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Tracing recharge to aquifers beneath an Asian megacity with Cl/Br and stable isotopes: the example of Dhaka, Bangladesh

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Abstract

Dhaka, the capital of Bangladesh, is home to a population of 15 million people, whose water supply is 85% drawn from groundwater in aquifers that underlie the city. Values of Cl/Br >500 are common in groundwater beneath western Dhaka in areas <3 km from the river, and in rivers and sewers around and within the city. The study shows that groundwater beneath western Dhaka is strongly influenced by infiltration of effluent from leaking sewers and unsewered sanitation, and by river-bank infiltration from the Turag-Buriganga river system which bounds the western limit of the city. River-bank infiltration from other rivers around Dhaka is minor. Values of Cl/Br and Cl concentrations reveal that 23% of wells sampled in Dhaka are influenced by saline connate water in amounts up to 1%. This residual natural salinity compromises the use of electrical conductivity of groundwater as a method for defining pathways of recharge by contaminated surface waters. Concentrations of As, B, Ba, Cd, Cu, F, Ni, NO_3, Pb, Sb, Se and U in groundwater samples are less than WHO health-based guideline values for drinking water.

Keywords: Bangladesh, Cl/Br, chloride, urban groundwater, stable isotopes

1. Introduction

Groundwater is ubiquitous in the sediments of the world's deltas, and most cities built on deltas tap that water for urban supply. Increasing populations and their increasing demand for water have led to increasing abstraction by such cities (Falkenmark and Widstrand, 1992, Foster, et al., 2011, Lundqvist, et al., 2003, McDonald, et al., 2011). As a consequence, over-exploitation of associated aquifers is common, leading to declining water levels and productivity, land subsidence, and a deterioration in water quality (Galloway and Burbey, 2011, Hoque, et al., 2007, Hosono, et al., 2011, Kagabu, et al., 2011, McDonald, et al., 2011, Morris, et al., 2003). Development continues despite recent discoveries of widespread pollution by arsenic (As) in the shallow aquifers of such deltas, particularly in SE Asia (Ravenscroft, et al., 2009).

Abstraction for city supply may induce recharge from nearby rivers; river-bank infiltration (Farnsworth and Hering, 2011) is expolited deliberately in some localities and may occur unwittingly in others (Hosono, et al., 2011). Where such infiltration has defined pathways that are monitored for water quality, the exploitation provides convenience, cleansing, and the ability to abstract groundwater at rates that exceed local rates of recharge by rainfall (Bourg and Bertin, 1993, Gunten and Kull, 1986, Schubert, 2002). Where river-bank infiltration is not effectively monitored, infiltration from contaminated and/or polluted rivers may adversely affect public water supply. Such is the case in many of the world's river-bank cities.

The city of Dhaka is surrounded by rivers (Fig. 1) that flow along fault-controlled lines (Hoque, et al., 1994, Morgan and McIntire, 1959). River-bank infiltration has therefore long been suspected to be an important source of recharge to the aquifers beneath Dhaka (Ahmed, et al., 1999). Using Cl/Br mass ratios, supplemented by stable isotope data and concentrations in groundwater of Cl, NO_3, and SO_4, this work confirms that such recharge occurs in westernmost Dhaka, but also shows that contaminated groundwater from leaking sewers has an equal impact on groundwater quality beneath that part of the city, and that groundwater elsewhere beneath Dhaka City has been little influenced by river recharge or human activity.

2. Study Area

2.1. Physiography

About 40% of Dhaka lies on the southern margin of an uplifted Pleistocene block termed the Madhupur Tract, and has elevations up to 15 m above sea level. These elevated regions are bounded by faults and lineaments (Hoque, et al., 1994). The Tract comprises the Madhupur Clay, a Pleistocene residuum, that is between 5 and 50 m thick (Hoque, et al., 2007, Rahman, et al., 2013) and overlies sands of the Dupi Tila Formation. The Madhupur Clay is thought to thin westward and allow the underlying Dupi Tila Formation to crop out along the bed of the Buriganga River that bounds the western side of Dhaka (Hasan, 1999). The remaining 60% of the city lies on Recent alluvium that occupies abandoned ancient or Recent river channels, and sediments of adjacent floodplains. This part has elevations no more that 1.5 to 3.5 meters above mean sea-level and is prone to flood in the monsoon season (Huq and Alam, 2003).

2.2. Hydrostratigraphy

Groundwater for Dhaka abstracted by water authorities comes from aquifers mostly > 100 m deep. Wells are screened in sands of the Plio-Pleistocene Dupi Tila Formation that underlies the Madhupur Clay or Recent alluvial deposits, or both. The Dupi Tila Formation sands contain laterally discontinuous silts and clays, which are common between 160 and 190 m depth (Fig. 1b). It is commonly assumed that these fine-grained units divide the aquifers into an Upper Dupi Tila Aquifer (UDTA) and a Lower Dupi Tila Aquifer (LDTA) (Ahmed, et al. 2011, DWASA and IWM 2008, Haque 2006, Hoque 2004, Rahman, et al. 2013). The UDTA comprises brown sands that were oxidised by flushing groundwater during low-stands of sea level (DPHE, 1999). These Pleistocene brown sands contain almost no organic matter (e.g., McArthur, et al., 2004), a fact that retards chemical evolution of the aquifer towards the suboxic/anoxic state. The LDTA comprises grey sands that remained below the depth of low-stand flushing and are less oxidised. Lateral discontinuity in clays and silt intercalations in the Dupi Tila Formation allows hydraulic connection between the UDTA and LDTA in some areas. A hydrogeological section constructed from borehole logs along a N-S traverse is shown in Fig. 1c.

Before human disturbance, the aquifers were confined beneath the Madhupur Clay. The UDTA is now unconfined (Hasan, 1999) owing to extensive drawdown of the water table during the last 30 years to a level below the base of the Madhupur Clay (Hoque, et al., 2007). The Madhupur Clay and local Recent alluvium restrict direct infiltration of water from surface sources. Nevertheless, local windows in these upper aquitards occur naturally along the rivers, as revealed by site investigation for bridge construction (Hasan, 1999). Other, more modern, windows may have been created through excavation of sand for the construction industry, dredging of the river bed to improve navigability, and from pumping of sand from rivers to infill lowlands for development.

2.3. Hydrology and hydrogeology

Dhaka, the capital of Bangladesh, is typical of the deltaic megacities in Asia. Over the past 300 years the city's footprint has grown from 1.5 km^2 to its present 360 km^2. The population is between 7 and 15 million, depending on where the city limits are placed, and is increasing at around 4% per year. Some 85% of Dhaka's inhabitants use groundwater for their supply of potable water. Exploitation of groundwater has resulted in the water table beneath Dhaka declining at a rate of 1.5 to 3.5 metres per year (Ahmed, et al., 1998, Hoque, et al., 2007), the decline reaching 60 m in some areas.

Dhaka City has a long-term mean annual rainfall of around 2,200 mm with 85% of this occurring between May and October, the monsoon season. The city is surrounded by four rivers: the Buriganga, Turag, Balu, and Lakhya, and one canal, the Tongi Khal (Fig. 1). The river courses are controlled by faults and lineaments (Hoque, et al., 1994, Khandoker, 1987). Many smaller rivers, streams, and canals are connected to these major rivers or to the Tongi Khal. The major rivers are, in turn, distributaries of the Brahmaputra River and the Old Brahmaputra River. River flow is seasonal, with high-monsoon flow being some 8 times that in the dry season in the Brahmaputra River as estimated from the data of Jian et al. (2009) and 5 times greater in the Buriganga River according to Rahman and Rana (1996) and Rahman (2011), giving flows as 700 m^3/sec for the monsoon season and 140 m^3/sec for the dry season.

The Buriganga and Turag Rivers, up to *ca.* 15 m deep, pass through urban and peri-urban areas and receive untreated solid and liquid waste from industry, and sewer and sullage water (Rahman and Bakri, 2010). The volume of waste-water changes little through the year, so the degree to which it contaminates/pollutes the rivers varies through the year in proportion to seasonal variations in baseline flow.

2.4 Urban water system and infrastructure

The public system of water supply in Dhaka dates from 1878 with exploitation of the UDTA. Since 2003, the LDTA has also been exploited (Haque, 2006). Abstraction has increased at a yearly rate of *ca* 5% since the 1980s. Some of the recent production wells, mostly since 2011, are screened in both upper and lower aquifers. By June 2013, the Dhaka Water and Sewerage Authority (DWASA) supplied around 325,000 outlets with 1900 million litres per day (MLD) of potable water sourced from 644 deep tube wells; some 1,150 registered private wells produce an estimated 700 MLD from the same aquifers. In addition, around 500 MLD of potable water is supplied from 4 surface-water-treatment plants. Potable water is delivered through 3,000 km of pipeline (DWASA, 2013).

The present sewerage system was initiated in 1923. Today, more than 30% of Dhaka is connected to a conventional sewer system running to a sewage-treatment plant in SE Dhaka. Most of the city's peripheral areas have yet to be included in this system. For sewage disposal, these other areas use septic tanks, or discharge straight to ground (Haq, 2006). Leakage from water-supply pipelines (Haq, 2006), and from sewers and storm drains contributes to aquifer recharge. The amount has not been quantified, but a value of 30% of recharge stated by van Wonderen (2003 p. 134) may be a maximum, given the low permeability of the Madhupur Clay in which most of the distribution pipes are emplaced.

3. Methods and Materials

Samples of water were collected from 44 DWASA wells and 40 private wells, 12 river locations around Dhaka, 5 sewers from 3 localities, including one from Dhaka's only sewage-treatment plant, and from 2 lakes (Fig. 1). The depth of the wells was between 33 and 335 m below ground level (mbgl), of which 29 were < 100 mbgl. Most of the shallow wells are from outside the city area, where people tap the upper part of the UDTA.

Most samples were collected during August 2012 and July 2013, at the height of the summer monsoon, when rivers were at maximum stage. Repeat sampling at 4 river localities (sites Tu, Bu1–3) along the Turag-Buriganga Rivers were made in the dry-season during January, 2014, and again in February, 2014.

Samples were collected in 15 mL polythene tubes, one acidified in the field with 0.15 mL of 50% Analar nitric acid, one unacidified. The cations analysis samples were analysed by standard methods using inductively coupled plasma atomic emission spectroscopy (ICP-AES). Analysis of As, B, Bi, Br, Cd, Cr, Co, Cu, I, Mo, Pb, Se, Sb, V, and U was by inductively coupled plasma mass spectrometry (ICP-MS) with detection limits for As and Br of around 0.1 and 1 µg/L respectively. Analysis for F, Cl, NO_3, and SO_4 was done by ion chromatography within three weeks of collection. Detection limits for NO_3 and SO_4 were < 0.05 mg/L. Analysis for $\delta^{18}O$ and $\delta^{2}H$ were done on unacidified filtered (0.22 µm) samples using a Picarro WSCRDS laser instrument. Analytical precision was better than 0.08‰.

The collected data are given in Table S1 of the electronic supplementary material (ESM). The stable isotopic data were augmented with stable isotopic compositions for 889 groundwater samples compiled from the literature; the sources are given in Table S2 of the ESM.

In order to assess the contribution to groundwater of sewage effluent and river water, contaminated wells are identified using Cl/Br values, following the development of this proxy to trace waste-water contamination of aquifers in Israel (Mazor and Mero, 1969, Nissenbaum and Magaritz, 1991, Vengosh and Pankratov, 1998), North America (Davis, et al., 2004, Davis, et al., 1998, Katz, et al., 2011, Panno, et al., 2006), Spain (Alcalá and Custodio, 2008), and the Bengal Basin (McArthur, et al., 2012). In addition, end-member mixing calculations were undertaken using Cl concentrations to quantify the contribution of contaminant water to groundwater. End-member values for Cl concentration were based on uncontaminated samples (reported in Table S1 of the ESM), contaminated samples of sewage effluent from West Bengal (McArthur, et al., 2012) and Israel (Vengosh and Pankratov, 1998), and contaminated river water. End-member values are given in Table 1.

Table 1 End-member values

End member	Cl (mg/L)	Cl/Br mass ratio
Groundwater and rain	1.0	165
Sewage effluent - high Cl/Br	300	1500
Sewage effluent - low Cl/Br	300	520
Buriganga River - high flow mean	4	520
Buriganga River - low flow mean	118	1360

4. Results

4.1. Cl/Br ratio and EC

Values of Cl/Br are plotted against Cl concentration and electrical conductivity (EC) in Fig. 2. The Cl/Br values of groundwater samples were classified as high, intermediate, normal, or pristine, based on the proximity of Cl/Br values to lines of mixing with effluent or sea-water (Fig. 2). The Cl/Br in groundwater ranges from 75 to 1335 with around 23 % of samples plotting close to the marine mixing line. There is no relation between EC and Cl/Br (Fig. 2b).

High Cl/Br in groundwater is found mainly in the westernmost part of Dhaka within 3 km of the Turag-Buriganga Rivers (Fig. 3a). Outside of this area, three high Cl/Br samples only (92, 96, 105) occur, and all are close to the Lakhya River (Figs. 1, 3a). Values of Cl/Br are intermediate in an area to the east of Dhaka International Airport, where extensive wetlands occur (Sultana, et al., 2009) and through which the highly contaminated Balu River flows.

Values of Cl/Br in groundwater decrease with increasing distance eastward of the sampled well from the Turag–Buriganga Rivers (Fig. 3b). This relation is not seen to the west of these rivers (Table S1 of the ESM). Increasing depth appears to be associated with a decreasing frequency of high Cl concentrations and high Cl/Br, as well as decreasing concentrations of SO_4 and NO_3, although low values of both are found at all depths, a fact that illustrates the limited reducing capacity of the organic-poor Dupi Tila sands (Fig. 4).

In comparison to other groundwater samples, those from Wells 15, 16, 27, 28, 29, 44, 65, and 69, all tapping the LDTA, have very low Cl/Br, of between 81 and 129 (Fig. 1), heavy $\delta^{18}O$ (> – 3.9‰) and $\delta^{2}H$ (> –21.2 ‰), and Cl concentrations $<$ 2.0 mg/L (Table S1 of the ESM). These groundwater samples are unaffected by human influence, are probably ancient, and so are termed here 'pristine' groundwater.

On a Cl v Cl/Br plot, river water samples plot along mixing lines between uncontaminated groundwater and sewage effluent (Fig. 2). The dry-season (January-February) values for Cl/Br in the Turag-Buriganga Rivers are similar to the Cl/Br of sewer waters from Dhaka and septic-tank outflows from West Bengal: dry-season data for other rivers were not collected. In the Turag–Buriganga Rivers, values of Cl/Br increase downstream (Fig. 3c). Lowest riverine concentrations of Cl are found in the monsoon-season Dhaleshwari River (< 1.5 mg/L; Table S1 of the ESM), which also has the lowest Cl/Br of all rivers (< 270). The two highest riverine concentrations of Cl, and highest Cl/Br, found in the monsoon season occur in the Balu River which had concentrations of Cl of 17 and 26 mg/L, and Cl/Br of 928 and 915.

4.2. Stable-isotopic composition

The $\delta^{18}O$ and $\delta^{2}H$ data of uncontaminated groundwater (Fig. 5a) plot close to, or slightly above, the local meteoric water line (LMWL) for Kolkata, West Bengal. The 8 pristine groundwater samples group close to or above the LMWL at distinctly heavier isotopic composition (Fig. 5). When plotted against depth, the pristine groundwater samples group with a subset of isotopically heavy groundwater from elsewhere across the Bengal Basin (Fig. 6; Table S2 of the ESM).

River waters plot along a well-defined linear array of distinctly lower slope than the LMWL (Fig. 5). Samples collected in August from the river during the monsoon (high-stage) are isotopically lighter than those collected during the dry season in January-February (low-stage). Groundwater samples 11, 18 and 19, which have high Cl/Br, plot on the river line, but others plot between the river line and the LMWL. The isotopically lightest river sample (De 2; $\delta^{18}O$ –10.3‰, $\delta^{2}H$ –71.2‰) is the Dhaleshwari, which also has the lowest Cl concentration of any river (0.8 mg/L), a concentration that reflects its predominant source, which is monsoon rain falling away from urbanization. The isotopically heaviest river is the Balu (Ba 1; δ^{18}O –2.9‰, δ^{2}H –19.1‰) (Ba 1), which is sourced from beels (shallow lakes) to the northeast.

Water samples from sewers have isotopic compositions similar to those of uncontaminated groundwater, or have a slight evaporative overprint (Fig. 5a).

4.3. Other Constituents

Pristine groundwater samples are all low in Cl (< 2.0 mg/L), have low Cl/Br (< 130), and contain neither SO_4 nor NO_3. All are from the LDTA. The maximum concentration of As in these pristine groundwater is 3.7 µg/L in water from Well 44, which also has the highest Cl concentration (1.9 mg/L) and highest Cl/Br (129) of all pristine groundwater. In all other chemical respects recorded here, these pristine groundwater samples differ little from other groundwater.

In other groundwater, concentrations of Cl, SO_4, NO_3, Fe, Mn, Na, Ca, and K vary laterally (Fig. 7), but show little relation to depth except a tendency compared to the UDTA for the LDTA to have lower concentrations of NO_3 and SO_4 and fewer occurrences of high-Cl water. Concentrations of Fe, Mn, Ca, and Mg tend to be higher in the south-west parts of the city (Fig. 7 and Table S1 of the ESM). In contrast, concentrations of PO_4 are higher in the north east of the city. Many wells contain detectable concentrations of Fe, but concentrations exceed 1 mg/L in only 4 wells, three of which have high Cl/Br.

Detectable concentrations of NO_3 were found in 8 well water samples, with concentrations ranging from 0.1 to 29 mg/L: previous reports of NO_3 in Dhaka groundwater include those of Hasan (1999) and Haque (2006). Of 8 wells with > 1 mg/L of NO_3, 5 are within 3 km of the Buriganga-Turag Rivers and have high Cl/Br. The shallowest of the NO_3-containing wells (Well 3; 85 mgbl) has the most NO_3 and concentrations decrease with increasing depth (Fig. 4). Groundwater containing NO_3 also contains SO_4, although SO_4 is also found in samples lacking NO_3. Some 50% of samples contain >1 mg/L SO_4, with a maximum concentration of 35.5 mg/L. Concentrations of SO_4 are higher in the UDTA than in the LDTA (Fig, 4). Waters high in SO_4 have high Cl/Br (Table S1 of the ESM).

Concentrations of total iodine exceed 100 µg/L in 7 groundwater samples, all of which contain > 50 mg/L of Cl and 5 of which plot close to the marine mixing line. Concentrations of K are highest in sewer water, reaching 13 mg/L. Groundwater samples contain < 3 mg/L of K, except for Well 14, which has 6 mg/L. The highest concentration of As in samples within the city limit is 3.7 µg/L in water from Well 44; two other well waters contained around 2 µg/L (Table S1 of the ESM). Concentrations up to 20 µg/L have been reported previously (Ahmed, et al., 2011).

5. Discussion

5.1. Groundwater uses and potential recharge

Groundwater flow to Dhaka's deep aquifers is now assumed to be radially inward (Fig. 8). The Buriganga River has been postulated often as a major source of recharge to Dhaka's aquifers (Ahmed, et al., 2011, Darling, et al., 2002, Hasan, 1999, Hoque, et al., 2007, van Wonderen, 2003). A connection between river and aquifer was proposed by Welsh (1977) on the grounds that seasonal fluctuations in the level of the water table reflected attenuated seasonal fluctuations in river stage. Since then, the Dupi Tila Aquifer has been recorded as cropping out in the bed of the Buriganga River. Furthermore, the existence of such a connection accords with the results of numerical modelling that assume a significant part of recharge occurs from the river (van Wonderen, 2003).

Other attempts to model recharge, attempts supported by the use of electrical conductivity of waters as a proxy for river-recharge by the Buriganga, were interpreted as showing recharge from the river (Ahmed, et al., 1999, Ahmed, et al., 2011, Hasan, 1999, Hasan, et al., 1998). Stable isotopic compositions of groundwater have also been used in an attempt to identify river recharge to the aquifers (Darling, et al., 2002).

5.2 Cl/Br as an indicator of recharge sources

In the eastern part of Dhaka, the paucity of high Cl/Br groundwater shows that sediments overlying the Dupi Tila aquifers largely protect it from infiltration by contaminated surface waters. The only high Cl/Br well water samples in eastern Dhaka are three that are sited close to the part of Lakhya River that bounds eastern Dhaka, and these isolated pockets of contaminated water must be related to infiltration through local windows in the Madhupur Clay.

High Cl/Br is confined almost entirely to westernmost Dhaka (Fig. 3) within 3 km of the Turag-Buriganga River. This distribution is interpreted as reflecting the influence on groundwater of infiltrating river water from the Buriganga and Turag Rivers, water from leaking sewers, and effluents from unsewered sanitation, all of which have higher Cl/Br than uncontaminated groundwater (Fig. 2). The distribution of high Cl/Br waters may partly reflect a westernmost thinning of the Madhupur Clay and so a westernmost limit to the protection against infiltration of contaminated surface water it affords to underlying aquifers (CDMP, 2009). Wells in peri-urban settings to the west of the Buriganga River tap the UDTA but have normal Cl/Br that is unrelated to their distance from the river. The absence of signs of river-bank infiltration to these wells may reflect the lower abstraction on the west side of the Buriganga compared to that on the east in the city of Dhaka.

The Cl concentrations in the Turag-Buriganga Rivers are 4.5 ± 1.5 mg/L and 131 ± 24 at high and low stage respectively. The Cl concentrations of rivers away from Dhaka (samples 60, 80) are < 1.5 mg/L (Table S1 of the ESM). It follows that the higher Cl concentration in Turag-Buriganga River water must be contaminant. For high Cl/Br groundwater, the proportion of water that is derived from contaminated sources can be quantified using end-member mixing calculations between uncontaminated groundwater (3 mg/L Cl) and contaminated water (300 mg/L Cl). A value of 300 mg/L is within the range of 250 to 400 mg/L recorded for sewage effluents from Israel (Vengosh and Pankratov, 1998), and is similar to the maximum concentration of 238 mg/L recorded for septic-tank effluent from West Bengal (McArthur, et al., 2012). On this basis, most wells contain less than 10% of effluent, with only 4 wells containing more. The highest is 42% in Well 76, but this well also has high Na (103 mg/L), high Cl (125 mg/L) and high SO_4 (33 mg/L), whilst having low K (2.1 mg/L), all of which suggest that the well is taping water contaminated by both saline connate water and waste-water.

The maximum concentration of Cl found in the Buriganga-Turag Rivers was 195 mg/L at low flow; the highest concentration at high flow was 6.2 mg/L. Taking a time-average Cl concentration over a year that is a mean of the mean-maximum and mean-minimum concentrations in the Turag-Buriganga (Table S1 of the ESM) gives a concentration around 60 mg/L Cl. Using this value as a contaminated end-member in the models that mix groundwater with river water results in the proportion of river water in well waters being as high as 50%.

These calculations show that Cl mixing models cannot precisely quantify the relative contributions to groundwater in westernmost Dhaka from river and waste-water sources without considering more information, such as seasonal variations in groundwater composition, and without considering other tracers. One other tracer of use is NO_3, since it is all but absent from river waters. Another is stable isotopic composition, given the clear difference between the isotopic composition of rivers and groundwater.

5.3. Isotopic indicators of recharge sources

All groundwater have isotopic compositions equal to, or more enriched (heavier) than, that of Kolkata's mean annual volume-weighted rain (Sengupta, et al., 2008, Sengupta and Sarkar, 2006). Of the waters high in Cl/Br, *i.e.* those from Wells 11, 18, and 19, plot on the line defining the isotopic composition of river water (Fig. 5). These waters must be derived largely from river recharge, and it is therefore no surprise that each is situated no more than 200 m from the Buriganga River (Fig. 1).

In contrast, Well 33, also with high Cl/Br, plots above the LMWL (*d*-excess of 12.2) and squarely amongst the low-Cl/Br well waters on Fig. 5. This stable isotopic composition suggests an insignificant contribution from river water. The well's concentration of Cl is 21 mg/L and mixing models suggest it contains a 6% contribution from waste-water, a suggestion supported by the fact that the well water also contains 12 mg/L of NO_3, a constituent all but absent from river water (Table S1 of the ESM). Similarly, water from Well 36 (Fig. 5) plots squarely on the modern LMWL, yet has a Cl/Br of 469 and a Cl concentration of 7.6 mg/L. This Cl concentration represents a contribution of around 2 to 3 % of waste-water.

Another high Cl/Br well in western Dhaka, Well 8, is some 2.6 km from the Buriganga River (Fig. 1). Water from this well has a Cl concentration of 74 mg/L, has 8.5 mg/L of NO_3, a Cl/Br of 1059, and contains 30.7 mg/L of SO_4. Its stable isotopic composition plots very close to the LMWL (Fig. 5). This well water shows little sign of being influenced by river water; based on Cl alone, about 25% of the water in this well is waste-water.

Finally, to illustrate further the sensitivity of Cl/Br as an indicator of human influence, water from Well 99 is discussed. This water has only 4.2 mg/L of Cl, yet its Cl/Br is 396 and it plots above the LMWL on Fig. 5. The isotopic composition above the LMWL (Fig. 5) precludes contamination by river water. At such low Cl concentrations, the uncertainty in the end-member Cl concentration adopted for uncontaminated groundwater in the mixing model has a large impact on the result of the mixing calculation; nevertheless, a contribution of around 1 % wastewater seems likely. Whilst small, such contributions might represent an advancing pollution front of which early warning is available through monitoring of Cl/Br.

These discussions, the range of isotopic composition in uncontaminated groundwater, and the different isotopic trends shown by river and groundwater, show that the degree of mixing of river recharge into groundwater cannot be assessed using only linear end-member mixing along the local or global meteoric water line, as proposed by Darling et al. (2002). Rather, the position of a characteristic water sample between the river line and the LMWL, or even a separate line, as yet undefined, for uncontaminated groundwater, should be used to obtain the proportion of river water in a well's water. Given today's analytical precision on isotopic analysis, such results are likely only to be semi-quantitative.

The distinctive composition, both isotopic and elemental, of the pristine groundwater beneath Dhaka is interpreted as showing that it is of a different age, and so origin, to most groundwater under Dhaka, and it is hypothesised that it may be isolated to some degree by persistent silt-clays that occur in, but largely between, the UDTA and the LDTA (Fig. 1). The implication of this hypothesis for the sustainability of abstraction is explored further below.

5.4. Electrical conductivity as an indicator of recharge sources

The electrical conductivity of groundwater is not related to Cl/Br (Fig. 2b), and therefore not directly related to human contamination; rather, high EC occurs in both contaminated (high

Cl/Br) and uncontaminated (normal Cl/Br) water, the latter owing to mixing with saline water. Pockets of saline groundwater have been reported from other parts of the Bengal Basin (Hoque, et al., 2003, McArthur, et al., 2012), including the eastern part of Dhaka (IWM, 2006). A Cl mass-balance shows that the maximum contribution of salt water to waters of normal or low Cl/Br is around 1 %. Given the above, the use of EC values alone as a tracer of a recharge plume from the Buriganga River to Dhaka's aquifers (e.g., Ahmed, et al., 2011, Hasan, 1999) is likely not to present an accurate picture of recharge.

5.5. Sustainability and drawdown

The presence beneath central Dhaka of a core of low-Cl, uncontaminated groundwater, that has a distinctive $\delta^{18}O/\delta^{2}H$ character, suggests a possible new interpretation for the groundwater dynamics beneath Dhaka. Groundwater piezometric contours in the UDTA are typically shown to be disposed approximately concentrically about central Dhaka, with highest drawdown beneath this central region (Fig. 7 of Hoque, et al., 2007; Fig. 8). With such heads, groundwater resource modelling assumes radial inflow to this central region. Given that this region contains unusual groundwater, largely in the LDTA, that lacks any chemical trace of human influence, it is postulated that the LDTA in this central region beneath Dhaka is hydraulically isolated. Such isolation would increase drawdown over that possible if hydraulic connections were present. Isolation may result from an appropriate disposition of the numerous intercalated silt-clays within (largely between) the aquifers, UDTA and LDTA (Fig. 1). The high drawdown in this central region may therefore be a consequence of the fact that water is being mined.

Pristine groundwater apart, when pumped, wells near the Buriganga River show less drawdown than do wells tapping the uncontaminated aquifer beneath central Dhaka (P. Ravenscroft, UNICEF, *pers. comm.* 2013). This lesser drawdown has been interpreted to show that the River Buriganga is recharging the aquifers beneath Dhaka. The data presented here suggests that recharge in western Dhaka is much supplemented by infiltration of water from leaking sewers and unsewered sanitation. Pumping wells close to the river may be acting as interceptors that restrict recharge to more distal locations. Pumping wells in the northern periphery of the water-table depression (Fig. 8) are also acting as barrier wells that prevent flow of mostly uncontaminated groundwater from the north.

In assessing future sustainability of the aquifers beneath Dhaka, recharge from both the Turag-Buriganga and leaking sewers needs to be accounted for. Beneath central Dhaka local isolation of the LDTA may also occur, making these low-Cl sources of water less sustainable than supposed. Elsewhere they are likely to be more sustainable because hydraulic conductivity and transmissivity of the overlying aquitards of westernmost Dhaka have been underestimated.

Nevertheless, many other effects on potentiometric surfaces exist e.g. atmospheric pressure, mechanical loading and de-loading due to rainfall, flooding, water table decline, and urbanisation (e.g., Sophocleous, et al., 2006, Steckler, et al., 2010) have not been fully evaluated so far and may be important for resource evaluation and groundwater flow characterisation (e.g., Harrington and Cook, 2011) of Dhaka's aquifers.

5.6. Ages

Few published ages exist for Dhaka groundwater. Groundwater at 137 mbgl on the northern edge of Dhaka contained 43.7 pMC (percent modern carbon) for dissolved inorganic carbon (Majumder, et al., 2011), which could indicate anything between modern recharge and an age of 2,500 years, depending on the corrections applied (e.g., Hoque and Burgess, 2012) to derive an age from this value. Groundwater ages at two locations within 4 km of the Buriganga River were estimated to *ca* 20 years (Darling, et al., 2002) using ^{3}H – ^{3}He data. The high Cl/Br identified in some groundwater samples (this study) is not in conflict with such values. The wells sampled here show no relation between the age of the well (years before 2012) and Cl/Br; rather, Well 15, delivering what is termed pristine groundwater, is 12 years old, making it one of the older of the wells sampled.

5.7 Compliance

Concentrations of As, B, Ba, Cd, Cu, F, Ni, NO_3, Pb, Sb, Se, and U in Dhaka groundwater samples do not exceed WHO guideline values for drinking water. There was no analysis for Hg. The limitation on As concentrations is noteworthy, given that As is a common pollutant in groundwater elsewhere in the Bengal Basin (Jakariya, et al., 2007, McArthur, et al., 2004, Ravenscroft, et al., 2009, van Geen, et al., 2003). Although below 10 μg/L, concentrations of As are higher in the LDTA than in the UDTA (Table S1 of the ESM), probably because of the greater sorption capacity for As of the sedimentary iron oxyhydroxides in the brown sands of the UDTA compared to the grey sands of the LDTA (DPHE, 1999; Stollenwerk, et al., 2007).

In 62% of Dhaka groundwater samples analysed here, concentrations of total iodine are < 10 µg/L. In 11 (15%) well waters, concentrations are > 40 µg/L. Only in these 11 are concentrations of iodine (I) sufficient to provide most of the recommended daily intake of iodine for humans of 50 – 250 µg/day, depending on maturity and gender (ICCIDD, 2013). These high-I wells are also high in Cl. The I/Cl mass ratios in these groundwater is around 3 x 10^{-3} and exceeds by more than a factor of 10^2 the I/Cl mass-ratio in sea water, so residual salt water alone has not sourced this iodine.

A tannery at Hazaribagh, in SW Dhaka, in the immediate vicinity of wells 9 and 10 (Fig. 1) discharges waste water to low ground and the Buriganga River (Zahid, et al., 2006). The one sample of tannery waste analysed (this study) contained 7,200 µg/L of Cr, 2,900 mg/L of SO_4, and 1,700 mg/L of Na. Water from Wells 9 and 10, and nearby Wells 11, 12, and 13, have water with high Cl/Br, but Cr concentrations of less than 0.3 µg/L. These values confirm the findings of Zahid et al. (2006) that local pollution by the tannery industry has not yet found a way to infiltrate the aquifer in this locale, despite the high Cl/Br in groundwater from these wells. Concentrations of Cr up to 16 μg/L are found in well water from the northern part of Dhaka, 5–10 km north of the tannery.

6. Conclusions

The value of Cl/Br in groundwater is a sensitive tracer that can identify the presence of as little as 1% waste-water. Using this tracer, it is shown that the aquifers beneath Dhaka are largely protected by their overlying aquitard, the Madhupur Clay and more recent deposits, from infiltration of contaminated water. Infiltration by waste-water, and contaminated river water in the Buriganga-Turag Rivers, is occurring to aquifers beneath westernmost Dhaka. Very limited infiltration of waste-water is occurring through wetlands to the east of Dhaka airport. More severe contamination has been recorded at three sites along the Lakhya River.

Using EC alone, the pathways or proportions of recharge from such sources cannot be estimated, owing to the presence beneath Dhaka of salt-water contamination that influences values of electrical conductivity.

Stable O- and H-isotopic data can be used to establish semi-quantitatively the proportion of groundwater that is derived from river recharge. The scale and source of recharge to Dhaka's aquifers needs to be further quantified. Infiltration from rivers surrounding Dhaka appears limited and should not, at present, be assumed large in the hope of increasing the long-term sustainability for Dhaka's supply of potable water.

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Figures

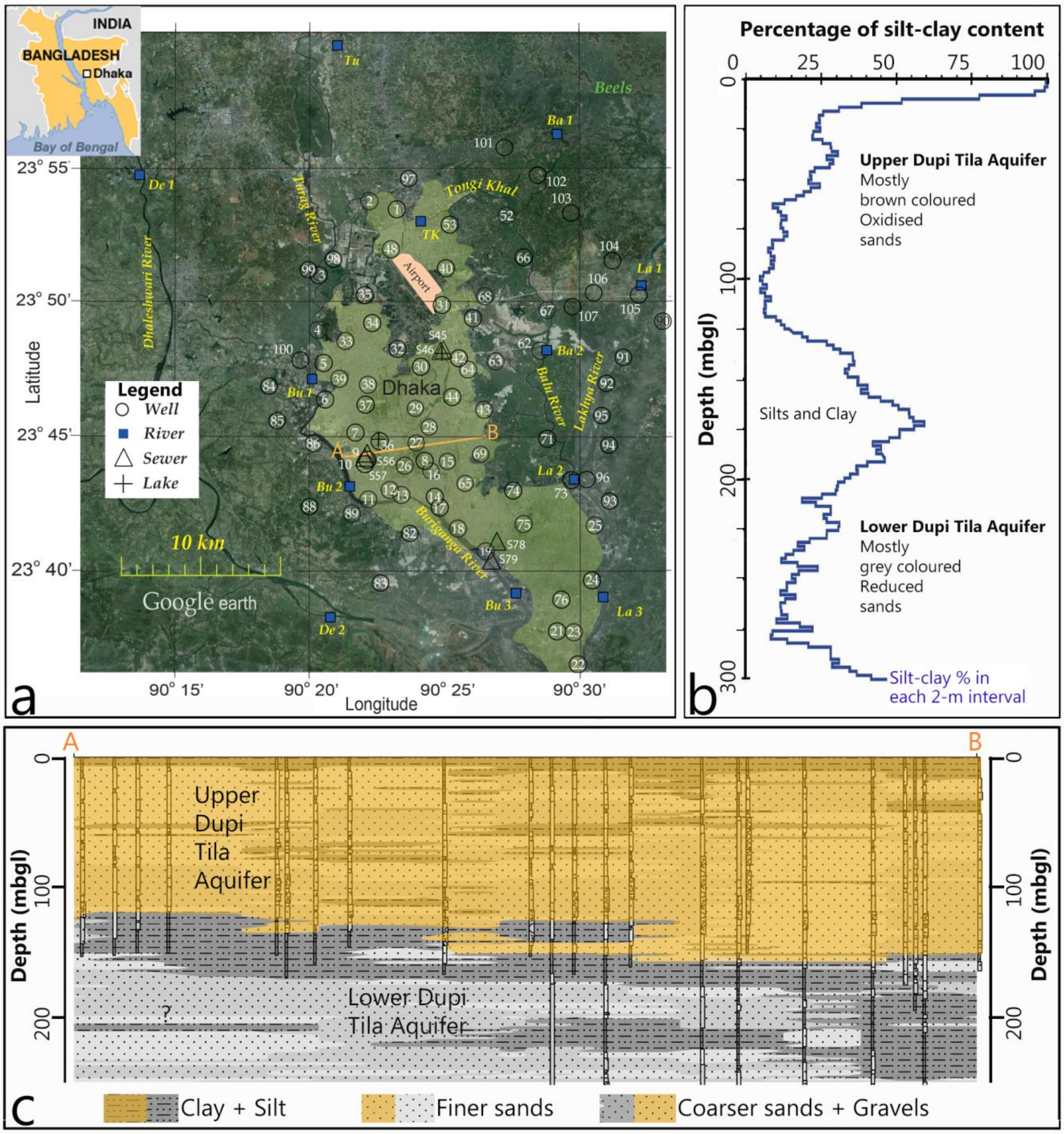

Fig. 1. a) Location, type, and sample/site numbers, of waters collected from Dhaka, Bangladesh. On the map, all the labels are sample numbers, except Tu, Bu1, Bu2 and Bu3 which are the sites where samples for both high- and low-flow condition were collected. Base image from Google Earth Pro, with permission. b) vertical profile through the aquifer showing the % of silt+clay with depth aggregated over 2-m slices for 128 drill logs for the city. c) Cross sections through the sediments beneath Dhaka along line in Fig. 1a. Latitude and longitude to the WGS84 co-ordinate system.

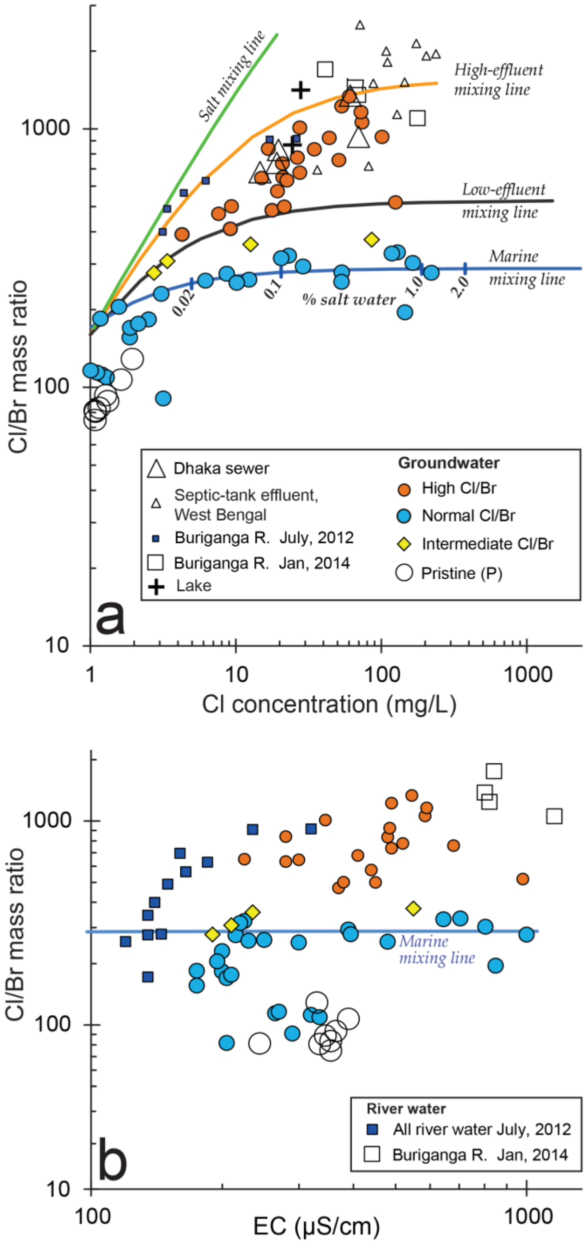

Fig. 2. a) Plot of Cl against Cl/Br mass ratio for groundwater, river water, and sewer water. End-members for mixing lines are given in Table 1. Uncontaminated groundwater end-members are Cl 1 mg/L and Cl/Br 165. Other end-members for mixing lines are given in the section *Methods and materials*. Values for septic tank effluent from West Bengal are from McArthur et al. (2012); b) Plot of Cl/Br mass ratio against measured electrical conductivity (EC), showing little relation between the two. Note symbols representing groundwater are same in both panels.

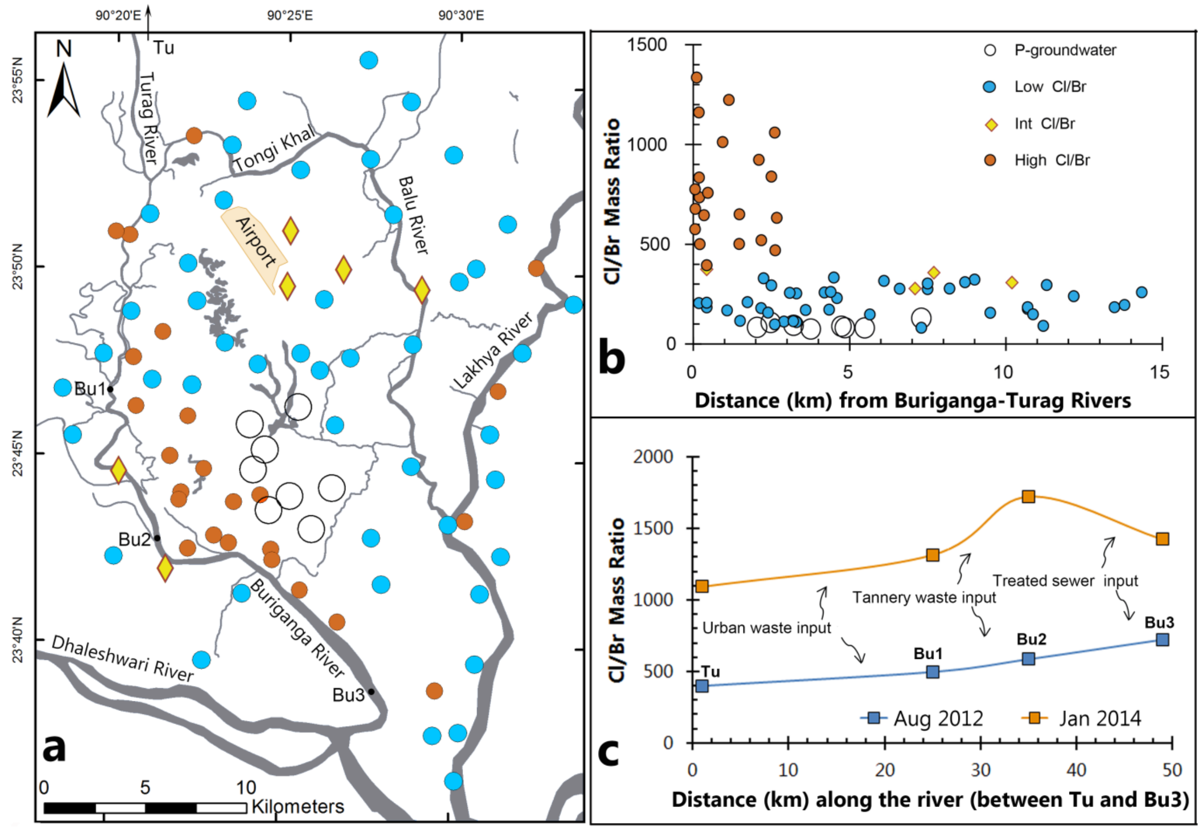

Fig. 3. a) Distribution of Cl/Br in well waters across Dhaka. b) Cl/Br in groundwater as a function of distance eastward of the Turag-Buriganga River; symbols as in Fig. 2. Lakhya River samples not included. c) Cl/Br mass ratio in the Turag-Buriganga River as a function of distance downriver from site Tu to Bu3 on Fig. 1.

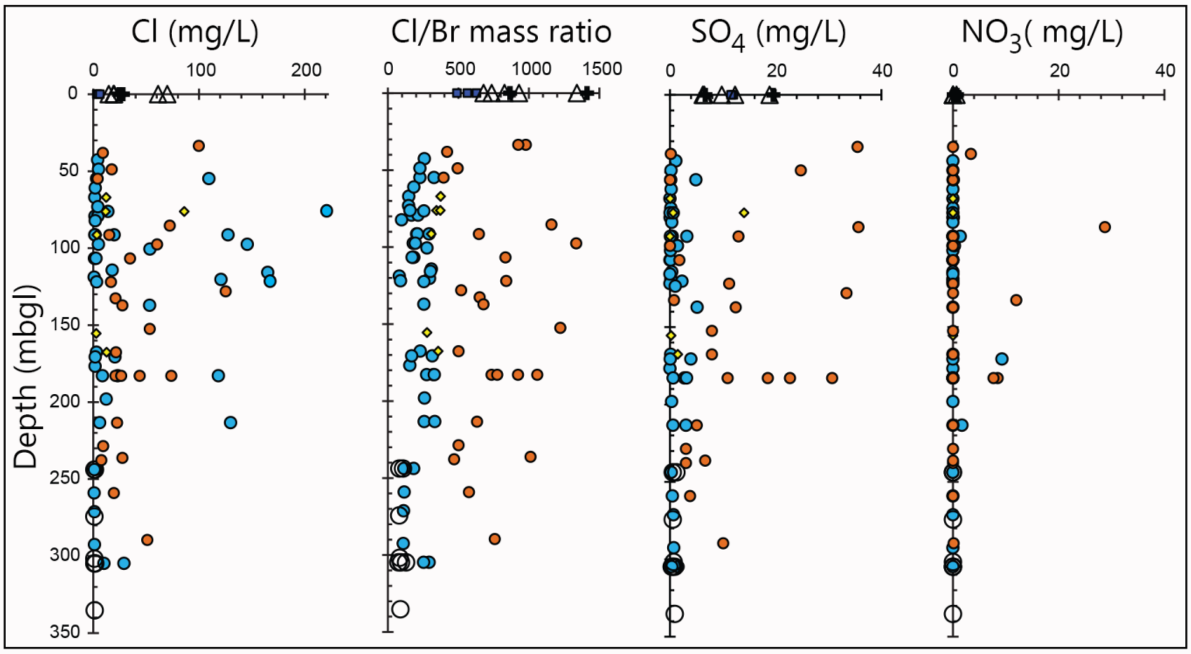

Fig. 4. Variation of Cl, Cl/Br, SO_4 and NO_3 with depth of well. Symbols as in Fig. 2.

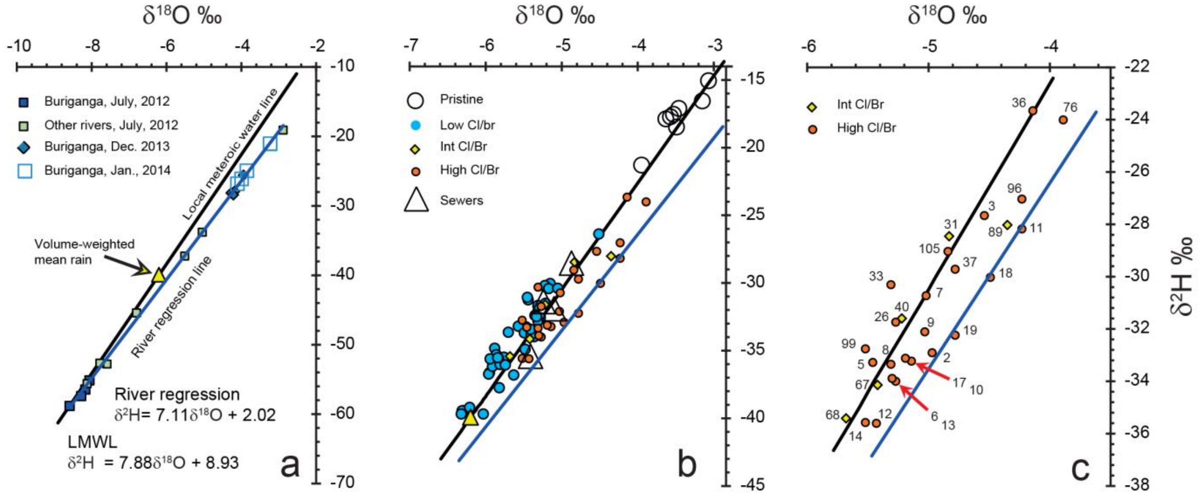

Fig. 5. a**)** Plot of δ^{18}O *vs* δ^{2}H for rivers, and the local meteoric water line (LMWL) of Sengupta and Sarkar (2006) as modified by Sengupta et al. (2008). b) Groundwater samples. c) Groundwater samples with high Cl/Br. For a discussion of numbered wells, see the text.

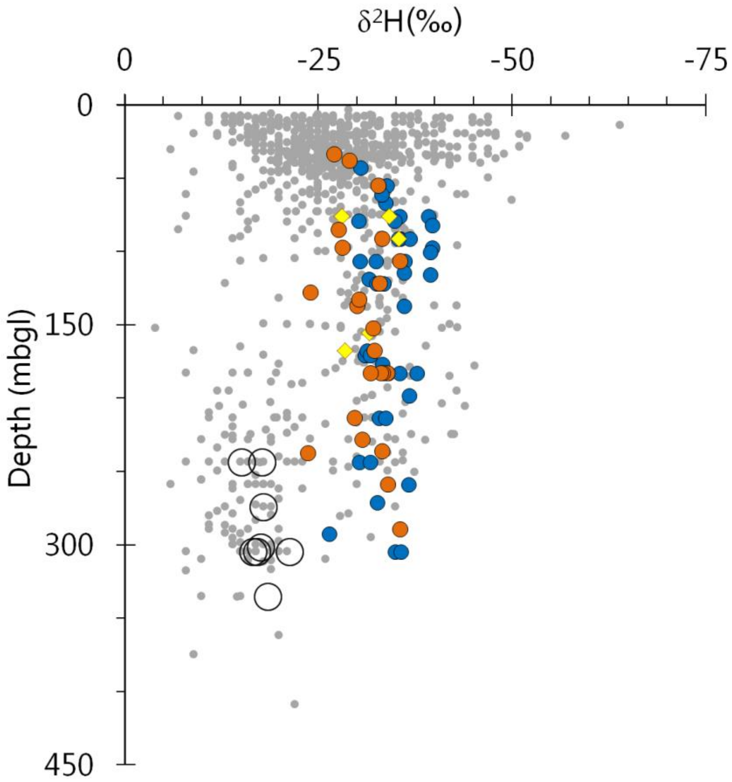

Fig. 6. Hydrogen-isotope composition of Dhaka groundwater samples plotted against depth, overlaid with 889 other isotopic data for groundwater samples from elsewhere in the Bengal Basin (grey dots); sources in Table S2 of the ESM.

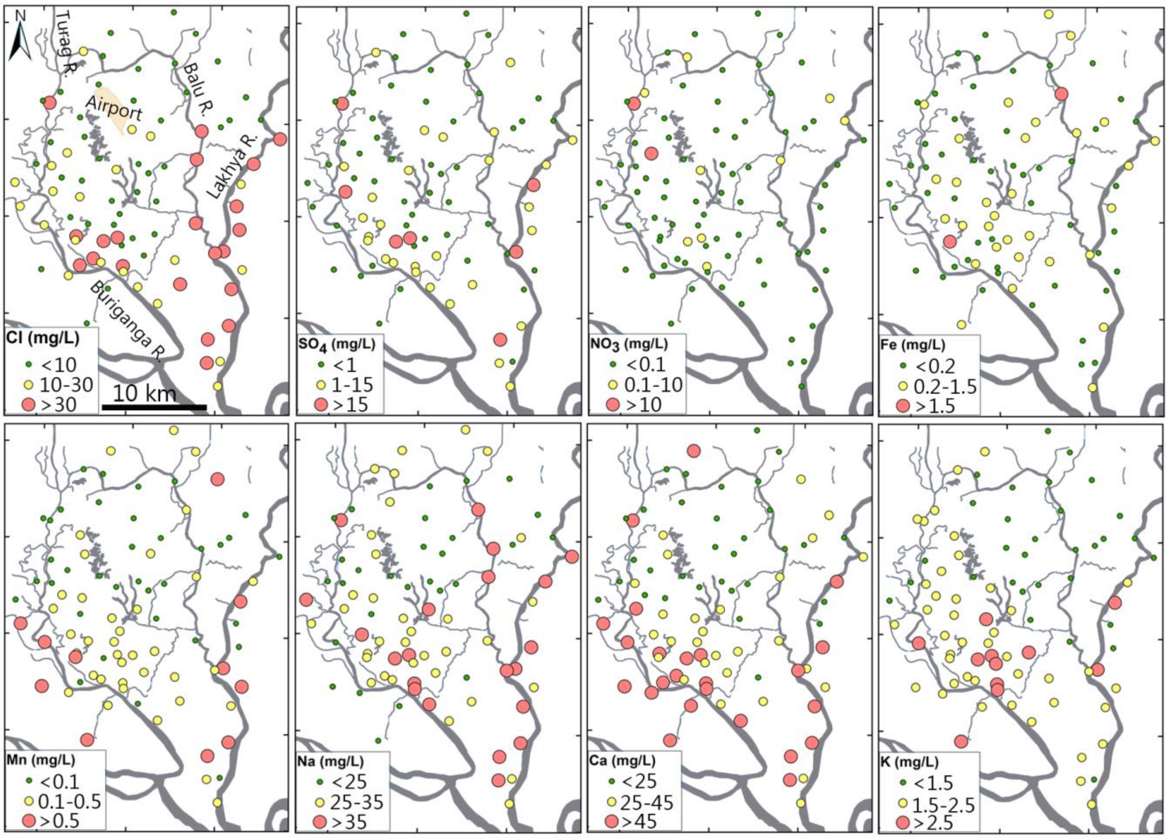

Fig. 7. Spatial distribution of constituents in groundwater.

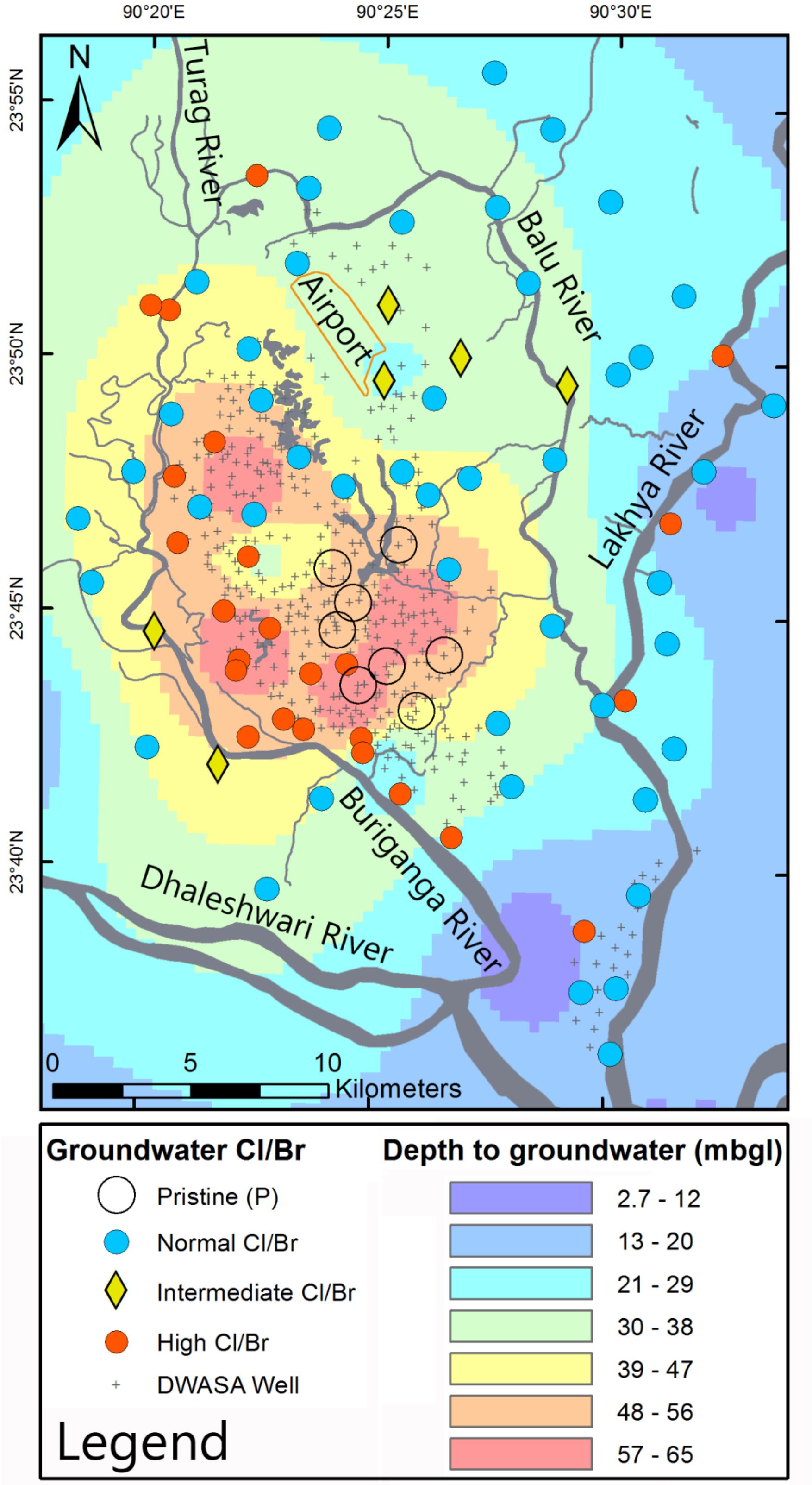

Fig. 8. Isopachs of depth to the water table beneath Dhaka, in April 2010, based on water level monitoring data of the Bangladesh Water Development Authority. Also shown are the Cl/Br ratios of groundwater samples and the locations of all DWASA production wells in Dhaka, some of which (Table S1 of the ESM) were sampled for this study.