent sediment resuspension-deposition events on bioavailability of polycyclic aromatic hydrocarbons in aquatic environments



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ABSTRACT

To investigate the effect of recurrent sediment resuspension-deposition events (RSRDEs) on bioavailability of polycyclic aromatic hydrocarbons (PAHs) in aquatic environments, a modified device was used to simulate three resuspension-deposition events with the sediment collected from the Yellow River. The results showed that the dissolved organic carbon (DOC)-water distribution coefficients of PAHs decreased with time during the first resuspension-deposition period. It indicates that some PAHs associated with organic carbon (OC) in suspended sediment (SPS) desorbed with the release of OC and became DOC-associated PAHs in the overlying water, then the PAHs desorbed from the DOC and became freely dissolved. After first 2-h suspension, only 1.90% of phenanthrene, 2.98% of pyrene, and 0.33% of chrysene in the overlying water came from pore-water; at least 61.6%, 89.6%, and 95.3% came from DOC-associated PAHs in SPS and the rests were released from the insoluble OC in SPS. The maximum desorption ratios in the original sediment were 20%, 12%, and 14% for phenanthrene, pyrene, and chrysene, respectively during the first resuspension-deposition event. The SPS concentration followed the sequence of the third > second > first resuspension event. This was because RSRDEs changed the SPS particle size and enhanced floc formation. There was no significant difference in the total dissolved PAH concentrations among the three resuspension events, while their freely dissolved concentrations followed the sequence of the third > second > first resuspension event. During deposition periods, more than half of the total/freely dissolved PAHs released during suspension still existed in the overlying water after 70-h deposition. This study suggests that the RSRDEs will increase the bioavailability of PAHs in aquatic environments, especially near the sediment-water interface, and the potential effects of PAHs during RSRDEs on fish/human in rivers and lakes should be considered in future management.

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1. Introduction

Hydrophobic organic compounds (HOCs) have a high affinity to suspended and deposited sediments in aquatic environments. Fine-grained bottom sediments can accumulate HOCs and act as an important sink for HOCs, thus reducing the bioavailability of HOCs to aquatic organisms (Lai et al., 2015; Xia et al., 2013; Zhang et al., 2014). However, once the sediment is disturbed, the resuspended particle will enter into the overlying water and may release contaminants, resulting in HOC being bioavailable in the overlying water. Therefore, sediment resuspension would exert great influences on HOC concentrations and water quality in

aquatic environments (Dong et al., 2015; Friedman et al., 2009; Huang et al., 2015; Pusceddu et al., 2005; Superville et al., 2015).

Sediment resuspension is a ubiquitous process in aquatic environments (Schoellhamer, 1996). It occurs when shear stress (friction of the water against the bottom) is high enough to move the surface sediment particles (Tengberg et al., 2003). Sediment resuspension event can be caused by waves (Ferré et al., 2008), storms (Siadatmousavi and Jose, 2015), currents (Ferré et al., 2008), dredging (Je et al., 2007), trawling (Durrieu de Madron et al., 2005; Pusceddu et al., 2005), ship traffic (Superville et al., 2015), and the operation of water conservancy projects (Dong et al., 2015). All these factors, especially anthropogenic activities, often occur regularly, leading to recurrent resuspension-deposition of sediment.

Three major hydrodynamic factors influencing sediment resuspension are duration, magnitude, and frequency of resuspension.

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Among them, the roles of sediment resuspension duration (Feng et al., 2008; Ståhlberg et al., 2006) and magnitude (Qian et al., 2011; Ståhlberg et al., 2006; Wang et al., 2009; Wang and Yang, 2010) have been studied comprehensively. However, the effect of resuspension frequency has been less understood and most researches focused on single resuspension or re-deposition event (Chalhoub et al., 2013; Kalnejais et al., 2007; Wang and Yang, 2010; Yang et al., 2008). Sediment resuspension-deposition often occurs repeatedly in river and lake systems. Based on the results reported by Schoellhamer (1996), the typical duration for most types of sediment resuspension event is hours, and sediment resuspension occurs hourly to weekly. Furthermore, the effect of recurrent sediment resuspension on water quality might be different from single resuspension event. For example, during the operations of trawling and ship traffic, the bottom sediments resuspend and release contaminants into overlying water. After that, the suspended sediments (SPS) deposited as a “new” layer on the surface of bottom sediments, which is different from the original surface layer of the bottom sediments. Once the hydrodynamic condition in the water-sediment systems is changed, the deposited sediment (the new layer) will resuspend again, and the effect of the second suspension event on the water quality and HOC concentrations may be different from the effect of the first suspension event.

However, very little work has been conducted to study the effect of recurrent sediment resuspension-deposition events (RSRDEs) on the HOC concentrations in aquatic environments. Only some researchers suggested that the effect of quiescent time between resuspension events on HOC release was significant (Schneider et al., 2007; Superville et al., 2015). In addition, the previous research mainly focused on the effect of sediment resuspension on HOC concentrations in the overlying water. Few researches have been reported on the variations of HOC bioavailability during the sediment resuspension-deposition events, which can reflect their bioaccumulation in and toxic potential to aquatic organisms.

Polycyclic aromatic hydrocarbons (PAHs), a class of HOCs, are recognized as chemicals of high concern due to their extensive sources, persistence, and potentially toxic effects on ecosystems and human health, and they are ubiquitous in aquatic environments (Liu et al., 2010; Xia et al., 2015; Zhang et al., 2014). Therefore, the PAHs including phenanthrene, pyrene, and chrysene were selected as a typical kind of HOCs to investigate the effect of RSRDEs on the bioavailability of HOCs in aquatic environments. Water and sediment samples were collected near the Xiaolangdi Dam of the Yellow River, the largest turbid river in the world. Three sediment resuspension-deposition events were conducted with these samples through using a modified turbulence-simulation device. The freely dissolved concentrations were used to indicate the bioavailability of PAHs. Both the freely and total dissolved concentrations of the three PAHs in the overlying water among the three resuspension and deposition events were compared. The concentration, particle size, and composition of SPS were analyzed, and the relative contributions of PAHs released from SPS and from pore-water to those in the overlying water during the first resuspension-deposition event were estimated. In addition, the physicochemical mechanisms (such as the desorption/re-sorption of PAHs as well as their equilibrium between SPS and water) regarding the variations in PAH bioavailability during the RSRDEs were also explored.

2. Materials and methods

2.1. Chemicals and glassware

Three standard PAHs (phenanthrene, pyrene, and chrysene) and three deuterated PAHs (*d*10-phenanthrene, *d*10-pyrene, and

*d*12-chrysene) were purchased from JT Baker (Philipsburg, NJ, USA). The physical-chemical properties of the studied PAHs are listed in Table S1 (Supplementary data). The internal standard (*m*-terphenyl) and the recovery standard (2-fluorobiphenyl) were obtained from J&K Scientific Ltd. (New Haven, C.T., USA). Other HPLC grade solvents including dichloromethane (DCM), methanol and hexane, were supplied by JT Baker (Philipsburg, NJ, USA). All glassware was pretreated as follows: soaked in an acid solution for 24 h, afterward washed with tap water, distilled water, and ultrapure water in sequence; then dried in an oven (105 °C) and heated at 450 °C in a muffle furnace for 4 h.

2.2. Sediment collection and pretreatment

The 0–10 cm surface sediment samples were collected from the Yellow River near the Xiaolangdi Dam (112°26′46.5″E, 34°55′18.5″N) in July 2013 using a Van Veen stainless steel grab sampler (Eijkelamp, Netherlands). The macroscopic gravel and macrofauna in the wet sediment were removed. Then parts of the homogenized wet sediment were used to analyze the physical-chemical properties and PAH concentrations in sediment and in pore-water. The physical-chemical properties of the sediment including particle size, composition, total organic carbon (TOC) content, black carbon (BC) content, and water content were determined. The sediment was first ground with agate mortar to pass through a 100 mesh stainless steel sieve. TOC contents in sediment were measured by an elemental analyzer (Vario El, Elementar Analysensysteme GmbH, Germany) after treated with HCl (1:1, v/v). BC contents were determined with the chemo-thermal oxidation method. Inorganic carbon in samples was firstly removed by HCl (1:1, v/v); amorphous organic carbon was subsequently removed in a thermal oxidation procedure at 375 °C in a tube furnace for 24 h in the presence of excess air. Then BC content in sediments was also determined with the elemental analyzer. The particle size of samples was determined with a Microtrac S3500 Laser Particle Size Analyzer (Microtrac Inc., Montgomeryville, PA). Due to the sandy nature of the sediment (Fig. 2), sediment pore-water was directly obtained by the filtering of the supernatants formed within minutes using 0.45- μ m nylon syringe filters without the aid of a centrifuge. Possible effect of the nylon filters on aqueous PAHs was considered and no significant effect was found based on the determination of freely dissolved PAH concentrations before and after filtration. The PAH concentrations in the pore-water and sediment were determined in triplicate. In addition, another part of the wet sediment was sieved successively through 300 μ m, 150 μ m, and 50 μ m sieves, then the sediment was classified into four categories: >300 μ m, 150–300 μ m, 50–150 μ m, and 0–50 μ m. The four categories were freeze-dried for 72-h prior to extraction.

2.3. Sediment resuspension experiment

The sediment resuspension simulation was conducted with a turbulence simulation device (Fig. 1). This device consists of a vertical acrylic cylinder (60 cm height and 25 cm diameter) and a set of oscillating vibratory grids driven by a variable speed motor (Wang et al., 2009). A total of 1 kg (wet weight) sediment was added into the cylinder. The ultrapure water (20 l) with 0.2 g/l NaN₃ (avoiding microorganism activities) was slowly injected into the cylinder to avoid disturbing the sediment. Then the cylinder was covered with a lid and kept under a static condition for 7 days. During sediment suspension, the perforated grids were oscillated with vertical amplitude of 3.2 cm (between 5.0 and 8.2 cm above from the sediment surface), and they were driven by the speed motor with 150 rpm, corresponding to 6.4 N/m², which was in the range of erosion shear stress (0.2–75 N/m²) found in natural rivers (Babaeyan-Koopaei

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