Impact of redox chemistry on the fate and transport of arsenic and uranium at an abandoned uranium mine

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Soil samples from the North Cave Hills abandoned uranium mine sites in Harding County, SD were analyzed to determine the chemical mechanisms controlling the transport of As and U, previously found at elevated concentrations at the mine site tailings and surrounding watershed. A two phased approach was undertaken to examine the soil chemistry and geochemical behavior of these redox sensitive species. First, the soil was characterized to determine the site-specific mineralogy and the As and U speciation. Second, changes in As and U speciation due to fluctuations in soil redox potential resulting from microbial Fe reduction were examined within simulated batch reactor experiments. Changes in aqueous and solid phase As, U, and Fe were monitored with or without the addition of lactate, acetate, or ethanol as the electron donor for microbial respiration.

X-ray absorption near-edge structure (XANES) analysis on anoxically collected soils indicates that As found in the mine tailings is most likely in a mineral form as opposed to adsorbed to iron oxy (hydr)oxides as observed within the downgradient watershed soils, and suggests that remobilization of As from the tailings is actively occurring. Elevated uranium concentrations were present in reduced sediments collected within a sedimentation pond at the base of the tailings, suggesting uranium immobilization due to reduction of U (VI) to insoluble U (IV) is occurring. Mineralogical characterization of iron-bearing clays and iron hydroxides was performed using x-ray diffraction (XRD), extended x-ray absorption fine structure (EXAFS), and electron microscopy. Preliminary results suggest that the anaerobic sedimentation pond efficiently sequesters U but is likely releasing As due to reductive processes. Results from the characterization of field soils and batch reactor experiments will be presented.

Fertile components of the NE Atlantic mantle – Minor elements and O-isotopes in olivine phenocrysts

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Primitive off-rift basalts in Iceland and Jan Mayen sample subcontinental lithospheric mantle (SCLM) in addition to the Iceland plume with recycled oceanic crust (ROC) and melt-depleted or primitive lower mantle material [1]. The SCLM/ROC-ratios are high at Jan Mayen (high La/Sm and 87/86Sr and low 206/238U, 187/188Os and 143Nd/144Nd), intermediate in the Snæfellsnes Flank Zone and low in the Southern Flank Zone, including Surtsey. The fertile components are also present in tholeiites in the Iceland rift zones and the NE Atlantic and Arctic ridges, despite larger proportions of melts from refractory asthenosphere. The SCLM/ROC-ratios are high where the distances to the nearest continental margin or the continental Jan Mayen Ridge are relatively short.

Minor element abundances and O-isotope ratios of olivine phenocrysts in the primitive off-rift basalts support the chemical provincialism. Ni, Cr and Mn form separate linear arrays as a function of forsterite content (Fo) for the Surtsey, Snæfellsnes and Jan Mayen sample suites. The Ni-Fo and Cr-Fo arrays have positive slopes, whereas the Mn-Fo array has negative slope. At a given Fo value, the Ni and Cr contents decrease and the Mn and Ca contents increase from Surtsey via Snæfellsnes to Jan Mayen. This may be caused by larger melt contribution from olivine-free pyroxenitic veins under Surtsey [2]. The SCLM-component, most prominent at Jan Mayen, may represent old depleted lithosphere, re-fertilized on a small scale, with olivine still present in the matrix between thin pyroxenitic or eclogitic veins.

The δ18O-ranges of the olivine phenocrysts in samples from Surtsey, Snæfellsnes and Jan Mayen are 5.3-4.8, 5.3-5.0 and 4.9-4.6, respectively. The δ18O-values are negatively correlated with La/Sm and 87Sr/86Sr, and positively correlated with 143Nd/144Nd, 187Os/188Os and 3He for these combined sample suites. The O-isotope data indicate that the SCLM- and ROC-like components have low and high δ18O, respectively.