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Stable isotope tracing of Mineral-Associated Organic Matter interactions with ambient water

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In natural systems, organo-mineral aggregation can influence the reactivity and sequestration of carbon (C), while biochemical, photochemical or redox transformations of particulate and dissolved organic matter (DOM) permit the incorporation of stable hydrogen ($^{2/1}$H) and oxygen ($^{18/16}$O) isotopes from ambient water. In a recent study [1], relatively high ^{18}/^{16}O ratios of DOM observed in a subalpine, oligotrophic lake were attributed to the input of mineral-bound DOM. This study highlights the potential for mineral-associated organic matter (MAOM) to be imprinted with the isotopic signatures of source water. However, there is still little information on the factors controlling the δ^2H and δ^{18}O compositions of MAOM. Aiming to fill this research gap, we performed sorption experiments to equilibrate goethite and montmorillonite with cyclodextrin or menaquinone in isotopically light water (δ^2H = - 460 ‰ and δ^{18}O = -196 ‰; pH 7) for 72 hours. The exchangeable fraction of H and O (*f*ex) and the non-exchangeable stable isotope compositions (δ^2H_n and $\delta^{18}O_n$) of the resulting MAOM were then determined by UniPrep-IRMS. Increasing organic C sorption by minerals was proportional to increases in the MAOM total H and O contents, and menaquinone was consistently more sorbed than cyclodextrin (5-10% vs. < 2% C w/w). Montmorillonite exhibited higher f_{ex}-H and f_{ex}-O (1-2%) compared to goethite (< 1%), and there was no exchangeable H detected for the menaquinone-goethite particulate. δ^2H_n values of MAOM were consistently lower than the pure compounds, likely reflecting the imprint of water-H into the organo-mineral framework. These findings suggest that the capacity for MAOM to be imprinted with $^{2/1}$H and $^{18/16}$O signatures of source water relies on the polarity and structure of the organic molecules and the mineral sorption capacities, and that stable isotope probing assays may be useful for determining MAOM formation processes. Ongoing, parallel investigation of MAOM in the dissolved phase and characterization by scanning electron microscopy, and energy dispersive X-ray spectroscopy and diffraction will further detail the mechanisms of organic-mineral interactions. These preliminary insights highlight the potential for $^{2/1}$H and $^{18/16}$O compositions to inform MAOM transformations and reactivity.

[1] Pilecky et al. (2023), *Science of the Total Environment* 891, 164622.