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Partitioning of selenium (Se) to natural colloidal phases and transport through steep redox gradients in a mesoscale laboratory lysimeter experiment

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The potential far-field transport of radionuclides from a deep geological repository for high-level radioactive waste into the biosphere and the accumulation in e.g. the food chain must be considered. Especially, the transport of long-lived fission products as redox-sensitive ^{79}Se(VI) is of great concern due to its high mobility in contrast to reduced forms as e.g. selenides. Mesoscale laboratory lysimeter with four different natural agricultural topsoil substrates were set up in a long-term experiment to investigate the upward transport of selenate and actinide tracers in Grimsel-Test site groundwater present below the exemption limit at ≤ ppq levels (Quinto et al., 2017) through the unsaturated zone. In the scenario based approach, the packed lysimeters were placed in a climate chamber at 10.5°C and were continuously fed for over 760 days with groundwater and maintained a constant water table in the evaporation driven systems. A redox interface formed after about 200 days at the capillary fringe and was observed by in-situ Eh probes and with sub-mm resolution using chemical optical sensor foils. Se as non-radioactive $Se^{(VI)}O_4^{2-}$ tracer was added after 500 days during an injection experiment in association with conservative tracers (Br, Cl). Soil cores were sampled at the end of the experiment for determining in situ K_d-values. Eh differences of 500-700 mV in a 7 cm soil section at the capillary fringe were detected. Pore waters below the interface in oxygen reduced environments showed higher concentrations of mobile organic matter (OM) and redox sensitive elements such as Fe and Mn. Conservative tracers showed recoveries of 80-90%. However, Se was retarded almost by factor 1000 under reducing conditions as shown by high solid phase concentrations in the lower soil core sections. Ultrafiltration experiments with lysimeter pore waters revealed a share of Se < 1 kDa of 56±18% likely associated to organic matter and in contrast to formed larger ternary metal-OM-$Se^{(IV)}$ colloidal phase complexes. Ultracentrifugation experiments supported this dataset by showing 36±12% Se (related to 1 kDa permeates) in supernatants in co-occurrence of OM. Thus, colloidal Se phases might contribute to the long-term upwards transport.

Quinto *et al.* (2017), *Anal Chem* **89**, 7182-89