

## **Silicon Isotope Doping Method for Measuring Silicate Reaction Rates in the Critical Zone**

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We used the Si isotope and measured silicate reaction rates at ambient temperature, near neutral pH, and close to equilibrium conditions, which are unattainable with the traditional Si concentration-based method. The isotope doping technique introduces an enriched rare isotope of an element to an experimental solution that is interacting with a mineral or a suite of minerals that have normal or natural isotopic compositions<sup>[1,2]</sup>. Although the method itself is not new, MC-ICP-MS technology now makes the measurement of a large number of non-traditional isotopes accessible, presenting a new opportunity to utilize the isotope doping method to advance the field of geochemical kinetics.

Results from experiments for albite, K-feldspar, and quartz<sup>[3,4]</sup> demonstrated that the isotopic contrast and analytical precision allow detection of the dissolution of a minute amount of silicate. Because the precipitation of Si-containing secondary phases or the reverse reaction consume silica but leaves the <sup>29</sup>Si /<sup>28</sup>Si ratios essentially unchanged in experimental solutions, dissolution rates were still measurable when secondary phase precipitation took place in experiments. This old story of “isotope ratio versus concentration” allowed our measurements of the simultaneous dissolution and precipitation rates at equilibrium, independently, and permitted testing of the applicability of the Principle of Detailed Balance. Many challenges remain in the discipline of geochemical kinetics. Innovation of the non-traditional stable isotope shows great promise for meeting some of the long-standing challenges.

<sup>1</sup>Gaillardet (2008). In Kinetics of water-rock interaction, 591-653

<sup>2</sup>Gruber et al. (2013). *Geochimica et Cosmochimica Acta*, 104, 261-280

<sup>3</sup>Liu et al. (2016). *Geochemical Perspectives Letters* 2, 78-86.

<sup>4</sup>Zhu et al. (2016). *Chemical Geology* 445, 146-163.