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Mobility of silver nanoparticles in porous media: The impact of biological interfaces, water saturation, and nanoparticle aging

Frederic Leuther[1], Laura Degenkolb[2,3], John Maximilian Köhne[1], George Metreveli [4], Sondra Klitzke[3], and Hans-Jörg Vogel[1]

[1]Helmholtz Centre for Environmental Research Leipzig-Halle, Department of Soil System Science, Theodor-Lieser-Straße 4, 06120 Halle, Germany

[2]Berlin University of Technology, Institute of Ecology, Department of Soil Science, Ernst-Reuter Platz 1, 10587 Berlin, Germany

[3]German Environment Agency, Section Drinking Water Treatment and Recource Protection, Schichauweg 58, 12307 Berlin, Germany

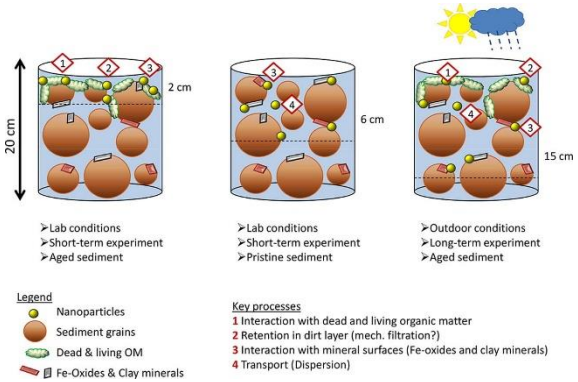
[4]University of Koblenz-Landau, Institute for Environmental Sciences, Group of Environmental and Soil Chemistry, Fortstraße 7, 76829 Landau, Germany

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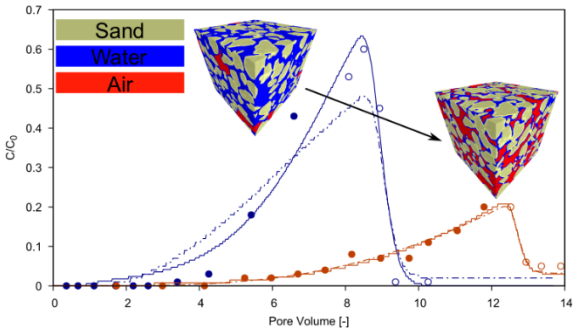
Overview

This presentation bases on two different studies:

1. **The fate of silver nanoparticles in riverbank filtration systems — The role of biological components and flow velocity (Degenkolb and Leuther et al, 2019, Science of The Total Environment, 134387)**



2. Transport and retention of sulfidized silver nanoparticles in porous media: The role of air-water interfaces, flow velocity, and natural organic matter (Leuther et al, WRR, under review)

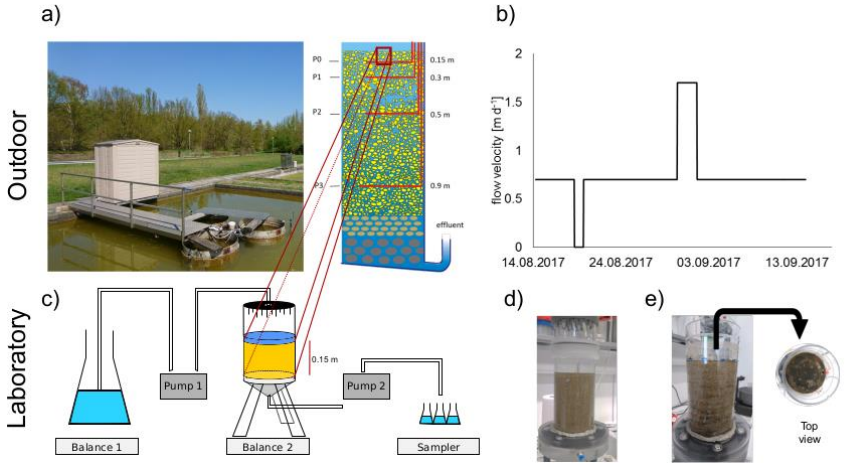


1. Impact of biological interfaces (substrate aging)

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We investigated Ag-NP (c = 0.6 mg L^{-1}) transport at different spatial and temporal scales in pristine and previously pond water-aged sediment columns:

- Transport of Ag-NP under near-natural conditions was studied in a long-term riverbank filtration experiment over the course of one month with changing flow scenarios (i.e. transport at 0.7 m d^{-1}, stagnation, and remobilization at 1.7 m d^{-1})
- To elucidate retention processes, we conducted small-scale lab column experiments at low (0.2 m d^{-1}) and high (0.7 m d^{-1}) flow rate using pristine and aged sediments.
- The riverbank filtration water contained i.a. Ca^{+}, K^{+}, Mg^{+}, Cl^{-}, SO_4^{2-} ions, TOC: ~5 mg L^{-1}, pH ~ 8

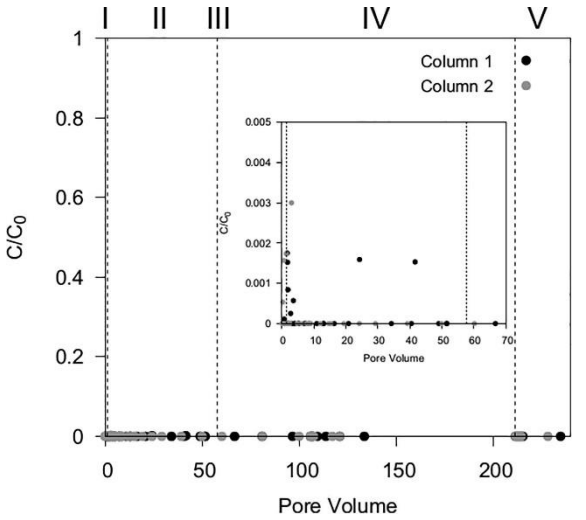


a) the near-natural riverbank filtration system, embedded in a slow sand filtration pond, b) the flow schedule of the outdoor experiment, c) the laboratory setup for the column experiments with pristine (d) and pond-aged sediment (e).

1. Impact of biological interfaces (substrate aging)

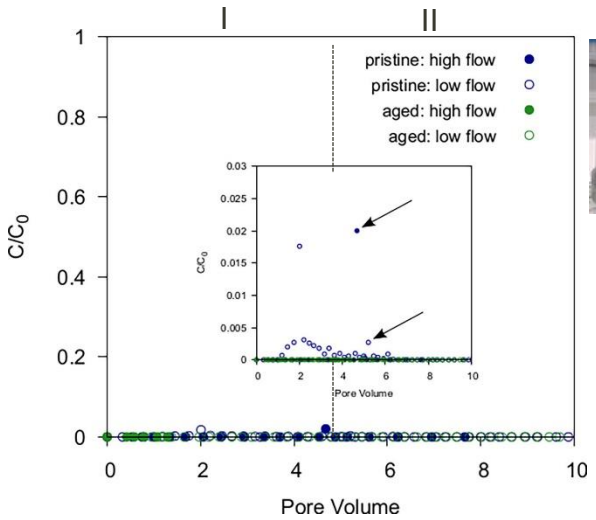
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- **Outdoor:** Breakthrough at 15 cm depth





- **Laboratory:** Breakthrough at 15 cm depth





The roman numerals refer to the flow schedule of the outdoor experiment:

I) 5 PV of NP application (j_{w1}=0.7 m d^{-1}), II) leaching (j_{w1}=0.7 m d^{-1}) and III) stagnation,

IV) leaching (j_{w1}=0.7 m d^{-1}), V) leaching (j_{w2}=1.7 m d^{-1})

I) 5 PV of NP application (low flow j_{w1}=0.2 m d^{-1} and high flow j_{w2}=0.7 m d^{-1}),

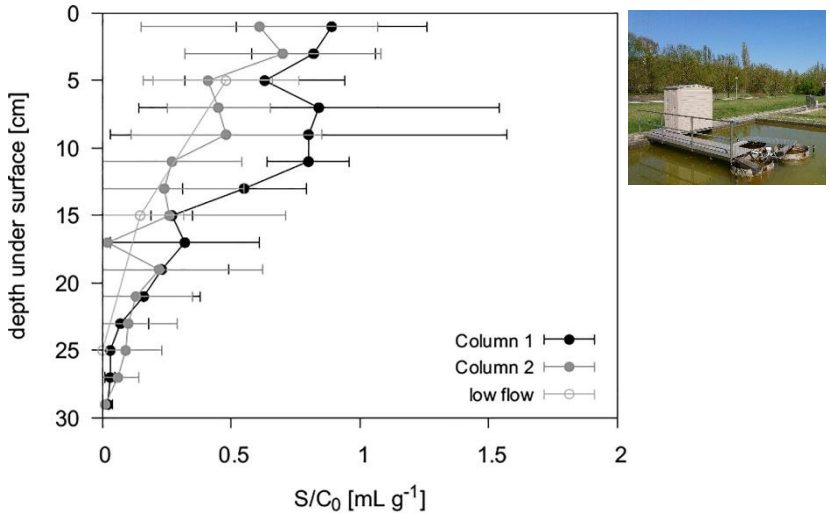
II) leaching (j_{w1}=0.7 m d^{-1} j_{w2}=1.7 m d^{-1})

Arrows = change from I to II. Indication of particle remobilization.

1. Impact of biological interfaces (substrate aging)

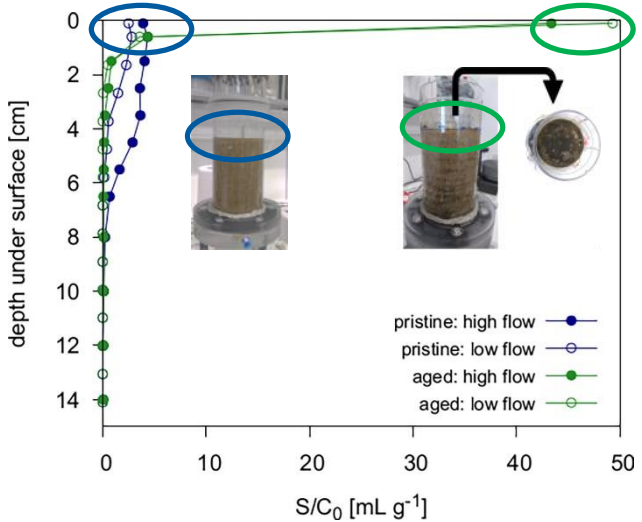
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- **Outdoor:** retention profiles 0 – 30 cm depth



The error bars margin the 95% confidence interval by two times standard error of 3–4 replicates. Column 1 and 2 show the results of the described experiment. For comparison, values gained by Degenkolb et al. (2018) are included as low flow (0.2 m d^{-1}) scenario.

- **Laboratory:** 0 – 15 cm depth



Ag NP retention profiles in pristine (blue) and aged (green) sediments under high flow (filled circles, 0.7 m d^{-1}) and low flow (open circles, 0.2 m d^{-1}) rate

1. Impact of biological interfaces (substrate aging)

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In the tested systems (outdoor and laboratory):

- Ag-NP accumulated in the upper centimeters of the sediment both in lab and outdoor experiments.
- In the lab study, retention of Ag-NP by attachment to biological components was very effective under high and low flow rate with nearly complete NP accumulation in the upper 2 mm. When organic material was absent, abiotic filtration mechanisms led to NP retention in the upper 5 to 7 cm of the column.
- In the long-term riverbank-filtration study, Ag-NPs were transported up to a depth of 25 cm.
- For the pristine sediment in the lab study and the outdoor experiments only erratic particle breakthrough was detected in a depth of 15 cm.

➢ physico-chemical interactions of Ag-NP with sediment surfaces are efficient in retaining Ag-NP under typical flow condition.

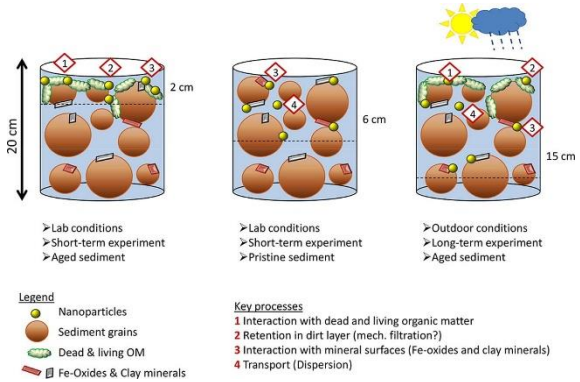
➢ organic material provides additional retention sites which increase the filtration capacity of the system.

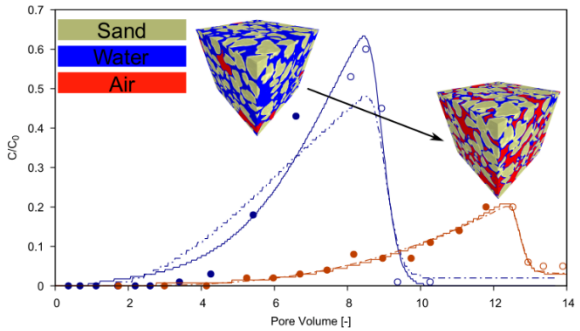
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2. **Transport and retention of sulfidized silver nanoparticles in porous media: The role of air-water interfaces, flow velocity, and natural organic matter (Leuther et al, WRR, under review)**





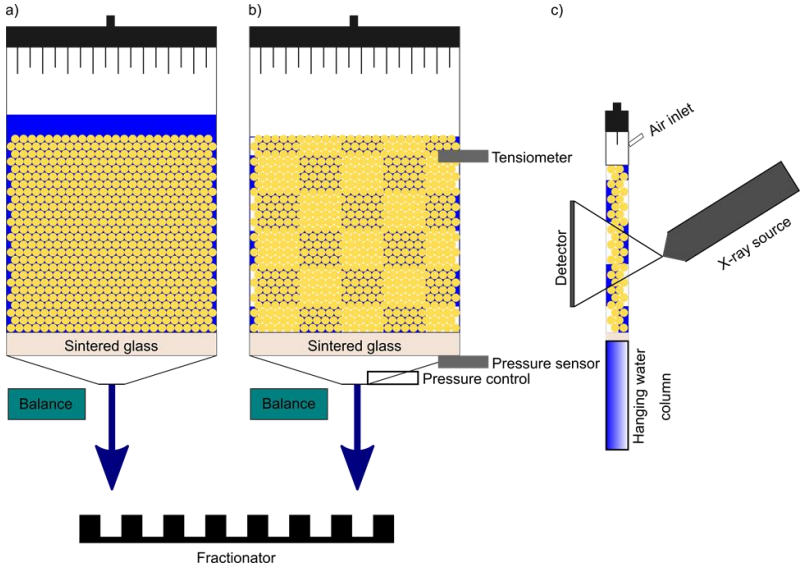
2. NOM-S-Ag transport: Impact of velocity and air-water-interfaces

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We investigated the mobility of sulfidized Ag-NPs (c = 1.0 mg L^{-1}) aged with natural organic matter (NOM 5 mg L^{-1}) in sand at different fluxes and water contents:

- Saturated transport experiments (a) at high and intermediate flux (inflow = unflow)
- Unsaturated experiments (b) with unit gradient in tension and water content along the profile. Established under high, intermediate, and low flux application
- X-ray microtomography: Determination of the distribution and quantity of air-water and solid-water interfaces under flow conditions

➢ **Research under review, data not shown**



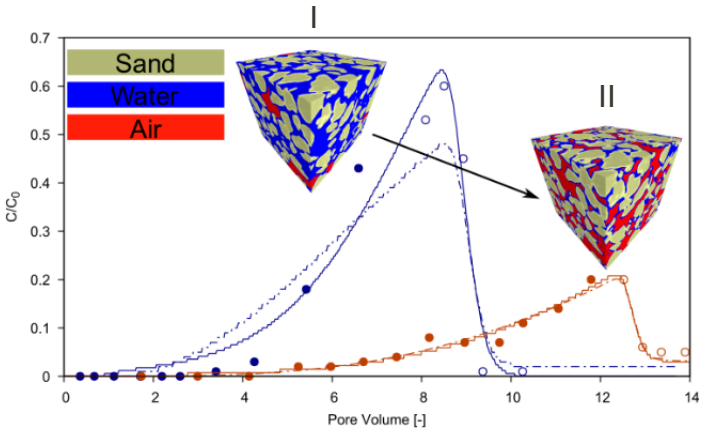
Set-up for a) saturated column experiments, b) unsaturated column experiments, and c) the flow tube installed inside the X-ray µCT. The different patterns indicate for different phase distributions of solid (yellow), water (blue), and air (white) during the experiments.

2. NOM-S-Ag transport: Impact of velocity and air-water-interfaces

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The transformation of Ag-NPs to NOM-S-Ag-NPs has an impact on particle mobility in porous media. There was a strong link between transport behaviour of nanoparticles and the structure of the transport media including the different interfaces:

- The retardation of particles was dependent on the transport velocity and the interfacial areas between the solid-water and air-water phase.
- The total amount of particle breakthrough was decreasing with decreasing water content and flux. The reduction was mainly dependent on the overproportioned increase of AWI, the decreasing transport velocity had a minor impact.



I) Saturated experiment at high flux (blue dots) and II) unsaturated experiment at low flux (red dots). Reconstructed, filtered, and segmented µCT sub-volume showing the phase distribution of sand, water, and air during water flow.

3. Acknowledgements and sources

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- Degenkolb, L., Leuther, F., Lüderwald, S., Philippe, A., Metreveli, G., Amininejad, S., Vogel, H.-J., Kaupenjohann, M. & Klitzke, S. (2019) The fate of silver nanoparticles in riverbank filtration systems—The role of biological components and flow velocity. *Science of The Total Environment*, 699, 134387. https://doi.org/10.1016/j.scitotenv.2019.134387