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Controls of climate and organic matter on uranium fluxes to lake sediments over the Holocene

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One of the main factors controlling the behavior of uranium (U) in water bodies is bottom water oxygenation, which enables the use of U isotopic ratios and concentration in oceanic sedimentary records as paleo-redox proxies [1]. Here, we investigated the mechanisms governing U accumulation in the sediments of Lake Nègre (2354 m above sea level, Mercantour Massif, France) over the past 9200 years. These sediments display exceptional natural U concentrations (300–1500 $\mu g.g^{-1}$), allowing for the first time the use of high-resolution X-Ray Fluorescence core-scanning for the measurement of U. Geochemical proxies (Ti content and K/Ti ratios) indicate that erosional sedimentary inputs were controlled by Holocene climatic variations. After a period of low erosion during the Holocene Climatic Optimum, a major regime shift was recorded at 4.2 kyr BP when terrigenous fluxes increased until present with high sensitivity to centennial-scale climatic variations. The temporal evolution of carbon to nitrogen (C/N) and bromine to organic carbon (Br/TOC) ratios indicate that sedimentary organic matter (OM) was mostly of terrigenous origin from the catchment soils until 2.4 kyr BP. From 2.4 kyr BP to present, lake primary production and soils equally contributed to sedimentary OM. The Fe/Mn core profile points out that the lake bottom water oxygenation remained constant over the past 9200 years. Uranium depositional conditions were also constant over this period as attested by the $^{238}U/^{235}U$ (expressed as $\delta^{238}U$) and ($^{234}U/^{238}U$) ratios. However, U fluxes to the sediments varied substantially and were correlated to terrigenous OM fluxes from 7 kyr BP to present. This correlation highlights that U supply to the lake was controlled by U scavenging in soils of the watershed followed by transport of U bound to detrital organic particles. The fluctuations of U sedimentary inputs thus appear to be controlled by climate-driven variations in terrigenous OM production and erosion, rather than by changes in bottom water oxygenation. This finding confirms that the use of U (and potentially other metals with high affinity to OM) concentrations alone should be used with caution for paleo-redox reconstructions.

[1] Lau *et al.* (2019) *The uranium isotope paleoredox proxy*. Cambridge University Press.