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Quantifying major element fluxes through a granitic forested watershed using reactive transport models highlights the importance of atmospheric inputs to ecosystem function

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In upland, steep terrain, soils are thin and forest ecosystems reach deep into weathering rock, where roots and water mobilize rock-derived nutrients. The chemical composition of plant biomass and stream water which drains these upland watersheds offer a record of this interplay between chemical weathering and ecosystem functioning. Numerical reactive transport models are increasingly utilized to quantify the geochemical evolution of natural ecosystems by incorporating experimentally constrained water-rock reactions. Exploring when and where these models fail to capture aspects of upland ecosystems opens the possibility to consider other physical, chemical and biological reactions not accounted for in the model that may have a significant influence on watershed structure and function. Here, we develop a model for a granitic weathering profile based on a small (0.54 km^2) watershed drained by Sapine creek, located on Mont Lozère in Les Cévennes, in southern France, which is part of the OZCAR French Critical Zone network. The model is allowed to reach steady state from an initially unweathered granite profile via constant rates of infiltration and uplift, taking approximately 350kya to achieve. Results are compared against solid phase elemental depletion measured between bedrock and soil samples as well as elemental ratios of the stream water. The resulting model weathering profile and aqueous discharge composition are in good agreement with our observations for the major elements Si, Na, K, and Mg. However, the simulations consistently fail to produce enough Ca accumulation in the shallow soils or stream water relative to measured values. Moreover, in our observed biomass composition (e.g. tree trunks, branches, plant litter) Ca is up to two orders of magnitude more concentrated than the other major elements. Thus, a significant mass influx of Ca exists for this watershed in addition to mineral weathering and precipitation. We suggest that atmospheric deposition of Ca is strongly tied to plant nutrient cycling. Here, we demonstrate the necessity and consider the appropriate methodology to incorporate atmospheric deposition into a reactive transport model such that the resulting weathering reaction network may be influenced by boundary conditions which are more complex than typical meteoric precipitation.