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Pyrite oxidation flux underestimated due to landscape-mediated microbial sulfate reduction

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Abstract

Oxidative weathering of pyrite consumes O_2 , generates dissolved sulfate (SO_4^{2-}) and acidifies carbonates to release CO_2 to the ocean-atmosphere system. Mountain belts are hotspots of chemical weathering and geological carbon cycling, but understanding the net role of orogenesis in modulating the evolution of atmospheric CO_2 and O_2 levels requires robust constraints on weathering pathways such as pyrite oxidation. Sulfur and water-normalized oxygen isotopic compositions of riverine SO_4^{2-} ($\delta^{34}S_{SO_4^{2-}}$ and $\Delta^{18}O$) are potentially powerful tracers to quantify pyrite oxidation, but they may be overprinted by microbial sulfate reduction (MSR)—a process often overlooked in terrestrial environments. To address this, we generated paired geomorphological, geochemical, and microbiological data from the headwaters of the Ganga River in the Himalayas. Our results indicate that MSR overprinting is maximized when low erosion rates are coupled to relatively low precipitation rates, revealing a landscape-mediated hydrological control on microbial terrestrial sulfur cycling. By applying this insight to global compilations of riverine $\delta^{34}S_{SO_4^{2-}}$, $\Delta^{18}O$ and major ion chemistry, we show that MSR overprinting leads to systematic shift in riverine sulfate isotopic signals, implying that global CO_2 emissions and O_2 consumption fluxes from pyrite weathering may be up to double previous isotope-based estimates.

Keywords: Sulfur Cycle, Microbial Sulfate Reduction, Pyrite Oxidation, Weathering and Erosion

Introduction

On timescales $\gtrsim 10^6$ years, atmospheric CO_2 levels (p CO_2) are governed by the balance between geologic sources—i.e., metamorphic degassing and oxidative weathering of pyrite and rock-derived organic carbon (OC_{petro})—and sinks—i.e., burial in marine sediments of pyrite, biospheric organic carbon (OC_{bio}) and carbonates whose formation is promoted by silicate rock weathering¹. Of particular interest here, oxidative pyrite weathering² generates sulfuric acid (H_2SO_4), which deprotonates to sulfate (SO_4^{2-}), acidifies

carbonate rocks, and releases CO_2 into the ocean-atmosphere system^{3,4}. Further, by also consuming atmospheric O_2 , pyrite oxidation represents a key process at the nexus of the global carbon, oxygen and sulfur biogeochemical cycles².

Mountain-building—with its tectonically driven crustal exhumation, steep topography and orographic precipitation—produces geologic carbon cycling hotspots that are traditionally considered net CO_2 sinks due to their elevated silicate weathering and OC_{bio} export fluxes^{5,6}. However, recent studies suggest that CO_2 emissions from metamorphic degassing⁷, pyrite weathering^{4,8} and OC_{petro} oxidation^{9,10} can approach and even exceed silicate weathering- and OC_{bio} export-driven CO_2 drawdown in mountain catchments^{7,11}. Beyond pCO_2 , any imbalance in pyrite oxidation relative to its burial in marine sediments also drives increases or decreases in atmospheric O_2 levels (pO_2)¹². Assessing the impact of mountain building on pCO_2 and pO_2 evolution therefore requires accurate constraints on individual source and sink fluxes, particularly pyrite weathering, which has historically received less attention than silicate weathering or OC cycling.

Terrestrial pyrite weathering can be quantified using dissolved riverine SO_4^{2-} fluxes, provided that other SO_4^{2-} sources—most importantly, dissolution of evaporites such as gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), which does not impact pO_2 or pCO_2 —are accurately unmixed. Relative source contributions are traditionally resolved using SO_4^{2-} stable-isotope compositions ($\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$; *Methods*)^{13–15}. Because SO_4^{2-} in evaporite minerals is typically ^{34}S - and ^{18}O -enriched relative to that derived from pyrite oxidation^{16,17}, elevated riverine $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ values are canonically attributed to higher relative evaporite contributions and *vice versa*^{13,14}. Assuming that $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ behave conservatively in major river systems, this approach predicts that oxidative pyrite weathering contributes only ~40-60% of total weathering-derived riverine SO_4^{2-} exported globally^{13,14}.

However, recent work suggests that riverine $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ may not conservatively track SO_4^{2-} sources due to overprinting from microbial sulfate reduction (MSR) in soils and aquifers^{18,19}. During MSR, SO_4^{2-} is reduced to sulfide (H_2S) through a series of reversible reactions that fractionate ^{34}S and ^{18}O (Fig. 1)^{20–23}. Since energetically unfavorable (i.e., small negative ΔG) terrestrial conditions would promote greater reversibility^{20,22,23}, H_2S is nearly quantitatively re-oxidized to SO_4^{2-} , thus dampening shifts in $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$. Nevertheless, equilibrium isotope exchange with intra-cellular water leads to ^{18}O enrichment in intermediate SO_3^{2-} ^{24,25} that is retained after re-oxidation to SO_4^{2-} . MSR could thus significantly increase riverine $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$, with or without a corresponding increase in $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$.

Still, quantifying MSR using geochemistry alone is challenging because its isotopic signature overlaps with that of evaporite dissolution¹⁸. Additionally, while MSR appears to be more prominent at lower elevations^{18,19}, little is known about its mechanistic controls—i.e., its quantitative response to landscape properties and hydroclimate. Orogenic weathering hotspots are marked by sharp gradients in physical erosion, temperature, and precipitation, which may drive large shifts in weathering pathways and river chemistry over relatively small spatial scales. Overall, the role of MSR in modulating riverine SO_4^{2-} isotope signals as well as its underlying geologic and climatic controls remain poorly understood.

To provide new insight, we report $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ from a suite of rivers spanning an erosional and climatic gradient in the Himalayan headwaters of the Ganga Basin. We integrate these results with concomitant measurements of major ion chemistry, ^{10}Be -derived erosion rates, and microbial marker gene abundances to independently quantify MSR overprinting and constrain how it responds to catchment geomorphology and hydroclimate. Finally, by extending this framework to global river chemistry compilations, we suggest that MSR is as important as lithological variability (i.e., pyrite weathering vs. evaporite dissolution) in controlling SO_4^{2-} isotope compositions, and we explore implications for global pyrite weathering flux—and thus CO_2 production and O_2 consumption—predictions.

Tectonic setting & study area

We investigated the Alaknanda and Bhagirathi catchments in the headwaters of the River Ganga in the Northwest Himalayas (Fig. 2). In addition to pyrite oxidation^{8,26}, these and neighboring catchments have been studied extensively to quantify silicate weathering^{27–29}, metamorphic degassing^{30,31}, OC_{petro} oxidation³², and OC_{bio} export fluxes^{11,33}, thus providing a robust geochemical framework to interpret results generated here. Moreover, despite covering a small geographic area, the Alaknanda and Bhagirathi basins exhibit significant hydroclimatic and geologic heterogeneity, making them an ideal case study for investigating MSR controls. Samples collected here represent catchment-averaged elevations of ≈ 1250 – 5300 m, slopes of 25 – 33° , mean annual temperatures (MAT) of 1 – 19°C , mean annual precipitation (MAP) of ≈ 750 – 1800 mm yr^{-1} , and glaciated area coverage of 0 – 36% (*Methods*). Moving from north to south, these catchments are underlain by (i) unmetamorphosed siliciclastic and carbonaceous sedimentary rocks of the Tethyan Sedimentary Series (TSS); (ii) high-grade gneisses, schists, granitoids and metacarbonates of the Higher Himalayan Crystalline Series (HHC); and (iii) low-to-medium grade schists, metasediments and dolomites of the Lesser Himalayan Series (LHS) (Fig. 2). The only riverine $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ values from these catchments reported to date range from -2% to $+10\%$, with lower values attributed to sedimentary and magmatic pyrite oxidation and higher values to evaporite dissolution, without considering the possibility of MSR²⁶. Crucially, despite this interpretation, no evaporite-bearing formations have been mapped in this region, but pyrite would be hosted throughout both basins within interbedded shales (TSS and LHS lithologies) and as magmatic sulfides (HHC lithologies).

Pyrite oxidation as the primary SO_4^{2-} source

In addition to pyrite oxidation and evaporite dissolution, riverine SO_4^{2-} in mountain catchments can be derived from precipitation and/or release of carbonate-associated sulfate during carbonate rock weathering. To unmix different end-member contributions in the Ganga headwaters, we first correct major ion concentrations (Na^+ , K^+ , Ca^{2+} , Mg^{2+} , SO_4^{2-} , Cl^-) for precipitation inputs (*Methods*)^{29,34}. As Himalayan precipitation is characterized by low SO_4^{2-} concentrations ($6.9_{-6.9}^{+7.4}$ μM ; $n = 29$; $\mu \pm 1\sigma$ throughout) and SO_4^{2-} to Cl^- molar ratios (0.5 ± 0.3 ; $n = 29$)³⁵, precipitation corrections typically don't exceed $\sim 10\%$ of dissolved riverine SO_4^{2-} concentrations (Table S1). We then employ an inversion-based lithology partitioning model (*Methods*). Crucially, our model only considers precipitation-corrected major ion concentrations as inputs; we do not include $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ or $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ as conservative tracers since our intention is to explicitly assess whether MSR modifies these values. Lithological end members considered here include: (i) granites and gneisses, (ii) shale, (iii) carbonate, (iv) pyrite and (v) evaporites.

We include pyrite as a separate end-member to ensure that the model is agnostic to the lithology in which it is hosted. This enables direct quantification of pyrite oxidation and avoids potential biases arising from incongruent weathering. Although evaporites have not been mapped in our study region, we nevertheless include this end-member in our inversion to be as conservative as possible when determining pyrite-weathering contributions to riverine SO_4^{2-} . Specifically, the presence of hot springs with high Cl^- concentrations implies halite (NaCl) dissolution, which could indicate deep brines or buried evaporite-bearing sequences. While hot-spring SO_4^{2-} concentrations are statistically positively correlated with those of Cl^- ($[\text{SO}_4^{2-}] = (0.04 \pm 0.02) \times [\text{Cl}^-] + (268.53 \pm 84.05)$; $R^2 = 0.48$; $p < 0.001$; Fig. S1a), the small slope and large intercept of this relationship suggest weak coupling and a substantial Cl^- -independent SO_4^{2-} component that is not derived from evaporite dissolution. Furthermore, hot-spring SO_4^{2-} and Ca^{2+} concentrations are only weakly correlated (Fig. S1b), suggesting that the Cl^- -independent SO_4^{2-} component is derived from pyrite oxidation rather than gypsum dissolution. This interpretation is supported by $\text{SO}_4^{2-}/\text{Cl}^-$ molar ratios, which are markedly higher in rivers (10.4 ± 7.9 ; $n = 26$; Table S1).

relative to hot springs ($0.3^{+1.0}_{-0.3}$; $n = 36$; Fig.S1a), further indicating a catchment-scale decoupling between SO_4^{2-} and Cl^- sources.

Even applying this conservative approach, mixing-model results indicate that pyrite oxidation is the dominant SO_4^{2-} source in all Ganga headwater samples analyzed here; f_{py} , the estimated fraction of SO_4^{2-} derived from pyrite weathering, ranges from ~ 0.73 to 1.0 (0.964 ± 0.004 ; $n = 26$; Table S1; Fig. 2). These results contrast with previous ^{34}S -based interpretations from some of the same catchments²⁶, where tributaries with $\delta^{34}\text{S}_{\text{SO}_4^{2-}} \geq 5\text{‰}$ were interpreted to represent evaporite dissolution. However, given the mixing model-predicted dominance of pyrite oxidation generated here, we instead explore the possibility that $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ in the Alaknanda and Bhagirathi catchments primarily reflect pyrite oxidation and MSR overprinting.

MSR modulation of sulfate isotopic signals

Ganga headwater $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ measured here ranges from -8.5 to 11.3‰ ($1.6 \pm 4.5\text{‰}$; $n = 26$) and -8.1 to 5.2‰ ($-4.0 \pm 3.2\text{‰}$; $n = 26$; Table S1; Fig. 3a). Our $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ values are lower than those previously reported from limited, low-elevation sites in these basins (3.4 ± 3.0 ; $n = 24$)²⁶. Because inversion model-derived $f_{\text{py}} \approx 1$ across our sample set—and because ^{34}S fractionation during pyrite oxidation is negligible^{36,37}—observed $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ variability likely reflects isotopic differences of weathered pyrite and/or MSR overprinting. Indeed, reported $\delta^{34}\text{S}$ for both Himalayan sedimentary and magmatic pyrite is highly variable, spanning -50 to $+10\text{‰}$ and -10 to $+10\text{‰}$ ¹⁸, and encompasses the $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ range observed here. Elevated $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ alone therefore does not require MSR.

In contrast, elevated $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ —despite predominantly pyrite-weathering derived SO_4^{2-} —strongly suggests re-oxidation of ^{18}O -enriched SO_3^{2-} (Fig. 1). Laboratory experiments indicate that oxygen atoms in SO_4^{2-} produced by pyrite oxidation are primarily derived from meteoric water ($\delta^{18}\text{O}_{\text{H}_2\text{O}} = -9.1$ to -13.6‰ in our study region), with $\sim 8\text{--}13\%$ possibly sourced from dissolved O_2 ($\delta^{18}\text{O}_{\text{O}_2} = +23.5\text{‰}$) in well-oxygenated environments^{36,38}. Because meteoric water becomes progressively ^{18}O -depleted with increasing elevation³⁹, we report $\delta^{18}\text{O}$ offsets between SO_4^{2-} and river water in which it is dissolved ($\Delta^{18}\text{O} = \delta^{18}\text{O}_{\text{SO}_4^{2-}} - \delta^{18}\text{O}_{\text{H}_2\text{O}}$; Table S1). Accounting for $\text{SO}_4^{2-}/\text{H}_2\text{O}$ and $\text{SO}_4^{2-}/\text{O}_2$ kinetic isotope effects³⁶, we predict primary pyrite-oxidation $\Delta^{18}\text{O}$ of $\sim 3.5\text{--}6.5\text{‰}$ in the Ganga headwaters. However, observed $\Delta^{18}\text{O}$ ranges from 4.6 to 14.3‰ (8.1 ± 2.2 ; $n = 26$; Fig. 3a), reaching substantially higher values than are possible during pyrite oxidation alone and requiring secondary processes to enrich ^{18}O .

In most landscapes, pyrite oxidation is largely confined to pores and micro-fractures along a subsurface weathering front⁴⁰. When combined with O_2 -dependent oxidation rates⁴¹, calculated in-situ pyrite oxidation fluxes suggest that these pores and micro-fractures contain low nanomolar dissolved O_2 concentrations. Therefore, pyrite oxidation itself, as well as plant respiration and/or microbially mediated organic matter decomposition, would rapidly consume O_2 and result in the formation of local anoxic micro-sites. We hypothesize that sulfate-reducing bacteria are present in these micro-sites, where reversible intra-cellular reactions cause post-weathering ^{18}O enrichment via SO_3^{2-} re-oxidation (Fig. 1).

To evaluate this hypothesis, we measured abundances of the *dsrB* marker gene (*Methods*)—which encodes the β -subunit of the dissimilatory sulfite reductase enzyme that catalyzes SO_3^- reduction to H_2S during MSR⁴². *dsrB* is detected across our elevation and erosional gradient (Table S1) and is positively correlated with $\Delta^{18}\text{O}$ ($R^2 = 0.46$, $p < 0.05$; Fig. 3b); this suggests a quantitative link between sulfate reducer abundance and oxygen-isotope overprinting, likely due to high intra-cellular MSR reversibility and thus SO_3^{2-} re-oxidation. Furthermore, to account for the influence of the sulfate reservoir and microbial community sizes on $\Delta^{18}\text{O}$ overprinting, we normalized $[\textit{dsrB}]$ to measured SO_4^{2-} concentrations ($202.6 \pm 122.5\text{ }\mu\text{M}$; $n = 26$; Table S1) and to 16S rRNA gene abundances ($25.7^{+87.7}_{-25.7} \times 10^4$ copies L^{-1} ; $n = 21$; Table S1), a proxy for total prokaryotic (bacterial and archaeal) abundance. Resulting $[\textit{dsrB}]/[\text{SO}_4^{2-}]$

ratios exhibit a stronger correlation with $\Delta^{18}\text{O}$ ($R^2 = 0.71$, $p < 0.001$; Fig. 3c) than un-normalized [*dsrB*], whereas [*dsrB*]/[16S rRNA] is negatively correlated with $\Delta^{18}\text{O}$ ($R^2 = 0.42$, $p < 0.05$; Fig. S2). Together, these findings indicate that the extent of $\Delta^{18}\text{O}$ overprinting is more strongly driven by sulfate reducer abundance relative to sulfate supply rather than their relative abundance within the overall microbial community.

Importantly, our interpretation that MSR occurs in close proximity to weathered pyrite is supported by isotope-mass balance calculations. For example, in rivers draining the Peruvian Andes, Ref. ¹⁵ concluded that SO_4^{2-} is conservatively transported across the mountain front with no recycling in floodplains ¹⁵. Although the absence of robust discharge measurements precludes similar calculations here, Bhagirathi main-stem samples collected along a transect containing no major tributaries (Fig. 2) exhibit consistent $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\Delta^{18}\text{O}$ values, ranging from 0.0 to 0.6 ‰ and 6.5 to 8.3‰. Together with Peruvian Andes results ¹⁵, this implies that overprinting occurs near the zone of pyrite weathering rather than further downstream in floodplains and riparian zones.

Still, in addition to sulfate reducers, *dsrB* can be found in aerobic sulfur oxidizers (e.g., *Chlorobi* and *Proteobacteria* species that oxidize H_2S , polysulfides, thiosulfate, or elemental sulfur to SO_4^{2-}) ⁴³. Similar to MSR, microbial sulfur oxidation (MSO) also results in $\Delta^{18}\text{O}$ overprinting. While these functions cannot be separated by the marker-gene approach employed here, $\Delta^{18}\text{O} \gtrsim 6.5\text{‰}$ is mainly observed in samples with $[\text{SO}_4^{2-}] \lesssim 200 \mu\text{M}$ (Table S1), the maximum value possible if all dissolved O_2 in initially air-equilibrated water were consumed by pyrite oxidation (i.e., closed-system dynamics) ⁴⁴. Because closed-system dynamics lead to anoxia, MSR and MSO (i.e., upon re-exposure of reduced sulfur to oxygen) would collectively contribute to $\Delta^{18}\text{O}$ overprinting in such settings. In contrast, $[\text{SO}_4^{2-}] \gtrsim 200 \mu\text{M}$ requires open-system O_2 transport to the pyrite-mineral surface, likely maintaining (sub)oxic conditions that inhibit both MSR and the availability of reduced sulfur for MSO, thereby limiting $\Delta^{18}\text{O}$ overprinting.

Geomorphic and hydrologic controls on MSR

Since MSR occurs in subsurface, anoxic environments, its activity should respond to geomorphic and hydrologic controls that promote long fluid transit times—which average ~ 45 days in Himalayan catchments ⁴⁵—and thus prolonged exposure to O_2 -consuming processes. Such controls include erosion rate ⁴⁶ and precipitation ⁴⁷, with steep, wet catchments described by short fluid transit times and *vice versa*. To quantitatively elucidate the relationship between geomorphology, MSR and $\Delta^{18}\text{O}$ in the Ganga headwaters, we estimated catchment-averaged erosion rates at each site by regressing k_{sn} , the normalized steepness index, to in-situ ^{10}Be -derived erosion rates measured at a subset of our sample sites (*Methods*; Fig. S3) ⁴⁸. We find that $\Delta^{18}\text{O}$ is negatively correlated with the logarithm of erosion rate ($R^2 = 0.55$; $p < 0.01$), with $\Delta^{18}\text{O} \gtrsim 8 \text{‰}$ only observed in streams draining catchments with $\lesssim 0.1 \text{ mm yr}^{-1}$ erosion (Fig. 4a). This relationship is corroborated by microbiological evidence, as high [*dsrB*] (both with and without $[\text{SO}_4^{2-}]$ normalization) coincides with low erosion rate (Fig. 3), suggesting that MSR—and thus elevated $\Delta^{18}\text{O}$ —is more prominent in less erosive landscapes. Furthermore, $\Delta^{18}\text{O}$ increases with mean annual temperature (MAT; $R^2 = 0.71$, $p < 0.001$; Fig. S4a) but decreases with glacial coverage ($R^2 = 0.59$, $p < 0.001$); the latter relationship may be driven by glacially enhanced erosion (Fig. S4b). Unlike for $\Delta^{18}\text{O}$ however, no statistical relationship exists between erosion rate and $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ (Fig. S5), likely due to high pyrite $\delta^{34}\text{S}$ variability ¹⁸ and sulfur re-oxidation dampening the expression of MSR ^{34}S fractionation.

The dependence of MSR and $\Delta^{18}\text{O}$ on erosion rate can be understood in terms of mechanisms that promote long transit times and anoxia in groundwater, which in the Himalayas is stored in fractured bedrock and discontinuous aquifers within porous alluvial sediments and sedimentary rocks ⁴⁵. In low-erosion catchments, transit times are also prolonged by the presence of thick, unsaturated regolith and

soils—i.e., the vadose zone—whose infiltration rates exceed precipitation and which would serve as a temporal buffer between precipitation, groundwater storage and runoff generation^{49,50}. Furthermore, warm and wet catchments with well-developed critical zones also support vegetation cover, OC-rich soils and heterotrophic microbial activity, thus accelerating dissolved O₂ consumption and promoting anoxia. In contrast, glaciated and high-erosion catchments are characterized by thin regolith, low vegetation cover and a (partly) crystalline basement (Fig. 2) that readily fractures in tectonically active settings, enhancing subsurface oxygenation⁵¹. Together, these factors limit the prevalence of anoxic conditions necessary to support MSR. However, overall microbial activity and vegetation cover are also inhibited in such landscapes, potentially increasing the *relative* importance of metabolic pathways such as MSR within limited available anoxic sites and resulting in some of the highest [*dsrB*]/[16S rRNA] observed here (Fig. S2). Still, the impact of MSR on $\Delta^{18}\text{O}$ overprinting in glaciated, high-erosion catchments with thin regolith and low vegetation cover is limited since the absolute abundance of sulfate-reducing bacteria is minimal.

Importantly, unlike with erosion rate or MAT, $\Delta^{18}\text{O}$ does not change monotonically with MAP (Fig. 4b). While $\Delta^{18}\text{O}$ does increase with MAP in drier, high-erosion catchments ($R^2 = 0.39$, $p < 0.2$) and across the dataset as a whole ($R^2 = 0.66$, $p < 0.001$), we observe the opposite trend in wetter, lower erosion catchments ($R^2 = 0.79$, $p < 0.05$). We suggest that this reversal reflects the vadose zone’s role in buffering fluid transport. Although decreasing erosion generally increases water transit times⁴⁶, deeper fractures and pore spaces could become saturated in low-erosion catchments characterized by high MAP, thereby activating shallow flow paths. Water transit times become shorter as a result⁵², reducing the time available for fluids to become anoxic and thus the likelihood of MSR. In contrast—beyond exhibiting overall lower MAP in our dataset (Fig. 2)—high-erosion landscapes contain less developed regolith, diminishing critical zone flow-path reorganization in response to porewater and fracture saturation.

Overall, we decipher a landscape-mediated hydrologic control on MSR, whereby thick, well-developed critical zones in low-erosion landscapes yield longer water transit times, increased dissolved O₂ consumption by respiration and a greater likelihood of MSR relative to high-erosion landscapes. In contrast, increased precipitation saturates pores, activates shallow fluid flow paths and results in shorter water transit times, decreasing dissolved O₂ consumption and the likelihood of MSR. These competing forcings lead to MSR-induced $\Delta^{18}\text{O}$ overprinting being maximized where low physical erosion rates are coupled to low-to-moderate MAP—a combination that prolongs sub-surface water transit times (Fig. 4b).

Implications for global pyrite oxidation flux estimates

To assess the global importance of terrestrial MSR, we compiled a dataset of simultaneous $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\Delta^{18}\text{O}$ and major ion chemistry from relatively pristine rivers with minimal anthropogenic input ($n = 393$ samples; *Methods*; Table S2). We find a positive correlation between $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\Delta^{18}\text{O}$ (Fig. 4c); such a relationship has been observed previously^{3,13,18} and is typically used to infer the relative contributions of pyrite oxidation and evaporite dissolution to riverine SO_4^{2-} . For example, Ref. ¹⁴ utilized $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ values and major ion chemistry of global rivers to predict that evaporite dissolution contributes to ~35% of global terrestrial SO_4^{2-} flux. After correcting the remaining “excess” SO_4^{2-} for anthropogenic⁵³, volcanic⁵⁴, and biogenic contributions⁵⁴, they estimated a global pyrite oxidation-derived SO_4^{2-} flux of ~1.3 Tmol yr⁻¹. Ref. ¹³ similarly applied a $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ vs. $\Delta^{18}\text{O}$ dual-isotope binary mixing model to estimate pyrite weathering contributions; using their end-member compositions, this approach predicts an average $f_{\text{py}} \sim 0.58$ for the global data compiled here (Fig. 3a). Assuming this dataset represents global rivers, this translates to a pyrite weathering-derived SO_4^{2-} flux of ~1.6 Tmol yr⁻¹.

However, here we argue that such unmixing overestimates evaporite contributions in the absence of explicit MSR constraints. We demonstrate this by subjecting the compiled ion chemistry data to

the same inversion-based lithology partitioning model used for Ganga headwater samples (allowing for site-specific variability in end-member compositions; *Methods*). Model results reveal that rivers whose dissolved SO_4^{2-} is dominated by pyrite weathering—here defined as $f_{\text{py}} \geq 0.8$ ($\sim 90\%$ of the dataset, see Fig. S6 for sensitivity test)—span the entire $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ vs. $\Delta^{18}\text{O}$ range (Fig. 4c). Thus, in addition to evaporite contributions, increases in $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\Delta^{18}\text{O}$ likely reflect variable extents of MSR-induced isotopic fractionation in different geomorphic and hydro-climatic settings, particularly in rivers where SO_4^{2-} is dominated by pyrite-weathering inputs. This result implies that using $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and/or $\Delta^{18}\text{O}$ values alone to partition SO_4^{2-} sources may systematically underestimate pyrite oxidation in modern weathering environments.

In contrast, by using a major-ion inversion model that explicitly includes pyrite oxidation as an end-member, here we suggest that f_{py} averages ~ 0.90 in the compiled dataset (Fig. 4c). Still, because studies included in our dataset that measure sulfate stable-isotope compositions may systematically target catchments dominated by pyrite oxidation, we also inverted the major ion chemistry of all stations from the Global River Chemistry (GLORICH) database⁵⁵ with ≥ 10 data points located in mountain catchments with low anthropogenic activity ($n = 235$ sites; see *Methods* for filtering criteria and inversion details). The filtered GLORICH database inversion—which displays broader geographical coverage than the sulfate stable-isotope containing dataset—predicts a slightly lower f_{py} value of 0.72 ± 0.17 (Fig. S7). Still, combining either the simultaneous $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\Delta^{18}\text{O}$ and major ion chemistry compilation or the screened GLORICH dataset—which we assume span the possible range of global f_{py} values—with recent total riverine SO_4^{2-} export estimates¹⁴ yields a pyrite weathering-derived SO_4^{2-} flux of 2.0 ± 0.5 to $2.5 \pm 0.3 \text{ Tmol yr}^{-1}$, which is ~ 1.5 to $\sim 1.9\times$ higher than previous estimates anchored to $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and/or $\Delta^{18}\text{O}$.

Nevertheless, this inversion-based pyrite oxidation flux estimate remains smaller than that predicted for pyrite burial in modern marine sediments ($\sim 7.0 \text{ Tmol yr}^{-1}$)¹². The global sulfur cycle today, therefore, likely results in a net atmospheric CO_2 consumption and O_2 accumulation, albeit with a smaller magnitude than previously estimated. On geologic timescales, the net impact on atmospheric O_2 would be tectonically regulated—reflecting the balance between continent-continent collision, which enhances the exhumation and oxidative weathering of pyrite-bearing sedimentary rocks, and flooded continental shelf area, the primary locus of pyrite burial. On the other hand, because CO_2 sources and sinks are finely balanced in mountain catchments, pyrite oxidation fluxes being systematically underestimated implies that mountain-building processes represent larger sources of atmospheric CO_2 than is currently believed. Taken together, we show that MSR is as important as lithology in determining the isotopic composition of terrestrially derived SO_4^{2-} , and we argue that future geologic carbon-budget reconstructions need to explicitly consider the role of MSR when quantifying pyrite oxidation fluxes.

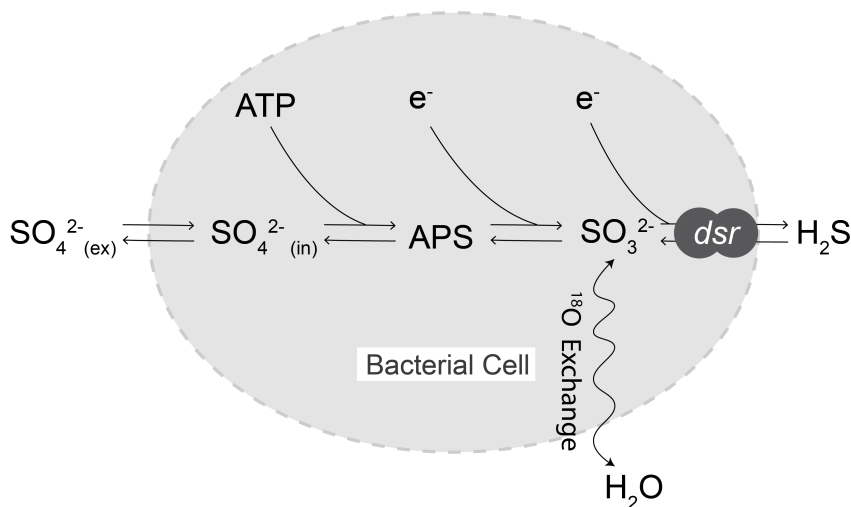


Fig. 1 | Schematic Diagram of the reversible MSR chain reaction. Importantly, this includes potential for equilibrium oxygen-isotope exchange between SO_3^{2-} and intracellular (meteoric) water. The SO_3^{2-} reduction step is catalyzed by dissimilatory sulfite reductase, whose marker gene (*dsrB*) is quantified in this study. Modified after Refs. [20,22](#)

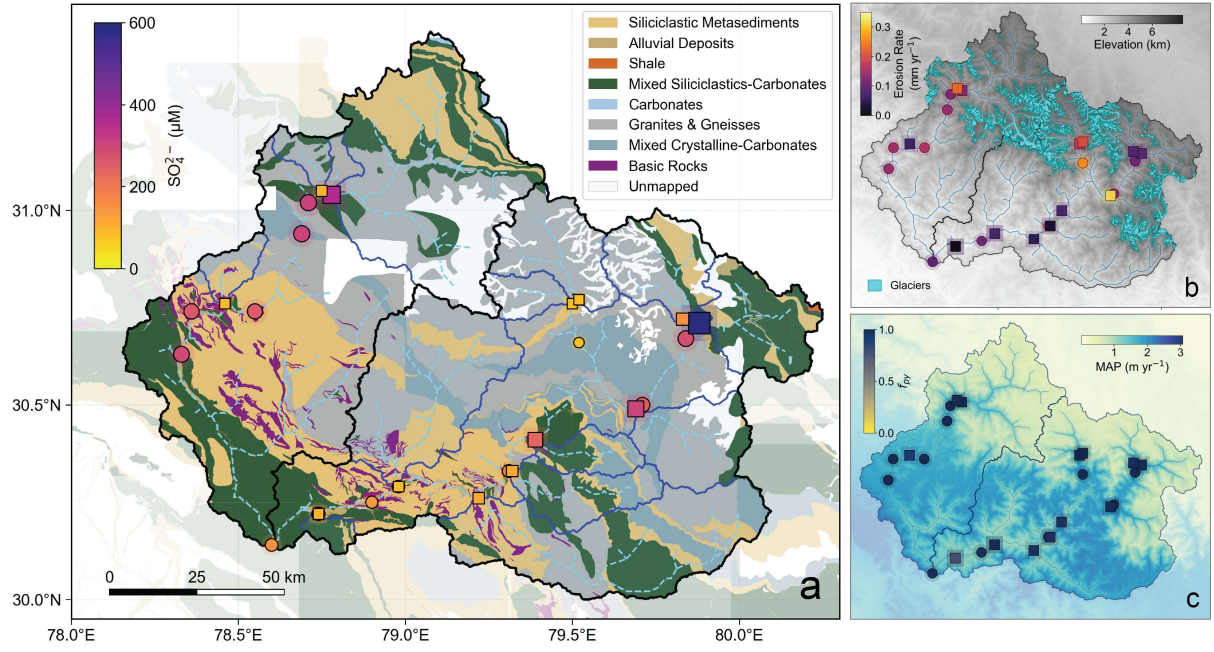


Fig. 2 | Study region maps. **a.** Lithology, as described in the Geological Survey of India database (<https://bhukosh.gsi.gov.in/>, accessed April, 2025) and reclassified into the lithological classes specified in the legend. Unmapped regions are areas permanently covered by glaciers. Also shown are major streams (cyan), major tributary catchment outlines (blue) and Bhagirathi (left) and Alaknanda (right) catchment outlines (black). Symbol colors indicate measured SO_4^{2-} concentrations at each sampling site. **b.** Elevation, including glacier extent (cyan). Symbol colors indicate measured or estimated catchment-averaged erosion rate at each sampling point (*Methods*). **c.** Mean Annual Precipitation (MAP; extracted from Ref. ⁵⁶). Symbol colors indicate the fraction of pyrite-derived SO_4^{2-} (f_{py}) at each sampling site as derived by inverting measured major ion concentrations (*Methods*). In all panels, square symbols indicate tributary samples and circles indicate main-stem samples.

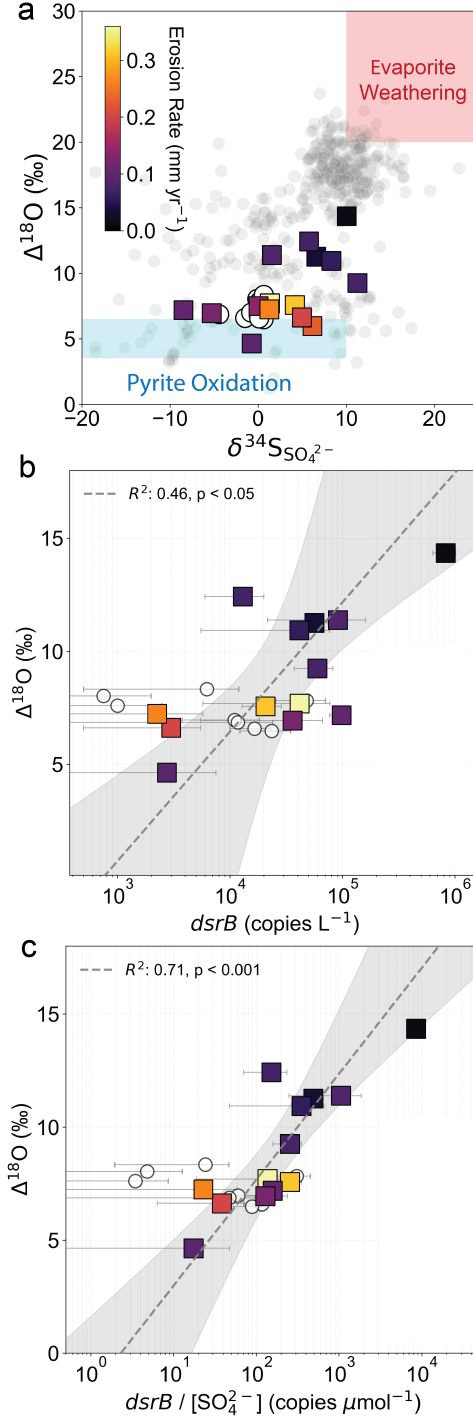


Fig. 3 | MSR controls on observed sulfate-isotope signals. **a.** $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ vs. $\Delta^{18}\text{O}$ of all global riverine SO_4^{2-} samples included in our database (grey circles; *Methods*; Table S2). Also shown are Ganga headwater samples analyzed here (white/coloured markers) and predicted pyrite oxidation and evaporite dissolution end-member compositions (coloured boxes; Ref. [18,36](#)). Ganga headwater $\Delta^{18}\text{O}$ as a function of **b.** dsrB marker-gene abundance (copies L⁻¹) and **c.** dsrB marker-gene abundance normalized to SO_4^{2-} concentration (copies μmol^{-1}). In all panels, square symbols indicate Ganga headwater tributary samples, white circles indicate Bhagirathi or Alaknanda main-stem samples, and symbol colours indicate measured or estimated catchment-averaged erosion rate at each sampling point (*Methods*). In panels **b** and **c**, dotted lines and grey envelopes represent logarithmic regressions and 95% confidence intervals, whereas error bars indicate $\pm 1\sigma$ uncertainty of replicate marker-gene qPCR measurements (*Methods*). Main-stream samples are excluded from each regression since they are subject to significant mixing of different water sources, although they broadly follow the expected relationship.

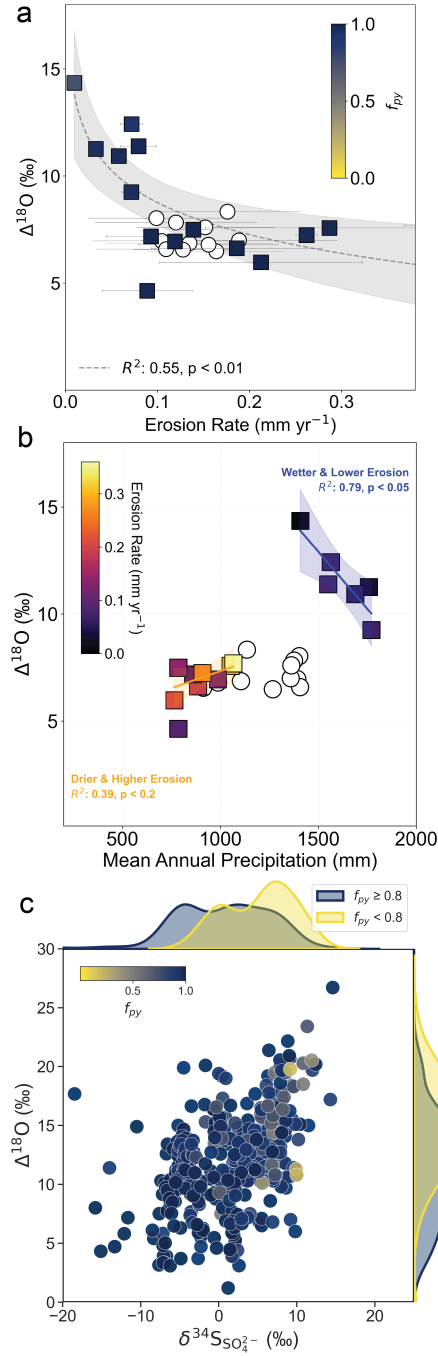


Fig. 4 | Landscape controls on observed sulfate-isotope signals. **a.** Measured or estimated catchment-averaged erosion rate vs. $\Delta^{18}\text{O}$ for all Ganga headwater SO_4^{2-} samples. Error bars indicate erosion-rate $\pm 1\sigma$ uncertainty (*Methods*). Symbol colours indicate the fraction of pyrite-derived SO_4^{2-} (f_{py}) at each sampling site as derived by inverting measured major ion concentrations (*Methods*). **b.** Mean Annual Precipitation (MAP) vs. $\Delta^{18}\text{O}$. Symbol colours indicate measured or estimated catchment-averaged erosion rate at each sampling point (*Methods*). Solid lines and envelopes represent ordinary least squares regressions and 95% confidence intervals for wet, low-erosion (blue) and dry, high-erosion (orange) tributaries. **c.** $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ vs. $\Delta^{18}\text{O}$ of all global riverine SO_4^{2-} samples included in our database that contain both stable-isotope and major-ion data (*Methods*; Table S2). Symbol colours indicate f_{py} as derived by inverting measured major ion concentrations (*Methods*). Kernel density estimates for $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\Delta^{18}\text{O}$ are also shown as marginal distributions, separated into samples with $f_{\text{py}} \geq 0.8$ (blue) and $f_{\text{py}} < 0.8$ (yellow). In panels **a** and **b**, square symbols indicate Ganga headwater tributary samples and circles indicate main-stem samples.

1 Methods

1 Sampling Methods

River water samples ($n = 26$) were collected at the monsoon onset in June 2024 from the Alaknanda and Bhagirathi main-stem as well as tributary catchments that span a range of lithology, elevation, and hydroclimatic parameters (Fig.1). River-water temperature, pH, total dissolved solids (TDS), and electrical conductivity (EC) were measured *in situ* using a Hanna portable multiparameter probe, which was calibrated with standard solutions prior to use. At each site, a measured quantity (~ 100 mL) of river water was passed through $0.22\ \mu\text{m}$ Millipore Sterivex filter cartridges. Effluent was sub-sampled into 4 mL amber vials without headspace for $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ analysis, whereas filter cartridges were placed on ice immediately after sampling and frozen at ca. $-5\ ^\circ\text{C}$ within ~ 8 hours (due to logistical constraints preventing storage at $-80\ ^\circ\text{C}$ in the field). Filter cartridges were subsequently shipped on dry ice and stored at $-80\ ^\circ\text{C}$ at ETH Zurich prior to DNA extraction and microbial marker gene analysis.

Additionally, ~ 2 L of river water was collected at each site using acid-washed HDPE bottles and subsequently filtered using a Tarson vacuum filtration unit and $0.22\ \mu\text{m}$ Millipore PES membrane filters. For major-ion measurements, effluent was sub-sampled into acid-cleaned HDPE bottles, which were pre-rinsed ($3\times$) with the sample water. Two aliquots were collected: one unacidified for major anion concentration measurements and one acidified with double-distilled HNO_3^- for major cation concentration measurements; both aliquots were preserved at $6\ ^\circ\text{C}$ until further analysis. A third aliquot was used for end-point alkalinity titrations within 12 hours of collection using $0.02\ \text{N}\ \text{H}_2\text{SO}_4$ and methyl orange indicator. The remaining ~ 1 L of effluent was passed through cartridges containing ~ 5 mL of Biorad AG 1-X8 anion-exchange resin over a ~ 5 -hour period (i.e., $\sim 200\ \text{mL}\ \text{hour}^{-1}$ flow rate) following standard procedures⁵⁷. Prior to fieldwork, all anion-exchange cartridges were pre-washed with 3 cartridge volumes each of $8\ \text{N}\ \text{HNO}_3^-$, $3\ \text{N}\ \text{HCl}$, and double-distilled water ($18.2\ \text{M}\Omega\text{-cm}$; henceforth “DDW”). After sample collection, cartridges were sealed with ~ 5 mL of residual sample water to prevent drying during storage.

Finally, for a subset of catchments ($n = 7$), sediment samples were collected from sand beds within active river channels for ^{10}Be measurements. Up to ~ 2 kg of sediments were collected at each sampling location, which were in close proximity to water sampling sites. Sampling locations were selected to minimize anthropogenic disturbance to sediment sourcing and transport processes relevant to ^{10}Be measurements (e.g., avoiding areas affected by construction activity or sand extraction). Collected sediments were oven-dried at $\sim 60\ ^\circ\text{C}$ and stored at room temperature prior to quartz extraction and ^{10}Be measurements.

2 Geospatial Analysis

The NASA Shuttle Radar and Topography Mission (SRTM) digital elevation model (DEM; 1 arc-second resolution) was used in ESRI ArcGIS to extract catchment areas and other geomorphic parameters (i.e., mean elevation, slope and area) upstream of each sampling point. Lithology data were downloaded from the Geological Survey of India’s database (<https://bhukosh.gsi.gov.in/>, accessed in April, 2025). Lithology is reported in 91 classifications, which we re-classify into 9 classifications (i.e., siliciclastic metasediments, alluvial deposits, shale, mixed siliciclastics-carbonates, carbonates, granites and gneisses, mixed crystalline-carbonates, basic rocks and unmapped) to preserve lithological distinctions across the study region without affecting overall clarity and resulting map interpretability. Unmapped regions are typically those permanently covered by glaciers. Glacial coverage was calculated as the fraction of catchment area upstream of each sampling point that is covered by glaciers as determined using the Randolph Glacier Inventory⁵⁸. Mean annual precipitation (MAP) and temperature (MAT) rasters were extracted from the WorldClim global climate database V2⁵⁶ at 30 seconds resolution and averaged over the catchment area

upstream of each sampling point. Finally, catchment-averaged normalized channel steepness index, k_{sn} ⁵⁹, was calculated upstream of each sampling point using Topotoolbox v2.2⁶⁰ with an upstream drainage area $>5 \text{ km}^2$ and a concavity index of $\theta = 0.45$.

3 Major Ion Measurements

Major cation (Na^+ , K^+ , Ca^{2+} , Mg^{2+}) and anion (F^- , Cl^- , NO_3^- , SO_4^{2-}) concentrations were measured by ion chromatography at IIT Kanpur using a Metrohm 930 Compact ion chromatograph (IC). Anion measurements utilized the Metrosep A Supp 5 column and a mobile phase that consisted of 3.2 mM Na_2CO_3 and 1.0 mM NaHCO_3 in Milli-Q water with 0.5% (v/v) acetonitrile at a flow rate of 0.7 mL min^{-1} ; the suppressor was regenerated using a solution of 50 mM H_2SO_4 and 5 g L^{-1} oxalic acid. Cations were measured using a Metrosep C 6 column, without suppression, and a mobile phase that consisted of 2.0 mM HNO_3 , 0.4 mM oxalic acid, and Pyridine-2,6-dicarboxylic acid at a flow rate of 0.9 mL min^{-1} . For all major ion measurements, 10 mg L^{-1} multi-element IC ion standard solutions were diluted gravimetrically to create a 5-point calibration curve ranging from 0.1–10 mg L^{-1} , which always displayed correlation coefficients (i.e., R^2 values) ≥ 0.99 for all ions. River water samples were diluted up to $5\times$ to ensure values fell within the analytical range defined by the calibration curve. A 5 mg L^{-1} standard solution was repeatedly analyzed to ensure accuracy and precision, both of which were always better than 5% ($\pm 1\sigma$ uncertainty). For circumneutral river waters such as those analyzed here (pH ~ 7), measured alkalinity was assumed to be equivalent to (unmeasured) bicarbonate (HCO_3^-) ion concentration following standard practice⁴⁸.

4 Weathering End-member Mixing Model

The proportion of major ions (i.e., Na^+ , K^+ , Ca^{2+} , Mg^{2+} , Cl^- and SO_4^{2-}) derived from weathering of different lithological end-members was constrained using a lithology partitioning model solved using a Monte Carlo inversion approach^{19,61,62}. This model assumes that (i) weathering-derived solutes behave conservatively and (ii) weathering is congruent, with the exception of pyrite, which is treated as a separate end-member that is agnostic to host lithology⁸. While these assumptions might not strictly hold in all cases due to (i) incorporation into secondary clays—particularly in low-lying catchments with thicker regolith—and (ii) incongruent weathering—for example, of different minerals within silicate lithologies—neither process involves SO_4^{2-} . Resulting sulfur-cycle interpretations are therefore robust to violations of both assumptions; we nevertheless implicitly account for such violations by assuming broad *a priori* end-member compositional ranges.

To solve our model, measured river-water major ion concentrations were first corrected for precipitation inputs, which are not explicitly included as an end-member and do not contribute to weathering. Specifically, similar to Refs. ^{19,29,63}, we first subtract the the median Cl^- concentration of Himalayan precipitation, Cl_p^- , from the Cl^- content of each sample. Then, for all other ions X , we subtract the product of the median X/Cl^- ratio in Himalayan precipitation and Cl_p^- . For samples with Cl^- concentrations lower than Cl_p^- , we instead multiply the median X/Cl^- ratio in Himalayan precipitation with measured Cl^- concentrations. That is, we let

$$X^* = X - \left(\frac{X}{\text{Cl}^-} \right)_p \times \begin{cases} \text{Cl}_p^- & \text{if } \text{Cl}^- \geq \text{Cl}_p^- \\ \text{Cl}^- & \text{if } \text{Cl}^- < \text{Cl}_p^- \end{cases} \quad (1)$$

where the superscript $*$ indicates precipitation-corrected concentrations. Based on reported Cl^- concentrations of Himalayan precipitation³⁵, such corrections are typically $<10\%$ of measured river-water ion concentrations (Table S1).

Second, all precipitation-corrected river-water species charge abundances were normalized to the sum of measured Na^+ , K^+ , Ca^{2+} , Mg^{2+} , and SO_4^{2-} charge as

$$\chi_{X^*} = \frac{|Z|[X]^*}{\Sigma^{\pm*}}, \quad (2)$$

where X is the species of interest (i.e., Na^+ , K^+ , Ca^{2+} , Mg^{2+} or SO_4^{2-}), $|Z|$ is the absolute value of species charge (i.e., 1 for Na^+ and K^+ ; 2 for Ca^{2+} , Mg^{2+} , and SO_4^{2-}), $[X]^*$ is the precipitation-corrected dissolved concentration of species X , and

$$\Sigma^{\pm*} = [\text{Na}^+]^* + [\text{K}^+]^* + 2[\text{Ca}^{2+}]^* + 2[\text{Mg}^{2+}]^* + 2[\text{SO}_4^{2-}]^*, \quad (3)$$

is the sum of precipitation-corrected charges. We explicitly include oxidation of pyrite—a common accessory mineral in sedimentary and magmatic rocks—as a separate end-member, which does not release base cations but rather achieves charge balance by providing proton equivalents and/or consuming alkalinity (i.e., HCO_3^-). Thus, for our model to be sensitive to pyrite oxidation contributions, SO_4^{2-} must be included in the definition of $\Sigma^{\pm*}$ in Eqs. 2-3.

Third, *a priori* end-member χ_X ranges were determined similar to Eq. 3 (but omitting precipitation corrections) using major-ion concentrations of each lithological end-member. Based on the study-area lithology and similar models from nearby catchments (i.e., Nepal Himalaya; Ref. 19), we consider the following end-members: (i) carbonate, (ii) gneiss/granite, (iii) pyrite, (iv) evaporite (v) and shale (Table S3). Although evaporites have never been explicitly mapped in our study region (Fig. 2)—and gypsum contributions are not evident in input from buried evaporites or deep brine (see *Main Text*)—we include an evaporite end-member in our inversion to be as conservative as possible. Following Ref. 61, “gneiss/granite” and “shale” are included as separate end-members to account for Mg-rich and Mg-poor silicates. The *a priori* end-member χ_X ranges used here are largely identical to those of Ref. 19, which in turn were compiled from previously reported Himalayan lithological compositions^{29,64–66}. The only exceptions are pyrite, which is included here as a separate source of pure SO_4^{2-} (c.f., inclusion within the shale end-member in Ref. 19), and evaporites, for which we allow greater compositional variability than in Ref. 19 to be more conservative in assessing deep brine inputs (see Table S3).

Fourth, end-member fractional contributions were determined for each measured river-water sample. Because our model contains six conservative tracers (i.e., $\chi_{\text{Na}^{+*}}$, $\chi_{\text{K}^{+*}}$, $\chi_{\text{Ca}^{2+*}}$, $\chi_{\text{Mg}^{2+*}}$, $\chi_{\text{SO}_4^{2-*}}$ and the sum-to-unity constraint) and five end-members, a non-negative linear regression problem is solved as

$$\min_{\mathbf{X}} \|\mathbf{A}\mathbf{X} - \mathbf{B}\|, \quad (4)$$

where $\mathbf{A}$ is the shape $[m \times n]$ design matrix of end-member χ_X values; $\mathbf{X}$ is the shape $[n \times p]$ matrix containing the model-estimated, non-negative fractional contributions of each end-member to each sample; $\mathbf{B}$ is the shape $[m \times p]$ matrix containing measured χ_{X^*} values for each sample; m is the number of conservative tracers; n is the number of end-members; and p is the number of measured river-water samples. Importantly, to include the sum-to-unity constraint, all entries in the final row of both $\mathbf{A}$ and $\mathbf{B}$ are set to unity; because we solve a linear regression problem rather than a direct matrix inversion, small deviations from the sum-to-unity constraint (typically $\sim 5\%$) are implicitly allowed.

To account for uncertainty in *a priori* end-member compositions, Eq. 4 was solved in Monte Carlo fashion (10,000 iterations) after pulling each entry in $\mathbf{A}$ from a uniform distribution bounded by the *a priori* end-member ranges (Table S3). Following standard practice^{19,61,62}, the top 1% of Monte Carlo

solutions with the lowest root mean square error (RMSE), defined as

$$\text{RMSE} = \left(\frac{\|\mathbf{AX} - \mathbf{B}\|^2}{p} \right)^{1/2}, \quad (5)$$

were then retained and used to calculate best-fit fractional end-member contributions to each sample, including uncertainty. In addition to fractional contributions, this model output also allows *a posteriori* χ_X ranges to be calculated for each lithological end-member (Table S4).

Finally, the fraction of pyrite-derived SO_4^{2-} in each sample, f_{py} , was calculated by multiplying the average fractional contribution of the pyrite-weathering end-member, X_{py} , by the average posterior modeled SO_4^{2-} composition of the pyrite end-member, $\chi_{\text{SO}_4^{2-}}^{\text{py}}$, normalized to the total SO_4^{2-} contribution from all end-members, as

$$f_{\text{py}} = \frac{X_{\text{py}} \cdot \chi_{\text{SO}_4^{2-}}^{\text{py}}}{\sum_j X_j \cdot \chi_{\text{SO}_4^{2-}}^j}, \quad (6)$$

where j represents a lithological end-member. All inversion calculations can be performed using the accompanying Python script.

We used this inversion-based lithology partitioning model to constrain f_{py} for all samples collected in the Alaknanda and Bhagirathi catchments within the Ganga headwaters as part of this study (Table S1), which are situated in broadly uniform lithology and geological setting (Fig.2). We then extended this framework to estimate f_{py} values in two global datasets:

First, we use a compiled dataset of simultaneous riverine $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ and major ion concentrations measurements (Table S2)^{3,13,18,67–79}. As for the Alaknanda and Bhagirathi basins, the inversion only includes major ion concentrations as inputs; we do not include $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ or $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ as conservative tracers since our intention is to explicitly assess whether MSR modifies these values. Unlike in the Alaknanda and Bhagirathi basins however, the rivers compiled here drain lithologically diverse settings. We account for this lithology end-member variability in the following ways: (i) our *a priori* Mg-poor “granite/gneiss” and Mg-rich “shale” end-member χ_X ranges are broadly defined such that they encompass a wide range of silicate end-member compositions; (ii) our *a priori* evaporite end-member χ_X ranges are modified to allow for all possible evaporite compositions (e.g., $\chi_{\text{SO}_4^{2-}}$ and χ_{Cl^-} are allowed to vary between 0.0 and 1.0; see Table S4); and (iii) the model is solved individually for each reported dataset, which is localized to a single catchment and/or geological setting (i.e., yielding setting-specific *a posteriori* end-member χ_X ranges). Additionally, as described above, by treating pyrite weathering as a separate end-member, our results are agnostic to the specific lithologies being weathered. The global mean f_{py} value (including $\pm 1\sigma$ uncertainty) based on the inversion of this dataset is then calculated as the average and standard deviation of f_{py} estimated for each sample (Fig.S7a).

Second, we use the Global River Chemistry (GLORICH) dataset⁵⁵. As GLORICH frequently contains time-series data—which better incorporate temporal variability—we solved our model for each individual sampling station rather than combining samples within a given catchment and/or geological setting. Additionally, since most samples are derived from floodplain-dominated and/or anthropogenically impacted sites—which would not yield pure weathering signals—we screened all data to only include individual mountainous sampling stations with ≥ 10 measurements and low anthropogenic activity ($n = 235$; see Table S5 for complete screening criteria). Lithology end-member variability was accounted for exactly as described above for the combined isotope and major ion dataset. To obtain a globally representative f_{py} estimate, we first calculated mean f_{py} values for each station as the average of all time-series

results (Fig.S7b), which in turn were used to calculate a global mean f_{py} (including $\pm 1\sigma$ uncertainty) as described above.

5 Isotope Measurements

To isolate SO_4^{2-} and remove all other anions that could interfere with oxygen-isotope measurements (i.e., nitrate, NO_3^-)⁵⁷, anion-exchange cartridges were eluted at ETH Zurich using 20 mL of 0.8 N NaCl in DDW. NaCl solution was chosen rather than more conventional HCl^{57,80} in order to avoid acidifying adsorbed HCO_3^- to CO_2 , which could bubble and disturb anion-exchange resin beds. Before eluting unknown samples, elution volumes were tested and verified using synthetic river water with known SO_4^{2-} , NO_3^- , and HCO_3^- concentrations; test results indicated that SO_4^{2-} is quantitatively eluted within ~ 15 mL and that NO_3^- is baseline separated by ~ 10 mL (Fig. S8). A diluted aliquot of the SO_4^{2-} -containing eluent for each sample was then analyzed by IC (Thermo Scientific Dionex Aquion with an AG9-HC guard column and an AS-9 HC analytical column) to ensure that (i) SO_4^{2-} was quantitatively eluted (i.e., by comparing to initial river-water concentrations) and (ii) no NO_3^- was co-eluted.

Then, 3 mL of 1 M barium chloride (BaCl_2) in DDW was added to the SO_4^{2-} -containing eluent to precipitate barite (BaSO_4), which was subsequently rinsed ($3\times$) with 0.1 N HCl to dissolve any potential co-precipitated barium carbonate (BaCO_3). After precipitation, supernatants were again analyzed by IC to ensure quantitative yields (i.e., no remaining dissolved SO_4^{2-}). Finally, all precipitates were analyzed by powder X-ray diffraction (XRD; Empyrean diffractometer; Cu $K\alpha$ radiation, $\lambda = 1.54184$; PIXcel1D-Medipix3 detector; 5° to 90° (2θ); 0.013° steps with a counting time of 128.5 s per step) to ensure that they contain pure barite.

Both $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ of precipitated barites were then measured at ETH Zurich. Following standard practice⁸¹, $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ was determined using 200-300 μg of powdered BaSO_4 wrapped in tin capsules along with an excess of vanadium pentoxide (V_2O_5) catalyst, which was converted to SO_2 analyte gas and analyzed using a Thermo Scientific Flash EA 1112 coupled to a Thermo Scientific Delta V isotope ratio mass spectrometer (IRMS). Each sample was measured in duplicate, and resulting $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ values are reported in units of “permil” (‰) relative to Vienna Canyon Diablo Troilite (VCDT). Calibration and scale-compression correction was achieved using a three-point calibration curve of international standard reference materials analyzed in the beginning, middle and end of each analytical session: NBS-127 ($\delta^{34}\text{S} = +21.1 \pm 0.21\text{‰}$), IAEA-SO-5 ($\delta^{34}\text{S} = 0.49 \pm 0.11\text{‰}$), and IAEA-SO-6 ($\delta^{34}\text{S} = -34.05 \pm 0.10\text{‰}$). Repeat measurements yielded a reproducibility typically better than 0.1‰ ($\pm 1\sigma$) for each reference material.

Similarly, $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ was determined using 200-300 μg of powdered BaSO_4 wrapped in silver capsules along with a $2\times$ excess of graphite powder (pre-combusted at 450°C) to ensure complete conversion to CO analyte gas. As detailed in Ref.⁸², CO was generated using a Thermo Scientific EA IsoLink OH high-temperature conversion (HTC) furnace equipped with a Costech Analytical ZeroBlank autosampler continuously flushed with reference He gas, subsequently purified using a custom-built cryogenic trapping system, and analyzed using a Thermo Scientific Delta V Plus IRMS. Each sample was measured in triplicate, and resulting $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$ values are reported in permil relative to Vienna Standard Mean Ocean Water (VSMOW). Blank corrections were achieved using silver capsules containing only graphite analyzed at the beginning of each analytical sequence. Calibration and scale-compression correction was achieved using a two-point calibration curve of in-house working standards, “H Barite” ($\delta^{18}\text{O} = +19.8 \pm 0.2\text{‰}$) and “VL Barite” ($\delta^{18}\text{O} = -16.7 \pm 0.1\text{‰}$), analyzed at the beginning, middle and end of each analytical session. In-house standards have been calibrated to the same international standard reference materials: NBS-127 ($\delta^{18}\text{O} = +8.59 \pm 0.26\text{‰}$), IAEA-SO-5 ($\delta^{18}\text{O} = 12.13 \pm 0.33\text{‰}$) and IAEA-SO-6 ($\delta^{18}\text{O} = -11.35 \pm 0.31\text{‰}$)⁸³. Repeat measurements yielded a reproducibility typically better than 0.2‰ ($\pm 1\sigma$) for each in-house standard.

Finally, $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ was determined using a Picarro high-precision vaporizer (including a salt liner to minimize salt precipitation and matrix effects) coupled to a Picarro L2130-i cavity ring-down spectroscopy (CRDS) isotope and gas concentration analyzer using N_2 as purge gas and operated in “express mode”. As detailed in Ref. ⁸⁴, twelve injections (2.1 μL each) were performed for each analysis; of these, the first six injections were used solely to flush the vaporizer and remove memory effects whereas the subsequent two individual pulses were not utilized for data reduction due to potential residual drift. Resulting $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ values are reported in permil relative to VSMOW. Calibration was achieved using a three-point calibration curve of IAEA international standard reference materials analyzed at the beginning of each analytical session ⁸⁴: SLAP-2 ($\delta^{18}\text{O} = -55.50 \pm 0.02\text{‰}$), GRESP ($\delta^{18}\text{O} = -33.40 \pm 0.04\text{‰}$) and VSMOW aliquot 2 ($\delta^{18}\text{O} = 0.0 \pm 0.3\text{‰}$). Drift correction was achieved by analyzing three in-house working standards (Siberian water, SIBW; Lake Zurich water, ZW; and Mediterranean Sea water, MEDW) at the beginning and end of each analytical session. Repeat measurements yielded a long-term reproducibility typically better than 0.25‰ ($\pm 1\sigma$) for each in-house standard.

6 DNA Extraction and Marker Gene Analyses

A DNeasy PowerSoil Pro Kit (Qiagen, Germany) was used to extract DNA from previously frozen Sterivex filters following the manufacturer’s protocol. Specifically, bead beating was performed on a FastPrep24 bead beater (MP Biomedicals, Germany), and the resulting DNA-containing eluent solution ($\sim 50\mu\text{L}$) was stored at -80°C until further analysis. DNA quantity was measured using the Qubit 4.0 fluorometer with the dsDNA HS Assay kit (Invitrogen, USA). Then, the V4 and V5 hypervariable regions of the 16S rRNA gene were amplified by quantitative polymerase chain reaction (qPCR) using the forward primer 515F (5'-GTGNCAGCMGCCGCGGTAA-3') ⁸⁵ and the reverse primer 926R (5'-CCGYCAATTYMTTTRAGTTT-3') ⁸⁶ performed on a LightCycler 480 II system (Roche, Switzerland) at the Genetic Diversity Center, ETH Zurich. Specifically, a solution containing forward and reverse primers (each at 10 μM in 0.5 μL), 5 μL SsoFast EvaGreen Supermix with Low ROX 2x (Bio-Rad, United States of America), and 1 μL Bovine Serum Albumin (10 mg mL^{-1}) was combined with 2 μL of template DNA and loaded onto well plates. The qPCR program included an initialization of 95°C for 300 seconds, 40 cycles of denaturation (10 seconds at 95°C), annealing (30 seconds at 50°C), and elongation (15 seconds at 68°C) with a melting curve of 15 seconds at 95°C and 60 seconds at 55°C and one acquisition per $^\circ\text{C}$ from 55°C to 95°C . 16S rRNA gene standards ranging from 2.088×10^5 to 2.088×10^6 amplicon copies per μL were prepared by serial dilution of a consensus 16S synthetic construct (details in Ref. ⁸⁷).

Similarly, the *dsrB* gene was quantified by qPCR on the same LightCycler 480 II System using *dsrB* F1ah/4RSI1af primer mixtures ⁸⁸ (Table S6). A solution containing the forward and reverse primers (each at 10 μM in 0.5 μL), 2 μL of template DNA, and the above-specified reagents was loaded onto well plates. Here, the qPCR program included an initialization of 95°C for 300 seconds, 45 cycles of denaturation (30 seconds at 95°C), annealing (30 seconds at 56°C), and elongation (20 seconds at 72°C) with a melting curve of 15 seconds at 95°C and 15 seconds at 55°C and one acquisition per $^\circ\text{C}$ from 55°C to 95°C . *dsrB* gene standards ranging from 6.24×10^1 to 6.24×10^5 gene amplicon copies per μL were prepared by serial dilution of an synthetic partial *dsrB* gene standard based on an uncultured bacterium from deep sea sediment (GenBank accession number MG743156.1; Table S6).

Copy numbers for both genes in normalized samples were derived from the crossing point values (Cp values) and the transformation factor from standard curves obtained from designed gene constructs of known copy numbers following Ref. ⁸⁹. The standard curves always displayed correlation coefficients (i.e., R^2 values) ≥ 0.985 for both *dsrB* and 16S rRNA. All qPCR results were corrected for procedural blank and reported in terms of copies L^{-1} of river water. Additionally, although high Himalayan waters would naturally have low biomass, we nevertheless exclude extremely low biomass samples ($n = 5$; here, defined

as $< 10^4$ 16S rRNA gene copies L^{-1} of river water) from our interpretations to be as conservative as possible, as *dsrB* was not detected in any of these samples. Notably, these are typically samples from high-erosion catchments characterized by low $\Delta^{18}O$, and their inclusion would not affect our conclusions.

7 Erosion Rate Determination

Quartz preparation followed established protocols^{90,91} at the Geological Institute, ETH Zurich. Samples were sieved to 250–500 μm , magnetically separated, and leached with HCl and H_2SiF_6 to remove non-quartz minerals. Remaining quartz was decontaminated of atmospheric ^{10}Be , fully dissolved in HF, and processed by cation and anion exchange chromatography to purify the ^{10}Be . Beryllium hydroxide ($Be(OH)_2$) was then precipitated, oxidized to BeO at 1000 °C, and pressed for AMS analysis. Resulting $^{10}Be/^{9}Be$ ratios were measured using a Multi Isotope Low-Energy AMS (MILEA) system at the Laboratory for Ion Beam Physics, ETH Zurich⁹². Measured $^{10}Be/^{9}Be$ ratios were normalized and corrected using a full process blank ratio of $5.2 \pm 2.1 \times 10^{-15}$. Normalization was performed relative to our in-house secondary ^{10}Be standards, S2007N and S2010N, with nominal values of $^{10}Be/^{9}Be = 28.1 \times 10^{-12}$ and 3.3×10^{-12} , respectively⁹³. S2007N and S2010N have been calibrated with our new primary standard ICN 01-5-1, with a nominal $^{10}Be/^{9}Be$ value of 2.709×10^{-11} ⁹⁴. The 1σ error of S2007N and S2010N are 2.7% and 2.2%⁹³, respectively. For the normalization of ^{10}Be results to S2007N and S2010N, we use the recommended ^{10}Be half-life of 1.387 ± 0.012 Ma^{95,96}.

Measured ^{10}Be concentrations range from $1.9 \pm 0.2 - 15.3 \pm 0.7 \times 10^4$ atoms g^{-1} of quartz (Table S1). Measured ^{10}Be concentrations were used to calculate erosion rates with the CRONUS-Earth online calculator⁹⁷ assuming a time-independent production scaling as described in Ref.⁹⁸ and a constant bedrock density of 2.65 g cm^{-3} . Following the recommendation of Ref.⁹⁹, no topographic shielding corrections were applied. Furthermore, snow- and ice-cover shielding effects are difficult to correct for in our study area due to a lack of detailed data on snow depth and historical glacial cover. Nevertheless, Ref.¹⁰⁰ investigated snow and ice cover shielding effects on ^{10}Be -derived erosion rates in the adjacent Yamuna catchment, which shares broadly similar lithology, topography, and hydroclimatic conditions with our study area. While not indicative of paleo-erosion, using present-day satellite-based snow- and ice-cover data, they estimated that such shielding corrections typically modify erosion rate estimates by less than $\sim 10\%$, and never by more than $\sim 20\%$. They further concluded that tectonics, rather than climate or glaciers, is the primary driver of erosion rates in such settings. As such, the absence of snow shielding corrections would not meaningfully affect the conclusions of our study.

For catchments lacking ^{10}Be measurements, erosion rates were estimated by regressing k_{sn} , the normalized steepness index (Table S1), with ^{10}Be -based erosion rates derived here. A power-law relationship between k_{sn} and erosion rates has been previously documented in Taiwan⁴⁸ and the adjacent Yamuna catchment¹⁰⁰ in the Himalayas. We observe a similar relationship in the Alaknanda and Bhagirathi catchments, and we apply the derived scaling relationship (Fig.S3) to predict erosion rates where direct constraints are absent, including propagated error on k_{sn} estimates ($\pm 1\sigma$; Table S1).

2 Supplementary Figures

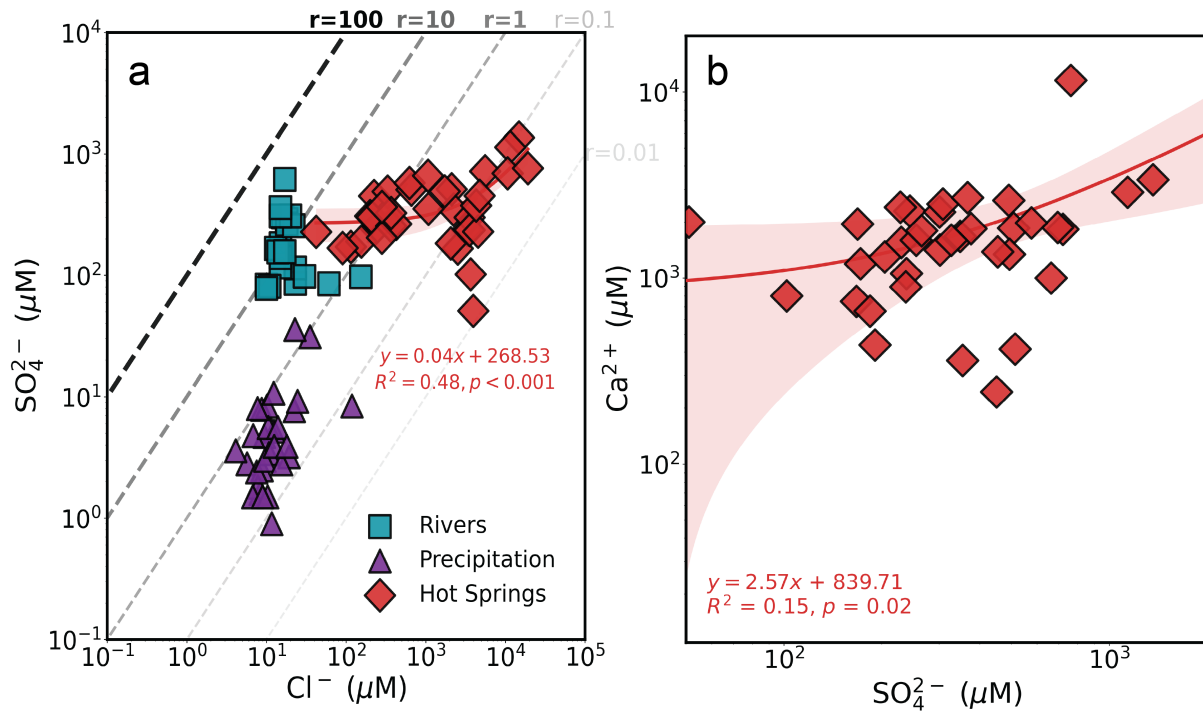


Fig. S1 | Evidence for no evaporite gypsum inputs. **a.** Dissolved Cl^- vs. SO_4^{2-} concentrations from rivers (this study, blue squares) as well as previously reported precipitation (purple triangles)³⁵ and hot spring (red diamonds)^{101–103} samples from our study area. Dashed lines indicate different $\text{SO}_4^{2-}/\text{Cl}^-$ ratios (i.e., $r = 0.01$ to 100 moving from light gray to black). Also shown are ordinary least squares linear regression results for hot-spring data (including 95% confidence interval, corresponding equation, R^2 , and p -value). **b.** Previously reported dissolved SO_4^{2-} vs. Ca^{2+} concentrations for hot springs^{101–103} in our study area. Also shown are ordinary least squares linear regression results (including 95% confidence interval, corresponding equation, R^2 , and p -value). Unlike for Cl^- vs. SO_4^{2-} , hot-spring SO_4^{2-} and Ca^{2+} concentrations are weakly correlated.

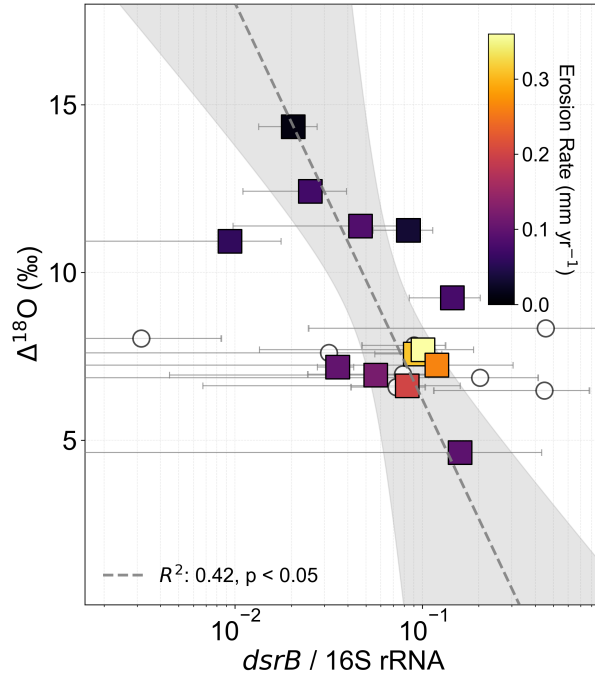


Fig. S2 | Microbial community composition controls on observed sulfate-isotope signals. Ganga headwater $\Delta^{18}\text{O}$ as a function of *dsrB* marker-gene abundance normalized to 16S rRNA marker-gene abundance (copies copy⁻¹). Square symbols indicate Ganga headwater tributary samples, white circles indicate Bhagirathi or Alaknanda main-stem samples, and symbol colours indicate measured or estimated catchment-averaged erosion rate at each sampling point (*Methods*). Additionally, dotted lines and grey envelopes represent log-linear regressions and 95% confidence intervals, whereas error bars indicate propagated $\pm 1\sigma$ uncertainty of replicate marker-gene qPCR measurements (*Methods*). Main-stream samples are excluded from each regression since they are subject to significant mixing of different water sources, although they broadly follow the expected relationship.

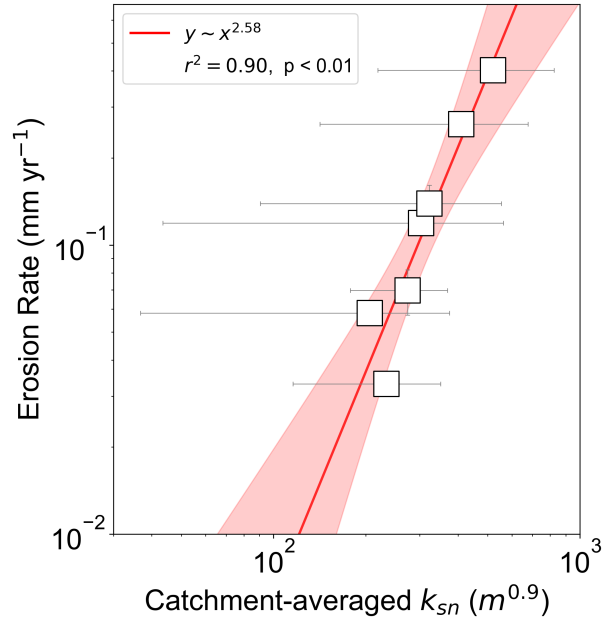


Fig. S3 | Catchment erosion-rate estimate calibration. Measured ^{10}Be -derived erosion rates plotted as a function of upstream catchment-averaged steepness indices (k_{sn} values; *Methods*) for all sampling points in which ^{10}Be data exist. Error bars represent $\pm 1\sigma$ error on catchment-averaged k_{sn} values and propagated uncertainty in ^{10}Be -derived erosion rates as calculated by the CRONUS-Earth online calculator⁹⁷. Also shown are log-log ordinary least squares linear regression results (including 95% confidence interval, corresponding equation, R^2 , and p-value). Regression results were used to estimate erosion rates using upstream k_{sn} values for all catchments in which no ^{10}Be data exist.

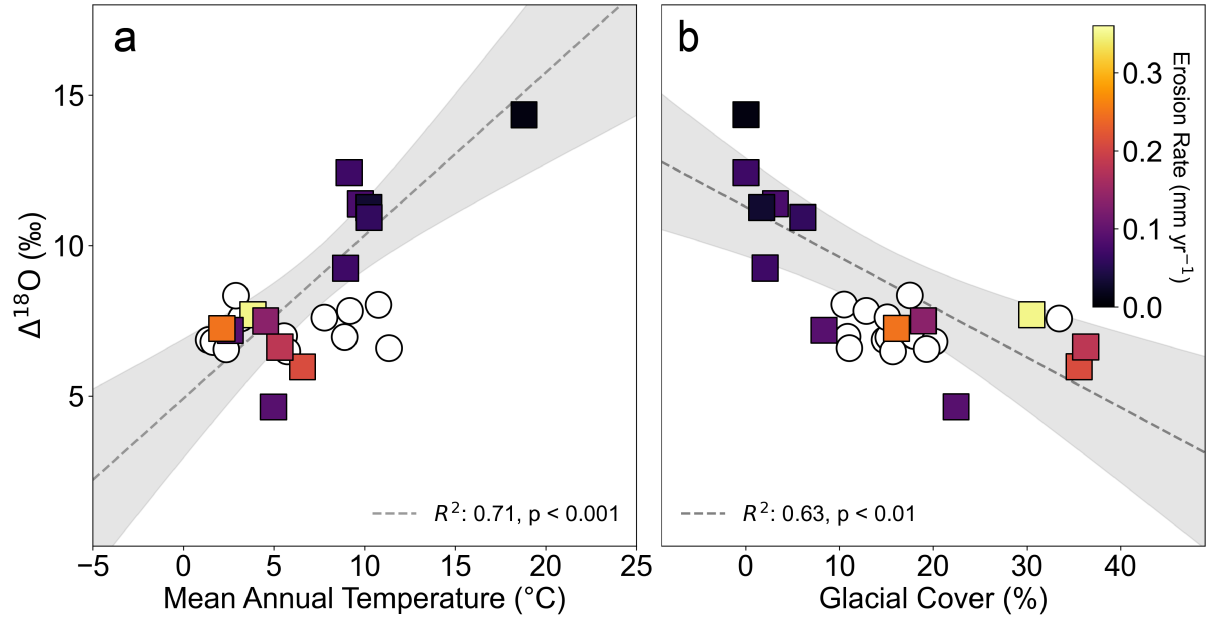


Fig. S4 | Environmental controls on observed sulfate-isotope signals. Ganga headwater $\Delta^{18}\text{O}$ as a function of **a.** catchment-averaged Mean Annual Temperature (MAT)⁵⁶ and **b.** percentage of catchment area covered by glaciers⁵⁸ (*Methods*). In both panels, square symbols indicate Ganga headwater tributary samples, white circles indicate Bhagirathi or Alaknanda main-stem samples, and symbol colours indicate measured or estimated catchment-averaged erosion rate at each sampling point (*Methods*). Additionally, dotted lines and grey envelopes represent linear regressions and 95% confidence intervals; propagated analytical error bars are always smaller than marker shapes. Main-stream samples are excluded from each regression since they are subject to significant mixing of different water sources, although they broadly follow the expected relationship.

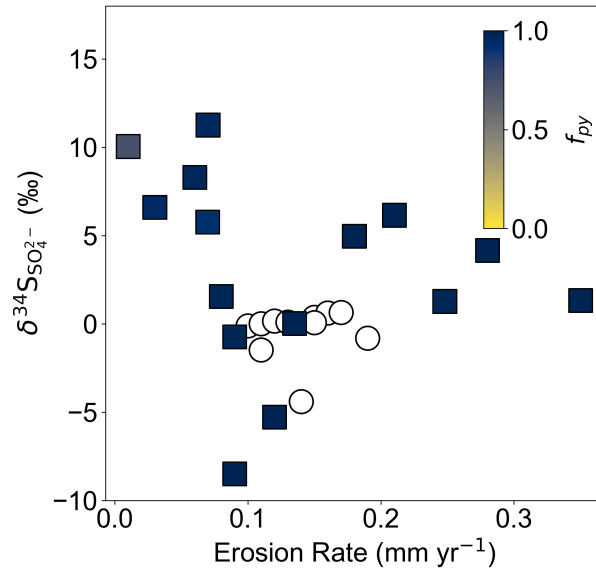


Fig. S5 | No erosion-rate control on observed sulfate $\delta^{34}\text{S}$ values. Measured or estimated catchment-averaged erosion rate vs. $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$. Square symbols indicate Ganga headwater tributary samples, white circles indicate Bhagirathi or Alaknanda main-stem samples, and symbol colours indicate the fraction of pyrite-derived SO_4^{2-} (f_{py}) at each sampling site as derived by inverting measured major ion concentrations (*Methods*). Unlike for $\Delta^{18}\text{O}$, no statistically significant correlation exists between erosion rate and $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$.

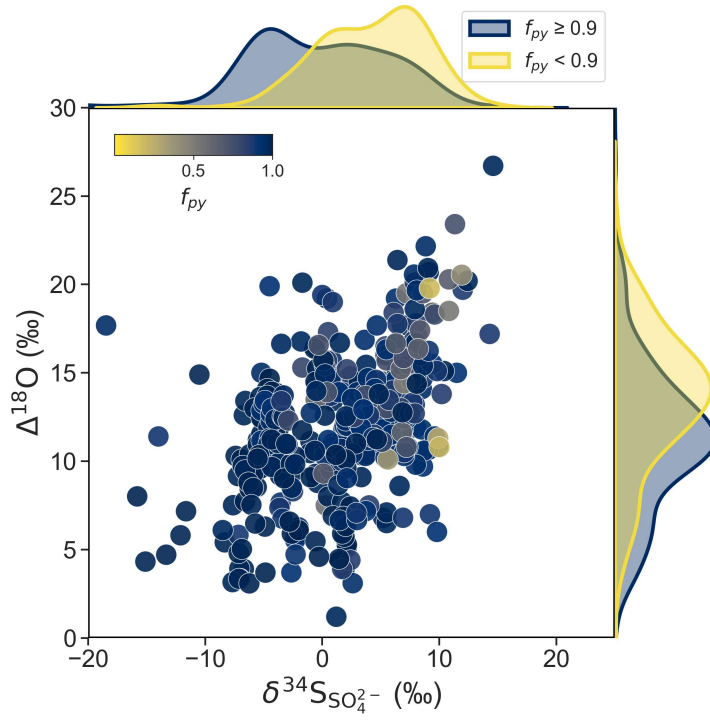


Fig. S6 | Sensitivity test of f_{py} cutoff. $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ vs. $\Delta^{18}\text{O}$ of all global riverine SO_4^{2-} samples included in our database that contain both stable-isotope and major-ion data (*Methods*; Table S2). Symbol colours indicate f_{py} as derived by inverting measured major ion concentrations (*Methods*). Kernel density estimates for $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\Delta^{18}\text{O}$ are also shown as marginal distributions, separated into samples with $f_{\text{py}} \geq 0.9$ (blue) and $f_{\text{py}} < 0.9$ (yellow; c.f., f_{py} of 0.8 in Fig. 4c). As was observed with a cutoff of $f_{\text{py}} = 0.8$, pyrite oxidation-dominated samples span the entire range of $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$ and $\Delta^{18}\text{O}$ observed in our dataset.

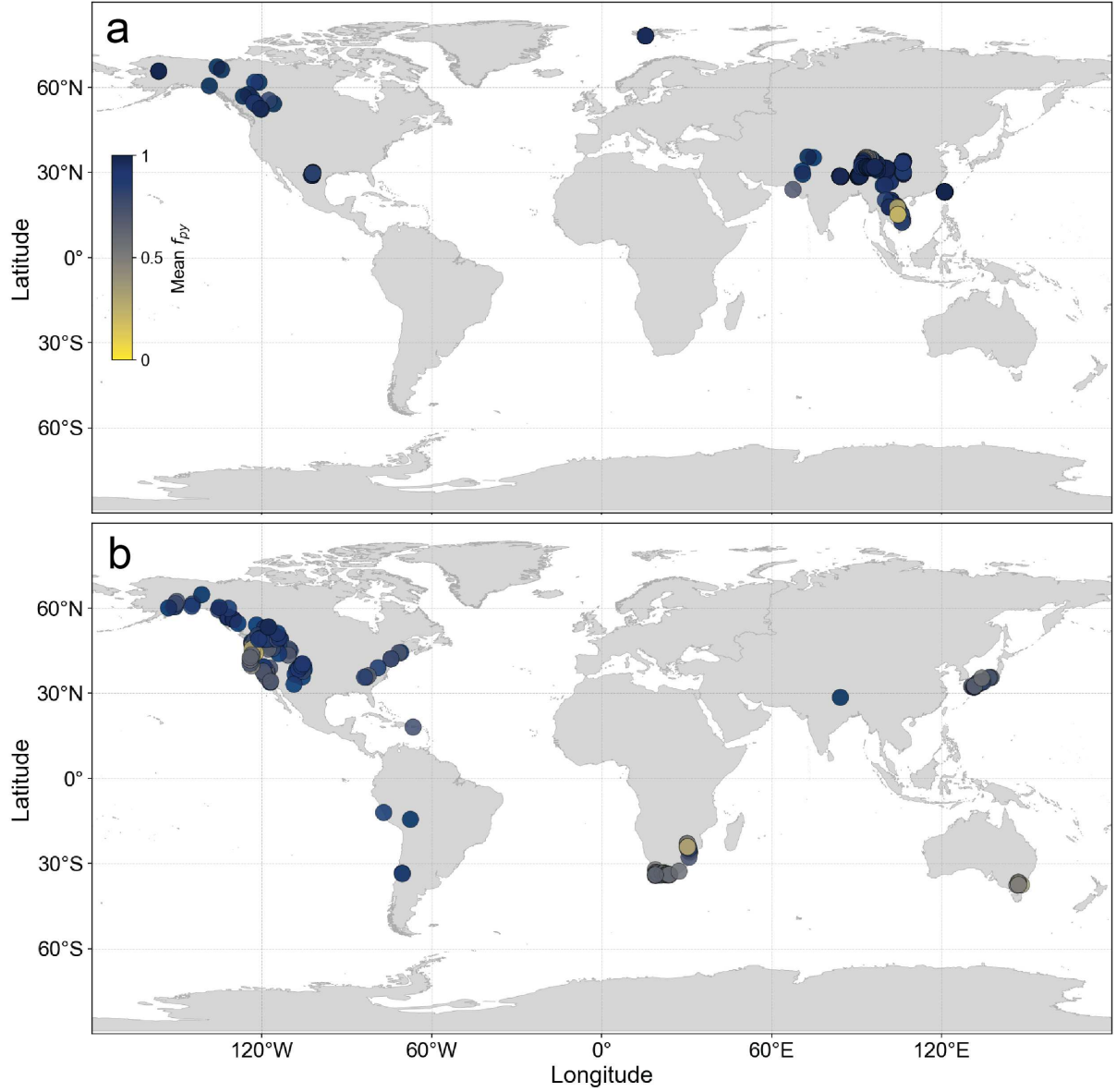


Fig. S7 | Inversion-model predicted global f_{py} distribution. Global map of **a.** sites of previously reported concomitant measurements of $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ and major ion concentrations (Table S2), and **b.** sampling stations from the GLORICH database⁵⁵ with ≥ 10 data points, filtered for mountain catchments with minimal anthropogenic influence ($n = 235$; see Table S5 for complete screening criteria). For both panels, marker colors indicate sampling station-specific f_{py} values (temporal averages for time-series sampling stations).

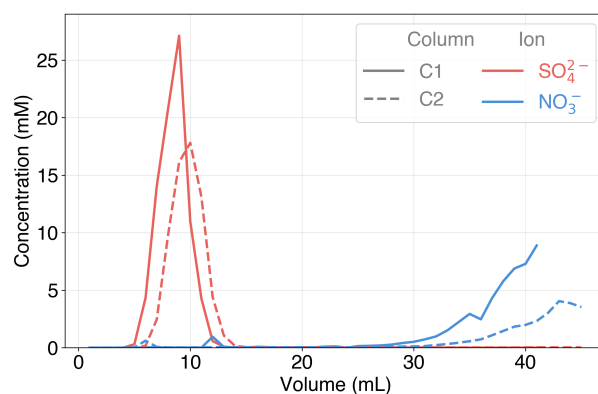


Fig. S8 | Anion-exchange resin elution test results. Two synthetic river water with known SO_4^{2-} , NO_3^- , and HCO_3^- concentrations were passed through anion-exchange columns. The columns (C1 and C2) were sequentially eluted in 1 ml steps to determine the window of SO_4^{2-} (red) recovery and confirm separation between SO_4^{2-} and NO_3^- (blue) peaks.

3 Supplementary Tables

Table S1: Sample sites in the Alaknanda and Bhagirathi catchments along with in-situ-measured physico-chemical parameters, major ion concentrations, and isotopic compositions— $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$, and $\delta^{18}\text{O}_{\text{H}_2\text{O}}$. Also included are *dsrB* and 16srRNA marker gene abundances; erosion rates constrained using ^{10}Be or inferred from regression with k_{sn} ; and major ion inversion model-constrained f_{py} values.

Table S2: Global compilation of concomitant $\delta^{34}\text{S}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{SO}_4^{2-}}$, $\delta^{18}\text{O}_{\text{H}_2\text{O}}$ and major ion concentrations measurements.

Table S3: *A priori* and *a posteriori* lithology weathering end-member compositions for Alaknanda and Bhagirathi catchments.

Table S4: *A priori* lithology weathering end-member compositions for inversions of global riverine chemistry.

Table S5: Parameters and thresholds used to screen the GLORICH river chemistry database for mountainous sampling stations with ≥ 10 measurements and low anthropogenic activity.

Table S6: Primer mixtures (*dsrB* F1ah/4RSI1af) and gene standard (GenBank accession number MG743156.1) used for *dsrB* qPCR quantification.

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Contributions

S.P. and J.D.H. conceived of the study; S.P. collected samples; S.P., L.B., A.S., N.H., and P.G. performed measurements; S.P., L.B., N.H., P.G., M.C., C.M., and J.D.H. analyzed results; S.P. and J.D.H. acquired funding; C.M., I.S.S., and J.D.H. provided reagents and instrumentation; S.P. and J.D.H. wrote the initial draft; all authors contributed to writing the final version.

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