Fate and transport in environmental systems: Integrating experiments, theory and reactive transport modeling

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Reactive transport modelling is a powerful, yet underutilized, tool in subsurface contaminant remediation science. Reactive transport models can be used to represent, analyze and, ultimately, predict the chemical reactivity and ecosystem functioning of subsurface environments. Therefore, they provide a means to assess to what extent biodegradation, abiotic reactions and physical transport processes determine the spatio-temporal distributions of contaminants and their degradation products. To build these models, mathematical expressions for the rates of the biogeochemical transformation processes that affect the distributions of relevant chemical constituents and microbial biomasses are needed. Here, an integrated, three-step approach is presented. (1) Kinetic experiments with simplified model systems are designed to identify possible reaction pathways, formulate rate expressions and obtain expected parameter ranges. (2) In situ potential rates and parameter values are constrained by kinetic experiments performed directly with natural sediment, soil or aquifer material. (3) Reactive transport modeling is used to verify the proposed rate expressions and to perform sensitivity tests. Once calibrated, reactive transport models can help explore biogeochemical activity in subsurface systems where many microbial, chemical and physical processes are coupled to one another. Various biogeochemical processes are used to illustrate the integrated approach, with an emphasis on processes that either control or depend on the local redox conditions. The strength of reactive transport modeling is further demonstrated for cases where slow microbial growth hinders a realistic simulation of the corresponding microbial processes in the laboratory.

Quaternary weathering rates and marine geochemical budgets

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Modern riverine chemistry and discharge is commonly used to estimate the long-term flux to the oceans of the many chemical and isotopic species that have their primary source in the continental crust. Here we suggest that measurements of many modern rivers, only a few thousand years after a glacial period, are not representative of the long-term flux and we quantitatively assess the implications of such a suggestion for key marine geochemical budgets.

The last deglaciation saw a profound change in the weathering regime in many parts of the planet - from intense physical grinding on a relatively cold, dry Earth to stronger chemical weathering on a warmer, wetter Earth. Experimental studies show that the fresh sediments produced in the glacial period would have undergone a pulse of rapid chemical weathering during the early post-glacial period. Moreover, these same experimental studies have also shown that both radiogenic and stable isotopes in this early chemical weathering pulse would match neither the bulk rock/soil composition nor the long-term weathering flux. Finally, the time constant of the pulse implies that today’s rivers still record more rapid weathering, and an unusual isotopic composition, than the long-term equivalents. The implication is that the material supplied by modern rivers to the oceans is not representative of the long-term flux.

These findings have major implications for long-standing problems in ocean chemistry. For example, while there is growing convergence between ocean floor hydrothermal fluid flux estimates based on independent tracer exchanges and simple thermal calculations, estimates based upon the oceanic 87Sr or Mg balances that rely on the quantification of the modern riverine flux are more than an order of magnitude greater. For elements such as Sr and Mg, with long oceanic residence times, modern riverine measured fluxes do not provide appropriate estimates of the long-term inputs. These ideas are relevant to many elements, and their isotope systems, with residence times in the oceans that are longer than the time period since the last deglaciation.