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Reactive transport simulations to identify key parameter for silver nanoparticle migration in soils

Dan Zhou (1), Joanna Makselon (2), Erwin Klumpp (2), Diederik Jacques (3), and Irina Engelhardt (1)

(1) Department of Hydrogeology, TU Berlin, Berlin, Germany, (2) Agrosphere (IBG-3), Institute of Bio- and Geosciences, Forschungszentrum Jülich GmbH, Germany, (3) Institute for Environment, Health and Safety (EHS), Belgian Nuclear Research Centre (SCK•CEN), Belgium

Silver nanoparticles (AgNP) are one of the most widely used engineered nanoparticles and can reach the subsurface via discharge from industry and waste water treatment plants. Current research indicates that AgNP have a higher mobility with increasing input concentration, flow velocity, grain size, decreasing ionic strength. Therefore, it can be speculated that hydrological changes e.g. rain intensity, evaporation, intense dry periods will directly influence AgNP mobility. However, subsequent investigations about the impact of hydrology on AgNP mobility are not published. In this study, AgNP transport was investigated to account for the influence of (i) interrupted flow and thus abrupt changes in flow velocity, (ii) variable water saturation, and (iii) variable ionic strength.

Laboratory column experiments were conducted with loamy sand and under variable saturated conditions. AgNP were injected at the top of the column. Then the column was continuously irrigated with KNO_3 solution. The irrigation period was followed by a drying period over 2.7 days (flow interruption). After the dying period a second irrigation with KNO_3 solution was conducted. During this second irrigation KNO_3 solution was varied in ionic strength.

Current numerical modeling studies describe nanoparticles transport with the colloid filtration theory (CFT). However, the CFT is not able to describe the influence of transient hydrochemistry (e.g. ionic strength) as the attachment and detachment coefficient is independent of the hydrochemistry. The presented numerical investigation use an extended approach that couple the CFT with a colloid release model (CRM) and the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory.

Measured and simulated breakthrough curves (BTC) showed a distinctive decrease of AgNP when the irrigation was stopped and a reduced mobility and high retention of AgNP in the soil. This effect is mainly attributed to a decrease in flow velocity, increased evaporation, and decreased water content. Interrupted flow and evaporation led to an increased electrical conductivity and ionic strength which in turn can compress the electrical double layer of AgN and thus enhance their mobility. However, our results display that changes in ionic strength influenced AgNP mobility only slightly. The numerical investigations showed that the key process affecting AgNP migration under variable saturated conditions is kinetic attachment at interfaces of the porous medium, the soil-water interface (SWI) and air-water-interface (AWI). Calculated values of the attachment coefficient at SWI were distinctively larger than the attachment coefficient at AWI. The simulation of retention profiles show that the AgNP concentration at SWI was less than at the AWI. This effect results from a reversible attachment at the SWI and irreversible attachment at AWI. After flow was restarted, flow velocity and water content raised again resulting in an increase of attachment at the SWI. During the previous drying period AgNP were irreversible attached at AWI and only a limited amount of AgNP was released when the flow was restarted due to the remaining air phase within the soil.