Role of iron oxides and organic matter in the accumulation of uranium in a peatbog

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A peatbog located in central France and impacted by both historical uranium mining activities and continued uranium leaching from granite rocks harbors U-enriched zones reaching a concentration of 4,000 mg/kg [1]. The mechanism leading to this remarkable accumulation is unknown. In order to unravel it and to evaluate the impact of proposed remediation strategies, extensive depth-resolved sampling of soil and porewater was carried out at three different areas as well as at a U-free background location in July and November 2011. Extensive physicochemical and microbial characterization of the four locations was conducted.

The porewater results show decreasing dissolved oxygen (DO) and increasing Fe(II) concentrations at 15-35 cm, suggesting that a redox transition zone (RTZ) occurs at that depth. The Fe(II) and total Fe profiles overlap exactly, suggesting that there is no measurable Fe(III) in the porewater. Secondly, a strong correlation was found between the U and Fe and U and total organic carbon (TOC) profiles, suggesting that U mobility may be controlled by adsorption to Fe oxides above the RTZ and by organic matter complexation in the porewater below that zone. Chemical extraction of U from soil revealed that U is concentrated at shallow depths (above 45 cm). Moreover, soil U occurs as a mixture of U(IV) and U(VI) and the profile of the ratio of U(VI)/U_{total} displays no clear trend as a function of depth. The persistence of U(VI) in a reduced environment may be attributable to its stabilization by complexation with organic matter. Finally, a clay layer, composed mainly of muscovite and quartz and rich in U, occurs at a depth of 10-25 cm in the peat soil. This unusual occurrence may have resulted from an anthropogenic input into the peat bog. The results to date suggest the involvement of iron oxides in immobilizing U in the oxic zone and the role of organic matter in binding U below the RTZ. Further mineralogical and microbial characterizations are underway to probe that current intrepretation of the data.

[1] Moulin (2008). PhD thesis, Ecole Centrale de Paris.

Abundances of sulfur, selenium and tellurium in mantle peridotites: constraints on planetary fractionation and late accretion

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The addition of a chondritic late veneer after core formation has been a popular explanation for the high abundances and mostly chondritic ratios of highly siderophile elements (HSE) in the silicate Earth. The HSE are mostly hosted by sulfides, which also control the mantle's budget of S, Se and Te [1]. S, Se and Te are moderately to highly siderophile at high P-T conditions [2], but they also are moderately volatile elements with very similar 50% condensation temperatures [3]. Thus, the detailed fractionation of these elements in the mantle, also in relation to HSE abundances, may provide insight into both the volatile element composition of a late veneer and the influence of metal-silicate partitioning on their mantle abundances. Until recently, the mantle abundances of Se and Te have not been well constrained and the influence of secondary alteration, partial melting and refertilization on S, Se and Te abundances are incompletely understood [1,4]. To address this problem, we have obtained new precise S, Se and Te data on post-Achean peridotites. Fresh samples of variable fertility were digested in a high pressure asher and S, Se and Te concentration data were obtained on the same digestion aliquot by isotope dilution ICPMS. Sulfur, Se and Te concentrations display excellent linear correlations, and decrease with decreasing Al_2O_3 contents. In spite of large concentration variations, Se/Te remains relatively constant (6.8 ± 3.1, 2sd, n=39, total range from 3.8 to 10.0) with variable fertility. S/Se in lherzolites also displays no systematic variation (2710±1580, n=27, most within 2000-4000), whereas harburgites tend to have higher values. Similar results (S/Se=3120 ± 500, Se/Te=8.6±2) have been obtained on lherzolites from Lherz (Pyrenees), which are believed to reflect melt refertilization [4]. The absence of substantial systematic variations of S/Se and Se/Te with fertility suggests that during moderate degrees of partial melting or refertilization no systematic fractionations occur between S, Se and Te. This appears to be at odds with Se and Te data on MORB [5], but the latter may have been influenced by crustal fractionation processes. We conclude that Se/Te in mantle peridotites is constant, but 20-30% lower than CI chondrite (Se/Te ≈ 9), whereas S/Se might be about 10-20% higher than in CI chondrites. If the experimental partition coefficients are valid [2], the Se and Te budget of the mantle should reflect the contribution from a late veneer, whereas the suprachondritic S/Se, possibly hints at significant residual S in the mantle after core formation. CI chondrite normalized Te/Ir_N, Se/Ir_N and S/Ir_N of the mantle are 0.5 to 0.6 and constrain the predominant composition of the late veneer. These values are consistent with slightly volatile depleted CM2 or transitional C1/C2 chondrites.

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