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The global imprint of shale weathering on molybdenum isotope ratios in river waters

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The molybdenum (Mo) isotope ratio (δ^{98}Mo) of marine sediments holds information on the redox state of the past ocean [1]. However, a robust interpretation of the sedimentary δ^{98}Mo record requires identifying the main controls on the δ^{98}Mo values of riverine inputs. There is a growing consensus that secondary mineral formation following rock weathering sets the δ^{98}Mo values of the dissolved riverine flux [2]. However, sedimentary rocks show marked variability in δ^{98}Mo values suggesting a potential lithological control. Here we assemble a δ^{98}Mo dataset (including river water and sediments) for large rivers spanning a broad range of sulfate abundance used here as an index of the extent of sulfide oxidation in the catchment.

Collectively, our results indicate that both rock source type and secondary processes control riverine dissolved Mo isotopes. First, the elemental and isotope partitioning of Mo between the river dissolved and solid loads is indicative of the impact of secondary weathering product formation. Second, the river solid Mo isotope signature likely reflects contribution from sulfide-rich shale in catchments where the weathering of such lithology is significant. The positive relationship found between dissolved and solid Mo isotope signatures indicates a similar lithological control for both river loads. Further, in most rivers, silicate weathering only cannot explain the dissolved Mo abundance, supporting the idea of the variable contribution from an additional Mo-rich source. Finally, dissolved Mo isotopes are related to sulfide oxidation tracers, further supporting the variable control of sulfide weathering on river dissolved Mo isotope signatures. The relationship between dissolved Mo isotopes and the weathering regime may be interpreted in terms of geomorphological and climatic controls on the relative roles of silicate vs. sulfide weathering at the global scale.

Overall, these findings indicate that the Mo isotope signature of rivers could be used as a tracer of sulfide oxidation in the Critical Zone, and that sulfide oxidation has most likely affected past river Mo fluxes, especially when global weathering intensities were low.

[1] Kendall et al. (2017), Reviews in Mineralogy and Geochemistry 82.1, 683-732.

[2] Revels et al. (2021), *Earth and Planetary Science Letters* 559, 116773.