Security of Deep Groundwater in the Coastal Bengal Basin Revealed by Tracers

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Abstract Uncertainty persists regarding the vulnerability of deep groundwater across Asia's megadeltas. In the coastal Bengal Basin aquifer system, shallow groundwater (<100 m) commonly features high salinity or arsenic concentrations, and deep, better-quality, groundwater supplies drinking water to >80 million people. Here we report new radiocarbon evidence from a network of nine dedicated, multilevel monitoring wells, which indicates residence times of between 10^3 and 10^4 years for groundwater at depths >150 m. Modern groundwater detected in some deep abstraction wells using anthropogenic tracers (SF_6, CFCs) is attributed to short circuiting of shallow groundwater within wells. Age-depth profiles and hydrochemical data in monitoring wells confirm the regional resilience of deep groundwater to ingress of shallow contaminated groundwater. Our results are consistent with high regional anisotropy in the aquifer and support continued use of deep groundwater though the potential for leakage of shallow contaminated groundwater in deep abstraction wells requires careful monitoring.

Plain Language Summary In the Bengal Basin of Bangladesh and India where shallow groundwater (<100 m below the land surface) commonly features high salinity or elevated arsenic concentrations, deep groundwater provides an invaluable source of drinking water to over 80 million people. Pressures on this resource are growing with rapidly increasing urban populations. Understanding how deeper groundwater (>100 m) is replenished and how vulnerable it is to the ingress of more saline and arsenic-rich groundwater from shallow and intermediate depths is critical to safeguarding public drinking water supplies. To date understanding of the vulnerability of deep potable groundwater here is largely based on modeling studies, with very few direct observations from samples taken from deep groundwaters. We assess the age and chemistry of deep groundwater in this region. Deep groundwater ages are between 1,000 and 10,000 years at monitoring sites and reflect long flow paths away from locations of modern replenishment. At a small proportion of wells, with a long history of pumping, there is evidence of a small amount of modern groundwater inflow to wells due to poor well seals but no evidence yet of salinity or arsenic ingress into the wider groundwater. Ongoing monitoring at deep sites needs to be prioritized in order to provide early warning of potential deterioration in the quality of deep groundwater.

1. Introduction

Deep groundwater in large deltaic aquifer systems across Asia provides a vital source of freshwater (Fendorf et al., 2010) and sustains drinking water supplies in saline coastal environments (MacDonald et al., 2016; Winkel et al., 2011). In coastal areas of West Bengal and Bangladesh, arsenic (As) contamination of the shallow (0–100 m) groundwater disadvantages over 80 million people (Ahmed et al., 2004; British Geological Survey/Department of Public Health Engineering, BGS/DPHE, 2001; Nickson et al., 1998). Groundwater is also saline at shallow-to-intermediate depths (typically 0–150 m) across much of the area (Figure 1b), making the deeper, fresh coastal groundwater (Figure 1) an invaluable source of water to meet growing demand in this region (Michael & Voss, 2008).

Regional-scale modeling (Burgess et al., 2010; Hoque et al., 2017; Michael & Voss, 2008, 2009b) and field studies (McArthur et al., 2016; Mihajlov et al., 2016; Mukherjee et al., 2007; Ravenscroft et al., 2013) have explored the vulnerability of the deep groundwater in the coastal Bengal Aquifer System (BAS) to contamination yet
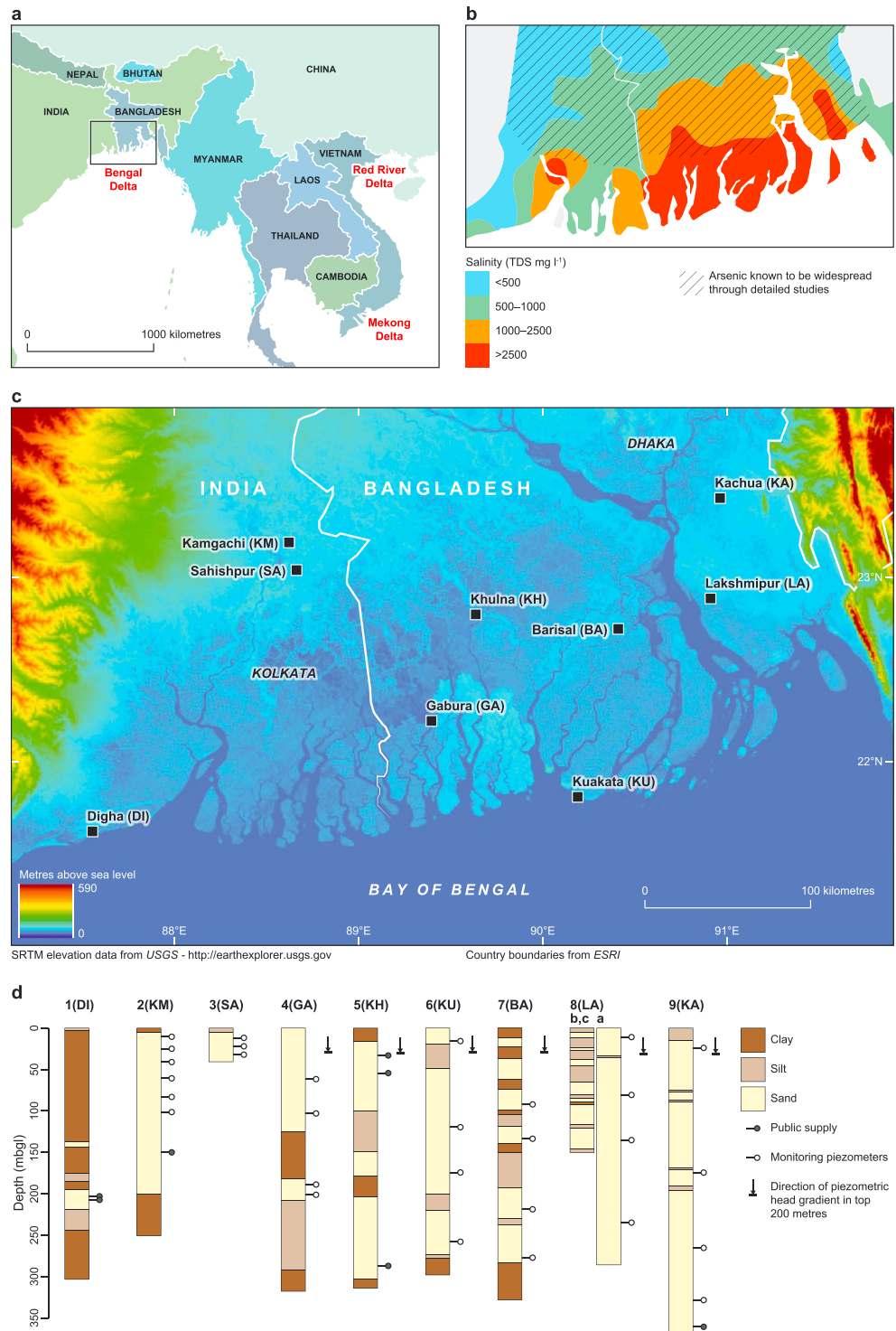
Figure 1. Location, salinity, elevation, and lithology of multilevel sites included in this study. (a) Location of the study area, Bengal delta, and other Asian megadeltas. (b) Salinity of the shallow groundwater resources (<200 mbgl) from MacDonald et al. (2016). (c) Map showing the location of the nine multilevel sample sites. The direction of piezometric head gradient is indicated (upward or downward) based on field observations at six sites, see supporting information for details, for site 5 (Khulna) piezometric data from BWDB was used (BWDB, 2013a). (d) Depth variations in simplified lithology and total depths for piezometers and public supply sites: DI (Public Health Engineering, 2016); SA (Mukherjee et al., 2011); KM (Neidhardt et al., 2013); GA-BA, LAa (BWDB, 2013b); LAb-c (BGS/DPHE, 2001); KA (Hoque & Burgess, 2012).

key knowledge gaps remain. These include the relative importance of site-specific, borehole- scale problems due to casing breaks at abstraction well sites (I. Choudhury et al., 2016) and aquifer-scale vulnerabilities (Hoque et al., 2017) to contamination from shallow groundwater and the long-term impacts of intensive pumping at depth (Khan et al., 2016; Knappett et al., 2016; Michael & Khan, 2016; Zahid et al., 2014). The presence of modern tracers (e.g., tritium and chlorofluorocarbons (CFCs)) in deeper groundwater systems (typically >150 m) has been attributed to deep pumping in the BAS and in many other aquifer systems globally (Jasechko et al., 2017; Lapworth et al., 2015; McMahon et al., 2013; Samborska et al., 2013) yet considerable uncertainty remains about the scale and nature of this pumping-induced contamination. Here we present new evidence from multiple tracers drawn from dedicated, depth-specific piezometers and actively pumped wells in the BAS, the world's largest deltaic aquifer system, to assess pumping-induced recharge processes within the deep fresh BAS.

The most practical solution to accessing As-safe and low salinity drinking water in the BAS remains exploitation of regionally extensive, deep groundwater (Benneyworth et al., 2016; Mukherjee, 2018; Ravenscroft et al., 2009; Zahid & Ahmed, 2006). Modeling (University College London, 2013) suggests that, on timescales of 50 to 100 years, the benefits of groundwater withdrawal outweigh the potential risks of contaminant breakthrough. Ravenscroft et al. (2013) showed evidence from rare, time series observations of the hydrochemistry of the BAS from 1998 to 2011, which suggest that current groundwater withdrawals are sustainable over decadal timescales. In contrast, caution has been expressed that the BAS is a finite and potentially vulnerable resource and, as such, exploitation should be restricted (Burgess et al., 2002; Burgess et al., 2010; Michael & Voss, 2008). Both approaches emphasize the necessity of rigorous monitoring.

Current knowledge of deep groundwater in the BAS is dominated by modeling studies (Burgess et al., 2010; Khan et al., 2016; Michael & Voss, 2008, 2009a; Zahid et al., 2014) and is inadequately constrained by field observations. The deep coastal BAS has been exploited for over 30 years and, in some urban areas such as Khulna, Lakshmipur, and Digha (Figure 1), there are some reports of increased salinity impairing drinking water supplies (K. Choudhury & Saha, 2004; IWM, 2011; Ravenscroft et al., 2009; Zahid & Ahmed, 2006). Mihajlov et al. (2016) showed little evidence of mixing from handpumps; however, the impact from larger production wells is poorly constrained.

Here we apply a range of groundwater residence time tracers including carbon-14 (^{14}C-DIC) and anthropogenic gases, for example, CFCs and sulfur hexafluoride (SF_6) to (1) characterize age-depth profiles within the top 360 m of the BAS; and (2) assess evidence of the vulnerability of deep groundwater to ingress from contaminated groundwater at shallow and intermediate depths. Samples were taken across the lower-deltaic and marine-influenced region of the BAS from both dedicated multilevel monitoring piezometers at nine locations, and pumped public water supply boreholes at four locations (Figure 1). Published groundwater ^{14}C-DIC, δ^{13}C-DIC, δ^{18}O, and δ^2H for the BAS were compiled for comparison ($n = 57$) and include six piezometer profiles, two of which extend into the deep BAS (Aggarwal et al., 2000; BGS/DPHE, 2001; Majumder et al., 2011; Zheng et al., 2005). A large proportion of data drawn from previous studies derives from abstraction wells with wide uncased intervals. Here we employ newly installed, dedicated multilevel piezometer installations with single, short screens (range of 1–10 m) to characterize vertical variations in hydrochemistry throughout the coastal BAS to a depth of 360 m. For the first time, we are able to compare new and historical residence time results from dedicated monitoring sites to those collected from pumped municipal sites (pumped at 20 L/s for large abstractions, and typically cased to ~200 mbgl) and are thereby able to characterize the regional vulnerability of the deep BAS to contamination from shallow groundwater.

2. Materials and Methods

2.1. Sampling Locations and Rationale

A total of nine multilevel sampling locations were selected across the coastal Bengal Aquifer System to provide geographical coverage of the region and sites of contrasting deep pumping history. At three locations (Khulna, Lakshmipur, and Digha) there has been decades of intensive pumping for drinking water. Downward hydraulic gradients between shallow and deep piezometers for Khulna and Lakshmipur (3 m and 5 m head difference, respectively, see supporting information) are comparable to downward gradients reported close to Dhaka (2–4.5 m) by Knappett et al. (2016). With the exception of site 1 Digha (DI) and

site 5 Khulna (KH), all sampled installations are purpose-built, multilevel monitoring wells and included sites used in previous hydrogeological studies [sites 3 Sahishpur (SA), 2 Kamgachi (KM), and 9 Kachua (KA) and four newly constructed multilevel monitoring stations (sites 4 Gabura (GA), 6 Kuakata (KU), 7 Barisal (BA), and 8 Lakshmipur (LA)] (BWDB, 2013a, 2013b). Details on the compilation of the lithology profiles for the nine sites are provided in the supporting information. Locations covered a range of pumping histories from the BAS and contrasting hydrogeologeat settings (see Figure 1). A summary of the hydrogeological setting is provided in the supporting information.

2.2. Environmental Tracers, Sampling, and Analysis

Sampling was undertaken at all nine locations for groundwater residence time tracers (CFCs, SF_6, ^{14}C-DIC) as well as stable isotopes ($\delta\,^{13}C$, $\delta\,^2H$, and $\delta\,^{18}O$) and aqueous hydrochemistry. All sampling was undertaken after pumping of at least three standing borehole volumes and monitoring for field chemistry parameters until stable results for dissolved oxygen (DO), specific electrical conductivity (SEC), and pH were obtained. Field alkalinity was determined by titration (duplicate) against sulfuric acid using a bromocresol green indicator. Sites were purged with a suction pump and sampling was undertaken immediately after with a peristaltic pump. All field measurements for DO, pH, and redox potential were carried out in a sealed flow cell. Field probes were calibrated daily. Samples for CFCs (125 mL) and SF_6 (500 mL) were taken by filling airtight glass sample bottles under water and using a jacket of water to exclude all air contamination of samples during sampling. 1 L ^{14}C-DIC samples were taken in Nalgene™ sample bottles, which were completely filled after being rinsed twice with sample water. Stable isotope samples ($\delta^{13}C$, $\delta\,^2H$, and $\delta\,^{18}O$) were stored (unfiltered) in 30 mL Nalgene™ bottles. Filtered samples (0.45 micron) for total dissolved anions (60 mL) and cations (30 mL) were taken and stored in Nalgene™ bottles. Cation samples were preserved with ultrapure Aristar™ grade HNO_3, whereas anion samples were filled to the top of the bottle to minimize air contact. DOC samples were filtered using 0.45 micron silver filters. Samples for arsenic speciation were taken at sites where SEC was <2,000 μS/cm and As speciation was stabilized in the field using ion exchange methods as per Watts et al. (2010).

Measurement of anions, cations, DOC, stable isotopes, and SF_6 and CFCs took place at British Geological Survey laboratories in the UK. Stable isotope analyses (δ^2H and $\delta\,^{18}O$) were carried out using standard preparation techniques followed by isotope ratio measurement on a VG-Micromass Optima mass spectrometer. Data considered in this paper are expressed in parts per-mil (‰) with respect to Vienna Standard Mean Ocean Water (V-SMOW). The $\delta^{13}C$ results were calibrated against Vienna Peedee Belemnite (VPDB) using the international standards NBS-19, IAEACO-8, and IAEA-CO-9. CFCs and SF_6 were measured by gas chromatography with an electron capture detector after preconcentration by cryogenic methods (Busenberg & Plummer, 1992, 2008). Measurement precision was within ±0.1‰ for $\delta^{18}O$, ±1‰ for δ^2H, and ± 5% for the CFC; overall method detection limits, including analytical and sampling errors, are 0.05 pmol/L for CFC-12, 0.1 pmol/L for CFC-11 and 0.1 fmol/L for SF_6. ^{14}C-DIC was analyzed by the NERC Radiocarbon Facility, East Kilbride. The samples were prepared by reduction with graphite at the NERC Radiocarbon Facility-East Kilbride and passed to the SUERC AMS Laboratory for ^{14}C analysis. Cations were analyzed by ICP-MS, anions by ion chromatography, and DOC using a Shimadzu TOC analyzer following acid sparging of DIC.

For $\delta^{13}C$, $\delta\,^{18}O$, and $\delta\,^2H$ repeat analysis on random samples gave 1σ of between 0.01‰ and 0.07‰, 0.2‰–0.4‰, and 0.05‰–0.1‰ respectively. The aquifer sediments of the BAS are dominated by siliceous material, fluvial in origin and are free from carbonates, therefore precluding the need for correction for aquifer-derived reservoir DIC (e.g., Pearson, 1965). Full details on well-established methods used to estimate groundwater ages in this region which account for dilution of ^{14}C-DIC by reference to $\delta^{13}C$ values (e.g., Hoque and Burgess, 2012) are provided in the supporting information. Mixing with bomb enriched radio carbon could potentially lead to an underestimation of age; however, results from the monitoring sites sampled here showed no evidence of this.

DOC, anion, and cation results were validated using independent NIST standards and Aquacheck™ samples and internal standards; all methods used are UKAS accredited. Two field procedural blanks for anions, cation, and DOC analysis were collected to validate the field procedures (results in the supporting information). Field As speciation results were cross-checked with total dissolved As results from an independent sample and found to be 100% ± 15% where total As concentrations were >0.4 μg/L.

3. Results

3.1. Hydrochemical Depth Profiles

Hydrochemical depth profiles (Figure 2) are used to explore the dual threats to deep groundwater posed by arsenic at shallow and salinity at shallow-intermediate depths, respectively, within the coastal BAS. Depth variations in As, specific electrical conductivity (SEC), dissolved organic carbon (DOC), total dissolved phosphorus (P), boron (B), sulfate (SO_4), and redox potential (Eh) are presented in Figure 2. At shallow depths (<50 m) the BAS is highly contaminated with As and has a low SEC (median of 67 µg/L and 910 µS/cm, respectively). At intermediate depths (50–150 m) As concentrations are generally lower; however, high salinity is prevalent (median As of 17 µg/L and SEC 7,300 µS/cm). Groundwater within the deep BAS (>200 m) typically has low As concentrations (<10 µg/L) and lower salinity (median 1200 µS/cm). One exception is Digha, which is very close to the coast and has elevated salinity at depth at two sites. DOC concentrations decrease with depth from a median of 1.9 mg/L in the shallow BAS to 0.8 mg/L in the intermediate and 1.0 mg/L in the deep BAS; boron depth profiles follow SEC trends. Dissolved oxygen and redox potential (Eh) show an overall decreasing trend with depth, with Eh values between 0 and +150 mV at depths >150 m in the BAS.

Inorganic As III and V dominate arsenic speciation in the BAS where As III is commonly reported as the most abundant form of As under reducing conditions (BGS/DPHE, 2001). We show that As III typically accounts for >90% dissolved As. Organic compounds (monomethylarsonic acid, MMA and dimethylarsinic acid, DMA) are found in concentrations of up to 10 µg/L in the shallow aquifer and are equivalent to concentrations of As V throughout the profile (Figure S2). Below 200 m, groundwater quality generally complies with World Health Organization (WHO) drinking water guidelines (World Health Organization, 2011) for the analytes reported with the exception of individual samples at four sites (5-Khulna, 6-Kuakata, 7-Barisal, and 9-Kachua) where total dissolved solids (TDS), B and As exceed WHO guidance values. Groundwater As concentrations of >1 µg/L below 200 mbgl are only observed at two sites in this study (6-Kuakata and 7-Barisal, 10.7 and 9.5 µg/L, respectively, Figure 2).

3.2. Groundwater Residence Time Tracers and Stable Isotopes

A suite of residence time tracers was used to assess age-depth profiles across the monitoring network and identify modern groundwater at depth within the coastal BAS. Radiocarbon ^{14}C-DIC activity and estimated groundwater ages, corrected using δ^{13}C-DIC (Hoque & Burgess, 2012), in five new depth profiles across the BAS as well as published piezometer profiles ($n = 8$), together with results from public supply abstraction wells, are presented in Figure 3. An overall decrease in ^{14}C activity, corresponding to an increase in age, is shown with depth between 100 and 250 mbgl; samples from pumped sites show consistently higher ^{14}C activity compared to samples from monitoring piezometers.

Modern tracer results from nine profiles for anthropogenic tracers (CFC-12, CFC-11, and SF_6) are presented in Figure 4. These tracers are detected to depths of 360 m in some abstraction wells and down to 100 m in monitoring piezometers. An overall reasonable agreement is observed between CFC-12 and CFC-11 results, $R = 0.76$ (Figure 4b).

Stable isotope ratios of C, O, and H (δ^{13}C-DIC, δ^{18}O, and δ^2H) in groundwater from this study and other published studies in the coastal BAS are presented in Figure S3. Stable isotope ratios show no systematic variation with depth and are consistent with values found in previous studies, including those from pumped municipal supply sites (see Figures S3a and S3c in the supporting information). Overall, there is a highly variable trend of enriched δ^{18}O with depth, with more variability found between 0–200 m at dedicated monitoring piezometers compared to abstraction wells (see Figure S3a). Groundwater stable isotope ratios (δ^{18}O versus δ^2H) plot alongside local and global meteoric water lines (Figure S3d).

4. Discussion

4.1. Hydrochemical Context to Deep Groundwater Vulnerability

Hydrochemical depth profiles provide the context for evaluating the vulnerability of deep groundwater to ingress of shallow contaminated groundwater. High arsenic is restricted to the shallow groundwater (<100 mbgl, see Figure 2) where higher DOC concentrations are found and redox conditions control the dissolution of arsenic bound to iron-oxyhydroxides (BGS/DPHE, 2001; Nickson et al., 2000; Smedley & Kinniburgh, 2002; van Geen et al., 2003). Evidence from small-scale studies (Harvey et al., 2002; Neumann
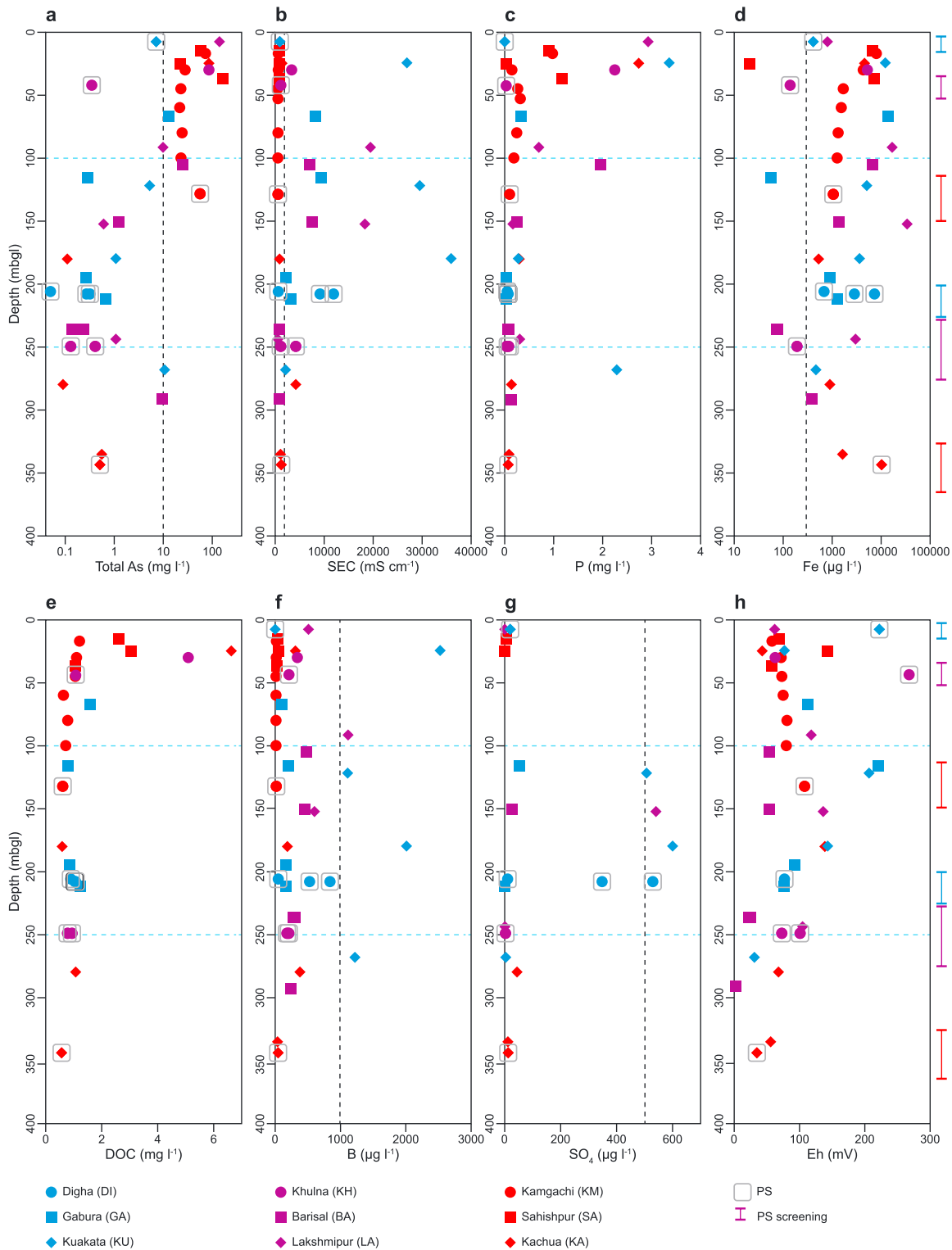
Figure 2. Dissolved hydrochemistry depth profiles from nine sites in the coastal aquifer system. (a) Total arsenic (μg/L), log scale for *x* axis. (b) Field specific electrical conductance (SEC) (μS/cm). (c) Phosphorus (mg/L). (d) Iron (Fe; μg/L). (e) Organic carbon (mg/L). (**f**) Boron (μg/L). (g) Sulfate (as SO_4 mg/L). (h) Redox potential (mV). All piezometer screens are 10-m long. PS denotes pumped public supply abstraction wells. PS well screening intervals shown on the right-hand side of Figures 2d and 2h, well screening range for DI = 200–220 m, KU = 2–10 m, KH = 35–55 m, KM = 115–150 m, KA = 326–365 m, and 220–280 m, mid screen point used for plots. Vertical dashed lines show World Health Organization drinking water guideline values (WHO, 2011), for SEC a drinking water guideline value of 1,500 μS/cm is used based on a total dissolved solids guideline of 1,000 mg/L and a conversion factor of 0.67. Data from the nine sites shown in Figure 1 are included in this plot. Horizontal lines show depth ranges for shallow, intermediate, and deep groundwater.
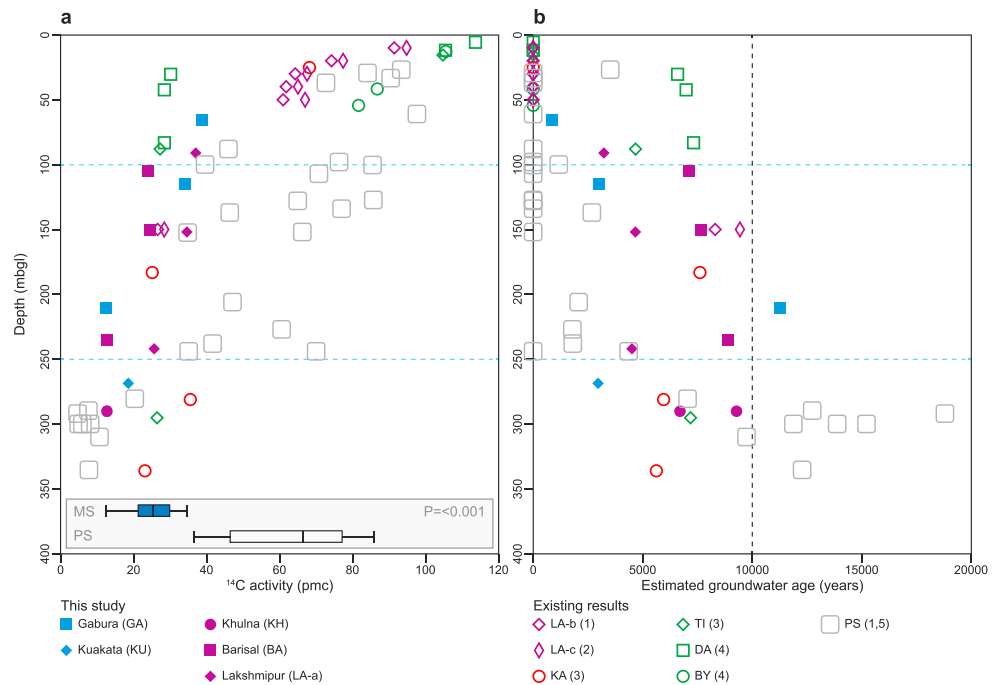
Figure 3. Depth variation of ^{14}C-DIC activity (pmc) and estimated groundwater age within the coastal aquifer system. (a) ^{14}C-DIC activity percent modern carbon (pmc) for data from this study and other published studies in coastal BAS. Published data sources: (1) Aggarwal et al. (2000); (2) BGS/DPHE (2001); (3) Hoque & Burgess (2012); (4) Zheng et al. (2005); (5) Majumder et al. (2011). Box plot of ^{14}C activity (pmc) in the 100–250 depth interval for MS and PS, P value from a t test for difference in median values shown ($P = <0.001$). (b) For all samples estimated groundwater age calculated by reference to the δ^{13}C-DIC values of samples (Hoque & Burgess, 2012). Profile data from monitoring piezometer sites: LA = Lakshmipur; BA = Barisal; KH = Khulna; GA = Gabura; KA = Kachua; TI = Titus; DA = Dari; BY = Baylakandi. All piezometer screens are 10-m long. PS denotes sites with published data from public supply abstraction wells, MS denotes data from monitoring piezometer sites.

et al., 2010) suggest that surface inputs of organic matter, induced by pumping, can lead to As mobilization in shallow aquifers (<100 m). More recent work by Shamsudduha et al. (2015) challenges this suggestion as a regional-scale phenomenon and shows, using statistical models from national data sets, that groundwater irrigation serves to flush As from the shallow groundwater system and redistribute it to the surface. Elevated salinity and associated high Cl, B, and SO_4 (Figure 2) are found at shallow (<100 m) to intermediate (100–200 m) depths at coastal sites and pose a potential threat to underlying fresh groundwater regionally in the coastal BAS. The source of high Cl groundwater was assessed using Cl:Br ratios. Cl:Br depth profiles indicate marine sources dominate profiles (e.g., profile Figure S1) but also provide evidence of wastewater Cl input at selected shallow sites (0–30 mbgl) and most notably at Sahishpur (SA) and Kamgachi (KA) in West Bengal, India (Katz et al., 2011; McArthur et al., 2012). There is no clear evidence of endogenous Cl inputs in this study, or indeed additional Cl inputs from fertilizer within the shallow aquifer (Alcalá & Custodio, 2008).

4.2. Groundwater Residence Time Depth Profiles in the Coastal BAS

Estimated ^{14}C-DIC ages and age-depth profiles indicate a trend of increasing age with depth (Figure 3). For monitoring piezometers, the oldest groundwater (11.2 ± 0.1 ka) is observed at Gabura (site 4, Figure 1) at a depth of 212 mbgl. Greater groundwater ages ranging from 12 to 18.8 ka were reported from municipal sites >300 mbgl in the Barisal division of Bangladesh from studies carried out in 2000 (Aggarwal et al., 2000) and 2006/2007 (Majumder et al., 2011). Sampling from dedicated multilevel piezometers in Barisal (site 7, Figure 1) give ages of between 7 and 9 ka for sites between 100 and 250 mbgl. Shallow groundwater (<100 mbgl) from dedicated piezometers has an age of less than 3 ka in our study, although published ages at other sites (e.g., Dari, Figure 3) are between 5 and 7 ka (Zheng et al., 2005).
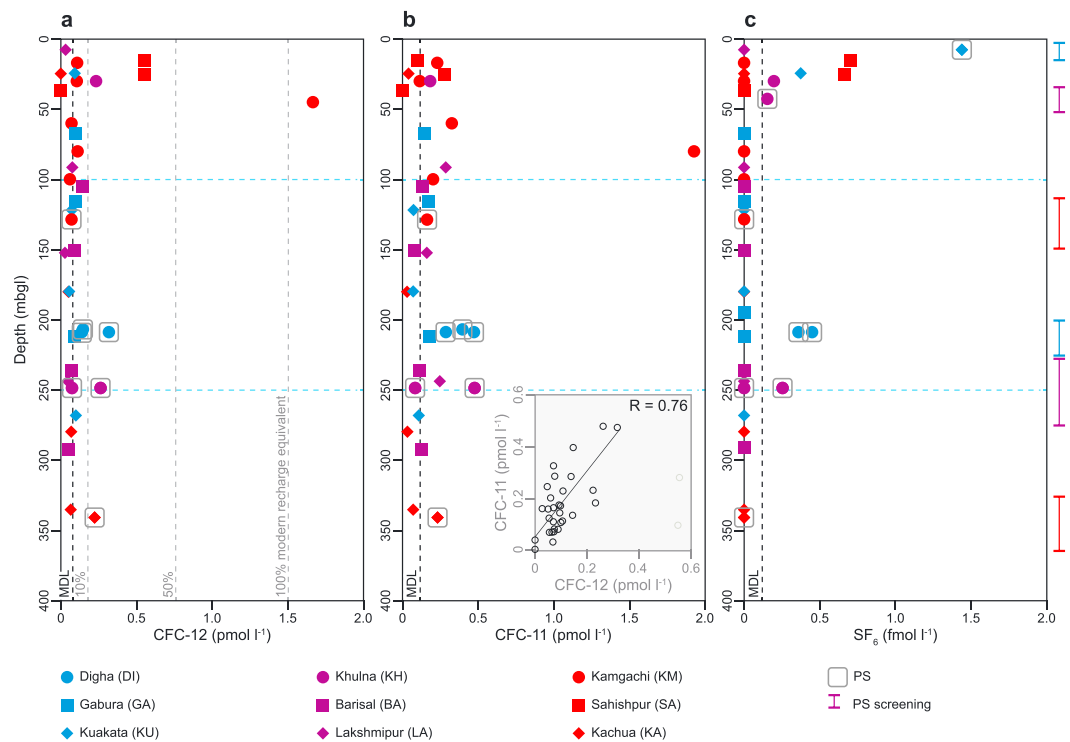
Figure 4. Chlorofluorocarbon (CFC-11 and CFC-12) and sulfur hexafluoride depth profiles for nine sites in the coastal BAS. (a) Chlorofluorocarbon-12 (CFC-12) depth profiles from nine sites across the coastal BAS. (b) Chlorofluorocarbon-11 (CFC-11) depth profiles from nine sites across the BAS, inlay shows a cross-plot of CFC-12 versus CFC-11 and shows good agreement for these two tracers, $R = 0.76$ for results 0–0.5 pmol/L excluding two outliers, gray symbols. (c) Sulfur hexa-fluoride (SF_6) depth profiles from nine sites across the coastal BAS. Percent modern atmospheric equivalent shown for comparison. All piezometer screens are 10-m long, PS denotes public supply abstraction wells, PS screening intervals shown on Figure 4c. Solid black vertical lines show method detection limits (MDL) for each tracer. See Figure 1 for site IDs and locations.

Samples from abstraction wells in general exhibit greater ^{14}C activity and thus younger bulk groundwater ages than samples from dedicated monitoring wells throughout much of the profile in Figures 3a and 3b. Indeed, a comparison of ^{14}C activities between pumped and monitoring sites, within the 100–250 mbgl depth interval, reveals greater activities at pumped sites (mean of 66 pmc and 33 pmc, respectively) and a significant difference was found between these groups ($P < 0.001$, rank sum test, Figure 3a). In contrast, Mihajlov et al. (2016) found no evidence for a significant contribution of recent recharge from handpump sites >150 mbgl in Araihaza. Screen intake lengths. Pumping rate and induced vertical fluxes are much greater in the case of public water supply wells, promoting the mixture of older less-enriched groundwater with more modern recent recharge containing ^{14}C including from thermonuclear testing (Fontes, 1992; Geyh, 2000).

Stable isotope ratios ($\delta^{18}O$ versus $\delta^{2}H$) for groundwater samples at all depths suggest that there has been lim-ited modification due to evaporation (Figure S3d) and that paleowaters have been recharged within the pre-vailing Indian monsoon regime ($\delta^{18}O$ fall between −7‰ and −2‰). This finding is consistent with other recent studies (Desbarats et al., 2014; Majumder et al., 2011; Mihajlov et al., 2016). Consistency in $\delta^{13}C$ values with depth and [14]C ($\delta^{13}C$ range between −20‰ and −5‰) also may suggest a relatively uniform source of DIC within the groundwater system over the last 20 ka (Aggarwal et al., 2004).

In this study CFC-11 and CFC-12 serve as semiquantitative tracers to constrain modern components of groundwater within a mixed composition. Direct comparisons with the conservative tracer SF_6 show similar depth profiles (Figure 4), demonstrating for the first time the effective use of SF_6 as a tracer of modern groundwater in this setting. Furthermore, a comparison of SF_6 with CFC data suggest that the overall impact of CFC degradation was small in this study.

At three public water supply wells (1-Digha, 5-Khulna, and 9-Kachua) within the BAS, anthropogenic gases are detected in concentrations that suggest up to 15% modern equivalent for CFC-12 (Figure 4a). This determination is confirmed independently by CFC-11 and SF_6 results. Together, these observations strongly imply vertical leakage of shallow modern groundwater has occurred in half of these public abstraction wells; these modern tracers are not, however, detected in the remaining sites. The results from the modern tracers support the ^{14}C results (Figure 3) and indicate a component of mixing with younger modern water in some deep pumped wells. Indeed, if this is the case, then the estimated ages for ^{14}C-DIC are a minimum since post 1950s recharge contains greater that 100% pmc.

4.3. Implications for Future Development and Monitoring of the Deep BAS

Use of deep groundwater for irrigation in most of the coastal BAS is limited and has declined since the early 1990s in Bangladesh but not in some parts of West Bengal, India (McArthur et al., 2016). Remote from cities, shallow groundwater levels over the last 20 years in the BAS have remained constant or risen (0.4 to 2.5 cm/a), commensurate with rising sea levels (Shamsudduha et al., 2009). Piezometric heads from multilevel monitoring sites presented here (Figure 1d) reveal downward hydraulic gradients within the uppermost 200 m below ground level (mbgl) of the BAS whereas deeper within the BAS >200 mbgl upward hydraulic gradients dominate (Burgess et al., 2017), see supporting information.

Deep groundwater in the coastal BAS has been exploited for drinking water supplies for several decades and will continue to be developed to sustain increasing demand for municipal, domestic, and industrial water in this region. Here we show that the majority of groundwaters at shallow depths (0–100 mbgl) have elevated arsenic concentrations (>50 µg/L) or high salinity at intermediate depths (100–250 mbgl). Due to the greater depth of the high-salinity groundwater and, in contrast to dissolved arsenic, the lack of sorption, breakthrough of salinity, where present, to deep pumping wells is a more immediate potential threat to the deeper groundwater and hence to drinking water quality in the coastal BAS than arsenic breakthrough (Figure 2). Where higher As concentrations are found in deep pumping wells in this study (e.g., Barisal and Kuakata, Figure 2) residence time indicators (Figure 3) and hydrochemical evidence (Figure 2) indicate that they are unlikely to be associated with the vertical migration of shallow modern water but may represent zones where there have been lower rates of groundwater flushing due to local variations in permeability (Cardenas & Jiang, 2010; Gassiat et al., 2013; Hoque et al., 2017). Indeed, this has been observed elsewhere in the shallow BAS (Shamsudduha et al., 2015).

Modern tracers were not detected at depth in monitoring piezometers but only at some pumped wells where a component of younger groundwater is detected. Results from pumped wells between 100 and 250 mbgl have significantly enriched ^{14}C values compared to monitoring piezometers, which is consistent with the evidence of modern residence time tracer results (Figure 4). The detection of modern tracers at some deep abstraction wells raises concerns about the vulnerability of these sites from shallow groundwater. The high salinity present at intermediate depths (Figure 2) rules out vertical migration of shallow groundwater within the aquifer system and strongly suggests that these observations reflect short circuiting of shallow groundwater within pumping wells rather than vertical mixing within the local or regional aquifer system (I. Choudhury et al., 2016).

Overall, within the top 100 m there is a sharp age-depth gradient followed by a gradual increase in age with depth between 100 and 300 mbgl, which is consistent with other recent studies (Hoque & Burgess, 2012; Mihajlov et al., 2016). There is one groundwater sample that gives age estimates exceeding 10 ka throughout the 0–360 m profile (Figure 3). Two groundwater age profiles obtained circa 15 years ago for Lakshmipur (Aggarwal et al., 2000; BGS/DPHE, 2001) are comparable (Figure 3). Data from monitoring piezometers show that estimated age-depth profiles vary considerably between sites, but there is no evidence of significant regional scale vertical mixing due to pumping. For example, at two sites in Khulna, both of which have been pumped from a depth of 290 m since 1980, only one showed evidence of CFC-12 contamination (Figure 4) and also had more enriched ^{14}C values (Figure 3). Further, there is no evidence of salinity breakthrough from the overlying saline aquifer system. Therefore, the modern water at depth is most likely explained by bypass flow via the well annulus from inadequate borehole construction. The multiple aquitard layers at Khulna (see Figure 1) have been shown to be locally continuous (Ravenscroft et al., 2009, and references therein) and as such are likely to provide a high level of protection to deep groundwater sources in this area.

5. Conclusions

Hydrochemical data from a network of multilevel monitoring wells in the coastal Bengal Aquifer System confirm the occurrence of both high concentrations of arsenic at depths from 0 to 150 mbgl and high salinity between 30 and 250 mbgl. Radiocarbon evidence from a network of multilevel monitoring wells in the coastal Bengal Aquifer System shows residence times of between 10^3 and 10^4 years for groundwater at depths >150 m. Anthropogenic tracers (SF_6, CFCs) of modern groundwater, detected in some deep abstraction wells, are attributed to short circuiting of shallow groundwater within the annulus of the well. It is therefore important to consider the vulnerability of deep groundwater arising from inadequately sealed boreholes, which is not observed at dedicated monitoring piezometers. The regional security of deep groundwater within the coastal Bengal Aquifer System from the ingress of shallow contaminated groundwater is confirmed by age-depth profiles. Our results are consistent with high regional anisotropy in permeability providing protection to deep groundwater and limiting the potential for vertical mixing. This conclusion supports the case for continued but carefully monitored development of deep groundwater. Given the importance of this resource, and expected future demand, monitoring needs to capture both short circuiting of vertical leakage, which may manifest itself over relatively short timescales in pumped sites, as well as local aquifer-scale pathways which may take much longer (many decades) to be observed at monitoring wells.

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