

Li isotope fractionation during biotite dissolution

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Silicate weathering controls the chemical composition of the oceans and the CO₂ level of the atmosphere on a geological timescale. Because lithium (Li) occurs predominantly in silicate minerals and is widely present as a trace element in biotite, where it can substitute for Mg, it has been known that Li isotopes are the most useful tracer of silicate weathering processes. Mineral dissolution at far from equilibrium does not fractionate Li isotopes [1] but diffusion-induced isotope fractionation occurs [2]. In this context, we dissolved biotite in a plug flow reactor at controlled pH and T = 25 °C to elucidate how and to what extent Li isotope fractionation occurs during biotite dissolution. Furthermore, we conducted a sequential leaching to better understand various potential sources of Li released during the dissolution. In the pH = 1 experiment, Li concentration increased to reach a steady-state values followed by decreased slowly towards near zero at the end of the experiment, while in the pH = 5 experiment Li concentration rapidly decreased until the end of the experiment. All leaching solutions, except one sample collected at 11 h in the pH = 5 experiment, during biotite dissolution are enriched in light isotope compared to the biotite sample, with 1.43‰ and 14.66‰ ranges of $\Delta^7\text{Li}_{\text{biotite-solution}}$ at pH = 1 and 5, respectively. At pH = 1, Li isotopic steady-state between the solution and biotite is established after ~16 h, while at pH = 5 it is never reached even after 4 months. Unlike Mg isotopes depending mostly on a balance between the relative proportions of labile and structural Mg with different $\delta^{26}\text{Mg}$ values [3], Li isotopes during biotite dissolution at pH = 5 cannot be explained by a balance between labile and structural Li although they display different $\delta^7\text{Li}$ values. This suggests that secondary minerals are formed during the incipient stage of dissolution, resulting in heavy Li isotope in solution, followed by leaching of secondary minerals driving the solution isotopically light.

[1] Pistiner and Henderson (2003), *Earth Planet. Sci. Lett.* **214**, 327-339. [2] Verney-Carron *et al.* (2011), *Geochim. Cosmochim. Acta* **75**, 3452-3468. [3] Ryu *et al.* (2016), *Chem. Geol.* **445**, 135-145.