Copper isotope fractionation during basalt weathering at pH = 0.3, 2, 5 and T = 25 ºC

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The geochemical cycling of copper in hydrosphere and soil environments involves the transport of Cu from rocks to waters by weathering. Understanding the mechanisms of isotope fractionation during continental weathering is a key point for application of Cu isotopes as a tracer for geochemical recycling. Previous experimental studies have focused on the leaching of sulfides at various conditions. However, to mirror the real process of continental weathering needs experimental studies of nature rocks. Here, we carried out leaching experiments for nature basalts (BHVO-2 and GBW07105) in Erlenmeyer flasks for 30 days at different pH values (0.3, 2, 5) and T = 25 ºC. For comparison, a chalcopyrite (Cpy) was also studied. The oxidative weathering of basalts produces a substantial fractionation of Cu isotope ($\Delta^{65}Cu_{aq\text{-}solid}$) between the leached solutions (Cu_{aq}) and the initial powders (Cu_{solid}). Cu initially released into solutions were isotopically heavier, with $\Delta^{65}Cu_{aq\text{-}solid}$ around +0.5‰ for GBW07105 (pH = 0.3) and around +0.6‰ for BHVO-2 (pH = 2). As the percentage of leached Cu increases, the $\delta^{65}Cu_{aq}$ values decrease first and then increase, as $\Delta^{65}Cu_{aq\text{-}solid}$ values vary from -0.4 to +0.5‰ for GBW07105 (pH = 0.3) and from -0.7 to +0.6‰ for BHVO-2 (pH = 2). However, the leached solutions are always enriched in heavy Cu isotope relative to the initial powders at pH = 2 for GBW07105 and pH = 5 for BHVO-2. Similarly, the Cu isotopic variation of the leached solutions from Cpy has a similar tendency with that of the leached solutions from BHVO-2 at pH = 2, and the leached solutions are also enriched in heavy Cu isotope at pH = 5. XRD patterns of the initial basalts and residual solids after each reaction interval do not reveal any phase changes. We conclude that the mechanism responsible for such Cu isotope fractionation is the relative rates of oxidation versus leaching, combining XRD of solids and isotopic analysis of the leached solutions. Our results show that weathering of nature rocks can produce both positive and negative fractionation, depending on the pH values. This has crucial important implications for using Cu isotopes to trace the palaeoenvironment.

Keywords: Cu isotopes; isotope fractionation; basalt; chalcopyrite; leaching experiments.