

Carbonate and silicate weathering in glacial environments and its relation to atmospheric CO₂ cycling in the Himalaya

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ABSTRACT. This paper presents new insights into the global carbon cycle related to CO₂ consumption from chemical denudation in heavily glacierised Himalayan catchments. Data from previous studies of solute concentrations from glacierised catchments were reprocessed to determine the regional scale of CO₂ consumption and solute hydrolysis. The results show that ~90% of the SO₄²⁻ is derived from crustal sulphide oxidation and ~10% from aerosols and sea salts. However, HCO₃⁻ flux calculation estimates contribution from sulphide oxidation to carbonate dissolution (SO-CD) (~21%), similar to the contributions from silicate dissolution and simple hydrolysis (~21 and ~20%, respectively). Furthermore, the atmospheric CO₂ consumption estimations suggests 10.6 × 10⁴ mole km⁻² a⁻¹ (19%) through silicate weathering, 15.7 × 10⁴ mole km⁻² a⁻¹ (28%) through simple hydrolysis, 9.6 × 10⁴ mole km⁻² a⁻¹ (17%) through SO-CD reaction and 5.9 × 10⁴ mole km⁻² a⁻¹ (11%) through carbonate carbonation reaction. Our solute provenance calculations clearly indicate that HCO₃⁻ production and CO₂ consumption via silicate weathering reactions is balanced by the simple hydrolysis and coupled SO-CD process. This shows a counter mechanism operating in subglacial environments of the Himalaya as a source of CO₂ to runoff rather than a sink.

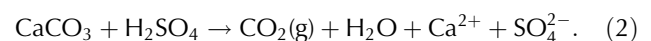
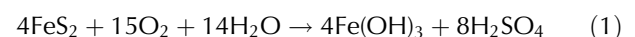
KEYWORDS: CO₂ consumption, glacierised catchments, Himalaya, silicate and carbonate weathering, sulphide oxidation

INTRODUCTION AND BACKGROUND

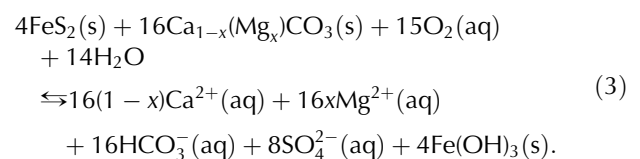
Weathering is a ubiquitous phenomenon that operates at different rates throughout the Earth's surface. Its feedback among glaciation, chemical weathering and carbon cycling has affected the global climate system over geological time scales (Gislason and others, 2009; Torres and others, 2017). It is widely accepted that silicate weathering has a profound influence on long-term (million years) CO₂ cycling (Urey, 1952; Berner and others, 1983; Sharp and others, 1995; Gislason and others, 2009; Elderfield, 2010). However, the rapid dissolution kinetics of carbonate weathering has not been given equal attention in studies of global CO₂ cycles (Liu and others, 2011; Torres and others, 2017). Particularly, importance of sulphuric-acid-generating processes (sulphide oxidation (SO)) requires more attention.

The weathering of trace sulphide and carbonate minerals, characterised as coupled sulphide oxidation and carbonate dissolution (SO-CD), provides a transient source of CO₂ (Raiswell, 1984; Tranter and others, 2002; Wadham and others, 2010; Torres and others, 2014), and thus becomes important to consider with respect to CO₂ flux (Eqns (1–3)). SO also includes transformation of redox-sensitive elements (sulphur, iron and oxygen) and, once coupled with the carbonate dissolution (CD) process, occurs more rapidly than with silicate minerals (Eqns (1) and (2)). Overall SO-CD (Eqn (3)) therefore supplies more minerals for reaction and affects the global CO₂ budget (Torres and others, 2016, 2017). The estimation of CO₂ release as a result of SO-CD involving glacial sediments is important to explain the

potential influence of chemical weathering in glacial–interglacial cycles of the Earth's climate (Hodson and others, 2000; Torres and others, 2017). This depends upon the burial and subsequent oxidation of the carbon and sulphur compounds involved in reaction (Berner and others, 2000).



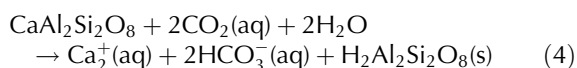
The release of CO₂ from Eqn (2) occurs immediately (Torres and others, 2014). Moreover, subglacial chemical weathering is equally affected by the carbonation of the rock surface via atmospheric CO₂, SO-CD reactions (Tranter and others, 2002; Wadham and others, 2010) and simple hydrolysis (Yde and others, 2005; Stachnik and others, 2016a). This SO-CD process dominates in the distributed drainage system of the subglacial environment (Tranter and others, 2002).



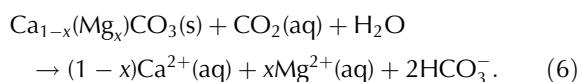
These reactions (i.e. SO, simple hydrolysis) also lead to the dissolution of silicates under subglacial conditions and in the proglacial zone, which have a similar effect as the dissolution of atmospheric CO₂ (Tranter and others, 2002). Present studies in

glacierised catchments require determination of the relationship between the aforementioned reactions to identify the relative importance of CO₂ sequestration via weathering fluxes.

In addition, the mechanism of long-term atmospheric CO₂ drawdown, regulated via climate–landform relations, temperature fluctuation and variable runoff, is thought to stabilise CO₂ concentrations in the atmosphere and thus global climate (Maher and Chamberlain, 2014). Siliciclastic/siliceous rock weathering enhanced by carbonic acids derived from atmospheric CO₂ dissolution causes a long-term CO₂ drawdown (Eqns (4) and (5)) due to CaCO₃ precipitation (Eqn 5; Sharp and others, 1995, Tranter and others, 2002). One mole of CO₂ returns to the atmosphere, whereas another is deposited as carbonate minerals in marine sediments (Eqn 5). However, the location and magnitude of the carbonate precipitation are poorly constrained. Quantification of these processes is equally important because they modify CO₂ feedback, which affects atmospheric loading and drives climate change processes.



The process leading to short-term consumption of CO₂ is the carbonation of carbonates (CC), which is represented as follows:



The release of CO₂ is initiated in first-order streams, where it begins to transform via biotic and abiotic reactions and is stored in terrestrial environments in various forms such as precipitated carbonates and transient CO₂ sinks in soils, ice and snow.

In the present paper, the role of the Himalayan glacierised mountain system in CO₂ cycling via glacial weathering is examined, highlighting the spatial variations in solute concentration and their relation to possible causal factors such as lithology and runoff. The quantitative assessment of long-term and short-term CO₂ consumption through silicate vs carbonate weathering and the relative contribution of

solute fluxes in glacial weathering reactions within this region are also determined. There is a lack of such comprehensive studies combining data from different glacierised basins in the Himalayan region.

STUDY SITES

This study was focused on the glacierised catchments of the central Higher Himalayan Crystalline (HHC) belt (Table 1). Available datasets from selected Indian Himalayan glacial catchments draining the crystalline belt of the Himalaya were compared. The location of the studied glacial catchments is shown in Figure 1. The study area is bounded between major thrust systems comprising complex metamorphic and granitic rocks (Valdiya, 1999; Yin, 2006; Fig. 2). Major rock types of the area consist of calc-silicate gneiss and calc-schist rocks interbedded with subordinate biotite-psammitic gneiss and granite pegmatite apatite veins. These are bounded by the floor thrust of the Main Central Thrust (MCT) in the South and the Trans Himadri Fault in the North (Heim and Gansser, 1939; Valdiya, 1998).

METHODS

Solute provenance calculations

For the requirement of working with a full ablation season of geochemical data, the Himalayan melt period of June to October was considered for this study. We assembled data of temporal and spatial solute fluxes from carefully selected central Himalayan glaciers between 1989 and 2012. We averaged all the solute data for the whole season collected near the snout of the glaciers. The Himalayan glaciers drain 80% of their discharge during the prevailing ablation periods and thus a majority of the major ion flux occurs during this period as well. The details of selected glaciers and sampling periods are as follows:

1. Meltwater samples were collected from the stream originating from the Chhota Shigri glacier at 3800 m a.s.l. for 10–15 d (from the last week of September to the first week of October) for each year between 2003 and 2007. A total of 40 samples were collected during each sampling year (Sharma and others, 2013).

Table 1. The focus of Indian Himalaya glaciers underlain by Higher Himalayan Crystalline bedrock of metamorphic and calcisilicatic lithologies

Glacier name	Area (km ²)	location		Geology	Reference
		Lat	Long		
Gangotri	549.25	30.93	79.28	Granite (tourmaline rich), phyllite, quartzite, schist, limestone and sulphide minerals	Singh and others (2014)
Dokriani	23	30.87	78.97	Vaikrita gniesses, shale and carbonates	Hasnain and Thayyen (1999)
Chhota Shigri	10	32.28	77.53	Metamorphites, migmatites and gniesses black slates, phyllites and fine-grained biotite-schists	Sharma and others (2013)
Bada Shigri	131	32.20	77.67	Metamorphites, migmatites and gniesses black slates, phyllites and fine-grained biotite-schists	Singh and Ramanathan (2015)
Patsio	7.82	32.79	77.35	Granite, granitic gneiss, quartzite, mica schist, magmatites	Singh and others (2015a)
Chaturangi	67.7	30.90	79.25	Granite (tourmaline rich), phyllite, quartzite, schist, limestone and sulphide minerals	Singh and others (2015b)
Satopanth	21	30.70	79.42	Biotite gneiss frequently interbedded with lime silicate marbles and fine grained hornblende gneiss	Chauhan and Hasnain (1993)

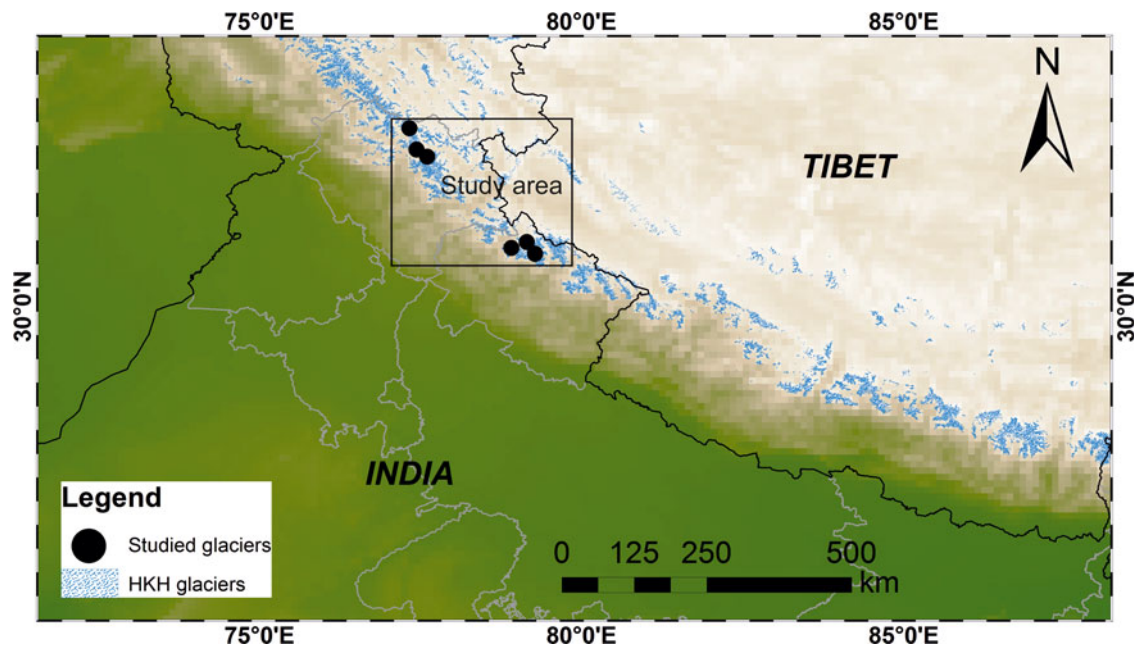


Fig. 1. Study area representing the glacial distribution of Hindu Kush Himalayan region and the studied glacierised catchments are marked as black circle symbol.

2. For the hydro-chemical studies of the Patsio glacier, a total of 72 meltwater samples were collected during September for three consecutive years: 2010, 2011 and 2012 (Singh and others, 2015a).
3. The Satopanth and Bhagirathi glaciers were sampled during the middle of the summer ablation period from 17 to 23 August 1989, and towards the end of the ablation period from 11 to 19 October 1991 (Chauhan and Hasnain, 1993).
4. Sampling of meltwater at the Bara Shigri glacier was carried out during the ablation period of July and September 2012 and from August to September 2013 (Singh and others, 2015a; b).

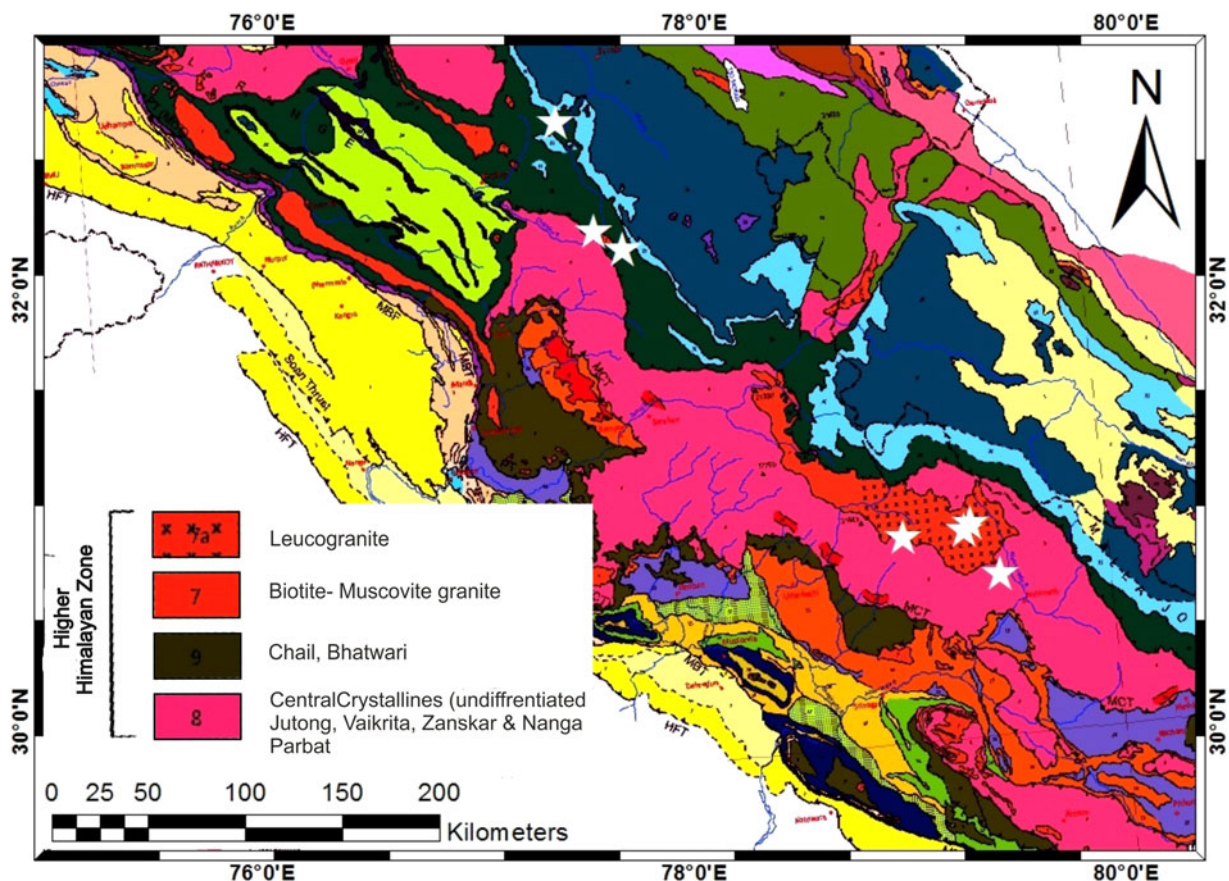


Fig. 2. Geological map of Western Himalaya showing major lithologies modified after Thakur and Rawat (1992). The studied glaciers are marked as star symbol.

- The Chaturangi glacier meltwater was sampled during August and September 2008 (Singh and others, 2015b).
- Gangotri glacier meltwater samples were collected during pre-monsoon (May–June), monsoon (July–August) and post-monsoon (15–30 September) periods during 2008 (Singh and others, 2014). For the successive year of 2009 (from 10 to 25 June) meltwater samples were collected for another study (Singh and others, 2014).
- The solute concentration of the Dokriani glacier used for the study was reported by Hasnain and Thayyen (1996) for the whole ablation season from June to September 1994 (Hasnain and Thayyen, 1999).

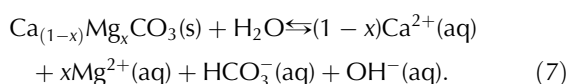
The major ion concentrations reported in mg L⁻¹ and µeq L⁻¹ were transformed to µmole L⁻¹ for calculations.

We apportioned the meltwater solute concentration into crustal, sea-salt and aerosol components before assessing the chemical weathering and atmospheric CO₂ consumption contributions. NO₃⁻ and Cl⁻ were assumed to be of atmospheric and marine origin and the sea-salt contributions of Ca²⁺, Na⁺, Mg²⁺ and K⁺ were estimated using standard sea-water ratios of SO₄²⁻/Cl⁻ (0.051) as used by Holland (1978). Although NO₃⁻ could also be produced in a glacial system (Wynn and others, 2007), no such study for the Himalayan region has been completed to date. As the site-specific snowpack-derived sulphate, nitrate and chloride data were not available for every site, the average concentration for the Himalayan snowpack was calculated from available datasets and the atmospheric SO₄²⁻/Cl⁻ ratio (0.44) was used for calculation (Supplementary Table S1 and references therein).

For determination of the provenance of the HCO₃⁻, we divided it into different categories following the procedure of Sharp and others (1995), Hodson and others (2000) and Tranter and others (2002). We improved these methods by including the simple hydrolysis reaction in the provenance budgets according to Hodson and others (2002).

Concentration associated with simple hydrolysis of carbonates (SHC)

We assumed that SHC accounts for ~110 µM of the total concentration of HCO₃⁻ as this has been widely reported for chemical weathering under subglacial conditions (Tranter and others, 2002; Wadham and others, 2010). Simple hydrolysis (Eqn 7) occurs during the initial stage of the meltwater and subglacial bedrock/sediment contact, and it plays an important role in chemical weathering (Tranter and Wadham, 2013).



Concentration associated with aerosol hydrolysis of HCO₃⁻

During the process of carbonate weathering, the neutralisation of nitric and sulphuric acid occurs. Each mole of nitrate produces one mole of HCO₃⁻ and each mole of sulphate (atmospherically derived) produces two moles of HCO₃⁻. Atmospheric SO₄²⁻ was deduced from the difference between the total SO₄²⁻ and the sea-salt-derived SO₄²⁻ concentration.

Concentration associated with SO-CD

The presence of sulphide and carbonate minerals in the rocks (although as a minor amount) can dominate weathering because of their nature of rapid dissolution. SO-CD therefore determines the crustally-derived sulphates and carbonates, such that two moles of HCO₃⁻ are produced with each mole of SO₄²⁻.

Concentration associated with carbonation of silicates

It was assumed that the Na and K that are crustally derived (i. e. *Na⁺ and *K⁺) were produced in the meltwater through the carbonation of aluminosilicate minerals. However, this reaction also contributes to the Ca²⁺ and Mg²⁺ fluxes. Therefore, the concentration of HCO₃⁻ derived from the carbonation of aluminosilicate minerals is 3.4 times the (*Na⁺ + *K⁺) concentration (Sharp and others, 1995; Hodson and others, 2000; Table 2) if the weathering environment conforms to global mean ratios of chemical weathering of silicate rocks (after Holland, 1978).

Concentration associated with CC minerals

The CC produces both crustal and atmospherically-derived HCO₃⁻ in equal proportions. The protons associated with nitrates and sulphates were assumed to produce crustally derived HCO₃⁻ only via carbonate weathering, and so the residual concentration represented the sum of SHC (defined above) and carbonate carbonation.

Concentration of HCO₃⁻ associated with atmospheric CO₂

The atmospherically-derived bicarbonate concentration was calculated as all of the silicate-derived HCO₃⁻ and half of the HCO₃⁻ associated with the CC.

Concentration associated with microbial oxidation of carbon

The synthesis of past geochemical data has underscored the potential for CO₂ sources to include organic matter oxidation in subglacial environments (Hodson and others, 2000; Wadham and others, 2010; Horan and others, 2017). However, recent studies have suggested that organic carbon as a solute source may actually be less important in subglacial environments than previously believed (Graly and others, 2017) particularly in silicate catchments typical of the Himalayan basins presented in this study. Additionally, dissolved organic carbon (DOC) is usually low in the Himalayan streams (Panwar and others, 2017). Therefore, we assumed that dissolved inorganic carbon (DIC) generated via organic carbon oxidation is negligible.

Calculation of saturation indices, pCO₂ and sulphate mass fraction (SMF)

Saturation indices were calculated to determine the tendency for dissolution/precipitation of minerals and the partial pressure of CO₂ (pCO₂) in the Himalayan glacierised basins. We used the PHREEQC software using the MINTEQA database (Parkhurst and Appelo, 2013). The concentration of dissolved constituents (P, Ca, K, Mg, Na, H₄SiO₄, SO₄²⁻, HCO₃⁻ and Cl⁻), water temperature and pH were used as

Table 2. Solute concentration of selected Indian Himalayan glaciers in the atmospheric, aerosol, marine and crystal provenance categories

Glacier name	Year of study	Solute component	SO ₄ ²⁻			HCO ₃ ⁻						
			Sea-salt	Aerosol	Crustal	Aerosol hydrolysis	SO-CD	Simple hydrolysis	Silicate derived	Carbonate carbonation	Atmospheric	SO-CD + CC
Gangotri	2008	Pre-monsoon	0.4	3.4	96.1	1.3	32.1	15.8	13.7	15.6	21.5	47.7
		Monsoon	0.3	2.7	97.0	0.7	26.7	15.5	17.1	15.3	24.7	42.0
		Post-monsoon	0.3	2.1	97.6	1.0	26.5	13.4	18.4	14.9	25.9	41.4
	2009		0.2	1.8	98.0	0.5	19.0	9.8	23.6	15.7	31.4	34.7
Dokriani	1994		0.1	0.8	99.1	0.2	24.1	15.7	22.1	10.5	27.4	34.6
Chota Shigri	2003		0.8	6.2	93.0	2.3	19.3	32.0	18.1	6.8	21.5	26.2
	2004		0.6	4.3	95.1	1.7	21.0	25.6	19.2	8.8	23.6	29.8
	2005		0.2	1.5	98.3	1.5	46.9	17.9	14.0	3.9	15.9	50.7
	2006		0.3	2.2	97.5	1.3	40.3	18.9	19.5	0.3	19.7	40.6
	2007		0.2	1.8	98.0	1.2	43.2	17.9	17.7	1.5	18.5	44.7
	2008		0.7	5.1	94.2	3.3	13.9	14.2	27.2	9.5	32.0	23.4
	2009		0.7	5.5	93.8	2.8	15.8	11.3	25.2	13.1	31.7	29.0
Bara Shigri	2012		1.3	10.3	88.4	5.2	15.2	13.5	27.2	7.8	31.1	23.0
	2013		2.4	18.6	78.9	4.1	7.3	28.7	29.8	0.7	29.4	8.0
Patsio	2010		2.4	18.3	79.3	4.8	12.1	34.2	23.8	0.8	24.2	12.9
	2011		0.7	5.2	94.2	0.8	13.2	19.3	26.6	8.9	31.1	22.1
	2012		0.3	2.5	97.2	0.5	12.5	27.1	25.6	5.8	28.5	18.3
Chaturangi			0.2	1.2	98.7	0.4	27.4	7.3	18.6	18.4	27.8	45.8
Satopanth	1989		11.2	85.1	3.7	5.8	0.3	44.8	10.2	19.2	19.8	19.4
	1991		0.0	0.0	100.0	0.0	6.7	16.5	30.0	11.3	35.6	18.0
		Average	1.2	8.9	89.9	2.0	21.2	20.0	21.4	9.4	26.1	30.6

Dataset presents per cent (%) contribution of ion to its average concentration. CH is for Central Himalaya, WH is for Western Himalaya.

input data. The H₄SiO₄ concentration was not available for the Dokriani glacier (Hasnain and Thayyen, 1999). We assumed the water temperature to be 0.01°C for the glacial meltwater (Stachnik and others, 2016b). Additionally, water temperature data were not provided in most previous studies from glacierised basins in the Himalaya. The logarithm of electron activity (pE) of 7.0 was used (Mitchell and Brown, 2008).

The SMF was calculated using Eqn (8). A SMF equal to 0.5 shows the predominance of SO coupled with CD. When the SMF is <0.5, CC increases in importance. An excess of SMF >0.5 suggests additional sources of sulphate originating from the SO coupled to the silicate weathering, carbonate precipitation, or Ca and Mg efflorescent salt dissolution (Cooper and others, 2002; Tranter and others, 2002).

The SMF was calculated using Eqn. (8) as follows:

$$\text{SMF} = \frac{[\text{SO}_4^{2-}]}{[\text{SO}_4^{2-}] + [\text{HCO}_3^-]}, \quad (8)$$

*SO₄²⁻ – the concentration of sulphate, corrected for atmospheric and marine input (in μeq L⁻¹), HCO₃⁻ – the concentration of bicarbonate (in μeq L⁻¹).

Further details of the calculations and normalisation of solute fluxes are presented in the Supplementary Appendix (A).

Atmospheric CO₂ flux calculation

The dataset reported from the Himalayan glacierised basins is further used to calculate the atmospheric CO₂ drawdown. CO₂ consumption was estimated from a flux calculation, requiring the combination of the atmospheric-derived HCO₃⁻ concentration and mean annual runoff. The mean annual runoff values are taken here as specific discharge (water discharge per unit basin area or 'R') of the studied basins.

RESULTS

The precipitation ionic ratios in the Himalayan catchments were used to quantify the atmospheric solute content. The ionic ratios of Na⁺/Cl⁻ and Mg²⁺/Cl⁻ in snow, ice and rain were near those observed in seawater (Supplementary Table S1A, B). Whereas the ratios of K⁺/Cl⁻, Ca²⁺/Cl⁻ and SO₄²⁻/Cl⁻ in snow, ice and rain tended to be orders of

magnitude higher. This suggests a marine and terrestrial contribution of ionic species. Ratios of all the major cations vs Cl⁻ for snow and ice were very similar, whereas the ratios for rain were higher by a factor of 2 and 4 for Mg²⁺/Cl⁻ and Ca²⁺/Cl⁻, respectively (Supplementary Table S1B).

Our calculations show that ~90% of the SO₄²⁻ is derived from crustal SO, while ~9% of the sulphates are formed via aerosols and only 1% via sea salts (Table 2). Similarly, to SO, dissolution of evaporites could also provide a source of Ca²⁺ and SO₄²⁻ but without the concurrent increase in CO₂ release. However, we think CD most likely controls the Ca²⁺ concentration in water due to negative saturation index with respect to calcite (Table 3). Also, evaporite dissolution is small in glacierised basin in the Himalaya comparing to CD and sulphide oxidation (e.g. Sharma and others, 2013; Singh and others, 2015a, b).

The provenance of HCO₃⁻ is complex. The mass-balance calculations presented here show the average value for all the studied glaciers. Aerosol hydrolysis contributes ~2%, SO/CD contributes ~21%, bicarbonates derived through silicates contribute ~21.4%, simple hydrolysis contributes 20%, carbonate carbonation contributes ~9.4%, while HCO₃⁻ derived via the atmosphere contributes ~26.1%. The mean HCO₃⁻ concentration derived via the carbonation process is ~30.6% (Table 2). These values do not accord with the results of Blum and others (1998) as they work further downstream where calcite oversaturation occurs, but quantify that an equal proportion of HCO₃⁻ is released from the weathering of calc-silicate rocks. Many rivers of the Himalayan system are typically oversaturated in terms of calcite, and Ca is being lost in the solid phase (Bickle and others, 2015). Bickle and others (2015) showed that calcite precipitation results in an overestimation of silicate-derived Ca and Mg (and Sr) by a factor of 2.

We therefore checked for calcite saturation using the PHREEQC geochemical model (Table 3). Our results reveal that water is usually undersaturated with respect to carbonate minerals (calcite and dolomite saturation indices <0), halite and gypsum, but oversaturated with respect of quartz (Fig. 3). Carbonate precipitation is therefore unlikely near the Himalayan glacierised basins as the SI for carbonates suggest dissolution rather than precipitation.

Based on the aforementioned solute provenances, the average CO₂ consumption attributed to silicate and

Table 3. Median (in bold) and min–max range of saturation indices and partial pressure of CO₂ (pCO₂) for meltwater from Himalayan glacierised basins

Glacier name	SI calcite	SI dolomite	SI gypsum	SI quartz	SI halite	pCO ₂
Gangotri (N = 12)	-3.5 -6.13; -2.46	-7.4 -12.54; -5.38	-3.1 -4.24; -2.55	0.1 -0.35; 0.59	-10.7 -12.6; -9.99	-2.9 -3.43; -2.15
Dokriani (N = 3)	-3.0 -3.9; -1.11	-7.3 -8.99; -3.84	-2.9 -3.54; -1.91	n.d. n.d.	-11.5 -12.43; -10.77	-2.8 -2.94; -2.57
Chhota Shigri (N = 21)	-2.6 -6.12; -0.29	-5.9 -12.96; -1.17	-3.4 -4.99; -2.61	0.1 -1.6; 0.63	-10.6 -12.83; -9.7	-3.2 -4.36; -2.67
Bara Shigri (N = 6)	-3.5 -4.3; -2.08	-7.8 -9.32; -4.91	-3.5 -4.35; -2.7	-0.6 -0.76; -0.43	-11.1 -11.74; -10.45	-3.0 -3.29; -2.79
Patsio (N = 9)	-1.9 -3.45; -1.25	-4.7 -7.63; -3.25	-2.5 -3.32; -2.34	-0.3 -0.67; -0.1	-10.8 -11.64; -10.44	-2.7 -3.08; -2.15
Chaturangi (N = 3)	-4.2 -4.95; -3.76	-8.9 -10.28; -7.95	-2.6 -2.8; -2.38	0.2 -0.03; 0.39	-10.4 -10.91; -10.13	-2.3 -2.33; -2.32

Sical, Sidol, Sigyp, SIhal, refer to the saturation indices of calcite, dolomite, gypsum and halite, respectively; pCO₂ refers to partial pressure of CO₂; n.d., not determined due to lack of Si concentration in water.

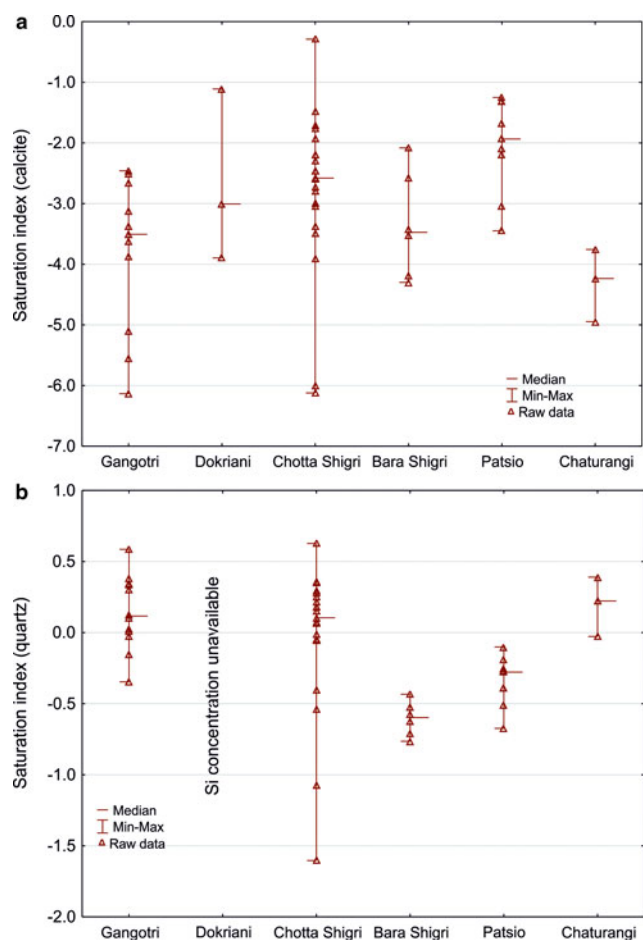


Fig. 3. Saturation indices calculations from PHREEQC software coupled to the MINTEQ database for calcite (a) and quartz (b). Note that y-axis have a different range of parameters.

carbonate weathering was calculated. The consumption of CO₂ through weathering of silicate minerals was found to be nearly $10.6 \times 10^4 \text{ mole km}^{-2} \text{ a}^{-1}$ (19%); whereas $15.7 \times 10^4 \text{ mole km}^{-2} \text{ a}^{-1}$ (28%) for simple hydrolysis reaction; $13.6 \times 10^4 \text{ mole km}^{-2} \text{ a}^{-1}$ (24%) derived from atmosphere; $9.6 \times 10^4 \text{ mole km}^{-2} \text{ a}^{-1}$ (17%) through SO-CD reaction; $5.9 \times 10^4 \text{ mole km}^{-2} \text{ a}^{-1}$ (11%) through carbonate carbonation and $1.4 \times 10^4 \text{ mole km}^{-2} \text{ a}^{-1}$ (2%) by the

aerosol hydrolysis. Table 4 shows variability between the western and central Himalayan glaciers.

DISCUSSION

Glacial catchments have high chemical denudation rates because of high water fluxes and suspended sediment loads (1.2–4 times greater than the global means; Anderson and others, 1997; Tranter and Wadham, 2013). Temperate glaciers in alpine catchments show higher chemical weathering rates due to intense physical weathering and, in our case, a favourable monsoonal climate (Sundriyal and others, 2018). The Himalayan glaciers are governed by strong seasonal discharge (Tipper and others, 2006b; Bookhagen and Burbank, 2010) and rapid retreat (Bolch and others, 2012), contributing to an increase in solute yields and CO₂ consumption. The HHC glaciers receive nearly ~80% of their precipitation from the southwest monsoon system (Bookhagen and Burbank, 2010). A potential change in global climate, such as the weakening of the global monsoon circulation, forest cover reduction, and an increase in aerosol concentration, evapotranspiration process and surface energy budget, has significantly changed the intensity of rainfall and moisture availability in the Himalayan region (Gaffen and Ross, 1999; Duan and Yao, 2003; Ramanathan and others, 2001, 2005). These climatic constraints have ultimately affected the weathering pattern of the region along with the role of temperature, which is behind the feedback mechanism of the climate–weathering relationship (Gabet and others, 2010). An analysis of large rivers indicates that a good correlation exists between CO₂ consumption and silicate weathering flux (Gaillardet and others, 1999) and its consumption in terms of chemical weathering is associated with the tectonic uplift of the Himalaya (Sharp and others, 1995). However, due to uplift, the increase in chemical weathering are not reflected in the long-term (>1 Ma) CO₂ budget, as the calculated CO₂ consumption rate only increases by a factor of 2 (Jacobson and Blum, 2003).

Chemical weathering and non-weathering sources of solute in the Himalaya – regional distribution

The contribution of solute from dissolution of aerosols and aeolian dust in our study is greater than that of other

Table 4. The average atmospheric CO₂ consumption rates attributed to carbonate and silicate weathering and their relative percentage values in selected Indian Himalayan glaciers

Glacier name	Year of study	CO ₂ flux (10 ⁴ mole km ⁻² a ⁻¹)					
		Aerosol hydrolysis	SO-CD	Simple hydrolysis	Silicate derived	Carbonate carbonation	Atmospheric
Dokriani	1994	0.07	8.59	5.59	7.89	3.75	9.76
Chota Shigri	2003	0.43	3.66	6.05	3.42	1.29	4.07
	2004	0.90	11.28	13.73	10.31	4.72	12.67
	2005	0.42	13.31	5.08	3.97	1.10	4.51
	2006	0.62	19.08	8.92	9.23	0.14	9.30
	2007	0.72	25.87	10.74	10.60	0.93	11.07
Patsio	2011	0.24	3.73	5.45	7.52	2.51	8.78
	2012	0.19	4.73	10.28	9.71	2.19	10.81
Satopanth	1989	10.00	0.44	77.13	17.52	32.99	34.02
	1991	0.00	5.79	14.34	26.09	9.86	31.02
Average flux		1.4	9.6	15.7	10.6	5.9	13.6
Per cent contribution		2.4	17.0	27.6	18.7	10.5	23.9

glacierised basins (Wake and Mayewski, 1993). The high SO₄²⁻/Cl⁻ ratio (Supplementary Table S1B) may be attributed to high altitude favouring an increase in SO₄²⁻ aerosol deposition (Polesello and others, 2007), enhanced by atmospheric circulation, domestic biomass burning (Shrestha and others, 2000), or an influx of CaSO₄ (Mayewski and others, 1983). As the sulphate produced is already oxidised, the SO-CD process may therefore be overestimated. Similarly, the great difference between the cation/Cl⁻ ratios in the rain vs those of snow and ice shows that wet and dry deposition may also contribute cations (Shrestha and others, 1997; Shrestha and others, 2000). Dissolution of CaSO₄ aerosol is also accompanied by dissolution of aged sea salts delivered flowing long transport from the sea (Geng and others, 2010), and evaporite deposits from more local stream waters and springs (Evans and others, 2002). However, during the monsoon, aerosols become depleted, whereas during the winter and the pre-monsoon period, the aerosol concentration increases (Shrestha and others, 2000, 2010). Furthermore, additional Cl⁻ from the Himalayan cold and warm springs has been shown to cause high concentrations and saturation with respect to halite (Evans and others, 2002). Meltwater from our glacierised basins show low Cl⁻ concentration and significant undersaturation with respect to halite (Table 3) suggesting that even if high Cl⁻ springs source the surface water, they likely have a little contribution to dissolved solids yields comparing with the bulk runoff from glacier-fed basins.

The cation denudation rates of the Himalayan glacial catchments are 1.2–4 times greater than global mean (Anderson and others, 1997). Considering the lithological aspect, the silicate weathering and particularly plagioclase weathering of the HHC rocks is the most rapid in the Himalaya (West and others, 2002). Previous studies of the Dokriani and Lirung glaciers of the higher Himalaya characteristically represent the HHC lithology releasing high carbonates via weathering fluxes and a proportionately low silicate flux compared with the Bore and Chinnya catchments of the Middle Hills Nepalese Himalaya (West and others, 2002). Our results also show that despite the high carbonate weathering, carbonate precipitation does not appear to occur because of undersaturation with respect to the carbonate minerals (Table 3). This is unlike the other studies that show carbonate precipitation forming travertine deposits in catchments with limited snow/ice cover in the Himalaya (Tipper and others, 2006a). The present study suggests the chemical weathering processes operating in the Himalaya are more complex than previously stated. The chemical composition studies at the Gangotri, Chhota Shigri, Bagni, Kafni, Dokriani and Patsio glaciers show neutral pH at all these glaciers with the control of carbonate weathering and SO as a dominating driving force (Fig. 4). The results presented here elucidate that, except for the Chhota Shigri glacier, the crustally derived sulphate fluxes on average are greater than those derived from other sources (Fig. 4). At the Chhota Shigri glacier, the presence of pyrite in small veins is a major source for SO and so bicarbonates are largely derived from carbonate weathering only and partly from silicate weathering (Sharma and others, 2013). It is interesting to note that the sea salt, aerosol and crustal proportions were different in 2006 and 2007 compared with 2003, 2004, 2005, 2008 and 2009. This difference may have been due to variable flow regime but at this point, it is difficult to explain because of the unavailability of long-term continuous data.

The HCO₃⁻ fluxes presented were derived predominantly from silicates and simple hydrolysis processes in the western Himalayan Glaciers (the Chhota Shigri, Bada Shigri and Patsio glaciers) compared with the central Himalayan glaciers (the Gangotri, Dokriani and Satopanth glaciers) (Fig. 4). The SO-CD-derived HCO₃⁻ fluxes show greater prominence in the central Himalaya as well as more carbonate carbonation. Overall, carbonate carbonation and SO-CD in the Himalayan system are clearly contributing larger HCO₃⁻ fluxes than the silicate-derived fluxes.

SO and silicate weathering

Glaciers of the HHC catchments (the Chhota Shigri, Patsio, Dokriani and Lirung glaciers) highlight the importance of the SO process as a primary driver of sulphates in meltwater (Fig. 4) as discussed in previous studies (West and others, 2002; Sharma and others, 2013; Singh and others, 2015a, b). SO creates strong sulphuric acid that may be associated with both carbonate and silicate weathering, leading to a possible decrease in the importance of long-term CO₂ consumption associated with the latter. The coupling of SO and silicate weathering under a subglacial drainage system suggests a long residence time for the water (Wadham and others, 2010). High saturation indices with respect to quartz suggest both dissolution and precipitation occur in the glacierised basins (Table 3). This agrees with other studies from the Yukon stating that both precipitation and dissolution reactions control silicate fluxes from subglacial conditions (Crompton and others, 2015).

SO-CD is an important process that releases CO₂ into the hydrological system, thereby offsetting the drawdown of atmospheric CO₂ by carbonation. The SO-CD reaction is therefore identified as a transient source of CO₂ to the atmosphere (Torres and others, 2014, 2017). Closer observation of bicarbonate and sulphate solute fluxes indicates a negative correlation ($r = -0.42$) between crustally derived SO₄²⁻ and hydrolysis-derived HCO₃⁻ and between SO-CD and silicate-derived HCO₃⁻ ($r = -0.63$) for all of the studied glaciers (Supplementary Table S4). Figure 4 shows that as the HCO₃⁻ produced from SO-CD becomes low, more silicate carbonation occurs consuming the CO₂ produced via the SO-CD reactions (Eqns 1 and 2) in the subglacial environment. This indicates an excess of CO₂ release from SO-CD being balanced with CO₂ consumption via carbonation during the silicate weathering process. However, available meltwater chemistry provenance models (e.g. Sharp and others, 1995; Hodson and others, 2000), which we have used in our study, appear not to include that source of CO₂ in glacierised basins. We propose that further works on solute provenance in glacierised catchments should include CO₂ release associated with SO-CD.

Comparing our study to those from different parts of the world (e.g. the Mackenzie River in Canada by Calmels and others, 2007; Liwa River in Taiwan by Das and others, 2012) also links silicate weathering and SO to the CO₂ release. The Mackenzie River weathering study estimates 62% of the carbonate weathering is via sulphuric acid (Calmela and others, 2007). The high SO rates at the Liwa River (Das and others, 2012) and the importance of SO coupled with carbonate carbonation reaction (Stachnik and others, 2016a, b) also indicate the likely importance of sulphides for simulating CO₂ release.

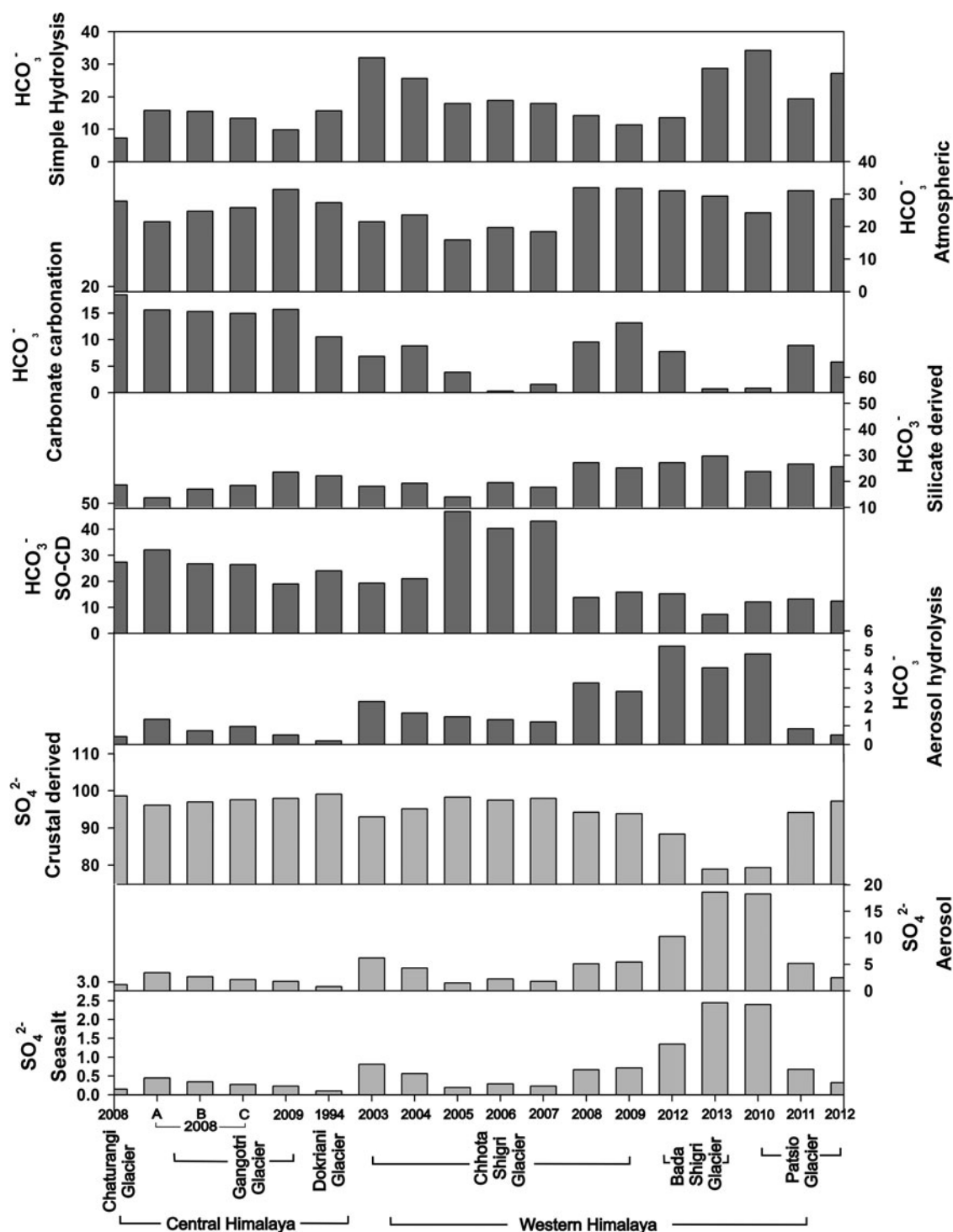


Fig. 4. Estimated solute fluxes of selected central Himalayan glacierised catchments showing relationship of partitioned atmospheric and crustal components. Provenance categories represent here the per cent (%) contribution of respective solute flux (estimated through Table 2). (a), (b) and (c) denote the pre-monsoon, monsoon and post-monsoon fluxes, respectively.

Carbon dioxide consumption in the Himalayan region

Geographical and geological features of a catchment have a pronounced effect on weathering in glacial environments (Hodson and others, 2000; Tranter and Wadham, 2013). A comparison of global CO₂ consumption rates attributed solely to Ca-Mg of silicate weathering is $\sim 9.0 \times 10^4$ mole km⁻² a⁻¹ (Gaillardet and others, 1999) and in the New Zealand Southern Alps 6.9×10^4 mole km⁻² a⁻¹ on the eastern side and 14×10^4 mole km⁻² a⁻¹ on the western side (Jacobson and Blum, 2003). This is similar to the

present study of CO₂ consumption in the Himalaya estimated from different concentrations (Table 4). This contribution of silicate weathering to CO₂ consumption in the Himalaya is similar to the average global CO₂ consumption through silicate weathering. Considering the study of the Indian Himalayan rivers (Krishnaswami and Singh, 2005) in the Ganga River catchment, the total consumption of atmospheric CO₂ from the weathering of silicate rocks was found to be 4×10^5 mole km⁻² a⁻¹, nearly ten times greater than the values of this study. This is because the bulk chemistry

of the large rivers reflects their silicate lithology and therefore underestimates the importance of carbonate weathering.

However, in our first-order catchments, we are not considering the production/burial or respiration/oxidation of organic matter and associated CO₂ released into the atmosphere in the watershed as well as the secondary mineral precipitation that is prevalent in the larger Himalayan river systems.

CONCLUSION

The findings of this study provide new insights into carbon cycling related to CO₂ consumption via weathering. We have demonstrated the consumption and/or release of CO₂ coupled to SO is equally responsible for regulating CO₂ cycling. Geochemical modelling suggests that the water is usually undersaturated with respect to carbonate minerals (calcite and dolomite), halite and gypsum but is oversaturated in terms of quartz. High saturation indices with respect to quartz suggest both dissolution and precipitation and therefore intensive carbonation of silicates via atmospheric CO₂. SO creates strong acid that may be associated with silicate weathering leading to a possible decrease in the importance of long-term CO₂ consumption via carbonation of silicates via atmospheric CO₂. By contrast, the saturation indices for carbonates suggest dissolution rather than precipitation producing high solute Ca²⁺ and SO₄²⁻ fluxes typical for glacial environments on account of the importance of SO coupled to carbonate weathering. We, however, postulate that in the Himalaya and Southern Alps silicate weathering and the SO-CD reaction are exchanging CO₂ and suggest that further consideration of CO₂ release from the SO-CD process could provide a better understanding of global CO₂ consumption by weathering.

Previous attempts have focused on long-term consumption (~millions of years) of CO₂ via silicate carbonation, but these show little influence is likely in context of future climate change scenarios because of its low importance as a weathering mechanism. However, chemical weathering of carbonates also causes CO₂ consumption, but this process is short-term (~thousands of years), making it more appropriate to include in climate change models. We believe that the potential of SO weathering reaction findings will be particularly helpful for understanding the present-day solute acquisition process in glacierised catchments of the Himalaya and invaluable for establishing how the long-term carbon cycle of the Earth is balanced.

SUPPLEMENTARY MATERIAL

The supplementary material for this article can be found at <https://doi.org/10.1017/aog.2019.5>

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