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

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Abstract

doi: 10.7185/geochemlet.1502

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.

Received 9 December 2014 | Accepted 23 January 2015 | Published 20 February 2015

Letter

10

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_2 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 >10⁴ 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 ⁶Li is preferentially incorporated into clays compared to ⁷Li, resulting in a strong enrichment of ⁷Li in waters (Burton and Vigier, 2011). Thus, it has been shown that the δ^7 Li composition of natural waters can be used as a proxy for chemical weathering rates at the catchment scale, since the heaviest δ^7 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 δ^7 Li composition of solid weathering products (*i.e.* soils, river sediments) is sensitive to chemical weathering conditions, where lower δ^7 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 δ^7 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 δ^7 Li values best explained by significant ⁶Li enrichment during clay formation (Burton and Vigier, 2011). The δ^7 Li values in bulk samples and clay-sized fractions evolve consistently as a function of time: both decrease between 35 and 25 ka,



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δ¹⁸O(‰)

¹⁸O(‰)

50

Guliva Ice Core



Silicate weathering rates

Figure 1 Relationship between $\delta^7 \text{Li}$ values of waters and clay minerals, and silicate weathering rates - estimated independently ($\delta^7 \text{Li} = [(^7 \text{Li}/^6 \text{Li}) / (^7 \text{Li}/^6 \text{Li})_{L:SVEC} - 1]x1000$) 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 ($\Delta^7 \text{Li}_{ay-water}$), 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 δ^7 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 δ^7 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 δ^7 Li ≥ 0 ‰ (Table S-3 and references therein). Thus, clay δ^7 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 (Kisakűrek *et al.*, 2005).



Figure 2 Clay δ^7 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 δ^7 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 $\delta^{18}O_{SMOW}$ record from the Guliya ice core on the Qinghai-Tibetan Plateau (right y-axis) (Thompson et *al.*, 1997). The grey curve is the $\delta^{18}O_{VPDB}$ 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 δ^{18} 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 Li⁺ ions in octahedral sites. Thus, to



Letter

I 2σ

3

-2

-3

-5

Clay 8⁷Li (‰)

13

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 δ^7 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, δ ⁷Li would be expected to correlate with depositional age. However, this is not observed as the oldest deposits (37-41 ka) display δ^7 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 δ^7 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), δ^7 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 δ^7 Li between 0.5 and 0.9 ‰ in the solid (Vigier *et al.*, 2008). This is well below the extent of the increase in δ^7 Li observed in clay-sized fractions (of ~7 ‰). Thus, temperature variations alone cannot account for the observed range in δ^7 Li values.

Several studies have shown that the δ^7 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; Kisakűrek et al., 2005; Vigier et al., 2009; Millot *et al.*, 2010). In addition, the low δ^7 Li values of clav-sized fractions support an enrichment in ⁶Li in secondary phases during chemical weathering (Kisakűrek *et al.*, 2005). Consequently, variations in δ^7 Li are best explained by changes in weathering conditions over the past 40 ka. In this case, the decrease towards low δ^7 Li compositions between 35 and 25 ka indicates an increase in weathering rates over this period of time. The following increase in δ^7 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 δ^7 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; Sanval and Sinha, 2010; Beukema et al., 2011). Consequently, the co-variation between δ^7 Li and δ^{18} O values indicates a synchronicity between changes in chemical weathering rates and monsoon intensity.

14



Letter

The strontium (Sr) isotope composition of bulk sediments also shows systematic variations with time, similar to that of δ^7 Li (Fig. 3). Strontium isotopes have been used to study changes in the provenance of sediments, on the basis of different ⁸⁷Sr/⁸⁶Sr ratios in the source rocks found in the catchment (Galy and France-Lanord, 1999). Average ⁸⁷Sr/⁸⁶Sr ratios for the Higher Himalaya Crystallines (HHC) and the Lesser Himalayas (LH) are significantly different: 0.76 ± 0.03 and 0.85 ± 0.09 , respectively (Rahaman *et al.*, 2009). Thus, the observed increase in ⁸⁷Sr/⁸⁶Sr 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 ⁸⁷Sr/⁸⁶Sr ratios, at odds with the data presented here (Fig. 3). Furthermore, if provenance controlled the ⁸⁷Sr/⁸⁶Sr 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 ⁸⁷Sr/⁸⁶Sr ratios of bulk sediments as a function of terrace deposition ages (same symbols as in Fig. 2). The error on ⁸⁷Sr/⁸⁶Sr ratios is smaller than the symbol size. The black curve represents the $\delta^{18}O_{SMOW}$ record from the Guliya ice core on the Qinghai-Tibetan Plateau (right y-axis) (Thompson *et al.*, 1997). The grey curve is the $\delta^{18}O_{VPDB}$ 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 ⁸⁷Sr/⁸⁶Sr ratios (Blum *et al.*, 1993; Colin *et al.*, 1999). For instance, Blum *et al.* (1993) have highlighted a significant dissolution of high ⁸⁷Sr/⁸⁶Sr phases in glaciated areas. Bulk sediments from the three regions studied show a correlation between their ⁸⁷Sr/⁸⁶Sr and mineralogical content, such as micas and orthoclase (Fig. S-7). This suggests that the relative dissolution of primary minerals controls the ⁸⁷Sr/⁸⁶Sr of the bulk sediment, rather than sediment provenance. Thus, the minimum in ⁸⁷Sr/⁸⁶Sr values associated with the lowest δ^7 Li values at 25 ka is consistent with more intense weathering conditions, when δ^{18} 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 δ^7 Li and δ^{18} 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 δ^7 Li from -4 ‰ to +3 ‰ corresponds to an increase of water δ^7 Li of 7 ‰ for the considered period. As a first approximation, using the relationship between water δ^7 Li and weathering rates (Vigier *et al.*, 2009; Millot *et al.*, 2010), this change in δ^7 Li_{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 Δ^7 Li_{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.

Acknowledgements

Letter

We would like to thank Yogesh Ray, P.O. Suresh, David Fink, Paul Hesse, Jan-Hendrik May and Oliver Korup for their assistance in the field. The Director of the Wadia Institute of Himalayan Geology is acknowledged for extending logistic support. Greg Ravizza, Ken Rubin, Doug Pyle, Chris Russo and Denys Vonderhaar at University of Hawai'i at Manoa, Aimeryc Schumacher at CRPG, and Paul Carr, Brian Jones, Venera Espanon, Jose Abrantes at University of Wollongong are thanked for their assistance with the analytical work. We would also like to thank John D. Jansen, Pete Burnard, Helen McGregor, Jan-Hendrik May, Tim Cohen, Allen Nutman and Colin Murray-Wallace for their comments on the manuscript. Finally, we would like to acknowledge Christian France-Lanord, Albert Galy, Marteen Lupker, Doug Burbank, François Chabaux, Cliff Riebe, Josh West and Niels Hovius for helpful discussions. A. Dosseto acknowledges an Australian Research Council Future Fellowship FT0990447 and a University of Wollongong URC Small Grant.

Editor: Eric H. Oelkers

Additional Information

Supplementary Information accompanies this letter at www.geochemicalperspectivesletters.org/article1502

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Cite this letter as: Dosseto, A., Vigier, N., Joannes-Boyau, R., Moffat, I., Singh, T., Srivastava, P. (2015) Rapid response of silicate weathering rates to climate change in the Himalaya. *Geochem. Persp. Let.* 1, 10-19.

References

- BAYON, G., DENNIELOU, B., ETOUBLEAU, J., PONZEVERA, E., TOUCANNE, S., BERMELL, S. (2012) Intensifying Weathering and Land Use in Iron Age Central Africa. *Science* 335, 1219-1222.
- BEAULIEU, E., GODDERIS, Y., DONNADIEU, Y., LABAT, D., ROELANDT, C. (2012) High sensitivity of the continental-weathering carbon dioxide sink to future climate change. *Nature Climate Change* 2, 346-349.



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- BERNER, R.A., LASSAGA, A.C., GARRELS, R.M. (1983) The carbone-silicate geochemical cycle and its effect on atmospheric carbon dioxide over the past 100 million years. *American Journal of Science* 283, 641-683.
- BEUKEMA, S.P., KRISHNAMURTHY, R.V., JUYAL, N., BASAVAIAH, N., SINGHVI, A.K. (2011) Monsoon variability and chemical weathering during the late Pleistocene in the Goriganga basin, higher central Himalaya, India. *Quaternary Research* 75, 597-604.
- BLUM, J.D., EREL, Y., BROWN, K. (1993) ⁸⁷Sr/⁸⁶Sr ratios of Sierra Nevada stream waters: Implications for relative mineral weathering rates. *Geochimica et Cosmochimica Acta* 57, 5019-5025.
- BOOKHAGEN, B., THIEDE, R.C., STRECKER, M.R. (2005) Late Quaternary intensified monsoon phases control landscape evolution in the northwest Himalaya. *Geology* 33, 149-152.
- BURTON, K.W., VIGIER, N. (2011) Lithium Isotopes as Tracers in Marine and Terrestrial Environments. In: Baskaran, M. (Eds.) *Handbook of Environmental Isotope Geochemistry*. Springer Berlin Heidelberg, 41-59.
- CLIFT, P.D., GIOSAN, L., BIUSZTAJN, J., CAMPBELL, I.H., ALLEN, C., PRINGLE, M., TABREZ, A.R., DANISH, M., RABBANI, M.M., ALIZAI, A., CARTER, A., LUECKGE, A. (2008) Holocene erosion of the Lesser Himalaya triggered by intensified summer monsoon. *Geology* 36, 79-82.
- COLIN, C., TURPIN, L., BERTAUX, J., DESPRAIRIES, A., KISSEL, C. (1999) Erosional history of the Himalayan and Burman ranges during the last two glacial-interglacial cycles. *Earth and Planetary Science Letters* 171, 647-660.
- DONNADIEU, Y., GODDERIS, Y., RAMSTEIN, G., NEDELEC, A., MEERT, J. (2004) A 'snowball Earth' climate triggered by continental break-up through changes in runoff. *Nature* 428, 303-306.
- FARRERA, I., HARRISON, S.P., PRENTICE, I.C., RAMSTEIN, G., GUIOT, J., BARTLEIN, P.J., BONNE-FILLE, R., BUSH, M., CRAMER, W., VON GRAFENSTEIN, U., HOLMGREN, K., HOOGHIEM-STRA, H., HOPE, G., JOLLY, D., LAURITZEN, S.E., ONO, Y., PINOT, S., STUTE, M., YU, G. (1999) Tropical climates at the Last Glacial Maximum: a new synthesis of terrestrial palaeoclimate data. I. Vegetation, lake-levels and geochemistry. *Climate Dynamics* 15, 823-856.
- GAILLARDET, J., DUPRE, B., ALLÈGRE, C.J. (1999) Geochemistry of large river suspended sediments: Silicate weathering or recycling tracer? *Geochimica et Cosmochimica Acta* 63, 4037-4051.
- GALY, A., FRANCE-LANORD, C. (1999) Weathering processes in the Ganges–Brahmaputra basin and the riverine alkalinity budget. *Chemical Geology* 159, 31-60.
- GOODBRED, S.J., KUEHL, S.A. (2000) Enormous Ganges-Brahmaputra sediment dischange during strengthened early Holocene monsoon. *Geology* 28, 1083-1086.
- GRANET, M., CHABAUX, F., STILLE, P., DOSSETO, A., FRANCE-LANORD, C., BLAES, E. (2010) U-series disequilibria in suspended river sediments and implication for sediment transfer time in alluvial plains: The case of the Himalayan rivers. *Geochimica et Cosmochimica Acta* 74, 2851-2865.
- KISAKŰREK, B., JAMES, R.H., HARRIS, N.B.W. (2005) Li and & Li in Himalayan rivers: Proxies for silicate weathering? *Earth and Planetary Science Letters* 237, 387-401.
- LABAT, D., GODDÉRIS, Y., PROBST, J.L., GUYOT, J.L. (2004) Evidence for global runoff increase related to climate warming. *Advances in Water Resources* 27, 631-642.
- MILLOT, R., VIGIER, N., GAILLARDET, J. (2010) Behaviour of lithium and its isotopes during weathering in the Mackenzie Basin, Canada. *Geochimica et Cosmochimica Acta* 74, 3897-3912.
- NORTH GREENLAND ICE CORE PROJECT MEMBERS (2004) High-resolution record of Northern Hemisphere climate extending into the last interglacial period. *Nature* 431, 147-151.
- OWEN, L.A., FINKEL, R.C., CAFFEE, M.W. (2002) A note on the extent of glaciation throughout the Himalaya during the global Last Glacial Maximum. *Quaternary Science Reviews* 21, 147-157.
- RAHAMAN, W., SINGH, S.K., SINHA, R., TANDON, S.K. (2009) Climate control on erosion distribution over the Himalaya during the past ~100 ka. *Geology* 37, 559-562.

- RAY, Y., SRIVASTAVA, P. (2010) Widespread aggradation in the mountainous catchment of the Alaknanda-Ganga River System: timescales and implications to Hinterland-foreland relationships. *Quaternary Science Reviews* 29, 2238-2260.
- RAYMO, M.E., RUDDIMAN, W.F. (1992) Tectonic Forcing of Late Cenozoic Climate. Nature 359, 117-122.
- RIEBE, C.S., KIRCHNER, J.W., GRANGER, D.E., FINKEL, R.C. (2001) Strong tectonic and weak climatic control of long-term chemical weathering rates. *Geology* 29, 511-514.
- SANYAL, P., SINHA, R. (2010) Evolution of the Indian summer monsoon: synthesis of continental records. Geological Society, London, Special Publications 342, 153-183.
- SINGH, A.K., PARKASH, B., MOHINDRA, R., THOMAS, J.V., SINGHVI, A.K. (2001) Quaternary alluvial fan sedimentation in the Dehradun Valley Piggyback Basin, NW Himalaya: tectonic and palaeoclimatic implications. *Basin Research* 13, 449-471.
- THOMPSON, L.G., YAO, T., DAVIS, M.E., HENDERSON, K.A., MOSLEY-THOMPSON, E., LIN, P.-N., BEER, J., SYNAL, H.-A., COLE-DAI, J., BOLZAN, J.F. (1997) Tropical Climate Instability: The Last Glacial Cycle from a Qinghai-Tibetan Ice Core. *Science* 276, 1821-1825.
- VIGIER, N., DECARREAU, A., MILLOT, R., CARIGNAN, J., PETIT, S., FRANCE-LANORD, C. (2008) Quantifying Li isotope fractionation during smectite formation and implications for the Li cycle. *Geochimica et Cosmochimica Acta* 72, 780-792.
- VIGIER, N., GISLASON, S.R., BURTON, K.W., MILLOT, R., MOKADEM, F. (2009) The relationship between riverine lithium isotope composition and silicate weathering rates in Iceland. *Earth* and Planetary Science Letters 287, 434-441.
- WALKER, J.C.G., HAYS, P.B., KASTING, J.F. (1981) A negative feedback mechanism for the longterm stabilization of Earth's surface temperature. *Journal of Geophysical Research* 86, 9776-9782.

Geochem. Persp. Let. (2015) I, 10-19 | doi: 10.7185/geochemlet.1502

18

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

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Supplementary Information

The Supplementary Information includes:

- ➤ Sample descriptions and methods
- ➤ Figures S-1 to S-8
- ► Tables S-1 to S-3
- > Supplementary Information References

Sediments were collected from three regions: the upper Yamuna River basin, the Alaknanda River basin and the Donga Fan. In these regions, sediments are derived from two main lithological units: (i) the Higher Himalaya Crystallines (HHC) to the north, in the upper part of the Yamuna and the Alaknanda, are composed mainly of high-grade metamorphic rocks and intrusions of leucogranites; (ii) the Lesser Himalaya (LH) in the outer part of the range is composed mainly of low-grade metasedimentary rocks, limestones and orthogneisses (Fort, 1989). The Yamuna and Alaknanda rivers mainly drain the HHC, and the LH to a lesser extent. Sediments coming from the Tethyan Sedimentary Series (TSS) area do not contribute significantly to their sediment budgets (Rahaman *et al.*, 2009). The Donga Fan collects sediments from the LH only (Singh *et al.*, 2001).

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The clay-sized fraction was selected as: (i) clays have high Li concentrations (20-90 ppm; Table S-1) and are susceptible to record environmental conditions (*e.g.*, temperature, chemistry) during their formation (Vigier *et al.*, 2008; Burton and Vigier, 2011). (ii) By choosing a narrow range of sediment particle size, geochemical variability as a result of hydrodynamic sorting during sediment transport is minimised, although this study focuses on fluvial terraces and alluvial deposits in or at the outer fringe of the Himalayan range where the flow is turbulent in mountainous rivers and there is therefore little hydrodynamic sorting (Lupker *et al.*, 2011).

Sample descriptions

Letter

Field description of sampling locations is presented below, subdivided in the three studied regions: (i) alluvial deposits from the Donga Fan, located at the foothills of the Himalaya between the Ganges and the Yamuna rivers, (ii) terraces of the Alaknanda River, in the Lesser Himalaya and (iii) terraces of the Yamuna River as it exits the Himalayan Range (Fig. S-1).



Figure S-1 Map of the study area. Yellow pushpins show the sample locations. The grey area shows the Donga Fan. The yellow star in the inset represents the location of the study area in India. Satellite image source: Cnes/Spot image from Google Earth™.



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Donga Fan

Two samples were collected from terraces of the Suarna River in the proximal part of the Donga Fan (Fig. S-2): IND02 and IND03 from terraces termed ST1 and ST6 and dated at 10.7 ± 2.4 ka and 29.4 ± 1.7 ka, respectively in Singh *et al.* (2001). IND02 is mostly coarse sand collected between subrounded cobbles, and IND03 is also mostly coarse sand collected between subrounded cobbles and gravels.

IND04 was collected from near the base of an alluvial fan deposit, termed section D3 and dated at 20.3 ± 7.5 ka (Singh *et al.*, 2001). It is composed of coarse grains with abundant subrounded gravels and cobbles. For all Donga Fan samples, sorting is absent and there is no sign of imbrication or other sedimentation features.

Yamuna River terraces

Samples were collected from a set of four terraces of the Yamuna River (Fig. S-2), where the river exits the Himalayan Range. IND06-4.90 and IND06-5.50 were collected at depth of 4.90 and 5.50 m, respectively, in the terrace termed YT2 and dated at 8.9 ± 1.9 ka (Singh *et al.*, 2001). At this site, sediment is mostly composed of subrounded cobbles with no significant sorting and silt to coarse sand material. IND07 was collected at a depth of 2.55 m in terrace termed YT3 and dated at 9.5 ± 2.3 ka (Singh *et al.*, 2001). Sediment is composed mostly of rounded cobbles with coarse sand and silt. IND08-2.60 was collected at a depth of 2.60 m in terrace termed YT4 and dated at 10.7 ± 2.2 ka (Singh *et al.*, 2001). Sediment is composed mostly of rounded cobbles and boulders with coarse sand and silt.



Figure S-2 Google Earth™ image of Donga Fan and Yamuna River sampling locations. The grey area shows the Donga Fan. Flow is from the top to the bottom of the image. Vertical exaggeration is 3x.



Letter

Alaknanda River terraces

Terraces of the Alaknanda River were sampled at four locations, from upstream to downstream: Gaucher, Nagrasu, Rudraprayag and Srinagar (Fig. S-3).

IND51 was collected from near the bottom of terrace T3 at Gaucher, dated at 37 ± 4 ka (Ray and Srivastava, 2010), in a sand layer showing cross-bedding and bounded by two horizons of rounded cobbles and gravels.

IND54 was collected from terrace T1 at Nagrasu, dated at 13 ± 2 ka (Ray and Srivastava, 2010), in a sand horizon bounded by rounded boulders and cobbles. The sampling depth was estimated to be 4 m.

IND57 was collected near the bottom of the terrace at Rudraprayag, 17.7 m below the surface of the terrace, in a massive sand horizon (thickness > 5 m) and dated at 41 \pm 5 ka (Ray and Srivastava, 2010). IND58 was collected in the vertical middle part of the terrace at Rudraprayag, dated at 35 \pm 6 ka (Ray and Srivastava, 2010), 6.5 m below the surface. The sample was mainly coarse sand collected between large rounded cobbles. IND59 was collected near the top of the terrace at Rudraprayag, dated at 18 \pm 3 ka (Ray and Srivastava, 2010), in a homogeneous sand horizon similar to that found in the lower part of the terrace. Sampling depth was 3.20 m.

At Srinagar, samples were collected from five terraces out of six described in Ray and Srivastava (2010): IND64 was taken in terrace T2, dated at 17 ± 2 ka (Ray and Srivastava, 2010), about 15 m below the surface. The sample was collected from a 20-30 cm-thick sand horizon showing no sedimentation features and bounded by poorly sorted rounded cobbles. IND65 was collected in terrace T5 about 10 m below the surface. This part of the terrace was dated at 14 ± 1 ka, while the lower part of T5 was dated at 15 ± 2 ka (Ray and Srivastava, 2010). The sample was taken from a sand horizon ~8 m thick, bounded by poorly sorted rounded cobbles and gravels. IND62 was collected from terrace T4, 20-30 m below the surface. No depositional age was determined for this terrace. However, Ray and Srivastava (2010) described terraces T2 through T5 as a cut-and-fill terrace. Thus, the age of T4 should be intermediate to that of T2 and T5, and a value of 16 ± 1 ka was assigned. The sample was taken from a 20-30 cm thick sand horizon overlain by poorly sorted subrounded cobbles and gravels.

IND72 was collected 6 m below the surface in terrace T6, dated at 26 ± 4 ka (Ray and Srivastava, 2010). The sample was taken from a sandy-silty horizon showing no sedimentation features and at least 10 m thick. IND73T was taken at a depth of 3.80 m in terrace T1 (two other samples, IND73M and IND73B, were collected at different depths but not analysed). This terrace was not directly dated but is younger than the cut-and-fill terrace, which youngest age is 14 ± 1 ka. Thus, an age of 7 ± 7 ka was assigned to IND73T.





Figure S-3 Google Earth[™] image of the Alaknanda River terrace sampling locations. From top to bottom: Gaucher (IND51), Nagrasu (IND54), Rudraprayag (IND57-59) and Srinagar (IND62, 64, 65, 72 and 73). Flow is from top to bottom of the image. Vertical exaggeration is 3x.

Methods

SI-5

Samples were air-dried and sieved at 2 mm. The size fraction smaller than 2 mm was retained and is termed "bulk" throughout this article. Mineralogical quantification was performed by X-ray diffraction at University of Wollongong. Dry bulk samples were crushed to <4 μ m with a Tollmill crusher (TEMA). The samples were then mounted in aluminium holders and placed in a Phillips 1130/90 diffractometer with Spellman DF3 generator set to 1 kW. They were loaded and analysed through an automatic sample holder. The 1 kW energy is achieved by setting the diffractometer to 35 kV and 28.8 mA. Samples were analysed between 4 and 70° 2-theta at 2° per minute with a step size of 0.02. Traces were produced through a GBC 122 control system and analysed using Traces, UPDSM and SIROQUANT softwares.

Bulk samples were analysed for particle size distribution using a Malvern Mastersizer at University of Wollongong. This was used to determine the quantity of bulk material required to obtain ~100 mg of clay-sized (<2 μ m) sediment. An aliquot of the required mass was sub-sampled and the clay fraction was separated by controlled centrifugation (Poppe *et al.*, 2001) (termed thereafter "clay-sized fraction").

For strontium isotope analysis, samples were prepared at the Wollongong Isotope Geochronology Laboratory, University of Wollongong. An aliquot of bulk sediment was ashed at 350 °C to destroy organic matter and then leached with 1.5 M HCl at ambient temperature in an ultrasonic bath to remove calcium



carbonates. The sample was then dissolved in a mixture of concentrated HF, HNO₃ and HCl at 130 °C overnight. Solutions were then dried at 80 °C and residues re-dissolved twice in concentrated HNO₃ to drive off fluorides. After drying, samples were redissolved in 1 M HNO₃ and loaded on an ion-exchange column packed with 0.2 mL of Eichrom Sr resin over 0.1 mL of Eichrom Pre-filter resin. Strontium was eluted in 0.02 M HNO₃. Isotopic measurements were performed on a ThermoScientific Neptune multi-collector ICPMS at the Research School of Earth Sciences, The Australian National University. Synthetic standard SRM987 was regularly analysed to assess accuracy. Total procedure blanks were <200 pg, which is negligible compared to the amount of Sr processed (between 2 and 10 μ g).

For lithium isotope analysis, sample preparation was undertaken at the School of Ocean and Earth Science and Technology, University of Hawai'i at Manoa for bulk sediments, and at the Centre de Recherches Pétrographiques et Géochimiques (CRPG) for clay-sized fractions. Aliquots of bulk sediment were weighed and dissolved in a mixture of concentrated HF, HNO₃ and HCl at 130 °C for 12 hours. Solutions were then dried at 80 °C and residues re-dissolved several times in concentrated HNO₃ to remove fluorides. After drying, residues were re-dissolved in 5 % HNO₃ and solutions were sub-sampled to determine Li concentrations.

For clay-sized fractions, exchangeable Li was removed using 1M acetate ammonium (10 ml for 50 mg of clay), agitated during 1 hour, repeated 3 times. The residue was separated by centrifugation and rinsed with water before dissolution with concentrated acids, following the same procedure as for bulk sediments.

Lithium concentrations for bulk sediments were determined using a ThermoScientific Element ICP-MS at the School of Ocean and Earth Science and Technology, University of Hawai'i at Manoa. Two reference materials (BCR-1, a basalt supplied by US Geological Survey, and JR-1, a rhyolite supplied by the Geological Survey of Japan) were used for calibration and run with Li concentrations of 20 and 200 ppb, respectively. An internal standard with 0.5 ppb of B, In and Sn was added to the samples to correct for beam drift during analysis. Blank correction was performed by analysing 5 % HNO₃ before each standard or sample measurement. Lithium concentrations for clay-sized fractions were determined using an Elan 6000 ICP-MS at the CRPG and the SARM French National Facilities.

Lithium isotopic analysis requires purification by chromatography before measurement. We used the procedure described in Millot *et al.* (2004), Vigier *et al.* (2008), where 2.4 mL of AG50XR cation exchange resin is used. Before loading samples on the column in 0.5 mL of 1 M HCl, the Li concentration in the solutions were adjusted to ~120 ppb.

The ⁷Li/⁶Li ratios were measured on a ThermoScientific Neptune Plus at the Centre de Recherches Pétrographiques et Géochimiques (CRPG) for clay-sized fractions, and on a Nu Instruments[™] high resolution multi-collector ICP-MS at the School of Ocean and Earth Science and Technology, University of Hawai'i



SI-8







Figure S-6 ⁸⁷Sr/⁸⁶Sr ratios in bulk sediment as a function of the distance from the divide for the Donga Fan (triangle), Yamuna (squares) and Alaknanda (diamonds). If ⁸⁷Sr/⁸⁶Sr ratios were controlled by sediment provenance only, *i.e.* which rock types they are derived from, Donga Fan sediments should display the highest ⁸⁷Sr/⁸⁶Sr values because they only originate from the LH where rocks are characterised by high ⁸⁷Sr/⁸⁶Sr ratios (see text for details). Similarly, the ⁸⁷Sr/⁸⁶Sr ratio should increase downstream for the Alaknanda as the river flows from the moderately radiogenic HHC into the very radiogenic LH. However the opposite is observed.



Letter

at Manoa, for the bulk sediments. To correct for mass bias, we used a standard bracketing technique, normalising isotope ratios to the LSVEC standard, in wet plasma, multicollection and low resolution. Solutions were diluted to 5 ppb, which typically resulted in a ⁷Li beam of 5 V. The carryover of the ⁷Li signal typically increased from 10 to 40 mV during the course of the day. This corresponds to a maximum contribution of analysis blank of 0.5 %. This contribution is deduced for each sample by measuring the dilution acid (0.05 M HNO₃) before each standard and sample analysis.

Total procedure blanks were below detection limit (20 pg). Accuracy of the isotope measurements was checked using Li-7N in-house standard solutions (Carignan *et al.*, 2007). The JG-2 granite reference material was also measured for validating the chemical procedure used for these samples. Mean δ^7 Li value for the Li7-N solution was 30.3 ± 0.21 ‰ (2 σ , *n*=7). The average δ^7 Li value for the JG-2 reference material was -1.3 ± 0.7 ‰ (2SD). The external analytical uncertainty on δ^7 Li was estimated by replicate analysis of two bulk sediments (± 0.5 ‰; 2SD).

The δ^7 Li values were calculated by normalising measured ⁷Li/⁶Li ratios to L-SVEC standard (Millot *et al.*, 2004):





Figure S-4 δ^7 Li values in clay-sized fractions (full symbols) and bulk sediments (open symbols). Clay-sized fractions show that δ^7 Li values reach a minimum shortly before the Last Glacial Maximum. This is supported by bulk sediments, although variations are much more dampened. Diamonds: Alakanda, squares: Yamuna, triangles: Donga Fan. The thick dark and light grey curves represent the schematic variation of δ^7 Li compositions with time for the clay-sized fraction and the bulk sediments, respectively. The horizontal grey line shows the average δ^7 Li value for the continental crust (Teng *et al.*, 2004, 2009).



Figure S-7 ⁸⁷Sr/⁸⁶Sr ratios vs (a) micas and (b) orthoclase contents in bulk sediments. The correlations suggest that ⁸⁷Sr/⁸⁶Sr values are controlled by the dissolution of biotite and muscovite, rather than changes in sediment provenance. Errors on ⁸⁷Sr/⁸⁶Sr ratios are smaller than the symbol size. Relative errors on mineral contents are less than 5 %.



Figure 5-8 Example of fluvial deposit sampled: IND64, Srinagar terrace T2, dated at 17 ± 2 ka (Ray and Srivastava, 2010). This terrace is part of a set of six terraces of the Alaknanda River in the Srinagar valley. The deposit consists of 15-20 m of rounded cobbles and boulders showing no preferential imbrication direction and intercalated with sand layers. There is no evidence of post-depositional alteration with neither soil development nor iron oxide staining of the sediment. The sample was collected 15 m below the deposit's surface, in a sand layer.

SI-9



Letter

deposits.
fluvial
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for
data
isotopic
strontium
and
Lithium
Table S-1

SI-II

Sample name	Site	Sampling depth (m)	Deposition age (ka)	2σ		Li (ppm)	2σ	δ ⁷ Li	2σ	⁸⁷ Sr/ ⁸⁶ Sr	2σ
IND02	Suarna T1	22	10.7	2.4	Bulk sediment	12.83	0.07	1.08	0.05	0.797089	0.000013
IND03	Suarna T5	4.65	29.4	1.7	Bulk sediment Clay-sized fraction	$17.61 \\ 46.30$	0.13	-3.97	0.50		
IND04	Donga Fan section D3	20	20.3	7.5	Bulk sediment	18.78	0.07	-0.37	0.02	0.783569	0.000015
IND06-4.90	Yamuna YT2	4.9	8.9	1.9	Bulk sediment	16.05	0.18	1.98	0.10	0.804151	0.000012
IND06-5.50	Yamuna YT2	5.5	8.9	1.9	Bulk sediment Clay-sized fraction	50.72		1.56 2.71	0.08 0.50		
IND07	Yamuna YT3	2.55	9.5	2.3	Bulk sediment	13.31	0.05	-0.18	0.01	0.786227	0.000011
IND08-2.60	Yamuna YT4	2.6	10.7	2.2	Bulk sediment	11.41	0.01	0.79	0.04		
replicate replicate					Bulk sediment Bulk sediment			0.83 0.76	0.04 0.04		
IND51	Gaucher T3	20	37	4	Bulk sediment			0.57	0.03		
IND54	Nagrasu T1	4	13	2	Bulk sediment Clay-sized fraction	60.87		-0.11 -1.65	0.01 0.50		
IND57	Rudraprayag bottom	17.7	41	5	Bulk sediment			0.73	0.04	0.912562	0.000011
					Clay-sized fraction	28.99		-0.99	0.50		
IND58	Rudraprayag middle	6.5	35	9	Bulk sediment	CC 07		1.23	0.06	0.858146	0.000013
					Cluy-sized fraction	10.00		-0.41	00.0		
IND59	Rudraprayag top	3.2	18	ŝ	Bulk sediment Clay-sized fraction	17.68		0.76	0.50		
IND62	Srinagar T4	20	16	-1	Bulk sediment	14.60	0.08				
					Clay-sized fraction	93.04		-0.80	0.50		
IND64	Srinagar T2	15	17	2	Bulk sediment	15.66	0.04	-0.46	0.02	0.761572	0.000010
					Clay-sized fraction	91.16		-2.60	0.50		
IND65	Srinagar T5	10	14		Bulk sediment	17.96	0.08	-1.54	0.08	0.774127	0.000009
	1				Clay-sized fraction	84.93		0.22	0.50		
IND72	Srinagar T6	9	26	4	Bulk sediment	23.53	0.04	-1.08	0.05	0.740728	0.000013
replicate					Bulk sediment Clay-sized fraction	72.46		-1.10 -4.26	0.06 0.50		
IND73T	Srinagar T1	3.8	7	7	Bulk sediment	18.34	0.01	1.64	0.08	0.772868	0.00000

Geochem. Persp. Let. (2015) I, 10-19 | doi: 10.7185/geochemlet.1502

Errors are on Li concentrations, δ Li and 87 Sr/ 86 Sr ratios are internal analytical uncertainties.

3



Letter

Table S-2 Mineralogical data.

				Mile_		A nor-	O"tho_	Kaoli-					
Sample name	Site	Quartz	Biotite	covite	Albite	thite	clase	nite	Chlorite	Gibbsite	Rutile	Titanite	Zircon
IND02	Suarna T1	82	0.4	6.9	4.9	1.1	1.7	\bigtriangledown	\bigtriangledown		\bigtriangledown		1.1
IND04	Donga Fan section D3	60	4.8	20	4.3	1.1	2.7	1.3	\triangleleft	1.1	1.4		1.3
IND06-4.90	Yamuna YT2	53	3.9	18	8.4	3.1	2.6	\bigtriangledown	7.8		\leq 1		\sim
IND07	Yamuna YT3	58	1.9	10	14	4.5		\bigtriangledown	8.8	1	\bigtriangledown		\checkmark
IND08-2.60	Yamuna YT4	82	0.2	8.4	2.2	1.7	2.9	\bigtriangledown			\bigtriangledown		1.8
IND64	Srinagar T2	68	1.4	1.9	11	6.7	5.1	\bigtriangledown	<1	0.4	<1	1.6	~ 1
IND65	Srinagar T5	46	1.9	6.6	19	15	4.6	\checkmark	3.1	0.7	<1	<1	<1
IND73T	Srinagar T1	62	3.1	7.2	10	3.9	3.3	1.2	5.4	0.3	<1	<1	<1
Unit: wt %													

Geochem. Persp. Let. (2015) I, 10-19 | doi: 10.7185/geochemlet.1502

Rock type	δ ⁷ Li (‰)	error	Reference
Amphibolite	4.8	1.4	Teng et al., 2008
Pegmatites	4.5	2.6	Teng et al., 2006
Gneiss	4.1	0.8	Teng <i>et al.,</i> 2008
Granulite	3.5	0.7	Teng <i>et al.,</i> 2008
Granulite (stronalite)	2.4	2.7	Qiu et al., 2011 Chem Geol
Granites	2.1	0.6	Teng <i>et al.,</i> 2009
Bulk continental crust	2.0	2.0	Liu and Rudnick, 2011
Regional amphibolites/schist	2.0	1.0	Liu <i>et al.,</i> 2010
Granites	1.5	1.0	Teng <i>et al.,</i> 2006
Schists	0.8	2.6	Teng <i>et al.,</i> 2006
Sub-greenschist facies	0.5	0.7	Qiu et al., 2011 EPSL
Mudrocks	0.4	1.2	Qiu <i>et al.,</i> 2009
Loess	0.2	1.3	Teng <i>et al.,</i> 2004
Greenschist facies	0.2	0.2	Qiu et al., 2011 EPSL
Granites	0.0	0.8	Teng <i>et al.,</i> 2004
Shales, greywackes and pelites	-0.3	0.9	Teng <i>et al.,</i> 2004
Amphibolite (kinzigite)	-1.3	0.7	Qiu et al., 2011 Chem Geol

Table S-3 Compilation of Li isotopic composition for various rock types.

Errors are 2 standard errors re-calculated from published values.

SI-13

Supplementary Information References

- BURTON, K.W., VIGIER, N. (2011) Lithium Isotopes as Tracers in Marine and Terrestrial Environments. In: Baskaran, M. (Eds.) *Handbook of Environmental Isotope Geochemistry*. Springer Berlin Heidelberg, 41-59.
- CARIGNAN, J., VIGIER, N., MILLOT, R. (2007) Three secondary reference materials for lithium isotope measurements: Li7-N, Li6-N and LiCl-N solutions. *Geostandards and Geoanalytical Research* 31, 7-12.
- FORT, P. (1989) The Himalayan Orogenic Segment. In: engör, A.M.C. (Eds.) Tectonic Evolution of the Tethyan Region. Springer Netherlands, 289-386.
- LUPKER, M., FRANCE-LANORD, C., LAVÉ, J., BOUCHEZ, J., GALY, V., MÉTIVIER, F., GAILLARDET, J., LARTIGES, B., MUGNIER, J.-L. (2011) A Rouse-based method to integrate the chemical composition of river sediments: application to the Ganga basin. *Journal of Geophysical Research* 116, F04012.
- MILLOT, R., GUERROT, C., VIGIER, N. (2004) Accurate and high-precision measurement of lithium isotopes in two reference materials by MC-ICP-MS. *Geostandards and Geoanalytical Research* 28, 153-159.
- POPPE, L.J., PASKEVICH, V.F., HATHAWAY, J.C., BLACKWOOD, D.S. (2001) A laboratory manual for X-Ray powder diffraction. USGS Open-File Report, 01-041.
- RAHAMAN, W., SINGH, S.K., SINHA, R., TANDON, S.K. (2009) Climate control on erosion distribution over the Himalaya during the past ~100 ka. *Geology* 37, 559-562.
- RAY, Y., SRIVASTAVA, P. (2010) Widespread aggradation in the mountainous catchment of the Alaknanda-Ganga River System: timescales and implications to Hinterland-foreland relationships. *Quaternary Science Reviews* 29, 2238-2260.
- SINGH, A.K., PARKASH, B., MOHINDRA, R., THOMAS, J.V., SINGHVI, A.K. (2001) Quaternary alluvial fan sedimentation in the Dehradun Valley Piggyback Basin, NW Himalaya: tectonic and palaeoclimatic implications. *Basin Research* 13, 449-471.
- TENG, F.Z., MCDONOUGH, W.F., RUDNICK, R.L., DALPÉ, C., TOMASCAK, P.B., CHAPPELL, B.W., GAO, S. (2004) Lithium isotopic composition and concentration of the upper continental crust. *Geochimica et Cosmochimica Acta* 68, 4167-4178.
- TENG, F.Z., RUDNICK, R.L., MCDONOUGH, W.F., WU, F.-Y. (2009) Lithium isotopic systematics of A-type granites and their mafic enclaves: Further constraints on the Li isotopic composition of the continental crust. *Chemical Geology* 262, 370-379.
- VIGIER, N., DECARREAU, A., MILLOT, R., CARIGNAN, J., PETIT, S., FRANCE-LANORD, C. (2008) Quantifying Li isotope fractionation during smectite formation and implications for the Li cycle. *Geochimica et Cosmochimica Acta* 72, 780-792.



Letter

