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Representing surface proton reservoirs in reactive transport simulations of alkalinity generation and export during enhanced weathering

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Reactive transport models (RTMs) are increasingly used to evaluate mineral-driven alkalinity generation and transient pH changes in soils. In soils, the alkalinity from mineral dissolution interacts with the surface proton reservoir (SPR) associated with (oxy)hydroxides, clays and organic compounds, consuming alkalinity prior to export and complicating quantification. However, most soil weathering studies represent cation exchange and surface buffering using empirical capacities that are not thermodynamically linked to molecular-scale surface reactions. Here we develop and evaluate a framework that uses molecular-scale constraints from surface complexation theory to partition operational cation exchange measurements into electrostatic and pH-dependent coordinative surface reservoirs that can be embedded within a reactive transport model. This enables RTM simulations to explicitly couple the depth-dependent SPR, cation exchange, alkalinity generation, and $CO_2(g)$ exchange. We apply this approach to contrasting silicate and carbonate amendments across representative soil and hydrologic conditions. Using distance-based generalized sensitivity analysis, we identify the multivariate control space involving SPR magnitude, baseline soil pH, soil moisture, and amendment rate that governs alkalinity export efficiency. We find that amendment-derived alkalinity is largely neutralized during early dissolution, with substantial impairment occurring below the shallow (>25 cm) soil zone. In deeply weathered soils, alkalinity impairment can extend over meters. These findings also highlight the depth-integrated SPR as a first-order limitation on transient changes to soil pH and alkalinity export, and suggest appropriate screening and monitoring strategies for CDR quantification. Monitoring frameworks that prioritize depth-resolved alkalinity fluxes and explicitly resolve hydrologic uncertainty will be critical for evaluating the capacity of soil amendments to generate sufficient alkalinity to support long-term marine storage.