A comprehensive predictive model for sulfate adsorption on oxide minerals

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This presentation provides a set of equations that enables the prediction of sulfate adsorption behavior on all oxide minerals over a wide range of pH, ionic strength, sulfate concentration, and solid/water ratio based on the extended triple-layer model (ETLM). Although surface complexation models including the ETLM have been frequently applied to describe sulfate-surface interactions on oxides, the proposed adsorption stoichiometries and equilibrium constants have been mutually inconsistent even in a specific sulfate–oxide system. Here, I show that the outer-layer capacitance (C2), a TLM parameter that has been traditionally set to 20 uF cm-2, has a significant impact on the model prediction of sulfate adsorption. With the capacitance value being the same as the inner-layer one (i.e., C2 = C1), which is a theoretically and spectroscopically reasonable assumption, our ETML calculation adequately represents all adsorption, surface titration, and proton co-adsorption data for various sulfate-oxide systems reported in the literature (oxides; goethite, ferrihydrite, r-alumina, gibbsite, and hematite) and those obtained in the present study (oxide; anatase). The combination of a monodentate-mononuclear inner-sphere and a bidentate-binuclear outer-sphere surface complex was found to explain all experimental data. Analysis of the retrieved equilibrium constants for the two surface species with the Born solvation theory led to the development of predictive equations for sulfate adsorption constants on all oxides. Considering the abundant and ubiquitous distribution of sulfate in natural environments, the proposed model should significantly contribute to a better understanding of the geochemical processes occurring at the mineral-water interface.

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