



Retention and remobilization mechanisms of environmentally aged silver nanoparticles in an artificial riverbank filtration system

Laura Degenkolb^{a,b,*}, George Metreveli^c, Allan Philippe^c, Anja Brandt^d, Kerstin Leopold^d, Lisa Zehlike^{a,b}, Hans-Jörg Vogel^e, Gabriele E. Schaumann^c, Thomas Baumann^f, Martin Kaupenjohann^a, Friederike Lang^g, Samuel Kumahor^e, Sondra Klitzke^{a,b,g}

^a Berlin University of Technology, Institute of Ecology, Department of Soil Science, Ernst-Reuter Platz 1, 10587 Berlin, Germany

^b German Environment Agency, Section Drinking Water Treatment and Resource Protection, Schichauweg 58, 12307 Berlin, Germany

^c University of Koblenz-Landau, Institute for Environmental Sciences, Group of Environmental and Soil Chemistry, Fortstraße 7, 76829 Landau, Germany

^d Ulm University, Institute of Analytical and Bioanalytical Chemistry, Albert-Einstein-Allee 11, 89081 Ulm, Germany

^e Helmholtz Centre for Environmental Research Leipzig-Halle, Department of Soil Physics, Theodor-Lieser-Strasse 4, 06120 Halle, Germany

^f Technical University of Munich, Institute of Hydrochemistry, Marchioninstr. 17, 81377 München, Germany

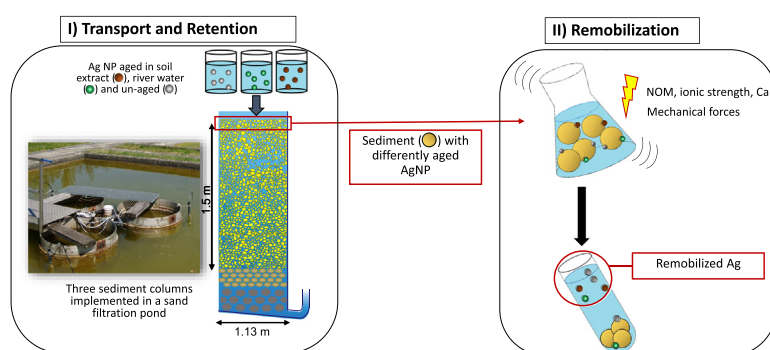
^g Albert-Ludwigs-Universität Freiburg, Chair of Soil Ecology, Alte Universität, Bertoldstraße 17, 79098 Freiburg i. Br., Germany



HIGHLIGHTS

- Outdoor, large-scale column experiments and laboratory batch studies
- Strong retention of Ag NP, but highest mobility of soil-aged Ag NP
- 50% of retained Ag is remobilizable by mechanical forces and hydrochemical changes.
- NOM and ionic strength reduction enhances NP mobility, Ca reduces it.
- Co-mobilization with natural colloids is an important remobilization mechanism.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 18 May 2018

Received in revised form 6 July 2018

Accepted 6 July 2018

Available online xxxx

Editor: D. Barcelo

Keywords:

Heteroaggregation

Nanoparticle transformation

Breakthrough

ABSTRACT

Riverbank filtration systems are important structures that ensure the cleaning of infiltrating surface water for drinking water production. In our study, we investigated the potential risk for a breakthrough of environmentally aged silver nanoparticles (Ag NP) through these systems. Additionally, we identified factors leading to the remobilization of Ag NP accumulated in surficial sediment layers in order to gain insights into remobilization mechanisms.

We conducted column experiments with Ag NP in an outdoor pilot plant consisting of water-saturated sediment columns mimicking a riverbank filtration system. The NP had previously been aged in river water, soil extract, and ultrapure water, respectively. We investigated the depth-dependent breakthrough and retention of NP. In subsequent batch experiments, we studied the processes responsible for a remobilization of Ag NP retained in the upper 10 cm of the sediments, induced by ionic strength reduction, natural organic matter (NOM), and mechanical forces. We determined the amount of remobilized Ag by ICP-MS and differentiated between particulate

* Corresponding author at: German Environment Agency, Section Drinking Water Treatment and Resource Protection, Schichauweg 58, 12307 Berlin, Germany.

E-mail addresses: laura.degenkolb@uba.de (L. Degenkolb), metreveli@uni-landau.de (G. Metreveli), philippe@uni-landau.de (A. Philippe), anja.brandt@uni-ulm.de (A. Brandt), kerstin.leopold@uni-ulm.de (K. Leopold), lisa.zehlike@uba.de (L. Zehlike), hans-joerg.vogel@ufz.de (H.-J. Vogel), schaumann@uni-landau.de (G.E. Schaumann), tbaumann@tum.de (T. Baumann), martin.kaupenjohann@tu-berlin.de (M. Kaupenjohann), fritzi.lang@bodenkunde.uni-freiburg.de (F. Lang), samuel.kumahor@ufz.de (S. Kumahor), sondra.klitzke@uba.de (S. Klitzke).

and ionic Ag after remobilization using GFAAS. The presence of Ag-containing heteroaggregates was investigated by combining filtration with single-particle ICP-MS.

Single and erratic Ag breakthrough events were mainly found in 30 cm depth and Ag NP were accumulated in the upper 20 cm of the columns. Soil-aged Ag NP showed the lowest retention of only 54%. Remobilization was induced by the reduction of ionic strength and the presence of NOM in combination with mechanical forces. The presence of calcium in the aging- as well as the remobilizing media reduced the remobilization potential. Silver NP were mainly remobilized as heteroaggregates with natural colloids, while dissolution played a minor role.

Our study indicates that the breakthrough potential of Ag NP in riverbank filtration systems is generally low, but the aging in soil increases their mobility. Remobilization processes are associated to co-mobilization with natural colloids.

© 2018 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

Riverbank filtration systems are natural systems that may ensure the cleaning of surface water for recharging groundwater. Due to their natural cleaning capacity, i.e. through mechanical filtering, sorption, and biodegradation processes, these systems have become increasingly important for drinking water production (Sprenger et al., 2017). The effectiveness of riverbank filtration in removing microorganisms such as pathogens (i.e. bacteria and viruses) from infiltrating surface water has long been recognized in Germany (Schmidt et al., 2003). Nowadays, there is a much larger number of potential pollutants in anthropogenically influenced water bodies which need to be removed before drinking water usage (Schmidt et al., 2003). One example is nanoparticles (NP) which are included in many industrial applications and consumer products ("Danish Consumer Council. The Nanodatabase." 2018). Amongst synthetic inorganic NP, silver nanoparticles (Ag NP) were the most frequently used nanomaterials in consumer products until 2015 (Vance et al., 2015).

After being released into rivers and lakes (Cleveland et al., 2012), Ag NP can undergo various transformation processes. In the following manuscript, the processes leading to the transformation of NP in the environment without complete loss of the original NP phase are termed "aging processes" (Schaumann et al., 2015). Surface oxidation, coating with natural organic matter (NOM), and cation-induced bridging between NP are aging phenomena which significantly influence the surface properties of NP (Darlington et al., 2009; Nel et al., 2006; Schaumann et al., 2015). Aging in different environmental media therefore has a strong impact on the fate of NP and their colloidal and chemical stability in natural surface water and porous systems. However, the fate of these environmentally modified NP is still underrepresented in the literature (Selck et al., 2016).

Recent studies conducted in complex aqueous systems suggest that unaged Ag NP mainly accumulate in upper sediment layers due to (hetero)aggregation processes and sorption to the solid phase (Coleman et al., 2013; Furtado et al., 2015). Additionally, an accumulation of NP in biofilms is often observed (Ferry et al., 2009; Nevius et al., 2012). Studies investigating the fate of Ag NP in soils have shown that horizontal and vertical translocation is very limited (Schlich et al., 2017) and a remobilization of Ag NP from the solid phase is negligible (Hoppe et al., 2015). Although there are experiments investigating the transport of NP in laboratory sediment columns (such as Zhang and Zhang, 2014 for Ag NP, Lv et al., 2016 for TiO₂, Fang et al., 2016 for CeO₂) and aquifers (Adrian et al., 2018), main retention mechanisms of riverbank filtration systems (such as filtration by biofilms or retention on clay minerals) are not taken into account in most of them. Studies that consider more environmentally relevant conditions suggest that the transport potential of engineered NP in the real environment will be much lower than predicted by laboratory experiments (Emerson et al., 2014). This highlights the need for more complex and realistic outdoor experiments. Therefore, the aim of our study was to investigate the breakthrough and retention of aged Ag NP in riverbank filtration systems and to understand the mechanisms that lead to their remobilization by answering the following questions:

- 1) *Does the aging of Ag NP in soils or rivers before reaching riverbank filtration systems affect their breakthrough and retention in natural sediments?* Recent studies about NP stability clearly show that a coating with NOM can enhance the colloidal stability of NP (Philippe and Schaumann, 2014). In our study, we focused on aging processes in rivers and soils, leading to the coating of NP with NOM and cation-induced aggregation, two important processes affecting NP stability and fate. Additionally, the dominating process in natural surface waters is heteroaggregation due to the high concentration of natural colloids in environmental systems compared to NP concentrations (Lowry et al., 2012; Praetorius et al., 2014). This might also lead to the co-transport of NP with natural colloids in porous systems (Makselon et al., 2017; Neukum et al., 2014). To account for this process, we used natural, colloid-containing pond water as mobile phase for the transport experiment.
- 2) *Which mechanisms can induce the remobilization of Ag NP retained in surficial sediment layers?* Long-term mesocosm studies show that the majority of NP in aquatic ecosystems is accumulated in upper soil and sediment layers (i.e. Lowry et al., 2012). Therefore, we hypothesized that a large part of Ag NP would be accumulated in the surficial sediment layers of riverbank filtration systems. In natural systems, these upper sediments are exposed to varying shear forces caused by turbulence in rivers and to changing hydrochemical conditions. While heavy rain falling could dilute river water and reduce ionic strength, flooding might transport a large amount of NOM from floodplain soils into rivers. Both ionic-strength reduction and enhanced NOM concentrations in combination with shear forces have been shown to cause disaggregation of Ag NP homoaggregates (Metreveli et al., 2015). Based on these findings, we aimed at identifying conditions that may cause a remobilization of Ag NP immobilized in the surface sediments of an artificial riverbank filtration system. To detect the main remobilization mechanisms, the properties of remobilized Ag were characterized. This included the differentiation between dissolved and particulate Ag as well the analysis of the aggregation state of Ag NP. Both characteristic are of high importance for a prediction of the fate of remobilized Ag and potential consequences of Ag remobilization.

Our study was undertaken in two parts:

1. The breakthrough and retention of Ag NP, aged in river water, soil extract, and ultrapure water respectively, were studied in a near-natural, water-saturated porous system. The experiments were conducted in an outdoor artificial riverbank filtration system. Compared to laboratory studies as used in the literature (i.e. El Badawy et al., 2013; Liang et al., 2013; Sargee et al., 2012) our system had larger dimensions and was influenced by atmospheric changes. The biological activity was driven by the surrounding pond water conditions.
2. In laboratory batch experiments, we systematically studied the effect of ionic strength, NOM, divalent cations, and mechanical forces on the extent of Ag remobilization. To gain a more mechanistic process

Download English Version:

<https://daneshyari.com/en/article/8858463>

Download Persian Version:

<https://daneshyari.com/article/8858463>

[Daneshyari.com](https://daneshyari.com)