

Distribution and activity of iron-oxidizing microorganisms in acidic geothermal environments

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Iron oxide mats of acidic geothermal springs in Norris Geyser Basin, Yellowstone National Park are inhabited by numerous deeply-rooted members of the domain *Archaea*. Microorganisms of the order Sulfolobales (e.g., *Metallosphaera yellowstonensis* and other novel phylotypes) have previously been shown to be important community members and are likely responsible for the oxidation of Fe(II) and subsequent formation of Fe(III)-oxide mats. Iron-oxidizing microorganisms are hypothesized to thrive in more oxic regions of Fe(III)-oxide mats and are likely most abundant at the water/mat interface where oxygen concentrations are high relative to deeper mat positions. The current study investigated the relationship among vertical distribution of specific microorganisms, mRNA transcript levels of genes involved in Fe(II)-oxidation, and corresponding oxygen gradients measured *in situ*.

Quantitative PCR primers designed around specific phylotypes were used to determine the vertical distribution of organisms within the Fe(III)-oxide mats. Similarly, mRNA transcripts of genes, thought to be important in Fe(II)-oxidation (i.e., Fe-specific heme copper oxidases), were amplified with primers designed for various Fe(II)-oxidizing microbial populations.

Results suggest that the spatial distribution of aerobic Fe(II)-oxidizing microbial populations correspond with measured oxygen gradients. Specifically, Fe(II)-oxidizing microorganisms become less abundant at depths below ~600 μm corresponding to depths where dissolved oxygen concentrations approach detection. Transcripts of genes (mRNA) previously shown to be involved in Fe(II)-oxidation are well represented in the top 600 μm -layer. This study highlights the important linkage among hydrodynamics, oxygen in-gassing and diffusion, and the spatial location of active Fe(II)-oxidizing microorganisms (and associated transcription of specific genes) in the biomineralization of Fe(III)-oxide phases in acidic geothermal environments.

Residence time analysis of metal-desorption and mineral-dissolution kinetics using a Damkohler approach

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Understanding processes that control coupled reaction and transport in contaminated and natural systems is important for predicting the behavior of metals. Mineral dissolution and metal desorption are two important processes that govern metal behavior. Reactive transport studies often assume that desorption occurs instantaneously and therefore apply thermodynamic equilibrium models, or that mineral dissolution is so slow that it is not relevant. This study investigates groundwater time scales where kinetic metal desorption and/or mineral dissolution are important mechanisms for accurate modeling of metal fate and transport. Compiled desorption reaction times for metal and metalloid rate constants show large variations, spanning over six orders of magnitude. Mineral dissolution rate constants span over thirteen orders of magnitude and overlap with the range of desorption rate constants.

The Damkohler number was used to calculate residence times where kinetic formulations for dissolution and desorption may be more accurate representations of metals behavior. According to this method, metal desorption kinetics are influential at residence times from a few days to ~2 years. For longer residence times, metal desorption should behave according to equilibrium desorption models. In contrast, kinetic mineral dissolution should be considered over nearly all residence times and length scales relevant to groundwater modeling. Groundwater models with residence times of minutes up to ~600 days should potentially consider both metal desorption and mineral dissolution rates, with greater residence times exhibiting equilibrium behavior and shorter residences resulting in negligible change in metal concentrations. Ultimately, this Damkohler analysis provides constraints on relevant metal release mechanisms for models of metal fate and transport, such as those used in the fields of carbon sequestration and acid mine drainage. Additionally, the results can be used in experimental design to determine the setup required to measure the desired variable, i.e. kinetic or equilibrium characteristics.