

## Rapid response of silicate weathering rates to climate change in the Himalaya

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### Abstract

Chemical weathering of continental rocks plays a central role in regulating the carbon cycle and the Earth's climate (Walker *et al.*, 1981; Berner *et al.*, 1983), accounting for nearly half the consumption of atmospheric carbon dioxide globally (Beaulieu *et al.*, 2012). However, the role of climate variability on chemical weathering is still strongly debated. Here we focus on the Himalayan range and use the lithium isotopic composition of clays in fluvial terraces to show a tight coupling between climate change and chemical weathering over the past 40 ka. Between 25 and 10 ka ago, weathering rates decrease despite temperature increase and monsoon intensification. This suggests that at this timescale, temperature plays a secondary role compared to runoff and physical erosion, which inhibit chemical weathering by accelerating sediment transport and act as fundamental controls in determining the feedback between chemical weathering and atmospheric carbon dioxide.

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### Letter

It has long been recognised that the dissolution of minerals, chemical weathering, can act as a long-term (>1 Ma) feedback on the Earth's climate (Walker *et al.*, 1981; Berner *et al.*, 1983; Donnadieu *et al.*, 2004). More recently, modelling studies have shown that the weathering engine can respond extremely rapidly

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(<100 yr) to climate change and may act as a key component in the atmospheric CO<sub>2</sub> level short-term regulation (Beaulieu *et al.*, 2012). However, the relative role of climatic parameters on chemical weathering is still debated. Model simulations and geochemical studies of river-born material highlight either a temperature dependence (Walker *et al.*, 1981), where warming promotes chemical weathering, or a predominance of other parameters like mechanical erosion (Raymo and Ruddiman, 1992; Gaillardet *et al.*, 1999; Riebe *et al.*, 2001; Donnadieu *et al.*, 2004) and vegetation (Bayon *et al.*, 2012). To address this question, another possible approach consists in investigating past continental environments through the study of marine or floodplain deposits. However, because in large river systems sediment transport and storage operates over >10<sup>4</sup> yr timescales (Granet *et al.*, 2010), a significant time lag may exist between the time when sediments acquired their geochemical characteristics, reflecting palaeoenvironmental conditions, and their final deposition.

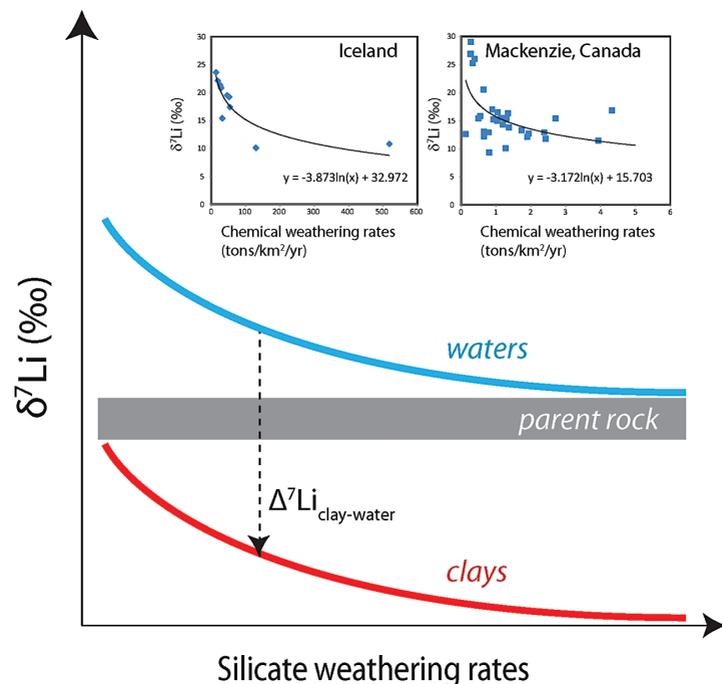
Here we use the lithium (Li) isotopic composition of clays from sedimentary records in Himalayan basins to determine how chemical weathering intensity has varied over the past 40 ka and particularly since the Last Glacial Maximum (locally older than 24 ka; Owen *et al.*, 2002). In order to minimise the time lag between source and deposit locations, we have focused on alluvial deposits located in the headwater areas of the Ganges and Yamuna Rivers.

While little isotope fractionation occurs during mineral dissolution clay formation induces strong fractionations at low temperature, whereby <sup>6</sup>Li is preferentially incorporated into clays compared to <sup>7</sup>Li, resulting in a strong enrichment of <sup>7</sup>Li in waters (Burton and Vigier, 2011). Thus, it has been shown that the  $\delta^7\text{Li}$  composition of natural waters can be used as a proxy for chemical weathering rates at the catchment scale, since the heaviest  $\delta^7\text{Li}$  compositions in water are associated with the areas characterised by low catchment-wide silicate weathering rates (Fig. 1). Several studies have also shown that the  $\delta^7\text{Li}$  composition of solid weathering products (*i.e.* soils, river sediments) is sensitive to chemical weathering conditions, where lower  $\delta^7\text{Li}$  values reflect more intensive leaching (Burton and Vigier, 2011).

To evaluate the reliability of the records studied, we investigated three different regions of the Himalaya and its piedmont in India: the upper Yamuna River basin, the Alaknanda River basin and the Donga Fan (Fig. S-1). Depositional ages for the alluvial deposits reported here were previously constrained by optically-stimulated luminescence (OSL) dating and range from 9 to 41 ka (Singh *et al.*, 2001; Ray and Srivastava, 2010). Both bulk sediments and clay-sized fractions were analysed for Li isotopes. In bulk sediments, mineralogical abundances and Sr isotopes were also measured (see Supplementary Information).

The  $\delta^7\text{Li}$  compositions of bulk sediments vary between -1.54 and +1.98 ‰, while clay-sized fractions show a much broader range. Most samples show low  $\delta^7\text{Li}$  values best explained by significant <sup>6</sup>Li enrichment during clay formation (Burton and Vigier, 2011). The  $\delta^7\text{Li}$  values in bulk samples and clay-sized fractions evolve consistently as a function of time: both decrease between 35 and 25 ka,

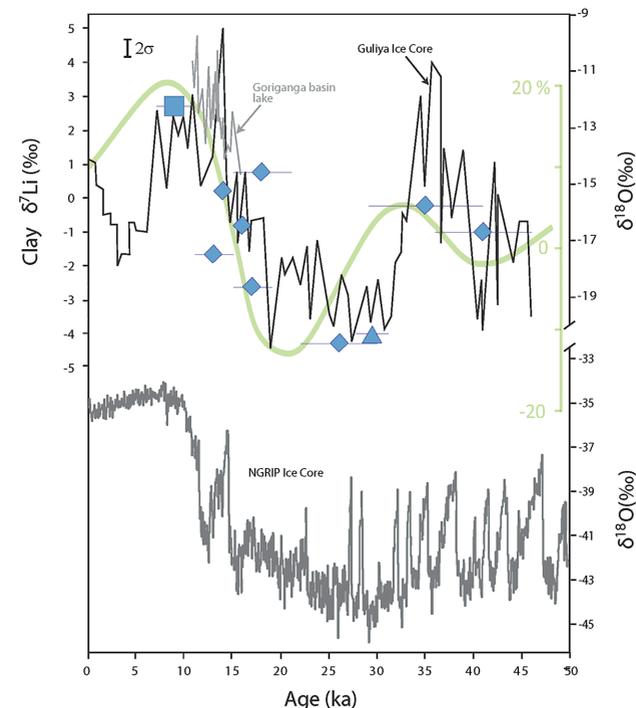




**Figure 1** Relationship between  $\delta^7\text{Li}$  values of waters and clay minerals, and silicate weathering rates - estimated independently ( $\delta^7\text{Li} = [({}^7\text{Li}/{}^6\text{Li}) / ({}^7\text{Li}/{}^6\text{Li})_{\text{L-SVEC}} - 1] \times 1000$ ) where L-SVEC is the standard). Most published studies highlight an inverse correlation between water  $\delta^7\text{Li}$  and weathering rates at the watershed scale. For a given Li isotope fractionation between clay and water ( $\Delta^7\text{Li}_{\text{clay-water}}$ ), clay  $\delta^7\text{Li}$  is expected to follow the same pattern with silicate weathering rates. Thus, low  $\delta^7\text{Li}$  values are indicative of fast weathering rates. Insets show data for river waters from the Mackenzie basin in Canada (Millot *et al.*, 2010) and from the basaltic basins of Iceland (Vigier *et al.*, 2009). The curves in the insets show a logarithmic regression through the data.

then increase between 25 and 10 ka (Fig. 2 and Fig. S-4). Several hypotheses can potentially account for the observed  $\delta^7\text{Li}$  variations: (i) change of sediment sources, (ii) changes in chemical weathering conditions (prior to deposition) and (iii) post-depositional alteration within the terraces.

Change of sediment sources is unlikely to control the  $\delta^7\text{Li}$  composition of clay-sized fractions because (i) the mineralogy of the clay-sized fraction is dominated by secondary clays that account for most of the Li budget; (ii) most igneous, metamorphic and sedimentary rocks have  $\delta^7\text{Li} \geq 0$  ‰ (Table S-3 and references therein). Thus, clay  $\delta^7\text{Li}$  compositions as low as -4 ‰ between 30 and 25 ka are significantly lower than both the average values for unweathered continental rocks and published values for Himalayan river bedload sediments (Kisakúrek *et al.*, 2005).



**Figure 2** Clay  $\delta^7\text{Li}$  as a function of terrace deposition ages (square: Yamuna, triangle: Donga Fan and diamonds: Alaknanda). The vertical bracket in the top left corner shows the external uncertainty for  $\delta^7\text{Li}$ . The light green curve represents the precipitation changes in the SW Indian Monsoon for South Asia (green axis graduated from -20 to 20 %; Sanyal and Sinha, 2010 and references therein). The black curve represents the  $\delta^{18}\text{O}_{\text{SMOW}}$  record from the Guliya ice core on the Qinghai-Tibetan Plateau (right y-axis) (Thompson *et al.*, 1997). The grey curve is the  $\delta^{18}\text{O}_{\text{VPDB}}$  record from lacustrine sediments in the Goriganga basin (right y-axis), 100 km east of the study area (Beukema *et al.*, 2011). Records from the Guliya ice core and the lacustrine sediments from the Goriganga basin show that relative changes in climatic conditions were consistent on both sides of the divide, at least for this period. Comparison with a global palaeo-climatic record is illustrated with the grey curve, showing the NGRIP ice core  $\delta^{18}\text{O}$  record from Greenland for the same period (North Greenland Ice Core Project members, 2004).

After deposition, weathering of alluvial sediments could bias the Li isotope composition of sediments and clays. However, several lines of evidence argue against this: (i) post-depositional alteration is expected to mainly affect the “exchangeable” Li. Experimental work has shown that Li is quickly incorporated into clay octahedral sites. It then remains into these sites even after intensive (hydrothermal) treatment (Vigier *et al.*, 2008). As a consequence, low-temperature water percolating through deposited sediment will principally, and quickly, react with exchangeable cations but not with  $\text{Li}^+$  ions in octahedral sites. Thus, to

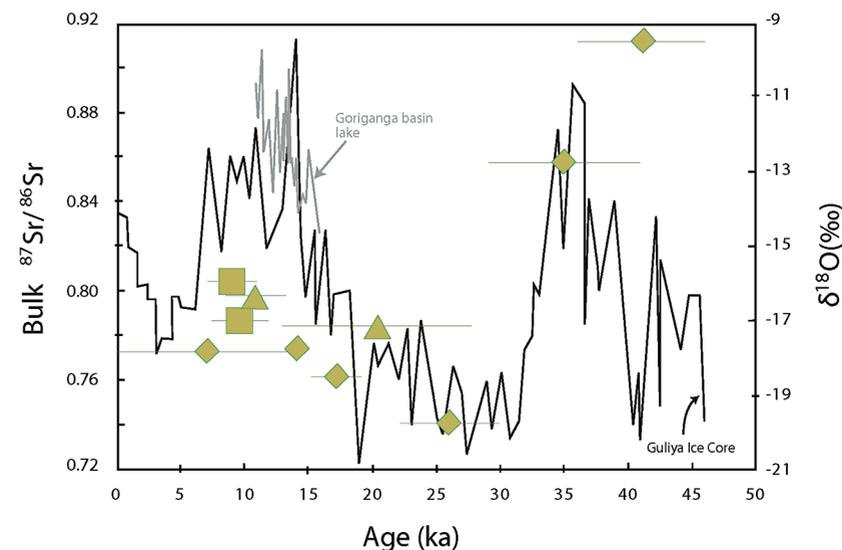


ensure that measured Li isotope compositions were not overprinted by any Li water-clay exchange during post-depositional alteration, the exchangeable Li was systematically removed during sample preparation (Supplementary Information). (ii) There is no simple relationship between  $\delta^7\text{Li}$  compositions and sampling depth (Fig. S-5). In fact, superficial alteration was carefully avoided by collecting samples located at significant depths (9 m on average), whereas soil development (if any) was restricted to the upper 2 m. Furthermore, all selected samples derive from undisturbed stratigraphic sections, with no sign of post-depositional alteration (Supplementary Information). (iii) If post-depositional alteration was significant,  $\delta^7\text{Li}$  would be expected to correlate with depositional age. However, this is not observed as the oldest deposits (37–41 ka) display  $\delta^7\text{Li}$  values that are similar to more recent sediments (Table S-1). Alternatively, the lack of relationship with depositional age could reflect complex lateral fluid flow. However, this water-sediment interaction would also only affect the exchangeable Li, which was removed as indicated above. (iv) Several samples with similar depositional ages but from different regions show consistent  $\delta^7\text{Li}$  values as a function of age (Table S-1).

As a first approximation, since Li isotope fractionation during clay formation is temperature-dependent (Vigier *et al.*, 2008),  $\delta^7\text{Li}$  variations in clays could potentially reflect mean temperature change since the Last Glacial Maximum (LGM). Warming since the LGM has been estimated between 4 and 7 °C (Farrera *et al.*, 1999). Experimental data indicate that this temperature increase could induce an increase in  $\delta^7\text{Li}$  between 0.5 and 0.9 ‰ in the solid (Vigier *et al.*, 2008). This is well below the extent of the increase in  $\delta^7\text{Li}$  observed in clay-sized fractions (of ~7 ‰). Thus, temperature variations alone cannot account for the observed range in  $\delta^7\text{Li}$  values.

Several studies have shown that the  $\delta^7\text{Li}$  values measured in river waters and soils decrease with increasing chemical weathering rates at the scale of the watershed or the soil profile (Fig. 1; Kisakúrek *et al.*, 2005; Vigier *et al.*, 2009; Millot *et al.*, 2010). In addition, the low  $\delta^7\text{Li}$  values of clay-sized fractions support an enrichment in  $^6\text{Li}$  in secondary phases during chemical weathering (Kisakúrek *et al.*, 2005). Consequently, variations in  $\delta^7\text{Li}$  are best explained by changes in weathering conditions over the past 40 ka. In this case, the decrease towards low  $\delta^7\text{Li}$  compositions between 35 and 25 ka indicates an increase in weathering rates over this period of time. The following increase in  $\delta^7\text{Li}$  between 25 and 10 ka then suggests that weathering rates have decreased since the end of the LGM. An intimate link between climate and weathering in this region appears when comparing clay  $\delta^7\text{Li}$  and the oxygen isotope compositions recorded by the Guliya ice core from the Qinghai-Tibetan Plateau (Fig. 2; Thompson *et al.*, 1997). This oxygen isotope record is consistent with an intensification of the monsoon between 25 and 10 ka ago, as suggested by most studies (Goodbred and Kuehl, 2000; Sanyal and Sinha, 2010; Beukema *et al.*, 2011). Consequently, the co-variation between  $\delta^7\text{Li}$  and  $\delta^{18}\text{O}$  values indicates a synchronicity between changes in chemical weathering rates and monsoon intensity.

The strontium (Sr) isotope composition of bulk sediments also shows systematic variations with time, similar to that of  $\delta^7\text{Li}$  (Fig. 3). Strontium isotopes have been used to study changes in the provenance of sediments, on the basis of different  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in the source rocks found in the catchment (Galy and France-Lanord, 1999). Average  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios for the Higher Himalaya Crystallines (HHC) and the Lesser Himalayas (LH) are significantly different:  $0.76 \pm 0.03$  and  $0.85 \pm 0.09$ , respectively (Rahaman *et al.*, 2009). Thus, the observed increase in  $^{87}\text{Sr}/^{86}\text{Sr}$  between 25 and 10 ka could be interpreted as an increasing contribution of sediments from the LH. However, as the monsoon intensified between 25 and 10 ka ago (Sanyal and Sinha, 2010), moisture penetrated further north, promoting erosion in the HHC, as demonstrated for the Sutlej River (Bookhagen *et al.*, 2005). More sediments from the HHC would have resulted in a decrease of sediment  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, at odds with the data presented here (Fig. 3). Furthermore, if provenance controlled the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio, sediments of the Donga Fan should show high values (because they only drain the LH) and the Sr isotopic composition of the Alaknanda River sediments should increase downstream (as more sediments are derived from the LH). However, our data show the opposite trend (Fig. S-6).



**Figure 3**  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of bulk sediments as a function of terrace deposition ages (same symbols as in Fig. 2). The error on  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios is smaller than the symbol size. The black curve represents the  $\delta^{18}\text{O}_{\text{SMOW}}$  record from the Guliya ice core on the Qinghai-Tibetan Plateau (right y-axis) (Thompson *et al.*, 1997). The grey curve is the  $\delta^{18}\text{O}_{\text{VPDB}}$  record from lacustrine sediments in the Goriganga basin (right y-axis) (Beukema *et al.*, 2011).



Alternatively, variations in Sr isotopes can reflect changes in weathering conditions since minerals with different susceptibility to weathering display different  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (Blum *et al.*, 1993; Colin *et al.*, 1999). For instance, Blum *et al.* (1993) have highlighted a significant dissolution of high  $^{87}\text{Sr}/^{86}\text{Sr}$  phases in glaciated areas. Bulk sediments from the three regions studied show a correlation between their  $^{87}\text{Sr}/^{86}\text{Sr}$  and mineralogical content, such as micas and orthoclase (Fig. S-7). This suggests that the relative dissolution of primary minerals controls the  $^{87}\text{Sr}/^{86}\text{Sr}$  of the bulk sediment, rather than sediment provenance. Thus, the minimum in  $^{87}\text{Sr}/^{86}\text{Sr}$  values associated with the lowest  $\delta^7\text{Li}$  values at 25 ka is consistent with more intense weathering conditions, when  $\delta^{18}\text{O}$  values of local and NGRIP ice cores both indicate cold conditions.

Taken together, Li and Sr isotope systematics suggest that chemical weathering conditions in the Himalayan range have been significantly affected by climatic variations between 40 and 10 ka. Moreover, synchronous variations (within errors) between  $\delta^7\text{Li}$  and  $\delta^{18}\text{O}$  values suggest that the response of transport and storage of weathering products to climate change was rapid (< a few thousand years). Our data indicate that chemical weathering rates significantly decreased between 25 and 10 ka. It is possible to estimate the corresponding magnitude of changes in silicate weathering rates using a constant clay-water fractionation: an increase of clay  $\delta^7\text{Li}$  from -4 ‰ to +3 ‰ corresponds to an increase of water  $\delta^7\text{Li}$  of 7 ‰ for the considered period. As a first approximation, using the relationship between water  $\delta^7\text{Li}$  and weathering rates (Vigier *et al.*, 2009; Millot *et al.*, 2010), this change in  $\delta^7\text{Li}_{\text{water}}$  would translate to a decrease in silicate weathering rates by an order of magnitude (using the regression for the Mackenzie data, Fig. 1 and  $\Delta^7\text{Li}_{\text{clay-water}} = -17$  ‰; Vigier *et al.*, 2008).

This decrease in chemical weathering rates coeval with global warming since the end of the LGM runs counter to the expectation that chemical weathering is promoted by warmer and wetter conditions (Walker *et al.*, 1981). This result highlights runoff and physical erosion as major controls on chemical weathering in the Himalayan region for the last 25 ka, climate warming playing a secondary role at this timescale. It has been proposed that a significant increase in erosion rates accompanied monsoon intensification in the Himalaya since the end of the LGM, mainly via increased runoff (Bookhagen *et al.*, 2005; Clift *et al.*, 2008) and its effect on landsliding and fluvial incision. The resulting increased sediment transport is likely to have reduced the average residence time of soils and sediments within the basins, which in turn would have limited chemical weathering. In contrast, at the LGM, the observation of more intense weathering could be explained by the high surface area of the regolith produced by glacial erosion, promoting mineral dissolution despite colder conditions.

Recently, Beaulieu *et al.* (2012) have shown that at the decadal/centennial scale, the chemical weathering response, dominated by carbonate weathering, is sensitive to both runoff and temperature. Distinguishing between the two factors can be challenging, since they are often linked to each other (Labat *et al.*, 2004).

Here, our results show that at the millennial scale, silicate chemical weathering rates in the Himalayan range are mainly driven by runoff and physical erosion, while temperature plays a secondary role.

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## Additional Information

**Supplementary Information** accompanies this letter at [www.geochemicalperspectivesletters.org/article1502](http://www.geochemicalperspectivesletters.org/article1502)

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