

## Stable and radiogenic isotope variations accompanying continental weathering

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Many natural isotope systems (both stable and radiogenic) are sensitive to variations in weathering process. The difficulty lies in distinguishing variations that result from weathering process from those caused by differences in rock type. One approach that circumvents this problem is the study of monolithologic catchments, where variations such as catchment age, runoff, physical and chemical weathering, vegetative cover and soil development can be related to riverine chemistry.

This study presents an overview of our recent work on radiogenic and stable isotope in rivers, soils and estuaries from catchments in Greenland, Iceland and Europe. Each case study illustrates the effects of weathering process on the riverine isotope signal, and the estuarine data indicates how this signal is transferred to the oceans. Aside from variations in rock type, the principal controls on isotope variations accompanying weathering are; (i) for many radiogenic isotope systems, preferential (incongruent) weathering of specific primary mineral phases, where those phases possess a markedly different parent/daughter and hence radiogenic isotope composition; (ii) for many stable isotope systems, preferential removal of an isotope into secondary phases formed during weathering, leaving residual waters depleted in that isotope. Recent studies have also shown the importance of biological removal and accompanying fractionation for certain isotope systems.

Taken together, these results clearly demonstrate that weathering processes can exert a significant effect on the isotope composition of natural waters, and for some of those isotopes marine sedimentary archives preserve a record of the changes in such processes with time. For marine records the challenge remains in unravelling the effects of weathering from those caused by variations in rock type, or a simple change in flux or geographical source.

## The CO<sub>2</sub> sealing efficiency of caprocks

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Shale lithotypes of various compositions occur abundantly in sedimentary basins and act as natural seals for petroleum and natural gas reservoirs over extended geologic periods. Based on their mechanical, petrophysical and chemical/mineralogical properties, shales are becoming increasingly important in the context of geological long-term storage of anthropogenic carbon dioxide in saline aquifers, depleted oil and gas reservoirs and coals.

Due to its chemical reactivity and physico-chemical properties, CO<sub>2</sub> is expected to differ substantially from other natural gas components in terms of transport behaviour and interaction with the mineral/water system. An experimental procedure has been developed to measure molecular diffusion of CO<sub>2</sub> in water-saturated shales. This non-steady state method provides information on the effective diffusion coefficients and the CO<sub>2</sub> storage capacity of the shales. Storage capacities were found to vary significantly but can be as high as 0.14 mmol CO<sub>2</sub>/g sample. Manometric sorption experiments with CO<sub>2</sub> at pressures up to 20 MPa have been performed on dry and moist shale samples to verify the findings noted above. The results of these experiments revealed unexpectedly high storage potentials of the same order of magnitude as those from the diffusion measurements. The CO<sub>2</sub> storage capacities do not correlate with organic carbon content. A separate set of experiments with pure clay minerals (Kaolinite, Illite, Montmorillonite...) revealed that some of these materials had high sorptive CO<sub>2</sub> storage potentials. Comparison of pre- and post-experiment X-ray diffraction patterns showed indications of changes in mineralogy

These findings provide a new view on the issue of caprock integrity. In addition to their sealing properties, natural shale sequences could represent a significant sink or buffer for carbon dioxide deposited in the subsurface by fixing and immobilising it and hence reduce the risk of leakage to the surface.