Geophysical Research Abstracts
Vol. 21, EGU2019-2181, 2019
EGU General Assembly 2019

Transport of sulfidized silver nanoparticles in unsaturated zone: The role of air-water interfaces, straining and NOM coating

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The transport and fate of silver nanoparticles (NPs) has been investigated mainly in saturated systems whereas the behavior in the unsaturated zone is still not fully understood. Column experiments at the scale of soil horizons revealed the importance of different types of retention mechanisms at the soil surface and along the flow path: grain size and grain surface roughness, water saturation, solution chemistry, geochemistry of porous substrate, stability and aggregation of NPs and the flow velocity. Yet, the detailed mechanisms and the relative importance of air-water and water-solid interfaces remained unclear. Furthermore, in transport experiments with silver NPs mostly original, pristine NPs are used. However, silver NPs, which are already sulfidized in sewer systems and wastewater treatment plants, can reach the natural compartments mostly in partly or completely sulfidized state. The application of sulfidized silver NPs representing more environmentally relevant form of silver is often missing in environmental studies.

In this study, we analyzed the transport and fate of sulfidized silver NPs in saturated and unsaturated conditions, under different flow velocities, and different ambient concentrations of dissolved organic matter. Larger column experiments were combined with small scale experiments to reveal the role of different water contents, water phase distributions, and flow velocities on NP transport. Transport experiments were conducted in larger sand columns with decreasing fluxes and water contents to determine NP breakthroughs, NP retention profiles, and hydraulic parameters. Flow experiments on a smaller scale were done inside an X-ray computed tomograph under the same conditions to determine phase distributions and geometry of solid, air, and water phases. In this way the total extent of water-solid and water-air interfaces was quantified and related to particle retention and breakthrough-characteristics of the larger column experiments.