

ケイ酸塩風化の見かけの活性化エネルギーに対する大気組成の影響 Effects of atmospheric composition on apparent activation energy of silicate weathering

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Silicate weathering is a major sink of atmospheric CO₂. Because CO₂ is an important greenhouse gas, silicate weathering regulates not only the partial pressure of atmospheric CO₂ (P_{CO_2}) but also the surface temperature (T). The apparent activation energy of silicate weathering represents the temperature dependence of silicate weathering and thus interrelates the intensity of silicate weathering, P_{CO_2} and surface temperature. It has been reported that solution composition can affect the apparent activation energy of dissolution/precipitation of silicates (e.g., Casey and Sposito, 1992; Lasaga, 1995; Cama et al., 1999). However, the relationship between the solution composition and the apparent activation energy of silicate reaction is not yet fully understood.

To investigate the apparent activation energy of silicate weathering in a natural weathering system, we formulated the apparent activation energy of silicate weathering in three different scales, namely, (i) dissolution/precipitation of each mineral, (ii) elemental loss as the net reactions of the minerals and (iii) weathering flux from a weathering profile, based on the rate expressions in the three scales. It was found that, due to the effects of solution composition on the apparent activation energy, the temperature dependence of atmospheric CO₂ ($\Delta H^*_{\text{CO}_2}$) affects the apparent activation energy of silicate weathering. Based on the formulated apparent activation energy, we estimated the apparent activation energy of silicate-weathering flux as a function of $\Delta H^*_{\text{CO}_2}$. Then, the compensation law between the pre-exponential factor and the apparent activation energy of silicate-weathering flux was introduced from the literature, leading to the establishment of the relationship between silicate-weathering flux (F_{CO_2}), T and $\Delta H^*_{\text{CO}_2}$.

Based on the F_{CO_2} - T - $\Delta H^*_{\text{CO}_2}$ relationship and the greenhouse effects of atmospheric CO₂ in the literature, we calculated the ratio of change in F_{CO_2} to that in P_{CO_2} as an indicator of silicate-weathering feedback in the Precambrian. The calculation revealed that when $P_{\text{CO}_2} > \sim 10^{-0.5}$ atm, the feedback is negative and independent of P_{CO_2} and surface temperature. On the other hand, when $P_{\text{CO}_2} < \sim 10^{-0.5}$ atm, the feedback is independent of P_{CO_2} but dependent on surface temperature; at low ($< \sim 30$ °C) and high ($> \sim 30$ °C) temperatures, the feedback is negative and positive, respectively. Due to the positive feedback, the conditions of $P_{\text{CO}_2} < \sim 10^{-0.5}$ atm and $T > \sim 30$ °C are unstable, and immediately change, with a slight change in P_{CO_2} , to either the conditions of $P_{\text{CO}_2} > \sim 10^{-0.5}$ atm or those of $P_{\text{CO}_2} < \sim 10^{-0.5}$ atm and $T < \sim 30$ °C. When $P_{\text{CO}_2} < \sim 10^{-0.5}$ atm and $< \sim 30$ °C, the feedback is not only negative, but also becomes more negative as temperature decreases, suggesting that global glaciations are harder to bring about than previously thought.

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