



Hydrochemical fingerprinting and effects of urbanisation on the water quality dynamics of the Quaternary aquifer of south Bengal Basin, India

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Abstract

Hydrochemical fingerprinting of groundwater helps to analyse the physico-chemical dynamics and assesses pumping-induced hydrochemical changes within a complex and stressed deltaic aquifer system of an urban-peri-urban area in south Bengal Basin in India using Kolkata and Howrah cities as case study. Statistical analysis of geochemical parameters of 287 groundwater samples shows that eight hydrochemical facies occur in the study area, which can be broadly categorized into fresh, blended and brackish water covering 53, 186 and 171 km² area, respectively, excluding the area covered by River Hugli. In the past two and half decades, due to over-withdrawal of groundwater, the groundwater trough in south-central Kolkata increased by 266 km² and resulted in the deterioration of fresh water (mean TDS = 518 mg/L) into blended (mean TDS = 722 mg/L) and brackish water (mean TDS = 1385 mg/L) due to hydraulic mixing, flushing and ion-exchange. A comparison of the common area of the hydrochemical facies of mid-nineties and present study reveals a reduction of area of fresh water by 74% and increase of area of blended and brackish water by 42% and 32%, respectively. Contamination of groundwater by anthropogenic arsenic clustering around a palaeo-channel named Tolly's Nala, wastewater from sewerage, copper, lead, cadmium and zinc surrounding the industrial areas are detected in wells where the upper aquitard is either absent or < 10 m in thickness. At the present rate of withdrawal, the quality of groundwater may further deteriorate at an accelerated pace and the groundwater of the entire area may become unsuitable for drinking in future.

Keywords Hydrochemical fingerprinting · Quaternary aquifer · Principal component analysis · Arsenic · Heavy metals · South Bengal Basin

Introduction

Since the early fifties, aquifers beneath urban and peri-urban areas in various river basins in India, especially those of Ganga, Brahmani-Baitarni and Indus are highly stressed. In each of these areas > 66% of water supply comes from groundwater while Mahi, Pennar and Sabarmati basins have groundwater supply > 40% (Fig. 1c, inset) (Patel and Krishnan 2009). The aquifer of south Bengal Basin, located in the lower deltaic plain of River Ganga, also has been subjected to tremendous stress as a result of huge exploitation of groundwater due to rapid population growth and

urbanisation (Sikdar 2001; Banerjee and Sikdar 2021). Decades of high-capacity pumping of groundwater has lowered the groundwater level by up to 14 m since 1950s', creating multiple cone-of-depressions in the Quaternary aquifer of Kolkata city (McArthur et al. 2018). Over the years, the exploitation of groundwater has significantly increased in the surrounding peri-urban areas. The groundwater has also been over-exploited in Howrah city. Bannerji and Mukhopadhyay (2018) elaborated that there is a significant gap between the treated surface water supply and demand for the people residing in Howrah city. Hence, the supply side is being managed by tapping the groundwater indiscriminately. Patra et al. (2018) indicated that the water level has depleted rapidly in Howrah city since 1996. The rate of fall of the piezometric surface in the urban and peri-urban areas of Kolkata and Howrah varies between 0.13 and 0.37 m/year (Banerjee and Sikdar 2021).

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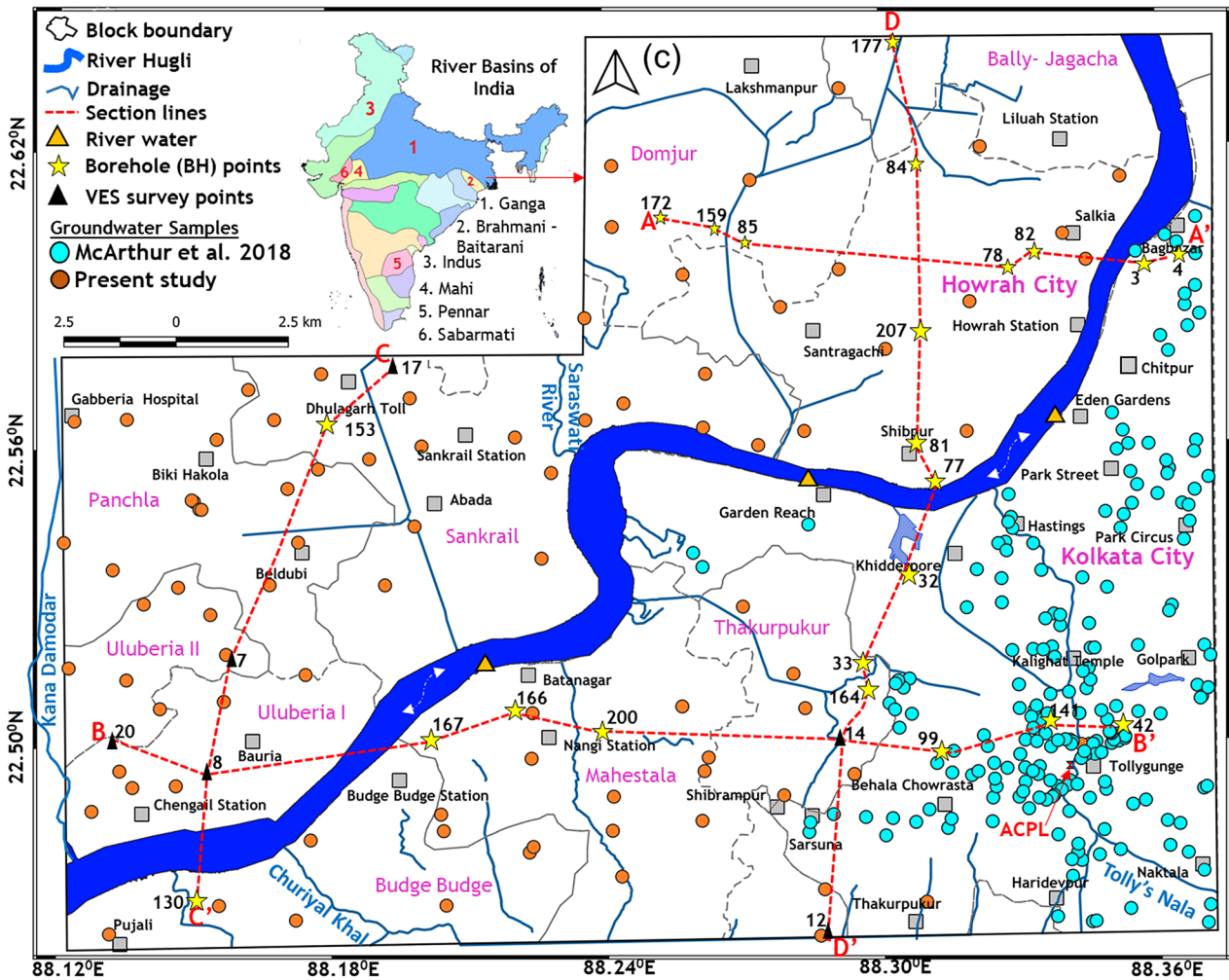
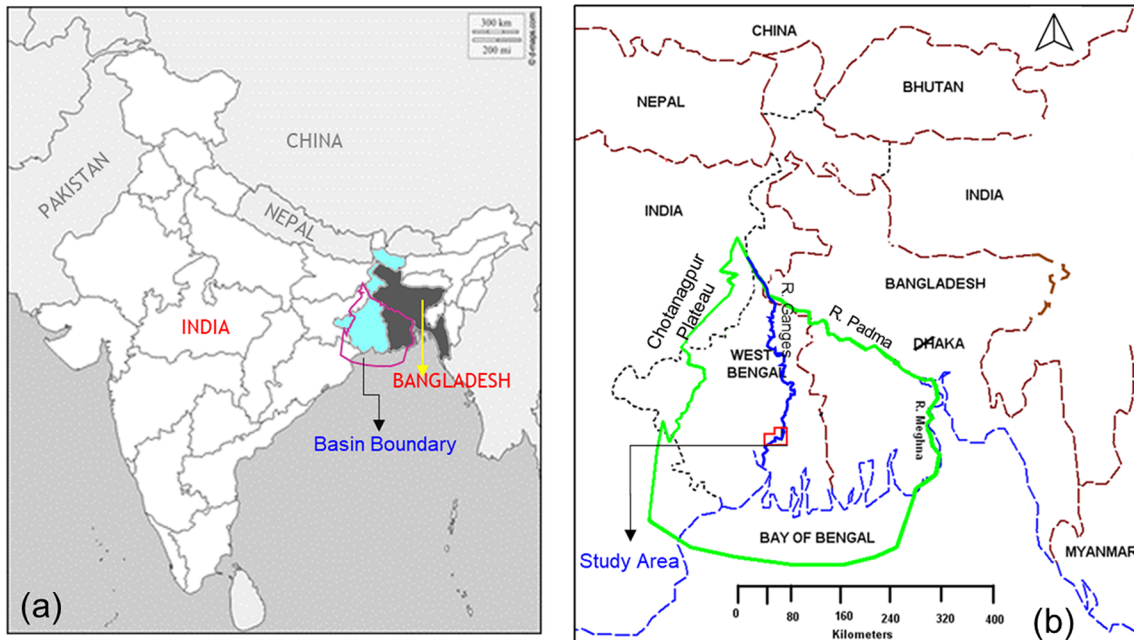


Fig. 1 **a** Location of the south Bengal Basin in the map of West Bengal (India) and Bangladesh; **b** area of the south Bengal Basin where green line indicates basin boundary, red boundary indicates study area, dotted blue line indicates coastline, brown line indicates international boundary and black dashed line indicates state boundary; **c** the study area showing major drainages and canals, block boundaries, section lines (AA', BB', CC', DD') and groundwater and river water sampling points. The map at inset shows the river basins in India (https://indiawris.gov.in/wiki/doku.php?id=river_basins, accessed on 19.5.2021)

In recent time, with rapid urbanisation and improved quality of life, the demand for groundwater resources is increasing (Ajdary and Kazemi 2014; Adimalla and Wu 2019; He et al. 2020, 2021). The over exploitation has put tremendous stress on the aquifer system resulting in the development of cone-of-depression in many cities like Cangzhou, Dezhou, Tianjin of North China Plain (Li et al. 2014); Dhaka, Bangladesh (Hoque et al. 2007); Tripoli City, North West Libya (Alfarrah and Walraevens 2018). The associated geo-hazards with the development of cone-of-depression are land subsidence (Su et al. 2020; Banerjee and Sikdar 2020), fissures, change of hydrochemistry and quality deterioration due to the change of recharge and discharge patterns. (Sun et al. 2007; Yuan et al. 2013; Su et al. 2020). Depletion of groundwater level and development of cone of depression due to the over withdrawal of groundwater in Upper Silesia, Poland (Samborska and Halas 2010) had resulted in rock-water interaction which ultimately induced changes in groundwater quality making it unsuitable for drinking purpose (Su et al. 2020). The groundwater quality of the carbonate aquifers in the karst topography area of Seville in Spain has also changed due to the intensive pumping (Martos-Rosillo and Moral 2015). As a result of continuous extraction of groundwater, the poor-quality groundwater at shallow aquifer had leaked into the middle and deeper aquifer of Shizuishan City, China resulted in the degradation of quality of the confined aquifer system (Wu et al. 2015). The depletion of groundwater levels and increased contamination due to urbanization and resultant exploitation of groundwater have been reported in places like London, UK (Marsh and Davies 1983), Dessau, Germany (Riemann 1997), Jakarta, Indonesia (Rismianto and Mak 1994), Dhaka, Bangladesh (Naik and Tambe 2008), Southern Delaware, USA (Ritter and Chirnside 1984), Hyderabad, India (Subrahmanyam 1996; Naik and Tambe 2008), and Tomsk, West Siberia, Russia (Lam 1999). Saline water ingress due to over-withdrawal of groundwater has been reported in Mar del Plata, Argentina (Bocanegra et al. 1993), Chennai, India (Rao and Thangarajan 1996; Naik and Tambe 2008; Manivannan and Elango 2019; Kanagaraj et al. 2018) and Valletta, Malta (Naik and Tambe 2008).

Apart from lowering of the piezometric surface, over pumping of groundwater due to urbanization has also

resulted in land subsidence (Sikdar et al. 1996; Chatterjee et al. 2007; Sahu and Sikdar 2011; Banerjee and Sikdar 2020, 2021) change of aquifer behavior and quality deterioration in the Quaternary aquifer system of south Bengal Basin (Banerjee and Sikdar 2021). Banerjee and Sikdar (2021) also related the increase of total dissolved solids (TDS) by an average of 370 mg/L and chloride (Cl) by an average of 89 mg/L in groundwater of the area in and around Kolkata and Howrah to over-withdrawal of groundwater. Other cities in India have also witnessed groundwater quality deterioration due to urbanization. Some notable examples are chloride and iron pollution in Jaipur (Rajasthan), metal pollution in Ludhiana (Punjab), chromium pollution in Faridabad (Haryana), major ion and metal pollution in Bhopal (Madhya Pradesh), nitrate pollution in Lucknow (Uttar Pradesh), nitrate and bacterial pollution in Tirupati (Andhra Pradesh), high iron (Fe) hazards, groundwater decline, saline water intrusion and contamination in Chennai (Tamil Nadu), groundwater decline and contamination in Hyderabad (Telangana) (Naik et al. 2008), and fluoride (F) contamination in Vellore (Tamil Nadu) (Kumar et al. 2014).

A general qualitative understanding of these impacts is known but the associated physico-chemical dynamics in the complex groundwater system are not well understood (Tyagi et al. 2008). Therefore, the objective of this research is hydrochemical fingerprinting of groundwater to understand the pumping-induced hydrochemical changes within a stressed aquifer system of an urban-peri-urban area in south Bengal Basin in India. This understanding will help city planners and policy makers worldwide to plan sustainable development of groundwater to prevent its quality deterioration which may have adverse impacts on human health.

Methods

Study area

The study area comprising 436 km², is a part of south Bengal Basin, located on the eastern and western banks of the River Hugli and covers Kolkata and Howrah cities and their peri-urban areas in S24-Parganas and Howrah districts, respectively (Fig. 1c). The area does not exhibit any marked topographical features. The landscape is mostly flat, and the elevation varies between 3 and 8 m above the mean sea level (msl). The subsurface lithological assemblages of the upper few hundred meters of the south Bengal Basin comprises a buried late Pleistocene landscape of palaeo-channels and palaeo-interfluvial overlain by Holocene channel/flood plain deposits (McArthur et al. 2018; Banerjee and Sikdar 2021). The study area lies under the fluvio-deltaic depositional environment. The peat layers in the upper horizons had been deposited as a result of

boggy and marshy conditions prevailing at the end of the sedimentation process (Sikdar 2000). The irregular nature of the subsurface lithology is characterised by the filled up palaeo-channels and a natural levee on both sides of the River Hugli.

The sedimentary architecture of the field area has been well elaborated using the borehole lithological data and geophysical data in Banerjee and Sikdar (2021) and in four cross sections of Fig. 2 in this research paper. A confining clay/silty clay layer of average 26.30 m thickness occurs at the upper part of sedimentary succession (Banerjee and Sikdar 2021). The clay isopach map (Fig. 5 of Banerjee and Sikdar 2021) reveals that the thickness of the upper confining bed is 21 m and 37 m towards the eastern and western sides of River Hugli, respectively. Below the upper confining bed, coarse clastic deposits of fine to coarse sand at places mixed with gravels, of about 300 m thick form the Quaternary confined aquifer beneath the study area (Sikdar 2000; Banerjee and Sikdar 2021). Spatially uncorrelated clay, silty clay and clayey silt lenses of various dimensions within the sandy layer make the aquifer composition heterogenous in nature (Sahu and Sikdar 2017). The grain size sharply increases to sand lithofacies within the study area at 60 m depth which continues up to 150 m depth covering an area > 400 km² (Banerjee and Sikdar 2021). Therefore, the screen of the

wells in and around Kolkata and Howrah cities is generally placed in between 80 and 140 m bgl.

The thick Holocene clay/silty clay and late Pleistocene Last Glacial Maximum Paleosol (LGMP) restricts any major infiltration of water from River Hugli in the study area (Sikdar and Dasgupta 1997). The upper confining clay/silty clay bed reduces to < 10 m or absent at places, e.g., along the course of Tolly's Nala, Mahestala and Budge Budge (M-B) in the southern and south-western parts of the study area (Fig. 1c; Fig. 5 of Banerjee and Sikdar 2021). The Holocene clay/silty clay and the LGMP were not deposited, and grey sands are present right below the Tolly's Nala bed, which provides a direct flow path for the Nala water to infiltrate into the underlying Quaternary aquifer system (McArthur et al. 2018).

In the Bengal Basin aquifer system, in general, and the present field area, in particular, the natural flow regime combines three scales of flow system (Ravenscroft 2003). The kilometer scale flow system in the shallow aquifer, is driven by local topography. The second scale flow system is controlled by micro-topographic features like channels, levees and ponds which laterally extend for few tens to hundreds of meters, and vertically for 10 to 20 m. The natural flow to the south in the Quaternary deeper aquifer system can be considered as the third scale of flow system. The groundwater

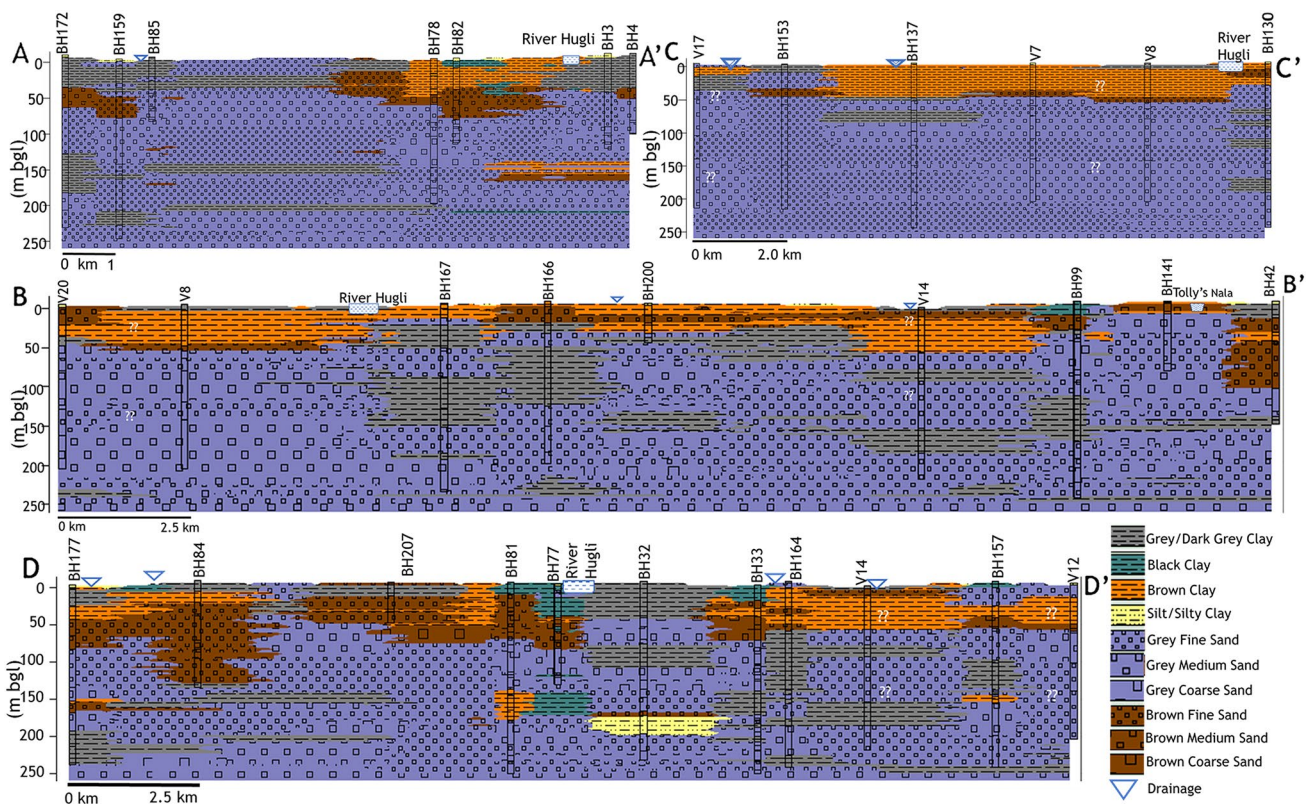


Fig. 2 Geological sections along **a AA'**, **b BB'**, **c CC'** and **d DD'** in the study area. Sections lines are given in Fig. 1c

flow systems of Bengal Basin are both local and regional in scale (Michael and Voss 2009; Sahu et al. 2013; Sikdar et al. 2013; Sikdar and Chakraborty 2017) where subsurface layers are considered as connected in nature. The area of the south Bengal Basin extends from the River Ganges and River Padma in the north to the Bay of Bengal in the south and from the outcrop of Precambrian crystalline bedrock of the Chotanagpur Plateau on the west, to the River Meghna in the east in Bangladesh (Fig. 1a,b) (Michael and Voss 2008).

Since historical time, steady urbanisation in and around the twin cities had been influenced by the easy availability of surface water from River Hugli and groundwater from the Quaternary confined aquifer (Banerjee and Sikdar 2021; Sikdar et al. 2001). In Kolkata, during the period 1980–2014, the built-up area had increased from 100 to 162 km² while the areas occupied by wetland and vegetation had reduced by 40 km² and 23 km², respectively (Mukherjee et al. 2018). During the period 1985–2016, in Howrah city, the built-up area had increased from 27 to 38 km², while the surrounding peri-urban and rural areas within the study area had increased from 22 km² to 62 km² (Banerjee and Sikdar 2021). This expansion of urbanised area by > 3 km²/year in and around the study area has increased the total groundwater abstraction manifold.

As a consequence, the groundwater flow pattern changed from an open system, which was from north to south in the mid-1950s, to a closed one in the mid-1980s with the formation of a groundwater trough. This closed groundwater flow system persisted in the 1990s with the increase in the area of the trough (Sikdar 2000). The trend of water level decline was about 0.33 m/year at the centre of the trough in Park Street and 0.11 m/year towards the periphery (CGWB 2006). With the increasing pumping, the area of the groundwater trough increased by 15 times at the rate of 8.6 km²/year between 1985 and 2016 resulting in a general west to east groundwater flow pattern (Banerjee and Sikdar 2021). In the pre-monsoon period of 2016, the deepest piezometric surface of 20.56 m bgl was recorded in the northern part of Kolkata in Chitpur area and the shallowest piezometric surface of 7.75 m bgl was recorded south-west of Kolkata city in M-B area (Fig. 1c) (Banerjee and Sikdar 2021).

In Kolkata and surrounding area, the general geochemical characteristics of the groundwater are fresh to brackish with high TDS and iron (Fe), occasional presence of arsenic (As), and high Cl and nitrate (NO₃) at places (Sikdar et al. 2001; CGWB 2006). The major types of groundwater found in the region are (a) Ca-Mg-HCO₃, (b) Na-HCO₃, (c) Ca-Mg-Cl, (d) Na-Cl (CGWB 2006). Groundwater in Howrah district is Na-HCO₃ and slightly alkaline in nature (CGWB 2004).

The wastewater impact on groundwater in the Bengal Basin had been uncared for in favour of investigation of the pollution by arsenic (DPHE 1999; 2001; Jakariya et al. 2007; Nickson et al. 2007; PHED 1991; Van Green et al. 2003).

Chloride and bromide (Br) mass ratios and concentrations in groundwater of Cl, NO₃, sulphate (SO₄) and ammonium (NH₄) have been used to show the extent to which groundwater in the Bengal Basin is contaminated by wastewater (McArthur et al. 2012a) and in Kolkata city (McArthur et al. 2018). The high-Cl sewage effluent has increased the Cl concentration in parts of the aquifer beneath the Kolkata city (McArthur et al. 2018; Banerjee and Sikdar 2021).

Groundwater in south Bengal Basin occurs in two principal settings: palaeo-channel and palaeo-interfluvial. Palaeo-channel setting consists of palaeo-valleys filled by grey sands since the end of the last glacial maximum. Palaeo-interfluvial setting comprises brown sand aquifer capped by a palaeosol (brown stiff clay) that formed between river channels prior to the LGM and are now buried by more recent sediments (McArthur et al. 2011). Arsenic occurs naturally in the sediments of the Bengal Delta and is released into the shallow groundwater in palaeo-channel setting by reduction of sedimentary FeOOH by dissolved organic matter.

Generally As pollution of groundwater occurs in alluvial aquifers worldwide (Ravenscroft et al. 2009) but is especially severe in Bengal Basin (PHED 1991; DPHE 1999; Van Green et al. 2003; Chakraborty et al. 2009; Mukherjee et al. 2007a, b; 2009; Jakaria et al. 2007; McArthur et al. 2008, Sikdar and Chakraborty 2010; McArthur et al. 2011; 2012a; 2012b; Sikdar et al. 2013; Ghoshal et al. 2015; Sahu and Sikdar 2011; Sikdar and Chakraborty 2017) where millions of consumers are drinking water containing > 10 µg/L As (WHO 2017; BIS 2015). In India, the problem of the occurrence of high As in groundwater has been observed in some areas of West Bengal located in the Indo-Gangetic delta plains affecting the districts of Nadia, Murshidabad, Malda, Bardhaman, Hugli, Howrah, North and South 24-Paraganas (<https://maps.wbphed.gov.in/arsenic/index.php> accessed on 12.08.2021). Water for domestic use is drawn from hand-pumped tubewells that usually tap the shallow aquifer within a depth of 50 m bgl and from deep wells (at depth > 80 m) fitted with hand-pump or motor-pump. Around 25% of wells tapping the shallow aquifer in Bengal Basin contain > 50 µg/L As (Sikdar et al. 2013). The deep Pleistocene aquifer is generally As-free, but at places it shows small but variable signs of being polluted with As at concentrations > 10 µg/L (Sikdar et al. 2013). Arsenic in deep aquifer was liberated in-situ by reduction of small amount of FeOOH in the grey Pleistocene sands by organic matter infiltrating from riverbeds during late Pleistocene or earliest Holocene times (McArthur et al. 2016).

Near Kolkata geogenic arsenic has been found in Rajarhat–Barasat area located in the east-northeastern side of the city (Sahu and Sikdar 2017). Chatterjee et al. (1993) and Sikdar (1996a, b) studying a case of As contamination of groundwater in Kolkata city, indicated that the source of arsenic in the confined groundwater at depth > 90 m bgl is

anthropogenic having concentrations > 10 µg/L (McArthur et al. 2018) in a few groundwater samples, which represents remnants of a pollution plume originating from an industrial site producing Paris Green ($\text{Cu}[\text{CH}_3\text{COO}]_2 \cdot 3\text{Cu}[\text{AsO}_2]_2$) between 1965 and 1985, an arsenical pesticide, located close to Tolly's Nala.

Groundwater chemistry

Two hundred eighty seven groundwater samples (Fig. 1) from different depths (32 to 229 m bgl) were considered for this study (Table S1) and the summary of the analysis is given in Table 1. The locations of the samples were recorded using a portable GPS device and are displayed in Fig. 1. Out of this, 201 water sample data of Kolkata city were taken from McArthur et al. (2018). The rest 86 samples were collected from the areas not covered by McArthur et al. (2018), and area to the west (Howrah) and south of Kolkata city (M-B area) in 2019 (Fig. 1c). These 86 samples were

analyzed for pH, cations, anions and As, and 16 samples for metals (Pb, Cu, Zn, Cr, Ni and Cd) in the laboratory of TÜV SÜD, India and National Institute of Roorkee, India. Total number of samples from Kolkata, Howrah and M-B area are 205, 58 and 24, respectively. Samples were collected from wells after purging in two 100 ml polythene bottles, one acidified in the field with 0.15 ml of 50% Analar® nitric acid for cation analysis and another unacidified for anion analysis. Analysis for all cations and anions were done by ion chromatography while metals were done by ICP-OES. TDS (mg/L) of the samples were calculated using the following formula given in APHA (2005):

$$\text{TDS} = 0.49 \times \text{HCO}_3 + \text{Na} + \text{K} + \text{Ca} + \text{Mg} + \text{Cl} + \text{SO}_4 + \text{NO}_3 + \text{F} \quad (1)$$

Table 1 Comparison of groundwater quality with drinking water standards, Indian (BIS 2015) and WHO (2017)

Sl. No	Parameters	No of samples	Minimum	Maximum	Mean	Indian standard	% compliance	WHO standard	% compliance
1	pH	106	6.9	8.4	7.5	6.5–8.5	100	7-Aug	89
2	TDS (mg/L)	287	176	3377	955	500	18	1000	66
3	Sodium [Na] (mg/L)	287	15	1116	192.4	–	–	200	66
4	Potassium [K] (mg/L)	287	1.1	76	6.3	–	–	–	–
5	Calcium [Ca] (mg/L)	287	4	323.6	106.1	75	39	75	39
6	Magnesium [Mg] (mg/L)	287	5	190	48.4	30	32	30	32
7	Chloride [Cl] / (mg/L)	287	17	1597.9	337.5	250	56	250	56
8	Bicarbonate [HCO_3] (mg/L)	287	101.7	884	491.6	–	–	–	–
9	Nitrate [NO_3] (mg/L)	287	0	21.9	1	45	100	50	100
10	Sulphate [SO_4] (mg/L)	286	0	203	21.7	200	99.7	250	100
11	Fluoride [F] (mg/L)	287	0	1	0.3	1	100	–	–
12	Bromide [Br] / (µg/L)	279	25	36,140	2123.4	–	–	–	–
13	Iron [Fe] (mg/L)	287	0	22.8	3	0.3	28	0.3	28
14	Arsenic [As] (µg/L)	287	0	79	3	10	94	10	94
15	Manganese [Mn] (mg/L)	287	0	2.8	0.3	0.1	36	0.1	36
16	Chromium [Cr] (µg/L)	223	0	11	0.8	50	100	50	100
17	Nickel [Ni] (µg/L)	221	0	8.5	0.6	20	100	20	100
18	Copper [Cu] (µg/L)	209	0	401.6	4.5	50	98	1000	100
19	Cadmium [Cd] (µg/L)	175	0	2.5	0.1	3	100	3	100
20	Lead [Pb] (µg/L)	223	0	47.6	2.8	10	96	10	96
21	Zinc [Zn] (µg/L)	16	4.2	5445.8	670.9	5000	94	3000	94
22	Carbonate [CO_3] (mg/L)	9	0	29	10	–	–	–	–

The analytical precision for the measurements of ions was checked by calculating the charge balance error (CBE) using the following formula:

$$CBE = \left\{ \left(\sum C - \sum A \right) / \left(\sum C + \sum A \right) \right\} \times 100 \quad (2)$$

where, $\sum C$ and $\sum A$ is the sum of concentrations of cations and anions, respectively, expressed in mill-equivalents/litre. If the CBE is within $\pm 10\%$, the analysis is assumed to be good.

Hydrochemical facies

The major cations (Ca^{2+} , Mg^{2+} , Na^+ and K^+) and anions (Cl^- , HCO_3^- and SO_4^{2-}) content of the 287 groundwater samples were plotted on the trilinear diagram of Piper (1944). Apart from the groundwater samples, three Hugli river water

samples of different seasons (McArthur et al. 2018) and one sea water sample (Garrels and Christ 1965) were also plotted on the Piper diagram (Fig. 3). A spatial hydrochemical facies map based on the analysis of 287 groundwater samples, collected during 2015 to 2019, was prepared using the Kriging method. This hydrochemical facies map was then compared with Fig. 8.7 of Sikdar (1996a) to understand whether any spatial variation of hydrochemical facies has occurred or not in the common area between the two study periods.

Principal component analysis

Principal component analysis (PCA) is a useful multivariate technique to understand the sources and influencing factors of hydrochemistry which has been used in several previous studies (Everest and Ozcan 2019; Kukrer and Mutlu 2019; Li et al. 2019; Shehzad et al. 2019; Wu et al. 2014). To understand the compositional data structure and to separate

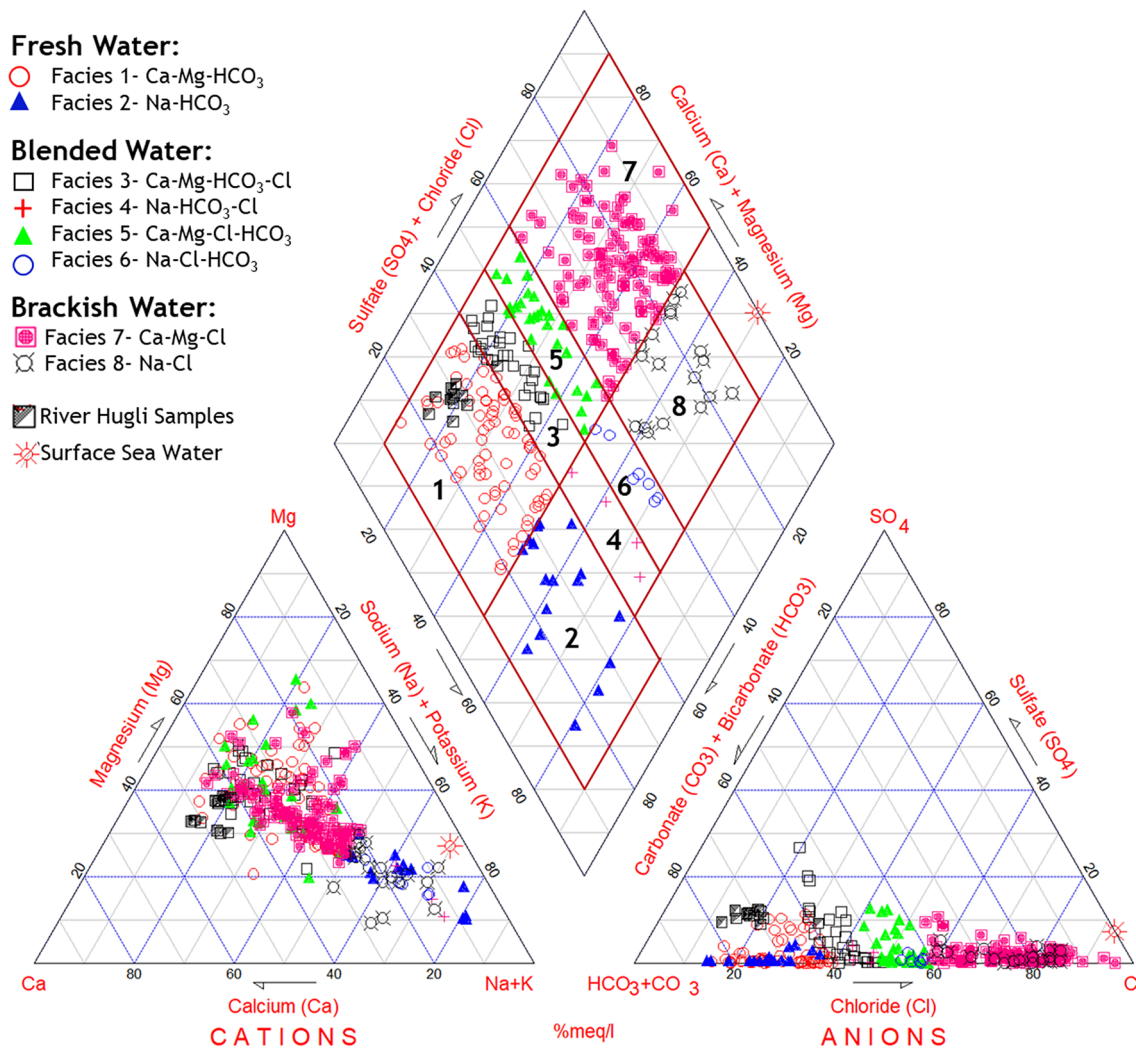


Fig. 3 Piper diagram showing the different hydrochemical facies

and estimate the relative importance of the factors controlling the chemical evolution of groundwater, Varimax-rotated R-mode factor analysis or PCA was carried out (Davis 1973) using 13 parameters (HCO_3^- , Cl^- , SO_4^{2-} , NO_3^- , Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Fe^{2+} , Mn^+ , F^- , As , Br^-) of 278 samples (Kolkata-204 samples, Howrah-53 samples, M-B Area-21 samples). Principal components are the Eigen vectors of a variance-covariance matrix. It transforms the original variables into some integrated variable, named principal components (PCs) (Bu et al. 2010; Ren et al. 2021). PCs are extracted as per the Kaiser criteria where the Eigen values of the correlation matrix are > 1 . The application of this criterion avoids any kind of necessary information loss during the transformation. The combined linear equation of the PCs can be expressed as (Wu et al. 2014):

$$y_i = c_{i1}x_1 + c_{i2}x_2 + \dots + c_{ip}x_p \tag{3}$$

where y is the PCs, c is the component loading, x is the origin variable, i is the component number and ranges from 1 to p , and p is the total number of variables.

A set of PC scores for each of the varimax-rotated factor PC axis, corresponding to individual samples was computed. Each of the PC axis or factors (with high loadings on one or more variables) represents an independent source of variation in the data matrix and may give some clues to genetic processes (Harman 1967).

Results

Hydrochemical facies

Table 1 shows the summary of chemical analysis of groundwater samples in the study area. Based on the Piper trilinear

diagram (Fig. 3) eight hydrochemical facies are identified. They are: Facies 1—Ca-Mg- HCO_3 , Facies 2—Na- HCO_3 , Facies 3—Ca-Mg- HCO_3 -Cl, Facies 4—Na- HCO_3 -Cl, Facies 5—Ca-Mg-Cl- HCO_3 , Facies 6—Na-Cl- HCO_3 , Facies 7—Ca-Mg-Cl and Facies 8—Na-Cl. The average composition of cations, anions and TDS of the eight hydrochemical facies are summarized in Table 2 and the spatial distribution and area covered by these hydrochemical facies are shown in Fig. 4. Facies 1 and 2 make up the freshwater category while Facies 7 and 8 belong to brackish water type. Facies 3 to 6 are categorized as blended water. The average composition of cations, anions and TDS of these three types of water are summarized in Table 2. Fresh, blended and brackish waters cover about 53 km², 186 km² and 171 km², respectively (Fig. 4, inset), excluding the area of 26 km² of River Hugli.

Fresh water

Facies 1 and 2 make up the freshwater category. Generally, the concentration of various parameters is lower than the desirable level of the Indian Standard (BIS 2015). This type of water has TDS values between 176 and 918 mg/L with an average value of 518 mg/L. The average HCO_3 content is 486 mg/L, much higher than the average Cl content of 69 mg/L. About 32% of the 287 samples belong to freshwater facies.

Blended water

Facies 3, 4, 5 and 6 belong to blended water type. Its overall quality is inferior to the freshwater type. The TDS value ranges from 303 to 1142 mg/L with an average of 722 mg/L. The average HCO_3 content is 521 mg/L and is close to that of fresh water, but the average Cl concentration (168 mg/L)

Table 2 Average composition of different hydrochemical facies

Groundwater type/hydrochemical facies	Average composition of hydrochemical facies						TDS (mg/L)
	Cations (percent equivalent)			Anions (percent equivalent)			
	Ca ²⁺	Mg ²⁺	N ⁺ + K ⁺	Cl ⁻	HCO ₃ ⁻	SO ₄ ²⁻	
Fresh water	31.4	15.0	53.7	12.3	86.0	1.7	518.4
Facies 1—Ca-Mg- HCO_3	35.0	16.7	48.3	12.5	85.6	1.9	498.2
Facies 2—Na- HCO_3	15.8	7.3	76.9	11.5	87.5	1.1	606.3
Blended water	35.0	16.1	48.9	23.1	73.0	3.9	721.8
Facies 3—Ca-Mg- HCO_3 -Cl	40.5	17.7	41.8	18.3	76.6	5.2	639.4
Facies 4—Na- HCO_3 -Cl	15.4	6.2	78.4	21.7	76.6	1.6	683.6
Facies 5—Ca-Mg-Cl- HCO_3	37.2	18.2	44.6	26.4	69.9	3.7	748.0
Facies 6—Na-Cl- HCO_3	15.8	6.8	77.4	30.1	69.1	0.8	968.5
Brackish water	30.5	13.5	56.0	52.3	45.3	2.4	1384.7
Facies 7—Ca-Mg-Cl	32.3	14.5	53.2	52.4	45.2	2.4	1348.0
Facies 8—Na-Cl	19.1	7.3	73.6	52.0	45.9	2.1	1611.2

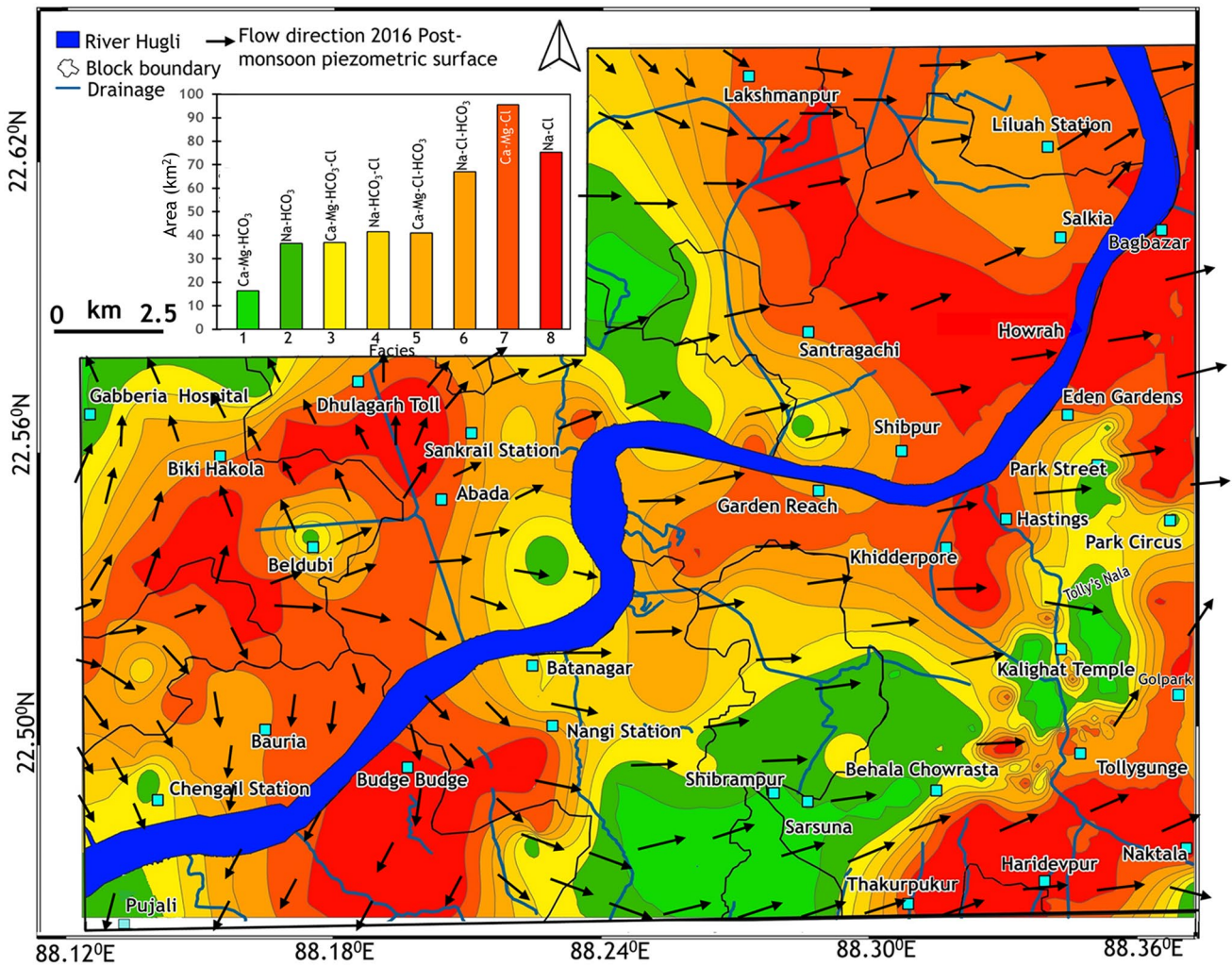


Fig. 4 Spatial distribution of different hydrochemical facies and area covered by different facies within the study area (inset). The arrows indicate the groundwater flow direction of 2016 post-monsoon period (after Banerjee and Sikdar 2021)

is more than double that of fresh water. About 23% of the 287 samples fall in the blended water category.

Brackish water

Facies 7 and 8 belong to brackish water type. The TDS value ranges from 355 to 3377 mg/L with an average of 1385 mg/L. The average HCO₃ content is 479 mg/L which is slightly lower than the value of fresh water, but the average Cl concentration (614 mg/L) increases sharply compared to that of the blended water type. About 45% of the 287 samples are under brackish water category.

The spatial distribution of different hydrochemical facies in the study area (Fig. 4) shows that fresh water occurs in some pockets in the south-western corner of Kolkata city near Behala Chowrasta-Sarsuna and north of Tollygunge area. Brackish water type occurs in the central and northern parts of Kolkata city, south of Tollygunge area and also in Budge

Budge. On the other side of River Hugli, brackish water occurs significantly in Howrah city and in the western part of study area surrounding Dhulagarh Toll. Figure 4 shows that the blended water covers the intermediate position between fresh and brackish water.

Principal component analysis

PCA has been carried out by computing 13 parameters of 278 groundwater samples. The outcome of the PC analysis (Table 3) reveals that cumulative eigen value of 4 loading axes is 67.18%. The first four factors (PC1 to PC4) were selected to represent the dominant hydrochemical processes, which have contributed significantly to the formation and chemistry of the present groundwater without suppressing much relevant information.

Table 3 Results of principal component analysis

Variables	Loading on PC axes			
	PC1	PC2	PC3	PC4
Na	0.481	0.468	0.012	0.554
K	0.093	0.273	0.623	0.056
Ca	0.88	0.105	0.149	0.109
Mg	0.834	0.254	0.17	0.097
Fe	0.084	-0.569	0.208	0.295
Mn	0.599	-0.075	-0.248	0.361
Cl	0.799	0.433	0.037	0.26
HCO ₃	0.039	0.039	0.167	0.864
SO ₄	0.328	-0.16	0.694	0.099
NO ₃	0.054	0.798	0.065	0.128
F	-0.699	0.119	-0.174	0.214
As	-0.074	-0.203	0.782	0.044
Br	0.278	0.755	-0.034	0.159
% eigen value	33.424	15.277	10.268	8.212
Cum%	33.424	48.701	58.97	67.182
Total initial eigen values	4.345	1.986	1.335	1.068

Bold indicates variables with significant loadings

Discussions

Stratigraphic short cut for aquifer contamination

The average thickness of the upper confining layer of clay/silty clay within the study area is 26.3 m (Banerjee and Sikdar 2021). But the upper confining bed is either absent or thinned down to < 10 m along the course of Tolly's Nala, Dhulagarh Toll and few areas in M-B (Fig. 5). The upper clay/silty clay bed thickness is also < 10 m where River Hugli detours from N-S direction to E-W direction (Fig. 5), near Shibpur and Hastings. The low-lying areas of M-B have number of small Nalas (channels) or remnant of past channels (Bandyopadhyay 1996), which debouches into River Hugli. The Holocene clay and the LGMP are absent and grey sands are present right below these channel beds, which provide a stratigraphic short cut for the contaminated surface water and wastewater to infiltrate into the underlying Holocene-Late Pleistocene aquifer (McArthur et al. 2018) resulting in groundwater quality deterioration.

Anthropogenic As and Pb, exceeding the acceptable limit (AL), have been detected at places in the area adjacent to the Tolly's Nala (Fig. 5) (Sikdar et al. 2001; McArthur et al. 2018). Pb > 10 µg/L and Cu > 50 µg/L have also been identified near Bagbazar. Copper and Pb above AL have been found in a well near Pujali (in the extreme south western corner; Fig. 5) where a thermal power plant and a coal handling plant are located. Interestingly, Cu and Zn above AL, and Pb and Cd above 50% of their respective AL have been also detected near Chengail which is located on the other

side of River Hugli and exactly opposite to Pujali indicating the movement of industrial contaminants in the aquifer system beneath the bed of River Hugli. Copper, Pb and As having concentrations between ≥ 50% of AL and below AL are detected at 1, 18 and 7 number of groundwater samples, respectively (Fig. 5). Though the upper aquitard thickness is < 10 m, but no heavy metals were detected in two groundwater samples collected from 80 m depth within a decade old industrial area near Dhulagarh in peri-urban Howrah (Fig. 5). The contaminants are possibly yet to migrate to the strainer depths of wells below 80 m bgl.

Mixing of fresh and brackish water

Groundwater flow pattern from the mid-eighties (Fig. 3 of Sikdar et al. 2001; Fig. 6 of Banerjee and Sikdar 2021) indicates a closed flow system with groundwater flowing from all directions towards the groundwater trough. Since then, the brackish groundwater from Howrah had moved into the aquifer beneath Kolkata and had deteriorated the quality (Sikdar et al. 2001; Sahu and Sikdar 2008; Sahu et al. 2016), confirmed by the fingerprinting of the hydrochemical facies. The groundwater flow directions of 2016 post-monsoon overlaid on the hydrochemical facies map (Fig. 4) indicates that the blended water has evolved due to the hydraulic mixing of fresh and brackish water within the aquifer.

The common area of the two studies (Sikdar 1996a, b and present study) is 159 km² including the area of River Hugli (8.57 km²), covering parts of Kolkata and Howrah cities (Fig. 6). Figure 6a (modified after Fig. 8.7 of Sikdar 1996a), shows that the groundwater below Howrah city was mainly brackish of Facies 7 and 8; all other facies occurred as small pockets. The hydrochemical facies map of the present study (Fig. 6b) clearly shows that the aquifer of Howrah city is still characterized by brackish water (Facies 7 and 8). Some small pockets of blended water (Facies 3, 4, 5 and 6) occur at the periphery indicating freshwater flow from the west (Fig. 4). Similarly, in Kolkata, a comparison between the two hydrochemical facies maps clearly highlight that the fresh water has deteriorated to blended water and further to brackish water in only two and half decades, confirming the influence of over-pumping on the groundwater chemistry. The aquifer beneath Howrah city still holds the brackish water while there is a possibility of gradual improvement of water quality due to the freshwater flow from the west (Fig. 4). The south-western part of the common area on both the sides of Tolly's Nala had fresh water during mid 90's. Within two and half decades, the freshwater has changed to blended and brackish water types with some remnants of fresh water surrounding Behala and Kalighat (Fig. 6).

The hydrochemical facies map prepared from the 287 groundwater samples collected during the period 2015 to

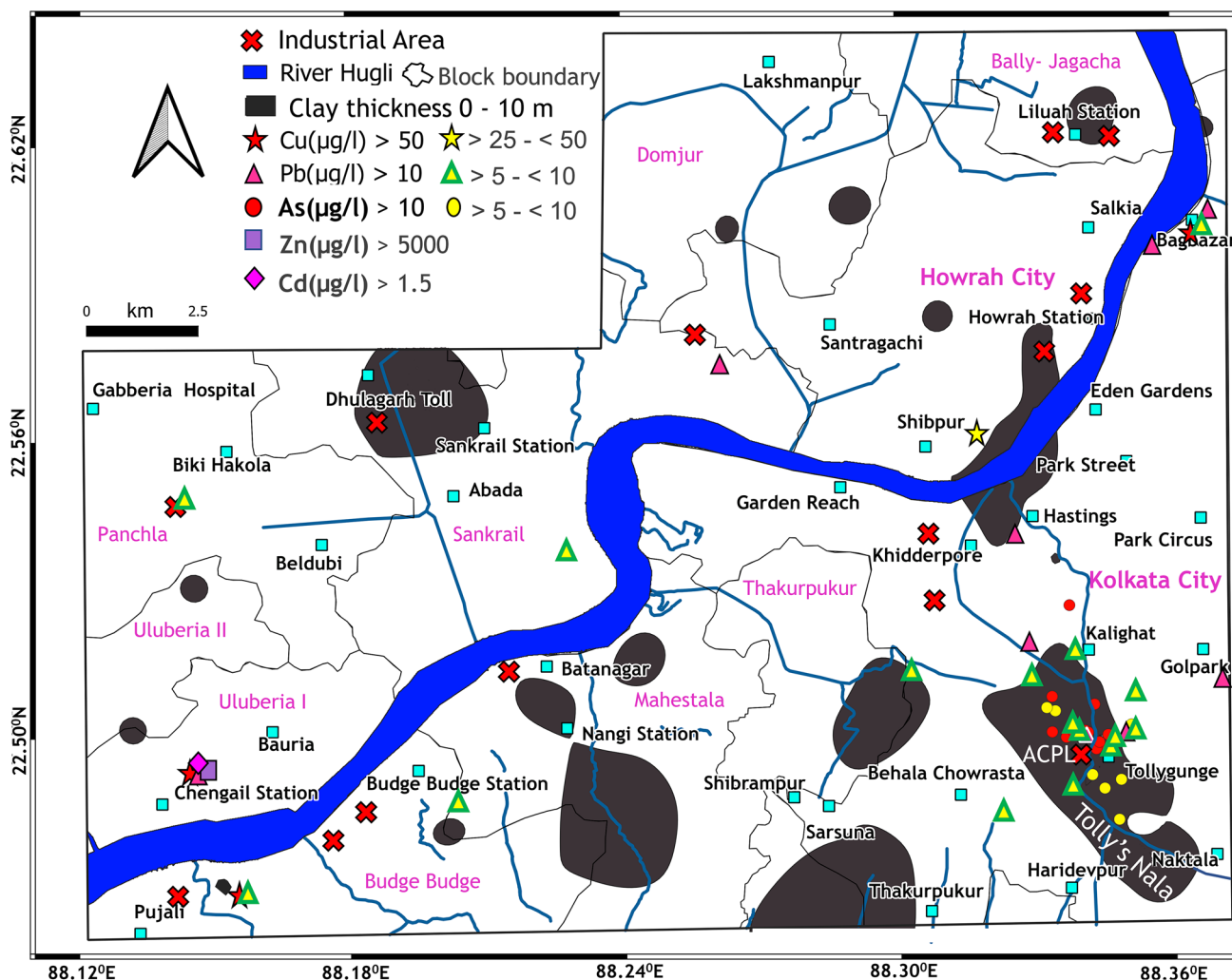


Fig. 5 Locations of wells having different heavy metal contamination above acceptable limit and 50% of the acceptable limit in the groundwater

2019, has been compared with Fig. 8.7 of Sikdar et al. (1996a) to understand whether any spatial variation of hydrochemical facies has occurred or not in the common area between the two study periods. The comparison shows that, in 1996, the area covered by freshwater was 77.64 km² while the same has been reduced to mere 19.82 km² resulting in a reduction of about 75%. This reduction of area has been compensated by the increase of blended water from 18.74 to 51.75 km² and of brackish water from 54 km² to 78.88 km² in the common area (Fig. 7).

Principal component analysis (PCA)

PC1 vs PC4

The PC1 and PC4 loading axes represent the Cl and HCO₃ dependent facies, respectively (Table 3). The plots of PC1 vs PC4 scores (Fig. 8), which together account for 41.64% of

the total variability, show two distinct clusters—(a) brackish water type in Quadrants I and II, and (b) freshwater type in Quadrants III and IV. Blended water encircles mainly freshwater samples and is populated in Quadrants III and IV. A small number of blended water (Facies 5: Ca-Mg-Cl-HCO₃, Facies 6: Na-Cl-HCO₃) samples occur with brackish water in Quadrant I, whereas a few groundwater samples of Facies 3: Ca-Mg-HCO₃-Cl, Facies 4: Na-HCO₃-Cl occur with brackish water in Quadrant II. The spatial distribution of hydrochemical facies map (Fig. 4) also replicates the outcome of PCA analysis and so blended water occurs between fresh and brackish water because of the hydraulic mixing within the aquifer system.

PC1 vs PC3

PC1 and PC3 axes together account for 43.69% of the total variability in the data structure. PC3 axis has high positive

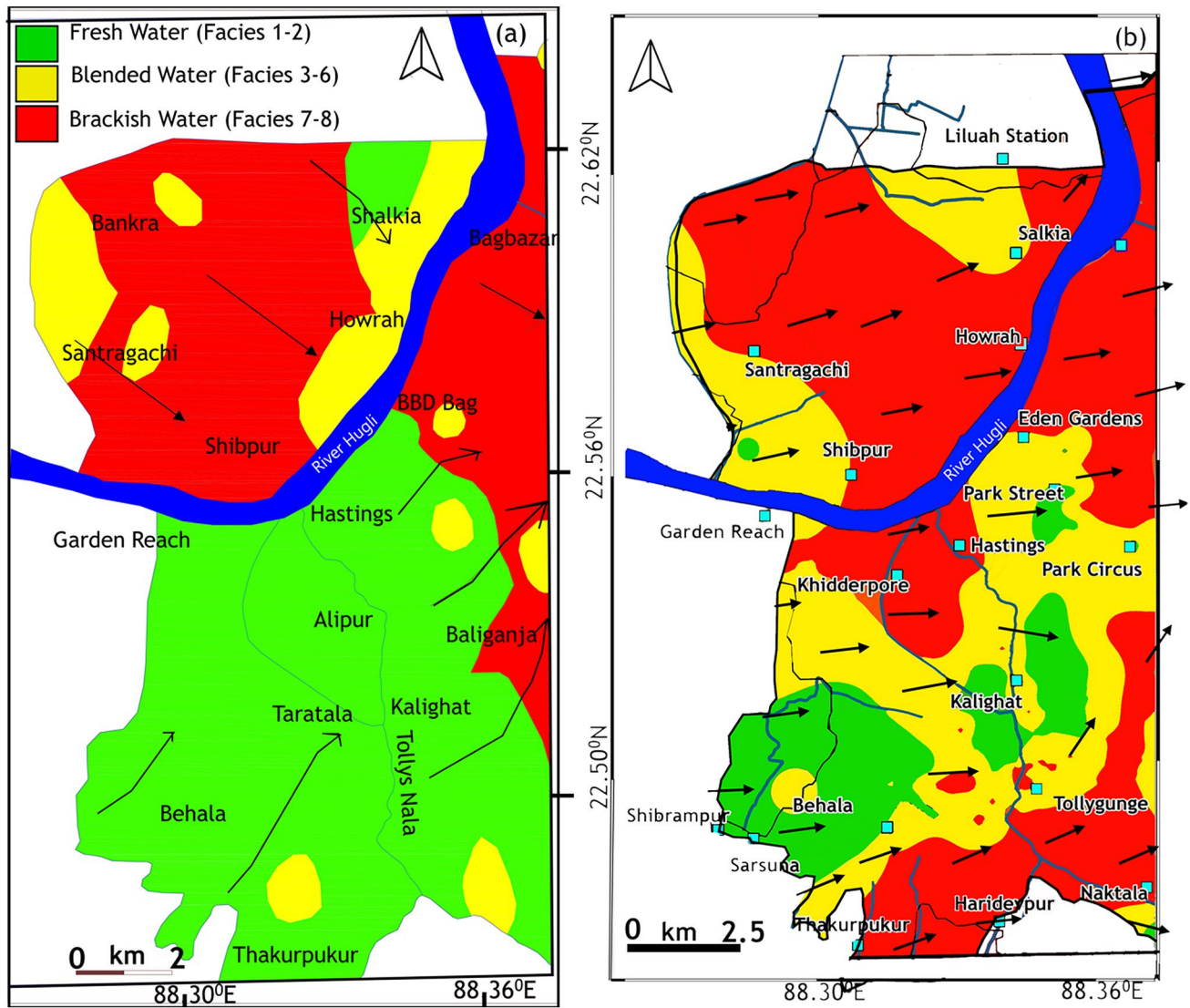


Fig. 6 Area covered by different facies—a comparison for the common study area of **a** Sikdar (1996a) and **b** present study. The arrows indicate the groundwater flow directions

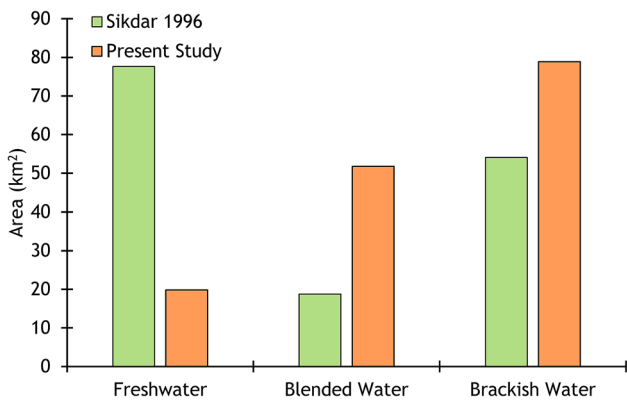


Fig. 7 Area covered by different facies—a comparison for the common study area of Sikdar (1996a) and present study

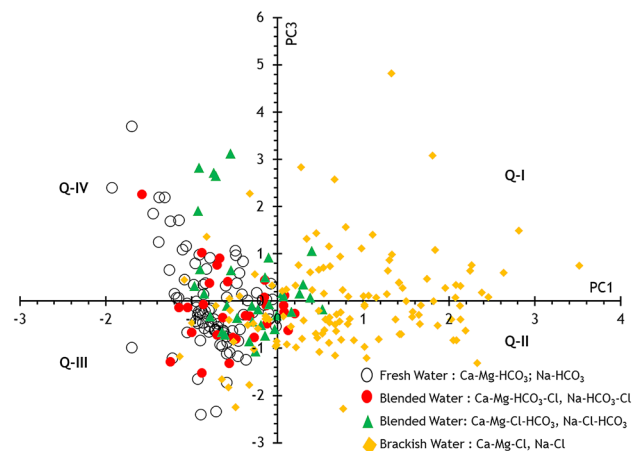


Fig. 8 Plots of PC scores—PC1 vs PC4

loadings on As (0.782) and SO₄ (0.694) and low negative loading on Mn (-0.248). The SO₄ vs Cl plots (Fig. 9) show that the salt-corrected concentrations of SO₄ are in deficit of upto 105 mg/L and 20% of the groundwater samples fall above the excess-SO₄ line. The removal of sulphate from groundwater takes place because of reduction of FeS₂ and H₂S (McArthur et al. 2018). Removal of sulphate from groundwater also sequesters the As in groundwater into the neo-formed FeS. This is clearly demonstrated by plotting separately the samples with ≥ 10 µg/L As in PC1 vs PC3 plots with reduced-SO₄ and excess-SO₄ (Fig. 10). Out of the 16 samples containing As ≥ 10 µg/L only 3 samples fall on the plots of PC1 vs PC3 scores of the samples with reduced-SO₄ (Fig. 10a). The rest 13 samples fall on the plots of PC1 vs PC3 scores of the samples with excess-SO₄ (Fig. 10b).

The groundwater samples with > 10 µg/L As cluster around Aceto Chemical Private Limited (ACPL) site located close to the Tolly’s Nala (Fig. 5). This As is a part of the past anthropogenic contamination from solid waste dump within the boundary of ACPL, a factory producing

Paris Green (Cu[CH₃COO]₂·3Cu[AsO₂]₂), an arsenical pesticide between 1965 and 1985 (Sikdar 1996a, b; McArthur et al. 2018). In 1993, As concentration was reported up to 39,000 µg/L (Chatterjee et al. 1993). Since then, the concentration of As has greatly reduced and presently the maximum concentration of As is 79 µg/L (KOL71 sample of McArthur et al. 2018). Dilution and dispersion of As and sorption of As in neo-formed pyrite in the late-Pleistocene sands have decreased the As concentration in the groundwater (McArthur et al. 2018). The microbial oxidation of the acetate or acetic acid of the effluent water of ACPL (Fig. 5) had influenced the reduction of SO₄ by the range of 105 mg/L and have also contributed to the low As concentration by attenuation of As during SO₄ reduction (McArthur et al. 2018). In PC3, the negative loading on Mn and positive loading on As indicates that As will not occur in groundwater until Mn (in sediments) reduction is completed. Therefore, wherever there is a Mn front in groundwater, As will not appear in groundwater. This reduction is driven by dissolved organic carbon (DOC) obtained from river-bank infiltration or infiltrated wastewater (McArthur et al. 2012).

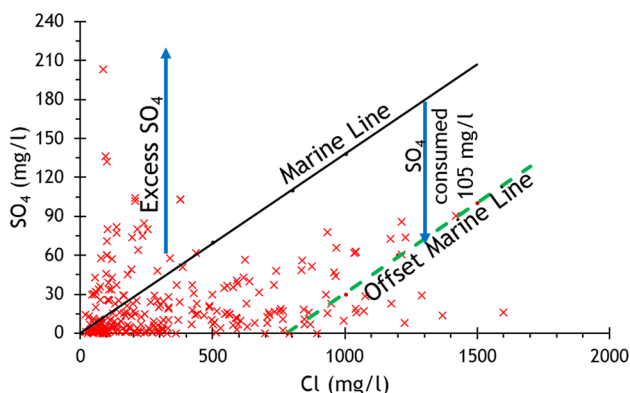


Fig. 9 cross plot of SO₄ and Cl for the differentiation of excess and reduced SO₄

PC1 vs PC2

PC2 shows high loading on Fe, NO₃ and Br. PC1 and PC2 axis together accounts for 48.70% of the total variability in the data structure. Since there are low agricultural activities within the study area, NO₃ in groundwater can only come from the wastewater (sewage and sullage) (McArthur et al. 2012a) recharge at places. Cl/Br mass ratios in groundwater are used to show the extent to which groundwater in the study area is contaminated by wastewater (Katz et al. 2011; McArthur et al. 2012a, 2016, 2018). About 64% samples of Kolkata, 17% samples of Howrah and 33% samples of M-B area occur above the marine mixing line whose Cl/Br is ≥ 288 (Fig. 12b). About 36% samples of Kolkata,

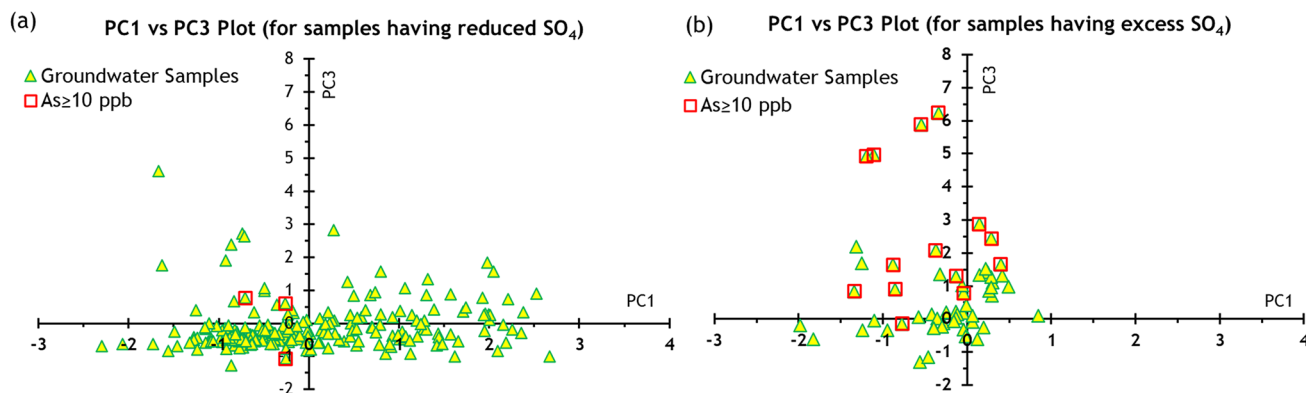


Fig. 10 Plots of PC scores—PC1 vs PC3 **a** for the samples having reduced-SO₄ with As ≥ 10 µg/L, **b** for the samples excess-SO₄ with As ≥ 10 µg/L

86% samples of Howrah and 67% samples of M-B area occur below the marine mixing line whose Cl/Br is < 288 (Figs. 11, 12a). The excess Br of these groundwater samples is derived from organic matter degradation. Therefore, the groundwater of Kolkata city is more contaminated by wastewater while the impact of organic degradation is more prominent in the rest of the study area. The groundwater samples having Cl/Br mass ratio above 308 (the upper limit of 288 ± 20 to allow for analytical uncertainty, McArthur et al. 2018), mostly cluster surrounding the Tolly's Nala and in Park Street-Park Circus area (Fig. 11), except few stray samples in the other parts of the study area. Samples having $\text{Cl/Br} > 308$ are contaminated by Cl of anthropogenic origin. In the area surrounding the Tolly's Nala, the upper confining clay bed is absent allowing easy infiltration of wastewater into the aquifer. A study on stable isotopes of groundwater of Kolkata city (McArthur et al. 2018) showed that the least depleted $\delta^{18}\text{O}$ of River Hugli water is -6.1‰ while

the volume-weighted mean annual value for $\delta^{18}\text{O}$ of local rainfall is -6.2‰ . More depleted groundwater than this are found in two clusters, located close to Tolly's Nala. The occurrence of these two clusters, close to Tolly's Nala, suggests direct infiltration of Nala water into the late Pleistocene aquifer. In Park Street-Park Circus area where the core of the groundwater trough exists (Sikdar 2000; Sikdar et al. 2001; Banerjee and Sikdar 2021), the presence of wastewater may be attributed to vertical leakage of water from the upper aquitard into the aquifer due to increase in the lithostatic pressure in both aquitard and aquifer as a result of heavy withdrawal of groundwater in that area.

Evolution of groundwater chemistry

In late Quaternary time, the south Bengal Basin including the study area was inundated with brackish to marine water (Sen and Banerjee 1990; Barui and Chanda 1992; Hait et al.

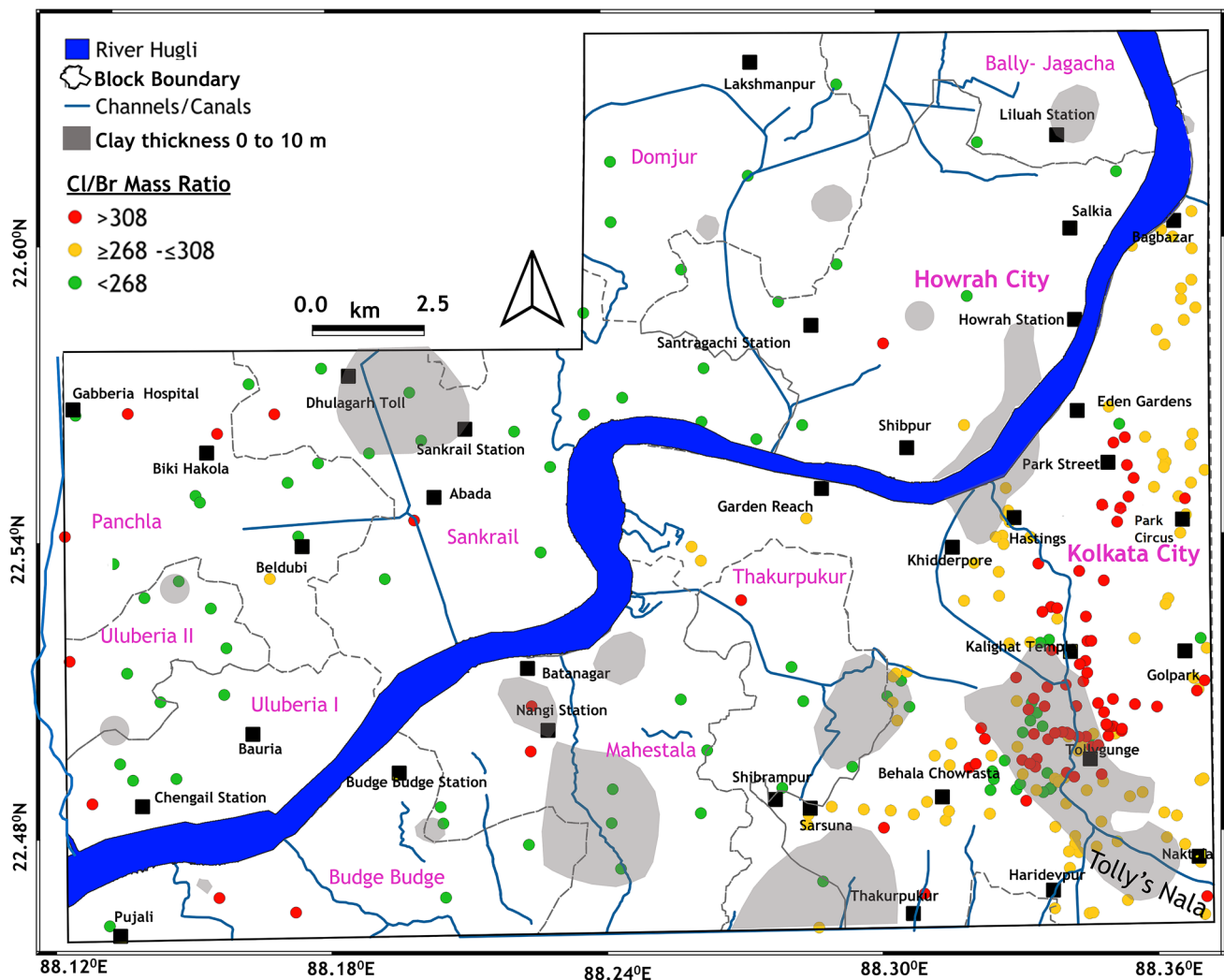


Fig. 11 Distribution of Cl/Br of groundwater, showing that most Cl/Br values > 308 occur close to Tolly's Nala

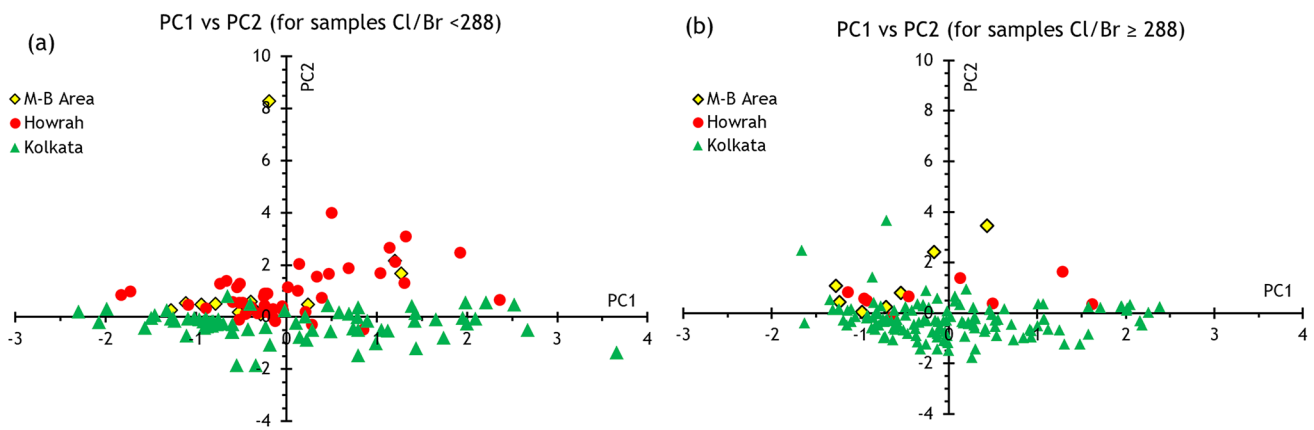


Fig. 12 Plots of PC scores—PC1 vs PC2 **a** for the samples having Cl/Br < 288, **b** for the samples Cl/Br ≥ 288

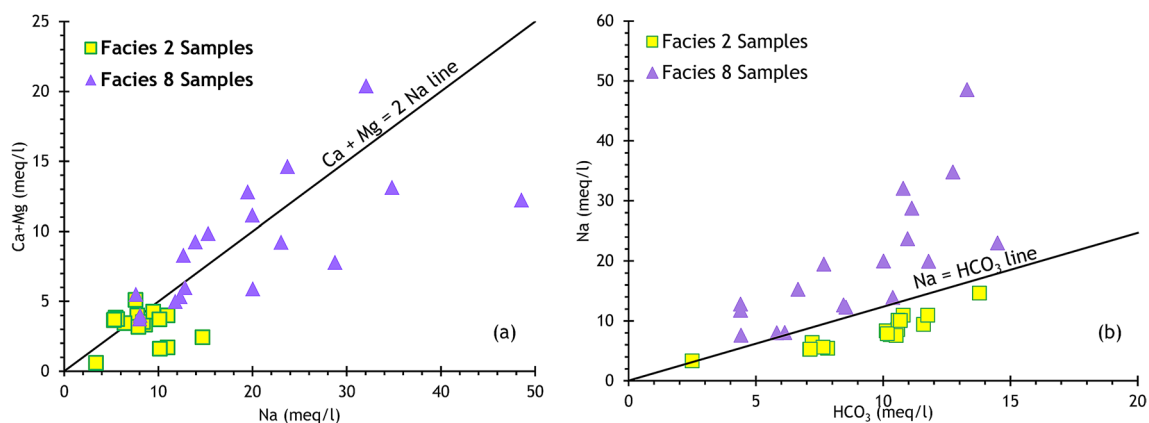
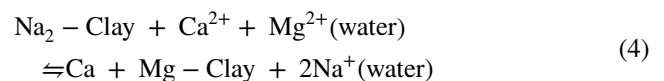


Fig. 13 Scatter plot of **a** Ca + Mg vs Na; **b** Na vs HCO₃ for Facies 2 and Facies 8 water samples

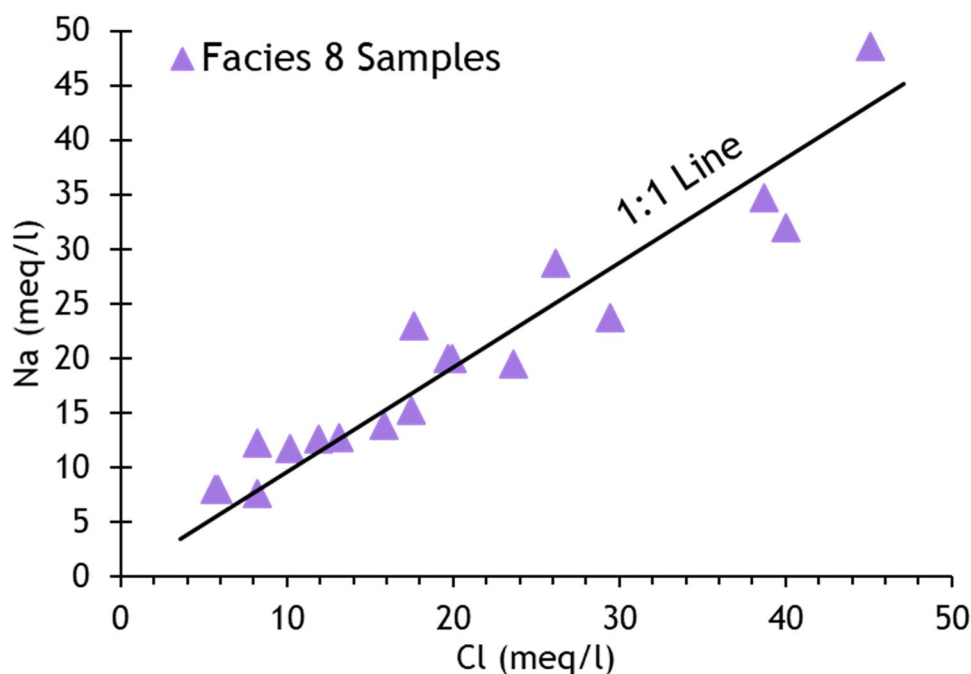
1994a, b, 1996; Sikdar et al. 2001). Therefore, the marine water having high-Cl content was probably entrapped during syn-sedimentation which was further modified in the aquifer under confined condition. Freshwater flushing from the recharge zone, which occurs further north of the present study area (Sikdar et al. 2001), had diluted the entrapped marine water. At places, dilution had also taken place from direct recharge of meteoric water where the upper confining clay/silty clay bed is absent or very thin (Fig. 5). This mixing produced the present-day brackish water. The mixing was non-uniform and is proved by the wide range of chloride content (127 to 1598 mg/L) in the present-day brackish water.

Occurrence of Facies 2 (Na-HCO₃ type) encircling small patches of Facies 1 (Ca-Mg-HCO₃ type) (Fig. 4) indicates active freshwater flushing in the aquifer system where ion exchange process takes place (Sikdar et al. 2001). The Ca + Mg vs Na plot (Fig. 13a) explains the presence of Ca²⁺ and Mg²⁺ in recharging water while Na⁺ gets absorbed in clay lenses. The reversible reaction is as follows



Na vs HCO₃ plots (Fig. 13b) show that Facies 2 groundwater samples fall below the Na = HCO₃ line, i.e., towards the side of HCO₃ axis while Facies 8 samples plot above the equiline. This has happened due to the carbonate alkalinity being balanced by the alkaline earth material. The occurrence of Facies 8 can be explained by restricted groundwater movement, no freshwater flushing, and over-exploitation of groundwater from the aquifer (Sikdar et al. 2001). The Na vs Cl plot (Fig. 14) shows that Facies 8 groundwater samples fall close to the 1:1 line; indicating active mineral dissolution mechanism within the aquifer (cf. Llyod and Heathcote 1985; Sikdar et al. 2001). Facies 7 groundwater samples have more Ca and Mg than Na with Cl. The base exchange reversible reactions within the aquifer matrix resulted in the formation of the present-day brackish water. Na⁻ ion of groundwater was exchanged by Ca⁻ and Mg⁻ ions of the

Fig. 14 Plot of Sodium vs chloride line for Facies 8 water samples



aquifer sediments. Hence Facies 7 occurs surrounding the Facies 8 (Fig. 4).

The groundwater trough of central Kolkata of mid-90's has increased by 266 km² at the rate of 8.60 km²/year (Banerjee and Sikdar 2021). The rate of expansion of groundwater trough between 1985 and 1994, 1994 and 2003, 2003 and 2016 were 2.03 km²/year, 9.14 km²/year and 12.73 km²/year, respectively. As a result, in 2016 the groundwater flows regionally from west to east (Fig. 4) that is from Howrah to Kolkata. The fingerprinting of the hydrochemical facies clearly reveals that the brackish water in and around Howrah city has moved into Kolkata's aquifer and changed the freshwater of south-central Kolkata into blended water and to some extent into brackish water. The Facies 7 (Ca–Mg–Cl) type of water has converted into Na–HCO₃ facies due to ion exchange process. The overall deterioration has taken place within the last two and half decades. This conclusively proves the predicted conclusion of Sikdar et al. (2001) that the native fresh groundwater of south western part of Kolkata will mix with the brackish water in the eastern half of the city due to the north-east and north ward flow directions and thus the fresh water (Facies 1) of south western part of Kolkata has changed into fresh water (Facies 2), blended water and brackish water.

Conclusions

The thickness of upper confining clay bed is extremely heterogeneous in nature and at places the clay bed is absent. Through these stratigraphic short cuts, industrial wastewater contaminated with toxic elements has already percolated down to the aquifer and deteriorated the groundwater at places. Anthropogenic As ≥ 10 $\mu\text{g/L}$ near the Tolly's Nala has been detected in several wells. Influence of wastewater from sewage and sullage having Cl/Br > 308 has also been detected in 30.5% wells, heavy metals like Cu > 50 $\mu\text{g/L}$ and Pb > 10 $\mu\text{g/L}$ have been detected in three and nine wells, respectively in the study area.

Due to the exponential growth of urbanization all across the area, the piezometric surface has lowered at the rate of 0.13 to 0.37 m/year. Through hydrochemical fingerprinting analysis it can be concluded that lowering of the piezometric surface has accelerated the deterioration of groundwater quality by mixing of groundwater of different facies and increasing the salinity. Therefore, with the present rate of groundwater abstraction, blended water may change into brackish water and the small patches of fresh water may change into blended water. Thus, the groundwater of the study area maybe completely devoid of freshwater and may be unsuitable for drinking in the near future. Stable and radioactive isotope studies and numerical groundwater modelling can be carried out as continuation of the present research work.

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Declarations

Competing interests The authors declare that they have no conflict of interest.

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