

# **Sulfate sulfur isotopes and major ion chemistry reveal that pyrite oxidation counteracts CO<sub>2</sub> drawdown from silicate weathering in the Langtang-Trisuli-Narayani River system, Nepal Himalaya**

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Himalayan weathering and the associated drawdown of carbon dioxide (CO<sub>2</sub>) have previously been implicated in Cenozoic cooling. However, the oxidation of pyrite (FeS<sub>2</sub>) in weathering systems can counteract the alkalinity flux from silicate weathering and, for the duration over which FeS<sub>2</sub>-derived sulfate (SO<sub>4</sub><sup>2-</sup>) accumulates in seawater, result in elevated pCO<sub>2</sub> values. In this work we evaluate the role of FeS<sub>2</sub> oxidation in connections between Himalayan weathering and Cenozoic climate by constraining the relative importance of sulfuric acid and carbonic acid weathering in the Langtang-Trisuli-Narayani River system of the Nepal Himalaya. River water samples were collected monthly or bimonthly in 2011 from 16 sites spanning from the High Himalayan Crystalline (HHC) formation through the Lesser Himalayan (LH) formation, with upstream influences from the Tethyn Sedimentary Series (TSS). Measurements of major ion concentrations and the <sup>34</sup>S/<sup>32</sup>S isotope ratio in dissolved SO<sub>4</sub><sup>2-</sup> suggest substantial FeS<sub>2</sub> oxidation throughout the studied region. A river inversion model with Monte Carlo error propagation shows that 62-101% of river SO<sub>4</sub><sup>2-</sup> is derived from the oxidation of sulfide minerals. Moreover, the data show that the fraction of H<sub>2</sub>SO<sub>4</sub>-driven weathering is lower during the monsoon season than during the non-monsoon season, which we attribute to changes in the relative importance of subsurface flow paths with short and long residence times. Inversion results indicate that the primary control on the <sup>34</sup>S/<sup>32</sup>S ratio of river SO<sub>4</sub><sup>2-</sup> is lithologically variable isotope composition, with lower model-constrained rock <sup>34</sup>S/<sup>32</sup>S ratios in the LH and TSS than in the HCC. Overall, we find that FeS<sub>2</sub> oxidation counteracts much of the alkalinity flux from silicate weathering such that chemical weathering in the Narayani River system exerts minimal impact on pCO<sub>2</sub> over timescales >5-10 kyr and <10 Myr. Moreover, reanalysis of prior datasets from the Himalaya using the same inversion model suggests similar dynamics are active in other drainages but that there is substantially less FeS<sub>2</sub> oxidation occurring in the Himalayan floodplain.