Geophysical Research Abstracts
Vol. 15, EGU2013-11476, 2013
EGU General Assembly 2013

A new methodology involving stable isotope tracer to compare short- and long- term selenium mobility in soils

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Selenium is an element of environmental concern given its dual beneficial and toxic character to animal and human health. Its radioactive isotope 79Se, a fission product of 235U, is considered critical in safety assessment of nuclear waste repositories in case of leakage and hypothetical soil contamination. Therefore, Se species transformations and interactions with soil components have to be clearly understood to predict its dispersion in the biosphere (e.g., accumulation in soils, migration to waters, transfer to living organisms). While natural Se interactions with soils run over centuries to millennia time scales, transformations and partitioning are generally studied with short-term experiments (often inferior to 1 month) after Se addition. The influence of slower, long-term processes involved in Se speciation and mobility in soils is thus not properly accounted for. We tested if using ambient Se would be relevant for long-term risk assessment while added Se would be more representative of short-term contamination impact.

For that purpose, we developed a new methodology to trace the differential reactivity of ambient and spiked Se at trace level (μg kg-1) in soils. It combined the use of a stable isotopically enriched tracer with our previous published analytical method based on specific extractions and HPLC-ICP-MS to determine trace Se species partition in different soil phases.

Given that soil extracts contains very high concentrations of various elements interfering Se (e.g., Fe, Cl, Br), the ICP-MS parameters and mathematical corrections were optimized to cope with such interferences. Following optimization, three correct and accurate (<2%) isotope ratios were obtained with 77Se, 78Se, 80Se and 82Se. The optimized method was then applied to an arable and a forest soil submitted to an aging process (drying/wetting cycles) during three months, to which 77Se(IV) was previously added. The results showed that ambient Se was at steady state in terms of water leachability, partition between soil solid phases (exchangeable Se and Se associated to organic matter) and speciation. At the opposite, the retention strength, solid phase partition and speciation of 77Se(IV) were modified during the experiment time-course and presented different kinetics. 77Se(IV) behavior tended to be similar to the one of ambient Se but still remained less strongly retained and chemically transformed at three months.

We concluded that kinetically limited processes are involved in Se retention and transformation in soils and that commonly used short-term experiments (<1 month) do not consider them properly. Otherwise, it seems more judicious to study ambient Se to infer the processes and parameters used in long-term risk assessment modeling. Since three correct and accurate Se isotope ratios were obtained, this new methodology can be further used to simultaneous monitor the reactivity of three different Se forms (e.g., added Se(IV), Se(VI) or Se(0), ambient Se), that will be useful for both soil Se contamination and supplementation contexts.