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Water Rock Interaction [WRI 14]

Chemical weathering of granitic rocks: experimental approach and Pb-Li isotopes tracing

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Abstract

In order to characterize water/rock interactions of granite, we performed laboratory experiments and Pb-Li isotopes tracing. The aim of the present work is to better constrain the processes of water/rock interactions both in terms of source and extent of weathering, by measuring major and trace elements as well as Pb and Li isotope signatures.

Keywords: weathering; granite; Pb isotopes; Li isotopes

1. Introduction and objective of the study

Weathering reactions, e.g. breakdown and alteration of rocks and minerals at the Earth's surface, supply solutes to both surface and ground waters. In a recent study [1] focusing on the lead geochemistry and Pb-isotope ratios of groundwaters along a small (53 km²) endoreic granitic catchment in India (Masheshwaram, Andhra Pradesh), we have shown that most of the lead in the groundwaters is of geogenic origin. Combining a weathering model and field observations, we were able to define a two steps weathering process that includes a control on the Pb-isotope ratios by accessory phases and by the main minerals from the granite in a second step of weathering. In order to go further and to better characterize water/rock interactions, we performed laboratory experiments with granite rock samples from this field site.

The aim of the present work is to better constrain the processes of water/rock interactions both in terms of (i) source (dissolution of different primary minerals and (ii) extent of weathering (precipitation of secondary minerals), by measuring Pb and Li isotope signatures. This is because 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 temperature mass fractionation which provides key information on the nature and extent of weathering processes.

2. Experimental setup and analytical methods

Laboratory experiments consisted in measuring the evolution through time of major and trace elements, as well as Pb and Li isotopic compositions of a rainwater solution in equilibrium with a granite powder. Experiments were carried out at 25°C with a solution/powder mass ratio of 10. 15 mL of reference solution TMRain-95 and 1.5 g of powdered granite were placed in screw-top Teflon® PFA beakers. The beakers were kept in a temperature-controlled oven, which temperature was maintained within 5% of target temperature over the total duration of the experiments. Aliquots of the solution (after filtration at 0.20 µm) in contact with the granite powder were periodically sampled (from weeks up to more than one year) and analyzed for lead and lithium isotopic compositions.

Lithium isotopic compositions were measured using a Neptune Multi-Collector ICP-MS at BRGM's Isotopic Geochemistry Laboratory. $^7\text{Li}/^6\text{Li}$ ratios were normalized to the L-SVEC standard solution (NIST SRM 8545) following the standard-sample bracketing method (see the method reported by [2]).

Lead isotopic compositions in waters were also measured using with the Neptune Multi-Collector ICP-MS in operation at the BRGM. According to the recent protocol developed by our group [3], the key parameter for the analysis of water by the mean of the MC-ICP-MS is the lead concentration and the cations matrix of the water sample. The high sensitivity of the Multi-Collector ICP-MS allows direct measurements at Pb concentration levels as low as 10 ng/mL and there is no effect of the matrix if the ion charge is lower than 400 mg/L [3]. With this method using multi-collection on Faraday cups, the TI correction technique is used for mass discrimination calculation. In such conditions, 2σ standard errors of 0.10% and 0.01% are achieved for $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$ ratios and $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ ratios respectively.

3. Results and comments

3.1. Major and trace elements

During the experiments, the evolution through time of the major and trace elements concentrations shows that major cations (Na, K, Mg and Ca) are released into solution during water/rock interaction, then these concentrations seem to reach stable values (Figure 1a). By contrast, anions concentrations are low in solution, with the exception of F (Figure 1b). Li and Pb are also released into solution during water/rock interaction, like Sr and U (Figure 1c). In addition, SiO₂, Al and Fe concentrations display evolution through time suggesting that in the first stage of water/rock interaction, these elements are released into solution, then due to secondary mineral formation incorporating these elements, the concentrations decrease.

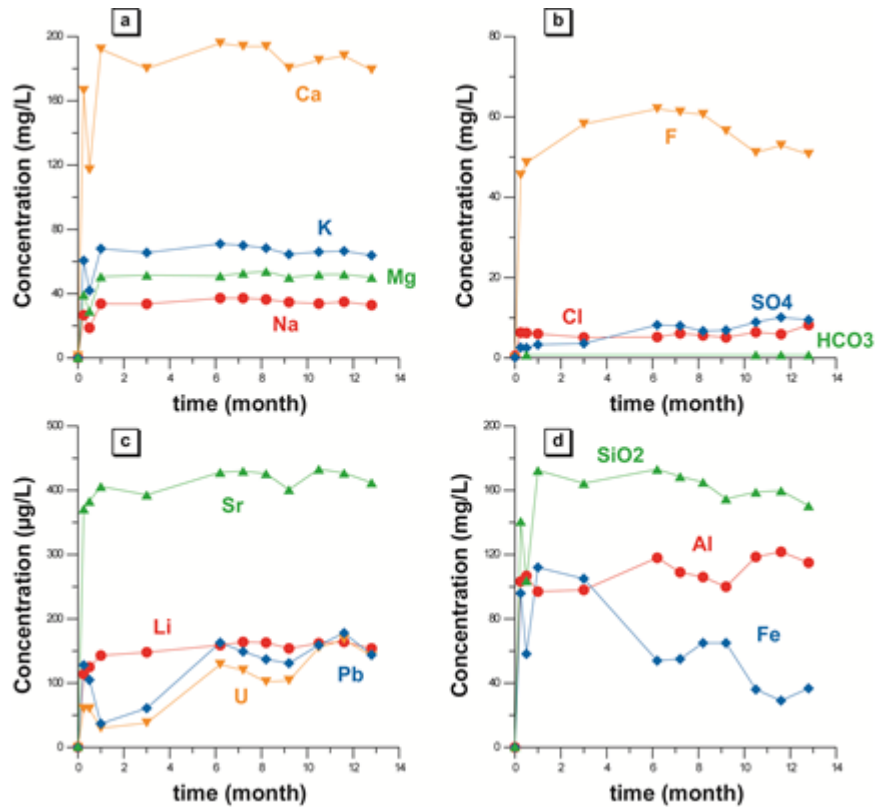


Fig. 1. Graphs showing the evolution over time of (a) cations concentrations (Na, K, Mg and Ca in mg/L); (b) anions concentrations (Cl, SO₄, HCO₃ and F in mg/L); (c) trace elements concentrations (Li, Pb, Sr and U in µg/L); (d) SiO₂, Al and Fe concentrations in mg/L.

3.2. Pb-Li isotopes tracing

Lead isotope tracing of these water/rock interactions shows that starting from the Pb isotope composition of the rain water solution (TMRAIN-95) having low $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios, the solution having interacted with the powdered granite shows a rapid increase of all isotope ratios through time (Figure 2a, b, c). After several weeks, the Pb isotopes ratios are very constant and do not vary through time radiogenic values are obtained for both $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios.

The evolution of Li and its isotopes ($\delta^7\text{Li}$) in solution (Figure 2d, e) as a function of time during water/rock interaction shows that Li concentration increases whereas Li isotopic signal decrease down to reach a constant value around $\sim +2\%$. This evolution suggests that first, dissolution processes are predominant and release Li into solution (consequently $\delta^7\text{Li}$ sharply decreases, whereas Li concentrations increase). This step is rather short. Then, during the second step, a steady state is reached between dissolution of granite, on the one hand, and precipitation of alteration minerals, on the other hand.

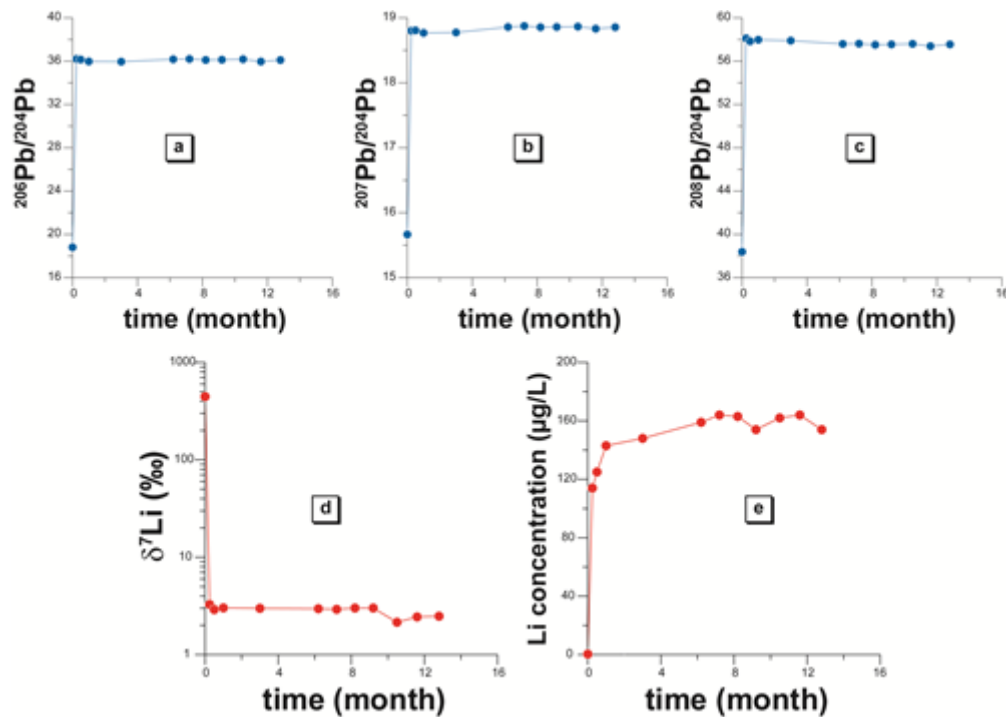


Fig. 2. Graphs showing the evolution over time of lead isotopes (a, b and c), lithium isotopes (d) and lithium concentrations (e).

4. Concluding remarks

Our data show that a radiogenic contribution of lead is observed during the experiments, in agreement with the field observations, and that the light lithium isotope (^6Li) is preferentially retained during uptake of Li into secondary minerals during weathering. If we compare the values obtained during our experiments with granite rock samples from the field site in India, the different minerals and the weathering model data [1]: we can observe that our experiments are in agreement with a major contribution of K-feldspar and plagioclase dissolution and to a lesser extent by biotite and apatite contribution. Further characterization of the secondary mineral phases after experiments is now needed in order to better constrain the minerals that can control Pb and Li isotopes during these experiments. The competition between dissolution and precipitation processes is likely to be the key factor controlling Pb and Li and their isotopes in these experiments.

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