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LITHIUM ISOTOPES IN LOW AND HIGH TEMPERATURE HYDROSYSTEMS

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1. Introduction
Assessing the origin and behaviour of lithium and the distribution of Li isotopes in hydro-systems is of major importance in order to increase our knowledge of the lithium cycling at the Earth’s surface. Lithium is a fluid-mobile element and due to the large relative mass difference between its two stable isotopes, it is subject to significant low and high temperature mass fractionation which provides key information on the nature of water/rock interaction processes. The main objective of the present work is to constrain the behaviour of Li and its isotopes by focusing on three different hydrosystems: rainwaters, river waters and deep geothermal waters.

2. Rainwaters
Firstly, we report Li isotope ratios in rainwater samples collected over a long time period (i.e. monthly rainfall events over 1 year) at a national scale (from both coastal and inland locations in France). A better understanding of the Li isotope signatures in rainwaters is needed to understand the origin of this element in the atmosphere and also to constrain the isotopic signature of the rainwater input to surface waters and/or groundwater bodies during recharge. The most striking outcome is that most Li present in rainwaters does not have a pure marine origin. This may be important in the understanding of dissolved Li distributions in ground- and surface waters, and should be taken into account in future studies.

3. River waters
Secondly, we report Li isotopic compositions measured in the rivers of the Mackenzie Basin (Canada) and we show that $^7$Li is significantly enriched in the dissolved load compared to sediments, and that $\delta^7$Li in the river waters can vary by 20% within a large river basin. This work also demonstrates that dissolved Li in river waters is essentially derived from the weathering of silicates and that Li isotopic ratios of the dissolved load depends on the weathering regime of silicates.

4. Geothermal waters
Finally, we also report Li isotope data for deep geothermal waters. One particularly important aspect of this work was to establish the nature, extent and mechanism of Li isotope fractionation as a function of temperature during water/rock interaction. The behaviour of Li and its isotopes have been characterized in geothermal systems located in volcanic island arc areas (Guadeloupe and Martinique islands). In addition, we report results of Li isotope exchange experiments during seawater/basalt interaction (from 25 to 250°C). These results confirm that Li isotopic exchange is strongly temperature dependent, and demonstrate the importance of Li isotopic fractionation during the formation of Li-bearing secondary minerals and allow us to determine the following empirical relationship between isotopic fractionation and temperature: $\Delta_{\text{solution}} / T - 8.093 = 7847 / T - 8.093$.

5. Concluding remarks
To summarize, this work shows that the fractionation of Li isotopes is dependent upon the extent of water/rock interaction both in terms of intensity (i.e. temperature in geothermal systems) and weathering regime of silicate rocks in surface waters systems (primary mineral dissolution and secondary mineral formation).

6. References
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