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Successive reduction of iodine by *Shewanella oneidensis*-mediated Fe(II)/Fe(III) redox cycling

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Iodine is a trace element that plays a crucial role in human health. High-iodine groundwater poses a severe negative impact on the health of millions of people worldwide. Iodate (IO_3^-) and iodide (I^-) are the two forms of inorganic iodine found in environment where I^- is more mobile than IO_3^-. Microbially mediated reduction of IO_3^- to I^- is critical for the formation and enrichment of I^- in groundwater. Ferrous iron (Fe(II)) produced by microbial Fe(III) reduction under anoxic condition and reactive oxygen species (ROS) generated from Fe(II) oxidation under oxic condition can mediate IO_3^- reduction and iodide I^- oxidation, respectively. However, how Fe(II)/Fe(III) cycling under redox fluctuating conditions impacts IO_3^-/I^- redox transformation remain unclear. In this study, *Shewanella oneidensis* MR-1 wildtype (WT) and its mutant ▵*dmsEFAB*, which lost the ability to enzymatically reduce IO_3^-, were chosen to conduct ferrihydrite/goethite/nontronite culture experiments under consecutive cycles of anoxic reduction of Fe(III) and re-oxidation of Fe(II) by O_2. The results showed that both surface-adsorbed and mineral structural Fe(II) chemically reduced IO_3^-. Chemical IO_3^- reduction by biogenic Fe(II) was slower than enzymatic IO_3^- reduction by WT. Compared to ▵*dmsEFAB* cultures, WT cultures all showed higher Fe(II) concentrations under anoxic conditions but lower cumulative •OH under oxic conditions, which imply the chemical reaction between I^- and ROS. I^- oxidation by ROS, however, did not lead to a significant production of IO_3^- compared with I^- formed under anoxic conditions. Consequently, Fe(II)/Fe(III) redox cycling successively reduced IO_3^- to I^-. Thus, Fe(III)-reducing bacteria are critically important drivers for I^- enrichment and mobility in high-iodine groundwater worldwide.