Hydrological and sedimentary controls leading to arsenic contamination of groundwater in the Hanoi area, Vietnam: The impact of iron-arsenic ratios, peat, river bank deposits, and excessive groundwater abstraction

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Abstract: Groundwater contamination by arsenic in Vietnam poses a serious health threat to millions of people. In the larger Hanoi area, elevated arsenic levels are present in both, the Holocene and Pleistocene aquifers. Family-based tubewells predominantly tap the Holocene aquifer, while the Hanoi water works extract more than 600,000?m^3/day of groundwater from the Pleistocene aquifer. Detailed groundwater and sediment investigations were conducted at three locations exhibiting distinct geochemical conditions, i.e., i) high levels of dissolved arsenic (av. 121??g/L) at the river bank, ii) low levels of dissolved arsenic (av. 21??g/L) at the river bank and, iii) medium levels of dissolved arsenic (60??g/L) in an area of buried peat and excessive groundwater abstraction. Seasonal fluctuations in water chemistry were studied over a time span of 14?months. Sediment-bound arsenic (1.3-22??g/g) is in a natural range. Arsenic correlates with iron ($r^2 > 0.8$) with variation related to grain size. Sediment leaching experiments showed that arsenic can readily be mobilized at each of the three locations. Low levels of arsenic in groundwater (< 10??g/L) generally exhibit manganese reducing conditions, whereas elevated levels are caused by reductive dissolution under iron- and sulphate reducing conditions. Average arsenic concentrations in groundwater are twofold higher at the river bank than in the peat area. The lower levels of arsenic contamination in the peat area are likely controlled by the high abundance of iron present in both the aqueous and sediment phases. With median molar Fe/As ratios of 350 in water and 8700 in the sediments of the peat area, reduced iron possibly forms new mineral phases that resorb (or sequester) previously released arsenic to the sediment. Despite similar redox conditions, resorption is much less significant at the river bank (Fe/As(aq) = 68, Fe/As(s) = 4700), and hence, arsenic concentrations in groundwater reach considerably higher levels. Drawdown of Holocene water to the Pleistocene aquifer in the peat area, caused by the pumping for the Hanoi water works, clearly promotes reducing conditions in Pleistocene groundwater. This demonstrates that excessive abstraction of water from deep wells, i.e., wells tapping water below the arsenic burdened depth, can cause a downward shift of iron-reducing conditions and concurrently mobilize arsenic along the way. Vertical migration of reduced groundwater may also impact aquifers under natural hydrological conditions. Seepage of DOC-enriched groundwater derived from degradation of organic matter in the clayey sediments at the river bank

was observed to enhance (and maintain) iron-reducing conditions in the aquifer where organic matter is scarce. Once the aquifer becomes reduced, arsenic is released from the aquifer solid-hosts but additionally derives from the arsenic-enriched groundwater seeping from the clay into the aquifer. This behaviour is an important mechanism for arsenic contamination in aquifers that might not necessarily contain enough organic matter in their sediments to induce reducing conditions independently. ?? 2007 Elsevier B.V. All rights reserved.
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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 (2), pp. 215-230
2. Berg, M., Stengel, C., ARS13-16 arsenic reference samples Interlaboratory Quality Evaluation (IQE) (2004) Report to Participants, Eawag, Swiss Federal Institute of Aquatic Science and Technology (Eawag), Dubendorf, Switzerland
3. 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 (13), pp. 2621-2626
4. Berg, M., Luzi, S., Trang, P.T.K., Viet, P.H., Giger, W., Stuben, D., Arsenic removal from groundwater by household sand filters: comparative field study, model calculations, and health benefits (2006) Environ. Sci. Technol., 40 (17), pp. 5567-5573
5. Berg, M., Stengel, C., Trang, P.T.K., Viet, P.H., Sampson, M.L., Leng, M., Samreth, S., Fredericks, D., Magnitude of arsenic pollution in the Mekong and Red River Deltas - Cambodia and Vietnam (2007) Sci. Total Environ., 372 (2-3), pp. 413-425
6. BGS, DPHE, Arsenic contamination of groundwater in Bangladesh. British Geological Survey Report WC/00/19 (2001) BGS Technical Report WC/00/19, , British Geological Survey, Keyworth, U.K. Keyworth, UK
7. Blute, N.K., Brabander, D.J., Hemond, H.F., Sutton, S.R., Newville, M.G., Rivers, M.L., Arsenic sequestration by ferric iron plaque on cattail roots (2004) Environ. Sci. Technol., 38 (22), pp. 6074-6077
8. Buschmann, J., Kappeler, A., Lindauer, U., Kistler, D., Berg, M., Sigg, L., Arsenite and Arsenate Binding to Dissolved Humic Acids: Influence of pH, Type of Humic Acid and Aluminum (2006) Environ. Sci. Technol., 40 (19), pp. 6015-6020
9. Buschmann, J., Berg, M., Stengel, C., Sampson, M.L., Arsenic and manganese contamination of drinking water resources in Cambodia: coincidence of risk areas with low relief topography (2007) Environ. Sci. Technol., 41 (7), pp. 2146-2152
10. Cao, T.H., Le, V.C., Nguyen, V.K., Bui, V.M., Ngo, N.A., Berg, M., Improving the supply water quality of Hanoi. Part 1: current situation of supply water and challenges for treatment technology (2005) Vietnam Water Suppl. Sewer., 7, pp. 31-35
11. Charlet, L., Polya, D.A., Arsenic in shallow, reducing groundwaters in southern Asia: An environmental health disaster (2006) Elements, 2 (2), pp. 91-96
12. Cheng, Z., Van Geen, A., Seddique, A.A., Ahmed, K.M., Limited temporal variability of arsenic concentrations in 20 wells monitored for 3?years in Araihazar, Bangladesh (2005) Environ. Sci. Technol., 39 (13), pp. 4759-4766
13. Coker, V.S., Gault, A.G., Pearce, C.I., van der Laan, G., Telling, N.D., Charnock, J.M., Polya, D.A., Lloyd, J.R., XAS and XMCD evidence for species-dependent partitioning of arsenic during microbial reduction of ferrihydrite to magnetite (2006) Environ. Sci. Technol., 40 (24), pp. 7745-7750
14. Dixit, S., Hering, J.G., Sorption of Fe(II) and As(III) on goethite in single- and dual-sorbate systems (2006) Chem. Geol., 228 (1-3), pp. 6-15
15. Dodd, M.C., Vu, N.D., Ammann, A., Le, V.C., Kissner, R., Pham, H.V., Cao, T.H., Von Gunten, U., Kinetics and mechanistic aspects of As(III) oxidation by aqueous chlorine, chloramines, and ozone: relevance to drinking water treatment (2006) Environ. Sci. Technol., 40 (10), pp. 3285-3292
16. 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 (9)
17. Duong, H.A., Berg, M., Hoang, M.H., Pham, H.V., Gallard, H., Giger, W., von Gunten, U., Trihalomethane formation by chlorination of ammonium- and bromide-containing groundwater in water supplies of Hanoi, Vietnam (2003) Water Res., 37

(13), pp. 3242-3252

18. Fette, M., Kipfer, R., Schubert, C.J., Hoehn, E., Wehrli, B., Assessing river-groundwater exchange in the regulated Rhone River (Switzerland) using stable isotopes and geochemical tracers (2005) Appl. Geochem., 20 (4), pp. 701-712
19. Harvey, C.F., Swartz, C.H., Badruzzaman, A.B.M., Keon-Blute, N., Yu, W., Ali, M.A., Jay, J., Ahmed, M.F., Arsenic mobility and groundwater extraction in Bangladesh (2002) Science, 298 (5598), pp. 1602-1606
20. Harvey, C.F., Ashfaque, 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 (1-3), pp. 112-136
21. Helz, G.R., Tossell, J.A., Charnock, J.M., Pattrick, R.A.D., Vaughan, D.J., Garner, C.D., oligomerization in as(Iii) sulfide solutions - theoretical constraints and spectroscopic evidence (1995) Geochim. Cosmochim. Acta, 59 (22), pp. 4591-4604
22. Herbel, M., Fendorf, S., Biogeochemical processes controlling the speciation and transport of arsenic within iron coated sands (2006) Chem. Geol., 228 (1-3), pp. 16-32
23. Horneman, A., Van Geen, A., Kent, D.V., Mathe, P.E., Zheng, Y., Dhar, R.K., O'Connell, S., Ahmed, K.M., Decoupling of As and Fe release to Bangladesh groundwater under reducing conditions. Part 1: evidence from sediment profiles (2004) Geochim. Cosmochim. Acta, 68 (17), pp. 3459-3473
24. 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 (6995), pp. 68-71
25. Islam, F.S., Pederick, R.L., Gault, A.G., Adams, L.K., Polya, D.A., Charnock, J.M., Lloyd, J.R., Interactions between the Fe(III)-reducing bacterium Geobacter sulfurreducens and arsenate, and capture of the metalloid by biogenic Fe(II) (2005) Appl. Environ. Microbiol., 71 (12), pp. 8642-8648
26. Kapaj, S., Peterson, H., Liber, K., Bhattacharya, P., Human health effects from chronic arsenic poisoning - a review (2006) J. Environ. Sci. Health Part A Toxic Hazard. Subst. Environ. Eng., 41 (10), pp. 2399-2428
27. 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 (13), pp. 2778-2784
28. Klump, S., Kipfer, R., Cirpka, O.A., Harvey, C.F., Brennwald, M.S., Ashfaque, K.N., Badruzzaman, A.B.M., Imboden, D.M., Groundwater dynamics and arsenic mobilization in Bangladesh assessed using noble gases and tritium (2006) Environ. Sci. Technol., 40 (1), pp. 243-250
29. Kocar, B.D., Herbel, M.J., Tufano, K.J., Fendorf, S., Contrasting effects of dissimilatory iron(III) and arsenic(V) reduction on arsenic retention and transport (2006) Environ. Sci. Technol., 40 (21), pp. 6715-6721
30. 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 (1), pp. 109-117
31. McArthur, J.M., Banerjee, D.M., Hudson-Edwards, K.A., Mishra, R., Purohit, R., Ravenscroft, P., Cronin, A., Chadha, D.K., Natural organic matter in sedimentary basins and its relation to arsenic in anoxic ground water: the example of West Bengal and its worldwide implications (2004) Appl. Geochem., 19 (8), pp. 1255-1293
32. Meharg, A.A., Scrimgeour, C., Hossain, S.A., Fuller, K., Cruickshank, K., Williams, P.N., Kinniburgh, D.G., Codeposition of organic carbon and arsenic in Bengal Delta aquifers (2006) Environ. Sci. Technol., 40 (16), pp. 4928-4935
33. Nguyen, V.D., Personal communication (2005) Northern Hydrogeological and Engineering Geological Division, , Vietnam Geological Survey, Hanoi, Vietnam
34. Nhan, D.D., Giap, T.V., Lieu, D.B., Loan, D.T., Anh, V.T., Minh, D.M., Thinh, N.T.H., Viet, D.D., (2005) Arsenic Contaminated Groundwater in the Hanoi Area and Its Studies Using Isotope Technique, , Institute for Nuclear Science and Technology, Hanoi, Vietnam
35. NHEGD, Annual Report 2001 (2002) Northern Hydrogeological and Engineering Geological Division, , Vietnam Geological

Survey, Hanoi, Vietnam

36. 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 (4), pp. 403-413
37. Polizzotto, M.L., Harvey, C.F., Li, G.C., Badruzzman, B., Ali, A., Newville, M., Sutton, S., Fendorf, S., Solid-phases and desorption processes of arsenic within Bangladesh sediments (2006) Chem. Geol., 228 (1-3), pp. 97-111
38. Postma, D., Larsen, F., Minh Hue, N.T., Duc, M.T., Viet, P.H., Nhan, P.Q., Jessen, S., Arsenic in groundwater of the Red River floodplain, Vietnam: Controlling geochemical processes and reactive transport modeling (2007) Geochim. Cosmochim. Acta, 71 (21), pp. 5054-5071
39. Redfield, A.C., The biological control of chemical factors in the environment (1958) Am. Sci., 46, pp. 205-221
40. Roman-Ross, G., Cuello, G.J., Turrillas, X., Fernandez-Martinez, A., Charlet, L., Arsenite sorption and co-precipitation with calcite (2006) Chem. Geol., 233 (3-4), pp. 328-336
41. Rowland, H.A.L., Polya, D.A., Lloyd, J.R., Pancost, R.D., Characterisation of organic matter in a shallow, reducing, arsenic-rich aquifer, West Bengal (2006) Org. Geochem., 37 (9), pp. 1101-1114
42. Rowland, H.A.L., Pederick, R.L., Polya, D.A., Pancost, R.D., Van Dongen, B.E., Gault, A.G., Vaughan, D.J., Lloyd, J.R., The control of organic matter on microbially mediated iron reduction and arsenic release in shallow alluvial aquifers, Cambodia (2007) Geobiology, 5 (3), pp. 281-292
43. Rozanski, K., Aragaus, L., Gonfiantini, R., Swart, P.K., Lohmann Kyger, C., McKenzie, J.A., Savin, S., Isotopic patterns in modern global precipitation (1993) Geophysical Monograph, 78, pp. 1-36. , Swart P.K., et al. (Ed), Americian Geophysical Union
44. Smedley, P.L., Kinniburgh, D.G., A review of the source, behaviour and distribution of arsenic in natural waters (2002) Appl. Geochem., 17 (5), pp. 517-568
45. Smith, A.H., Lingas, E.O., Rahman, M., Contamination of drinking-water by arsenic in Bangladesh: a public health emergency (2000) Bull. World Health Organ., 78 (9), pp. 1093-1103
46. Spycher, N.F., Reed, M.H., As(Iii) and Sb(Iii) sulfide complexes - an evaluation of stoichiometry and stability from existing experimental-data (1989) Geochim. Cosmochim. Acta, 53 (9), pp. 2185-2194
47. Stuben, D., Berner, Z., Chandrasekharam, D., Karmakar, J., Arsenic enrichment in groundwater of West Bengal, India: geochemical evidence for mobilization of As under reducing conditions (2003) Appl. Geochem., 18 (9), pp. 1417-1434
48. Trafford, J.M., Lawrence, A.R., Macdonald, D.M.J., Nguyen, V.D., Tran, D.N., Nguyen, T.H., The effect of urbanisation on the groundwater quality beneath the city of Hanoi, Vietnam (1996) BGS Technical Report WC/96/22, , British Geological Survey, Keyworth, UK
49. Trang, P.T.K., Berg, M., Viet, P.H., Van Mui, N., Van Der Meer, J.R., Bacterial bioassay for rapid and accurate analysis of arsenic in highly variable groundwater samples (2005) Environ. Sci. Technol., 39 (19), pp. 7625-7630
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 (5)
51. Van Geen, A., Rose, J., Thoral, 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 (17), pp. 3475-3486
52. Wasserman, G.A., Liu, X.H., Parvez, F., Ahsan, H., Factor-Litvak, P., van Geen, A., Slavkovich, V., Graziano, J.H., Water arsenic exposure and children's intellectual function in Araihazar, Bangladesh (2004) Environ. Health Perspect., 112 (13), pp. 1329-1333
53. Water-Master-Plan, Water master plan of Hanoi City for the period of 1993-2010 (1993) Vol. 1, The Social Republic of

Vietnam, Hanoi People's Committee. The Republic of Finland, Finnish International Development Agency FINNIDA, Hanoi, Vietnam

54. White, D.E., Hem, J.D., Waring, G.A., Chemical composition of subsurface water, Data of Geochemistry (1963) US Geological Survey
55. Zheng, Y., Stute, M., van Geen, A., Gavrieli, I., Dhar, R., Simpson, H.J., Schlosser, P., Ahmed, K.M., Redox control of arsenic mobilization in Bangladesh groundwater (2004) Appl. Geochem., 19 (2), pp. 201-214