



Research article

Effects of human management on black carbon sorption/desorption during a water transfer project: Recognizing impacts and identifying mitigation possibilities



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ABSTRACT

Water resources management is an important public concern. In this study, we examined the extent of sorption/desorption of trace pollutants to soil black carbon (BC) in the water level fluctuation zone (WLFZ) of the middle route of the South to North Water Transfer Project in China. In addition, we investigated the main management measures affecting these processes during the project. The results showed that the pseudo second-order model adequately describes the sorption/desorption of phenanthrene on the soil BC in the WLFZ. Water level fluctuation may indirectly influenced BC sorption/desorption by altering water chemistry. Water level residence time had negative effects on BC sorption in short-term experiments (days to months), but the impact gradually diminished with increased residence time. The results suggested that long-term field monitoring of water chemistry is urgent. During the initial period of water transfer, delaying the water supplies as drinking water source or directly irrigating crops could mitigate the adverse impacts. Future research should focus on the water-soluble products of BC degradation. The findings of this study should be useful in improving sustainable management of water resources for water transfer projects.

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1. Introduction

Sustainable management of natural water resources is a worldwide problem. Many water transfer projects have been initiated to mitigate the crisis of water resource shortage. This human water management creates an artificial annual water level fluctuation zone (WLFZ) within water transfer projects. These WLFZs have received considerable attention with a focus on reducing contamination in aquatic environments (Anbumozhi et al., 2005; Mitsch et al., 2008). Soil sorption/desorption is a key process in determining the fate of pollutants and quantifying the risk of contamination (Jonker and Koelmans, 2002).

Carbonaceous geosorbents, such as black carbon (BC) are ubiquitous in the environment and frequently occur in soils and sediments. They play important roles in sorption/desorption of

hydrophobic organic contaminants (HOCs), such as polycyclic aromatic hydrocarbons (PAHs) (Cornelissen et al., 2005). During the past decades, many studies have identified the importance of BC to PAHs sorption/desorption in soils and sediments and shown high and nonlinear sorption of PAHs to BC. These observations have been summarized in several review studies (Ahmad et al., 2014; Cornelissen et al., 2005; Koelmans et al., 2006). To our knowledge, most research in the literature has focused on the isotherms of sorption/desorption and relatively few studies describe the velocity using kinetic models. This topic is important as the velocity of sorption/desorption affects the mobility of contaminants (Martínez-Hernández et al., 2016). In addition, most studies have used synthetic/pyrolytic BC. However, research has shown that soil BC differs significantly from synthetic/pyrolytic BC (Cheng et al., 2008). Even though some studies have used soil BC (Chai et al., 2011; Yang et al., 2008), our previous study indicated that soil BC in the WLFZ is different from natural soil BC (Hao, et al., 2017). Since BC physicochemical properties influence its sorption properties (Cornelissen et al., 2005), the sorption/desorption of soil BC in the

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WLFZ may have different characteristics to those found in past studies. Furthermore, most sorption measurements were carried out at relatively high pollutant concentrations, which does not reflect the actual circumstances within water transfer projects, therefore hampering the assessment of ecological risk and management of water resources.

The fluctuation and residence time of water level are controlled artificially for water transfer projects. Survey results have found that long-term water level fluctuations can alter characteristics of the water chemistry, such as pH and ionic strength (Dinka et al., 2004; Tulipani et al., 2014). Furthermore, BC sorption/desorption is influenced by these factors (Luo et al., 2011; Wu and Sun, 2010). Similarly, Nguyen and Lehmann (2009) emphasized that water regime may play an important role in BC degradation, while BC degradation may further impact its adsorption (Cheng and Lehmann, 2009). Theoretically, these measures will directly or indirectly influence BC sorption/desorption. It is not clear whether these influences will be positive or negative as, to date, these impacts have not been studied and remain unclear.

In this study, we selected the middle route of the South to North Water Transfer Project (SNWDP) in China. This project is typical of water transfer projects worldwide (Li et al., 2013). In addition, the SNWDP is also a source of drinking water. Given sustainable management of water resources, the process determining PAHs transport behavior must be understood in detail (Schwarzenbach et al., 2006). However, very little is known about the extent of trace pollutants sorption/desorption in soils of the WLFZ of the SNWDP as well as the influences of human management on these processes. The purposes of this study were (1) to determine the processes of sorption/desorption of PAHs to soil BC in the WLFZ; (2) to investigate the impacts of human management measures on these processes; and (3) to propose feasible countermeasures for sustainable development. This study will provide a better understanding of the transport behaviors of PAHs in the aquatic system and help improve water resource management during water transfer projects.

2. Materials and methods

2.1. Study area and samples

The study site is located between 32°36′–33°48′N and 110°59′–111°49′E and is the water source area of the Middle Route of the SNWDP in China. The project transfers water to North China, especially to Beijing and Tianjin, to mitigate water resource shortages. Its ecological and socioeconomic importance has received recent attention. Water levels have fluctuated regularly between statutory high and low levels throughout 60 years in the researched area. An artificial WLFZ with a maximum upright fall of 30 m is formed annually. A detailed description of the study area can be found in our previous publication (Hao et al., 2017).

Different soil samples (0–15 cm) were collected in July 2013 and 2014 from upstream and downstream along the WLFZ. Soil samples were air-dried, gently crumbled, and sieved through a 2-mm sieve. All soil physicochemical properties are listed in Table S-1 in the supporting information. Soil BC was extracted from soil samples by using the wet chemical oxidation method. For a detailed description, please refer to our previous paper (Hao et al., 2017). The main sequential steps were as follows: (a) Removal of carbonates and silicates; (b) Removal of organic carbon; (c) Analysis of BC. The extracted BC from different soil samples was labelled with BC1–BC9, respectively. In our previous

studies, the part of soil BC (from 2013) was used to study the physicochemical properties of BC and the isotherms of sorption/desorption. In present study, the left was used for kinetic experiments. Water samples were collected in July 2014 at a depth of approximately 10 cm using previously acid-washed high-density polyethylene (HDPE) bottles. The containers were rinsed thrice with sample water on site. Water samples were filtered (0.45- μm Millipore nitrocellulose filter) in the field, acidified with ultrapure HNO_3 to $\text{pH} < 2$, and stored at 4 °C until further processing.

2.2. Experimental design

2.2.1. Sorption/desorption experiments of kinetics, pH and ionic strength

Phenanthrene was selected as a representative of PAHs to determine sorption/desorption. All sorption/desorption experiments were conducted using batch systems. Batch experiments were carried out by BC suspension with different amounts of phenanthrene solutions in 0.01 mol L⁻¹ NaCl and 200 mg L⁻¹ NaN₃ background electrolyte solution. All experiments were conducted in triplicate. Relative deviations between the replicates were below 10%. A blank sample was run routinely along with the determination in order to remove possible interferences. The sorption/desorption experiment is described in detail in our previous paper (Hao et al., 2017); in this paper, only new procedures are described.

Preliminary experiments were conducted to determine the saturation concentration needed for each BC prior to reach apparent equilibrium. The kinetic experiments were conducted using the optimum phenanthrene level and the natural pH. Samples were taken at .02, .03, .08, .13, .17, .25, .33, .50, .75, 1, 2, 4, 8, 12, 24, 48, and 60 h and separated by centrifugation. The pH ranges were adjusted from 2.0 to 9.0 using 0.1 M HCl or NaOH. The CaCl₂ concentrations were varied from 0.5 to 50 mM. Phenanthrene concentrations were analyzed by HPLC. Detailed analytical methods are available in Hao et al. (2017).

2.2.2. Residence time in the water level experiments

To simulate submerged conditions, filtered water samples were used as background solution. Dried BC samples were added to clean glass vials containing filtered water and 50 mg NaN₃ L⁻¹ to minimize biological activity. The vials were capped and incubated at room temperature prior to the sorption/desorption experiment. To avoid photolysis, all vials were kept in the dark and externally sealed with photographic negative materials. The samples were set at intervals of 5, 30, 60, 90, and 100 days. After residence time has elapsed, the suspension containing BC was taken from each vial for the sorption/desorption experiments, using the same procedure as described previously.

2.3. Data processing

Four models were used to fit the sorption/desorption data measured in this work. The pseudo first-order kinetic equation is described as follows:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (1)$$

where q_e (mg g⁻¹) and q_t (mg g⁻¹) are the amounts of adsorbed adsorbate at equilibrium and at time t , respectively, and k_1 (min⁻¹) is the rate constant of pseudo first-order adsorption.

The pseudo second-order model equation is expressed as:

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