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1 Calcium Isotope Variations along the Hydrothermal Pathway

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Hydrothermal fluid geochemistry provides insight into water-rock-interactions during hydrothermal activity. Seawater underlies progressive chemical changes along its pathway through oceanic crust and is modified to a hydrothermal fluid under exceptional temperature and pressure conditions. In this study, we investigate the evolution of seawater within hydrothermal systems with respect to calcium.

The Ca isotope compositions of 21 fluid samples from the Logatchev Hydrothermal Field at the Mid-Atlantic Ridge (15˚N) were determined. The $\delta^{44/40}$Ca ratios of the fluids are inversely correlated to their corresponding Ca and Mg contents and thus to the fluid amounts as deduced by Mg endmember calculations. Binary mixing calculations lead to a Ca concentration of 32 mM and a $\delta^{44/40}$Ca value of -0.97 $\pm$ 0.11 per mill ($2\sigma_m$) relative to seawater for the hydrothermal endmember. Similar to Ca, the Sr concentrations and ^{87}Sr/^{86}Sr ratios are inversely correlated. A plot of $\delta^{44/40}$Ca against ^{87}Sr/^{86}Sr displays a two-component mixing curvature that yields endmember values of ^{87}Sr/^{86}Sr = 0.7037 $\pm$ (8) and $\delta^{44/40}$Ca = -0.92 $\pm$ 0.15 per mill. Both approaches combined result in a value of -0.95 $\pm$ 0.09 per mill (95 % confidence level) for the Ca isotope composition of the hydrothermal endmember. With regard to Sr, a two-endmember mixing with seawater and MORB as endmembers can be assumed. The Ca isotope ratios in contrast exhibit a significant deviation of about 0.2 per mill between the Ca isotope signature of MORB ($\delta^{44/40}$Ca = -1.17 $\pm$ 0.05 per mill ($2\sigma_m$))

(Richter et al., 2003) and that of the hydrothermal endmember as determined in this study. The difference might be based on either a different Ca isotope composition of the bed-rock, in particular with regard to the ultramafic-hosted Logatchev Hydrothermal Field or fractionation processes along the hydrothermal pathway. Currently, no Ca isotope data exist for ultramafic rocks. But it is assumed that the bulk silicate Earth displays a value of about –1.1 per mill relative to seawater.

The second suggestion, namely a potential fractionation during the formation of hydrothermal precipitates such as anhydrite and calcium carbonate would serve as an explanation when assuming a Rayleigh fractionation law. This approach would predict a Ca isotope composition of hydrothermal precipitates that are around -0.5 per mill lighter than the original solution from which they precipitated. Nonetheless host-rocks from the Logatchev Hydrothermal Field have to be analysed to verify one of these explanations.