

Hydrogen thresholds and steady-state concentrations associated with microbial arsenate respiration

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Abstract: H_2 thresholds for microbial respiration of arsenate (As(V)) were investigated in a pure culture of *Sulfurospirillum arsenophilum*. H_2 was consumed to threshold concentrations of 0.03-0.09 nmol/L with As(V) as terminal electron acceptor, allowing for a Gibbs free-energy yield of 36-41 kJ per mol of reaction. These thresholds are among the lowest measured for anaerobic respirers and fall into the range of denitrifiers or Fe(III)-reducers. In sediments from an arsenic-contaminated aquifer in the Red River flood plain, Vietnam, H_2 levels decreased to 0.4-2 nmol/L when As(V) was added under anoxic conditions. When As(V) was depleted, H_2 concentrations rebounded by a factor of 10, a level similar to that observed in arsenic-free controls. The sediment-associated microbial population completely reduced millimolar levels of As(V) to arsenite (As(III)) within a few days. The rate of As(V)-reduction was essentially the same in sediments amended with a pure culture of *S. arsenophilum*. These findings together with a review of observed H_2 threshold and steady-state values suggest that microbial As(V)-respirers have a competitive advantage over several other anaerobic respirers through their ability to thrive at low H_2 levels. ?? 2007 American Chemical Society.

Index Keywords: Arsenic compounds; Concentration (process); Electrons; Hydrogen; Microorganisms; Sediments; Anaerobic respirers; Electron acceptor; Pollution control; arsenic acid; hydrogen; Arsenic compounds; Concentration (process); Electrons; Hydrogen; Microorganisms; Pollution control; Sediments; anoxic conditions; aquifer pollution; arsenate; floodplain; Gibbs free energy; hydrogen; microbial community; respiration; river; steady-state equilibrium; threshold; anoxia; aquifer; article; concentration (parameters); microbial respiration; nonhuman; reduction kinetics; *Spirillum*; steady state; *Sulfurospirillum arsenophilum*; Arsenates; Biodegradation, Environmental; Chromatography, Gas; Environmental Pollutants; Epsilonproteobacteria; Geologic Sediments; Hydrogen; Methane; Thermodynamics; Vietnam; Asia; Eurasia; Red River [Asia]; Southeast Asia; Viet Nam; *Sulfurospirillum arsenophilum*

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References:

1. BGS, D.P.H.E., (2001) Arsenic Contamination of Groundwater in Bangladesh, , Kinniburgh, D. G, Smedley, P. L, Eds, British Geological Survey: Nottingham
2. Smedley, P.L., Kinniburgh, D.G., A review of the source, behaviour and distribution of arsenic in natural waters (2002) Appl. Geochem, 17, pp. 517-568
3. Ng, J.C., Wang, J., Shraim, A., A global health problem caused by arsenic from natural sources (2003) Chemosphere, 52, pp. 1353-1359
4. WHO Guidelines for Drinking-Water Quality. 1: Recommendations, 3rd ed.
5. WHO: Geneva, 1993Smith, A.H., Lingas, E.O., Rahman, M., Contamination of drinking-water by arsenic in Bangladesh: A

public health emergency (2000) Bull. W. H. O, 78, pp. 1093-1103

6. Ahmann, D., Roberts, A.L., Krumholz, L.R., Morel, F.M.M., Microbe grows by reducing arsenic (1994) Nature, 371, p. 750
7. Laverman, A.M., Switzer Blum, J., Schaefer, J.K., Phillips, E.J.P., Lovley, D.R., Oremland, R.S., Growth of strain SES-3 with arsenate and other diverse electron acceptors (1995) Appl. Environ. Microbiol, 61, pp. 3556-3561
8. Dowdle, P.R., Laverman, A.M., Oremland, R.S., Bacterial dissimilatory reduction of arsenic(V) to arsenic(III) in anoxic sediments (1996) Appl. Environ. Microbiol, 62, pp. 1664-1669
9. Newman, D.K., Kennedy, E.K., Coates, J.D., Ahmann, D., Ellis, D.J., Lovley, D.R., Morel, F.M.M., Dissimilatory arsenate and sulfate reduction in *Desulfotomaculum auripigmentum* sp. nov (1997) Arch. Microbiol, 168, pp. 380-388
10. Oremland, R.S., Dowdle, P.R., Hoefft, S., Sharp, J.O., Schaefer, J.K., Miller, L.G., Blum, J.S., Wallschlaeger, D., Bacterial dissimilatory reduction of arsenate and sulfate in meromictic Mono Lake, California (2000) Geochim. Cosmochim. Acta, 64, pp. 3073-3084
11. Oremland, R.S., Stolz, J.F., The ecology of arsenic (2003) Science, 300, pp. 939-944
12. Brannon, J.M., Patrick Jr., W.H., Fixation, transformation, and mobilization of arsenic in sediments (1987) Environ. Sci. Technol, 21, pp. 450-459
13. Masscheleyn, P.H., Delaune, R.D., Patrick Jr., W.H., Effect of redox potential and pH on arsenic speciation and solubility in a contaminated soil (1991) Environ. Sci. Technol, 25, pp. 1414-1419
14. Ahmann, D., Krumholz, L.R., Hemond, H.F., Lovley, D.R., Morel, F.M.M., Microbial mobilization of arsenic from sediments of the Aberjona watershed (1997) Environ. Sci. Technol, 31, pp. 2923-2930
15. Newman, D.K., Ahmann, D., Morel, F.M.M., A brief review of microbial arsenate respiration (1998) Geomicrobiol. J, 15, pp. 255-268
16. Oremland, R.S., Stolz, J.F., Arsenic, microbes and contaminated aquifers (2005) Trends Microbiol, 13, pp. 45-49
17. Oremland, R.S., Stolz, J.F., Hollibaugh, J.T., The microbial arsenic cycle in Mono Lake, California (2004) FEMS Microbiol. Ecol, 48, pp. 15-27
18. Oremland, R.S., Kulp, T.R., Switzer Blum, J., Hoefft, S.E., Baesman, S., Miller, L.G., Stolz, J.F., A microbial arsenic cycle in a salt-saturated extreme environment (2005) Science, 308, pp. 1305-1308
19. Appelo, C.A.J., Postma, D., (2005) Geochemistry, Groundwater and Pollution, , 2nd ed, A. A. Balkema Publishers: Leiden, The Netherlands
20. Christensen, T.H., Bjerg, P.L., Banwart, S.A., Jakobsen, R., Heron, G., Albrechtsen, H.-J., Characterization of redox conditions in groundwater contaminant plumes (2000) J. Contam. Hydrol, 45, pp. 165-241
21. Chapelle, F.H., Haack, S.K., Adriaens, P., Henry, M.A., Bradley, P.M., Comparison of Eh and H₂ measurements for delineating redox processes in a contaminated aquifer (1996) Environ. Sci. Technol, 30, pp. 3565-3569
22. Cord-Ruwisch, R., Seitz, H.-J., Conrad, R., The capacity of hydrogenotrophic anaerobic bacteria to compete for traces of hydrogen depends on the redox potential of the terminal electron acceptor (1988) Arch. Microbiol, 149, pp. 350-357
23. Lovley, D.R., Goodwin, S., Hydrogen concentrations as an indicator of the predominant terminal electron-accepting reactions in aquatic sediments (1988) Geochim. Cosmochim. Acta, 52, pp. 2993-3003
24. Conrad, R., Compensation concentration as critical variable for regulating the flux of trace gases between soil and atmosphere (1994) Biogeochemistry, 27, pp. 155-170
25. Conrad, R. Soil microorganisms as controllers of atmospheric trace gases (H₂, CO, CH₄, OCS, N₂O, and NO). Microbiol. Rev. 1996, 60, 609-640 Postma, D., Jakobsen, R., Redox zonation: Equilibrium constraints on the Fe(III)/SO₄-reduction interface (1996) Geochim. Cosmochim. Acta, 60, pp. 3169-3175
26. Jakobsen, R., Albrechtsen, H.-J., Rasmussen, M., Bay, H., Bjerg, P.L., Christensen, T.H., H₂ concentrations in a landfill

- leachate plume (Grindsted, Denmark): In situ energetics of terminal electron acceptor processes (1998) *Environ. Sci. Technol.*, 32, pp. 2142-2148
27. Hoehler, T.M., Alperin, M.J., Albert, D.B., Martens, C.S., Thermodynamic control on hydrogen concentrations in anoxic sediments (1998) *Geochim. Cosmochim. Acta*, 62, pp. 1745-1756
28. Yang, Y., McCarty, P.L., Competition for hydrogen within a chlorinated solvent dehalogenating anaerobic mixed culture (1998) *Environ. Sci. Technol.*, 32, pp. 3591-3597
29. Heimann, A.C., Jakobsen, R., Experimental evidence for a lack of thermodynamic control on hydrogen concentrations during anaerobic degradation of chlorinated ethenes (2006) *Environ. Sci. Technol.*, 40, pp. 3501-3507
30. Marsh, T.L., McInerney, M.J., Relationship of hydrogen bioavailability to chromate reduction in aquifer sediments (2001) *Appl. Environ. Microbiol.*, 67, pp. 1517-1521
31. Stolz, J.F., Ellis, D.E., Switzer Blum, J., Ahmann, D., Lovley, D.R., Oremland, R.S., *Sulfurospirillum barnesii* sp. nov. and *Sulfurospirillum arsenophilum* sp. nov., new members of the *Sulfurospirillum* clade of the ? Proteobacteria (1999) *Int. J. Syst. Bacteriol.*, 49, pp. 1177-1180
32. Santini, J.M., Stolz, J.F., Macy, J.M., Isolation of a new arsenate-respiring bacterium - physiological and phylogenetic studies (2002) *Geomicrobiol. J.*, 19, pp. 41-52
33. Herbel, M.J., Blum, J.S., Hoeft, S.E., Cohen, S.M., Arnold, L.L., Lisak, J., Stolz, J.F., Oremland, R.S., Dissimilatory arsenate reductase activity and arsenate-respiring bacteria in bovine rumen fluid, hamster feces, and the termite hindgut (2002) *FEMS Microbiol. Ecol.*, 41, pp. 59-67
34. Berg, M., Tran, H.C., Nguyen, T.C., Pham, H.V., Schertenleib, R., Giger, W., Arsenic contamination of groundwater and drinking water in Vietnam: A human health threat (2001) *Environ. Sci. Technol.*, 35, pp. 2621-2626
35. Johnson, D.L., Pilson, M.E.Q., Spectrophotometric determination of arsenite, arsenate, and phosphate in natural waters (1972) *Anal. Chim. Acta*, 58, pp. 289-299
36. Anderson, C.R., Cook, G.M., Isolation and characterization of arsenate-reducing bacteria from arsenic-contaminated sites in New Zealand (2004) *Curr. Microbiol.*, 48, pp. 341-347
37. Heimann, A.C., Jakobsen, R., Filtration through nylon membranes negatively affects analysis of arsenic and phosphate by the molybdenum blue method (2006) *Talanta*, , doi:10.1016/j.talanta.2006.11.012
38. Parkhurst, D.L., Appelo, C.A.J., (1999) User's guide to PHREEQC (Version 2), , U.S. Geological Survey
39. Stookey, L.L., Ferrozine - a new spectrophotometric reagent for iron (1970) *Anal. Chem.*, 42, p. 779
40. Wilhelm, E., Battino, R., Wilcock, R.J., Low-pressure solubility of gases in liquid water (1977) *Chem. Rev.*, 77, pp. 219-262
41. Schink, B., Energetics of syntrophic cooperation in methanogenic degradation (1997) *Microbiol. Mol. Biol. R.*, 61, pp. 262-280
42. Hoehler, T.M., Alperin, M.J., Albert, D.B., Martens, C.S., Apparent minimum free energy requirements for methanogenic Archaea and sulfate-reducing bacteria in an anoxic marine sediment (2001) *FEMS Microbiol. Ecol.*, 38, pp. 33-41
43. Hoehler, T.M., Alperin, M.J., Albert, D.B., Martens, C.S., Acetogenesis from CO₂ in an anoxic marine sediment (1999) *Limnol. Oceanogr.*, 44, pp. 662-667
44. Lovley, D.R., Minimum threshold for hydrogen metabolism in methanogenic bacteria (1985) *Appl. Environ. Microbiol.*, 49, pp. 1530-1531
45. Luijten, M.L.G.C., Roelofsen, W., Langenhoff, A.A.M., Schraa, G., Stams, A.J.M., Hydrogen threshold concentrations in pure cultures of halo-respiring bacteria and at a site polluted with chlorinated ethenes (2004) *Environ. Microbiol.*, 6, pp. 646-650
46. Lovley, D.R., Phillips, E.J.P., Lonergan, D.J., Hydrogen and formate oxidation coupled to dissimilatory reduction of iron or

- manganese by *Alteromonas putrefaciens* (1989) *Appl. Environ. Microbiol.*, 55, pp. 700-706
47. Caccavo Jr., F., Blakemore, R.P., Lovley, D.R., A hydrogen-oxidizing, Fe(III)-reducing microorganism from the Great Bay estuary, New Hampshire (1992) *Appl. Environ. Microbiol.*, 58, pp. 3211-3216
 48. Sung, Y., Fletcher, K.E., Ritalahti, K.M., Apkarian, R.P., Ramos-Hernández, N., Sanford, R.A., Mesbah, N.M., Löffler, F.E., *Geobacter lovleyi* sp. nov. strain SZ, a novel metal-reducing and tetrachloroethene-dechlorinating bacterium (2006) *Appl. Environ. Microbiol.*, 72, pp. 2775-2782
 49. Chapelle, F.H., Lovley, D.R., Competitive exclusion of sulfate reduction by Fe(III)-reducing bacteria: A mechanism for producing discrete zones of high-iron ground water (1992) *Ground Water*, 30, pp. 29-36
 50. Blodau, C., Gatzek, C., Chemical controls on iron reduction in schwertmannite-rich sediments (2006) *Chem. Geol.*, 235, pp. 366-376
 51. Hering, V., Conrad, R., Kinetics of H₂ oxidation in respiring and denitrifying *Paracoccus denitrificans* (1991) *FEMS Microbiol. Lett.*, 78, pp. 259-264
 52. Löffler, F.E., Tiedje, J.M., Sanford, R.A., Fraction of electrons consumed in electron acceptor reduction and hydrogen thresholds as indicators of halorespiratory physiology (1999) *Appl. Environ. Microbiol.*, 65, pp. 4049-4056
 53. Smatlak, C.R., Gossett, J.M., Zinder, S.H., Comparative kinetics of hydrogen utilization for reductive dechlorination of tetrachloroethene and methanogenesis in an anaerobic enrichment culture (1996) *Environ. Sci. Technol.*, 30, pp. 2850-2858
 54. Kassenga, G., Pardue, J.H., Moe, W.M., Bowman, K.S., Hydrogen thresholds as indicators of dehalorespiration in constructed treatment wetlands (2004) *Environ. Sci. Technol.*, 38, pp. 1024-1030
 55. Heimann, A.C., Batstone, D.J., Jakobsen, R., *Methanosarcina* spp. drive vinyl chloride dechlorination via interspecies hydrogen transfer (2006) *Appl. Environ. Microbiol.*, 72, pp. 2942-2949
 56. Stumm, W., Morgan, J.J., (1996) *Aquatic Chemistry*, 3rd ed, Wiley: New York
 57. Krauskopf, K.B., Bird, D.K., (1994) *Introduction to Geochemistry*, 3rd ed, McGraw-Hill
 58. Thauer, R.K., Jungermann, K., Decker, K., Energy conservation in chemotrophic anaerobic bacteria (1977) *Bacteriol. Rev.*, 41, pp. 100-180
 59. Schnörrer, A., Schink, B., Svensson, B.H., *Clostridium ultunense* sp. nov., a mesophilic bacterium oxidizing acetate in syntrophic association with a hydrogenotrophic methanogenic bacterium (1996) *Int. J. Syst. Bacteriol.*, 46, pp. 1145-1152
 60. Cord-Ruwisch, R., Lovley, D.R., Schink, B., Growth of *Geobacter sulfurreducens* with acetate in syntrophic cooperation with hydrogen-oxidizing anaerobic partners (1998) *Appl. Environ. Microbiol.*, 64, pp. 2232-2236
 61. Seitz, H.-J., Schink, B., Pfennig, N., Conrad, R., Energetics of syntrophic ethanol oxidation in defined chemostat cocultures (1990) *Arch. Microbiol.*, 155, pp. 82-88
 62. Mukhopadhyay, R., Rosen, B.P., Phung, L.T., Silver, S., Microbial arsenic: From geocycles to genes and enzymes (2002) *FEMS Microbiol. Rev.*, 26, pp. 311-325
 63. Croal, L.R., Gralnick, J.A., Malasarn, D., Newman, D.K., The genetics of geochemistry (2004) *Annu. Rev. Genet.*, 38, pp. 175-202
 64. Conrad, R., Wetter, B., Influence of temperature on energetics of hydrogen metabolism in homoacetogenic, methanogenic, and other anaerobic bacteria (1990) *Arch. Microbiol.*, 155, pp. 94-98
 65. Sierra-Alvarez, R., Cortinas, I., Yenal, U., Field, J.A., Methanogenic inhibition by arsenic compounds (2004) *Appl. Environ. Microbiol.*, 70, pp. 5688-5691
 66. Islam, F.S., Gault, A.G., Boothman, C., Polya, D.A., Charnock, J.M., Chatterjee, D., Lloyd, J.R., Role of metal-reducing bacteria in arsenic release from Bengal delta sediments (2004) *Nature*, 430, pp. 68-71
 67. Van Geen, A., Rose, J., Thorat, S., Garnier, J.M., Zheng, Y., Bottero, J.Y., Decoupling of As and Fe release to Bangladesh

groundwater under reducing conditions. Part II: Evidence from sediment incubations (2004) *Geochim. Cosmochim. Acta*, 68, pp. 3475-3486

68. Gault, A.G., Islam, F.S., Polya, D.A., Charnock, J.M., Boothman, C., Chatterjee, D., Lloyd, J.R., Microcosm depth profiles of arsenic release in a shallow aquifer, West Bengal (2005) *Mineral. Mag.*, 69, pp. 855-863