References

1. Neumann, R. B. *et al. Nature Geosci.* **3,** 46–52 (2010).
2. Argos, M. *et al. Lancet* **376,** 252–258 (2010).
3. Harvey, C. F. *et al. Science* **298,** 1602–606 (2002).
4. Sengupta, S., McArthur, J. M., Sarkar, A. & Leng, M. J. *Environ. Sci. Technol.* **42,** 5156–5164 (2008).
5. McArthur J. M. *et al. Appl. Geochem.* **19,** 1255–1293 (2004).
6. Klump, S. *et al. Environ. Sci. Technol.* **40,** 243–250 (2006).
7. Swartz, C. H. *et al. Geochim. Cosmochim. Acta* **68,** 4539–4557 (2004).
8. Harvey, C. F. *et al. Chem. Geol.* **228,** 112–136 (2006).

J. M. McArthur[1]*, P. Ravenscroft[2] and O. Sracek[3]
[1]Earth Sciences, University College London, Gower Street, London WC1E 6BT, UK, [2]AMEC Entec, 17 Angel Gate, City Road, London EC1V 2SH, [3]Department of Geology, Faculty of Science, Palacký University, 17. listopadu 12, 771 46 Olomouc, Czech Republic.
*e-mail: j.mcarthur@ucl.ac.uk

Authors' reply — McArthur *et al.* argue that a single process, termed aquifer flushing, explains the pattern of dissolved arsenic concentrations we observe in groundwater in Bangladesh. We concur that rice-field recharge has the potential to flush out arsenic-contaminated groundwater at shallow depths; we have shown that rice-field recharge carries little arsenic or biologically available organic carbon that could mobilize arsenic from the aquifer[1–3]. However, we contend that their interpretation explains neither the origin of the high arsenic concentrations in groundwater located at intermediate depths, nor why concentrations decline at greater depths.

We argue that McArthur *et al.*'s interpretation is inconsistent with the physics that force groundwater layering. Pond recharge must flow horizontally beneath low-arsenic recharge from rice fields, to reach the irrigation wells and river channels where it discharges. Thus pond recharge should predominately occupy arsenic-contaminated intermediate depths[2,4].

In their Fig. 1a, McArthur *et al.* compare water isotope data from one location with arsenic concentration data from multiple locations, where flow patterns differ. In fact, the δ^{18}O minimum does align with the arsenic peak, within the resolution of the data, when data from the same wells are compared, contradicting their assertion of a mismatch (see Supplementary Information and their Fig. 1c). Our interpretation, however, does not rely on this alignment.

Plumes of different solutes that originate from the same source are often not collocated. Solutes follow different patterns because surface sorption retards transport to varying degrees and many solutes, including arsenic, are mobilized from the aquifer. Reactive transport of most solutes in groundwater is not explained by 'endmember' mixing, as suggested by McArthur *et al.* Indeed, according to their proposed model of linear mixing, the consistent decline in arsenic concentrations with depth, below its peak, implies more flushing towards the bottom of the aquifer, an implication that we feel is physically implausible.

In their Fig. 1b, McArthur *et al.* do not present our measurements[5] from 30 m and below, arguing that they are of different derivation and age. However, the origin of the contaminated groundwater is the question being pursued; all samples have different ages; and ponds existed before the advent of irrigation pumping. When the complete data set is plotted, the trends apparent in their Fig. 1b are no longer evident (see Supplementary Information).

Our interpretation — that water within the intermediate contaminated zone originates from pond recharge — is also supported by chemical analysis showing that ponds provide dissolved organic carbon that is biologically available; carbon-dating analysis suggesting that organic carbon concentrations are maintained by old organic carbon released from the aquifer; and the observation that only pond water from early in the dry season can provide the isotopically light recharge matching contaminated groundwater. ❐

References

1. Harvey, C. F. *et al. Science* **300,** 584D–U3 (2003).
2. Neumann, R. B. *et al. Nature Geosci.* **3,** 46–52 (2010).
3. Neumann, R. B. *et al. Environ. Sci. Technol.* **45,** 2072–2078 (2011).
4. Harvey, C. F. *Nature* **454,** 415–416 (2008).
5. Swartz, C. H. *et al. Geochim. Cosmochim. Acta* **68,** 4539–4557 (2004).

Additional information

Supplementary information accompanies this paper on www.nature.com/naturegeoscience.

Rebecca B. Neumann[1,2], Khandaker N. Ashfaque[1], A. B. M. Badruzzaman[3], M. Ashraf Ali[3], Julie K. Shoemaker[4] and Charles F. Harvey[1]*
[1]Massachussetts Institute of Technology, Cambridge, Massachusetts 02139, USA, [2]University of Washington, Seattle, Washington 98195, USA, [3]Bangladesh University of Engineering and Technology, Dhaka 1000, Bangladesh, [4]Harvard University, Cambridge, Massachusetts 02138, USA.
*e-mail: charvey@mit.edu