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Biotransformation involved in sustained reductive removal of uranium in contaminant aquifers

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RESULTS TO DATE: This report summarizes progress made from June 2003 to July 2004. During this period research focused on further understanding the factors controlling the growth and activity of dissimilatory metal reducers in subsurface environments and the application of these findings to better design of strategies for in situ bioremediation of uranium.

Field studies focused on a new strategy for better sustaining microbial uranium reduction. In the previous field study(1), which was published during this reporting period, once Fe(III) was depleted near the injection gallery sulfate reducers were able to outcompete the Fe(III) reducers for the injected acetate and consumed the acetate at the point of injection, leaving no acetate for the Fe(III) reducers further downgradient. This was problematic because the sulfate reducers were ineffective in U(VI) reduction. Thus, as Fe(III) was depleted near the injection gallery U(VI) concentrations in the groundwater began to rise. To circumvent this in our next experiment, the acetate concentration injected into the aquifer was increased. This permitted consumption of acetate by the sulfate reducers at the point of injection while leaving acetate to be transported further downgradient to promote the activity of the dissimilatory metal reducers. The results suggested that this approach was successful, but there are issues with preferential flow paths of the injectate which need to be resolved for large-scale application of this approach. Two manuscripts describing the geochemistry and microbiology of this field trial are in preparation.

An unexpected result of our field experiment is that there has been no remobilization of the reduced uranium once the acetate injection was stopped. We continue to monitor this.

Analysis of the solid-phase uranium during the field experiment demonstrated that although U(IV) was being precipitated as the result of U(VI) reduction, there were also significant quantifies of U(VI) associated with the solid phase of the sediments (5). Laboratory studies were conducted to further evaluate the persistence of this U(VI). Studies in which sediments were incubated under strict anaerobic conditions demonstrated that the solid-phase U(VI) was resistant to microbial reduction. U(VI) adsorption onto iron oxides and hydroxides and clays in sediments is well known. However, previous studies of in situ uranium bioremediation have primarily focused on the reduction of dissolved U(VI) to insoluble U(IV) because immobilization of contaminant uranium is of paramount importance. Our results suggest that although dissimilatory metal-reducing microorganisms can effectively reduce soluble U(VI), U(VI) associated with the solid phase is not microbially reducible. The inability of microorganisms to reduce U(VI) adsorbed to sediments is not a limitation during the acetate injection phase of in situ bioremediation because this U(VI) is already immobile. However, it does have an impact on strategies for eventually extracting the immobilized uranium. For example, we have previously been proposed that once uranium has been immobilized in a discreet zone via U(VI) reduction, it could be resolubilized and extracted by reoxidizing precipitated U(IV) to soluble U(VI). The finding that a high proportion of the immobilized uranium is likely to be in the form of U(VI) suggests that oxidation of U(IV) should be supplemented with procedures for extracting U(VI), such as the use of bicarbonate, a common extraction technique for in situ uranium mining. In a similar manner, injection of bicarbonate, upgradient of a zone of acetate injection could solubilize U(VI) from sediments followed by reductive precipitation within the acetate injection zone to remove uranium which otherwise may later serve as a source of dissolved U(VI) in the groundwater via desorption. This potential strategy is currently under investigation.

Supplemental funding was supplied to this grant to determine if electrodes might serve as an electron donor to promote microbial U(VI) reduction. It was determined that electrodes could promote U(VI) reduction in contaminated aquifer sediments and that this process depended upon the presence of

microorganisms. This may represent an alternative to the addition of acetate to promote in situ bioremediation of uranium. A poster on these results was presented at the ASM meeting and a manuscript describing these results is in preparation.

Several other studies were completed and the results published. These included a study demonstrating the role of structural Fe(III) in clays in serving as an electron acceptor for metal reducers in subsurface environments (6) and a study providing field and laboratory evidence that stimulating the activity of Geobacter species in the subsurface can be a good strategy for in situ bioremediation of vanadium because Geobacter species can use vanadium as an electron acceptor (4). Also a study evaluating the potential for promoting in situ uranium bioremediation in the low-pH zones of the NABIR FRC was published (7). Two invited reviews that incorporated results from our studies under this grant were also published (2, 3).

1. Anderson, R. T., H. A. Vrionis, I. Ortiz-Bernad, C. T. Resch, A. Peacock, R. Dayvault, S. Marutzky, D. R. Metzler, K. Karp, M. Lowe, D. C. White, P. E. Long, and D. R. Lovley. 2003. Stimulating the in situ activity of Geobacter species to remove uranium from the groundwater of a uranium-contaminated aquifer. Appl. Environ. Microbiol. 69:5884-5891. 2. Lovley, D. R. 2003. Cleaning up with genomics: applying molecular biology to bioremediation. Nature Microbiol. Rev. 1:35-44. 3. Lovley, D. R., D. E. Holmes, and K. P. Nevin. 2004. Dissimilatory Fe(III) and Mn(IV) reduction. Adv. Microb. Physiol.:(in press). 4. Ortiz-Bernad, I., R. T. Anderson, H. A. Vironis, and D. R. Lovley. 2004. Vanadium respiration by Geobacter metalllireducens: a novel strategy for the in situ removal of vanadium from groundwater. Appl. Environ. Microbiol. 70:3091-3095. 5. Ortiz-Bernad, I., R. T. Anderson, H. A. Vrionis, and D. R. Lovley. 2004. Resitance of solid-phase U(VI) to microbial reduction during in situ bioremediation of uranium-contaminated groundwater. Appl. Environ. Microbiol. 70:(in press). 6. Shelobolina, E. S., R. T. Anderson, Y. N. Vodyanitskii, R. Yuretich, and D. R. Lovley. 2003. Importance of clay size minerals for Fe(III) respiration in a petroleum-contaminated aquifer. Geobiology 2:67-76. 7. Shelobolina, E. S., K. R. O'Neil, K. T. Finneran, L. A. Hayes, and D. R. Lovley. 2003. Potential for in situ bioremediation of a low-pH, high-nitrate uranium-contaminated groundwater. Soil and Sediment Contamination 12:865-884.

DELIVERABLES: 1. Anderson, R. T., H. A. Vrionis, I. Ortiz-Bernad, C. T. Resch, A. Peacock, R. Dayvault, S. Marutzky, D. R. Metzler, K. Karp, M. Lowe, D. C. White, P. E. Long, and D. R. Lovley. 2003. Stimulating the in situ activity of Geobacter species to remove uranium from the groundwater of a uranium-contaminated aquifer. Appl. Environ. Microbiol. 69:5884-5891. 2. Lovley, D. R. 2003. Cleaning up with genomics: applying molecular biology to bioremediation. Nature Microbiol. Rev. 1:35-44. 3. Lovley, D. R., D. E. Holmes, and K. P. Nevin. 2004. Dissimilatory Fe(III) and Mn(IV) reduction. Adv. Microb. Physiol.:(in press). 4. Ortiz-Bernad, I., R. T. Anderson, H. A. Vironis, and D. R. Lovley. 2004. Vanadium respiration by Geobacter metalllireducens: a novel strategy for the in situ removal of vanadium from groundwater. Appl. Environ. Microbiol. 70:3091-3095. 5. Ortiz-Bernad, I., R. T. Anderson, H. A. Vrionis, and D. R. Lovley. 2004. Resitance of solid-phase U(VI) to microbial reduction during in situ bioremediation of uranium-contaminated groundwater. Appl. Environ. Microbiol. 70:(in press). 6. Shelobolina, E. S., R. T. Anderson, Y. N. Vodyanitskii, R. Yuretich, and D. R. Lovley. 2003. Importance of clay size minerals for Fe(III) respiration in a petroleum-contaminated aquifer. Geobiology 2:67-76. 7. Shelobolina, E. S., K. R. O'Neil, K. T. Finneran, L. A. Hayes, and D. R. Lovley. 2003. Potential for in situ bioremediation of a low-pH, high-nitrate uranium-contaminated groundwater. Soil and Sediment Contamination 12:865-884.