



## Transport and retention of silver nanoparticles in soil: Effects of input concentration, particle size and surface coating

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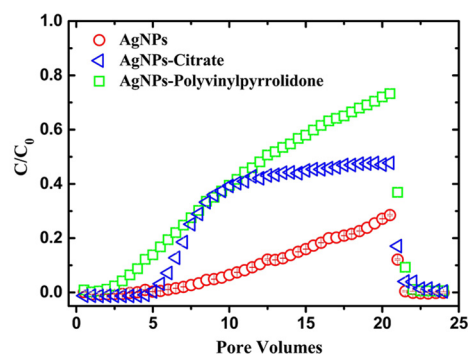
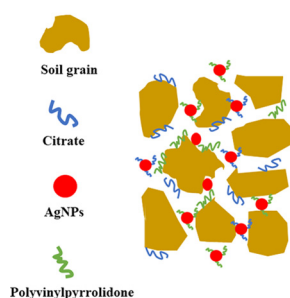
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### HIGHLIGHTS

- Migration of AgNPs in the Ultisol was low due to high surface area and retention sites.
- Increased transport occurred at higher particle concentration and smaller particle size.
- Surface coatings promoted AgNPs transport by effectively blocking the solid-phase sites.
- The 1- or 2-species model successfully described the behaviors of AgNPs in soil columns.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Soils are considered as a major sink for engineered nanoparticles (ENPs) because of their inevitable release to the subsurface environment during production, transportation, use and disposal processes. In this context, the transport and retention of silver nanoparticles (AgNPs) with different input particle concentration, particle size, and surface coating were investigated in clay loam using water-saturated column experiments. Our results showed that the mobility of AgNPs in the soil was considerably low, and >73.9% of total injected AgNPs (except for no coating condition) was retained in columns. This is primarily due to the high specific surface area and favorable retention sites in soil. Increased transport of AgNPs occurred at higher input concentration and smaller particle size. The presence of surface coatings (i.e., polyvinylpyrrolidone (PVP) and citrate) further promoted the transport and reduced the retention of AgNPs in soil, which is likely due to their effective blocking of the solid-phase sites that are originally available for AgNPs retention. Although the shape of retention profiles (RPs) of AgNPs was either hyperexponential or nonmonotonic that is different from the colloid filtration theory prediction, the 1-species (consider both time- and depth-dependent retention) and/or 2-species (account for the release of reversibly deposited AgNPs) model successfully described the transport behaviors of AgNPs in soil columns under all the investigated conditions. This study proves the applicability of mathematical model in predicting the fate and transport of ENPs in real soils, and our findings presented herein are significant to ultimately develop management strategies for reducing the potential risks of groundwater contamination due to ENPs entering the environment.

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## 1. Introduction

After over 20 years of basic and applied research, nanotechnology is undergoing rapid development and industrialization. The overall market value of products incorporating nanotechnology is projected to be about \$3 trillion by the year of 2020 (Roco, 2011). Engineered nanoparticles (ENPs) are increasingly incorporated into many industrial and consumer products. According to the Consumer Products Inventory (CPI), currently there are 1814 consumer products in 32 countries (Vance et al., 2015) in a wide range, such as textile, paint, clothing, sunscreen, cosmetics, antimicrobial agents, medicine, food additives, pesticides, etc. (Council NR, 2012; Kah et al., 2013; Kah and Hofmann, 2014). In particular, silver nanoparticles (AgNPs) are the most frequently used nanomaterial type, accounting for 435 products (or 24%) of the total nanomaterials produced (Vance et al., 2015). With the increasing application of AgNPs environmental seepage are inevitable, which may occur through multiple pathways, such as atmospheric deposition, irrigation with wastewater effluent, land application of sewage sludge, and stormwater runoff (Gardea-Torresdey et al., 2014; Gottschalk et al., 2009; Klaine et al., 2008; Wiesner et al., 2006). The released AgNPs would eventually enter the major repository, that is, soil. In this regard, a thorough understanding of the fate and transport of AgNPs in soils is of critical significance for their benign use, risk assessment, and waste management.

During the last decade, most of the research on the fate and transport of AgNPs were performed using well-defined model porous media (e.g., quartz sand and glass bead) (El Badawy et al., 2013; Liang et al., 2013b; Lin et al., 2011; Mitzel and Tufenkji, 2014; Tian et al., 2010). While natural soils are highly complex and heterogeneous with respect to their physicochemical characteristics, such as varied soil pH, soil organic matter (SOM) and cation exchange capacity, mineral composition and specific surface area, broad grain size distribution, as well as complex pore structure (Wang et al., 2014a, 2014b, 2015). For example, AgNPs dissolution was only detected in 6 soils of 16 soil suspensions with a wide range of physicochemical properties, which was possibly attributed to the strong partitioning of dissolved  $\text{Ag}^+$  ions to soil grains (median  $K_d = 1791 \text{ L kg}^{-1}$ ) (Cornelis et al., 2012). Moreover, the partitioning process became more significant predominantly with increased SOM content. X-ray microtomography analysis suggested that AgNPs transport in natural soil was strongly influenced by soil aggregate size (Sagee et al., 2012). And they found that chemical interactions between AgNPs and soil grains were of pivotal importance as a retention mechanism. Retained AgNPs were strongly associated with soil colloids composed of Si, Fe, Al, and organic matters (Makselon et al., 2018). Therefore, information obtained from those studies in model porous media may have limited applicability for predicting the fate and transport of AgNPs in natural soils (Cornelis et al., 2012, 2013; Darlington et al., 2009; Jaisi and Elimelech, 2009; Sagee et al., 2012; Yopasá Arenas et al., 2018). In addition, most AgNPs incorporated into commercial products are typically modified with capping agents, such as ligands (e.g., citrate) and polymers (e.g., polyvinylpyrrolidone, PVP). These coatings are thought to strongly affect the surface chemistries, subsequently, impact on their mobility, reactivity, and toxicity in natural environments (El Badawy et al., 2011, 2012; Levard et al., 2012).

Laboratory column experiments were conducted to systematically investigate the effects of input particle concentration, particle size, and surface coating on the transport and retention of AgNPs in soil, which was a complement to our previous work (Wang et al., 2014a) examining effects of soil grain size, solution ionic strength and composition, flow rate, and capping agent concentration. Again, attempts were made to better understand the mechanisms behind AgNPs mobility using 1-species and/or 2-species transport model. The overall goal of this study is to deepen our current knowledge of key physicochemical factors controlling the fate and transport of AgNPs in real subsurface environments (i.e., soil and sediment).

## 2. Materials and methods

### 2.1. Chemicals

Deionized water (resistivity:  $18.2 \text{ M}\Omega \text{ cm}$ , Milli-Q, Millipore, USA) was utilized for preparing all aqueous solutions. Analytical reagent-grade  $\text{KNO}_3$ ,  $\text{AgNO}_3$  and citric acid were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Biotech-grade PVP (MW =  $40,000 \text{ g mol}^{-1}$ ) was purchased from Biosharp (Hefei, China).

### 2.2. Soil

Surface soil (0–20 cm) was sampled from the Ecological Experiment Station of Red Soil, Chinese Academy of Sciences (Yujiang County, Jiangxi, China). Prior to use, the soil was air-dried, gently ground, and sieved to obtain median grain size of  $650 \mu\text{m}$  (soil size distribution of 600–710  $\mu\text{m}$ ). The soil was classified as clay loam with 30% sand, 43% silt, and 27% clay. Additional properties were documented in our previous publications (Wang et al., 2014a; Wang et al., 2014b) and also retrieved in Supporting Information (SI) Table S1. Briefly, this soil had a pH of 4.6, organic matter content of  $6.6 \text{ g kg}^{-1}$ , cation exchange capacity of  $8.5 \text{ cmol kg}^{-1}$ , and a specific surface area of  $24 \text{ m}^2 \text{ g}^{-1}$ . The clay fraction consisted of kaolinite, montmorillonite and hydromica. The surface charge of the soil in 1 mM  $\text{KNO}_3$  solution was determined to be  $-15.0 \pm 1.1 \text{ mV}$  using a NanoBrook 90Plus PALS apparatus (Brookhaven Instruments, USA).

### 2.3. AgNPs suspensions

Silver nanoparticles (AgNPs) with two different sizes were used in this study. Commercial AgNPs (denoted as small AgNPs) were purchased from the Nanjing XFANO Materials Tech Co., Ltd. (Nanjing, China). In-house AgNPs (denoted as large AgNPs) were synthesized as per Yu et al. (2014) method with a slight modification, details were described in the Supporting Information Text S1. AgNPs stocks were stored at  $4 \text{ }^\circ\text{C}$  in the dark before use. Aliquots of AgNPs stock solution were transferred into 1 mM  $\text{KNO}_3$  electrolyte solution containing 0.5% PVP or 0.5% citrate. Subsequently, the mixture of AgNPs with 0.5% PVP/citrate coating was vigorously stirred for 1 h, followed by sonication for 30 min prior to the column transport experiments. The pH value of AgNPs solutions was unbuffered and ranged between 4.5 and 5.3.

The scanning spectra of small and large AgNPs were obtained using an UV-Vis spectrophotometer (UV-2700, Shimadzu corporation, Japan) at the wavelength ranging from 200 to 800 nm. Their zeta potentials and hydrodynamic diameters ( $D_h$ ) were examined with NanoBrook 90Plus PALS analyzer (Brookhaven, USA) at room temperature. Morphological images of AgNPs were taken using a transmission electron microscope (TEM, JEM-2100, Japan), in which particle size distribution was calculated using Nanomeasurer 1.2 software based on randomly selected ~100 particles from 10 TEM images.

### 2.4. Column transport experiments

Transport experiments were performed in duplicates using glass chromatography column with 1.2-cm inner diameter and 10-cm in length. Both ends of column were capped with polytetrafluoroethylene and stainless-steel mesh (80- $\mu\text{m}$  opening) to support the packed bed. The vertically-oriented columns were dry-packed by carefully adding 1–2 cm increments of soil grains ( $650 \mu\text{m}$ ) followed by gentle vibration to eliminate any layering (Wang et al., 2012a). The packed height was around 8.5 cm. A well-established procedure of saturating and conditioning the packed column was applied to stabilize the soil colloids and to avoid pore clogging (Wang et al., 2014a, 2014b, 2015). Detailed protocol was stated in SI Text S2. A nonreactive tracer (bromide) experiment was performed to obtain the fluid pore-water velocity and

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