



Effect of Tenax addition amount and desorption time on desorption behaviour for bioavailability prediction of polycyclic aromatic hydrocarbons

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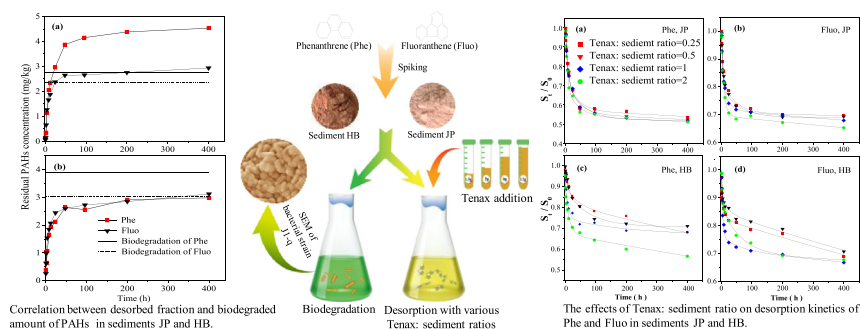
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HIGHLIGHTS

- Desorption kinetics of PAHs were fitted well by first-order three-fraction model.
- Tenax: sediment ratio of 1 could satisfactorily predict bioavailability of PAHs.
- Tenax showed better correlation with biodegradation of high molecular weight Fluo.
- The rapidly desorbing fraction was estimated over a 24 h Tenax desorption period.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, Tenax consecutive extractions of polycyclic aromatic hydrocarbons (PAHs) were conducted in two spiked sediments to investigate the influence of different Tenax addition amounts and desorption times on the rapidly desorbing fraction of PAHs, and to determine a reliable method for estimating PAHs bioavailability. The results indicated that a large Tenax addition amount has a positive effect on the desorption of PAHs from sediments. The desorption amounts of target PAHs compounds (3-ring phenanthrene and 4-ring fluoranthene) increased as the Tenax: sediment ratios increased from 0.25 to 2 in two spiked sediments. The highest desorption percentages of phenanthrene and fluoranthene were 48.91% and 34.70% for Jialing industrial park sediment, and 43.36% and 33.24% for Huanghuayuan bridge sediment, respectively. The results of desorption kinetics were suitably fitted with first order three-compartment model to estimate the rapidly desorbing fraction. Moreover, the Tenax: sediment ratio of 1 and desorption time of 24 h were found to be suitable for the desorption of phenanthrene and fluoranthene from sediments. The PAHs in sediments were biodegraded well by the bacterial strain J1-q. Comparing the maximum biodegraded amount of target PAHs in 30 days and the desorbed fraction over 400 h, the results showed that Tenax had better correlation with the high molecular weight fluoranthene than with the low molecular weight phenanthrene.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a kind of persistent organic pollutants, which pose a risk for human health and ecological

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environment due to their toxic, mutagenic and/or carcinogenic properties (Kohlmeier et al., 2008; Nzila, 2018; Zhang et al., 2017). PAHs have both natural (e.g., forest fires and volcanic activities) and anthropogenic sources (e.g., traffic, fossil fuel combustion, emissions from power plants, etc.) (Duan et al., 2013; Lau et al., 2014). Due to their hydrophobicity, the majority of PAH compounds have strong sorption tendency with natural organic matter in soils/sediments (Falciglia et al., 2018). As a result, soils/sediments act as a sink and are enriched with high concentrations of PAHs (Congiu and Ortega-Calvo, 2014). Recently, many researchers have used the bioavailability of PAHs to evaluate the environmental risks and pollution levels of contaminants, which provided the foundation for qualitative prediction of bioremediation of PAHs in sediment/soil (Fonseca et al., 2011; Wang et al., 2017; Yang et al., 2010). The term “bioavailability” can be defined as the extent to which the target compounds are available to microorganisms for the biodegrading process (Barnier et al., 2014). Bioavailability of PAHs is affected by adsorption-desorption, extraction, biodegradation, soil/sediment properties, aging time, etc. (Cui et al., 2013; Duan et al., 2014; Enell et al., 2005; Ma et al., 2012; Rhodes et al., 2010; Spasojevic et al., 2015). The extent of biodegradation of PAHs is controlled by the adsorption-desorption phenomenon due to the hydrophobicity of PAHs and their interaction with organic matter (Clegg et al., 2014; Congiu and Ortega-Calvo, 2014; Rhodes et al., 2010). Previous studies have reported that the release of PAHs from sediment was restricted after adsorption on organic compounds in sediments or soils, thus limiting their bioavailability (Barnier et al., 2014; Bogan and Sullivan, 2003).

Investigating the desorption process of PAHs in sediment/soil is important for predicting the bioavailability of contaminants. Up to now, two-fraction and three-fraction models have been applied widely (Hulscher et al., 2003; Rhodes et al., 2010; Shor et al., 2003; Spasojevic et al., 2015). The two-fraction model includes rapidly and slowly desorbing fractions, whereas the three-fraction model includes rapidly, slowly and very slowly desorbing fractions (Greenberg et al., 2005; Huang et al., 2017; Rhodes et al., 2012). The rate constant of very slow desorption is at least an order of magnitude lower than the corresponding value for slow desorption (Greenberg et al., 2005). Moreover, the very slow desorption stage controls the release of PAHs from sediments into solution. The rapidly desorbing fractions diffuse from soil/sediment surface to aqueous phase with a rapid rate (Cornelissen et al., 1997a). Several recent studies have demonstrated that the rapidly desorbing fraction has a correlation with the bioavailable fraction in both natural contaminated soils and spiked sediments (Cornelissen et al., 1997b; Sinche et al., 2017; You et al., 2006). The rapidly desorbing fraction has been predicted using suitable extraction techniques. Recently, various extracting agents have been applied to predict the bioavailability of PAHs, such as methyl- β -cyclodextrin (MCD), hydroxypropyl- β -cyclodextrin (HPCD), mild organic solvents, XAD resin, Tenax polymer resin, etc. (Li et al., 2015; Wang et al., 2017; Yang et al., 2013; Yang et al., 2011; Zhou et al., 2013). Consecutive Tenax extraction has been applied to investigate the desorption behaviour of PAHs and is considered as the main technique for predicting the bioavailability of PAHs (Liu et al., 2011; Sun et al., 2014).

It has been reported that an insufficient or excess Tenax: sediment/soil ratio would underestimate or overestimate the PAHs bioavailability from sediments and soil (Bernhardt et al., 2013). Therefore, a suitable Tenax: sediment/soil ratio is a critical factor for predicting the bioavailability. Recently, researchers have studied bioavailability of PAHs using Tenax extraction with a specific Tenax: sediment/soil ratio (Guo et al., 2016; Liu et al., 2011; Yang et al., 2010). However, relatively little research has focused on the effect of different Tenax addition amounts on the PAHs desorption from sediments. Moreover, using an appropriate extraction time to estimate the rapidly desorbing fraction could minimize the risk of underestimation of PAHs bioavailability (Barnier et al., 2014). Previous reports have suggested that Tenax extraction of different PAH-contaminated soils/sediments requires different

extraction times for estimating the rapidly desorbing fraction (Barnier et al., 2014; Cornelissen et al., 2001; Shor et al., 2003). Cornelissen et al. (2001) reported that an extraction time of 6 h was not suitable for certain contaminated industrial soils, which is inconsistent with other report (Yang et al., 2010). There is still some debate regarding the suitable extraction time to estimate the rapidly desorbing amount of PAHs.

The desorption of PAHs in sediments was conducted using consecutive Tenax extraction with four Tenax: sediment ratios. The desorption was then correlated with the biodegradation potential to predict the bioavailability of PAHs. The objectives of this study are to (i) investigate the effect of various Tenax: sediment ratios on rapidly desorbing fraction of PAHs to determine the optimum Tenax: sediment ratio, (ii) determine the correlation between the desorption and biodegradation potential to predict the bioavailability of PAHs, and (iii) use different desorption kinetics models to investigate the desorption behaviour and define the optimum desorption time to estimate the rapidly desorbing fraction from sediment.

2. Materials and methods

2.1. Chemicals

Tenax-TA adsorbent polymer resin (60–80 mesh) was purchased from Shanghai ChengMai Technology Co., Ltd. (Shanghai, China). Before use, the Tenax-TA beads were pretreated with acetone and hexane (1:1, v/v), then dried overnight at 75 °C in previous reports (Barnier et al., 2014; Wang et al., 2017). Phenanthrene, fluoranthene, phenanthrene-d10 and chrysene-d12 (purity >99%) were obtained from TianChang Chemicals Co. (Anshan, China). Acetone, hexane, dichloromethane, and other solvents were obtained from ChuanDong Chemical Engineering Co., Ltd. (Chongqing, China). All other solvents and chemicals were of analytical grade or chromatography grade purity.

2.2. Sediments and spiking

2.2.1. Sediments

The tested sediments were collected from two sites that are representative of PAHs pollution at the surface (0–20 cm): former Jialing industrial park (JP) and Huanghuayuan bridge (HB) along Jialing River in Chongqing, China. The sediment were air dried at room temperature in darkness, homogenized, sieved to 2 mm particle size, and stored in brown glass bottles at 4 °C. The main properties of the sediments are listed in Table 1. The contents of total organic carbon in sediments were measured by the Walkley–Black procedure (Chakraborty et al., 2014). And the amounts of humic acid were measured with the dilute alkali method set by International Humic Substance Society (Swift, 1996).

The initial concentrations of 3-ring and 4-ring PAH compounds were 31% and 29% of the total concentrations of PAH in sediment JP, respectively, while the corresponding values in sediment HB were 30% and 35%, respectively. The target PAH compounds in this study were chosen

Table 1
Main characteristics of the sediment samples.

Sediment type		JP	HB
Particle size analysis (%)	Clay	21.5	26.7
	Fine sand	16.3	15.2
	Silt	35.6	41.9
Initial concentration analysis	\sum 16 PAHs (mg/kg)	6.0	6.3
	Phenanthrene % ($W_{\text{Phenanthrene}}/W_{\sum 16 \text{ PAHs}}$)	26.9	26.8
	Fluoranthene % ($W_{\text{Fluoranthene}}/W_{\sum 16 \text{ PAHs}}$)	10.3	9.8
pH	8.4	8.4	
Total organic carbon (%)	2.3	1.6	
Humic acid (mg/g)	9.6	8.4	

JP, Jialing industrial park sediment site; HB, Huanghuayuan bridge sediment site.

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