Dynamics of contaminant sorption on mineral surfaces due to multiple redox cycling of natural iron-rich clay minerals

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Iron-rich clay minerals are rechargeable sources of electrons, which can determine the fate of nutrients and contaminants in natural and engineered systems. Due to their natural abundance, and structural and surface-dependent reactivities, these iron-rich clay minerals can sequester contaminants. Furthermore, climate and environmental changes can lead to frequent redox cycling of these minerals, plausibly altering the dynamics of the associated contaminants. Previous studies explored the reactivities of the natural iron-rich clay minerals in their native and reduced forms and their impact on contaminant cycling in soils and water. However, the mechanism of the coupled reactivity of the natural iron-rich clay minerals and associated contaminants, especially under multiple redox cycles, remains elusive.

In this study, we examined the reactivity and, thus, sorption capacity of a natural iron-rich clay mineral- nontronite (NAu-2) under multiple redox cycles and when exposed to geogenic contaminants such as arsenic (As) and cobalt (Co). We exposed NAu-2 to two repeated reduction and oxidation cycles, resulting in four samples of NAu-2, i.e., natural, reduced, oxidized, and rereduced. CBD (Citrate-Bicarbonate-Dithionite) treatment was employed under anoxic conditions for reduction and H_2O_2 under oxic conditions for oxidation steps. As(III) and Co(II) at different concentrations were applied to four NAu-2 samples (0.1 to 1 g/L) at pH 7.0±0.02 under anoxic conditions. We used ICP-OES to quantify the dissolved concentrations of iron, As, and Co. Solid mineral samples with and without exposure to contaminants were assessed with XRD, TEM, and XAS analysis.

We will present a quantitative study amongst four NAu-2 samples and their impact on As(III) and Co(II) sorption at nearneutral pH conditions. We will further discuss the reaction mechanisms with adsorption isotherms. By probing the reactivity of natural iron-rich clay minerals coupled with geogenic contaminants under multiple redox cycles, we will better understand the surface and structural processes occurring at the mineral surfaces and concurrently infer the fate of the contaminants in changing environments.