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# Humic acid transport in saturated porous media: Influence of flow velocity and influent concentration

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## ABSTRACT

Understanding the transport of humic acids (HAs) in porous media can provide important and practical evidence needed for accurate prediction of organic/inorganic contaminant transport in different environmental media and interfaces. A series of column transport experiments was conducted to evaluate the transport of HA in different porous media at different flow velocities and influent HA concentrations. Low flow velocity and influent concentration were found to favor the adsorption and deposition of HA onto sand grains packed into columns and to give higher equilibrium distribution coefficients and deposition rate coefficients, which resulted in an increased fraction of HA being retained in columns. Consequently, retardation factors were increased and the transport of HA through the columns was delayed. These results suggest that the transport of HA in porous media is primarily controlled by the attachment of HA to the solid matrix. Accordingly, this attachment should be considered in studies of HA behavior in porous media.

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## Introduction

The presence of humic substances in natural waters can cause various environmental and health problems. Humic acids (HAs) are the dominant type of aqueous organic molecules in most surface water and near-surface groundwater systems, and their behavior can significantly enhance or delay contaminant transport. HAs have a high affinity for aqueous metal cations; therefore, the fate of these metals can be controlled by the behavior of HA (Pandey et al., 2003; Ge et al., 2007; Wang et al., 2009; Zhang et al., 2009; Brigante et al., 2010; Chang Chien et al., 2010; Cao et al., 2010). The presence of HAs has been found to enhance the aqueous solubility of organic pollutants and to facilitate transport of such pollutants through soils (Williams et al., 2000) and other porous and fracture systems (Murphy et al., 1990; Ding and Wu, 1995; Murphy and Zachara, 1995; Haberhauer et al., 2002; Franchi and O' Melia, 2003). Soluble HAs have also been shown to increase the retention of organic pollutants (Conte et al., 2001), which in turn reduces the mobility of these

pollutants. Moreover, through their interaction with actinide ions such as the uranyl ion (Cherwinski et al., 1994), HAs may exert a crucial influence on the mobility of uranium (Artinger et al., 1998, 2002). However, whether HA enhances or retards the transport of pollutants depends on the solid matrix of the media and the solution properties. Therefore, HA mediated migration is important for the risk assessment of contaminant environmental behavior, and the accurate modeling of contaminant transport relies on the ability to describe the transport properties of HAs.

HAs comprise a subclass of humic substances that generally display macromolecular and colloidal characteristics. HAs contain both hydrophobic and hydrophilic moieties as well as many functional groups such as carboxylic, phenolic, carbonyl and hydroxyl groups connected to aliphatic or aromatic carbons (Hering and Morel, 1988). The existence of such carboxylic and phenolic groups results in HA being predominantly negatively charged in aqueous solutions (Cornel et al., 1986). These unique properties affect the transport of HA in different environmental media. Accordingly,

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understanding the fate of HA in the environment, especially its transport in porous media, can provide important and practical evidence for accurate prediction of organic/inorganic contaminant transport in different environmental media and interfaces. Among the different environmental media and interfaces, chemical transport in porous media is very important and has long been the focus of studies conducted by hydrologists and soil scientists due to its crucial applications in civil and agricultural engineering.

Most studies of the environmental behavior of HA that have been conducted to date have focused on the adsorption of HA onto solid surfaces under different conditions (Vermeer et al., 1998; Avena and Koopal, 1999; Specht et al., 2000; Pehlivan and Arslan, 2006; Moura et al., 2007; Wang et al., 2010). However, the transport of HA in a water-filled porous solid system and its governing mechanisms are still not well understood. The factors controlling the transport of HA in porous media may be classified into two types: (1) mechanical factors such as molecular shape, flow rate and HA concentration; and (2) physicochemical factors, such as ionic strength, pH and porous media surfaces. These factors are interrelated since molecular shape is primarily dependent on the solution pH and ionic strength. Weng et al. (2002) evaluated the transport of HA in a copper contaminated acid sandy soil and found that its breakthrough curves were characterized by a rapid, relatively sharp front followed by a plateau at a lower HA concentration than in the influent solution. Wei and Shao (2007) found that the transport of HA through soil columns varied greatly with soil types and the transport was mainly influenced by clay content and cation exchange capacity, which enhanced the adsorption of humic acid, and thus, retarded the transport in soils. Wei et al. (2010) studied the HA transport through different porous media at different solution pH and ionic strength conditions. They found that decreasing pH and increasing ionic strength increased adsorption, and therefore, delayed the transport of HA in porous media. However, few studies have been conducted to evaluate the effects of mechanical factors on transport of HA in porous media, which are essential for understanding HA behaviors in porous media and for controlling pollution related with HA transport. Therefore, this study was conducted to investigate the transport of HA in different porous media, and to discuss the effects of flow rate and influent HA concentration on the transport behavior. Our objective is to understand how HA transport responds to mechanical factors of different porous media.

## 1. Materials and methods

HA was prepared from a commercial humic substance (China Medicine Group, Shanghai Chemical Reagent Corporation, China) and then purified by mixing with deionized water, after which

the pH was adjusted to 10 with 0.1 mol/L NaOH. After centrifugation, the supernatant was removed and precipitated by acidification to pH 1.5 with concentrated HCl. Next, HA was dissolved and precipitated three times, followed by dialysis to reduce the salt content. The external water was replaced in the first 5 hr and then every 12 hr until no chloride was detected. Finally, the product was collected by vacuum filtration and oven dried at 70 to 80°C for 50 hr (Wei et al., 2010).

Glass columns with a 1.9-cm inner diameter and 11.0-cm length were uniformly packed with quartz sand (grain diameters ranging from 0.5 to 1.0 mm) and river sands (grain diameters ranging from 0.5 to 1.0 mm or <0.5 mm). The average grain densities were 2.43, 2.54 and 2.52 g/cm<sup>3</sup> for quartz sand, 0.5–1.0 mm river sand and <0.5 mm river sand. The experimental design and conditions are shown in Table 1. A previous study showed that the surface tension of HA is larger at pH 8 than at lower pH values (Yates and von Wandruszka, 1999), and the fluorescence intensity is larger at pH 8 than at lower or higher pH values (Chen and Kenny, 2007). These results suggest that low pH enhances the aggregation of HA in solution. Thus, a solution pH of 8 was chosen to conduct the experiments. Prior to use, the sand was thoroughly cleaned by washing with 0.1 mol/L Na<sub>2</sub>S<sub>2</sub>O<sub>4</sub> for 2 hr to remove surface metallic compounds and then with 5% H<sub>2</sub>O<sub>2</sub> solutions for 3 hr to remove organic impurities, followed by washing with 12 mol/L HCl overnight and then with deionized water for 5 hr (Wei et al., 2010).

The experimental setup used for the transport experiments is illustrated by Wei et al. (2010). Standard gravimetric methods were used to determine the column packing density. All column experiments were performed in duplicate. Prior to injection of the HA solution, a HA-free KCl solution that had been adjusted to the desired ionic strength and pH was introduced to the top of the column. At least 50 pore volumes of this solution were then applied to thoroughly flush the column and stabilize the pH of the system. The pore volumes were calculated as the ratio of the volume of solution leached through the column in any given time interval and the total volume of solution present inside the column at the beginning of the transport experiment (Shukla, 2013). The flow velocity of the solution was controlled by adjusting the inflow water head. After equilibrium of the column, the KCl solution was terminated, and the HA solution was injected into the column at a constant rate. At the column outlet, the effluent suspension was collected at regular time intervals and analyzed for HA concentration by light

**Table 1 – Experimental design.**

Factor	Solid matrix	pH	Ionic strength (mol/L KCl)	Flow velocity (cm <sup>3</sup> /min)	Influent concentration (g/kg)
Flow velocity	Quartz sand	8	0.001	1.12–5.68	5
	0.5–1.0 mm river sand	8	0.001	0.86–6.43	5
	<0.5 mm river sand	8	0.001	1.33–7.36	5
Influent concentration	Quartz sand	8	0.001	4.73	10
		8	0.001	4.73	2
	0.5–1.0 mm river sand	8	0.001	2.26	10
		8	0.001	2.26	2
	<0.5 mm river sand	8	0.001	3.16	10
		8	0.001	3.16	2

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