

Advanced Analytical Approach for Characterizing Uranium-Organic Matter Interactions

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Uranium plays a major role in both natural and anthropogenic environments, where its speciation influences its mobility and ecological impact. Natural organic matter (NOM) is ubiquitous in natural aqueous systems, and forms highly mobile nanoscale colloids that are able to increase the transport of pollutants. Thus, understanding the interactions between uranyl ions (UO_2^{2+}) and NOM nano-colloids is essential for predicting its behavior in natural subsurface systems.

In order to explore uranium speciation in presence of NOM colloids in a soil interstitial water, we employed an advanced analytical approach to examine quantitatively the distribution of uranium between the dissolved and colloidal phases in the presence of NOM.

Asymmetric flow field-flow fractionation (AF4) coupled with inductively coupled plasma mass spectrometry (ICP-MS) was used to characterize the size distribution of the colloidal fractions and qualify uranium association within each fraction. An insight in the calibration procedure of AF4/ICP-MS coupling allows determining the quantitative distribution of U(VI) along the colloidal size continuum.

This procedure is applied to determine uranium speciation in a suspension of in-field sampled NOM colloids through experimental isotherms. We show that uranium distributes equally between the dissolved and the colloidal phases, with the balance influenced by soil lixiviation conditions. However, this distribution remains principally related to the NOM compounds and relatively stable across the dissolved uranium concentration range considered (1 $\mu\text{g/L}$ – 1 mg/L).

Additionally, ultra-high-resolution mass spectrometry (Orbitrap) was used to determine the detailed composition of NOM and identify the families of organic compounds likely involved in uranium complexation. Orbitrap analysis reveals that uranyl interacts with a wide range of organic compounds but shows a particular affinity for a specific sub-fraction of NOM.

These results will be used to feed a modeling approach of U(VI)-NOM interactions by integrating experimental data with thermodynamic simulations. Complexation constants for U(VI)-NOM association in both colloidal and dissolved fractions are evaluated from experimental isotherms, helping to predict their impact on uranium mobility in natural environments.