Temporal trends in concentrations and speciation of sulfur and selenium in rainwater

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Abstract

Atmospheric deposition is a main source of many chemical elements to terrestrial systems, including (micro)nutrients such as sulfur and selenium. Due to stricter controls on industrial emissions, especially in Europe and the United States, concentrations of (micro)nutrients in wet deposition have decreased over the last few decades. Apart from lower inputs to soils, declining industrial emissions may also lead to a higher relative importance of natural sources and pathways in biogeochemical cycles, which potentially changes the distribution of such (micro)nutrients in terrestrial systems. We therefore investigate biogenic sources of sulfur, selenium, and other trace elements in rainfall and their temporal dynamics at different locations in Europe. Hence, we examine variations in concentrations and speciation of these elements over time using (HPLC)-QQQ-ICP-MS and HG-AFS, in addition to studying sulfur and carbon isotopic signatures in rainfall. First results show that total selenium and sulfur concentrations correlate significantly with bromine and dissolved organic carbon contents in rainfall. In marine-influenced locations, these relationships are stronger than in more continental locations. Furthermore, concentrations of sulfur, selenium, and bromine in rainfall were generally higher at sites closer to the coast than in more continental sites. Initial selenium speciation analyses using HG-AFS revealed seasonal differences: the ratio of selenite to total selenium (Se[IV]/Setot) was higher in summer compared to winter. These data indicate that despite the strong oxidation potential of the atmosphere in summer, reduced selenium species are present, pointing to a higher share of biogenic sources during summer. Isotope analysis as well as comprehensive speciation analyses of selenium, sulfur and bromine in rainfall are currently underway. Mechanistic understanding as well as quantification of biogenic inputs of (micro)nutrients via atmospheric deposition is of key importance for predicting biogeochemical cycling of these elements in marine and terrestrial systems.

Keywords: elemental speciation, rainfall, time series analyses, sulfur, selenium

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