

# Palaeo-hydrogeological control on groundwater As levels in Red River delta, Vietnam

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**Abstract:** To study the geological control on groundwater As concentrations in Red River delta, depth-specific groundwater sampling and geophysical logging in 11 monitoring wells was conducted along a 45 km transect across the southern and central part of the delta, and the literature on the Red River delta's Quaternary geological development was reviewed. The water samples ( $n = 30$ ) were analyzed for As, major ions,  $\text{Fe}^{2+}$ ,  $\text{H}_2\text{S}$ ,  $\text{NH}_4$ , ?? $^{18}\text{O}$  and ??D, and the geophysical log suite included natural gamma-ray, formation and fluid electrical conductivity. The SW part of the transect intersects deposits of grey estuarine clays and deltaic sands in a 15-20 km wide and 50-60 m deep Holocene incised valley. The NE part of the transect consists of 60-120 m of Pleistocene yellowish alluvial deposits underneath 10-30 m of estuarine clay overlain by a 10-20 m veneer of Holocene sediments. The distribution of ?? $^{18}\text{O}$ -values (range -12.2? to -6.3?) and hydraulic head in the sample wells indicate that the estuarine clay units divide the flow system into an upper Holocene aquifer and a lower Pleistocene aquifer. The groundwater samples were all anoxic, and contained  $\text{Fe}^{2+}$  (0.03-2.0 mM), Mn (0.7-320 ?M),  $\text{SO}_4$  (<2.1 ?M-0.75 mM),  $\text{H}_2\text{S}$  (<0.1-7.0 ?M),  $\text{NH}_4$  (0.03-4.4 mM), and  $\text{CH}_4$  (0.08-14.5 mM). Generally, higher concentrations of  $\text{NH}_4$  and  $\text{CH}_4$  and low concentrations of  $\text{SO}_4$  were found in the SW part of the transect, dominated by Holocene deposits, while the opposite was the case for the NE part of the transect. The distribution of the groundwater As concentration (<0.013-11.7 ?M; median 0.12 ?M (9 ?g/L)) is related to the distribution of  $\text{NH}_4$ ,  $\text{CH}_4$  and  $\text{SO}_4$ . Low concentrations of As (??0.32 ?M) were found in the Pleistocene aquifer, while the highest As concentrations were found in the Holocene aquifer. PHREEQC-2 speciation calculations indicated that  $\text{Fe}^{2+}$  and  $\text{H}_2\text{S}$  concentrations are controlled by equilibrium for disordered mackinawite and precipitation of siderite. An elevated groundwater salinity (Cl range 0.19-65.1 mM) was observed in both aquifers, and dominated in the deep aquifer. A negative correlation between aqueous As and an estimate of reduced  $\text{SO}_4$  was observed, indicating that Fe sulphide precipitation poses a secondary control on the groundwater As concentration. ?? 2008 Elsevier Ltd. All rights reserved.

**Index Keywords:** Aquifers; Calcite; Clay deposits; Clay minerals; Concentration (process); Electric conductivity; Estuaries; Gamma rays; Geophysics; Groundwater; Hydrogeology; Iron compounds; Iron ores; Manganese; Manganese compounds; Offshore oil well production; Oil well logging; River control; Rivers; Sedimentation; Underground reservoirs; Wells; Alluvial deposits; Aqueous; Electrical conductivities; Flow

systems; Gamma-ray; Geological controls; Geological developments; Geophysical loggings; Geophysical logs; Groundwater samplings; Higher concentrations; Holocene; Holocene sediments; Hydraulic heads; Hydrogeological; Low concentrations; Major ions; Monitoring wells; Negative correlations; Phreeqc; Pleistocene; Pleistocene aquifers; Red river deltas; Secondary controls; Speciation calculations; Sulphide precipitations; Vietnam; Water samples; Groundwater resources; aquifer; arsenic; electrical conductivity; Holocene; hydrogeology; iron; oxygen isotope; paleohydrology; Pleistocene; precipitation (chemistry); salinity; siderite; speciation (chemistry); sulfur compound; well logging; Asia; Eurasia; Red River Delta; Southeast Asia; Viet Nam

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#### References:

1. Ahmed, K.M., Bhattacharya, P., Hasan, M.A., Akhter, S.H., Alam, S.M.M., Bhuyian, M.A.H., Imam, M.B., Sracek, O., Arsenic enrichment in groundwater of the alluvial aquifers in Bangladesh: an overview (2004) *Appl. Geochem.*, 19, pp. 181-200
2. Akai, J., Izumi, K., Fukuhara, H., Masuda, H., Nakano, S., Yoshimura, T., Ohfuri, H., Kurumi Akai, K., Mineralogical and geomicrobiological investigations on groundwater arsenic enrichment in Bangladesh (2004) *Appl. Geochem.*, 19, pp. 215-230
3. Amini, M., Abbaspour, K.C., Berg, M., Winkel, L., Hug, S.J., Hoehn, E., Yang, H., Johnson, C.A., 2008. Statistical modelling of global geogenic arsenic contamination in groundwater. *Environ. Sci. Technol.* 42Badloe, C., Nguyen, T.P.T., Nguyen, Q.H., 2004. Random survey of arsenic contamination in tubewell water of 12 provinces in Vietnam and initially human health arsenic risk assessment through food chain. In: Proc. Third Scientific Conf. Hanoi University of Science, Multidisciplinary Scientific Session "Environmental Science - Technology and Sustainable Development", 16 November, CETASD, Hanoi, VietnamBerg, M., Tran, H.C., Nguyen, T.C., Viet, P.H., 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
4. 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, pp. 413-425
5. Bostick, B.C., Chen, C., Fendorf, S., Arsenite retention mechanisms within estuarine sediments of Pescadero, CA (2004) *Environ. Sci. Technol.*, 38, pp. 3299-3304
6. Boyd, W., Lam, D.D., Holocene elevated sea levels on the north coast of Vietnam (2004) *Aust. Geogr. Stud.*, 42, pp. 77-88
7. Charlet, L., Polya, D.A., Arsenic in shallow, reducing groundwaters in southern Asia: an environmental health disaster (2006) *Elements*, 2, pp. 91-96
8. Cline, J.D., Spectrophotometric determination of hydrogen sulfide in natural waters (1967) *Limnol. Oceanogr.*, 14, pp. 454-458
9. Delemos, J.L., Bostick, B.C., Renshaw, C.E., St?rup, S., Feng, X., Landfill-stimulated iron reduction and arsenic release at the Coakley Superfund Site (NH) (2006) *Environ. Sci. Technol.*, 40, pp. 67-73
10. Funabiki, A., Haruyama, S., Nguyen, V.Q., Viet, P.H., Dinh, H.T., Holocene delta plain development in the Song Hong (Red River) delta, Vietnam (2007) *J. Asian Earth Sci.*, 30, pp. 518-529
11. Gani, M.R., Alam, M.M., Fluvial facies architecture in small-scale river systems in the Upper Dupi Tila Formation, northeast Bengal Basin, Bangladesh (2004) *J. Asian Earth Sci.*, 24, pp. 225-236
12. Goodbred, 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. Guillot, S., Charlet, L., Bengal arsenic, an archive of Himalaya orogeny and paleohydrology (2007) *J. Environ. Sci. Health, A*, 42, pp. 1785-1794
14. Hanebutth, T.J.J., Saito, Y., Tanabe, S., Quang, L.V., Quang, T.N., Sea levels during late marine isotope stage 3 (or older?) reported from the Red River delta (northern Vietnam) and adjacent regions (2006) *Quatern. Int.*, (145-146), pp. 119-134

15. Harvey, C.F., Swartz, C.H., Badruzzaman, A.B.M., Keon-Blute, N., Yu, W., Ali, M.A., Jay, J., Ahmed, M.F., Groundwater arsenic contamination on the Ganges Delta: biogeochemistry, hydrology, human perturbations, and human suffering on a large scale (2005) *C. R. Geosci.*, 337, pp. 285-296
16. Hori, K., Tanabe, S., Saito, Y., Haruyama, S., Viet, N., Kitamura, A., Delta initiation and Holocene sea-level change: example from the Song Hong (Red River) delta, Vietnam (2004) *Sediment. Geol.*, 164, pp. 237-249
17. Hossain, F., Hill, J., Bagtzoglou, A.C., Geostatistically based management of arsenic contaminated ground water in shallow wells of Bangladesh (2007) *Water Resour. Manage.*, 21, pp. 1245-1261
18. 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
19. Jian, Z., Huang, B., Kuhnt, W., Lin, H.-L., Late Quaternary upwelling intensity and East Asian monsoon forcing in the South China Sea (2001) *Quatern. Res.*, 55, pp. 336-370
20. Kitazawa, T., Pleistocene macrotidal tide-dominated estuary-delta succession, along the Dong Nai River, southern Vietnam (2007) *Sediment. Geol.*, 194, pp. 115-140
21. Lam, D.D., Boyd, W.E., Holocene costal stratigraphy and the sedimentary development of the Hai Phong area of the Bac Bo plain (Red River delta), Vietnam (2003) *Aust. Geogr.*, 34, pp. 177-194
22. Lambeck, K., Yokoyama, Y., Purcell, T., Into and out of the last glacial maximum: sea-level change during oxygen isotope stages 3 and 2 (2002) *Quatern. Sci. Rev.*, 21, pp. 343-360
23. Langmuir, D.L., Mahoney, J., Rowson, J., Solubility products of amorphous ferric arsenate and crystalline scorodite ( $\text{FeAsO}_4\{\text{radical dot}\}2\text{H}_2\text{O}$ ) and their application to arsenic behavior in buried mine tailings (2006) *Geochim. Cosmochim. Acta*, 70, pp. 2942-2956
24. Larsen, F., Pham, N.Q., Dang, N.D., Postma, D., Jessen, S., Pham, V.H., Nguyen, T.B., Refsgaard, J.C., Controlling geological and hydrogeological processes in an arsenic contaminated aquifer on the Red River flood plain, Vietnam (2008) *Appl. Geochem.*, 23 (11), pp. 3099-3115
25. Li, Z., Saito, Y., Matsumoto, E., Wang, Y., Tanabe, S., Quang, L.V., Climate change and human impact on the Song Hong (Red River) delta, Vietnam, during the Holocene (2006) *Quatern. Int.*, 144, pp. 4-28
26. Liew, P.M., Kuo, C.M., Huang, S.Y., Tseng, M.H., Vegetation change and terrestrial carbon storage in eastern Asia during the last glacial maximum as indicated by a new pollen record from central Taiwan (1998) *Global Planet. Change*, (16-17), pp. 85-94
27. Lowers, H.A., Breit, G.N., Foster, A.L., Whitney, J., Yount, J., Uddin, Md.N., Muneem, Ad.A., Arsenic incorporation into authigenic pyrite, Bengal Basin sediment, Bangladesh (2007) *Geochim. Cosmochim. Acta*, 71, pp. 2699-2717
28. Mathers, S., Zalasiewicz, J., Holocene sedimentary architecture of the Red River delta Vietnam (1999) *J. Coast. Res.*, 15, pp. 314-325
29. 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
30. 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. Invest. Rep.*, pp. 99-4259
31. Petersen, H.I., Andersen, C., Anh, P.H., Bojesen-Koefoed, J.A., Nielsen, L.H., Nytoft, H.P., Rosenberg, P., Thanh, L., Petroleum potential of Oligocene lacustrine mudstones and coals at Dong Ho, Vietnam - an outcrop analogue to terrestrial source rocks in the greater Song Hong Basin (2001) *J. Asian Earth Sci.*, 19, pp. 135-154
32. Petersen, H.I., Vu, T., Nielsen, L.H., Nguyen, A.D., Nytoft, H.P., Source rock properties of lacustrine mudstones and coals (Oligocene Dong Ho Formation), onshore Song Hong Basin, northern Vietnam (2005) *J. Petrol. Geol.*, 28, pp. 19-38

33. Polya, D.A., Gault, A.G., Diebe, N., Feldman, P., Rosenboom, J.W., Gilligan, E., Fredericks, D., Cooke, D.A., Arsenic hazard in shallow Cambodian groundwaters (2005) *Mineral. Mag.*, 69, pp. 807-823
34. Postma, D., Larsen, F., Hue, N.T.M., 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, pp. 5054-5071
35. Ravenscroft, P., McArthur, J.M., Hoque, B.A., Geochemical and palaeohydrological controls on pollution of groundwater by arsenic (2001) Fourth Int. Conf. Arsenic Exposure and Health Effects, , Chappell W.R., Abernathy C.O., and Calderon R.L. (Eds), Elsevier Science, Ltd, Oxford
36. Ravenscroft, P., Burgess, W.G., Ahmed, K.M., Burren, M., Perrin, J., Arsenic in groundwater of the Bengal Basin, Bangladesh: distribution, field relations, and hydrogeological setting (2005) *Hydrogeol. J.*, 13, pp. 727-751
37. Rodríguez Lado, L., Polya, D., Winkel, L., Berg, M., Hegan, A., Modelling arsenic hazard in Cambodia: A geostatistical approach using ancillary data (2008) *Appl. Geochem.*, 23 (11), pp. 3009-3018
38. Rost, K.T., Pleistocene paleoenvironmental changes in the high mountain ranges of central China and adjacent regions (2000) *Quatern. Int.*, (65-66), pp. 147-160
39. 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, pp. 1101-1114
40. Rowland, H.A.L., Pederick, R.L., Polya, D.A., Pancost, R.A., 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, pp. 281-292
41. 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
42. Shah, B.A., Role of Quaternary stratigraphy on arsenic-contaminated groundwater from parts of Middle Ganga Plain, UP-Bihar, India (2008) *Environ. Geol.*, 53, pp. 1553-1561
43. 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
44. Stanger, G., A palaeo-hydrogeological model for arsenic contamination in southern and south-east Asia (2005) *Environ. Geochem. Health*, 27, pp. 359-367
45. Stollenwerk, K.G., Breit, G.N., Welch, A.H., Yount, J.C., Whitney, J.W., Foster, A.L., Uddin, M.N., Ahmed, N., Arsenic attenuation by oxidized aquifer sediments in Bangladesh (2007) *Sci. Total Environ.*, 379, pp. 133-150
46. Stookey, L.L., Ferrozine - a new spectrophotometric reagent for iron (1970) *Anal. Chem.*, 42, pp. 779-781
47. Stumm, W., Morgan, J.J., (1981) *Aquatic Chemistry*. second ed., , Wiley & Sons, New York
48. Swartz, C.H., Blute, N.K., Badruzzaman, B., Ali, A., Brabander, D., Jay, J., Besancon, J., Harvey, C.F., Mobility of arsenic in a Bangladesh aquifer: Inferences from geochemical profiles, leaching data, and mineralogical characterization (2004) *Geochim. Cosmochim. Acta*, 68, pp. 4539-4557
49. Tanabe, S., Hori, K., Saito, Y., Haruyama, S., Le, Q.D., Sato, Y., Hiraide, S., Sedimentary facies and radiocarbon dates of the Nam Dinh-1 core from the Song Hong (Red River) delta, Vietnam (2003) *J. Asian Earth Sci.*, 21, pp. 503-513
50. Tanabe, S., Hori, K., Saito, Y., Haruyama, S., Van, P.V., Kitamura, A., Song Hong (Red River) delta evolution related to millennium-scale Holocene sea-level changes (2003) *Quatern. Sci. Rev.*, 22, pp. 2345-2361
51. Tanabe, S., Saito, Y., Quang, L.V., Hanebuth, T.J.J., Quang, L.N., Kitamura, A., Holocene evolution of the Song Hong (Red River) delta system, northern Vietnam (2006) *Sediment. Geol.*, 187, pp. 29-61
52. Tran, N., Ngo, Q.T., Do, T.V.T., Nguyen, D.M., Nguyen, V.V., Quaternary sedimentation of the principal deltas of Vietnam

- (1991) J. Southeast Asian Earth Sci., 6, pp. 103-110
- 53. Tran, N., Mai, T.N., Chu, V.N., Hoekstra, P., Weering, V.Tj., van den Bergh, J.H., Dinh, X.T., Vu, V.P., Holocene sedimentary evolution, geodynamic and anthropogenic control of the Balat river mouth formation (Red River-delta, northern Vietnam) (2002) Z. Geol. Wiss., Berlin, 30, pp. 157-172
  - 54. Traynor, J.J., Sladen, C., Seepage in Vietnam - onshore and offshore examples (1997) Mar. Petrol. Geol., 14, pp. 345-362
  - 55. Vu, T.C., 1996. Salinity intrusion in the Red River delta. Seminar on Environment and Development in Vietnam, December 6-7, Australian National University. (See coombs.anu.edu.au/~vern/env\_dev/seminar96.html)Winkel, L., Berg, M., Amini, M., Hug, S.J., Johnson, C.A., 2008. Predicting groundwater arsenic contamination in Southeast Asia from surface parameters. Nature Geosci. 1, 536-542Yokoyama, Y., Lambeck, K., Deckker, P.D., Johnston, P., Fifield, L.K., Timing of the last glacial maximum from observed sea-level minima (2000) Nature, 406, pp. 713-716
  - 56. Zheng, Z., Li, Q., Vegetation, climate, and sea level in the past 55,000 years, Hanjiang Delta, southeastern China (2000) Quatern. Res., 53, pp. 330-340

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