Contents lists available at ScienceDirect

Geoderma

journal homepage: www.elsevier.com/locate/geoderma

Transport of silver nanoparticles in intact columns of calcareous soils: The role of flow conditions and soil texture

Samaneh Rahmatpour^a, Mohammad Reza Mosaddeghi^{a,*}, Mehran Shirvani^a, Jiří Šimůnek^b

^a Department of Soil Science, College of Agriculture, Isfahan University of Technology, Isfahan 84156-83111, Iran
^b Department of Environmental Science, University of California Riverside, Riverside, CA, USA

ARTICLE INFO

Handling Editor: A.B. McBratney Keywords: Ag nanoparticles Breakthrough curve Retention profile HYDRUS-1D One-site kinetic attachment model Flow condition

ABSTRACT

Growing production of manufactured nanomaterials has increased the possibility of contamination of groundwater resources and soils by nanoparticles (NPs). It is crucial to study the fate of NPs in subsurface porous media in order to evaluate and control their risks to ecosystems and human health. Hence, this study was conducted to investigate the transport and retention of polyvinylpyrrolidone (PVP) stabilized silver nanoparticles (AgNPs, a diameter of 40 nm) under saturated and unsaturated conditions in intact columns of two calcareous sandy loam (TR) and loam (ZR) soils. Furthermore, similar experiments were conducted using sand quartz as a reference medium. A pulse of the AgNP suspension with an input concentration (C_0) of 50 mg L⁻¹ was injected into the columns for 3 pore volumes. The transport of bromide (Br), as a non-reactive inert tracer, was also examined. High mobility of AgNPs was observed through the sand columns due to unfavorable conditions for AgNP deposition on the quartz sand surfaces. Nearly all AgNPs introduced into the columns of both soils were retained in the soil. Percentages of AgNPs leached out of the columns were < 1% of the total injected mass in both soils. Hyperexponential retention profiles (RPs) were observed in both soils and maximum concentrations of 100–130 mg kg⁻¹ were determined near the columns' inlet. However, slightly stronger retention of AgNPs and greater maximum retained concentrations on the solid phase (S_{max}) in the ZR soil compared with the TR soil may be attributed to smaller grain sizes of the ZR soil. Hydrodynamic forces adjacent to the solid surfaces near the column inlet can provide a viable explanation for the hyperexponential shape of RPs. The one-site kinetic attachment model in HYDRUS-1D, which accounted for time- and depth-dependent retention, was successfully used to analyze the retention of AgNPs. The results showed that the degree of saturation had little effect on the mobility of AgNPs through undisturbed soil columns. Our results suggested the limited transport of AgNPs in neutral/alkaline calcareous soils under both saturated and unsaturated conditions.

1. Introduction

Silver nanoparticles (AgNPs) are most commonly used for their strong antimicrobial and antiviral properties in medicine, dentistry, cosmetics, jewelry, photography, textile, and food packaging (Cornelis et al., 2013). Discharged nanoparticles (NPs) find their way into the soils during their manufacturing, transportation, application, and disposal (Pachapur et al., 2016), and soils and aquifers act as primary filter systems to remove particulate materials from percolating water and protect water resources (Kasel et al., 2013a; Liang et al., 2013a). Many previous studies have shown the negative effects of AgNPs on human health and the environment (e.g., He et al., 2011; Carbone et al., 2014). Hence, a detailed understanding of the processes governing the transport and retention of AgNPs in soil is required to accurately assess the fate and distribution of AgNPs and to design effective strategies for

reducing their toxicity in the environment.

According to the particle filtration theory, retention and mobility of NPs in porous media are controlled by physicochemical properties of both NPs and collector surfaces, such as the grain size (texture), input concentration, mineralogical composition, the presence of humic acid, the type of coatings or stabilizing agents, the solution chemistry, as well as by hydrodynamic forces (Lin et al., 2011; Liang et al., 2013a,b; Braun et al., 2015). The filtration theory has commonly been employed to describe the deposition of NPs on porous media surfaces. However, studies with NPs and natural colloids often showed deviations in predictions from filtration theory. For example, the retention of NPs may occur even under unfavorable conditions for attachment due to local surface charge heterogeneities (Lin et al., 2011), aggregation (Bradford et al., 2006), and straining in small pores and at locations of high surface roughness (Kasel et al., 2013a). Additionally, an increase in the

* Corresponding author. *E-mail address*: mosaddeghi@cc.iut.ac.ir (M.R. Mosaddeghi).

https://doi.org/10.1016/j.geoderma.2018.02.016





GEODERM/

Received 2 June 2017; Received in revised form 3 February 2018; Accepted 8 February 2018 0016-7061/ © 2018 Elsevier B.V. All rights reserved.

NPs transport due to stabilizing agents causing short-range repulsive forces (El Badawy et al., 2013), retarded and asymmetric BTCs, and uniform, non-monotonic, and hyperexponential retention profiles (RPs) (Liang et al., 2013a) are examples of violations of the filtration theory.

Most researchers have studied the transport of AgNPs in highly idealized systems consisting of repacked, homogeneous, coarse-textured porous media (e.g., glass beads, quartz sands) under water-saturated or unsaturated conditions (e.g., El Badawy et al., 2013; Fang et al., 2013; Liang et al., 2013b). While these studies have provided valuable insights into the mechanisms controlling the transport of AgNPs, they do not exactly resemble natural soil conditions. Soil components such as clay minerals, organic matter, and carbonates would interact with the NPs and subsequently have a dominant effect on their fate, mobility, and potential risks to reach groundwater resources. For example, recent batch studies have shown that the retention of NPs in natural soils is correlated with the clay content (Cornelis et al., 2012). Simple porous media are not able to account for the complexity and heterogeneity (such as a wide particle size distribution, irregular grain shapes, variations in surface chemical heterogeneity, and a complex pore structure) of natural soils (Sagee et al., 2012; Liang et al., 2013a).

Unfortunately, to date, only a few column studies have focused on studying the transport of AgNPs in natural systems, leading to a serious lack of information about their fate and distribution in the environment. In natural soils, clay content and type, soil pH, the presence of organic material, and the ionic strength of the soil solution can affect mobility and bioavailability of AgNPs (Liang et al., 2013a). The information on the fate and behavior of AgNPs in soils will ultimately determine the environmental risk of AgNPs applications. Only recently, have some studies involving the transport of AgNPs in intact soil columns been conducted (Nowack and Bucheli, 2007; Sagee et al., 2012; Cornelis et al., 2013; Liang et al., 2013a; Braun et al., 2015). Neukum et al. (2014) suggested that the transport of AgNPs in sandstones is affected by the pore size distribution, mineralogy, and solution chemistry. Liang et al. (2013a) observed a significant retardation in breakthrough curves (BTCs) and hyperexponential RPs of AgNPs in natural (undisturbed) soils. Sagee et al. (2012) reported high mobility and early breakthrough of AgNPs in a disturbed sandy clay soil. They found that mechanical straining and chemical interactions between AgNPs and the soil surfaces played key roles in retention of AgNPs. Cornelis et al. (2013) suggested that straining was enhanced following fast heteroaggregation between negatively charged AgNPs and positively charged soil colloid sites and that the mobility of polyvinylpyrrolidone (PVP) coated AgNPs was significantly reduced in the natural soils. Despite the frequent existence of unsaturated water flow conditions in natural soils, information is scarcely available on the transport of NPs through unsaturated soils. Colloid retention under unsaturated flow conditions is more complicated due to the presence of air in the soil system and due to water flow being constrained to smaller pores. Colloid retention in unsaturated porous media under electrostatically unfavorable deposition conditions has been attributed to attachment to the solid-water interface (SWI), attachment to the air-water interface (AWI), deposition on solid surfaces, retention at the solid-air-water triple point and, straining in thin water films (Chen and Flury, 2005; Bradford and Torkzaban, 2008; Torkzaban et al., 2008; Fang et al., 2013; Kumahor et al., 2015). Under unsaturated conditions, colloid transport may increase due to moving AWI (McCarthy and McKay, 2004) or decrease due to greater colloid attachment to stationary AWI (Chen et al., 2008). However, most previous studies on colloid transport reported that colloid retention increased in unsaturated porous media. Bradford et al. (2002) reported that the retention of colloids under unsaturated flow conditions was controlled by the spatial distribution of water saturation and may increase due to limited preferential flow and a greater contact time between the media matrix and the colloids. Kumahor et al. (2015) also investigated the transport of citrate-coated AgNPs in unsaturated sand columns. They suggested that a non-equilibrium interaction

occurred at the SWI, while an equilibrium sorption to the AWI led to retardation of AgNPs. They concluded that reversibility of attachment to the AWI was sensitive to the properties of the AWI and AgNPs, and to the solution chemistry. Moreover, Chen and Flury (2005) concluded that colloids were repelled from the AWI. Fang et al. (2013) showed that decreasing water saturation had little effect on the mobility of TiO₂ NPs through packed sand columns owing to net repulsive interactions between negatively charged AWI and TiO₂ NPs.

To the best of our knowledge, no previous study has evaluated the transport of AgNPs in calcareous soils, despite their widespread occurrence in the arid and semi-arid regions around the world. Calcareous soils contain a high amount of calcium carbonate, and Ca²⁺ bridging due to the high activity of Ca²⁺ cations may play an important role in NPs retention. Divalent cations such as Ca²⁺ and Mg²⁺ have been shown to be more effective in NPs retention than monovalent cations like Na⁺ (El Badawy et al., 2010; Liang et al., 2013a). In general, it can be expected that alkaline soils have a high potential to retain NPs compared to acidic soils because the pH value of these soils (7.0 to 8.5) is closer to the pH value at the point of zero charge (pH_{PZC}) of NPs. Also, the high Ca²⁺ concentration in calcareous soils could lead to aggregation of NPs (Hotze et al., 2010). Moreover, only a few studies have evaluated the transport and retention of AgNPs in undisturbed and/or unsaturated soil systems.

The primary objective of this research is thus to evaluate the effects of the degree of soil saturation and soil texture on the transport and retention of AgNPs in intact columns of calcareous soils. To allow for direct comparisons with previous studies, similar AgNP transport experiments were also conducted in sand columns. In addition, AgNP transport and retention data were modeled using HYDRUS-1D to better understand their transport and retention mechanisms. This information is needed to properly assess the risk of exposure of ecosystems to AgNPs and to develop strategies for waste management and remediation.

2. Materials and methods

2.1. Synthesis of AgNPs

All chemicals reagents used in our experiments were of analytical grade and were used as received without further purification. A sonochemical method was applied for preparing AgNPs in aqueous polyvinylpyrrolidone (PVP) solutions (Zhu et al., 2010). In a typical preparation, 0.05 g of PVP was added to 100 mL of aqueous solution containing 0.1 g of AgNO₃. The PVP acts as both reducing and capping agent. The mixture was stirred for complete dissolution and agitated under sonication. Ultrasound irradiation was carried out with a multiwave ultrasonic generator (Sonicator 3000; Bandeline, MS 72, Germany), equipped with a converter/transducer and titanium oscillator (horn), 12.5 mm in diameter, operating at 20 kHz with a maximum power output of 60 W. The operating condition was at 5 s pulse on and 5 s pulse off with amplitude of 72% at 25 °C for 20 min.

The suspension for each experiment was freshly prepared (in order to minimize the dissolution of AgNPs) by dilution of the concentrated stock suspension of stabilized AgNPs into selected electrolyte solutions to achieve an approximate concentration of 50 mg L^{-1} . The stock suspension was sonicated for 30 s with the probe before dilution. During injection into the columns, the suspension was continuously sonicated in a sonication bath.

2.2. Characterization of AgNPs

Transmission electron microscope (TEM) images of the synthesized AgNPs were obtained on a Philips CM30 instrument at an accelerating voltage of 150 kV. An average hydrodynamic diameter and surface charge characteristics of the AgNPs were determined using the Malvern ZEN 3600 Nano ZS Zetasizer (Malvern Instruments Ltd., Malvern, UK). Zeta potential (ζ) was measured with the same equipment between

Download English Version:

https://daneshyari.com/en/article/8894070

Download Persian Version:

https://daneshyari.com/article/8894070

Daneshyari.com