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Release of pentachlorophenol from black carbon-inclusive sediments under different environmental conditions

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ABSTRACT

To investigate the feasibility of using black carbon (BC) in the control of hydrophobic organic contaminants (HOCs) in sediment, we added BCs from various sources (rice straw charcoal (RC), fly ash (FC) and soot (SC)) to sediment to create different BC-inclusive sediments and studied the release of pentachlorophenol (PCP) in the sediments under different condition. Different pH values had no obvious effect on the release of PCP in BC-inclusive sediment, but solid/liquid ratio, temperature, salinity and dissolved organic matter (DOM) content had significant influences on the release of PCP in all sediments except the RC-inclusive sediment. Adding 2% RC to sediment resulted in a 90% decrease in PCP release, which was a greater decrease than observed with FC- and SC-inclusive sediments. Therefore, from the standpoint of HOC release, the application of RC is feasible for organic pollution control in the water environment.

1. Introduction

Recently, black carbon (BC) has received much attention from environmental chemists because of its widespread distribution in sediments and its potential role in controlling the fate and toxicity of many hydrophobic organic contaminants (HOCs), although BC content is commonly low in sediments (Cornelissen et al., 2005; Lohmann et al., 2005; Cui et al., 2009; Xia et al., 2010; Lou et al., 2011). To date, there is increasing recognition that the strong interaction between HOCs and BC could greatly reduce the mobility, bioavailability and environmental risk of HOCs in sediments (Vinturella et al., 2004; Cornelissen et al., 2005; Lohmann et al., 2005; Ferguson et al., 2008; Cui et al., 2009; Lou et al., 2011).

The fate of HOCs is primarily dominated by the environmental conditions and the chemical nature of the pollutants (Fisher et al., 1999; Mäenpää et al., 2008). In real aquatic system, environmental factors such as the re-suspension of sediment, pH value, temperature, salinity and dissolved organic matter (DOM) content change widely, causing pollutants adsorbed by sediment to release to the overlying water, resulting in secondary pollution. For example, the solid/liquid ratio can influence the concentration of pollutants in water (Shrotri et al., 1998; Vilar et al., 2007), pH values may alter the surface properties of sediment and existing forms of pollutants (Shimizu et al., 1992; Stapleton et al., 1994; You et al., 2010), temperature plays an important role in affecting the sorptive behavior of pollutants to sediment (Xu and Li, 2009), salinity is able to change the sorptive strengths of pollutants (Stapleton et al., 1994; You et al., 2010; Uchimiya et al., 2011), and the sorptive properties of sediments are affected by DOM, which competes with sediments to adsorb contaminants (Kwon and Pignatello, 2005; Cao et al., 2008; Xu and Li, 2009; Uchimiya et al., 2011).

However, studies about the influences of different environmental factors on the release of HOCs by BC-inclusive sediments are scarce. There are neither published reports concerning the release of HOCs, nor information about the effects of various environmental conditions on the release of HOCs in different BC-inclusive sediments. Therefore, it is necessary to investigate the effects of different environmental factors on the release of HOCs in BC-inclusive sediments to provide a theoretical basis for the application of BC to treat sediment polluted with HOCs.

To clarify the release of HOCs from BC-inclusive sediment, we treated sediment with different BCs, including rice straw charcoal, fly ash, and diesel soot. Simultaneously, we investigated the effects of solid/liquid ratio, pH, temperature, salinity, and DOM content on the release of HOCs in sediments containing BC. In this paper, pentachlorophenol (PCP) was chosen as a representative HOC because of its stable aromatic ring structure and high chlorine content (Piringer and Bhattacharya, 1999; Scelza et al., 2008).

2. Materials and methods

2.1. Chemicals and materials

Pentachlorophenol (PCP), with a purity of >98%, was purchased from Sigma Aldrich (China) and prepared in a concentrated stock

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solution with methanol. Sediment was obtained with a clam sampler from 1 to 10 cm surface sediment of the Qiantang River, China. Physical and chemical properties of the sediment are shown in Table 1. Pollutants were present in the sediment, including polychlorinated biphenyl (PCB) (0.0459 mg kg^{-1}), Cr (0.0429 mg kg^{-1}), Zn (0.1683 mg kg^{-1}), Cd (0.0021 mg kg^{-1}), and Cu (0.0764 mg kg^{-1}), but the sediment did not contain a detectable quantity of PCP.

Air-dried rice straw collected from the Hua-jia-chi Campus of Zhejiang University in China was burned on a stainless steel plate in an open field under uncontrolled conditions on a still and sunny afternoon. Fly ash produced during the combustion of coal (at 1400 °C) was collected on electrostatic filters at the Hangzhou Thermoelectric Plant, Zhejiang province, China. Soot was purchased from the Degussa Company, Germany. To remove mineral materials, 2 M HCl solution (200 mL) was added into each BC sample (10 g) for 24 h at 298 K, and the supernatant was removed through centrifuging the HCl solution and BC sample at 4800 rpm for 20 min. This procedure was repeated 4 times, and then 1 M: 1 M HCl-HF solution was used instead of 2 M HCl to conduct this procedure five times. After that, all the treated BCs were washed about five times by distilled water to remove residual acids, Si, and soluble salts (Chun et al., 2004; Lou et al., 2011). The treated rice straw charcoal, fly ash, and soot were oven-dried overnight at 105 °C, and they were called RC, FC, and SC. Characteristics of the BCs are shown in Table 2.

2.2. Spiking of BC-inclusive sediments with PCP

BC-inclusive sediments used in the experiment were prepared by mixing sediment and BC at different ratios. The percentages of BC added to sediment were 0%, 0.5%, and 2.0% (w/w). Sediment without BC is referred to as CK; sediments containing 0.5% and 2% RC, FC, and SC are referred to as R0.5, R2, F0.5, F2, S0.5, and S2, respectively. The sediments containing BC were thoroughly mixed and then spiked with the PCP stock solution to achieve a concentration of 50 mg kg^{-1}. PCP-enriched, BC-inclusive sediments were adjusted to the maximum water holding capacity (WHC) of 60% using distilled water and held at 288 ± 1 K for 7 d in the dark to reach equilibrium (Yang et al., 2009; Rhodes et al., 2010; Lou et al., 2011). In order to inhibit the activity of microorganisms, NaN_3 was added to all the BC-inclusive sediment.

2.3. Effect of solid/liquid ratio

After a 7-d incubation, 2 g of contaminated BC-inclusive sediments (dry weight) were weighed in 50-mL glass centrifuge vials. Distilled water (pH 7.0) was added at ratios of 1:1, 1:2.5, 1:5, and 1:10 (solid/liquid, W/V). The vials were sealed with caps and shaken at 200 rpm on a horizontal shaker at 288 ± 1 K for 24 h in the dark. Three replicates were conducted for each design.

2.4. Effect of pH value

Distilled water at different pH values was added to 50-mL glass centrifuge vials containing 2 g incubated BC-inclusive sediments (dry weight) at a solid-liquid ratio of 1:2.5. To simulate the pH value of natural water, pH values ranged from 5.0 to 9.0 and were adjusted by NaOH (0.1 M) or HCl (0.1 M). The equilibrium adsorption experiments were conducted as described in Section 2.3. Each experiment was done in triplicate.

2.5. Effect of temperature

Different temperatures were studied to investigate the change in pollutant concentrations between different seasons. Distilled water (pH 7.0) was added to vials containing contaminated BC-inclusive sediments at a ratio of 1:2.5 (W/V, solid: liquid). The vials were sealed and shaken at 200 rpm on a horizontal shaker at 288 ± 1 K, 298 ± 1 K, and 308 ± 1 K for 24 h in the dark. Three replicates were conducted.

2.6. Effect of salinity

NaCl solutions at concentrations of 0%, 1%, 2.5%, 5%, and 10% were added to vials of BC-inclusive sediment at a solid/liquid ratio of 1:2.5. Samples were shaken as described in Section 2.3. Each experiment was performed in triplicate.

2.7. Effect of DOM

The DOM was extracted from the organic fertilizer (collected at Siling field, Hangzhou, Zhejiang province, China) in the dark for 30 min with distilled water (solid:water = 1:10) at 200 rpm. The suspensions were centrifuged for 30 min at 1500 g and filtered through 0.45 μm polysulfone membrane filters (Kaiser and Guggenberger, 2000). The extracted DOM solution were diluted to 1%, 2.5%, 5%, and 10%, whose TOC value were 208, 526, 1045, and 2079 mg L^{-1}, respectively. The pH value of the DOM solution was adjusted to 7.0 before conducting the experiment. DOM solution was added to vials of BC-inclusive sediment at a solid/liquid ratio of 1:2.5 (W/V). Three replicates were performed.

2.8. PCP analysis

After establishment of equilibrium, water and sediments were separated by centrifugation at 3000 rpm for 20 min, and the supernatant liquid was filtered through a 0.45 μm millipore membrane. PCP concentrations of the filtered liquids were analyzed using high performance liquid chromatography (HPLC) with a diode array UV detector and a C18 reversed-phase column (ODS, 5 μm, 2.1 × 250 mm). The mobile phase used was methanol and 1% acetic acid (90:10, V/V), the flow rate was1.0 mL min^{-1}, and the injection volume was 20 μL. The UV wavelength for the detection of PCP was set at signal wavelength of 220 nm with a bandwidth of 20 nm, and a reference wavelength of 300 nm with 50 nm bandwidth (Chen et al., 2004; Lou et al., 2011). All data were subjected to analysis of variance by using ORIGIN 8.0.

3. Results and discussion

3.1. Effect of solid/liquid ratio

The influence of solid/liquid ratio on the release of PCP in PCP-BC-sediment systems is shown in Fig. 1a. It is obvious that the released concentration of PCP decreased with decreasing solid/liquid ratio, and the addition of BC could reduce the release of PCP from

Table 1
Physical and chemical properties of sediments.

Humic acid	Kerogen (%)	BC	Moisture content (%)	Cation exchange capacity (cmol kg^{-1})	Organic matter (g kg^{-1})	pH	Sand	Silt (%)	Clay
0.57 ± 0.02	1.72 ± 0.01	0.37 ± 0.04	55.01 ± 0.61	10.94 ± 0.32	9.64 ± 0.08	6.76	7.12 ± 0.20	13.84 ± 0.12	79.04 ± 0.85