

Arsenic in groundwater of the Red River floodplain, Vietnam: Controlling geochemical processes and reactive transport modeling

Postma D., Larsen F., Minh Hue N.T., Duc M.T., Viet P.H., Nhan P.Q., Jessen S.

Institute of Environment and Resources, Technical University of Denmark, Building 115, DK 2800 Kgs Lyngby, Denmark; Research Centre for Environmental Technology and Sustainable Development (CETASD), Hanoi University of Science, VNU, Viet Nam; Hanoi University of Mining and Geology, Viet Nam

Abstract: The mobilization of arsenic (As) to the groundwater was studied in a shallow Holocene aquifer on the Red River flood plain near Hanoi, Vietnam. The groundwater chemistry was investigated in a transect of 100 piezometers. Results show an anoxic aquifer featuring organic carbon decomposition with redox zonation dominated by the reduction of Fe-oxides and methanogenesis. Enhanced P_{CO_2} pressure causes carbonate dissolution to take place but mainly in the soil and unsaturated zone. The concentration of As increases over depth to a concentration of up to 550 $\mu\text{g/L}$. Most As is present as As(III) but some As(V) is always found. Arsenic correlates well with NH_4 , relating its release to organic matter decomposition and the source of As appears to be the Fe-oxides being reduced. Part of the produced Fe(II) is apparently reprecipitated as siderite containing less As. Results from sediment extraction indicate most As to be related to the Fe-oxide fractions. The measured amount of sorbed As is low. In agreement, speciation calculations for a Fe-oxide surface suggest As(III) to constitute only 3% of the surface sites while the remainder is occupied by carbonate and silica species. The evolution in water chemistry over depth is homogeneous and a reactive transport model was constructed to quantify the geochemical processes along the vertical groundwater flow component. A redox zonation model was constructed using the partial equilibrium approach with organic carbon degradation in the sediment as the only rate controlling parameter. Apart from the upper meter a constant degradation rate of 0.15 C mmol/L/yr could explain the redox zonation throughout the aquifer. Modeling also indicates that the Fe-oxide being reduced is of a stable type like goethite or hematite. Arsenic is contained in the Fe-oxides and is first released during their dissolution. Our model further suggests that part of the released As is adsorbed on the surface of the remaining Fe-oxides and in this way may be retarded. ?? 2007 Elsevier Ltd. All rights reserved.

Index Keywords: arsenic; concentration (composition); decomposition; dissolution; floodplain; flow modeling; groundwater; groundwater flow; methanogenesis; organic carbon; reduction; transport process; vadose zone; water chemistry; Asia; Eurasia; Hanoi; Southeast Asia; Viet Nam

Year: 2007

Source title: *Geochimica et Cosmochimica Acta*

Volume: 71

Issue: 21

Page : 5054-5071

Cited by: 45

Link: [Scorpus Link](#)

Correspondence Address: Postma, D.; Institute of Environment and Resources, Technical University of Denmark, Building 115, DK 2800 Kgs Lyngby, Denmark; email: djp@er.dtu.dk

ISSN: 167037

DOI: 10.1016/j.gca.2007.08.020

Language of Original Document: English

Abbreviated Source Title: *Geochimica et Cosmochimica Acta*

Document Type: Article

Source: Scopus

Authors with affiliations:

1. Postma, D., Institute of Environment and Resources, Technical University of Denmark, Building 115, DK 2800 Kgs Lyngby, Denmark
2. Larsen, F., Institute of Environment and Resources, Technical University of Denmark, Building 115, DK 2800 Kgs Lyngby, Denmark
3. Minh Hue, N.T., Research Centre for Environmental Technology and Sustainable Development (CETASD), Hanoi University of Science, VNU, Viet Nam
4. Duc, M.T., Research Centre for Environmental Technology and Sustainable Development (CETASD), Hanoi University of Science, VNU, Viet Nam
5. Viet, P.H., Research Centre for Environmental Technology and Sustainable Development (CETASD), Hanoi University of Science, VNU, Viet Nam
6. Nhan, P.Q., Hanoi University of Mining and Geology, Viet Nam
7. Jessen, S., Institute of Environment and Resources, Technical University of Denmark, Building 115, DK 2800 Kgs Lyngby, Denmark

References:

1. Akai, J., Izumi, K., Fukuhara, H., Masuda, H., Nakano, S., Yoshimura, T., Ohfuji, H., Akai, K., Mineralogical and geomicrobiological investigations on groundwater arsenic enrichment in Bangladesh (2004) *Appl. Geochem.*, 19, pp. 215-230
2. Anawar, H.M., Akai, J., Sakugawa, H., Mobilization of arsenic from subsurface sediments by effect of bicarbonate ions in groundwater (2004) *Chemosphere*, 54, pp. 753-762
3. Appelo, C.A.J., Postma, D., (2005) *Geochemistry Groundwater and Pollution*. second ed., , Balkema Publisher 649 pp
4. Appelo, C.A.J., Van Der Weiden, M.J.J., Tournassat, C., Charlet, L., Surface complexation of ferrous iron and carbonate on ferrihydrite and the mobilization of arsenic (2002) *Environ. Sci. Technol.*, 36, pp. 3096-3103
5. Aurillo, A.C., Mason, R.P., Hemond, H.F., Speciation and fate of arsenic in three lakes of the Aberjona watershed (1994) *Environ. Sci. Technol.*, 28, pp. 577-585
6. 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
7. Cline, J.D., Spectrophotometric determination of hydrogen sulfide in natural waters (1967) *Limnol. Oceanogr.*, 14, pp. 454-458
8. Cullen, W.R., Reimer, K.J., Arsenic speciation in the environment (1989) *Chem. Rev.*, 89, pp. 713-764
9. Dowling, C.B., Poreda, R.J., Basu, A.R., Peters, S.L., Aggarwal, P.K., Geochemical study of arsenic release mechanisms in

the Bengal Basin groundwater (2002) *Water Resour. Res.*, 38, p. 1173. , 10.1029/2001WR000968

10. Dzombak, D.A., Morel, F.M.M., (1990) *Surface Complexation Modeling-Hydrous Ferric Oxide*, Wiley
11. Gault, A.G., Polya, D.A., Charnock, J.M., Islam, F.S., Lloyd, J.R., Chatterjee, D., Preliminary EXAFS studies of solid phase speciation of As in a West Bengali sediment (2003) *Miner. Mag.*, 67, pp. 1183-1191
12. Goodbred Jr., S.L., Kuehl, S.A., The significance of large sediment supply, active tectonism, and eustasy on margin sequence development: late quaternary stratigraphy and evolution of the Ganges-Brahmaputra delta (2000) *Sediment. Geol.*, 133, pp. 227-248
13. Hansen, L.K., Jakobsen, R., Postma, D., Methanogenesis in a shallow sandy aquifer, R??m??, Denmark (2001) *Geochim. Cosmochim. Acta*, 65, pp. 2925-2935
14. Harvey, C.F., Swartz, C.H., Badruzzaman, A.B.M., Keon-Blute, N., Yu, W., Ali, A., Jay, J., Ahmed, M.F., Arsenic mobility and groundwater extraction in Bangladesh (2002) *Science*, 298, pp. 1602-1606
15. Harvey, C.F., Ashfaq, K.N., Yu, W., Badruzzaman, A.B.M., Ali, M.A., Oates, P.M., Michael, H.A., Ahmed, M.F., Groundwater dynamics and arsenic contamination in Bangladesh (2006) *Chem. Geol.*, 228, pp. 112-136
16. Heimann, A.C., Blodau, C., Postma, D., Larsen, F., Pham, H.V., Pham, Q.N., Jessen, S., Jakobsen, R., Hydrogen thresholds and steady state concentrations associated with microbial arsenate respiration (2007) *Environ. Sci. Technol.*, 41, pp. 2311-2317
17. Herbel, M., Fendorf, S., Biogeochemical processes controlling the speciation and transport of arsenic within iron coated sands (2006) *Chem. Geol.*, 228, pp. 16-32
18. Horneman, A., van Geen, A., Mathe Kent, D.V., Zheng, Y.P.E., Dhar, R.K., O'Connell, S., Hoque, M.A., Ahmed, K.M., Decoupling of As and Fe release to Bangladesh groundwater under reducing conditions Part I: evidence from sediment profiles (2004) *Geochim. Cosmochim. Acta*, 68, pp. 3459-3473
19. Hudson-Edwards, K.A., Houghton, S.L., Osborn, A., Extraction and analysis of arsenic in soils and sediments (2004) *Trends Anal. Chem.*, 23, pp. 745-752
20. Islam, F.S., Gault, A.G., Boothman, C., Polya, D.A., Charnock, J.M., Chatterjee, D., Lloyd, J., Role of metal-reducing bacteria in arsenic release from Bengal delta sediments (2004) *Nature*, 430, pp. 68-71
21. Jakobsen, R., Postma, D., Redox zoning, rates of sulfate reduction and interactions with Fe-reduction and methanogenesis in a shallow sandy aquifer, R??m??, Denmark (1999) *Geochim. Cosmochim. Acta*, 63, pp. 137-151
22. Jakobsen, R., Cold, L., Geochemistry at the sulfate reduction-methanogenesis transition zone in an anoxic aquifer-a partial equilibrium interpretation using 2D reactive transport modeling (2007) *Geochim. Cosmochim. Acta*, 71, pp. 1949-1966
23. Keon, N.E., Swartz, C.H., Brabander, D.J., Harvey, C., Hemond, H.F., Validation of an arsenic sequential extraction method for evaluating mobility in sediments (2001) *Environ. Sci. Technol.*, 35, pp. 2778-2784
24. Kj??ller, C., Postma, D., Larsen, F., Groundwater acidification and the mobilization of trace metals in a sandy aquifer (2004) *Environ. Sci. Technol.*, 38, pp. 2829-2835
25. Langmuir, D.L., Mahoney, J., Rowson, J., Solubility products of amorphous ferric arsenate and crystalline scorodite ($\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$) and their application to arsenic behavior in buried mine tailings (2006) *Geochim. Cosmochim. Acta*, 70, pp. 2942-2956
26. Larsen, O., Postma, D., Kinetics of bulk dissolution of lepidocrocite, ferrihydrite and goethite (2001) *Geochim. Cosmochim. Acta*, 65, pp. 1367-1379
27. McArthur, J.M., Ravenscroft, P., Safiulla, S., Thirlwall, M.F., Arsenic in groundwater: testing pollution mechanisms for sedimentary aquifers in Bangladesh (2001) *Water Resour. Res.*, 37, pp. 109-117
28. McArthur, J.M., Banerjee, D.M., Hudson-Edwards, K.A., Mishra, R., Purohit, R., Ravenscroft, P., Cronin, A., Chahda, D.K.,

- Natural organic matter in sedimentary basins and its relation arsenic in anoxic ground water: the example of West Bengal and its worldwide implications (2004) *Appl. Geochem.*, 19, pp. 1255-1293
29. Nickson, R., McArthur, J., Burgess, W., Ahmed, K.M., Ravenscroft, P., Rahman, M., Arsenic poisoning of Bangladesh groundwater (1998) *Nature*, 395, p. 338
 30. Nickson, R.T., McArthur, J.M., Ravenscroft, P., Burgess, W.G., Ahmed, K.M., Mechanism of arsenic release to groundwater, Bangladesh and West Bengal (2000) *Appl. Geochem.*, 15, pp. 403-413
 31. Oremland, R.S., Dowdle, P.R., Hoefft, S., Sharp, J.O., Schaefer, J.K., Miller, L.G., Switzer Blum, J., Wallschlaeger, D., Bacterial dissimilatory reduction of arsenate and sulfate in meromictic Mono Lake, California (2000) *Geochim. Cosmochim. Acta*, 64, pp. 3073-3084
 32. Parkhurst, D.L., Appelo, C.A.J., User's guide to PHREEQC (Version 2)-a computer program for speciation, reaction-path, 1D-transport, and inverse geochemical calculations (1999) U.S. Geol. Surv. Water Resour. Inv. Rep., pp. 99-4259
 33. Pedersen, H.D., Postma, D., Jakobsen, R., Larsen, O., Fast transformation of iron oxyhydroxides by the catalytic action of Fe(II) (2005) *Geochim. Cosmochim. Acta*, 69, pp. 3967-3977
 34. Pedersen, H.D., Postma, D., Jakobsen, R., Release of arsenic associated with the reduction and transformation of iron oxides (2006) *Geochim. Cosmochim. Acta*, 70, pp. 4116-4129
 35. Polizzotto, M.L., Harvey, C.F., Li, G., Badruzzman, B., Ali, A., Newville, M., Sutton, S., Fendorf, S., Solid-phases and desorption processes of arsenic within Bangladesh sediments (2006) *Chem. Geol.*, 228, pp. 97-111
 36. Postma, D., Pyrite and siderite formation in brackish and freshwater swamp sediments (1982) *Am. J. Sci.*, 282, pp. 1151-1183
 37. Postma, D., Jakobsen, R., Redox zonation: equilibrium constraints on the Fe(III)/SO₄-reduction interface (1996) *Geochim. Cosmochim. Acta*, 60, pp. 3169-3175
 38. Postma, D., Boesen, C., Kristiansen, H., Larsen, F., Nitrate reduction in an unconfined sandy aquifer: water chemistry, reduction processes, and geochemical modeling (1991) *Water Resour. Res.*, 27, pp. 2027-2045
 39. Radu, T., Subacz, J.L., Phillippi, J.M., Barnett, M.O., Effects of dissolved carbonate on arsenic adsorption and mobility (2005) *Environ. Sci. Technol.*, 39, pp. 7875-7882
 40. Sengupta, S., Mukherjee, P.K., Pal, T., Shome, S., Nature and origin of arsenic carriers in shallow aquifer sediments of Bengal Delta, India (2004) *Environ. Geol.*, 45, pp. 1071-1081
 41. Seyler, P., Martin, J.-M., Biogeochemical processes affecting arsenic species distribution in a permanently stratified lake (1989) *Environ. Sci. Technol.*, 23, pp. 1258-1263
 42. 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
 43. Stanger, G., A palaeo-hydrogeological model for arsenic contamination in southern and south-east Asia (2005) *Environ. Geochem. Health*, 27, pp. 359-367
 44. Stookey, L.L., Ferrozine-a new spectrophotometric reagent for iron (1970) *Anal. Chem.*, 42, pp. 779-781
 45. Stumm, W., Morgan, J.J., (1981) *Aquatic Chemistry*. second ed., , Wiley & Sons, New York 780 pp
 46. Suter, D., Siffert, C., Sulzberger, B., Stumm, W., Catalytic dissolution of iron(III) (hydr)oxides by oxalic acid in the presence of Fe(II) (1988) *Naturwissenschaften*, 75, pp. 571-573
 47. Swartz, C.H., Blute, N.K., Badruzzman, B., Ali, A., Brabander, D., Jay, J., Besancon, J., Harvey, C., Mobility of arsenic in a Bangladesh aquifer: inferences from geochemical profiles, leaching data, and mineralogical characterization (2004) *Geochim. Cosmochim. Acta*, 68, pp. 539-4557
 48. Swedlund, P.J., Webster, J.G., Adsorption and polymerization of silicic acid on ferrihydrite and its effect on arsenic

adsorption (1999) *Water Res.*, 33, pp. 3413-3422

49. Tessier, A., Campbell, P.G.C., Bisson, M., Sequential extraction procedure for the speciation of particulate trace metals (1979) *Anal. Chem.*, 51 (7), pp. 844-851
50. van Geen, A., Zheng, Y., Versteeg, R., Stute, M., Horneman, A., Dhar, R., Steckler, M., Ahmed, K.M., Spatial variability of arsenic in 6000 tube wells in a 25 km² area of Bangladesh (2003) *Water Resour. Res.*, 39, p. 1140. , 10.1029/2002WR001617
51. van Geen, A., Zheng, Y., Cheng, Z., Aziz, Z., Horneman, A., Dhar, R.K., Mailloux, B., Ahmed, K.M., A transect of groundwater and sediment properties in Araihasar, Bangladesh: further evidence of decoupling between As and Fe mobilization (2006) *Chem. Geol.*, 228, pp. 85-96
52. Wenzel, W.W., Kirchbaumer, N., Prohaska, T., Stingeder, G., Lombic, E., Adriano, D.C., Arsenic fractionation in soils using an improved sequential extraction procedure (2001) *Anal. Chim. Acta*, 436, pp. 309-323
53. Yu, W.H., Harvey, C.M., Harvey, C.F., Arsenic in groundwater in Bangladesh: a geostatistical and epidemiological framework for evaluating health effects and potential remedies (2003) *Water Resour. Res.*, 39, p. 1146

Download Full Text: 0560.pdf