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# Evolution of noncrystalline uranium in lake sediments over 3,300 years

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The long-term behavior of uranium (U) in contaminated soils and sediments downstream former U-mines is a widespread environmental challenge. Although reducing subsurface environments are generally considered as favorable for U immobilization, it has been shown that U is often present in the form of potentially labile noncrystalline U(IV) species in contaminated anoxic sediments [*e.g.*, 1, 2] and soils [*e.g.*, 3]. The fate of such mononuclear or polymeric U(IV) species and their ability to eventually transform into less soluble minerals at the centennial or millennial scale remains poorly documented, which thus deserves further research to improve the management of contaminated sites.

Here, we will present our recent findings [4] on the evolution of noncrystalline U(IV) species over 3,300 years in the sediments of Lake Nègre (2354 m above sea level, Mercantour Massif, France) that display exceptional natural U concentrations (up to 760 µg/g), comparable to those found in contaminated systems. Uranium isotopic ratios ($\delta^{238}U$ and ($^{234}U/^{238}U$)) are constant along the sediment core, indicating that U sources and deposition patterns remained constant over the considered period, and allowing to highlight the effect of early diagenesis on U solid speciation. We used Extended X-ray absorption fine structure spectroscopy (EXAFS) at the U $L_3$-edge and Transmission Electron Microscopy to determine U speciation along the sediment core. The obtained results show that U was initially deposited in the sediments as mononuclear U(IV) bound to organic matter, and then evolved in less than 700 years into an authigenic polymeric/nanocrystalline coffinite ($U^{IV}SiO_4 \cdot nH_2O$)-like phase. We thus reveal the low temperature formation of a coffinite-like phase likely favored by high biogenic silica supply from diatoms to the sediment pore waters. However, selective extractions show that the formation of polymeric U-silicates does not substantially enhance U scavenging, underlining the need for sustainable reducing conditions to allow long-term U sequestration in organic and silica-rich environmental conditions.

[1] Morin *et al.* (2016) *Geochem. Persp. Let.* 2, 95-105; [2] Stetten *et al.* (2018) *Geochim. Cosmochim. Acta* 222, 171-186; [3] Stetten *et al.* (2018) *Environ. Sci. Technol.* 52(22), 13099-13109; [4] Lefebvre *et al.* (2021) *Proc. Natl. Acad. Sci. USA* 118(4), e2021844118.