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An isotopically heavy source of nickel: Release of nickel during birnessite transformation into todorokite

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Nickel is a bio-essential element in the oceans and thought to have played an important role in the evolution of Earth's atmosphere due to its requirement by methanogenic archaea [1]. Therefore Ni and its isotopes show growing promise as tracers for key biogeochemical processes. For Ni to be developed into a viable proxy further understanding of its modern biogeochemical cycling is required. An outstanding conundrum of the Ni cycle is the flux and isotope imbalance of the marine budget. Current estimates show the output flux of Ni to marine sediments is 10.6×10^8 mol/yr double the input flux from mostly rivers at 5.3×10^8 mol/yr [2].The isotope composition of known sources is also considerably lighter than known sinks [2,3]. To balance the marine budget, recent studies suggest there could be an undiscovered isotopically heavy benthic flux of Ni into the ocean, generated during the diagenetic remobilisation of Ni in marine sediments, but whether and to what extent such a flux exists are unknown [2,3].

In marine sediments Ni is scavenged by Mn oxides like birnessite, which transforms into todorokite during oxic diagenesis [4]. To investigate the isotopic composition of Ni during diagenesis, we transformed a synthetic Ni-Mn oxide that is analogous to marine birnessite into todorokite via a mild reflux procedure [5,6] and measured the Ni isotopic composition of the solution and solid phase. Nickel released to solution is heavier than the mineral with an isotope fractionation factor of $\delta^{60}Ni_{sol\text{-}min} = 0.9 \pm 0.08$‰ ($\delta^{60}Ni_{sol\text{-}min} = \delta^{60/58}Ni_{solution} - \delta^{60/58}Ni_{mineral}$) after 4 weeks. Here we show for the first time, a heavy source of Ni is released during diagenesis of Mn oxides and suggest this could form an isotopically heavy benthic flux required to balance the Ni marine budget. We also use our observations to address large variations of Ni isotope compositions measured in oceanic Mn-rich sediments.

[1] Konhauser *et al.* (2009) *Nature* 458(7239):50-753 [2] Gueguen & Rouxel (2021) *Chemical Geology* 563:120050. [3] Little *et al.* (2020) *EPSL* 547:1646. [4] Bodeï *et al.* (2007) *GCA* 71(23):5698-5716. [5] Atkins *et al.* (2014) *GCA* 144:109-125 [6] Atkins *et al.* (2016) *GCA* 189:158-183.