Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.38166

Critical Zone processes place limits on carbon dioxide removal by enhanced rock weathering.

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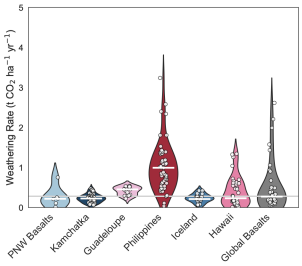
Enhanced Rock Weathering is a proposed strategy for carbon dioxide removal that relies on the dissolution of silicate minerals, typically basalt, when added to soils. Globally significant quantities of CO_2 can only be sequestered if excess alkalinity is generated by weathering of added rock minerals and if that alkalinity is conserved as it is transported to the oceans. While small-scale field trials and models attempt to capture local alkalinity generation after addition of crushed rock, its downstream delivery is modulated by hydrological and geochemical processes that unfold across watersheds. We synthesized data from natural volcanic watersheds as a model to evaluate the fate of alkalinity along complete reactive pathways from soil to river. Globally, natural basalt weathering accounts for 0.04 to 0.07 Gt CO_2 yr^{-1} CO_2 uptake. Fluxes from EW would have to be at least an order of magnitude greater to be climate-relevant. We examine why natural systems despite having ≈ 150 times greater rock loading than EW trials appear to be limited in CO_2 uptake to less than 1 t CO_2 ha^{-1} yr^{-1} and in most cases lower. Several processes limit alkalinity export in natural systems: Secondary mineral formation including oxy-hydroxides and clays generate acidity even if no base cations are consumed, and some clay formation reactions directly consume base cations. Carbonate minerals can form in a range of settings from soils to ground water to riparian zones and beyond. Transport processes can also limit alkalinity export. Soil water fluxes limits alkalinity export from soils. Alkalinity is strongly diluted at high discharge implying that stream hydrochemical data must capture the full range of hydrologic conditions to make accurate assessments of export. Because the net alkalinity export from large watersheds determines the impact of weathering reactions on the global CO_2 budget, these observations provide field-constrained upper limits on enhanced weathering derived alkalinity fluxes and highlight the need to incorporate whole watershed processes into future assessments of carbon dioxide removal. EW data that focus on the near-field subsurface zone often substantially overestimate the CO_2 uptake capacity of basalt applications at the relevant watershed scale.



Compilation of basaltic weathering rates from stream export data. Horizontal white lines and corresponding values indicate median weathering rates for each category. The grey solid line represents the median for a global compilation (grey violin) spanning basaltic regions.