

Re-assessing the weathering signature of continental waters

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Chemical weathering mediates Earth's carbon cycle and hence global climate over geological time-scales. Ca and Mg from silicate minerals are released to the solute phase during dissolution with carbonic acid and subsequently buried as marine carbonate. This mechanism has provided the climatic feedback that has maintained Earth's climate equable over geological history. Quantitative models of contemporary silicate weathering processes coupled to estimates of modern day carbon fluxes associated with silicate weathering are therefore fundamental to understanding Earth's carbon cycle, and the feedbacks between the carbon cycle, climate and chemical weathering.

Estimating the Ca and Mg released from silicate weathering is not straightforward because their fluxes are dominated by carbonate weathering. Instead, contemporary silicate weathering fluxes are typically quantified based on Na and K fluxes in river waters because these elements are considered to be derived from silicate weathering (after correction for salt dissolution and rainfall inputs). Silicate Ca fluxes are based on the product of the Na flux and an average Ca/Na ratio of silicate rocks. This relies on the assumption that Na and K are predominantly released by silicate mineral dissolution.

However, it has been proposed that Na-Ca exchange reactions with clay on mineral surfaces could account for 80% of the Na in rivers waters. At present, none of the methods to estimate silicate weathering fluxes and associated CO₂ consumption account for cation-exchange reactions largely because physical and chemical weathering were assumed to be steady state processes implying that cation exchange has no net influence on weathering fluxes.

In tandem, there are numerous reports of stable isotope fractionation of the elements Mg and Li that are inferred to be induced by clay minerals. At present it is not clear whether this fractionation is associated with mineral surfaces (exchange) or structural incorporation into the clays. Here we will report Mg and Li isotope analyses on dissolved, exchangeable and structural Mg and Li from a series of large river systems to determine whether Mg and Li isotopes are fractionated by exchange processes and in turn whether they can help quantify the significance of exchange processes on controlling the chemistry of natural waters, and the apparent proportions of carbonate to silicate weathering.