

Title: Thorium (IV) binding to organic and inorganic ligands: Marine colloidal organic matter, marine polysaccharides and hematite

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Abstract: The fate of metals in aquatic systems is governed by a combination of two factors: (1) element speciation, and (2) the processes that act to transport those species. In most aquatic systems, natural organic matter (NOM) constitutes an important pool of ligands for binding to metals. The effects of NOM on metal ion speciation and sorption to mineral surfaces are not yet fully understood. Factors which impact metal speciation are complex functions of ion and mineral chemistry, the environment in which the mineral and ions reside, and the source, molecular weight distribution, fraction and composition of the NOM. In ternary systems (e.g. metal ion, organic ligands, metal oxide), metal ion speciation and sorption behavior may be the consequence of the formation of solution phase species, solution phase binary complexes, binary and/or ternary surface complexes, the regulation of mineral surface charge by NOM sorption, as well as other processes.

In this research, a ternary system comprised of thorium (IV), two samples of marine colloidal organic matter (COM) and hematite were examined. Historically, the model used to describe thorium (IV) interactions with marine ligands was based on non-differentiated scavenging, with thorium distribution among particulate, colloidal and dissolved ligands a function of ligand concentration. However, more recent data show that Th(IV) binds more strongly with certain components of the ligand pool.

This project examined the binding of Th(IV) to COM and fractionated COM, as well as the role COM plays in the sorption of Th(IV) onto hematite. A more thorough understanding of the chemistry of this ternary system will provide insights into metal speciation, the impact natural organic matter has on the fate and transport of metals in the environment and the role thorium plays as a tracer for constituents of the carbon cycle.

The objectives of this research were: (1) to evaluate the impact of organic ligands on metal speciation, including metal sorption to mineral surfaces, as represented by thorium sorption to hematite and (2) to demonstrate that a highly reactive fraction of marine colloidal organic matter controls the complexation of thorium to this ligand pool.

Experimental results of component characterization, Th/COM binding, COM fractionation, batch binary sorption and batch ternary sorption experiments are discussed. The complexation of Th(IV) by whole and fractionated COM was interpreted by a graphical (Schubert's) method, a non-linear regression (modified Schubert's) method and a discrete ligand method. The binding of Th(IV) to COM was found to be strong, with the complexation abilities of the COM being associated with an operationally defined polysaccharide enriched COM fraction.

These experiments allowed for a characterization of the Th/COM/hematite system to be performed based on the results of the component and binary examinations. Concurrent to the experiments a series of chemical models was developed to assist in

understanding the complexities of the system. The results of these experiments indicated that marine COM enhances the binding of Th(IV) to hematite and that an accurate simulation of the ternary system requires that a ternary reaction be modeled, in addition to all of the component and binary system reactions.