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Hafnium and neodymium isotope measurements in seawater - a powerful combination of geochemical tracers

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Dissolved hafnium (Hf) in seawater is a promising tracer for ocean circulation, water mass mixing and element source provenance. However, the processes involved in the transport of Hf to the oceans and the potential sources of dissolved Hf are not well constrained. Studies of marine ferromanganese crusts (Fe-Mn crusts), which have been interpreted to record the seawater Hf isotope composition, suggested rivers, eolian input of continental material, young volcanogenic material, or oceanic crust-seawater interaction as potential Hf sources. In combination with neodymium (Nd), which is a better-established water mass tracer, these isotope systems can provide valuable information on ocean circulation and continental weathering inputs into the ocean.

We present the first direct measurements of Hf isotopes in seawater. These data allow us to draw conclusions about the pathways of Hf in the aquatic environment and to assess the importance of the various possible sources, as well as the influence of the style of weathering. The MC-ICPMS measurements of seawater Hf from the NW Pacific and the Arctic Ocean show distinct variations in the $^{176/177}$Hf ratio. In the ε_{Hf} notation, for which the measured $^{176/177}$Hf is expressed relative to the CHUR $^{176/177}$Hf (chondritic uniform reservoir), Pacific samples range from ε_{Hf} = 3.5 to 8.6 (average ε_{Hf} = 5.9). Possible source rocks for this data range are the circum-Pacific arc volcanics. The Arctic seawater, with a much lower average of ε_{Hf} = 0.6, varies from ε_{Hf} = -2.9 to 6.8. Hf measurements for the largest Arctic rivers display an even larger spread than seawater from ε_{Hf} = -7.0 (Mackenzie) to ε_{Hf} = 1.5 - 3.0 (Ob, Lena, Yenisei). Thus, a large part of the variation in Arctic seawater ε_{Hf} data can be explained as a result of a mixed riverwater signal. The highest Arctic seawater Hf values, which have been measured in samples from the Alaskan coast, indicate the advection of seawater Hf from the N Pacific to the Arctic Ocean through Bering Strait.

To explain the seawater Hf-Nd systematics, their nature in the mantle and the continental crust need to be considered. The Lu-Hf and Sm-Nd element ratios behave similarly in the major geochemical processes within the earth, and hence Hf and Nd isotopes show a strong correlation for terrestrial samples (continental crust and oceanic basalts). This is in contrast to the Hf and Nd isotope compositions derived from Fe-Mn crusts and Mn nodules. Although they are correlated along an array, there is a distinct offset from the terrestrial Hf-Nd isotope array towards higher ε_{Hf} for a given ε_{Nd}. This offset has been explained as a result of incomplete weathering processes. In the continental crust, where the lowest ε_{Hf} and ε_{Nd} values have been determined, Hf contrary to Nd is mainly incorporated into zircons. Since this is an erosion-resistant mineral, the low ε_{Hf} is strongly retained during weathering processes, resulting in a decoupling of Hf and Nd. Only in the case of strong mechanical weathering and the breakdown of zircon, low ε_{Hf} will be released into the aquatic cycle. For basaltic rocks on the other hand, with higher ε_{Hf} and ε_{Nd} values, the release of both elements during erosion is similar. Thus, there is less discrepancy between Fe-Mn crusts and the corresponding basaltic source rocks in Hf-Nd isotopes. The combined Hf-Nd isotope data from our NW Pacific seawater samples overlay the Pacific Fe-Mn crusts data, indicating the importance of the source rocks and the style of weathering for the resulting seawater Hf and Nd isotope signature. The Arctic Hf-Nd seawater data show a clear offset from the terrestrial array, in agreement with the domination of continental crust surrounding the Arctic Ocean. Comparable Atlantic Fe-Mn crusts are slightly higher in ε_{Hf} than the Arctic seawater. This difference can be explained as a result of the cold Arctic climate, causing stronger mechanical erosion and the partial release of low ε_{Hf} from the source rocks.