Response to Comments on "Arsenic Mobility and Groundwater Extraction in Bangladesh"

Aggarwal *et al.* (*1*) and van Geen *et al.* (*2*) point to selected data in (*3*) to craft contrasting regional generalizations about the cause of high arsenic concentrations in groundwater in Bangladesh. Aggarwal *et al.* argue that arsenic mobilization is related to continuing groundwater recharge, but that groundwater flow is not influenced by groundwater extraction, whereas van Geen *et al.* argue that arsenic mobilization occurs in stagnant groundwater. Both claims are contradicted by multiple sources of hydraulic and geochemical data from our site (*4*), contain serious inconsistencies, and ignore basic hydrologic processes.

Aggarwal *et al.* (*1*) argue that extensive extraction of groundwater for irrigation does not affect groundwater flow at our study site. They incorrectly assert that we inferred (*4*) the effects of pumping from water isotopes and water table movement. Three points are important to consider. First, our value of pumping-induced flow was calculated from knowledge of the location and pumping rates of irrigation wells [figure 4 in (*4*)]. There is no need to infer pumping-induced flow through indirect means. Second, Aggarwal *et al.* provide no physical principle supporting their contention that irrigation withdrawal does not cause groundwater flow. Pumping from a well does draw water to the well screen and must drive flow through the aquifer. Third, site data clearly show the effects of pumping. Hydraulic head declines sharply at irrigation onset (Fig. 1A), and flow converges at the depth of the arsenic peak later in the irrigation season (Fig. 1B), two effects for which we see no plausible explanation other than pumping.

Despite the physical implausibility of their contention that pumping does not affect flow, Aggarwal *et al.* interpret water isotopes (tritium and stable isotopes) from other areas of Bangladesh to support their argument. Contrary to their interpretation, the tritium data are consistent with increased groundwater flow to depth after the onset of groundwater irrigation. None of the wells below 25 m had tritium values above 1 tritium unit (TU) in 1979. However, four wells showed measurable tritium in 1999, consistent with migration of young water to extraction depths after the marked increase in irrigation with pumped groundwater from 1979 to 1999. As for the stable isotope data, irrigation could create heavier groundwater if increased evaporation fractionates irrigation water before it reinfiltrates the aquifer. From their plot of paired deuterium and oxygen isotope ratios in 1979 and 1999 [figure 1 in (*5*)], Aggarwal *et al.* conclude that "pumping since the mid-1970s did not significantly change stable oxygen and hydrogen isotope compositions..." However, simple statistical tests confirm that the 1999 waters do differ from the 1979 waters, in direct conflict with their inference. A *t* test on the mean residuals around the meteoric line (*6*) rejects the hypothesis that the two data sets derive from the same population ($P < 0.01$). Most of the 1979 data plot above the meteoric line, and most of the 1999 plot below. This shift in stable isotopes provides compelling evidence for the effects of irrigation pumping because three processes work to mute the change. First, crop transpiration, which accounts for much of the evaporation during irrigation, will not fractionate return water (*7*). Second, irrigation wells were sampled in 1979, clearly indicating that these data do not represent preirrigation conditions. Third, recharging irrigation water is greatly diluted by surface and monsoon waters.

Aggarwal *et al.* also argue that pumping does not affect flow by relying on a single groundwater hydrograph collected 40 km from our site on the other side of the Ganges River. However, 183 of the 252 groundwater hydrographs collected over the 10-year period (1988 to 1997) show trends of falling minimum water levels (*8*). According to the Bangladesh Ministry of Water Resources, "The impact of increasing groundwater development can often be clearly observed from hydrograph" (*9*). Furthermore, at Sreenagar, 4 km from our site, the water table minimum during the irrigation season dropped 1 m over the last 17 years ($P < 0.001$). The water table minimum is a far better indicator of pumping withdrawals than the mean and variance used in (*1*), which largely reflect rainfall and flood level during the wet season. However, even the minimum cannot be directly equated with flow below the water table. Evaporation in the flat topography of Bangladesh can cause water table decline by simply removing water from the near surface without driving groundwater flow. In contrast, pumping must drive flow, but the resulting water table decline may be substantially diminished by infiltration of irrigation return water. These hydrologic dynamics, compounded by exchange with ponds and rivers and rapid lateral flow induced by pumping, argue strongly for relying on direct measurement of pumping rates.

Van Geen *et al.* (*2*) overlook our hydraulic data and most of our chemical data to dispute the importance of pumping. First, they avoid the physical fact that pumping must affect groundwater flow. Hydraulic data in Fig. 1 provide an example at the arsenic peak. Second, they discount bomb levels of ^{14}C in dissolved inorganic carbon (DIC) at 19 m, despite the fact that at this depth arsenic levels sharply increase towards the peak. Tritium measurements support these radiocarbon measurements and show that a portion of the groundwater throughout the arsenic peak, to 61 m, must be young (*10*). Third, they base their argument on the false premise that the onset of pumping must precede the radiocarbon age of the bulk dissolved carbon for pumping to play a role in mobilization.

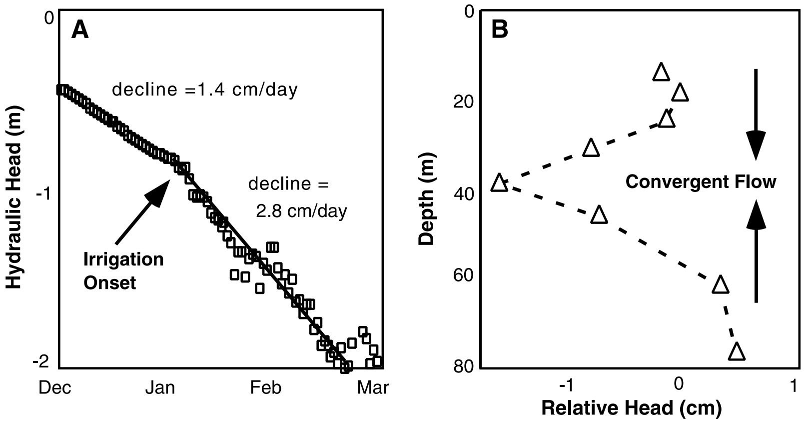

Fig. 1. (**A**) Hydraulic head at 32 m, the arsenic peak, 2001-2002. (**B**) Vertical head profile in March 2002 showing convergence at ~40 m.

Pumping may transport carbon, perhaps from nearby sediment, that can drive mobilization regardless of its bulk radiocarbon age. Pumping also transports young carbon from the surface and mixes it with old. Indeed, van Geen *et al*. do not object to our inferences that (i) pumping has altered groundwater flow; (ii) clearly, DIC has been transported by groundwater flow to the depth of the arsenic peak because the DIC radiocarbon age is much younger than the local sediment age, and (iii) this carbon was involved in arsenic mobilization. By accepting these inferences, van Geen *et al*. apparently contradict their claim that arsenic was mobilized in stagnant water unaffected by pumping.

In addition to neglecting the data and processes noted above, van Geen *et al*. disregard the accepted fact that dissolved carbon contains a mixture of radiocarbon ages as a necessary consequence of solute mixing and exchange with solid carbon. Five types of geochemical and hydrologic evidence indicate that beneath 19 m, a portion of the DIC and methane derives from recently produced organic carbon: (i) Tritium data show that recent recharge constitutes at least a portion of the groundwater to 61 m (*10*). Surface carbon transported by this recent recharge is likely also modern. (ii) Pumping rates are sufficient to have drawn water from the surface into the arsenic peak. (iii) The calcium peak, which very closely follows the arsenic peak, is likely the product of carbonate dissolution driven by production or inflow of DIC. Calcium concentrations indicate that 30% of the DIC is derived from carbonate. Assuming that this carbonate has a radiocarbon age 3000 years, the remaining 70% of DIC must be primarily modern, as demonstrated by the mixing line in figure 3 in (*4*). (iv) The DOC is much older than the DIC and methane in the same groundwater, so if DOC of this age has been converted, substantial young carbon must also have also been incorporated to obtain the observed bulk radiocarbon ages. (v) The convergence of groundwater from above and below the arsenic peak (Fig. 1B) accelerates the lateral flow within a streamtube passing through this depth, and likely mixes young carbon from above and old carbon from below. In essence, invading young carbon must mix with resident old carbon, yet van Geen *et al*. ignore this fact and present no hypothesis to reconcile their claim of stagnant groundwater in light of the measured DIC age being a fraction of the age of DOC and sediment.

Van Geen *et al*. argue against the importance of groundwater flow regionally by selectively focusing on only eight out of more than 50 samples from IAEA data, and five out of 49 unpublished values, where arsenic concentration is high but tritium is undetected. For these samples, they boldly claim to know arsenic concentrations decades before water was actually sampled, neglecting the possibility that solutes that interact with arsenic, or simply arsenic itself, have been transported in the old water. They then extrapolate their inference from these selected recent samples to generalize that arsenic concentrations have not changed throughout the whole country during the last 30 years of pumping.

We disagree with the contention that arsenic concentrations are immune to groundwater dynamics throughout Bangladesh. Data from our site point to the importance of transport and mixing due to flow and show no simple uniform relation between arsenic concentrations and water age. Arsenic levels are low at shallow depths where water is youngest and most affected by pumping, but are also low where water is oldest, deep in the aquifer. The highest concentrations occur where young and old water appear to mix, and tritium is still detected. Furthermore, mean arsenic levels in irrigation wells (120 ppb, $n = 23$), which receive the most rapid flow from the surface, is significantly ($P < 0.05$) lower than in domestic wells (200 ppb, $n = 32$). This is consistent with the hypothesis that, although flow to irrigation wells may help mobilize arsenic, pumping has begun to flush arsenic from streamtubes leading to these irrigation wells. In some areas, irrigation pumping may be flushing arsenic from the system or introducing oxidants that immobilize arsenic. The groundwater flow systems in Bangladesh are complex, and conditions vary. Natural topographic flow in some areas may control arsenic levels, as per Aggarwal *et al*., whereas in other stagnant areas (i.e., below clay layers), arsenic mobilized by detrital carbon (*11*) may remain constant, as per van Geen *et al*. However, data support neither of those two situations at our site.

Our interpretation that the band of high arsenic has been caused by an inflow of carbon should not be misinterpreted to mean that wherever wells are pumping, they are currently increasing arsenic concentrations. Such a misreading both oversimplifies and overextends our results. Likewise, our findings that arsenic levels are low at shallow depths most affected by irrigation, and lower in irrigation than household wells, should not be misconstrued to argue that irrigation pumping will remedy the problem across Bangladesh. We do, however, hope that our paper will help to dispel assumptions that groundwater arsenic concentrations are immune to changes in groundwater flow, recharge, and consequent chemical changes. Billions of dollars have been spent tracking the movement of groundwater contaminants under the premise that groundwater flow affects solute concentrations. We see no reason why Bangladesh would be different—ample evidence documents strong spatial gradients in dissolved arsenic and groundwater extraction guarantees groundwater flow.

C. F. Harvey
Ralph M. Parsons Laboratory
Department of Civil &
Environmental Engineering
Massachusetts Institute of Technology
Cambridge, MA 01239, USA
E-mail: charvey@mit.edu

C. Swartz*
Ralph M. Parsons Laboratory
Massachusetts Institute of Technology

A. B. M. Badruzzaman
Bangladesh University of Engineering and Technology
Dhaka 1000, Bangladesh

N. Keon-Blute
W. Yu
Ralph M. Parsons Laboratory
Massachusetts Institute of Technology

M. A. Ali
Bangladesh University of Engineering and Technology

J. Jay
Ralph M. Parsons Laboratory
Massachusetts Institute of Technology

R. Beckie
Department of Earth and Ocean Sciences
University of British Columbia
BC V6T 1Z1, Canada

V. Niedan
D. Brabander†

P. Oates
K. Ashfaque
Ralph M. Parsons Laboratory
Massachusetts Institute of Technology

S. Islam
Department of Civil and
Environmental Engineering
University of Cincinnati
Cincinnati, OH 45221, USA

H. Hemond
Ralph M. Parsons Laboratory
Massachusetts Institute of Technology

M. F. Ahmed
Bangladesh University of
Engineering and Technology

---

*Present address: Silent Spring Institute, Newton, MA 02458, USA. †Present address: Environmental, Coastal and Ocean Sciences Department, University of Massachusetts, Boston, MA 02125, USA

References and Notes

1. P. K. Aggarwal, A. R. Basu, K. M. Kulkarni, *Science* **300**, 584 (2003); www.sciencemag.org/cgi/content/full/300/5619/584b.
2. A. van Geen, Y. Zheng, M. Stute, K. M. Ahmed, *Science* **300**, 584 (2003); www.sciencemag.org/cgi/content/full/300/5619/584c.
3. P. K. Aggarwal *et al.*, "Isotope hydrology of groundwater in Bangladesh: Implications for Characteriza tion and Mitigation of Arsenic in Groundwater" (IAEA-TC Project Report: BGD/8/016, International Atomic Energy Agency, Vienna, 2002; www.iaea.org/programmes/ripc/ih/publications/bgd_report.pdf).
4. C. F. Harvey *et al.*, *Science* **298**, 1602 (2002).
5. A. R. Basu, S. B. Jacobsen, R. J. Poreda, C. B. Dowling, P. K. Aggarwal, *Science* **296**, 1563a (2002).
6. Data extracted from figure 1 in (*5*).
7. J. R. Gat, *Annu. Rev. Earth Planet. Sci.* **24**, 225 (1996).
8. Water Resources Planning Organization, Ministry Water Resources, People's Republic of Bangladesh, *National Water Management Plan* (Annex C, Appendix 9, 2000), vols. 1 to 11.
9. Water Resources Planning Organization, Ministry Water Resources, People's Republic of Bangladesh, *National Water Management Plan* (Annex C, Appendix 6, p. 52, 2000), vols. 1 to 11.
10. Tritium values were 6.5 TU at 19 m; 4.10 at 24 m; 0.42, 0.74, and 0.93 in three wells at 32 m; 0.20 at 38 m; 0.81 at 61 m; and 0.02 at 107 m, with measurement errors of approximately 0.1 TU. These values indicate that most of the groundwater at 19- and 24-m depths infiltrated in the past 50 years, and that from 24 m to 61 m groundwater contains a portion of recent recharge. Analysis performed by W. Aeschbach-Hertig and R. Kipfer, EAWAG and ETH, Zurich.
11. P. Ravenscroft *et al.*, in *Arsenic Exposure and Health Effects IV*, W. Chappell, C. Abernathey, R. Calderon, Eds. (Elsevier, Oxford, 2002).

23 January 2003; accepted 11 March 2003