U-series disequilibrium investigation of a weathering profile in a tropical granitoid watershed, Luquillo, Puerto Rico

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Understanding how weathering profiles develop over time is a key to understanding biogeochemical cycling and the controls on chemical fluxes from land to the oceans. Isotopic tracers that identify and date chemical transformations in those profiles can provide essential knowledge for understanding profile development and calculating watershed scale weathering rates.

We have investigated the use of U-series disequilibrium to examine weathering profile development in the Rio Icacos watershed in the Luquillo Mountains of Puerto Rico. In order to further constrain interpretations based on U-series data, we have coupled the U-series analyses with analyses of trace element concentrations and Ge/Si and $^{87}Sr/^{86}Sr$ ratios. Used together, these geochemical tracers provide a powerful tool for understanding weathering reactions, chemical transfers within and out of the profile, and the timing of those chemical transfers.

$^{234}U/^{238}U$ activity ratios in soil, saprolite and pore water samples reveal a complex history of U transformations dominated by two distinct weathering fronts in the Rio Icacos profile. Significant disequilibria exists between deep saprolite samples (8 m) and pore waters from the same depth, with higher $^{234}U/^{238}U$ in pore waters reflecting α-recoil emplacement of ^{234}U during weathering. $^{234}U/^{238}U$ activity ratios in both saprolite samples and pore waters decrease towards the surface of the profile. $^{234}U/^{238}U$ activity ratios in the soil (top 50 cm) are distinct in that the pore waters and soil samples have very similar $^{234}U/^{238}U$ activity ratios, a lack of fractionation reflecting more intense weathering conditions in this zone. Trace element and REE abundance patterns throughout the profile illustrate redistribution of elements from shallower saprolite to a zone of accumulation between 4 and 7 m depth. These patterns also reveal a distinct enrichment in the top 30 cm of the profile indicating a significant atmospheric contribution that must be taken into account. Finally, simple modeling exercises are undertaken to examine possible scenarios for the observed patterns in U/Th and $^{234}U/^{238}U$ and $^{230}Th/^{238}U$ activity ratios.

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Marine $^{87}Sr/^{86}Sr$ record mirrors the evolving upper continental crust

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Prior the discovery of hydrothermal vents in 1977 temporal variations in the marine $^{87}Sr/^{86}Sr$ were generally thought to reflect changes in the composition of the continental crust (Brass, 1976). Since 1977 the marine $^{87}Sr/^{86}Sr$ record generally has been interpreted as two-component mixture of radiogenic continental and unradiogenic hydrothermal fluxes of Sr to seawater (e.g., Palmer and Edmond, 1989).

We have quantitatively investigated the relationship between riverine $^{87}Sr/^{86}Sr$ and the geologic makeup of drainage basins. We find striking positive correlations between dissolved riverine $^{87}Sr/^{86}Sr$ and average bedrock ages in the Fraser and Mississippi river basins. These findings point to a predominant geologic control of riverine $^{87}Sr/^{86}Sr$. A global analysis of bedrock geology indicates that sediments and volcanic rocks are generally young (average ages of 246 ± 42 and 331 ± 55 Myr, respectively) whereas plutonic and metamorphic rocks are much older (1745 ± 248 Myr). A GIS analysis of 19 large-scale drainage basins that cover the entire non-glaciated Earth surface, combined with a Sr isotope database for 112 rivers draining these basins reveals a positive correlation between bedrock age (or lithology) and dissolved $^{87}Sr/^{86}Sr$. This confirms that the relationships found in individual drainage basins are characteristic for large-scale drainage basins in general.

One endmember interpretation of the marine $^{87}Sr/^{86}Sr$ record casts temporal variations in the marine $^{87}Sr/^{86}Sr$ as variations in the average bedrock age (or lithology) of the continental crust. Ill-constrained temporal variations of unradiogenic hydrothermal fluxes to the ocean are, for now, neglected. Such an analysis indicates that the eroding crust has undergone progressive rejuvenation from the early Cambrian to the Late–Mid Jurassic, followed by only minor changes until the trend reversed in the Eocene. Interestingly, the Paleozoic and Mesozoic trend broadly resembles temporal variations in the makeup of the continental bedrock as reconstructed by Ronov and his coworkers (Ronov et al., 1989) and quantitatively evaluated by Bluth and Kump (Bluth and Kump, 1991).

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