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Strontium Isotope Systematics During Chemical Weathering of Granitoids: Importance of Relative Mineral Weathering Rates

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Introduction

The application of strontium isotopes as tracers of biogeochemical processes on both the catchment and global scales has increased considerably in recent years. In such studies it is generally assumed that for a given composition and age of bedrock there is a characteristic $^{87}Sr/^{86}Sr$ released by mineral weathering. But recently many researcher's results indicate that non-stoichiometric release of Sr from dissolving minerals occurs during initial stages of dissolution, and the $^{87}Sr/^{86}Sr$ ratio released by natural weathering of crystalline rocks can deviate significantly from bedrock values (Blum and Erel, 1997; Bullen et al., 1997; Brantley et al., 1998). Furthermore, ambiguities remain concerning the factors controlling the release of strontium and temporal evolution of $^{87}Sr/^{86}Sr$ released during *in situ* chemical weathering of silicate rocks. Clearly there is still a need for additional information about the behaviour of Sr in mineralogically-complex weathering systems.

Methods

In this study we primarily investigate the mineral and strontium isotopic composition, major and trace element concentrations of three weathering profiles developed on granitic parent rocks in southern China, and use the $^{87}Sr/^{86}Sr$ ratios of exchangeable and whole-sample digestion Sr to evaluate the relative weathering rates of several important granitic minerals.

Results and Discussion

The weathering profiles studied are developed on granite, granite porphyry and metamorphic rocks, respectively, which have different maturity controlled by the balance between erosion and chemical weathering. With the increase of chemical weathering intensity (CIA increases), alkalis and alkaline earths (e.g., Rb, Sr, Na, K, Cs, Ca, Ba) are rapidly leached away, resulting in the significant decrease of element/Ti ratios from the bottom to top of the profiles. Meanwhile, Rb/Sr ratios increase with increasing CIA, implying the differential losses of Rb and Sr. A positive correlation was observed between the $^{87}Sr/^{86}Sr$ ratios of whole-sample digestion Sr and Rb/Sr (or $^{87}Rb/^{86}Sr$) ratios, which is resulted from the weathering of a homogeneous source rock.

The Ca/Si ratios of weathered samples steeply decrease with CIA during early stages of weathering, and calculations indicate that calcite in the parent rocks was completely removed during incipient weathering of granitoids. With the increase of chemical weathering intensity (in particular when CIA is less than 70), the Sr/Ca ratios of weathered samples significantly decrease compared to the parent rock values, implying the preferential losses of Sr over Ca. While Rb/K ratios in these profiles almost keep constant regardless of the weathering intensity, suggesting the congruent release of Rb and K from the weathering profiles.

The most striking is the fluctuation of whole-sample digestion $^{87}Sr/^{86}Sr$ throughout the weathering profiles. According to the temporal evolution of $^{87}Sr/^{86}Sr$, along with the mineralogical variations in weathering profiles, the whole weathering process can be divided into four distinct stages. During incipient weathering of granitoids (CIA<65), the whole-sample digestion $^{87}Sr/^{86}Sr$ slightly decreases, which is affected by partially weathering of biotite to vermiculite. And then a dramatic and systematic increase is observed in the $^{87}Sr/^{86}Sr$ ratio of whole-sample digestion Sr with increasing weathering extent, which correlates with enhanced plagioclase weathering rate. In the third stages, the $^{87}Sr/^{86}Sr$ ratio gradually decreases indicative of the weathering of K-feldspar. Finally all minerals release Sr at a steady-state rate, therefore the $^{87}Sr/^{86}Sr$ ratios of both whole-sample digestion Sr and ammonium-acetate extractable Sr keep constant with increasing CIA. Accordingly the $^{87}Sr/^{86}Sr$ ratios dissolved strontium in stream water or groundwater fluctuates in the shape opposite to that of whole-sample digestion Sr.

In summary, the temporal evolutions of $^{87}Sr/^{86}Sr$ ratios of whole-sample digestion Sr and exchangeable Sr are controlled by the change of relative weathering rates of several important granitic minerals.

The above results imply that Sr release during initial weathering of granitoids is highly non-stoichiometric relative to the bedrock mineralogy, and weathering-derived $^{87}Sr/^{86}Sr$ values fluctuate with increasing age, and will not be equal to the bulk Sr signature. This factor must be considered in the investigation of nutrient cycling, weathering fluxes, and the chemical evolution of the oceans based on strontium isotopes.

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