



Differences in transport behavior of natural soil colloids of contrasting sizes from nanometer to micron and the environmental implications

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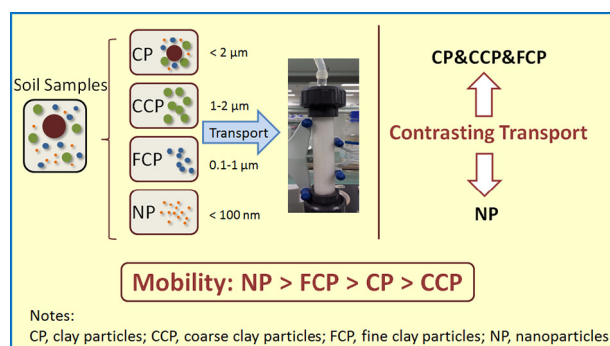
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HIGHLIGHTS

- Transport of soil nanoparticles and clay fractions exhibited different behaviors.
- Ripening was observed for Inceptisol nanoparticles while blocking for Oxisol nanoparticles in BTCs at pH 5 and 30 mM NaCl.
- Secondary energy minimum and physical straining lead to the retention of clay fractions.
- Primary energy minimum and straining induced by simultaneous aggregation cause the retention of nanoparticles.
- Soil nanoparticles have higher mobility than that of bulk clay fractions.

GRAPHICAL ABSTRACT



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ABSTRACT

Transport behaviors of nanoparticles (<math>< 100 \text{ nm}</math>) and clay fractions (clay particles, <math>< 2 \mu\text{m}</math>; coarse clay particles, 1–2 μm and fine clay particles, 0.1–1 μm) extracted from two natural soils (Inceptisol from Jilin and Oxisol from Hainan, China) were investigated in saturated sand columns at 1–30 mM NaCl and pH 5–9. Increasing NaCl concentrations decreased the mobility, while increasing pH increased the mobility of soil particles of various sizes. At pH 5 and 30 mM NaCl, nanoparticles and clay fractions exhibited the different transport behaviors, and ripening was observed for Inceptisol nanoparticles while blocking for Oxisol nanoparticles in breakthrough curves (BTCs). The effluent mass recoveries (MRs) of nanoparticles were much more than that of clay particles for both two soils (>1.9-fold) at all tested conditions, except for Inceptisol at pH 5 and 30 mM NaCl (with comparable MR). According to Derjaguin-Landau-Verwey-Overbeek (DLVO) calculations and particle-collector size ratios, both secondary energy minimum and physical straining led to the retention of clay fractions at pH 5 and 30 mM NaCl, whereas primary energy minimum and straining induced by simultaneous aggregation caused the retention of nanoparticles. The experimental attachment efficiency between soil particles of various sizes and sand collector for both two soils was in the order nanoparticles <math>< \text{fine clay particles}</math> <math>< \text{clay particles}</math> <math>< \text{coarse clay particles}</math>, indicating that soil nanoparticles had greater mobility than clay fractions. Consequently, nanoparticles are suggested to have greater risks to transport contaminants to surface and groundwater, once they are released from soils.

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1. Introduction

Mobile clay colloids ($<2\ \mu\text{m}$), one of the most active (both physically and chemically) soil components, are ubiquitous in natural subsurface environments. They have high specific surface areas ($10\text{--}800\ \text{m}^2\ \text{g}^{-1}$) and a stronger affinity for a variety of nutrients and contaminants (often serving as carriers) than bulk soils (Kretzschmar et al., 1999; Qafoku, 2010). These soil colloidal fractions, to a great extent, could determine the fate and transport of pathogens, nutrients, heavy metals and organic contaminants, and contribute to environmental pollution by spreading them to adjacent water bodies or groundwater (McCarthy and Zachara, 1989; Grolimund and Borkovec, 2005; Sen and Khilar, 2006; Cai et al., 2013a, 2013b). A comprehensive understanding of the colloidal transport behaviors is thus critical for predicting their environmental fate and risk.

Over the past decades, the transport of colloids in porous media has been extensively investigated at both the laboratory and field scale (Molnar et al., 2015). However, much of previous knowledge about colloid transport is derived from experiments with typical engineered nanoparticles (e.g., carbon nanotube, silica, TiO_2 , graphene oxide) (Jaisi et al., 2008; Wang et al., 2012; Chen et al., 2012; Liu et al., 2013), mineral colloids (e.g., montmorillonite and ferrihydrite) (Tian et al., 2015; Liao et al., 2017), and more recently cotransport of different types of colloids (Walshe et al., 2010; Cai et al., 2013a, 2013b; Wang et al., 2015a; Yang et al., 2016; Afrooz et al., 2016). These previous investigations have advanced our understanding of the mechanisms about transport/cotransport of one/two colloids, but the results cannot be readily extrapolated to natural soil colloids that possess complicated physicochemical properties (McCarthy and McKay, 2004; Qafoku, 2010). A limited number of studies indicated that soil colloids are highly mobile in soils (Grolimund et al., 1998), and can be attached to sand surfaces but re-entrained into bulk solution in response to fluctuations in solution chemistry (Zhou et al., 2011). However, these studies have merely focused on the transport of the entire soil colloidal fraction, and made no attempt to separate fractions of various sizes such as clay ($<2\ \mu\text{m}$), coarse clay ($1\text{--}2\ \mu\text{m}$), fine clay ($0.1\text{--}1\ \mu\text{m}$) and particularly nanoparticles ($<100\ \text{nm}$).

With the widespread use of engineered nanomaterials imposing risks to living organisms, the research focus has shifted to nanoparticles derived from the natural environment (Hochella et al., 2008; Theng and Yuan, 2008; Pan and Xing, 2012; Zeng et al., 2014; Bakshi et al., 2015). Our previous studies showed that the nanoparticles extracted from soils remain dispersed and stable in solution after 100 days (Li et al., 2012; Zhu et al., 2014) and are highly mobile in quartz sand columns, and enhance the water solubility and mobility of hydrophobic organic contaminants such as phenanthrene (Li et al., 2013). Soil nanoparticles may interact with the porous media via retention mechanisms different from those with bulk clay colloids. In addition, differences in physicochemical properties such as specific surface area and organic matter content between nanoparticles and bulk clay colloids would lead to their contrasting transport behaviors as well as colloid-associated transport of nutrients and contaminants.

The transport of colloids is influenced by a variety of factors including size, shape, and surface characteristics (roughness and chemical heterogeneity) of colloids and media, colloid concentration, and solution chemistry like ionic strength (IS) and pH (Gao et al., 2011). Although the effects of these factors on transport of engineered nanoparticles are well-known (Wang et al., 2016), experiments on natural soil colloids remain scarce. These factors can affect colloid attachment/retention and hence mobility in soil matrix.

There are various ways to assess colloid attachment. Straining is a mechanism of attachment that occurs with colloids in pore throat too small to pass (Bradford et al., 2002, 2007) and also at the wedged pore spaces such as grain-to-grain contacts (Li et al., 2006). Straining is thought to be significant when particle-collector diameter ratio is greater than the theoretical limit (1.7×10^{-3}) (Bradford et al., 2002,

2003). This ratio is often calculated to identify the role of straining. The classic Derjaguin-Landau-Verwey-Overbeek (DLVO) theory is also employed to explain colloid attachment mechanisms (Ryan and Elimelech, 1996). Under environmentally relevant conditions, porous media (e.g., quartz sand) and soil colloids both often possess net negative charges on the surface, leading to unfavorable attachment conditions (Wan and Tokunaga, 2002). A repulsive energy barrier (Φ_{max}) could exist (in DLVO interaction energy curves) between colloids and porous media. Colloidal particles can attach onto the surfaces of porous media in the secondary minimum ($\Phi_{\text{min}2}$) or overcome the energy barrier to attach in the primary minimum, or both if they are under the unfavorable conditions (Johnson et al., 2007; Shen et al., 2007, 2008). Solution IS and pH are often selected to examine their effects on the transport and retention of colloids due to their key role in mediating these processes.

It was hypothesized that the transport of natural soil nanoparticles would behave different with their bulk clay fractions in response to various environmental conditions. Thus the main objectives of this study were: (i) to examine the effects of IS and pH on the transport; (ii) to elucidate particle retention mechanisms by DLVO and experimental attachment efficiency calculations. In addition, their environmental implications for facilitated transport of contaminants were highlighted.

2. Materials and methods

2.1. Extraction and preparation of soil clay fractions and nanoparticles

Two surface soil samples (0–20 cm) were collected from an Inceptisol (from Jilin, China, $44.5^\circ\ \text{N}$, $126.1^\circ\ \text{E}$) and an Oxisol (from Hainan, China, $19.5^\circ\ \text{N}$, $109.7^\circ\ \text{E}$) (USDA, 2006). The selected soil properties were measured using the standard methods (Sparks et al., 1996) and provided in Table S1 (Supplementary materials). Combined with wet sieving, sedimentation, siphoning and centrifugation methods, a series of procedures were conducted to extract three soil clay fractions i.e., clay ($<2\ \mu\text{m}$), coarse clay ($1\text{--}2\ \mu\text{m}$) and fine clay ($0.1\text{--}1\ \mu\text{m}$) (Tang et al., 2009). Hereafter, we define the above three clay-sized particles together as “clay fractions”. Details of extraction and preparation of three soil clay fractions are given in Supplementary materials. Ultrasonic dispersion and centrifugation methods were used to extract soil nanoparticles below $100\ \text{nm}$ (Li et al., 2012). Briefly, 3 g soil was dispersed in a 100-mL glass-cooling cell containing 80 mL water, and then sonicated by an Ultrasonic Processor VCX750 (Sonics, Newtown, CT, USA) at 60 kJ ultrasonic energy to provide optimum dispersion conditions, whilst keeping the temperature below $20^\circ\ \text{C}$. After sonication, the suspensions were transferred into 50-mL tubes and centrifuged at 3500g for 24 min three times. The supernatants were collected and stored at $4^\circ\ \text{C}$ until use or freeze-dried for further analysis.

2.2. Characterization of soil particles and quartz sand

Concentrations of soil particles were determined by a standard gravimetric method. The stock suspensions were diluted as required before use. The hydrodynamic diameter of nanoparticles and the zeta potentials of all fractions were analyzed by a Zetasizer (Nano ZS90, Malvern, U.K.) based on a dynamic light scattering (DLS) technique and the Helmholtz-Smoluchowski equation, while the size distributions of the clay fractions were determined using a Laser Particle Analyzer (Coulter LS230, Beckman Coulter Inc., USA). Pulverized quartz-sand particles ($<2\ \mu\text{m}$) were suspended in solutions with corresponding IS and pH conditions and sonicated for 1 h. Thereafter, aliquots (5 mL) of the supernatant were used for zeta potential measurement in place of quartz collector surface (Zhou et al., 2011). The pH was measured with a pH meter (S975 SevenExcellence, METTLER TOLEDO, Switzerland). The specific surface area of each freeze-dried fraction was determined by a N_2 sorption isotherm with a multi-point BET method using a Surface Area and Porosimetry Analyzer (Tristar 3020,

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