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The importance of dissolved sulfhydryl binding sites on the speciation of aqueous metals in lake waters: Results from pristine and human-impacted lakes

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Sulfhydryl binding sites (R-SH) are ubiquitous on dissolved organic molecules in natural aqueous environments, within complex solid organic frameworks, and on microbial cell walls. Although their concentrations are typically lower than that of other ligands, sulfhydryl binding sites likely play an outsized role in metal binding due to their high affinity to bind chalcophile elements. Few studies have measured sulfhydryl concentrations in natural environmental samples due to the difficulty, until recently, of measuring total sulfhydryl concentrations on solids or in aqueous samples, and none have quantified the impact that these binding sites have on metal speciation. In this study, recent advances in measuring dissolved total sulfhydryl site concentrations were applied to a set of water samples from pristine and human-impacted lakes to determine the role of dissolved sulfhydryl binding sites in metal speciation in each lake. In addition to reporting sulfhydryl site concentrations in each sample, we also measured the pH, dissolved organic carbon concentration, the total dissolved concentrations of a wide range of elements and anions, and the total dissolved binding site concentrations. Chemical analyses were used in conjunction with Visual MINTEQ to model the speciation of the dissolved elements in each lake sample. Measurable sulfhydryl concentrations were recorded in all lake samples, with significantly greater sulfhydryl concentrations in the pristine compared to the human-impacted lakes. Speciation modeling indicates that aqueous element-sulfhydryl complexes represent a significant proportion of the total dissolved element budget for a range of elements in both the natural and human-impacted lakes, with greater proportions observed in the pristine lake samples. For example, modeling results indicate that the pristine lake waters have an average of 47% of the total dissolved Zn present in solution as an aqueous Zn-sulfhydryl complex compared to a lower average of 29% for human-impacted lakes. Overall, the results of this study demonstrate that it is imperative to measure at least total aqueous sulfhydryl site concentrations in environmental aqueous samples in order to accurately model dissolved element speciation in those waters and thereby to assess and model contaminant mobility and bioavailability.