



Antagonistic effect of humic acid and naphthalene on biochar colloid transport in saturated porous media



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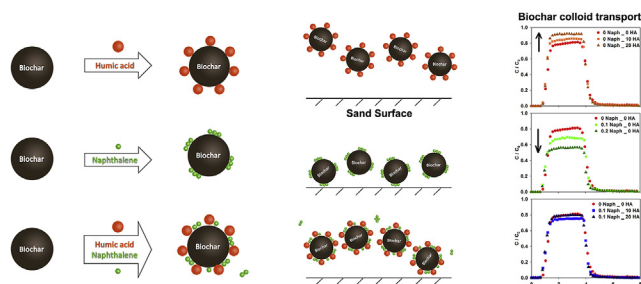
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HIGHLIGHTS

- Humic acid increased biochar colloid transport in saturated porous media.
- Naphthalene decreased the mobility of biochar colloid in porous media.
- Naphthalene and humic acid showed the antagonistic effect on biochar colloid transport.
- Biochar colloid breakthrough curves were well described by the two-site kinetic retention model.

GRAPHICAL ABSTRACT



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ABSTRACT

Biochar is a carbon-enriched material derived from organic material pyrolysis under no/limited oxygen, which is widely used for soil amendment, carbon sequestration, and contaminated soil remediation. This study aims to explore the interplay effect of humic acid (HA) and naphthalene on transport of biochar colloid (BC) in saturated porous media. A series of column experiments were conducted to study BC mobility at different concentrations of HA (0, 10, and 20 mg L⁻¹) and naphthalene (0, 0.1, and 0.2 mg L⁻¹). The results showed that increasing HA concentration promoted BCs mobility in porous media by increasing the electrostatic and steric interaction between BCs and collectors. However, the presence of naphthalene reduced the mobility of BCs with naphthalene increasing from 0 to 0.2 mg L⁻¹, because the nonpolar naphthalene adsorbed onto the biochar surface and shielded the negative charge of BCs. The maximum breakthrough C/C_0 of BCs was increased from 0.7 to 0.8 with increasing HA concentration from 0 to 20 mg L⁻¹ in the presence of 0.1 mg L⁻¹ naphthalene. This meant that HA still played the role to increase the electrostatic repulsion between BCs with HA and collectors when naphthalene was adsorbed on BCs. BCs breakthrough curves were well described by the two-site kinetic retention model including one reversible retention site and another irreversible retention site. The antagonistic effects of naphthalene and HA on BC transport suggested that the mobility of colloidal biochar particles in naphthalene-polluted soil was dependent on the coupled effects of naphthalene and natural organic matter.

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1. Introduction

Biochar is a carbon-enriched material from thermal degradation of organic materials in the absence of oxygen (Lehmann et al., 2011; Lehmann and Joseph, 2015). It is commonly derived from agricultural residue (e.g., grass, crop residue, and woody biomass) or organic waste (e.g., manure, sewage sludge, municipal wastes, industrial organic solid) to improve soil quality, increase carbon sequestration, enhance crop production, and immobilize environmental contaminants (Lehmann and Joseph, 2015; Cernansky, 2015; Qian et al., 2015).

Biochar has strong affinity with environmental contaminants due to its porous carbonated structure, high specific surface area, large micropore, and aromatic functional group. A number of recent studies has shown that biochar strongly adsorbed heavy metals (Uchimiya, 2011; Yang et al., 2014; Qian et al., 2016), as well as other organic compounds (Ahmad et al., 2014; Mohan et al., 2014), especially polycyclic aromatic pollutants (Qiu et al., 2009; Uchimiya, 2011; Qian et al., 2015; Zand and Grathwohl, 2016).

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic contaminants, which contain two or more than two benzene rings (Haritash and Kaushik, 2009; Waqas et al., 2015). At many locations, PAHs exceed the drinking water limit to make them carcinogenic, mutagenic, and toxic (Manoli and Samara, 1999). Because of their high hydrophobicity, PAHs may easily accumulate in water and soils all over the world (Manoli and Samara, 1999; Zhang et al., 2011). Many researchers suggested that the application of biochar into soils remarkably immobilized PAHs leaching in the porous media (Chen and Yuan, 2011; Mohan et al., 2014; Reddy et al., 2014; Zand and Grathwohl, 2016). On the other side, the colloidal and nano-sized range of biochar under two pyrolysis temperatures (350 and 550 °C) showed strong mobility in porous media under saturated water flow conditions (Wang et al., 2013a). Due to strong affinity of biochar to organic compounds, the colloidal and nano-sized fraction of biochar may serve as a carrier for PAHs in porous media, and facilitate PAHs transport with saturated water flows.

Since PAHs can be well adsorbed onto nano and colloidal particles such as dissolved organic matter (DOM), the fate of PAHs is highly related to the leaching behavior of DOM in subsurface (Prechtel et al., 2002; Laegdsmand et al., 2003; Kai et al., 2006, 2007). Natural organic matter is ubiquitous in environment and mainly composed of humic substances (HS), which include three major fractions (1) humic acid (HA); (2) fulvic acid (FA); and (3) humin. A number of studies have reported that HA may enhance the stability and transport of the colloids at various solution pH values in aqueous phase and porous media. This may occur due to macromolecular nature of HA and its attachment to the negatively charged surfaces of colloids, HA may also increase the steric and electrostatic repulsion between colloids and collectors (Hu et al., 2010; Dong and Lo, 2013; Zhang et al., 2013; Cheng and Saiers, 2015).

Naphthalene is one typical organic compound of flat PAHs with two benzene rings (Samanta et al., 2002), which has widely been used in industry. To study the effect of the PAHs on colloidal biochar particles, we used naphthalene as a model for PAHs. HA has strong adsorption capacity towards naphthalene (Xing, 2001), and inhibited the adsorption of naphthalene onto biochar (Zhang et al., 2015). However, according to our knowledge, there is no existing study related to the interplay of HA and naphthalene (hydrophobic organic compound) on BCs transport in saturated porous media. Therefore, the objective of this study is to investigate the effect of HA and naphthalene concentration, both independently and in combination, on the transport and retention of BCs in porous media under water-saturated condition.

2. Materials and methods

2.1. Biochar colloids

Biochar in this study was derived from wheat straw. To produce biochar from agricultural residuals, wheat straw (Zhengzhou, Henan province) was pyrolyzed in a muffle furnace under anaerobic 600 °C conditions for 1 h. The C, H, N, and S contents of the derived biochar were determined by elemental analyzer (Flash 2000, Thermo, USA). The spectra of Fourier transform infrared spectroscopy (FTIR) were recorded between 400 and 4000 cm^{-1} with a Spectrum Spotlight 200 FTIR microscopy system (PerkinElmer, USA). The static contact angles of wheat straw biochar were determined by the sessile drop method using a goniometer (POWEREACH, JC2000D2). The biochar was grounded into biochar colloids (particle size around 200–900 nm) by ball mill to get the appropriate particle size. The morphology of BCs was determined using transmission electron microscopy (JEOL JEM-1230, JPN). The hydrodynamic diameter and electrophoretic mobility of BCs suspensions were determined by a Zetasizer (Nano ZS90, Malvern Instruments Limited).

2.2. Naphthalene, HA, and porous media

Naphthalene (99%, J & K Scientific Ltd, Beijing, China) was used to prepare stock solution for column transport experiments (Caball et al., 2009). HA stock solution (Sigma Aldrich, USA) was sonicated in a water bath for 10 min and was passed through 0.45 μm Nylon Membrane filter (Jinlong, Tianjin, China) before using in the experiment.

Quartz sand (0.425–0.60 mm, Sinopharm, China) was used as model porous medium in the column experiments. To remove organic matters and oxides from sand surface, the sand was soaked in 37% HCl solution for 24 h. After thoroughly washing the sand through DI water, they were dried in an oven at 105 °C for 24 h and then baked at 600 °C for 4 h (Chen et al., 2015). The colloidal particles from ground quartz sand were used as surrogates for the sand surface to obtain the electrophoretic mobility of the sand. The quartz sand was firstly ground into small particles with an agate mortar, and the small particles were suspended in solutions with certain ionic strengths and pH conditions and the suspensions were sonicated for 10 min. After the suspension was settled down for a while, the colloidal quartz particles suspended in the supernatant were measured for the electrophoretic mobility of sand (Wang et al., 2011; Zhang et al., 2010; Zhou et al., 2011).

2.3. Column experiments

Column experiments were conducted in the stainless steel column of 2.5-cm inner diameter and 12.5-cm long. The columns were wet packed with clean sand in an increment of 2 cm thick layers. The average bulk density and porosity of the column was around 1.42 g cm^{-3} and 0.46, respectively. A peristaltic pump was used to provide continuous water supply in an upward direction, and the average pore water velocity was maintained to 26 cm h^{-1} with the flow resident time of 0.48 h in the column. The tracer column experiment was performed by injecting 3 pore volumes (PVs) of 3 mM NaNO_3 solution into the column and then eluted with 5 PVs of DI water to know the hydrodynamics of column experiment. NO_3^- concentrations in the effluents were measured by the spectrophotometer (TU-1900, Persee, China) at 235 nm (Shang et al., 2010). The dispersivity of the column was obtained by fitting NaNO_3 breakthrough curves (BTCs) through convection-dispersion equation (CDE) using Hydrus-1D (Šimunek and Šejna, 1998).

During the experiments, the column was equilibrated by

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