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Lithium isotope fractionation during low temperature leaching of basaltic glass: an experimental study

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Chemical erosion of silicates and in particular basalts has a major impact on the CO_2 cycle and needs to be quantified in order to understand its role on past, present and future climate (e.g. [1]). For this, lithium isotopes are powerful tracers as several field studies highlighted significant differences of isotopic compositions between drained rocks and river waters (e.g. [2]). However, the link between weathering mechanisms (diffusion, dissolution, secondary phase formation) and Li isotope signature of waters remain poorly understood.

Experimental studies provide a way to better constrain and quantify Li isotope fractionations during weathering, and therefore to interpret natural observations. At present experimental weathering of basalt, weathered basalt, and sediments were performed either at high temperature and high pressure, or in rock-dominated conditions, which favours secondary phase precipitation (e.g. [3], [4]).

In this study, we performed short-term ($<$ 1 week) experiments of basaltic glass alteration in water-dominated conditions, at various temperatures (25, 50 and 90°C) and pH (3 to 9). The objective is to determine Li isotope fractionation associated with the leaching and diffusion mechanisms. Since alteration solutions were highly diluted, a Li-enriched synthetic basaltic glass was used ($\sim$ 4480 ppm Li).

Li isotopes were measured with a Neptune MC-ICP-MS after a separation chemistry as described in [5]. Uncertainties on δ^7Li range from 0.2 to 1.4‰ ($2\sigma_m$, depending on the Li concentration during the measurement). Before the experiments we first checked the isotopic homogeneity of the synthetic basaltic glass by SIMS using a CAMECA IMS-3f. The standard-deviation on 34 in-situ δ^7Li measurements of various grains is less than 1.1‰

Results for the 90°C experiments show that the leaching solutions are all enriched in ^{6}Li (δ^7Li range between 4.9 and 10.3‰) relative to the synthetic basaltic glass (δ^7Li = 13.0‰), whatever the solution pH. A model that couples the formation of a leached layer by alkalis diffusion and the dissolution was developed. This model is based on Boksay's equations ([6]) but is here applied to Li isotopes. The good consistency of the simulated and measured δ^7Li allows the isotopic fractionation associated to Li diffusion in the leached glass (D_{7Li}/D_{6Li}) to be determined. The ongoing analyses of experiments performed at 25 and 50°C should provide additional constraints on the effect of temperature on diffusion coefficients and isotopic fractionation. Applied to natural field, these results demonstrate that, for basaltic terrains, the enrichment of ^{7}Li systematically observed in natural waters is only caused by the precipitation of secondary phases (clay minerals). However, apparent isotopic fractionation ($\Delta^7\mathrm{Li}_{water-sediments}$) may be underestimated without considering the effect of diffusion.

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