

## **Thermodynamic and molecular study of vanadium adsorption onto clay mineral surfaces**

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The enrichment of V in sediment profiles and the isotopic fractionation of V have been utilized in recent years to trace redox variations in paleo-oceans. However, the adsorption behavior of pentavalent V, V(V), to the surfaces of clay minerals, the major component of suspended sediments in rivers and deep ocean sediments, remains unclear. To fill this gap, the adsorption behavior of V(V) to three clay minerals (kaolinite, montmorillonite, and illite) were investigated in this study. A surface complexation model was developed to estimate the binding constants of V onto clay minerals based on pH adsorption edge experiments. Synchrotron extended X-ray adsorption fine structure (EXAFS) analyses were performed to constrain the local adsorption structure of V onto the three studied clay minerals. We found that for all three clay minerals, V adsorption reaches a maximum at pH 4 and then decreases with increasing pH. Permanently charged sites on the clay mineral surfaces primarily adsorb V at low pH, while amphoteric sites are responsible for V adsorption in the circumneutral pH range. EXAFS analysis shows that V(V) binds to kaolinite mainly through the substitution of Al in the octahedral sheet, while V binds to montmorillonite and illite through both Al substitution and adsorption onto Si sites. Our experimental results provide fundamental information for tracing the behavior of V on clay surfaces and thus are critical for the reconstruction of paleo-ocean redox conditions using V enrichment in shales.