Oxidation of uranium oxide aerosol particles in the near-surface environment

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Between 1958 and 1984, microscopic particles of depleted uranium (DU) oxide were emitted as an aerosol from a manufacturing plant located in a populated area of Colonie, NY, and deposited in sediments, soils, and interior dusts. During this period, plant workers and nearby residents were exposed to significant amounts of DU through inhalation [1]. This study focuses on a interior dusts and a dated sediment core from a nearby reservoir that contains a detailed historical record of particle deposition [2].

We have identified more than 50 discrete, DU-bearing particles with diameters <1 to 30 μm (mean = 5 μm). Particle morphologies include spheres, irregular shards, and agglomerates of micron- to submicron-size particles. Particle compositions determined by electron microprobe range from 45-100 wt % UO_2, with minor Si, Ca, Pb, and Fe.

U L_{III}-edge μ-XANES spectra were compared between DU particles collected from interior dusts (dry) and from sediments (wet). Absorption edge profiles and energies of the dust sample spectra are similar to the $U(IV)O_2$ model compound, indicating a preponderance of U(IV). In contrast, the edge positions of sediment sample particles are shifted to higher energies, suggesting a higher proportion of U(VI) relative to the dust samples. In one case, the absorption edge position of a sediment particle was nearly identical to the U(VI) model compound metaschoepite. These results suggest that U oxidation state and phase relations of these particles are functions of environment (wet vs. dry) and exposure age.

[1] Parrish *et al.* (2008) *Science of the Tot. Environ* **390,** 58-68. [2] Arnason & Fletcher (2003) *Environ. Pollution* **123,** 383-391.

The Holocene geochemical fingerprint of outer Alps denudation

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Lake Bourget is a hard water fjord-type lake located at the footstep of French northern Alps. Since the Early Holocene, sedimentation is mainly related to biogenic calcite precipitation. Superimposed to this autochthonous fraction, variable amounts of detrital siliceous material are delivered into the lake through major river Rhône floods. During such flood event, the sediment-providing catchment area covers a surface of 2000 km^2, and includes the Mt Blanc massif. This conformation allows reconstructing both the quantity and quality of erosion products at the exit of the alpine range all along the Holocene period.

We show that most of the Holocene was a period of reduced solid discharge, which may be related to particularly low denudation rates. In contrast, since 2.7 ka cal. BP, the solid discharge in Lake Bourget dramatically increases and reaches a maximum during the so-called Little Ice Age period (1300-1900 AD). As 2.7 ka cal BP corresponds to the onset of Iron Age, one may question whether such a rise in the detrital input is to be related to natural or anthropic factors. Indeed, it has been proven that tillage using iron tools is much more efficient and affects a substantially deeper soil layer than it did using bronze tools. To address this problem we applied a source-to-sink geochemical and mineralogical approach in order to track the origin of detrital sediments all along the Holocene. Coupling clay mineralogy, major elements contents and Sr and Nd isotope geochemistry, we were able to demonstrate that periods of enhanced detrital fluxes correspond to higher relative contribution of the Mont Blanc massif to the solid discharge. Based on this result, we argue that the rise in erosion rates deciphered from our detrital signal during the last third of Holocene is rather due to climatic forcing. We propose a pervasively advanced position of Mt Blanc glacier as a primary forcing of enhanced denudation.