



Lithium isotope behaviour during weathering in the Ganges Alluvial Plain

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Abstract

The Ganges river system is responsible for the transportation of a large flux of dissolved materials derived from Himalayan weathering to the oceans. Silicate weathering-driven cooling resulting from uplift of the Himalayas has been proposed to be a key player in Cenozoic climate variation. This study has analysed Li isotope ($\delta^7\text{Li}$) ratios from over 50 Ganges river waters and sediments, in order to trace silicate weathering processes. Sediments have $\delta^7\text{Li}$ of $\sim 0\text{‰}$, identical to bulk continental crust, however suspended sediment depth profiles do not display variations associated with grain size that have been observed in other large river systems. Dissolved $\delta^7\text{Li}$ are low ($\sim 11\text{‰}$) in the Ganges headwaters, but reach a constant value of $21 \pm 1.6\text{‰}$ within a relatively short distance downstream, which is then maintained for almost 2000 km to the Ganges mouth. Given that Li isotopes are controlled by the ratio of primary mineral dissolution to secondary mineral formation, this suggests that the Ganges floodplain is at steady-state in terms of these processes for most of its length. Low $\delta^7\text{Li}$ in the mountainous regions suggest silicate weathering is therefore at its most congruent where uplift and fresh silicate exposure rates are high. However, there is no correlation between $\delta^7\text{Li}$ and the silicate weathering rate in these rivers, suggesting that Li isotopes cannot be used as a weathering-rate tracer, although they do inform on weathering congruency and intensity. The close-to-constant $\delta^7\text{Li}$ values for the final 2000 km of Ganges flow also suggest that once the size of the alluvial plain reached more than ~ 500 km (the flow distance after which riverine $\delta^7\text{Li}$ stops varying), the Ganges exerted little influence on the changing Cenozoic seawater $\delta^7\text{Li}$, because riverine $\delta^7\text{Li}$ attained a near steady-state composition.

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1. INTRODUCTION

Chemical weathering of silicate rocks is a significant long-term removal process of atmospheric CO_2 , both through supply of cations and alkalinity for marine carbon-

ate precipitation, and through supply of nutrients for organic carbon burial (Walker et al., 1981; Berner et al., 1983; West et al., 2002, 2005; Berner, 2003). As such, it has long been postulated as a key long-term climate control and stabilisation mechanism. In the Cenozoic, in particular, it is questioned whether the observed ~ 40 Myr cooling trend and CO_2 decline were caused by uplift of the Himalayas, which increased the supply of primary material for weathering, which in turn removed more atmospheric

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CO₂ (Raymo et al., 1988; Raymo and Ruddiman, 1992; Raymo, 1994). The question of whether tectonic processes can strongly affect CO₂ drawdown is critical for understanding the Earth's climate system, because it may suggest that a temperature-driven stabilising feedback on CO₂ via weathering (the Earth's “weathering thermostat”) is at times a relatively weak control on climate compared to a supply-driven or organic carbon-driven control (Gíslason et al., 2006; Gíslason et al., 2009; Oelkers et al., 2012; West, 2012; Maher and Chamberlain, 2014).

Evidence for the controls on weathering initially focussed on marine Sr isotope ratios, which started to increase approximately 40 Ma ago (McArthur et al., 2001). This increase was interpreted as a greater input of radiogenic continental material, implying greater weathering and CO₂ drawdown, likely due to Himalayan uplift. However, it has since become clear that the interpretation of the ⁸⁷Sr/⁸⁶Sr curve is not straightforward, because of the effect of metamorphosed radiogenic Himalayan carbonates (Palmer and Edmond, 1992; Galy et al., 1999; Oliver et al., 2003; Bickle et al., 2005). It is therefore not clear how much of the increase in seawater ⁸⁷Sr/⁸⁶Sr is due to weathering of continental silicates, and how much due to metamorphosed carbonates (Galy et al., 1999), the latter of which would not affect atmospheric CO₂ on timescales >~10,000 years, and may even be a source of CO₂, if the dominant source of acidity is sulphuric acid (Torres et al., 2014). Equally, seawater osmium isotopes show a punctuated increase from ~60 Ma (Peucker-Ehrenbrink et al., 1995; Peucker-Ehrenbrink and Ravizza, 2000), and here debate has focussed on the control radiogenic black shales in Himalayan lithologies may exert on seawater ¹⁸⁷Os/¹⁸⁸Os ratios (Pierson-Wickmann et al., 2002a,b). Hence, both of the above radiogenic isotope systems can at best only provide somewhat ambiguous interpretations as to whether weathering rates, or rock weatherability, increased due to the Himalayas, because of the effect of individual Sr- and Os-rich lithologies.

More recently, lithium isotopes have been used to try to answer this question. Seawater $\delta^7\text{Li}$ appears to have started increasing between 50 and 60 Ma, with behaviour more similar to Os than Sr isotope records (Hathorne and James, 2006; Misra and Froelich, 2012). As a stable isotope system, Li is virtually unaffected by the lithologies undergoing weathering, and is isotopically fractionated by the weathering process. Specifically, Li in rocks is overwhelmingly dominated by the silicate fraction, meaning that Li isotopes are probably the only proxy that solely traces silicate weathering (Kisakürek et al., 2005; Millot et al., 2010). The Li isotope ratio of silicate rocks describes a very narrow range ($\delta^7\text{Li}_{\text{continental crust}} \sim 0.6 \pm 0.6\text{‰}$ (Teng et al., 2004; Sauzeat et al., 2015); $\delta^7\text{Li}_{\text{basalt}} = \sim 3\text{--}5\text{‰}$ (Elliott et al., 2006; Tomascak et al., 2008)), relative to that reported in river waters ($\delta^7\text{Li} = 2\text{--}44\text{‰}$; global mean 23‰ (Huh et al., 1998, 2001; Kisakürek et al., 2005; Pogge von Strandmann et al., 2006, 2010; Vigier et al., 2009; Millot et al., 2010; Wimpenny et al., 2010; Witherow et al., 2010; Rad et al., 2013; Dellinger et al., 2014; Bagard et al., 2015; Liu et al., 2015; Pogge von Strandmann and Henderson, 2015; Wang et al., 2015; Henchiri et al.,

2016). This high variability in rivers is caused by weathering processes: dissolution of silicates causes no isotope fractionation, but secondary minerals formed during weathering preferentially take up ⁶Li, driving residual waters isotopically heavy (Huh et al., 2001; Pistiner and Henderson, 2003; Vigier et al., 2008; Pogge von Strandmann et al., 2010; Wimpenny et al., 2010a). Therefore, surface water $\delta^7\text{Li}$ is controlled by the ratio of primary mineral dissolution (low $\delta^7\text{Li}$, high [Li]) relative to secondary mineral formation (driving waters to high $\delta^7\text{Li}$, and low [Li]). This ratio has also been described as the weathering congruency (Misra and Froelich, 2012; Pogge von Strandmann et al., 2013), weathering efficiency (Pogge von Strandmann and Henderson, 2015) or weathering intensity (Pogge von Strandmann et al., 2010; Dellinger et al., 2015). When riverine $\delta^7\text{Li} = \text{rock } \delta^7\text{Li}$, then weathering is congruent (water chemistry = rock chemistry), efficient (cations are not retained in clays, but are delivered to the oceans) and low intensity (little clay formation, and a low weathering to denudation ratio – bearing in mind that “weathering intensity” has been defined differently by different authors).

If, as seems likely, the ~9‰ increase from early Cenozoic seawater to the present day was controlled by changes in weathering rates or processes, it would imply an increasing relative proportion of secondary mineral formation (Misra and Froelich, 2012; Bouchez et al., 2013; Li and West, 2014; Wanner et al., 2014; Vigier and Godderis, 2015). Initially, this was interpreted as directly due to mountain uplift (Misra and Froelich, 2012). However, High Himalayan rivers tend to have low $\delta^7\text{Li}$ values suggesting little clay formation (Kisakürek et al., 2005), and in general uplifting and denuding terrains tend to have lower $\delta^7\text{Li}$ than flatter terrains, because of the continuous exposure of isotopically light primary rock, and lower rock–water interaction times in the former, and longer water–rock interaction times leading to greater precipitation of secondary minerals in the low-lying areas (Dellinger et al., 2015; Pogge von Strandmann and Henderson, 2015). This has led to the proposal that the seawater $\delta^7\text{Li}$ curve is charting the evolution of the Himalayan floodplains (or others, such as those of the Andes), rather than the mountain uplift and exposure itself (Pogge von Strandmann and Henderson, 2015). This also agrees with the concept that more CO₂ is drawn down in these floodplains, rather than in the mountains (West et al., 2002; Lupker et al., 2012).

Therefore, studies have begun examining the Ganges-Brahmaputra floodplains to examine weathering and Li behaviour in low-lying floodplain settings. Sample sets taken ~10 years apart (in the late 1980s and late 1990s) from Rishikesh in the Ganges headwaters have lower $\delta^7\text{Li}$ than samples taken close to the river mouth (Huh et al., 1998; Bagard et al., 2015). To date, the highest sample resolution is from Bagard et al. (2015), which comprises four samples along the ~2500 km of the Ganges, with $\delta^7\text{Li}$ increasing from ~10 to 24‰.

This study has increased this sample resolution by an order of magnitude, with an average sample spacing of ~50 km along the Ganges and its tributaries, using samples that have previously been analysed for silicon isotopes (Frings et al., 2015). We have also analysed the Li isotope

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