Weathering processes and supergene formation of uranium bearing minerals at U-mines in the Saint-Sylvestre Area (French Massif Central)

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In France, uranium mines were exploited between 1945 and 2001 leading to the production of 76,000t of U but also 200 Mt of waste rocks. We present the supergene evolution of waste rock piles generated during mining operations at the Vieilles Sagnes site (Fanay) located in the Saint-Sylvestre leucogranite complex (northwestern Limousin, French Massif Central). This two-mica granite was emplaced at 324 ± 4 Ma and hosts an important U-ore mineralization deposited at 270-280 Ma, due to sustained hydrothermal circulation.

At the Vieilles Sagnes site mining operations between 1957 and 1965 generated waste rock piles of dominantly granitic rocks that have been exposed to supergene processes since the mining time. The U content of the blocks in the waste rock pile is 10-120 ppm, while in the ore body it is around 1000-3000 ppm.

To assess their supergene evolution and the neo-formation of uranium bearing phases two trenches were escavated through the waste rock pile, enabling the sampling of both superficial blocks and samples from the interior of the pile. Granitic blocks, proto-soils and alteration products that form the matrix of the waste rock pile, as well as paleo-soils underlying the rock pile were sampled.

The micromorphology, mineralogy and geochemistry of the samples representative of the different horizons of the profile investigated were examined by optical microscopy, and SEM observations, XRD and ICPMS whole rock analyses, respectively.

The degree of weathering of the granitic blocks is evidenced by the occurrence of chlorite and kaolinite alteromorphs after biotite and smectite and kaolinite after feldspar. The liberation of Fe gives rise to the local formation of Fe-phosphates and sulphates. Magmatic and hydrothermal U-bearing minerals release U as alteration and arenisation of the granites take place. So far no coffinite or uraninite from the initial mineralization have been observed.

Preliminary results suggest that the migration of uranium and formation of uranyl phosphates is at least partly associated with the supergene formation of Fe-phosphates. U-sorption onto clay minerals also seems to play an important role in decreasing U-mobility during meteoritic weathering.

Non-mass-dependent oxygen isotope enrichments in O_3 and CO_2: New insights from experiments, observations, and modeling

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Results from recent laboratory experiments, atmospheric observations, and photochemical kinetics modeling provide new insights inot the anomalous ^{17}O and ^{18}O enrichments in ozone and their transfer to carbon dioxide. In particular, new observations of the triple oxygen isotope composition of stratospheric CO_2 reveal surprisingly large anomalous enrichments that vary with latitude, altitude, and season. The triple isotope slopes were 1.95±0.05(1σ) in the middle stratosphere and 2.22±0.07 in the Arctic vortex versus 1.71±0.03 from previous observations and a factor of 4 times larger than the mass-dependent value of 0.52. Kinetics modeling of laboratory measurements of CO_2-ozone isotope exchange demonstrates that non-mass-dependent isotope effects in ozone formation alone quantitatively account for the ^{17}O anomaly of CO_2 in the laboratory, resolving long-standing discrepancies between models and laboratory measurements. Model sensitivities then provide a framework for understanding ^{17}O in stratospheric CO_2 and the larger three isotope slopes observed there than in laboratory experiments and, thus, a firmer foundation for the many biogeochemical and paleoclimate applications of ^{17}O anomalies in tropospheric CO_2, O_2, minerals sulfates, and fossil bones and teeth, all of which derive from isotope exchange between ozone and stratospheric CO_2. Finally, measurements of the pressure and bath gas dependence of the ^{17}O and ^{18}O enrichments in ozone in bulk photochemistry experiments in the laboratory [1] provide new insight into and constraints on the physical chemical origin of the dynamically-driven symmetry isotope effects in ozone formation.

[1] Feilberg, Wiegel, & Boering (2013), *Chem. Phys. Lett.* **556**, 1-8.

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