

Nitrogen reduction on metal sulfide surfaces under hydrothermal conditions

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A hypothesis in this research is that there may of been a transition period on early Earth where inorganic iron-sulfur clusters were adapted for use by the biological world. It has been proposed by others that prebiotic chemistry originated on the surface iron sulfides within the environment of hydrothermal vents (i.e. the 'iron-sulfur world') on the ocean floor [1]. Currently our studies are investigating prebiotic relevant chemistry on iron sulfide minerals under hydrothermal conditions.

Under current study is the conversion of dinitrogen to ammonia, presumably a key reaction that would precede the assembly of biologically relevant molecules. Prior studies have shown that dinitrogen can be converted to ammonia in the presence of iron sulfide under hydrothermal conditions [2]. Current literature lacks studies that have investigated the surface chemistry involved in this chemical transformation. In this work we present results from laboratory based experiments using *in situ* attenuated total reflection Fourier transform infrared spectroscopy (ATR-FTIR) to investigate the fixation of dinitrogen under hydrothermal conditions in the presence of iron sulfide and hydrogen sulfide at pressures up to 100 bar and temperatures up to 150°C. Using ATR-FTIR we will present data that helps to determine the the composition of the iron sulfide surface during the conversion of the dinitrogen to ammonia under hydrothermal reaction conditions. Results for the conversion of nitrite and nitrate to ammonia on iron sulfide surfaces will also be presented that help to elucidate key redox active reaction sites on the sulfide surfaces. Experimental results will be focused on FeS and FeS₂ mineral phases.

[1] Wachtershauser, G. (1988) *Systematic & Applied Microbiology* **10**, 207–210. [2] Schoonen, M.A.A. & Xu, Y. (2001) *Astrobiology* **1**, 133–142.

Interactions between climate change, land use and the biological silica buffer in wetlands and forests

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We have studied the storage and recycling of amorphous biogenic silica in boreal wetlands and temperate forests and croplands. From both studies it is clear that climate, hydrology and land use changes can impact strongly on the biological silica buffer in these ecosystems.

In boreal wetland ecosystems, factors such as hydrosere, permafrost, climate and human interference may disturb the prevailing mire vegetation, whereby a new dominant assemblage can develop. We observed that diatoms thrive during periods associated with major vegetation transitions, creating isolated and shallow peat layers with significantly elevated amorphous Si content. We also observed high amorphous Si storage associated with sedge vegetations. Modeling indicates that the biological Si buffer impacts strongly on watershed Si fluxes.

In the temperate Scheldt river basin, we observed that biological, reactive Si storage in forests strongly exceeds storage in cropland ecosystems. In cropland dominated watersheds, strong erosion is associated with significant transport of amorphous Si in rivers during precipitation peaks. At base-flow, we observe significantly lower dissolved Si fluxes from old agricultural watersheds compared to forested watersheds. These results have inspired the development of a new conceptual model for Si fluxes after deforestation: initially, fast recycling and erosion of amorphous Si will increase Si fluxes from deforested watersheds. However, on long-term, loss of biological control on Si fluxes may lead to lower Si fluxes from agricultural watersheds.