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Provenance of High Arsenic Holocene Sediment and Low Arsenic Pleistocene Sediment in Bangladesh Aquifers: A Preliminary Assessment

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Geogenic arsenic (As) in drinking water, especially in groundwater, has impacted the health of over 100 million people worldwide, with an estimated 20 to 50 million people at risk in Bangladesh alone. It is understood that a combination of sluggish flow and reducing chemical environment in sedimentary aquifers releases As to groundwater. The role of the aquifer sediment's origin on As occurrence in groundwater is incompletely known because conventional wisdom suggests there is sufficient As in geological material and that biogeochemical reactions for mobilization are the primary control of groundwater As. This view, while correct for some specific aquifers, does not consider the heterogeneity of As in the upper crust and its implications at the basin scale.

The geochemical cycle of As leads to enrichment of As in the upper crust (Rudnick and Gao, 2003). Sedimentary processes also influence the distribution of As. Under low temperature conditions, anoxic marine sediments have functioned as widespread sinks with typically small As enrichment, in many cases also associated with organic matter. This is evident from the As enrichment in carbonaceous shales (Tourtelot, 1964). Thermal alteration within the earth preferentially partitions arsenic, as an incompatible element, into the fluid phase resulting in As-rich fluids that deposit As in favorable environments along the flow path (Goldhaber et al., 2003). Sedimentary and hydrothermal processes are especially notable where high As anomalies have been reported in association with metal (gold, antimony and copper) ore (van Hees et al., 1999, Hu et al., 2004) and coal deposits (Yudovich and Ketris, 2005). Mining and mineral processing activities accelerate the movement of As from these enriched solid sources to aqueous environments and ecosystems. A parallel process, albeit at a slower rate through the weathering and transport from enriched sources to the low gradient depositional environments, is likely to lead to As enrichments downstream (Saunders et al., 2005, Stanger, 2005, Guillot and Charlet, 2007). Specifically, these studies recommended that the search for the original source of As should be conducted in ophiolitic, As-rich, arc-related rocks in the Indus-Tsangpo suture zone (Hattori and Guillot, 2003), where As was thought to be originally accumulated in oceanic ferro-manganoan sediments of the eastern Paleo-Tethys (Stanger, 2005). To resolve the importance of the impact of these processes, this study assesses the origin of the sediment in two aquifers of Bangladesh that contain distinctly different As levels: the Holocene aquifer with elevated groundwater and sediment extractable As and the Pleistocene aquifer with low groundwater and sediment extractable As (Table 1).

Table 1. Summary of As data from Aquifer Sands in Bangladesh

	Bulk As				Extractable As				Ratio of extractable As			
	n	mean	min	max	n	mean	min	max	n	mean	min	max
High As Holocene Aquifer	101	4.0	0.4	55.0	70	2.0	0.05	40.2	63	0.37	0.01	0.97
Low As Pleistocene Aquifer	27	1.5	0.3	5.4	16	0.1	0.01	0.5	15	0.09	0.00	0.37

* Source of Data: BGS and DPHE 01; Swartz 04, Zheng 05, Stollenwerk 07, Seddique 08, Selim Reza 10, Uddin 11;
* Locations of cores in Fig. 4a;
* Extractable As are mostly by oxalate, except for Zheng 05 which used phosphate;
* Bulk As, extractable As, and the ratio of the two between Holocene and Pleistocene aquifers are significantly different at 99% confidence level. Two-tailed student t-test p-values are 0.003, 0.003, and 5E-08, respectively.

The contrast in As content between the Holocene and the Pleistocene aquifers was attributed to a longer history (Zheng et al., 2004) and more vigorous flushing (van Geen et al., 2008) of the Pleistocene aquifer during the low sea level periods during glacial intervals. However, this explanation assumes that the sediments from both aquifers are derived from a similar source and thus with comparable As contents in similar mineral assemblages before flushing. In this study we will compare the origin of low-As Plio-Pleistocene aquifer and high-As Holocene aquifer sediments in the Ganges-Brahmaputra basin by determining the age of detrital components with samples (n=20) from two sediment cores collected in Bangladesh

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(Fig. 1). New data from Ar-Ar dating of individual detrital muscovite combined with bulk sediment and carbonate $^{87}Sr/^{86}Sr$ will provide constraints to sediment sources and thus the ultimate geological source of As for Asian deltas.

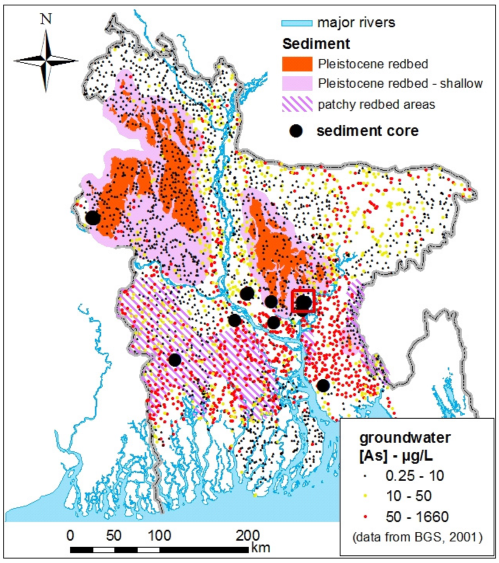

Fig. 1. Location of sediment cores used for provenance study (solid circle with red square, same study sites from (Zheng et al., 2005) overlain groundwater As distribution map in Bangladesh. Three types of Pleistocene red beds where low As groundwater is found are indicated (Hoque et al., 2011).

Key words: arsenic, geological source, Ar-Ar age of muscovite, Sr isotope, Bangladesh, sediment provenance

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