

Sr isotope evolution during chemical weathering of granites

— impact of relative weathering rates of minerals

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Abstract The Sr isotopic systematics in the weathering profiles of biotite granite and granite porphyry in southern Jiangxi Province were investigated. The results showed that during the chemical weathering of granites, remarked fractionation occurred between Rb and Sr. During the early stages of chemical weathering of granites, the released Sr/Si and Sr/Ca ratios are larger than those of the parent rocks, and the leaching rate of Sr is higher than those of Si, Ca, K, Rb, etc. Dynamic variations in relative weathering rates of the main Sr-contributing minerals led to fluctuation with time in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of inherent and released Sr in the weathering crust of granite. Successive weathering of biotite, plagioclase and K-feldspar made $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the weathering residues show such a fluctuation trend as to decrease first, increase, and then decrease again till they maintain stable. This work further indicates that when Sr isotopes are used to trace biogeochemical processes on both the catchment and global scales, one must seriously take account of the preferential release of Sr from dissolving solid phase and the fluctuation of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios caused by the variations of relative weathering rates of Sr-contributing minerals.

Keywords: Sr isotope, chemical weathering, weathering rate, non-stoichiometry, granite.

In recent years, Sr isotopes have been widely used to trace various biogeochemical processes on both the catchment and global scales^[1, 2]. In such studies, it is generally assumed that for a given composition and age of bedrock there is a characteristic $^{87}\text{Sr}/^{86}\text{Sr}$ released by mineral weathering, which is independent of the age of the weathering surface. However, several recent investigations suggest that Sr will be incongruently released from dissolving minerals (e.g. feldspars)^[3], and with increasing weathering age, and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of Sr released from weathering soil tends to decrease gradually^[4–7]. Meanwhile, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in riverine weathering fluxes can reflect the types and rates of weathering reactions in the weathering rocks and soils in a river basin^[8].

But, even up to now, little work has been done on the mechanism of variation of $^{87}\text{Sr}/^{86}\text{Sr}$ in the processes of formation and development of a natural weathering profile, which represents the whole process of continental chemical weathering from fresh parent rock to surface soil, particularly on the laws of Sr release and Sr isotope evolution during natural weathering of mineralogically-complex silicate rocks. In this paper, detailed studies have been carried out on the distributions of Rb and Sr and their isotopes in two well-developed granite weathering profiles in southern Jiangxi Province in an attempt to reveal the fluctuation of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio during chemical weather-

ing. Moreover, it is also pointed out that variations in relative weathering rates of different Sr-contributing minerals are the key factor leading to such $^{87}\text{Sr}/^{86}\text{Sr}$ fluctuation.

1 Sampling and analytical method

1.1 Sample collection

The studied area includes Longnan ($24^{\circ}54'\text{N}$, $114^{\circ}47'\text{E}$) and Xunwu ($24^{\circ}57'\text{N}$, $115^{\circ}38'\text{E}$) in southern Jiangxi Province, where there is warm and humid climate and gentle hill-marked topographical features and various types of lateritic weathering mantles are widespread. Two typical weathering profiles (a biotite granite weathering profile and a granite porphyry weathering profile) were investigated in this study^[9]. The biotite granite weathering profile (JL-1), located in Jiangtou Village, Dongjiang, Longnan, develops on a medium-grained biotite granite (134—155 Ma B.P.) composed mainly of quartz (25%—30%), plagioclase (30%), K-feldspar (35%), biotite (5%) and muscovite (2%). The granite porphyry weathering profile (JX-3), located in an ion-adsorbed type of REE quarry in southeast Xunwu, develops on an Upper Jurassic granite porphyry (135—160 Ma B.P.), and in the parent rock, the phenocryst is composed largely of quartz (15%—30%), K-feldspar (15%—40%), plagioclase (40%), biotite (2%) and the matrix is felsic. Both profiles are 25 m thick in vertical cut.

Samples were systematically taken from each of the horizons described as the humic soil layer, lateritic layer, weathered layer, sub-weathered layer, weathering front and parent rock. After quartering on the spot, the samples were sealed in polyethylene bags and taken back to the laboratory. Owing to intense chemical weathering, no fresh parent rock was exposed at the bottom of profile JX-3.

1.2 Analytical method

After plant fragments removed, all the samples were air-dried and ground to < 200 mesh in an agate mortar. Major elements were analyzed by wet chemical methods with errors less than 0.5%. After the samples were decomposed in Teflon bomb, trace elements were analyzed by ICP-MS, with an accuracy less than 5%. For quality control, blanks and known standards for each element were treated exactly as the samples were.

After the samples were acid-dissolved, Sr was separated from other ions by the cation exchange method and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were measured on an MAT-260 mass spectrometer. NBS987 yielded a standard value of $0.710\,25 \pm 2$ during the period of analysis, and the average analytical error was ± 0.00002 (2σ).

2 Results

2.1 Variations of Rb and Sr contents in the weathering profiles

Listed in table 1 are the contents of Si, Ca, K, Sr and Rb and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the weathering profiles. After normalization to the most conservative element $\text{Ti}^{[9]}$, the mobility of alkali and alkali-earths during weathering can be discussed. The results showed that with the increase of

chemical weathering intensity, the elements K, Ca, Na, Mg, Rb, Sr, Cs, and Ba were rapidly leached away from the profiles. Studies on element distributions in different grain-sized fractions indicated that Rb and Sr are relatively enriched in the fine grain-sized fractions, and Rb is much richer than Sr in clay-sized materials.

Chemical weathering always results in fractionation between Rb and Sr. As can be seen from table 1, with increasing CIA (chemical index of alteration) in the biotite granite profile (JL-1), the Rb/Sr ratios of the weathered samples increase rapidly, indicating that the leaching rate of Sr is greater than that of Rb. While in the granite porphyry profile (JX-3), at the initial stage of weathering ($\text{CIA} < 75$), the Rb/Sr ratios tend to increase with increasing CIA. With further intensification of chemical weathering, the Rb/Sr ratios decrease rapidly, implying that the activity of Rb is stronger than that of Sr. Changes in distributions of Rb and Sr in the weathering profiles may be attributed to their modes of occurrence, medium conditions, the formation of secondary minerals (e.g. carbonates), especially the migration of clay-sized materials within the weathering profiles, and in addition, to their differences in chemical properties^[7,10–13].

Differences in activity of Rb and Sr must lead to the variations of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio during chemical weathering. And there is always a positive correlation between $^{87}\text{Sr}/^{86}\text{Sr}$ and Rb/Sr (fig. 1). Minerals with high Rb/Sr ratios usually have high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and these minerals are more resistant to chemical weathering^[14]. During chemical weathering, Sr is different in activity from Rb, and there would occur fractionation between common Sr and radiogenic Sr^[11]. In the case of little external disturbance, if ^{87}Sr in the weathering crust was mainly derived from decay of Rb, the decomposition of Rb-bearing minerals will lead to contemporaneous loss of both Rb and ^{87}Sr from the weathering profiles, hence resulting in a better positive correlation between Rb/Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (fig. 1).

2.2 Fluctuations of $^{87}\text{Sr}/^{86}\text{Sr}$ in the weathering profiles

The most striking contrast is the obvious fluctuations in whole-sample digestion $^{87}\text{Sr}/^{86}\text{Sr}$ ratios with increasing CIA throughout the weathering profiles (fig. 2). According to the degrees of chemical weathering of the samples, in conjunction with variations in mineral composition and content in the profiles^[9], the evolution of $^{87}\text{Sr}/^{86}\text{Sr}$ with time in the whole process of chemical weathering can be divided into four distinct stages. (i) The slightly decreasing stage of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ($\text{CIA} < 65$). In the initial stage of granite weathering, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios show a tendency of slightly decreasing, particularly in the biotite granite weathering profile. (ii) The rapidly increasing stage of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio ($65 < \text{CIA} < 70$). With intensification of chemical weathering, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the weathered samples tend to increase rapidly till their maximum values occur. (iii) The $^{87}\text{Sr}/^{86}\text{Sr}$ decreasing stage. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the weathering residues tend to decrease gradually from their maximum values till reaching constant values. (iv) The relatively stable stage ($\text{CIA} > 90$). At this time, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Sr released during weathering are generally equal to those of the weathering residues^[6].

Table 1 The major element contents and Sr isotopic ratios in the granite porphyry (JX-3) and biotite granite (JX-1) weathering profiles, southern Jiangxi, China^{a)}

Sample No.	Depth ^{b)} /m	CIA ^{c)}	SiO ₂ (%)	CaO (%)	K ₂ O (%)	Sr/10 ⁻⁶	Rb/10 ⁻⁶	Rb/Sr	⁸⁷ Sr/ ⁸⁶ Sr
JX-3-1	top soil	97.48	68.02	0.06	0.15	4.166	12.612	3.027	0.716687
JX-3-2	0.3	97.28	58.95	0.07	0.12	5.086	12.101	2.379	0.724535
JX-3-3	0.8	97.30	58.45	0.08	0.12	3.883	13.164	3.390	—
JX-3-4	1.8	97.82	60.68	0.03	0.13	6.725	11.946	1.776	0.721599
JX-3-5	2.3	97.20	63.06	0.07	0.09	2.416	12.799	5.298	—
JX-3-6	3.8	95.04	64.20	0.10	0.43	21.330	22.152	1.039	0.721236
JX-3-7	4.8	92.89	61.43	0.07	0.84	18.211	37.071	2.036	0.718928
JX-3-8	5.8	83.08	64.95	0.10	2.10	25.945	86.857	3.348	0.724353
JX-3-9	7.0	88.09	65.17	0.07	1.64	15.859	61.174	3.857	—
JX-3-10	8.0	86.92	65.86	0.08	1.63	16.029	62.274	3.885	0.724128
JX-3-11	9.1	80.53	66.87	0.10	3.03	17.483	108.508	6.206	—
JX-3-12	10.2	77.56	66.65	0.40	3.12	15.758	123.692	7.849	0.739809
JX-3-13	11.5	73.92	68.09	0.10	4.14	26.618	163.493	6.142	—
JX-3-14	13.0	68.25	70.16	0.60	4.61	14.268	191.214	13.402	0.764481
JX-3-15	14.5	80.25	68.01	0.30	2.72	7.355	117.190	15.933	—
JX-3-16	16.0	71.63	72.87	0.10	3.90	10.892	166.155	15.255	0.772282
JX-3-17	17.5	70.66	70.82	0.11	4.46	13.508	198.559	14.699	0.769460
JX-3-18	19.0	65.57	69.92	0.20	6.40	26.700	229.027	8.578	0.744755
JX-3-19	23.5	63.33	68.61	0.80	6.60	24.593	231.383	9.408	0.746505
JL-1-17	top soil	78.22	68.29	0.07	3.94	3.681	449.571	122.133	1.181719
JL-1-16	0.4	84.44	63.74	0.08	3.40	2.625	382.737	145.805	1.01225
JL-1-15	0.8	82.66	65.47	0.07	3.35	2.504	371.541	148.379	—
JL-1-14	1.5	78.50	74.41	0.10	3.08	2.265	332.729	146.900	1.128163
JL-1-13	2.5	78.19	71.75	0.07	3.81	3.282	405.958	123.692	—
JL-1-12	4.0	73.11	73.94	0.06	4.26	3.116	461.053	147.963	—
JL-1-11	6.5	74.74	75.37	0.07	3.75	3.723	395.613	106.262	—
JL-1-10	8.5	75.21	70.64	0.06	4.58	3.380	484.247	143.268	1.194982
JL-1-9	10.0	73.77	75.98	0.02	3.89	2.957	414.288	140.104	—
JL-1-8	11.5	71.51	75.64	0.07	4.45	4.137	492.205	118.976	1.242312
JL-1-7	13.0	65.63	74.32	0.07	4.45	7.179	515.177	71.762	1.133989
JL-1-6	14.0	62.98	74.62	0.10	4.36	12.738	535.597	42.047	0.920487
JL-1-5	15.0	61.98	74.83	0.11	4.08	13.475	517.454	38.401	—
JL-1-4	17.0	59.15	76.47	0.10	4.30	14.605	518.868	35.527	0.926357
JL-1-3	19.0	60.47	75.59	0.11	4.14	17.941	474.391	26.442	0.894698
JL-1-2	22.5	58.89	76.75	0.10	4.09	13.542	477.224	35.240	0.951822
JL-1-1	parent rock	56.12	76.25	0.11	4.43	15.419	528.512	34.277	0.950386

a) Major and trace elements were analyzed at the Institute of Geochemistry, CAS. Sr isotopic ratios were analyzed at the Institute of Mineral Deposits, Chinese Academy of Geological Sciences. —, Not analyzed; b) the depth refers to the vertical distance from the surface downwards; c) CIA (chemical index of alteration) = $[\text{Al}_2\text{O}_3/(\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O})] \times 100$, where the oxides are given in molar proportions, CaO^* represents CaO in silicate minerals with the exception of that in carbonate and apatite, and CIA can reflect the degree of chemical weathering of samples. The data in the table are from Ma Yingjun^[9].

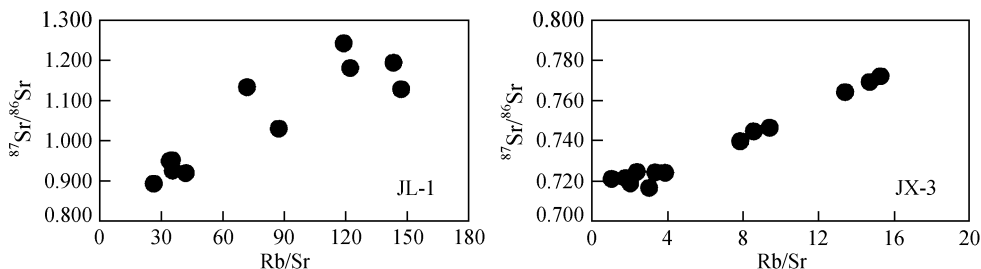


Fig. 1. Rb/Sr versus $^{87}\text{Sr}/^{86}\text{Sr}$ correlation diagram in the weathering profiles. JL-1, the biotite granite weathering profile; JX-3, the granite porphyry weathering profile.

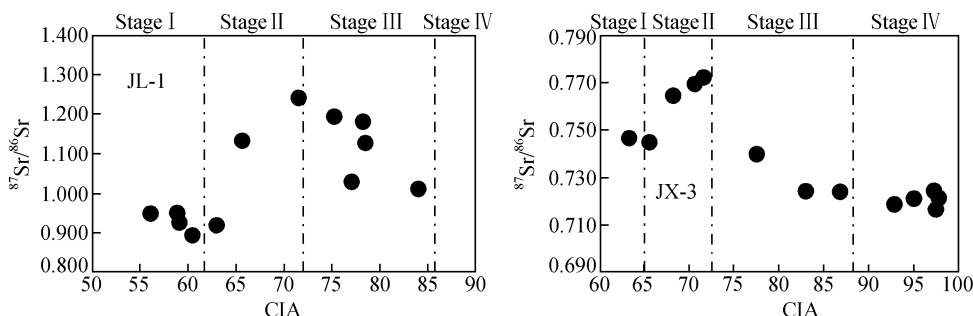


Fig. 2. Variations of $^{87}\text{Sr}/^{86}\text{Sr}$ in the weathering profiles with CIA. JL-1, the biotite granite weathering profile; JX-3, the granite porphyry weathering profile.

Comparatively speaking (fig. 2), $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the granite porphyry weathering profile ($63 < \text{CIA} < 98$) just slightly decreased at its first stage because no parent rock samples were collected from the bottom of the profile. While the biotite granite weathering profile ($56 < \text{CIA} < 85$) did not yet develop to its forth stage, the stable stage of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio could not yet be observed.

3 Discussion

3.1 Nonstoichiometry of strontium release during rock weathering

When Sr isotopes are used to trace the cycling of materials on the Earth's surface, it is generally assumed that Sr release from dissolving solid phase is stoichiometric, and there is no preferential release of Sr in comparison with other cations and also no variation with time in release rates of cations. However, recent experiments made by Brantley et al.^[3] showed that non-stoichiometric release of Sr from dissolving feldspars occurred during the initial period of dissolution and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of released Sr are not equal to ratios in the dissolving minerals.

As shown in fig. 3, with increasing CIA in the biotite granite weathering profile, the Sr/Si ratio of the samples tends to decrease gradually. In the case of $\text{CIA} < 70$, the Sr/Si ratio decreases more rapidly, indicating that the Sr/Si ratio released during chemical weathering is larger than that of the parent rock. Especially during the early period of chemical weathering the releasing rate of Sr is much greater than that of Si. Meanwhile, the gradual decrease of Sr/Ca ratio in the weathering residues suggests that Sr was released in preference to Ca. Comparatively, no remarkable

variation is observed in Rb/K ratio in the profiles, which implies that both Rb and K are released in stoichiometric proportions during chemical weathering of granite.

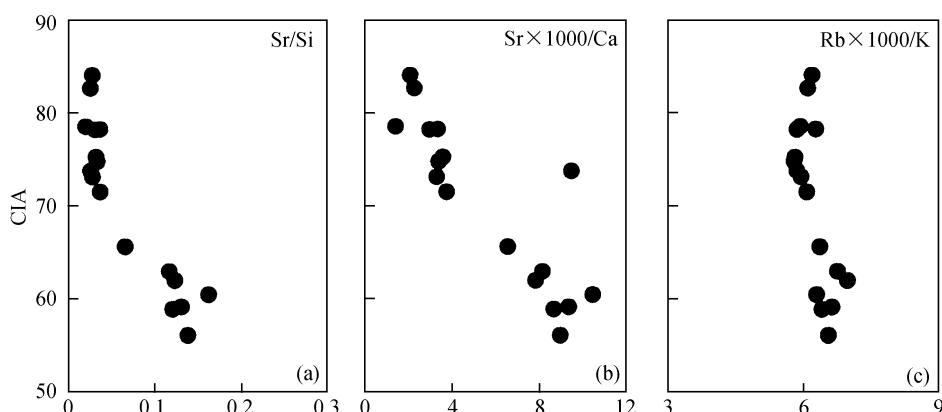


Fig. 3. Variations in Sr/Sr, Sr/Ca and Rb/K in the biotite granite weathering profile with CIA. The element ratios in the figure are calculated on a molar basis.

Preferential release of Sr is also observed during chemical weathering of granite porphyry (table 1). But when the chemical weathering developed to a quite intense degree (e.g. CIA>90), the Rb/K ratio of the weathered samples would rise rapidly, implying that the leaching rate of K is greater than that of Rb in the soil layer, which may be related with the preferential adsorption of Rb on clays^[10].

In general, during the early period of chemical weathering, Sr would be released in preference to Si, Ca, K, Rb, etc. from dissolving granites into surface runoff. In granites, difference in dissolving rates of different minerals and differences in mineral micro-structures (e.g. inclusions, lattice defects, etc.) are the main factors leading to the nonstoichiometric release of Sr^[3,7,15]. Preferential release of Sr from the weathering crust will necessarily lead to deviations in Sr concentrations and ⁸⁷Sr/⁸⁶Sr ratios in soils from those of the parent rocks^[5–8].

3.2 Variations in ⁸⁷Sr/⁸⁶Sr ratio and relative weathering rate of minerals

The fact that ⁸⁷Sr/⁸⁶Sr ratios in the granite weathering profiles vary from one stage to another (fig. 2) indicates that the ⁸⁷Sr/⁸⁶Sr ratio of Sr released during chemical weathering is not equal to that of the parent rock, and that dynamic variations in relative weathering rate of different minerals during weathering may be the main factors leading to the variations of ⁸⁷Sr/⁸⁶Sr with time.

⁸⁷Sr/⁸⁶Sr ratios in the primary minerals of granites tend to increase progressively in order of plagioclase→K-feldspar→biotite^[5, 7, 14]. So during chemical weathering, with the variation of predominating weathering mineral phases, the ⁸⁷Sr/⁸⁶Sr ratio of Sr released from weathering rocks will vary continuously. In the following section, a detailed discussion will be given to the impact of relative weathering rates of minerals on the Sr isotopic composition during weathering based on studies of variations in mineral composition and contents in the weathering profiles^[9].

At the first stage of granite weathering (fig. 2), ⁸⁷Sr/⁸⁶Sr ratios in the weathered samples tend

to decrease gradually. This phenomenon is particularly noticeable in the biotite granite weathering profile. In the process of alteration of biotite to vermiculite Fe^{2+} in octahedral lattice site will be oxidized to Fe^{3+} . To maintain charge balance in the whole mineral, K^+ and Sr^{2+} of the interlayer cations will always be expelled rapidly, leading to the loss of radiogenic Sr with a high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio^[7]. At the same time $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the residues tend to decrease gradually. Generally, the rate of alteration of biotite to vermiculite is at least 4—8 times the weathering rate of plagioclase in fresh parent rocks^[7, 8].

With intensification of the chemical weathering of granites (Stage II), plagioclase weathering became dominant. In the process of decomposition of plagioclase common Sr with a low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio would be leached away, thus leading to the rapid increase of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in weathering residues. As can be seen from fig. 2, the CIA values lie between 70 and 75 in both the weathering profiles when the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of weathered samples reach their highest values. Studies on the variations of trace elements and mineral composition in the weathering profiles indicated^[9] that in the case of CIA=70, plagioclase has been already decomposed completely, during which the Sr with a low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio has also been largely leached away. It is obvious that the extensive and rapid decomposition of plagioclase is an essential factor leading to the rapid increase of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios at this stage.

At the third stage of granite weathering (fig. 2), $^{87}\text{Sr}/^{86}\text{Sr}$ ratio tends to decrease gradually. Variations in trace elements and mineral composition in the weathering profiles provide evidence^[9] suggesting that at this stage ($70 < \text{CIA} < 90$) the weathering rates of K-contributing minerals (e.g. K-feldspar, illite and residual biotite) were enhanced relatively, leading to the release of K, Rb, and radiogenic Sr with a high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio. As a result, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the weathering residues tend to decrease gradually.

With further intensification of chemical weathering (e.g. stage IV in profile JX-3 as shown in fig. 2), $^{87}\text{Sr}/^{86}\text{Sr}$ ratio generally remains unchanged, indicating that most of Rb- and Sr-contributing primary minerals in the weathering profiles have been completely decomposed and the residual Rb and Sr are being released congruently along with other cations from the weathering profiles. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of Sr released at that time is equal to that of the weathering materials.

As is known from the principle of mass conservation, corresponding to the fluctuations of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in weathering residues, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Sr lost along with weathering fluxes (e.g. river water, underground water, etc.) would necessarily show such a similar trend as to vary from one weathering stage to another in weathering profiles, though the variation trend of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios should be opposite to that of the weathering residues. Of course, the migration of Sr within the weathering profiles, exchange reaction between Sr and clay minerals, and the input of atmospheric Sr may influence the isotopic composition of Sr in the weathering crust and surface runoff^[5—7, 16, 17]. However, the impact of atmospheric deposition is usually restricted to the surface soil layers of the profiles, and what is mainly influenced by the atmospheric deposition is the iso-

topic ratio of exchangeable Sr. In addition, in most cases, the exchangeable Sr only accounts for about 1% of the total Sr^[7]. So in this study atmospheric deposition is not responsible for the variations in ⁸⁷Sr/⁸⁶Sr ratios in the weathering profiles.

4 Conclusions and implications

To sum up, the following conclusions can be drawn: (i) In the process of natural weathering of granites remarked fractionation occurred between Rb and Sr, and the release of Sr from the dissolving rocks was obviously nonstoichiometric. (ii) During chemical weathering of granite dynamic variations in relative weathering rate of different minerals led to variations with time in ⁸⁷Sr/⁸⁶Sr ratios of the weathering residues and the ⁸⁷Sr/⁸⁶Sr ratios of Sr released from weathering crust. Only when the weathering crust evolved to a certain stage, could Sr be released congruently from it along with other cations at a steady-state rate.

This work further suggests that not only the types of weathering rocks and continental weathering rates can change global riverine Sr fluxes^[4, 14], but variations in relative weathering rates of Sr-contributing minerals during chemical weathering of mineralogically-complex rocks may also be an important mechanism affecting ⁸⁷Sr/⁸⁶Sr ratios in Earth's surface runoff. For this reason, in estimating resources and fluxes of nutrient cations or reconstructing the history of continental weathering and paleo-environmental change based on strontium isotopes, it is imperative to consider whether Sr is released in preference to other cations during chemical weathering of soils and whether the releasing rate of Sr varies with time. Only after a proper correction for the Sr isotopic ratios in terms of different dissolving rates of Sr-contributing minerals, can a true conclusion be drawn.

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