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### Quantifying the effect of weathering processes on the chemical signal to the oceans

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Radiogenic isotope records of seawater potentially provide fundamental information on ocean circulation, erosional sources, and weathering processes, but the challenge remains to distinguish between such effects. The largest changes in the radiogenic isotope chemistry of seawater have actually been in the North Atlantic, rather than Pacific or Indian Oceans, suggesting, at first sight, a major change in erosional flux associated with Quaternary glaciation. However, recent studies have shown that the weathering and erosional processes accompanying glaciation may themselves have played a role in generating this chemical signal.

Here we present multi-isotope data (both short- and long-lived isotopes) from rivers and sedimentary records, and show how these can be used to obtain information on weathering processes and their timescales, and erosional transport mechanisms, and to resolve those changes caused by variations in erosional source from those caused by changes in weathering congruence. This will include: coupled Hf-Nd and Sr measurements of river waters draining silicate rocks that provide the first direct evidence for incongruent weathering of a radiogenic Hf phase. Re-Os measurements of rivers draining basaltic rocks, which also show clear evidence for incongruent weathering, yielding very radiogenic Os isotope compositions. This is because the phases that are the most susceptible to chemical weathering, such as glass, olivine and plagioclase, also possess exceptionally high Re/Os ratios, yielding radiogenic Os isotope compositions in very short periods of time. U-series isotopes for the same basaltic rivers, show fractionation of <sup>234</sup>U relative to <sup>238</sup>U ( $\alpha$ -recoil effects) and U relative to Th, and indicate short weathering timescales. Li-isotopes show covariations with U-series isotopes, suggestive of variations in weathering intensity. Finally, we will show how high-resolution Ar chronology on detrital phases can be used to resolve those changes caused by variations in erosional source on a large scale from those caused by changes in weathering congruence.

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### Silicate weathering mechanisms inferred from direct measurements of <sup>176</sup>Hf/<sup>177</sup>Hf ratios in river waters

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Chemical weathering of silicate rocks represents an important sink for atmospheric CO<sub>2</sub> on geologic timescales and therefore has played a significant role in the Earth's climate. Previously, many attempts to reconstruct the link between climate change and crustal weathering have focused upon measurements of radiogenic isotope ratios, such as Sr and Os, in ancient seawater records. Recent investigations of deep-sea ferromanganese crusts and nodules have suggested that the Hf isotopic composition of past oceans may provide a more sensitive tracer of silicate weathering. On a  $\epsilon_{Nd}$  vs.  $\epsilon_{Hf}$  plot, ferromanganese precipitates define a 'seawater' array, characterised by radiogenic (high) Hf isotope values, which is distinct from the terrestrial array defined by oceanic basalts, continental crustal rocks and sediments. Previous studies have proposed that the preferential release of dissolved radiogenic Hf into river waters during weathering of the continental crust may account for these high <sup>176</sup>Hf/<sup>177</sup>Hf ratios in seawater. Until now, however, it has not been possible to test this hypothesis directly because of analytical difficulties in measuring <sup>176</sup>Hf/<sup>177</sup>Hf ratios in low level samples such as waters.

Here, we report the first measurements of <sup>176</sup>Hf/<sup>177</sup>Hf ratios in river waters, which demonstrate that preferential weathering of radiogenic mineral phases occurs during chemical erosion of silicate rocks.

Five rivers (Lépage, Moselotte, Vologne, Moselle, Avières) have been sampled from the Moselle River watershed (Eastern France), which comprise mostly silicate rocks (granite, gneiss, sandstone) of Variscan age (~300 Ma). After pre-concentration onto organo-metallic compounds and chemical separation, <sup>176</sup>Hf/<sup>177</sup>Hf ratios in dissolved river fractions were measured by MC-ICPMS. The Hf isotopic composition ( $\epsilon_{Hf}$ ) for dissolved phases of Moselle rivers varies significantly (from  $-13.91 \pm 1.64$  to  $-0.63 \pm 1.56$ ), while suspended sediments are isotopically homogeneous, displaying lower  $\epsilon_{Hf}$  values (from  $-14.65 \pm 0.74$  to  $-10.82 \pm 0.74$ ). Examination of these results with other isotopic (Nd, Sr) and major element data suggests that variations of <sup>176</sup>Hf/<sup>177</sup>Hf ratios in Moselle rivers are due to the selective dissolution of specific mineral phases during silicate weathering. Isotopic composition of dissolved Hf exhibits good correlation with (Mg+K)/Si ratios, a potential indicator of biotite weathering in silicated drainage basins.