



Distinguishable Transport Behavior of Zinc Oxide Nanoparticles in Silica Sand and Soil Columns



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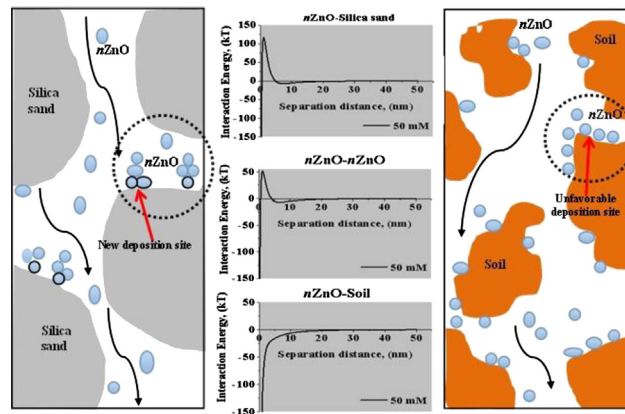
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HIGHLIGHTS

- *n*ZnO were mobile in both silica sand and soil columns even at IS 50 mM.
- *n*ZnO outflow decreased with C_0 in silica sand, while it increased with C_0 in soil.
- DLVO theory explained the difference in *n*ZnO transport between silica sand and soil.
- The two-site kinetic attachment model provided a good description for *n*ZnO transport.

GRAPHICAL ABSTRACT



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ABSTRACT

As part of ongoing risk assessments of ZnO nanoparticles (*n*ZnO) in the natural environment, transport behaviors of *n*ZnO in soil need investigation. This work comparatively studied the transport and retention behavior of *n*ZnO in silica sand versus soil, where the effect of input concentration ($C_0 = 34 \sim 430 \text{ mg L}^{-1}$) and ionic strength (IS = 1 ~ 50 mM) were investigated. In silica sand, *n*ZnO were highly mobile, especially at low C_0 and the efflux of *n*ZnO generally decreased with increasing C_0 at all tested IS. Conversely, at low C_0 , *n*ZnO were almost entirely immobile in soil and the efflux of *n*ZnO increased with C_0 at all tested IS. In both media, the retention profiles (RPs) were generally hyper-exponentially shaped suggesting *n*ZnO easily deposited near the column inlet. As indicated by DLVO calculations, previously deposited *n*ZnO on the silica sand surface acted as new deposition sites due to the lower energy barrier (Φ_{\max}) between *n*ZnO-*n*ZnO than between *n*ZnO-silica sand. However, previously deposited *n*ZnO in soil could enhance Φ_{\max} between the soil surface and *n*ZnO which resulted in unfavorable sites for *n*ZnO deposition. The two-site kinetic attachment model provided good descriptions for the breakthrough curves of *n*ZnO. Detachment was more significant than attachment in silica sand ($k_{1d} > k_{1a}$), whereas soil surface attachment of *n*ZnO were strong and irreversible ($k_{1a} > k_{1d}$). The straining interaction parameter (k_{2str}) increased with increasing C_0 in silica sand, but decreased with C_0 in soil. Such differences were mainly responsible for the various transport behaviors of *n*ZnO in silica sand and soil.

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1. Introduction

Zinc oxide nanoparticles (*n*ZnO) are currently one of the most widely used engineered nanoparticles (ENPs) in the world; finding many diverse applications as sunscreens, cosmetics and coatings, and being produced globally in large (between 101 and 1000 t/year) quantities (Piccinno et al., 2012). Increasingly, *n*ZnO are being introduced into the soil, sediment and water environments (Gottschalk et al., 2013), where they pose a potential hazard to environmental and human health because of their perceived toxicity (Weinberg et al., 2011). Toxicology studies have demonstrated that *n*ZnO have a measurable negative impact on marine organisms (Peng et al., 2011), soil bacterial communities (Ge et al., 2011), cause DNA damage to human epidermal cells (Sharma et al., 2009) and ryegrass (Lin and Xing, 2008). Since soil, sediment and water environments serve as the major sinks for ENPs considered as 'nanowaste', it is important that the effects of ENPs on both soil and water processes should be better understood.

For these reasons, *n*ZnO transport, fate and retention behavior in porous media has become an important topic (Ben-Moshe et al., 2010; Kanel and Al-Abed, 2011; Petosa et al., 2012; Jiang et al., 2012, 2013). Ionic strength (IS), ion valence, pH, biofilm, humic acid and polymer coatings all strongly influence *n*ZnO transport. High flat breakthrough plateaus ($C/C_0 = 0.8 \sim 0.6$) were observed at low IS both in NaCl (1–5 mM) and CaCl₂ (0.1–0.5 mM) solution, though ripening was observed at higher IS (10–20 mM NaCl and 1 mM CaCl₂) (Jiang et al., 2012). Elution of pristine *n*ZnO (0.1 mM NaCl) from clean silica sand was higher at pH 3 and 11, and lower or negligible at pH 7 and 9, respectively (Kanel and Al-Abed, 2011). *Escherichia coli* biofilm enhanced *n*ZnO retention in quartz sand under various IS (0.1–5 mM NaCl) and flow rates (4 and 8 m d⁻¹) (Jiang et al., 2013). Compared to bare *n*ZnO, *n*ZnO elution significantly increased in the presence of 60 mg L⁻¹ humic acid (Ben-Moshe et al., 2010). Polymer-coated *n*ZnO were much less likely to aggregate and exhibited significant transport potential even at high IS (100 mM NaNO₃) (Petosa et al., 2012). However, most of these experiments involved only well-defined regular porous media of quartz sand or glass beads. Soil is one of the most complicated porous media, with unique properties which significantly differentiate it from well-defined regular porous media to such an extent; that the transport behaviors of nanoparticles in soils can potentially be very different and even opposite to those observed in sands.

To date, there are very few studies in the literature which have reported the transport of *n*ZnO in natural soils. Zhao et al. (2012) found that almost all *n*ZnO were retained in saturated sandy-loam soil columns even at low IS (0.1 mM NaCl), a result that directly conflicted with those obtained in well-defined porous media. Contradictory results between well-defined porous media and soil have also been observed for other ENPs. Quevedo and Tufenkji (2012) showed that the retention of carboxylated quantum dots (CdTe and CdSe) was very low (24 and 17%, respectively) in quartz sand columns at 100 mM KCl, but increased to 90 and 43%, respectively at a 10 fold lower IS (10 mM), in a loamy sand. Decreasing the flow velocity had little effect on the transport of fullerene nanoparticles (*n*C₆₀) in Ottawa sand, which was essentially pure quartz, but significantly inhibited the transport in Lula soil (Zhang et al., 2012). This observed difference was attributed to the smaller grain size, more irregular and rougher shape and greater heterogeneity of the Lula soil.

Previous studies in porous media have indicated that increasing the IS of the influent solution could decrease transport of colloids and ENPs, while increasing the influent concentration could promote transport (Bradford and Bettahar, 2006; Bradford et al., 2009; Chowdhury et al., 2011; Wang et al., 2012; Kasel et al., 2013). One of the important factors for predicting the fate of particles in the environment is the prevalence for straining. Straining occurs within mobile-water conduits that are too narrow to permit particles to pass, and is related with physico-chemical solution properties and hydrodynamic force (Bradford et al., 2009; Tufenkji and Elimelech, 2005). For collector grains which are rough

and irregular in shape, straining is believed to be particularly important when the ratio of the particle to the median grain diameter exceeds 0.002 (Bradford et al., 2003). Higher concentrations should produce a more rapid filling (blocking) of favorable attachment sites on the collector surface than lower concentrations (Chowdhury et al., 2011). Concentration dependent straining behavior was significant at lower influent concentrations, but due to repulsive colloid (aqueous phase)–colloid (strained) interactions straining was hindered at higher concentrations (Bradford and Bettahar, 2006). However, to date the related effect of influent concentration and solution IS on *n*ZnO transport behavior has not been reported.

Therefore, the overall objective of this study was: (1) to comparatively study the transport and retention behavior of *n*ZnO in pure silica sand versus soil; and (2) to examine the effect of input concentration (C_0) and solution IS on the transport and retention behavior of *n*ZnO. The input concentrations of *n*ZnO ranged from 34 to 430 mg L⁻¹, at three different IS (1, 10 and 50 mM). The transport and retention mechanisms of *n*ZnO in both silica sand and soil were comparatively discussed and breakthrough data were simulated using the two-site kinetic deposition model; where fitted model parameters were used to gain insight into the different mechanisms controlling *n*ZnO transport in silica sand and soil.

2. Materials and methods

2.1. Porous media

Silica sand (Yonghua Chemical Technology Ltd, China) used for all *n*ZnO transport experiments, consisted of 99.8% SiO₂ (quartz) and trace amounts of metal oxides, with a mean diameter of 325 μm. To remove metal impurities, silica sand was cleaned sequentially with concentrated hydrochloric acid (HCl), 10% sodium hydroxide (NaOH) and rinsed repeatedly with deionized distilled water (DDW) until the pH was near 7, before being oven-dried (105 °C). The zeta potential of silica sand was determined on NaNO₃ suspensions of crushed sand which were sonicated for 1 h on a Zetasizer 3000 HSa (Malvern Instruments, UK). The zeta potential of the silica sand was -33.3 ± 1.6 , -31.1 ± 1.5 and -20.5 ± 0.8 mV at an IS of 1, 10 and 50 mM, respectively, indicating that the silica sand surface was negatively charged. Red soil (0–15 cm) was sampled from Quzhou, Zhejiang province of southern China, air dried, extrogenous impurities removed and sieved (<1 mm) prior to use. Soil pH was 4.88, which was measured at a soil to DDW ratio of 1:5 (w/v) by an Orion (Model 250A+) pH meter. Soil organic matter (SOM) was 1.54% using the Walkley-Black procedure (Nelson and Sommers, 1982). Cation-exchange capacity (CEC) of the soil was 2.51 cmol kg⁻¹ using the BaCl₂ method (Rhoades, 1982). Soil sand, silt and clay content measured by the pipette method (Konert and Vandenberghe, 1997) were 75.3%, 16.3% and 8.4%, respectively. The average particle diameter of the soil was 135 μm; calculated as $175 \mu\text{m} \times \text{sand \%} + 20 \mu\text{m} \times \text{silt \%} + 1.5 \mu\text{m} \times \text{clay \%}$ (Ley et al., 1994). The zeta potential of the soil was measured on a 1:5 (w/v) extract in a supporting electrolyte solution (NaNO₃) following ultrasonication for 30 min and filtering through a 0.45 μm filter. The soil zeta potential was -12.4 ± 1.0 , -7.8 ± 0.5 and -4.1 ± 0.2 mV at an IS of 1, 10 and 50 mM, respectively, indicating the surface of soil was also negatively charged.

2.2. *n*ZnO

A concentrated *n*ZnO suspension ($\geq 40\%$ (w/v), without dispersant) was obtained commercially (Hangzhou Wanjing New Material Ltd, China) and used without further surface modification. The surface morphology of pristine *n*ZnO were characterized using SEM analysis. For each experiment, the concentrated *n*ZnO suspension was added to the background electrolyte solution to achieve the desired concentration and IS, followed by stirring and immediate bath sonication for 15 min at room temperature. Under the conditions used in this study, *n*ZnO

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