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Modeling the transport of sodium dodecyl benzene sulfonate in riverine sediment in the presence of multi-walled carbon nanotubes

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ABSTRACT

The environmental risks of carbon nanotubes have received considerable attention. In this work, the effects of multi-walled carbon nanotubes (MWCNTs) on the adsorption of sodium dodecyl benzene sulfonate (SDBS) by riverine sediment and the transport of SDBS in sediment were studied. MWCNTs could significantly increase the adsorption capacity of the sediment for SDBS, thus affecting the transport of SDBS in sediment. Maximum adsorption capacity of the sediment for SDBS increases from 2.29 to 2.99 mg/g with the increasing content of MWCNTs from 0% to 1.5%. Breakthrough curves (BTCs) of SDBS obtained from the column experiments were described by the convection-dispersion equation, Thomas model, and Yan model. The estimated retardation factor R increases with the incorporation of MWCNTs, either in water or sediment. Additionally, the value of R is well correlated to the content of MWCNTs in sediment. Compared with Thomas model, Yan model is more suitable for fitting the BTCs with all the values of $R^2 \geq 0.951$, but it tends to overestimate the maximum adsorption capacity when the content of MWCNTs in sediment is relatively higher. The results of SDBS retention in sediment indicate that MWCNTs can increase the accumulation of SDBS in the top sediment layer, while they can impede the transport of SDBS into deeper sediment layer when incorporated into the sediment. These effects should be taken into consideration for risk assessment of CNTs in the aquatic environment.

1. Introduction

Carbon nanotubes (CNTs), composed of carbon atoms in a periodic hexagonal arrangement, are hollow cylinders with a diameter in the nanometer range. Single-walled nanotubes (SWCNTs) and multi-walled nanotubes (MWCNTs) are two main types of CNTs. Since their observation was first reported by Iijima in 1991 (Iijima, 1991), CNTs have been attracting much attention of researchers because of their unique mechanical, thermal, optical, and electronic properties, as well as many potential applications (Popov, 2004; Zhang et al., 2007; Huang et al., 2008; Tang et al., 2008; De Volder et al., 2013). Current production capacity of CNTs worldwide has exceeded 5000 tonnes per year, and is increasing at an annual growth rate of 32.5% (Patel, 2011; De Volder et al., 2013). Increasing production and application of CNTs will inevitably result in the release of these nanomaterials into the environment. In a multimedia environment (atmosphere, soil, water, and sediment), mass accumulation of CNTs was mostly in soil and sediment (Yang et al., 2010; Liu and Cohen, 2014). Based on the research of Koelmans et al. (2009), the estimated concentrations of manufactured carbon-based nanoparticles in aquatic sediment are ranging from 1.2 to 2000 μg per kilogram of the dry sediment. And it is likely that the concentrations of CNTs in sediment will increase in the future.

CNTs have strong adsorption affinity for various organic and inorganic contaminants (Gong et al., 2009; Song et al., 2017a, 2017b). As sediment is also the ultimate reservoir of various contaminants in aquatic ecosystem, the interaction between CNTs and contaminants may alter the fate and transport of these contaminants, significantly influencing their mobility, toxicity, and bioavailability (Xu et al., 2012a; Zeng et al., 2013a, b; Cheng et al., 2016). For example, Sun et al. (2015) found that CNTs released

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into sediment would increase the adsorption capacity of Cd(II) by sediment. Fang et al. (2013) demonstrated that TX100 suspended MWCNTs could facilitate the transport of phenanthrene in soil columns, while Li et al. (2013) reported that 5 mg/g CNTs could significantly retain polycyclic aromatic hydrocarbons in soil. Recent research by Liang et al. (2016) showed that CNTs could enhance the mobility of tetrabromobisphenol A in saturated porous media. Zhang et al. (2017) also observed facilitated transport of chlordecone and sulfadiazine in the presence of CNTs in soil. However, studies investigating the effect of CNTs on the transport of contaminants in real riverine sediment were insufficient.

Since sodium dodecyl benzene sulfonate (SDBS) is commonly used to increase the dispersity and stability of CNTs in aqueous solutions, most of the current studies focused on the effect of SDBS on the properties, transport, and fate of CNTs (Tian et al., 2011; Ju et al., 2012; Wusiman et al., 2013). However, few studies investigated the effect of CNTs on the transport and fate of SDBS. As an anionic surfactant, SDBS is usually present in detergent, soap, as well as cosmetic, and widely used as emulsifier, dispersant, lubricant, and preservative in industrial processes (Myers, 2005; Taffarel and Rubio, 2010). Because of its extensive applications, a large amount of SDBS is released into the aquatic environment, causing serious environmental problems. The adverse effects of the surfactant on the aquatic environment and human health have been studied and reported elsewhere. According to the available literature, SDBS exhibits toxic effects towards algae, benthic invertebrates, fishes, and human cells (Qv and Jiang, 2013; Mu et al., 2014; Zhang et al., 2015, 2016). Considering the ecological and human health risks of SDBS, the environmental behavior of SDBS in the presence of CNTs in the aquatic environment should be studied.

In this study, research on the transport of SDBS in riverine sediment in the presence of MWCNTs was conducted. The objectives of the present study were (1) to investigate the effect of MWCNTs on the adsorption of SDBS by sediment, and (2) to study the transport of SDBS in the presence and absence of MWCNTs in riverine sediment by column experiments and numerical modeling.

2. Materials and methods

2.1. Chemicals, sediment, and carbon nanotubes

SDBS ($C_{18}H_{29}NaO_3S$, AR) was purchased from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. All other reagents in this study were of analytical grade or better and commercially available. Surface sediment samples (0–15 cm) were collected from Changsha section of the Xiangjiang River in Hunan Province, China. Sediment samples were air-dried at room temperature and then crushed in a porcelain mortar. Subsequently, the samples were sieved over a one mm mesh sieve and homogenized prior to use. Sediment properties including pH, zeta potential, electrical conductivity, organic carbon content, cation exchange capacity, and texture (sand, silt, and clay) were measured with the methods mentioned in previous literature (Song et al., 2017b). Industrial grade MWCNTs with an outer diameter of 10–20 nm and a length of 5–10 μm were used in this study. They were purchased from Chengdu Organic Chemistry Co., Chinese Academy of Sciences, Chengdu, China.

2.2. Batch adsorption experiments

Kinetics experiments of SDBS adsorption onto MWCNTs, sediment, and sediment-MWCNTs mixtures (content of MWCNTs in sediment, w/w: 0.5%, 1.0%, and 1.5%) were performed in 250 mL conical flasks containing 20 mg/L SDBS on a shaker at 180 rpm, 25 ± 1 °C. Absorbent dosage of MWCNTs was 0.3 g/L, while the dosages of sediment, and sediment-MWCNTs were both 20 g/L. The samples were taken out from the flask after predetermined time intervals (from 30 s to 300 min), and the concentrations of SDBS were determined by high performance liquid chromatography (HPLC, Agilent 1100, USA) equipped with UV–vis variable wavelength detector (VWD) and reversed-phase C18 column. Methanol (90%, v/v) was used as the mobile phase at a flow rate of 1 mL/min with constant detection wavelength at 224 nm.

Adsorption isotherm experiments were conducted in conical flasks containing SDBS solutions of different concentrations (from 10 to 80 mg/L) on a shaker at 180 rpm, 25 ± 1 °C. The dosages of absorbents were the same as those in kinetics experiments. After a 2 h equilibrium, samples were taken out and the concentrations of SDBS were determined by the above-mentioned analytical method of HPLC.

2.3. Column experiments

Column transport experiments of SDBS in various sediment columns were carried out under saturated flow conditions. A Teflon column with a length of 300 mm and an inner diameter of 24 mm was used in the experiments. The column packing was based on previously reported methods with appropriate modifications (Zhuang et al., 2003; Tričković et al., 2016). Concretely, a stainless steel wire mesh with pore size of 0.14 mm was placed at the bottom of the column, and then a quartz sand (25–50 mesh) layer of 10 mm was added for supporting the sediment particles. Subsequently, ultrapure water was introduced into the column from the bottom to a certain height with a peristaltic pump (DDB-300, Zhisun Equipment Co., Ltd., Shanghai, China). After that, sediment was slowly poured into the column by 5–6 mm increments until the column was packed to a height of 48 mm. During the packing process, the sediment in the column was stirred with a glass rod to ensure homogeneous packing and to avoid air entrapment. After the sediment column was prepared, a potassium bromide (KBr) solution was used as a conservative tracer for characterizing the sediment column and the hydraulic conditions.

Two sets of column transport experiment, denoted as Set I and Set II, were conducted. In Set I, 50 mg/L SDBS, 50 mg/L SDBS containing 0.3 g/L MWCNTs (reached adsorption equilibrium in advance), and 32 mg/L SDBS (an equilibrium concentration of 50 mg/L SDBS containing 0.3 g/L MWCNTs) were pumped respectively to the top of the sediment columns. For Set II, sediment in the column was mixed with MWCNTs, and the contents of MWCNTs were 0.5%, 1.0%, and 1.5% (w/w), respectively. In this set, 50 mg/L SDBS solutions were pumped to the columns by peristaltic pump and transported from the top down. The duration of each column transport experiment was 24 h for both Set I and Set II. During the column experiments, a constant water head of 150 mm was maintained and the effluent was collected at regular intervals for the measurement of SDBS concentrations.

At the end of the column experiment, sediment in the column was air-dried and equally divided into 6 segments. Then SDBS concentrations in these segments were analyzed based on previously reported method (Santos et al., 2007). In short, SDBS in the sediment was extracted with methanol. For each extraction, 20 mL methanol was added into the beaker containing the sediment, followed by shaking the beaker on a shaker for 5 min. Afterwards, the sample was sonicated for 30 min and then centrifuged to collect the supernatants. After filtered through 0.45 μm filter membrane, the SDBS concentration in the filtrate was determined by HPLC.

2.4. Numerical modeling

The obtained data of adsorption kinetics were fitted with