



Assessing the role of climate on uranium and lithium isotope behaviour in rivers draining a basaltic terrain

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ABSTRACT

This study presents uranium (U) and lithium (Li) isotope and major and trace element data for the dissolved load, suspended particles and bedload from rivers draining dominantly basaltic catchments on the island of Sao Miguel in the Azores Archipelago. Uranium activity ratios are at secular equilibrium in the bedload, but are higher for suspended material, suggesting sorption of uranium from the solution onto particle surfaces. The ($^{234}\text{U}/^{238}\text{U}$) of the riverine dissolved phase varies between 1.02 and 1.86, and lies on a mixing trend between the values expected from chemical weathering of basalt (high [U]; ($^{234}\text{U}/^{238}\text{U}$) at secular equilibrium) and an endmember with low [U] and high ($^{234}\text{U}/^{238}\text{U}$), which probably reflects α -recoil effects associated with physical weathering and/or soil formation. Lithium isotope ratios are consistently lighter in suspended material than the corresponding bedload, suggesting preferential retention of ^6Li in the suspended load. In turn, the $\delta^7\text{Li}$ of the dissolved load is always isotopically heavier, ranging from 5.9 to 36.2‰, although the lightest values are affected by hydrothermal input. The lithium isotopic composition of the dissolved load reflects the balance of primary mineral dissolution to secondary mineral formation. Comparison of the behaviour of U and Li isotopes between Sao Miguel, Iceland, and other basaltic terrains suggests that whilst U isotopes are dominated by weathering regimes dependent on factors such as weathering rates, temperature, runoff and climate, neither weathering intensity nor climate exerts a direct influence on Li isotope behaviour; rather the formation of secondary minerals, which is indirectly controlled by climate, weathering, primary mineralogy and biology, appears to play the dominant role.

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1. Introduction

The chemical weathering of continental Ca–Mg silicates acts a major long-term mechanism for the consumption of atmospheric CO_2 , and therefore is a dominant natural climate control process (Walker et al., 1981; Berner et al., 1983; Berner, 2004). The chemical weathering of basalts, in particular, is thought to have a much greater effect on global CO_2 budgets than would be expected from the global extent of basaltic terrains (Gislason et al., 1996; Louvat and Allegre, 1998; Gislason et al., 2006; Louvat et al., 2008; Gislason et al., 2009). For example, estimates for present-day CO_2 consumption due to silicate weathering on land suggest that around 35% may be attributable to basaltic rocks, even though they constitute less than 5% of the continental area (Dessert et al., 2003).

However, as yet there are few unambiguous tracers of continental silicate weathering both at the present-day and in the past. Radiogenic isotopes such as strontium have been used to examine continental weathering processes (Palmer and Edmond, 1989, 1992; Allegre et al., 1996), but the interpretation of the Sr-isotope input to the oceans is not unequivocal, because variations in seawater $^{87}\text{Sr}/^{86}\text{Sr}$ which arise due to changes in the continental weathering flux can rarely be distinguished from those caused by variations in isotope composition, due to changes in lithology or weathering process (Palmer and Edmond, 1992; Taylor and Lasaga, 1999; Oliver et al., 2003; Bickle et al., 2005). Therefore tracers of chemical weathering processes which are independent of the composition of the continental source rocks are required. Uranium isotopes circumvent this problem by being independent of composition, because rocks older than ~ 1 Myr can be assumed to be in secular radioactive equilibrium, giving them an activity ratio equal to 1. Surface waters generally have a ($^{234}\text{U}/^{238}\text{U}$) activity ratio (where parentheses indicate activity ratio) greater than 1, because the energetic α -decay of ^{238}U to ^{234}U in rocks causes preferential enrichment of surrounding

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fluids in ^{234}U due to α -recoil (Kigoshi, 1971; Andersson et al., 1995; Ray et al., 1996; Riotte and Chabaux, 1999; Porcelli et al., 2001; Vigier et al., 2001; Chabaux et al., 2003; Riotte et al., 2003; Robinson et al., 2004; Swarzenski et al., 2004; Maher et al., 2006; Vigier et al., 2006; Andersen et al., 2009; Bourdon et al., 2009). Thus mechanical (physical) weathering and soil formation will raise the ($^{234}\text{U}/^{238}\text{U}$) activity ratio of surface waters, due to an increase in the surface area to grain size ratio (e.g. in glacial tills or soil solutions (Kronfeld and Vogel, 1991; Robinson et al., 2004; Pogge von Strandmann et al., 2006a; Andersen et al., 2009). Conversely, congruent chemical weathering of silicate rocks will lower the activity ratio of fluids towards secular equilibrium. The U activity ratio in surface waters can thus provide information on the balance of physical to chemical weathering.

The stable isotopes of lithium (^6Li and ^7Li) also provide information on chemical weathering processes (Huh et al., 1998, 2001, 2004). Congruent dissolution of rocks is thought to cause negligible isotope fractionation; however, secondary minerals preferentially incorporate ^6Li , driving the isotope composition of the residual surface waters to heavier values (Huh et al., 1998, 2001; Kisakurek et al., 2004). Studies have also shown that the Li isotope budget of rivers is dominantly determined by silicate rocks, even in carbonate-dominated catchments (Kisakurek et al., 2005). Thus the Li isotope ratio of rivers provides information on the balance of primary silicate rock dissolution to secondary mineral formation (Huh et al., 2001; Kisakurek et al., 2005; Pogge von Strandmann et al., 2006a). The degree of isotope fractionation also depends on the mineralogy of clay minerals undergoing formation, e.g. little or no Li isotope fractionation accompanies physisorption onto smectite, but isotopic fractionation can be significant during chemisorption onto kaolinite and gibbsite (Zhang et al., 1998; Pistiner and Henderson, 2003; Vigier et al., 2008).

The Azores archipelago provides the opportunity to study the effects of weathering and erosion of largely monolithological basalts in a temperate climatic setting with relatively high rainfall. In terms of weathering rates, the Azores have virtually identical chemical weathering rates to the Amazon and Iceland (Gaillardet et al., 1999; Louvat et al., 2008) (although physical weathering rates are lower), which bears out the significant influence that small basaltic islands are thought to have on weathering and erosion on a global scale (Milliman and Syvitski, 1992).

This study presents U and Li isotope data for the dissolved and suspended load of rivers draining dominantly basaltic catchments on the island of Sao Miguel in the Azores. These results provide insights into the elemental and isotope behaviour of U and Li in a temperate

climate, and allow comparison with their behaviour in contrasting climatic and hydrologic regimes in Iceland and elsewhere.

2. Geology, climate and river setting

The Azores archipelago is a group of volcanic islands, situated ~1500 km due west of Lisbon (Portugal) in the middle of the Atlantic Ocean. The islands are distributed in a chain which extends for over 620 km in a WNW–ESE direction. The total land area of the archipelago is 2247 km², of which 760 km² are made up by the largest island of Sao Miguel, which is located at the eastern end of the island chain. The islands are situated on the Azores plateau. This region is thought to be the location of the triple junction between the African, Eurasian and North American plates. The plateau was formed by still-active hot-spot volcanism and is mainly composed of alkali basalts (White et al., 1979).

Exposed rock ages in Sao Miguel range from the ~4 Ma Nordeste volcanic complex in the north-east of the island to historical ages in the centre (Fig. 1). Generally, the eastern and western ends of the island are the oldest, with the central Pico and Agua de Pau being more recent (Moore, 1990; Cruz et al., 1999; Guest et al., 1999).

The climate of the Azores archipelago is maritime and temperate. Mean temperatures range from between 13 °C in January and 23 °C in August. Mean annual precipitation increases westwards across the archipelago, with 700 mm rainfall per year on Sao Miguel (Instituto de Meteorologia (Portuguese Meteorological Office)). The island is densely vegetated – generally by cloud forest above the 500 m contour line and by agriculture closer to the coast.

The only permanent rivers are in the oldest eastern part of the island and around the calderas, which have often formed lakes. The recent volcanic soils in the west and centre of the island drain too rapidly to carry permanent streams. The rivers are generally located in steep, narrow gorges. The lack of wide valleys, meandering streams and flood plains points to the relative immaturity of the island's geomorphology and to the ease with which the basalt can be eroded.

The mean physical erosion rate of rivers on Sao Miguel has been estimated to be between 170 and 500 t/km²/yr (Louvat and Allegre, 1998), which is almost up to three times the world average for physical denudation (Meybeck, 1988). The mean chemical erosion rate for Sao Miguel is 26–50 t/km²/yr (Louvat and Allegre, 1998), up to twice that of the global average (Gaillardet et al., 1999).

In this study, rivers have been largely sampled from the older eastern part of the island – both from Nordeste, and the slightly

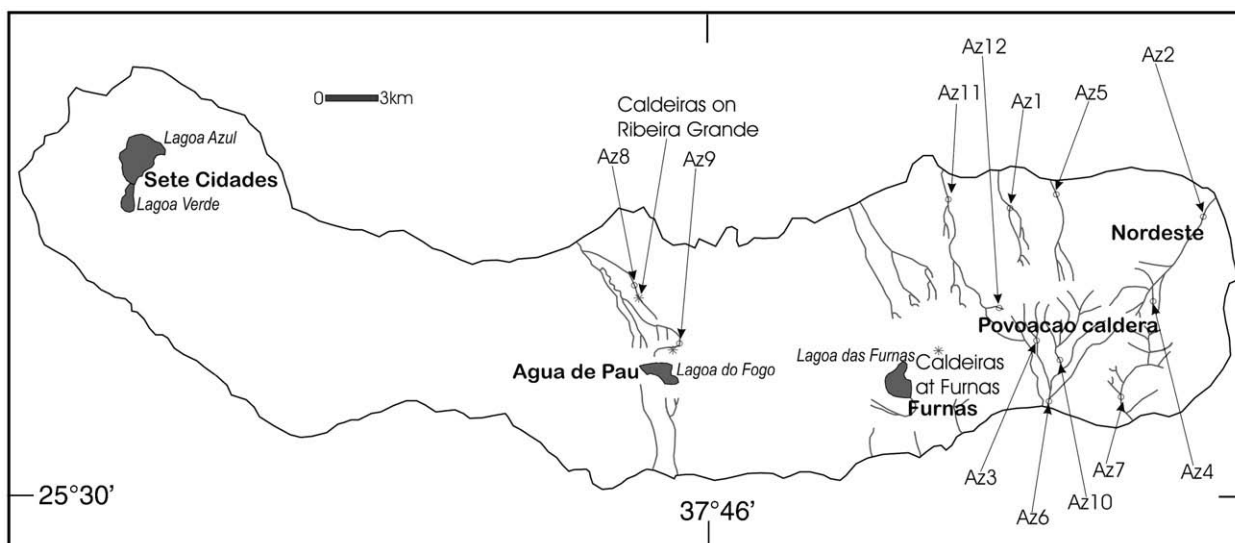


Fig. 1. Map of sample locations.

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