Earth and Planetary Science Letters 427 (2015) 136–148

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

www.elsevier.com/locate/epsl

Silicate weathering in the Ganges alluvial plain

Patrick J. Frings [a,*], Wim Clymans [a], Guillaume Fontorbe [a], William Gray [b], Govind J. Chakrapani [c], Daniel J. Conley [a], Christina De La Rocha [d,1]

[a] *Department of Geology, Lund University, Sölvegaten 12, 223 62, Lund, Sweden*
[b] *Department of Earth Science and Marine Science Institute, University of California, Santa Barbara, CA 93106, USA*
[c] *Department of Earth Sciences, Indian Institute of Technology, Roorkee, India*
[d] *IUEM, CNRS UMR 5639, University of West Brittany, Plouzané, France*

ARTICLE INFO

Article history:
Received 17 February 2015
Received in revised form 20 June 2015
Accepted 24 June 2015
Available online 16 July 2015
Editor: M. Frank

Keywords:
silicon isotopes
silicate weathering
dissolved silica
Ganges
lowland weathering

ABSTRACT

The Ganges is one of the world's largest rivers and lies at the heart of a body of literature that investigates the interaction between mountain orogeny, weathering and global climate change. Three regions can be recognised in the Ganges basin, with the Himalayan orogeny to the north and the plateaus of peninsular India to the south together delimiting the Ganges alluvial plain. Despite constituting approximately 80% of the basin, weathering processes in the peninsula and alluvial plain have received little attention. Here we present an analysis of 51 water samples along a transect of the alluvial plain, including all major tributaries. We focus on the geochemistry of silicon and its isotopes. Area normalised dissolved Si yields are approximately twice as high in rivers of Himalaya origin than the plain and peninsular tributaries (82, 51 and 32 kmol SiO_2 km^{-2} yr^{-1}, respectively). Such dissolved Si fluxes are not widely used as weathering rate indicators because a large but variable fraction of the DSi mobilised during the initial weathering process is retained in secondary clay minerals. However, the silicon isotopic composition of dissolved Si (expressed as $\delta^{30}Si$) varies from +0.8‰ in the Ganges mainstem at the Himalaya front to +3.0‰ in alluvial plain streams and appears to be controlled by weathering congruency, i.e. by the degree of incorporation of Si into secondary phases. The higher $\delta^{30}Si$ values therefore reflect decreasing weathering congruency in the lowland river catchments. This is exploited to quantify the degree of removal using a Rayleigh isotope mass balance model, and consequently derive initial silica mobilisation rates of 200, 150 and 107 kmol SiO_2 km^{-2} yr^{-1}, for the Himalaya, peninsular India and the alluvial plain, respectively. Because the non-Himalayan regions dominate the catchment area, the majority of initial silica mobilisation from primary minerals occurs in the alluvial plain and peninsular catchment (41% and 34%, respectively).

1. Introduction

On geological timescales, Earth's climate is regulated by a balance between silicate weathering reactions that consume atmospheric CO_2 and a continuous input of carbon from volcanic and metamorphic degassing (Walker et al., 1981). Given that degassing rates vary on long timescales but climate has remained broadly stable, a negative feedback between the rate of CO_2 removal and atmospheric CO_2 concentrations must exist (Berner and Caldeira, 1997). The dependency of silicate weathering rates on climate is the strongest contender for such a feedback, although the exact nature of this dependency remains elusive. River geochemistry is an integrative function of catchment solute inputs and biogeochemical cycling, and so is an excellent tool with which to quantify and understand weathering rates, provided inputs of solutes from non-silicate sources, biological cycling and human activity can be successfully de-convolved.

Driven by the hypothesis that the long-term global cooling since the early Eocene, illustrated, for example, by marine sediment records (Zachos et al., 2001) can be attributed to the Himalayan orogenesis over the same period (Raymo and Ruddiman, 1992), many investigations focus on the major and trace element and isotope geochemistry of the mountainous headwaters of large rivers. As weathering processes in these regions become better understood their role as important long-term CO_2 sinks is increasingly questioned due to their apparently modest silicate weathering rates and small surface areas (see e.g. Moore et al., 2013). However, there is also a growing awareness that lowland regions

---

[*] Corresponding author.
E-mail address: patrick.frings@geol.lu.se (P.J. Frings).

[1] Presently no affiliation.

http://dx.doi.org/10.1016/j.epsl.2015.06.049

of large rivers potentially play an important role in the generation of river weathering fluxes. The deposition and active reworking of freshly eroded and highly weatherable material in a system with water and sediment residence times substantially longer than the upstream source areas creates potential for additional silicate weathering and CO_2 consumption (Bouchez et al., 2012; Lupker et al., 2012). Unfortunately, the capacity of the lowland regions to modify weathering fluxes at the whole-basin scale is poorly understood. For example, the Ganges is one of the world's largest river systems, lies at the heart of the uplift-climate hypothesis (Raymo and Ruddiman, 1992), and has a vast alluvial plain in the foreland basin of the Himalaya. Yet few studies have attempted to budget silicate weathering in the alluvial plain although it constitutes the majority of the Ganges basin. Those that have are often complicated by the anthropogenically perturbed solute and sediment budgets, or the presence of evaporite soils (Rengarajan et al., 2009).

Examination of the biogeochemical cycling of silicon (Si) can provide insights into weathering processes in the Ganges alluvial plain (hereafter 'GAP') because Si derives ultimately – and exclusively – from silicate minerals. Indeed, the flux of dissolved Si (DSi) from a catchment has often been used as a surrogate for silicate weathering rates (Edmond and Huh, 1997; Rengarajan et al., 2009; White and Blum, 1995). However, DSi fluxes are not widely used since a large but variable amount of DSi mobilised during solubilisation of primary minerals typically ends up locked into clay minerals or cycling biologically, meaning what is observed in a river does not necessarily directly reflect the initial weathering. If we are to quantify weathering fluxes, the information we really want is the rate at which Si is mobilised from primary minerals, before some fraction of it is incorporated into secondary clays and biogenic silica. This could be further converted into a silicate–cation weathering rate if the Si/cation ratio of the parent material is known (although the fraction of silicate hydrolysis driven by inorganic acids and the fraction of silicate-hosted cations incorporated into secondary clays must be accounted for by other methods). Previous work on Si geochemistry in the Ganges basin has focused mostly on the behaviour of Si isotopes in the upper end of the basin (Fontorbe et al., 2013), or in groundwater of the Ganges–Brahmaputra delta (Georg et al., 2009); knowledge of Si geochemistry and its relation to silicate weathering in the GAP is limited.

Here, based on samples from 51 sites in the Ganges basin, we use an approach that exploits the fractionation of silicon isotopes during removal of Si from solution to constrain initial DSi mobilisation rates. Our results show that a majority of initial Si mobilisation occurs in lowland regions of the catchment, albeit at lower surface area-normalised rates than the Himalaya. Overall, this suggests that any role the Himalayan orogeny plays as a driver and determinant of global climate change has been modulated by the evolution and functioning of its alluvial plains over time.

2. Study area

The Ganges (Fig. 1) drains a basin of $\sim 0.98 \times 10^6$ km^2 and has a mean annual discharge in Bangladesh of $\sim 490 \times 10^9$ m^3 yr^{-1}. Conventionally the Ganges basin is subdivided into three broad physiographic regions: the Himalayan orogeny to the north, the hills and plateaus of Peninsular India to the south, with the GAP lying between. Climate within the Ganges basin varies widely, from tropical/sub-tropical in the south to areas with sub-zero mean annual temperatures in the high Himalaya (Tripathy and Singh, 2010). The most important hydrological aspect is the annual monsoon which occurs progressively from the southwest between approximately June and September and provides ~80% of the rainfall in the basin (Dalai et al., 2002; Tripathy and Singh, 2010). Rainfall is not evenly divided between the three regions; the Himalaya have average runoff of ~ 100 cm yr^{-1}, Peninsular India ~ 30 cm yr^{-1} and the GAP ~ 46 cm yr^{-1}.

Ganges catchment streams draining the Himalaya are well studied (e.g. Bickle et al., 2003; Chakrapani, 2005; Dalai et al., 2002; Fontorbe et al., 2013; Galy and France-Lanord, 1999) and are generally characterised by rapid physical and chemical erosion rates, weathering fluxes dominated by carbonate weathering and minor contributions from silicate weathering and hot springs. Besides the headwaters of the Ganges itself, major tributaries draining into the Ganges from the Himalayan orogeny include the Yamuna, Ramganga, Ghaghara (also known as the Karnali), the Gandak (the Narayani) and the Kosi. The Himalaya account for 17% of the total surface area of the basin, and 34% of total runoff, and are thought to supply ~44% of dissolved Si (Galy and France-Lanord, 1999).

The southern part of the Ganges basin partly drains a plateau of the Bundelkhand crystalline granites and Vindhyan Precambrian, shales, sandstones and sedimentary carbonates. Together, they constitute a peripheral cratonic bulge that forms part of the Indian shield. The south–western basin also partly drains the Deccan Traps, solidified flood basalts from the late Cretaceous. The shield is relatively low-elevation and is covered in part by deeply weathered soils (laterites) as well as saline and alkaline soils. As a result, the headwaters of southern tributaries of the Ganges have different chemistries compared to the Himalayan streams, e.g. their cation compositions are often dominated by Na^+ (Rengarajan et al., 2009) instead of Ca^{2+}. Major rivers draining peninsular India include the Chambal, Betwa and Ken, tributaries to the Yamuna with their headwaters in the Deccan Traps, and the Tons (also known as the Tamas) and the Son (the Sone) (Fig. 1). Peninsular India accounts for ~31% of the Ganges basin surface area, and 19% of the runoff. Galy and France-Lanord (1999) estimate that peninsular catchments supply ~30% of total exported DSi.

The GAP is essentially the result of the infilling of the active foreland basin by fluvial sediments, of both Himalayan and cratonic origin, up to several kilometres thick. The ~300 km wide GAP lies in a south-easterly direction and consists of massive beds of clays, sand and gravels, mostly Himalayan, and currently aggrades by $\sim 65 \times 10^6$ tons yr^{-1} (Sinha, 2005). It is a region of low elevation, limited topography and high population density. Many streams drain the GAP directly; the largest of these is the 900 km long Gomati (also referred to as the Gomti), which originates ~50 km from the Himalaya and drains an interfluve between the Ramganga and the Ghaghara. Other notable tributaries include the Punpun from the south and the Buri Gandak from the north. The GAP accounts for 52% of the total surface area of the Ganges basin. The alluvial plain supplies about 33% of total Ganga discharge and ~26% of DSi (Galy and France-Lanord, 1999), and disproportionately contribute Na, Cl and Sr (Rai et al., 2010).

An unusual feature of the GAP is the presence of saline/alkali soil salts and carbonates, locally called *kankar*, that are thought to form through repeated annual wetting and drying of depressions or seasonally endorheic areas. Locally, they can strongly influence the water chemistry but their composition is poorly constrained (Galy and France-Lanord, 1999; Rengarajan et al., 2009; Sarin et al., 1989). Large-scale human activity has altered the hydrology of the region through irrigation, including the construction of canals, groundwater abstraction and damming for hydropower or water management, although this is still somewhat limited in scope.

3. Materials and methods

3.1. Fieldwork

Samples were collected at 51 sites within the Ganges fluvial network, including 19 on the Ganges mainstem itself. They span