

# Equilibrium Si isotope fractionation during adsorption of monosilicic acid on Fe(III)-oxyhydroxide surface

YUN LIU\* AND HONG-TAO HE

State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, China.

\*liuyun@vip.gyig.ac.cn

Adsorption process is ubiquitous in Earth's surface environments and has an important effect on the change of Si isotope compositions of pore water, river and sea water. In this study, we focus on monosilicic acid ( $\text{H}_4\text{SiO}_4$ ), which is the most important aqueous Si-bearing species, adsorbed on Fe(III)-oxyhydroxide and its consequence on Si isotope fractionations. Based on quantum chemistry calculation, we find a large equilibrium Si isotope fractionation (i.e., -3.0‰) between a stable adsorbed bidentate Si surface complex ( ${}^2\text{C}>\text{Fe}_2\text{O}_2\text{Si}(\text{OH})_2$ ) and aqueous  $\text{H}_4\text{SiO}_4$  solution, which considerably differs from previously experimental results (ca. -1.08‰). To explain this large discrepancy, we investigate a transient monodentate Si surface complex ( ${}^1\text{V}>\text{Fe}_2\text{OSi}(\text{OH})_3$ ) and find that it has much smaller Si isotope fractionation. A kinetic isotope fractionation effect for the formation of  ${}^1\text{V}>\text{Fe}_2\text{OSi}(\text{OH})_3$  surface complexes is also calculated.