"Green Rust" in the Lab and in the Soil

In their report "Abiotic selenium redox transformations in the presence of Fe(II,III) oxides," S. C. B. Myneni *et al*. state (*1*, p. 1109) that the reduction of selenium (Se)(VI) by iron (Fe)(II,III) oxides ("green rust") in an artificially constituted system "provides direct evidence for the formation of reduced Se species in anoxic sediments." By extrapolating their experiments, done under highly specialized laboratory conditions, to encompass suboxic sediments in general, Myneni *et al*. appear to challenge a body of evidence which demonstrates that the bulk of Se(VI) reduction in such natural systems is directly mediated by bacteria (for example, through dissimilatory selenate reduction).

The evidence presented by Myneni *et al.* to support their contention for abiotic reduction in sediments was a selective comparison of their rate constants to some of those in the literature. However, agreement of rate constant data provides only circumstantial evidence and does not constitute a rigorous proof for the underlying mechanism. We concede that some amount of abiotic reduction of Se(VI) can theoretically occur in natural systems. However, Myneni *et al*. have yet to show conclusively that abiotic Se(VI) reduction actually occurs in recent sediments. Furthermore, they also would have to demonstrate that its quantitative significance is of a magnitude comparable to that achieved by the direct bacterial reduction of this element in such environments.

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Response: We thank Oremland *et al.* for reiterating the importance of microbial reactions involving Se(VI) in soils and sediments, which several researchers have shown for many species of microorganisms. Our report neither contradicts these previous observations nor suggests that bacterial processes are not important for Se(VI) reduction in nature. Instead, we showed that Se(VI) can be reduced abiotically under various laboratory conditions by green rust, an Fe(II,III) oxide that can exist in some types of anoxic sediments.

By oxidizing organic matter and depleting oxygen, microorganisms create anoxic conditions in soils and sediments, which may further lead to the formation of several reductants, such as NH_4^+, various organic acids, and Fe-oxyhydroxides. Thus the redox transformations of major and trace elements (including Se) and organic compounds in sediments may be facilitated directly by the microorganisms (for example, by dissimilatory selenate reduction) and by the inorganic and organic reductants formed in the sediments as a result of microbial processes. Our report discusses Se(VI) reactions with green rust, a potential reductant formed as a result of biogeochemical processes in sediments. While the spectroscopic and microscopic data in our report provide direct evidence for such reactions, the thermodynamic and kinetic information that we discussed supports the possibility that green rust plays a role in Se(VI) reduction in these samples.

Procedures that have been used to distinguish biotic and abiotic reactions—for example, autoclaving (*2*) or the addition of chemicals—not only change the activity of microorganisms, but also affect the rate at which oxygen is consumed and organic matter decays in soils and sediments. This further affects sediment redox conditions (*1*) and modifies the concentration of natural reductants. Thus, such procedures provide only indirect evidence for microbial reduction of Se(VI) and do not rule out other mechanisms.

In summary, Se(VI) reduction in a sediment can occur through different pathways, and the magnitude of Se(VI) reduction through any of these pathways is determined by Se(VI) reduction rates, the reductant's concentration, and the activity of microorganisms. In addition, the rate at which anoxic conditions are imposed in the sediment is also critical. All these factors have to be evaluated when a contaminant transformation in sediments is evaluated.

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Editor's Summary