



# Partitioning of Ag and CeO<sub>2</sub> nanoparticles versus Ag and Ce ions in soil suspensions and effect of natural organic matter on CeO<sub>2</sub> nanoparticles stability

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## HIGHLIGHTS

- Dissolved organic matter stabilizes CeO<sub>2</sub> nanoparticles in the aqueous phase.
- Ag<sup>+</sup> and Ce<sup>3+</sup> ions interact more strongly with soil constituents than nanoparticles.
- Correlations between Ag or CeO<sub>2</sub> nanoparticles in soil suspension and soil properties.
- Different interactions between Ag/Ce nanoparticles/ions and suspended/organic matter.

## ARTICLE INFO

### Article history:

Received 26 October 2017

Received in revised form

1 February 2018

Accepted 21 February 2018

Available online 22 February 2018

### Keywords:

Silver

Cerium oxide

Nanoparticles

Environmental fate

Soil

Stability

## ABSTRACT

This study examined the solid-liquid distribution of 14.8-nm Ag and 6.2-nm CeO<sub>2</sub> nanoparticles in soil suspensions and compared it to that of Ag<sup>+</sup> and Ce<sup>3+</sup> ions, to better understand their environmental behaviour and fate. After 24 h incubation, more than 51% or 29% of the spiked amounts of Ag or CeO<sub>2</sub> nanoparticles, respectively, can be retrieved in the liquid phase of (re)suspended soils. The Ag or Ce concentration remaining in solution depends on the incubation time and was influenced by soil properties. Significant correlations are obtained between, on the one hand, the relative amounts of Ag or CeO<sub>2</sub> nanoparticles in suspension and the soil-pH, CEC, texture, suspended matter, nitrogen, phosphorus, TOC and main and trace elements content on the other hand. The presence of dissolved natural organic matter stabilizes CeO<sub>2</sub> nanoparticles in the aqueous phase. In soil suspensions, Ag<sup>+</sup> and Ce<sup>3+</sup> ions seemingly interact more strongly with soil constituents compared to their nanoparticle counterparts, rendering the Ag and CeO<sub>2</sub> nanoparticles to be more stable and potentially bioavailable.

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## 1. Introduction

Due to their small size (<100 nm), engineered nanoparticles (ENPs) often possess unique size-related properties (e.g., high surface-to-volume ratio and surface reactivity) that are not observed in bulk materials (Akaighe et al., 2013). ENPs have received much attention due to their rapidly increasing

applications in industry and consumer products (Handy et al., 2008; Klaine et al., 2008; Voelker et al., 2015). The growing use of ENPs increases the risk of accidental or intentional emission to aquatic or terrestrial environments, such as soils, where they might be transported via pore water into groundwater (Liang et al., 2013a). Hence, there is a public concern about the potential harmful effects of ENPs on the environment and human health (Klaine et al., 2008; Gottschalk et al., 2013; Durenkamp et al., 2016). The extent of such adverse effects depends on the concentration, bioavailability and transformations of ENPs in the environment (Cornelis et al., 2013, 2014). Therefore, it is important to gain

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extensive knowledge on the environmental behaviour and fate of ENPs (Fang et al., 2009; Sagee et al., 2012; Schaumann et al., 2015).

Metallic ENPs, including Ag, Au, Fe, CeO<sub>2</sub>, TiO<sub>2</sub> and ZnO, are the most widely utilized ENPs (Liu et al., 2012; Tourinho et al., 2012; Benoit et al., 2013; Peijnenburg et al., 2016). Ag ENPs have been used in numerous commercial products, such as cosmetics, textiles and medical products, mostly because of their antimicrobial properties (Benoit et al., 2013; Liang et al., 2013b). Ag ENPs have been shown toxic though to a wide range of aquatic and terrestrial organisms (Durenkamp et al., 2016; Tourinho et al., 2012; Levard et al., 2012). The high oxygen storage and UV absorbing capacity of CeO<sub>2</sub> ENPs have led to their use as catalysts in the automotive industry for reducing NO<sub>x</sub> and particulate emissions of diesel engines, in fuel cells, polishing materials and in a variety of pharmaceutical and agricultural products (Cornelis et al., 2011; Li et al., 2011). CeO<sub>2</sub> ENPs have also been demonstrated to induce toxicological effects in bacteria, algae and human cells (van Hoecke et al., 2009; Rogers et al., 2010; Zhang et al., 2011a, 2016).

As a major environmental compartment, the soil is considered a potential sink for ENPs (Cornelis et al., 2014; Peijnenburg et al., 2016). Main exposure pathways of terrestrial environments to ENPs include the application of biosolids from wastewater treatment to agricultural land as soil amendments, leakage from landfills, deposition of airborne particulates, accidental spills or the deliberate use in soil remediation or agrochemicals (Klaine et al., 2008; Limbach et al., 2008; Gottschalk et al., 2009). Consequently, soils are expected to be a source of ENPs to aquatic environments through, e.g. erosion and surface runoff (Whitley et al., 2013). Still, limited information on the behaviour and fate of metallic ENPs in the soil compartment has been generated (Schaumann et al., 2015; Tourinho et al., 2012; Peijnenburg et al., 2016).

The stability of nanoparticles in complex aquatic matrices such as soil solutions, is a primary determinant of their fate, bioavailability and toxicity (Liu et al., 2012; Gimbert et al., 2007; Klitzke et al., 2015). Preferential partitioning of ENPs towards the solid phase reduces their mobility and bio-accessibility, whereas well-dispersed nanoparticles could persist longer in the environment, be transported more easily over longer distances, and potentially be involved in particle-facilitated contaminant transport (Fang et al., 2009; Gimbert et al., 2007). The nature and extent of their partitioning between solid and liquid soil compartments, which is suggested to be highly affected by the solution chemistry and soil properties, is therefore an essential aspect to consider in order to assess the potential environmental risks ENPs may pose (Sagee et al., 2012; Schaumann et al., 2015; Cornelis et al., 2012; Van Koetsem et al., 2017). Soils are comprised of a large collection of reactive surfaces, which could impact the bioavailability and stability of nanoparticles through e.g. deposition (Batley et al., 2013). Additionally, environmental conditions such as pH, ionic strength, dissolved organic carbon and biofilm coating have been demonstrated to affect the behaviour of several ENPs (Klitzke et al., 2015; Ben-Moshe et al., 2013; Han et al., 2016). For instance, at ionic strengths higher than the critical coagulation concentration, ENPs will quickly aggregate and settle out of solution. Natural organic matter on the other hand can bind onto nanoparticles and stabilize them in suspension, thereby increasing their mobility (Liu et al., 2012; Liang et al., 2013b; Whitley et al., 2013; Johnson et al., 2009). However, the majority of studies examining the retention and transport behaviour of ENPs have utilized highly idealized systems of model porous media, such as well-defined glass beads or homogeneous quartz sand (Liang et al., 2013b; Li et al., 2011; Han et al., 2016; Petosa et al., 2010). Although studies employing such simplified systems have provided fundamental mechanistic insights and valuable knowledge on physicochemical parameters affecting nanoparticle transport and deposition, they are not able to

account for the full complexity and heterogeneity of natural soil systems (Liang et al., 2013a; Cornelis et al., 2013; Schaumann et al., 2015). Several authors have investigated the solid-liquid distribution of ENPs using natural soils or sediments (Cornelis et al., 2012; Van Koetsem et al., 2015). Specifically concerning Ag nanoparticles, Klitzke et al. (2015) studied the interactions of pH, dissolved organic carbon and Ag nanoparticles, while Hashimoto et al. (2016) compared the chemical speciation of Ag nanoparticles and ionic Ag under aerobic and anaerobic conditions in soils. However, in regard to CeO<sub>2</sub> nanoparticles, less scientific knowledge is currently available. Fang et al. (2016) investigated the effect of macropores on the transport of CeO<sub>2</sub> nanoparticles in both quartz sand and soil. However, to the best of our knowledge, information about the interactions between CeO<sub>2</sub> nanoparticles and pH, dissolved organic carbon in soils is still lacking. In addition, there is still a paucity of information regarding the factors affecting the partitioning behaviour of ENPs and, to a further extent, their transport behaviour and general environmental fate, especially in natural soil systems. Schaumann et al. (2015) and Peijnenburg et al. (2016) also emphasized this current lack of knowledge in their reviews.

Our study examined the solid-liquid partitioning of Ag and CeO<sub>2</sub> ENPs as well as their ionic counterparts (Ag<sup>+</sup> and Ce<sup>3+</sup>) by subjecting soil suspensions to different settling forces. Suspended particles (including ENPs) and organic compounds are removed from the liquid phase to a different extent by the application of gravitational and centrifugation force. The correlation between ENPs remaining in suspension and the amount of total organic carbon (TOC) and suspended matter (SM) remaining in the liquid phase will be indicative of the association between ENPs and organic or suspended matter. The ENP soil partitioning is explored at selected time points in order to gain insight in the partition kinetics. Additionally, the impact of more soil properties on the ENP soil partition is evaluated to better understand their physicochemical behaviour and fate in aquatic and terrestrial environments.

## 2. Materials and methods

### 2.1. Soil sampling and characterization

The top 0–25 cm layer of three soils was collected in the region of Ghent (Belgium) and used throughout the study. The soils were characterized according to standard methods described elsewhere (Salehi et al., 2011; Jones and Willet, 2006; Gee and Bauder, 1986; van Reeuwijk, 2002; Sartori et al., 2013; van Ranst et al., 1999). More details about soil sampling and characterization are provided in the [Supplementary Information](#).

### 2.2. Nanoparticle and ionic solutions

Citrate-stabilized Ag ENPs of average particle size 14.8 nm (concentration: 100 mg L<sup>-1</sup>, ζ-potential: -50 ± 5 mV, pH: 6–8) and CeO<sub>2</sub> ENPs of average particle size 6.2 nm (concentration: 50 g L<sup>-1</sup>, ζ-potential: +40 ± 5 mV) were obtained as aqueous dispersions from PlasmaChem (Berlin, Germany). More details about the ENP dispersions and characterization are provided in the [Supplementary Information](#). Ionic Ag as AgNO<sub>3</sub> (Plasma HIQU, 10 g L<sup>-1</sup> in 2–5% HNO<sub>3</sub>) and Ce as Ce(NO<sub>3</sub>)<sub>3</sub> (Plasma HIQU, 10 g L<sup>-1</sup> in 2–5% HNO<sub>3</sub>) reference standard solutions were acquired from Chem-Lab NV (Zedelgem, Belgium) and used as the ionic spikes of Ag<sup>+</sup> and Ce<sup>3+</sup> in the batch experiments.

### 2.3. Batch experiments

Ultrapure water (18.2 MΩ cm<sup>-1</sup>) (Milli-Q®, Millipore, Billerica,

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