



Hydrochemical systematics and isotope ($\delta^{18}\text{O}$, δD and ^3H) variations of aquifer system of southern Bengal Basin: implications for groundwater pollution

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Abstract

Hydrogeological, hydrochemical and isotopic traits of the groundwater in the Quaternary aquifer system in an urban-periurban locality within and encircling the Kolkata-Howrah twin city in the south Bengal Basin have been synthesised to explain the present- and paleo-hydrological processes, surface and groundwater interaction and mixing dynamics of contamination of groundwater. Rock-weathering, evaporation, ion-exchange and active mineral dissolution are the key processes commanding the groundwater chemistry. Freshwater flushing from the recharge zones had thinned the entrapped sea water which has generated the present-day brackish water by a non-uniform fusion. The best-fit line of the plots of δD and $\delta^{18}\text{O}$ of groundwater samples displays a slope lower than that of local meteoric water line (LMWL) and global meteoric water line (GMWL) which hints that isotopic constitution of the groundwater of the present area is primarily formed by evaporation before or in the recharging process. A wide range of $\delta^{18}\text{O}$ values in groundwater suggests that these waters are not blended enough to remove dissimilarities in isotope configuration of recharge water. This also suggests that many groundwaters are a result of mixing of present-day recharge and an older integrant recharged under previously cooler climatic conditions. The groundwater samples are more depleted of oxygen at the shallower level. The depleted samples cluster around the *Tolly's nala (canal)* where upper aquitard is missing or < 10-m thick. The tritium values range between 0.70 and 15.02 which indicate the occurrence of 'sub-modern', 'a mix of modern and sub-modern water' and 'modern water'. It indicates mingling of isotope-depleted water from the Hugli River by means of *Tolly's canal* with relatively less-depleted groundwater of Kolkata's late Pleistocene aquifer. The tritium values and Cl/Br ratio of groundwater samples adjoining *Tolly's canal* and elsewhere refer the direct infiltration of 'modern wastewater and freshwater' which mixes with the 'sub-modern water' in the aquifer system.

Keywords Groundwater · Hydrological processes · Wastewater · Stable and radioactive isotopes · Recharge sources · Bengal Basin

Introduction

The demand for groundwater in India and elsewhere is increasing by leaps and bounds owing to the rising population, industrialisation, unpredictable rainfall and change of climate (Banerjee and Sikdar 2022a, 2020; Battista

and de LietoVollaro 2017; Sahu and Sikdar 2017; Yan et al. 2016; Howard and Gelo 2002). The increased exploitation of groundwater is leading to declining groundwater levels, land subsidence and water quality degradation from anthropogenic and geogenic sources (Sikdar et al. 2022; Banerjee and Sikdar 2022a, b; Krishana et al. 2021; Galloway and Burbey 2011; Kagabu et al. 2011; Hosono et al. 2011; McDonald et al. 2011; Hoque et al. 2007; Morris et al. 2003). Isotopic signatures are widely used for analysis of hydrological processes like transport, recharge and discharge areas, and mixing mechanism since they give a conventional fingerprint which is inherent to the water molecules (Li et al. 2014; Criss 1999; Clark and Fritz 1997; Gat 1996; Gat and Gonfiantini 1981). Furthermore, the isotope (δD and $\delta^{18}\text{O}$) composition of water at various locations is influenced by

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climatic activities that reveal its origin (Li et al. 2014; Clark and Fritz 1997). Similar methods were employed in Dhaka, Bangladesh (Hoque et al. 2014), sections of south Kolkata, India (McArthur et al. 2018) and Delhi, India (Kumar et al. 2011) to locate recharge zones (Sikdar and Sahu 2009).

The South Bengal Basin's Quaternary aquifer, which is in the lower deltaic plain of Ganga River, is severely stressed because of unregulated abstraction of subsurface water required for growing population and urbanisation (Banerjee and Sikdar 2022a, b, 2021; Sikdar et al. 2001). The area with megacities, urban-peri-urban areas, agrarian economy and groundwater-dependant industries cannot be sustained if the problems of groundwater in the area are overlooked and undermined.

The aquifer system lying under the two metropolis of south Bengal Basin—Kolkata-Howrah and their surrounding peri-urban areas is experiencing several problems, like (i) declining water level, (ii) land subsidence, (iii) change of groundwater flow direction, (iv) water quality deterioration, (v) salinisation of freshwater, (vi) at places, aquifer changing from confined nature to unconfined and (vii) expansion of groundwater depression (Sikdar et al. 2022; Banerjee and Sikdar 2022a, b, 2021; McArthur et al. 2018). The piezometric surface lying within the extent of the study area has dropped by 34 cm in 2016 due to the groundwater overdraft of $408 \times 10^3 \text{ m}^3/\text{day}$. The water level has been found to drop by 13–37 cm/year on a long-term basis (Sikdar et al. 2022; Banerjee and Sikdar 2021). As a result, Kolkata and Howrah are on the verge of a catastrophic water crisis (Sikdar et al. 2022; Banerjee and Sikdar 2022a, b, 2021; McArthur et al. 2018; Mukherjee et al. 2018; Sahu and Sikdar 2017, 2011; Sikdar et al. 2001; Sikdar 1996a).

Stable isotopes, δD (D/H) and $\delta^{18}\text{O}$ ($^{18}\text{O}/^{16}\text{O}$) and radioactive isotope, tritium (^3H) are useful to study the present and paleo-hydrological processes and surface water-groundwater interaction (Bahir and Misdaq 2007; D'Alessandro et al. 2004; Bourg et al. 2001). Hydrogeological processes and identification of groundwater recharge source have been studied by different researchers by analysing environmental tracers (Meredith et al. 2013; Gonfiantini et al. 1998) including Indo-Gangetic basin (O'Keeffe et al. 2020; Lapworth et al. 2020, 2017; Bonsor et al. 2017; MacDonald et al. 2016; Kumar et al. 2011). However, published works on $\delta^{18}\text{O}$, $\delta^2\text{H}$ and ^3H distributions in the Bengal Basin are scanty. Several researchers have applied stable isotope techniques to understand hydrogeochemical processes of the aquifer in Bengal Basin (Sikdar and Sahu 2009; Sikdar and Chakraborty 2008; Klump et al. 2006; Harvey et al. 2005; Stüben et al. 2003; Basu et al. 2002; Aggarwal et al. 2000; Shivanna et al. 1999) and in Kolkata city (McArthur et al. 2018) by using isotope signature but in a piecemeal manner.

Studies of stable isotope signatures along with the identification of recharge areas for Kolkata, one of the largest

megacities of South Bengal Basin (McArthur et al. 2018), are limited. Comprehensive research related to recharge mechanism, groundwater mixing dynamics, wastewater contamination in view of the analysis of radioactive isotope—tritium, for the densely populated Kolkata city, are scanty. Similar studies of stable and radioactive isotopes are available (Sikdar and Sahu 2009) in and around East Calcutta Wetland (ECW), lying adjacent to the eastern periphery of the city limit. Such studies in and around the rural and peri-urban areas of Howrah city and blocks of Howrah district, Mahestala-Budge Budge areas, have never been carried out. The increased groundwater requirement due to rapid urbanisation and industrialisation of these rural and peri-urban areas demand such kind of scientific studies for understanding the groundwater dynamics. The basin scale studies in Bengal Basin were carried out by several researchers and elaborated the variation on stable and radioactive isotopes in groundwater with respect to depth of the aquifer, recharge areas, season. But the variation in the isotope values, for smaller heterogenous complex hydrogeological system beneath the twin cities and its' peri-urban areas in South Bengal Basin has never been examined. This research paper attempts to find the answers these novel questions.

Hence, objectives of this study are (a) to use stable and radioactive isotopes combined with hydrogeological and hydrochemical information for better understanding of groundwater system dynamics in the complex groundwater system beneath the world's largest delta, (b) to use tritium (^3H) dating of groundwater for identifying the recharge zones for groundwater and potentially toxic elements, (c) to explain the present hydrological processes, surface and groundwater interaction and mixing dynamics of contamination of groundwater, (d) to improve the overall knowledge and understanding in the relevant hydrogeological field for the megacities of the south Bengal Basin as well as the adjacent rural and peri-urban areas.

Study area

The research area is 436 km^2 located on both banks of the meandering Hugli river in the south Bengal basin. The area includes the twin cities Kolkata and Howrah, and their peri-urban areas in South 24-Parganas and Howrah districts (Fig. 1). The increase of built-up area between 1985 and 2016 is about 19% (109 to 192 km^2) which has been nearly compensated by the reduction of vegetation cover by 11%, agricultural land by 7% and waterbodies by 1.47% in the study area (Banerjee and Sikdar 2021). Groundwater is abstracted from wells where a screen is mainly placed at 80 to 140 m bgl and between 180 and 240 m bgl at places (Sikdar et al. 2022; Banerjee and Sikdar 2022a, 2021).

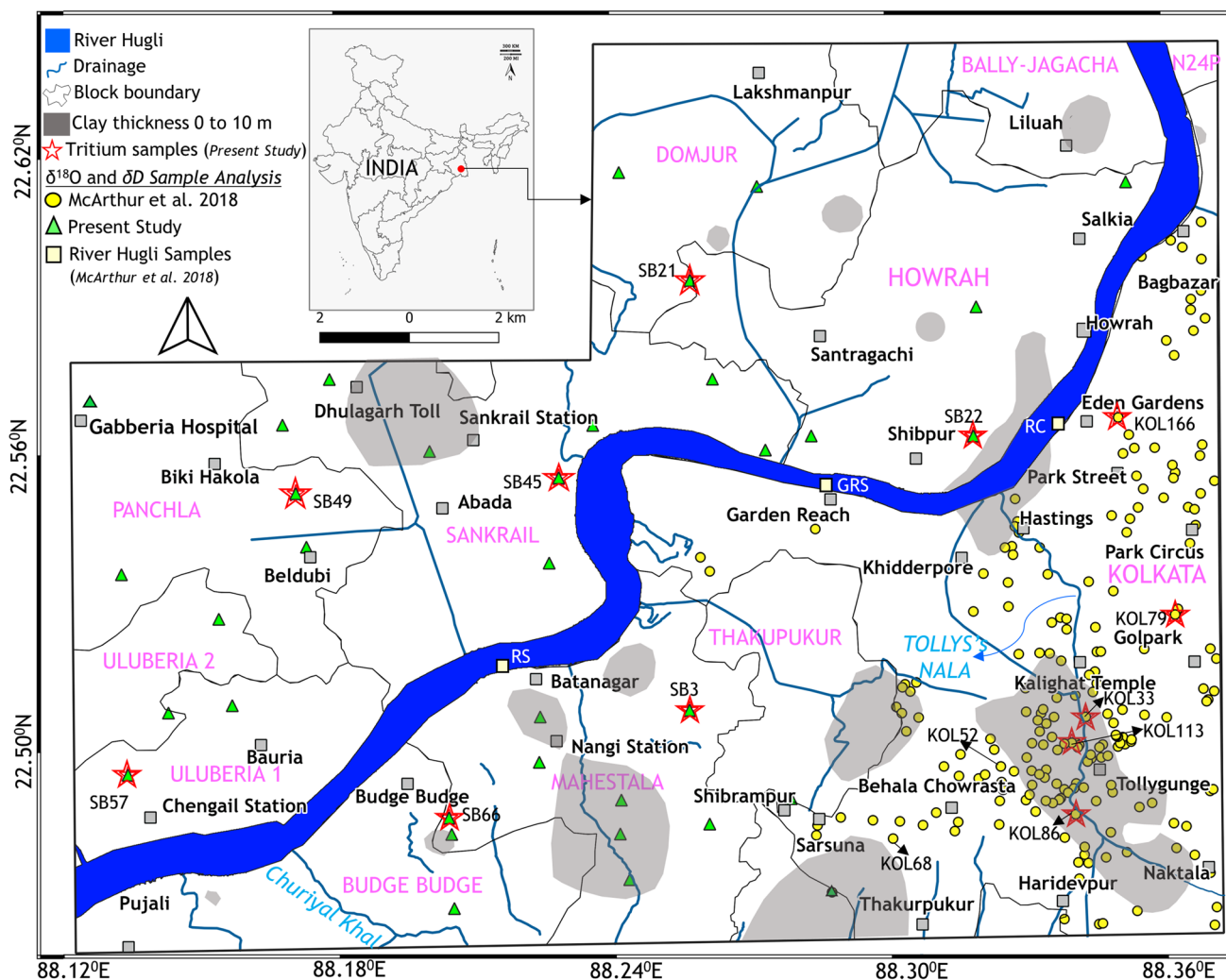


Fig. 1 Map of study area along with locations of samples analysed for stable and radioactive isotopes. The map also shows the areas where upper clay thickness is between 0 and 10 m

Geomorphologically, the study area lies under the fluvio-deltaic environment of deposition. The meandering River Hugli, the lifeline of the twin cities, flows from north-east corner to south-west corner of the present study area. Topographically, the area is almost flat, with many depressions. The surface elevation ranges between 1.5 and 9 m amsl with an average of 6 m amsl. The regional slope is from north to south. The major geomorphic units exposed are broadly classified into different units, e.g. deltaic plains, inter-distributary marshes, paleo-channels, younger levees adjacent to the River Hugli and older levees on both sides of the old Tolly's Canal (Figs. 1 and SI-1) (Banerjee and Sikdar 2021, 2022a, b). Generally, the riverbank of the River Hugli is elevated, having its natural slope away from the riverbed, that is, towards east in Kolkata and towards west in Howrah.

The climate of the study area is predominantly influenced by the northeast and southwest monsoons. Hot lengthy summer with occasional nor'westers, prolonged monsoon from June to October, mild winter and a brief spring are the characteristic features of the climate in the area. The mean annual rainfall of the area is 1668 and 1619 mm during 1966–2018 and 1998–2018 respectively (Banerjee and Sikdar 2022a, 2021; IMD 2020). The area receives maximum rainfall during peak-monsoon period (July–September). In the last few decades, the long-term rainfall in this area shows a decreasing tendency (Banerjee and Sikdar 2022a, 2021).

Geological settings

The sediments of south Bengal Basin were deposited in a fluvio-deltaic environment and have relatively flat landscape

(Sikdar et al. 2022, 2013). The sub-surface sedimentology of a few hundred metres consists of Late Pleistocene topography containing paleo-channels and paleo-interfluves covered by sediments of Holocene channels/floodplains (Sikdar et al. 2022; McArthur et al. 2018).

The stratigraphy of this sedimentary deposit (Fig. SI-3) was developed using borehole lithology and geophysical logging by Banerjee and Sikdar (2022a, 2021) and Sikdar et al. 2022. At the upper part of the sediment distribution, there is a confining clay/silty clay layer (aquiclude) with an average thickness of 26.30 m (Banerjee and Sikdar 2022a, 2021). Below the upper confining bed, the Quaternary confined aquifer occurs, which is about 300-m thick. It consists of sands of various grades mixed with gravels at places (Banerjee and Sikdar 2022a, 2021; Sikdar 2000). This aquifer is heterogeneous in nature as the lenses of fine-grained sediments in the sandy horizon are spatially uncorrelated (Sikdar et al. 2022; Sahu and Sikdar 2017). The Holocene grey clay and late Pleistocene Last Glacial Maximum Paleosol (LGMP) of the upper part of the sediments prevent infiltration of Hugli river water into underlying aquifer (Sikdar et al. 2022; Sikdar and Dasgupta 1997). The overlying cover of aquiclude is thinned down to < 10 m or ceases to exist in some areas; examples include areas along the *Tolly's nala (canal)*, *a palaeo-channel* and Mahestala and Budge Budge (M-B) area in the southern and south-western sections respectively of the present study area (Figs. 1 and 5 of Banerjee and Sikdar 2021). Grey sands occur underneath the *Tolly's canal* instead of the grey clay/silty clay and LGMP. This grey sand has given a direct path for contaminated water from the canal to enter the aquifer below (McArthur et al. 2018).

In Kolkata and surrounding area, the general characteristics of the aquifer are fresh and saline in nature, high TDS, occasional As and Cl, high Fe content with presence of NO₃ in water (CGWB 2006). The discharge rate, transmissivity and storage coefficient of the aquifer beneath Kolkata vary from 20 to 70 m³/h, 1400 to 5610 m²/day and 33 × 10⁻³ to 20 × 10⁻⁵ respectively (Banerjee and Sikdar 2021; Sikdar 1996a, b; CGWB 2006). On the other hand, in Howrah district, discharge, transmissivity and storativity vary from 36 to 211 m³/h, 446 to 2514 m²/day and 45 × 10⁻⁴, respectively (CGWB 2004).

Impacts of urbanisation

Between 1980 and 2014, the built-up area in Kolkata increased by 62 km² while wetland and vegetation cover decreased by about 38.5 km² and 23.5 km² respectively (Mukherjee et al. 2018). During the period 1985–2016, the built-up area of Howrah increased by 11 km², while the adjacent peri-urban and rural areas increased by 40 km² (Banerjee and Sikdar 2021). Such widespread urbanisation has put a significant stress on total groundwater demand.

It has been estimated that the groundwater abstraction in Kolkata Municipal Corporation (KMC) had jumped from 55 to 219 million litres/day (mld) between 1956 and 1992 (Banerjee and Sinha Roy 1992). The surface water supply had reduced the groundwater withdrawal to 144.3 mld during 2006 (CGWB 2006). The groundwater withdrawal by motor pump fitted wells, both KMC wells and private-owned, was estimated at around 305 mld. Although, the extrapolated groundwater extraction is about 320 mld through 2020 to 2025 period (CGWB 2006). The total requirement of drinking water for the entire population of Kolkata city (consisting of 4.6 million residents and a floating population of 6 million) happens to be about 1262 mld (<https://www.downtoearth.org.in/news/city-of-sorrows-12134>). Out of the total water supply of around 1209.6 mld, surface water is 1096.2 mld and the rest is groundwater. As per official record, the demand and supply gap is 37.8 mld. Keeping in mind that 35% supply water loss is caused by pipeline leakage, Kolkata can be labelled a 'water-negative' city rather than being 'water-positive'.

Deterioration of Quaternary aquifer system

The indiscriminate abstraction of groundwater has resulted in a significant fall in piezometric levels, resulting in the formation of a groundwater depression in central and south-central Kolkata and changing the directions of the groundwater flow (Sikdar et al. 2022; Banerjee and Sikdar 2021; Sikdar 1996b). Decades of high-capacity pumping has caused the decline of piezometric surface by approximately 14 m and the rate varies from 0.08 to 0.37 m/year (Sikdar et al. 2022; Banerjee and Sikdar 2022a, b, 2021; McArthur et al. 2018). Between 1985 and 2016, the groundwater trough has increased in area by 15 times at an annual rate of 8.6 km². Thus, the core of the trough has shifted eastward, causing regional groundwater flow inside the city to shift from west to east (Banerjee and Sikdar 2021; Fig. SI-4). In Howrah city, the piezometric surface is declining since 1996 (Patra et al. 2018). Over-withdrawal of groundwater has also brought in water quality deterioration, changes in hydrochemical facies, mixing of wastewater and other potentially toxic elements (PTE), a change in the nature of the aquifer at places from confined to unconfined and an increase in the risk of land subsidence in south Bengal Basin (Sikdar et al. 2022; Banerjee and Sikdar 2022a, b, 2021).

Tolly's canal receives the majority of south Kolkata's contaminated water. Chloride, NO₃, SO₄ and NH₄ concentrations in groundwater, as well as Cl/Br mass ratios, have been utilised to demonstrate the extent to which wastewater contaminates the Bengal Basin aquifer (McArthur et al. 2012) and Kolkata city (Banerjee and Sikdar 2022b; McArthur et al. 2018).

Churiyal Khal (canal) (Fig. 1) receives surface runoff (rainwater) and carries municipality discharge of the M-B area. Few localised small north–south trunk channels connect the *canal* and occasionally these channels along with connecting branches discharge polluted water to the neighbouring low-lying lands or water bodies. During monsoon, water logging occurs in pockets in several areas. The drains' size and hydraulic capacity are insufficient to convey the sewage load. Due to over-exploitation of groundwater, both rainwater and sewage water from the twin cities may infiltrate into the aquifer at places where the upper aquitard is absent or thin (< 10 m) or a local unconfined aquifer condition has developed (Banerjee and Sikdar 2022b, 2021).

Methods

Groundwater chemistry

Total 231 and 56 number of pumping wells tapping groundwater between the depth of 33 and 140 m bgl and below the depth of 180 m bgl, respectively, was sampled for chemical analysis. Groundwater is not being tapped in between 140 and 180 m bgl due to the increase of finer particles (silty sand to silty clay) (Banerjee and Sikdar 2021) within the study area. Out of 287 wells, only nine wells are tapping water above 50 m bgl which is statistically insignificant for analytical purpose. The strainers of the pumping wells are placed based on the grain size of the sediments (medium to coarse sand). Therefore, the samples are mainly collected from the depth range of 80–140 m bgl and 180–230 m bgl. Results of chemical analysis of 287 groundwater samples (Fig. 1 of Banerjee and Sikdar 2022a, b; Fig. SI-1) collected from depths between 32 and 229 m bgl have been used in this study. Out of these, 205, 58 and 24 samples are from Kolkata, Howrah and M-B area respectively. The locations of the samples were recorded using a portable GPS device and are shown in Fig. SI-1. Results of two hundred and one samples have been taken from McArthur et al. (2018), while the remaining 86 samples have been collected by the present authors in 2019 from localities not covered by McArthur et al. (2018). These areas are Howrah in the west and M-B area to the south-west of Kolkata (Fig. 1).

After purging, the samples are collected in two 100-mL polythene bottles from wells fitted with hand pumps or motor pumps. Prior to collection, sampling bottles were soaked in 1:1 diluted HCl solution for 24 h, washed three times with deionised waters and were washed again before filling the sampling bottles with the sample water. During sampling, the samples were filtered with Whatman 40 filter paper. In the laboratory, the samples were filtered using 0.45- μ m Millipore filter paper and acidified with ultra-pure nitric acid for cation analyses. For cation analysis, one

sample is acidified with 0.15 mL of 50% Analar® nitric acid, while the other is not acidified for anion analysis. Decision for the time for purging depends on two factors: depth of piezometric surface and depth of well with the aim that the water sample is collected directly from the aquifer not from the well casing. Analysis of the 86 samples has been carried out at the laboratory of TÜV SÜD, India, and National Institute of Hydrology, Roorkee, India, for cations, anions and metals/metalloid (Fe, Mn and As). Ion chromatography is used to analyse cations and anions, while ICP-OES is used to analyse metals. The total dissolved solid (TDS, mg/L) of these samples is calculated using the following APHA (2005) methodology:

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

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

$$\text{CBE} = \{(\Sigma C - \Sigma A)/(\Sigma C + \Sigma A)\} \times 100 \quad (2)$$

where ΣC and ΣA are 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.

Isotope analysis

For groundwater samples taken from 33 to 229 m bgl (Fig. 1), two types of isotope analysis were carried out: stable isotope ($\delta^2\text{H}$ and $\delta^{18}\text{O}$) and radioactive isotope [Tritium (^3H)]. For stable isotopes, 235 groundwater samples (Kolkata—201, Howrah—23 and M-B area—11) and three Hugli river water were taken into account. Stable isotope sample analysis data of 200 groundwater samples from Kolkata city and three samples from River Hugli were taken from McArthur et al. (2018). Remaining 35 samples of groundwater were collected by the present authors in acid washed polyethylene Tarson bottles. Out of these, 12 samples were used for tritium (^3H) analysis. For stable isotope analysis, 100 mL of sample and for tritium analysis, 500 mL of sample were collected. These bottles were washed twice with the respective well water at the site. Sample bottles were tightly sealed and brought to the laboratory at the shortest possible time in ice-box for analysis to avoid any losses due to diffusion and evaporation. Other parameters of these samples, such as pH and electrical conductivity (EC), were also measured.

In the year 2019, $\delta^2\text{H}$, $\delta^{18}\text{O}$ and ^3H were analysed at the Nuclear Hydrology Laboratory, National Institute of Hydrology Roorkee, India (NIH). Continuous flow isotope ratio mass spectrometry and dual inlet isotope ratio mass spectrometry were used to analyse $\delta^{18}\text{O}$ and $\delta^2\text{H}$ using standard operating procedures (Brenninkmeijer and Morrison 1987;

Epstein and Mayeda 1953). The data are displayed on the following δ scale in concentrations per mil (‰) relative to Vienna Standard Mean Ocean Water (VSMOW):

$$\delta = \left(R_{\text{sample}} / R_{\text{standard}} - 1 \right) \times 1000\text{‰ VSMOW}.$$

where R_{sample} is the $^{18}\text{O}/^{16}\text{O}$ or $^2\text{H}/^1\text{H}$ ratio in water sample and R_{standard} is the corresponding ratio in VSMOW standard.

The all-inclusive accuracy, founded on ten repeated calculations for each of the samples, is $< \pm 1.0\text{‰}$ for $\delta^2\text{H}$ and $< \pm 0.1\text{‰}$ for $\delta^{18}\text{O}$. To condense ^3H in 500 mL of water sample, electrolytic reduction was applied using a 20-cell standard tritium enrichment unit at temperatures ranging from 0 to 5 °C to achieve maximal tritium fractionation and enrichment. As the electrolytic process progresses, the sample volume decreases to 25 mL. The activity of tritium in the reinforced samples, including the standards, was determined in tritium unit (TU) with a 2-sigma error using an ultra-low-level liquid scintillation counter (Quantulus Wallac model 1220). The minimum detection limit is estimated to be 0.4 TU (1 σ criteria).

GMWL and LMWL

McArthur et al. (2018); Sikdar and Sahu (2009); Sikdar and Chakraborty (2008); Klump et al. (2006); Sengupta and Sarkar (2006); Mukherjee (2006); Harvey et al. (2005); Stüben et al. (2003); Basu et al. (2002); Aggarwal et al. (2000); and Shivanna et al. (1999) have published work on $\delta^{18}\text{O}$, $\delta^2\text{H}$ and ^3H distributions in Bengal Basin. Mukherjee (2006) collected 64 groundwater samples, seven river-water and 14 rainwater samples from Gangetic portion of West Bengal during May–August of 2003–2005 and analysed $\delta^{18}\text{O}$ and $\delta^2\text{H}$ compositions to obtain a local meteoric water line (LMWL). The corresponding equation for this LMWL is $\delta\text{D} = 7.24 \delta^{18}\text{O} + 7.73$ ($r^2 = 0.93$). LMWLs are site-specific and act as the covariation of hydrogen and oxygen stable isotope ratios over a long time. Sengupta and Sarkar (2006) also obtained a LMWL whose equation is $\delta\text{D} = 7.88 \delta^{18}\text{O} + 8.93$ ($r^2 = 0.99$). Majumder et al. (2011) studied 328 numbers of groundwater samples to appraise the pattern of groundwater flow in Bengal Delta aquifers of Bangladesh applying environmental isotope (^2H , ^{18}O , ^{13}C , ^3H and ^{14}C) techniques. A distinct LMWL ($\delta^2\text{H} = 7.7 \delta^{18}\text{O} + 10.7\text{‰}$) was established implementing linear correlation analysis methods to the monthly weighted rainfall isotopic compositions ($\delta^{18}\text{O}$ and $\delta^2\text{H}$) (Majumder et al. 2011). Sikdar et al. (2013) displayed the plots of δD drawn against $\delta^{18}\text{O}$ for groundwater samples fetched from both shallow together with deep levels in May 2009 from Moyna, N-24 Parganas that defined a line that falls under the LMWL of Sengupta and Sarkar (2006). These LMWLs nearly match the global meteoric water line (GMWL) of Craig (1961) ($\delta\text{D} = 8 \delta^{18}\text{O} + 10$). Craig's GWML displays the relations between δD and $\delta^{18}\text{O}$ in fresh surface waters all over the world.

In the current study, the GMWL of Craig (1961) ($\delta\text{D} = 8 \delta^{18}\text{O} + 10$) and the LMWL of Sengupta and Sarkar (2006) ($\delta\text{D} = 7.88 \delta^{18}\text{O} + 8.93$) were used to investigate the isotopic data of water samples in and around Kolkata-Howrah cities.

Results

Ion chemistry

Ion chemistry of groundwater of the study area has been elaborated in Banerjee and Sikdar (2022a, b). However, the results of TDS, chloride, sodium, calcium, fluoride and bromide are presented here to discuss the hydrochemical processes and relation with the isotopic variations. TDS concentrations range from 176 to 3377 mg/L. TDS levels of 500 mg/L, the acceptable limit (AL) and > 2000 mg/L, the permissible limit (PL), of BIS (Bureau of Indian Standard) (2012) are found in approximately 18% and 6.6% of the total number of samples, respectively. TDS $< \text{AL}$ concentration were found in 11, 3.5 and 3.5% samples in Kolkata, Howrah and M-B areas respectively. The concentration of TDS $> \text{PL}$ is observed up to a depth of 137 m. In the depth span of 195 to 229 m bgl, only 12% of samples with TDS > 500 mg/L exist. Samples having Cl < 25 mg/L account for about 56.5%, whereas Cl > 1000 mg/L is found to occur in samples up to a depth of 137 m. Excessive Cl and TDS concentrations were found in groundwater samples obtained from the M-B region at lower depths, i.e. < 40 m.

Sodium concentration ranges between 15 and 1116 mg/L, the mean value being 192 mg/L. The mean concentration of Na up to 137-m depth is 202 mg/L and between 183 and 229 m depth, it is around 154 mg/L. Calcium concentration varies from 4 to 324 mg/L with a mean of 106 mg/L. The mean concentration of Ca up to 137 m bgl is 112 mg/L whereas the same is 82 mg/L from depth 183 to 229 m. Fluoride concentrations are ≤ 1 mg/L with mean of 0.31 mg/L. The F and Ca concentrations in groundwater show an inverse correlation in the study area (Banerjee and Sikdar 2022b).

Isotope analysis

The summary of the isotope analysis of 235 samples (Fig. 1) is given in Table 1. $\delta^{18}\text{O}$ and δD have average values of -5.0‰ and -33.7‰ respectively. In the shallow part of the entire area (depth, 33–137 m bgl), the mean value of $\delta^{18}\text{O}$ and δD is -5.2‰ and -35.2‰ respectively. Between 183 and 229 m bgl, the mean values of $\delta^{18}\text{O}$ and δD are -4.0‰ and -26.0‰ respectively. Table 1 shows the range and mean values of $\delta^{18}\text{O}$ and δD for groundwater and river water (river Hugli samples) collected during pre-monsoon, monsoon and post-monsoon periods. Water samples taken from boreholes with upper clay thickness < 10 m

Table 1 Summary of stable isotope composition of groundwater within the study area

Water samples tapping from (m bgl)	Number of samples	Range		Average	
		$\delta^{18}\text{O} \text{‰}$	$\delta\text{D} \text{‰}$	$\delta^{18}\text{O} \text{‰}$ (average)	$\delta\text{D} \text{‰}$ (average)
Entire aquifer section(33–229 m)	235	– 3.0 to – 8.0	– 19.7 to – 54.7	– 5.0	– 33.7
Shallower aquifer (33–137 m)	196	– 3.0 to – 8.0	– 20.0 to – 54.7	– 5.2	– 35.2
Deeper aquifer (183–229 m)	39	– 3.3 to – 6.7	– 19.7 to – 46.0	– 4.0	– 26.0
Entire aquifer section (33–229 m) _ pre-monsoon	84	– 3.0 to – 8.0	– 21.4 to – 54.7	– 5.0	– 34.3
Entire aquifer section (33–229 m) _ monsoon	52	– 3.4 to – 7.6	– 20.0 to – 53.2	– 5.2	– 34.9
Entire aquifer section (33–229 m) _ post-monsoon	99	– 3.3 to – 7.7	– 19.7 to – 53.1	– 4.9	– 32.5
Entire aquifer section (33–229 m) of boreholes having upper clay thickness > 10 m (confined system)	162	– 3.0 to – 8.0	– 21.4 to – 54.7	– 4.9	– 33.0
Entire aquifer section (33–229 m) of boreholes having upper clay thickness < 10 m (unconfined system)	73	– 3.3 to – 7.7	– 19.7 to – 53.1	– 5.2	– 35.2
River Hugli	12	– 6.1 to – 7.9	– 41.6 to – 55.3	– 6.8	– 47.9
River Hugli _ pre-monsoon (Jan–April)	6	– 6.1 to – 7.9	– 44.4 to – 55.3	– 7.0	– 50.0
River Hugli _ early monsoon (July)	3	– 6.4 to – 6.6	– 41.6 to – 44.1	– 6.5	– 43.1
River Hugli _ late monsoon (September)	3	– 6.5 to – 7.7	– 45.1 to – 54.2	– 6.9	– 48.4

(unconfined system) and > 10 m (confined system) were separated to determine the range and the mean value of $\delta^{18}\text{O}$ and δD (Table 1). Summary of isotopic composition of groundwater samples for Kolkata, Howrah and M-B area from shallow (33 to 137 m bgl) and deeper (183 to 229 m) aquifer is given in Table 2.

The stable isotope plots' best fit lines (Fig. 2) have a slope that is lower than both the GMWL of Craig (1961) and the LMWL of Sengupta and Sarkar (2006). The following are the obtained LMWL equations for the groundwater samples taken in the areas of Kolkata, Howrah and M-B:

$$\delta\text{D} = 7.4899\delta^{18}\text{O} + 4.1990; R^2 = 0.97; (\text{Kolkata})$$

$$\delta\text{D} = 8.2842\delta^{18}\text{O} + 5.2304; R^2 = 0.94; (\text{Howrah})$$

$$\delta\text{D} = 6.4935\delta^{18}\text{O} - 1.6542; R^2 = 0.82; (\text{M-B})$$

Depth plots drawn against the concentration of $\delta^{18}\text{O}$ and δD (Fig. 3a, b) show that the groundwater samples < 140 m bgl are more depleted (more negative values of $\delta^{18}\text{O}$) of oxygen than the samples from deeper level (> 183 m bgl). Two samples, KOL52 and KOL68 with screen placed at deeper level (> 140 m bgl), have $\delta^{18}\text{O}$ value of – 6.5 and – 6.7‰, respectively (Fig. 3b).

Tritium (TU) content of the 12 groundwater samples varies between 0.70 and 15.02 TU with error of 0.10 to 0.20 (Fig. 3c). The sample having TU > 15 was collected during peak monsoon period and from the unconfined aquifer system adjacent to Tolly's canal. The depth profile shows higher TU value at depths between 75 and 140 m bgl while samples having low TU values occur at shallow (75–90 m bgl) and deep (200–225 mbgl) depths. There are

Table 2 Summary of isotopic composition of groundwater samples for Kolkata, Howrah and M-B area from shallow and deeper aquifer

Area	Number of samples	Depth of well (m bgl) _ shallow aquifer	$\delta^{18}\text{O} \text{‰}$	$\delta\text{D} \text{‰}$	$\delta^{18}\text{O} \text{‰}$ (average)	$\delta\text{D} \text{‰}$ (average)
Kolkata	176	57–137	– 3.0 to – 8.0	– 20.0 to – 54.7	– 5.3	– 35.9
M-B	7	33–115	– 3.4 to – 5.8	– 25.8 to – 41.9	– 4.4	– 30.7
Howrah	13	33–122	– 3.4 to – 5.1	– 23.1 to – 39.2	– 4.1	– 29.1
Area	Number of samples	Depth of well (m bgl) _ deeper aquifer	$\delta^{18}\text{O} \text{‰}$	$\delta\text{D} \text{‰}$	$\delta^{18}\text{O} \text{‰}$ (average)	$\delta\text{D} \text{‰}$ (average)
Kolkata	25	183–229	– 3.3 to – 6.7	– 19.7 to – 46.0	– 4.1	– 25.4
M-B	4	213–226	– 4.2 to – 4.4	– 28.6 to – 29.6	– 4.3	– 29.1
Howrah	10	200–213	– 3.3 to – 4.2	– 22.5 to – 30.1	– 3.8	– 26.4

Fig. 2 Cross-plot of $\delta^{18}\text{O}$ vs δD based on stable isotope analysis of samples from the present region of investigation. GMWL is the global meteoric water line of Craig (1961) while LMWL is the local meteoric water line of Sengupta and Sarkar (2006) for Barasat, some 25 km northeast of the present region of investigation

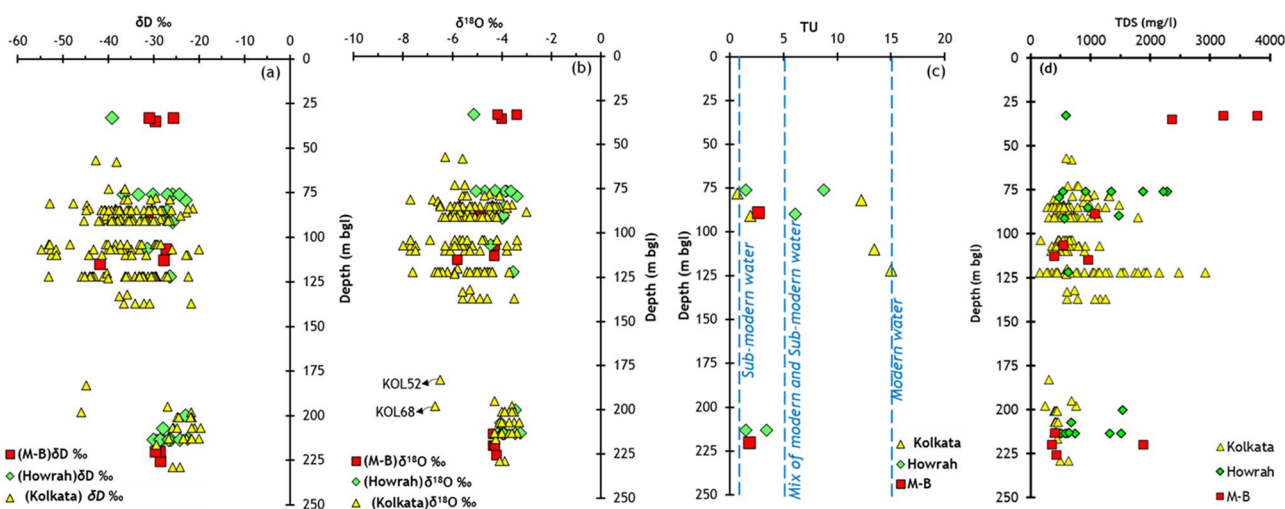
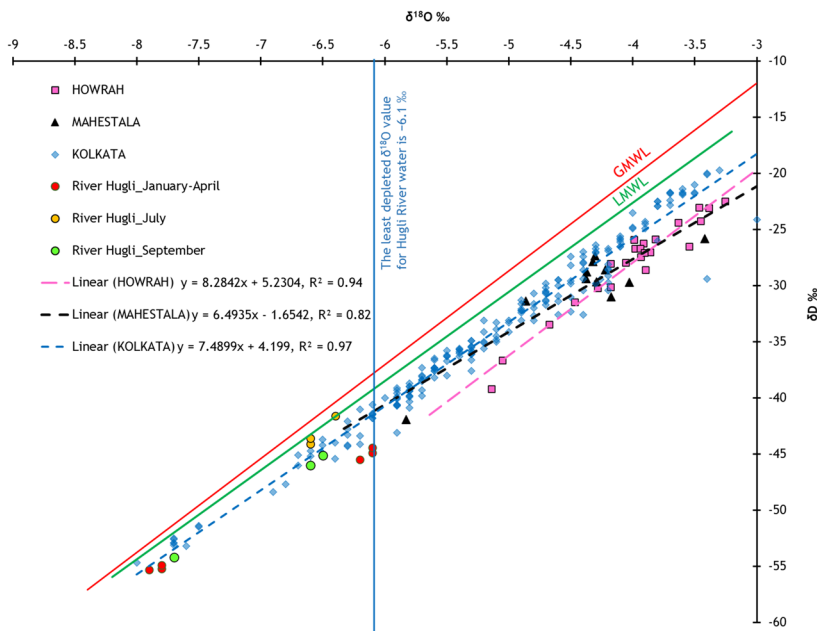


Fig. 3 Depth profile of **a** δD , **b** $\delta^{18}\text{O}$, **c** tritium (TU) and **d** TDS values

total 30 samples out of 235 samples which have depleted $\delta^{18}\text{O}$ having value of < -6.1 ‰. All these 30 samples are from Kolkata and mainly congregate on each side of *Tolly's canal* (Fig. 4).

Discussions

Hydrochemical systematics

Gibbs diagram (Gibbs 1970) which is commonly used to demonstrate the link between water composition and aquifer

lithological properties (Marandi and Shand 2018) has three fields (evaporation dominance, rock dominance or rock–water interaction and precipitation dominance). The present area's groundwater samples lie in between evaporation dominance and rock dominance (Fig. 5). The ratio of Na^+ to $(\text{Na}^+ + \text{Ca}^{2+})$ is in the range of 0.25 and 0.9, meaning that sodium concentrations in groundwater are frequently greater than calcium concentrations. The $\text{Cl}^- / (\text{Cl}^- + \text{HCO}_3^-)$ ratio ranges between 0.05 and 0.90, suggesting that the amount of Cl^- and HCO_3^- in groundwater varies. Therefore, the main process controlling the groundwater chemistry in the studied region is rock weathering in conjunction with evaporation.

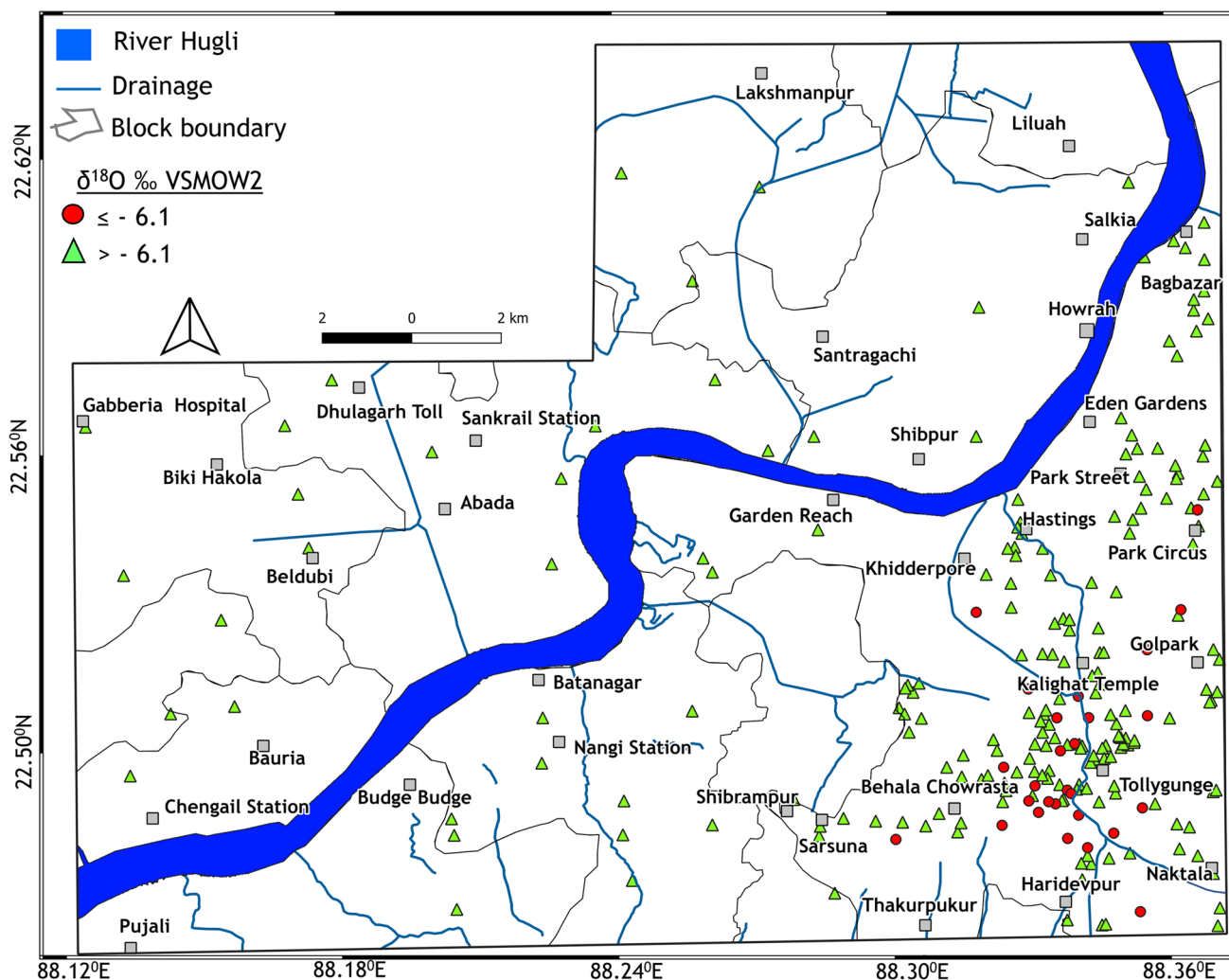


Fig. 4 Spatial distribution of groundwater samples having depleted and enriched $\delta^{18}\text{O}$

The samples do not plot on the field of precipitation dominance suggesting that precipitation has little impact on groundwater composition. Some water samples were plotted outside of the three fields (Fig. 5a), indicating that some other factor, such as ion exchange, may influence the chemical composition of groundwater.

The Facies 8 (Na-Cl) groundwater samples plot around the line of equality in the Na versus Cl plot (Fig. 14 of Banerjee and Sikdar 2022a), indicating that mineral dissolution is occurring in the aquifer (Sikdar et al. 2001; cf. Llyod and Heathcote 1985).

Silicate weathering, evaporite dissolution and ion exchange processes in groundwater are further investigated using the Ca/Na vs. Mg/Na plot (Fig. 6) and the Na/Cl vs. Cl plot (Fig. 7). Figure 6 displays that silicate weathering is the most important mechanism influencing water chemistry. Figure 7 also indicates that silicate weathering is the dominant process for the samples having Na/Cl ratio greater

than that of rainfall (0.86) (Hem 1989) but those with $\text{Na}/\text{Cl} > 2$ indicates the twin effect of silicate weathering and ion exchange processes (Herczeg and Edmunds 2000).

The south Bengal Basin, including the current region, was submerged in brackish to seawater during the Late Quaternary period (Sikdar et al. 2001; Hait et al. 1996a, b, 1994a, b; Barui and Chanda 1992; Sen and Banerjee 1990). As a result, seawater with high Cl concentration was probably trapped in the sediments as they were deposited and later modified in the aquifer in a confined condition. Later the trapped seawater was diluted by freshwater infiltrating into the aquifer through the recharge zones present in the north of the current area (Sikdar et al. 2001). Also, direct recharge of meteoric water in localised pockets where the upper aquitard is either absent or exceedingly thin because of scouring action of past channels caused dilution of the trapped seawater (Fig. SI-2). Thus, the mixing of seawater and freshwater gave rise to the brackish water of present-day. This mixing

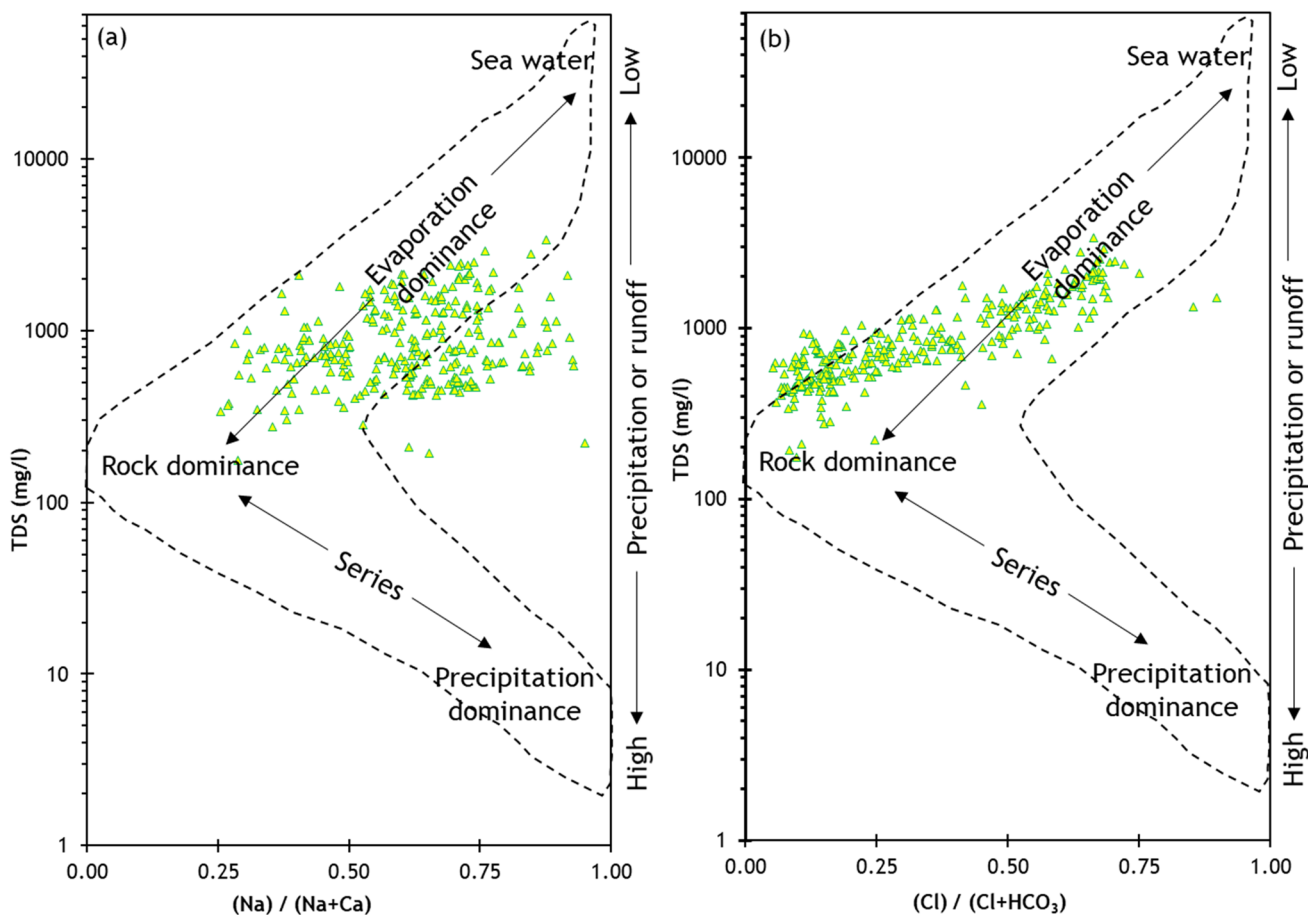


Fig. 5 Gibbs diagrams: **a** TDS vs. $(Na)/(Na + Ca)$ and **b** TDS vs $Cl/(Cl + HCO_3)$ showing the mechanisms controlling groundwater chemistry

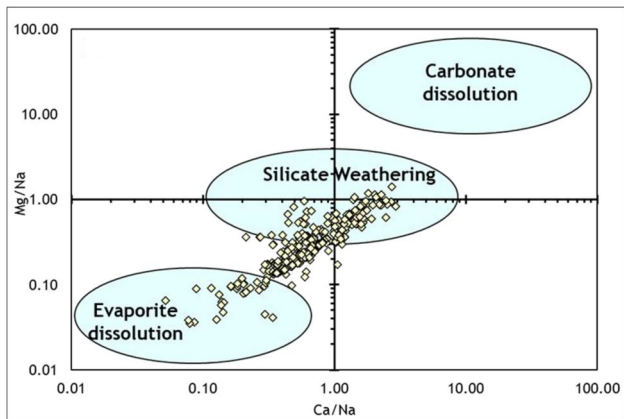


Fig. 6 Scatter diagram of Ca/Na vs Mg/Na explaining the geochemical processes in groundwater

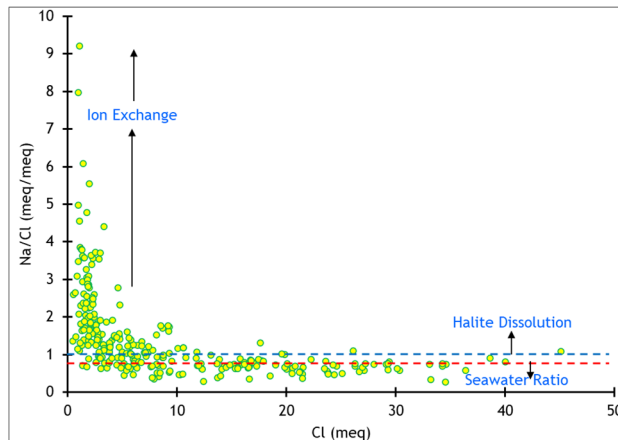


Fig. 7 Scatter diagram of Cl vs Na/Cl representing the geochemical processes in the aquifer

was non-uniform because of the wide variation in the concentration of Cl (127 to 1598 mg/L) of the brackish water (Banerjee and Sikdar 2022a). The majority of the samples of groundwater occur in the salinisation field, implying limited freshening of modified connate water (Fig. 8). Plots of

groundwater samples near the marine water mixing line (red line in Fig. 8) indicate a delayed phase of aquifer flushing by freshwater. The presence of Facies 2 ($Na-HCO_3$) water surrounding tiny areas of Facies 1 ($Ca-Mg-HCO_3$) water

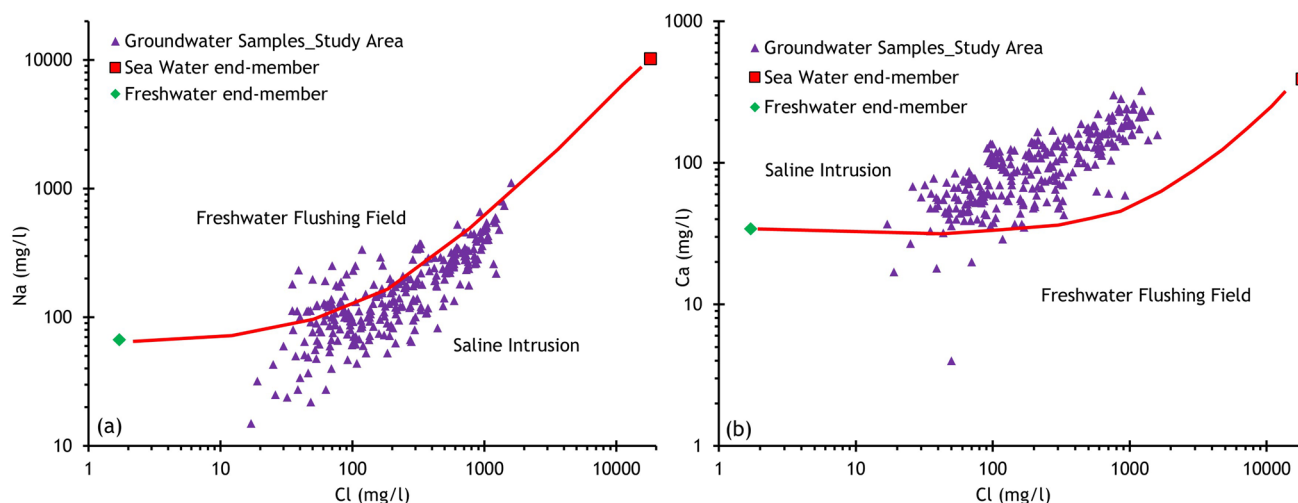


Fig. 8 Scatter diagram of Cl vs. **a** Na, **b** Ca. The red line represents mixing line between dilute groundwater end-member (Na 67, Cl 1.8, Ca 36 mg/L) and seawater end-member (Na 10155, Cl 18290, Ca

388 mg/L) (from Ravenscroft et al. 2013; McArthur et al. 2012; Sikdar and Sahu 2009; Ravenscroft and McArthur 2004)

(Fig. 4 of Banerjee and Sikdar 2022a; Fig. SI-5) suggests dynamic flushing of freshwater in the aquifer system where ion exchange occurs (Sikdar et al. 2001).

Isotope signature

Stable isotopic signatures of recharge sources

A study on stable isotopes in Hugli river water (McArthur et al. 2018) found that the least depleted $\delta^{18}\text{O}$ is -6.1‰ , whereas the volume-weighted annual mean value for $\delta^{18}\text{O}$ of local rainfall is -6.2‰ . Hugli river water has $\delta^{18}\text{O}$ and δD values ranging from -6.1 to -7.9‰ and -41.6 to -51.3‰ , respectively. The values of $\delta^{18}\text{O}$ and δD fluctuate seasonally in response to varying proportions of base-flow and rainfall-runoff that reaches the river (McArthur et al. 2018). The stable isotope plots of Hugli river water (Fig. 2) are below the GMWL and LMWL suggesting evaporation has an enrichment effect.

Previous isotopic studies in South 24-Parganas district of West Bengal (Misra 2001; Shivanna et al. 1999) indicate that $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of shallow groundwater (aquifer depth < 70 m) along with deep groundwater (aquifer depth > 70 m) exist in two separate groups but occur on the LMWL or above it, revealing that evaporation has not affected these waters. Moving north-west from the previously studied South-24 Parganas, in the study area, stable isotope data plots indicate a different hydrogeological condition. The best-fit line of $\delta^2\text{H}$ — $\delta^{18}\text{O}$ plots of the present area's groundwater has a slope lower than LMWL and GMWL (Fig. 2) indicating that evaporation of rainwater occurred before or at the time of infiltration and/or intermixing of the infiltrated water with the pre-existent soil moisture

which had faced various cycles of evaporation and wetting (Allison 1982).

Groundwater samples collected between 33 and 137 m bgl are slightly more depleted (mean $\delta^{18}\text{O} - 5.2\text{‰}$) compared to samples collected between 183 and 229 m bgl (mean $\delta^{18}\text{O} - 4.0\text{‰}$). Groundwater samples collected from the unconfined system are slightly more depleted (mean $\delta^{18}\text{O} - 5.2\text{‰}$) than those from the confined system (mean $\delta^{18}\text{O} - 4.9\text{‰}$). This suggests that evaporation occurs prior to infiltration of rainwater into the aquifer system. However, the range and average mean of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of shallow and deep groundwater, samples from unconfined and confined system (Table 1) are almost similar. No significant isotopic variation is observed as the recharge area for both are quite close, the local climatic condition is quite similar and similarity in hydrostratigraphy. The wide range of $\delta^{18}\text{O}$ (-3 to -8‰) and $\delta^2\text{H}$ (-19.7 to -54.7‰) values indicates groundwater recharged from multiple recharge sources with varied residence time and different isotopic signatures. The study area has several recharge mechanisms. They are (a) recharge to the locally unconfined system through direct precipitation; (b) recharge to the locally unconfined system through river water—thirty groundwater samples of Kolkata having depleted $\delta^{18}\text{O}$ ($< -6.1\text{‰}$) cluster around *Tolly's canal* resulted from the mixing of isotopically depleted Hugli river water and less-depleted groundwater from late Pleistocene aquifer; (c) several distant recharge areas for the confined system; (d) artificial groundwater recharge systems in the city areas; (e) return flow from the irrigation practice in the rural areas.

The deuterium excess (d-excess) gives information about source of the initial moisture, effects of monsoon evaporation and moisture recirculation from large inland water

bodies (Krishana et al. 2021; Négrel et al. 2007; Clark and Fritz 1997; Dansgaard 1964). The plots of d-excess and $\delta^{18}\text{O}$ of groundwater samples are presented in Fig. 9. The d-excess values range between -2.2 and 9.3‰ and the mean is 6.5‰ . The poor correlation (R^2 varies between 0.02 and 0.19) between $\delta^{18}\text{O}$ and D-excess (Fig. 9) reflects the influence of evaporation of surface water before infiltration (Liet al. 2014). The d-excess of Hugli River water samples ranges from 6.8 to 7.4‰ with a mean value of 7.03‰ . This suggests that the river water is subjected to evaporation resulting in the stable isotope composition occurring below the GMWL and the LMWL (Fig. 2). The large range of $\delta^{18}\text{O}$ values (Table 1) in groundwater indicates that there has not been enough mixing of Hugli River water to homogenise variances in recharge water's isotopic composition.

Tritium isotope: age indicator

Tritium (^3H) dating of groundwater has been used in the current study to detect the recharge zones and the source of contaminants. ^3H is produced in the environment by natural reactions between cosmic neutrons and the atmospheric ^{14}N . Once created, it joins together with oxygen to create a water molecule, which then falls as rain. The tritium content in the groundwater in the recharge region is about equal to the tritium value of the recharging water.

After being recharged, the concentration of ^3H begins to decline in accordance with its 12.32-year half-life. The ^3H level thus falls exponentially over time away from the recharge zone as groundwater moves towards the discharge zone. Groundwater devoid of ^3H is anticipated to be older than 50 years.

Tritium content of groundwater from shallow and deep aquifer in and around East Calcutta Wetland (ECW), adjacent to the present study area, varies from <0.76 TU to 6.6

TU and <0.53 TU to 4.5 TU, respectively (Sikdar and Sahu 2009). The tritium value of the present study ranges between 0.7 and 8.74 TU, except two samples having tritium > 10 TU located at the bank of *Tolly's Canal* where locally unconfined aquifer system exists. Six samples out of twelve are having tritium ranges between 0.7 and 2.6 TU. This low TU value indicates that this groundwater has long residence time and that recharging may have occurred further away. The ^3H content of today's precipitation in India is expected to be approximately 1.33 to 6.32 TU at Kozhikode, Kerala; 1.59 to 9.21 TU at Hyderabad (www.isohis.iaea.org); and 5.3 TU in 1995 in Dhaka, Bangladesh (Stute 1999). In the Tuscon basin, southwest USA tritium in rain increased because of industrial tritium release in 1970s, which was ultimately washed out by 1991. Since then, the ^3H content in rain is 4.6 to 6.7 TU, which represents natural cosmogenic production (Eastoe et al. 2004). The groundwater in the present study area has a ^3H value range that is similar to that of rainwater from Kozhikode, Tuscon basin and ECW which suggests that the aquifer is directly recharged by monsoonal precipitation.

According to Abdullah et al. (2018), groundwater age estimation using tritium provides semi-quantitative 'ball park' values as follows:

<0.8 TU denotes sub-modern water (pre-1950s).

0.8 to 5 TU denotes mixture of sub-modern and modern water.

>5 to 15 TU denotes modern water (<5 to 10 years).

15 to 30 TU denotes bomb tritium.

30 TU denotes recharge generation from the 1960s to 1970s.

Tritium content of the 12 groundwater samples varies between 0.70 and 15.02 TU with error of 0.10 to 0.20 TU (Figs. 3c and 10). The sample having TU > 15 was collected during peak monsoon period and from the unconfined aquifer system adjacent to *Tolly's canal*. The depth profile (Fig. 3c) shows that samples with higher TU value occur at depth < 140 m bgl while those with lower TU value occur at shallow and deep depths.

Figure 10 reveals that samples KOL33, KOL86 and KOL113, all of which are found adjacent to the *Tolly's canal*, exhibit $\delta^{18}\text{O} < -6.1\text{‰}$ and $^3\text{H} > 5$ TU. The age of these groundwater samples is <5 to 10 years. The absence of the upper aquitard has allowed the modern water to infiltrate into the late Pleistocene aquifer samples from wells SB49 (Dhulagori Toll) and SB57 (Chengail) (Fig. 1) exhibit ^3H value of 6.05 and 8.74 TU respectively. These values indicate the existence of modern water in the aquifer system in both of the places. At Dhulagori, the near-absence of the upper aquitard has allowed direct infiltration of modern water (Fig. SI-2). At Chengail, modern water may not have directly infiltrated from the surface

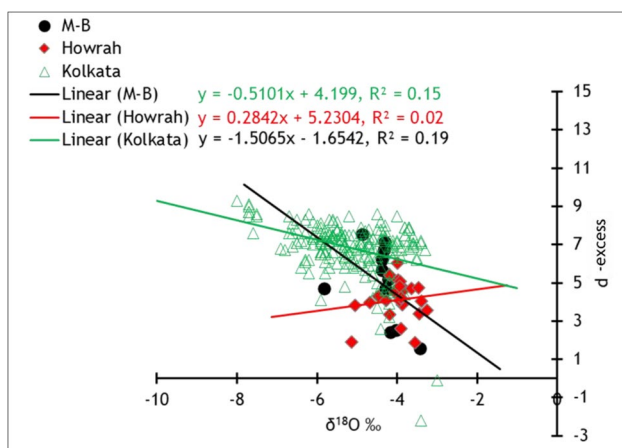


Fig. 9 Cross-plot diagram of d-excess vs. $\delta^{18}\text{O}$ in groundwater samples of the study area

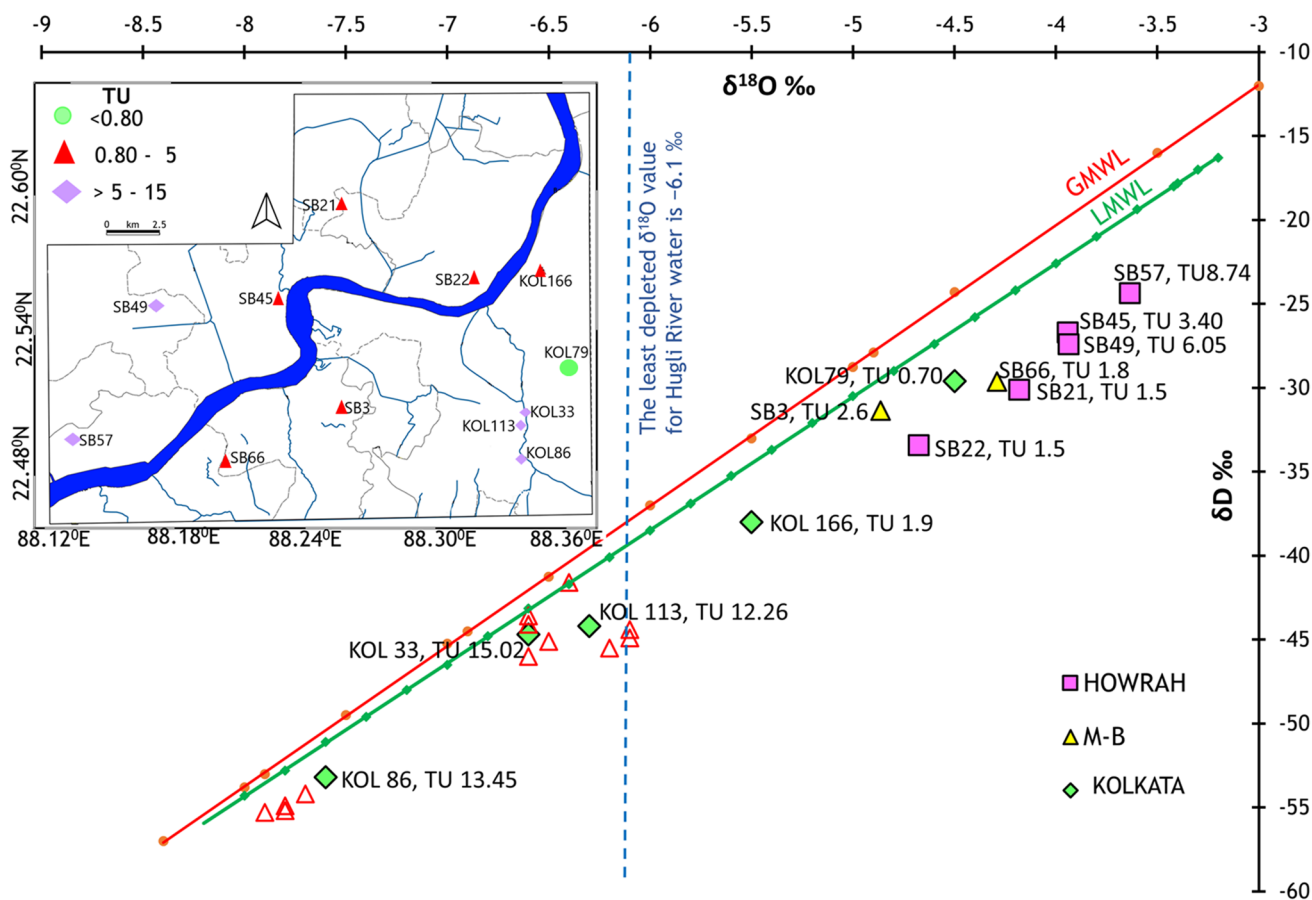


Fig. 10 Cross-plot diagram of $\delta^{18}O$ vs. δD for the samples having tritium analysis data. The location of the samples and range of TU values are given in the inset map

into the aquifer because of the presence of the upper aquitard but may have migrated from neighbouring Pujali area where the upper aquitard is practically absent (Figs. 1 and SI-2) Numerical groundwater modelling (Fig. 15 of Sikdar et al. 2022) has also shown that modern water with potentially toxic elements (PTEs) has migrated from Pujali to Chengail. The 3H value of sample KOL79 (Figs. 1 and 10) is 0.70 TU and the $\delta^{18}O$ is -4.5‰ indicating the presence of sub-modern water. The 3H content of the remaining seven samples ranges between 1.5 and 3.4 TU indicating a mix of modern and sub-modern water (Fig. 3c).

Figure 11 shows tritium levels in groundwater together with salinity (as measured by electrical conductivity). The salinity induced by salt dissolution rises with the age of groundwater. However, higher salinity and tritium levels suggest evaporative enrichment within the aquifer system (Krishana et al. 2021).

Cl/Br and tritium as an indicator of recharge sources

In the present area, Cl/Br mass ratios and Cl concentrations in groundwater are used as a proxy for wastewater

infiltration (Banerjee and Sikdar 2022b; McArthur et al. 2018, 2016, 2012; Katz et al. 2011). The average Cl/Br ratio is higher in samples at depths < 140 m bgl than the groundwater samples at > 180 m bgl for Kolkata, Howrah

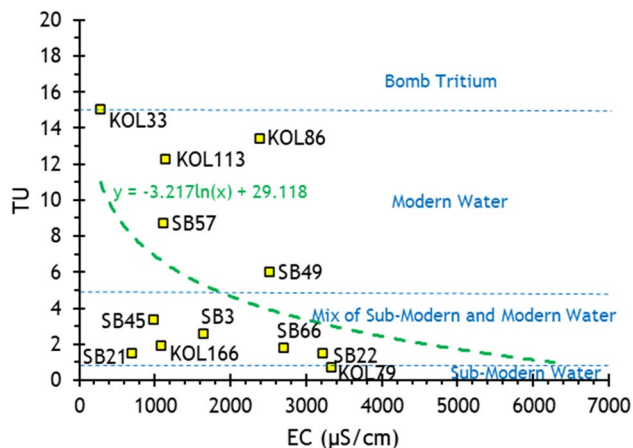


Fig. 11 Cross-plot diagram of tritium content (TU) vs. electrical conductivity (EC) in groundwater. The green broken line is the logarithmic trendline whose equation is $y = -3.21\ln(x) + 29.118$

and M-B areas (Fig. 8a of Banerjee and Sikdar 2022b). Out of the 279 samples examined, around 29% have $Cl/Br < 268$ (lower limit of 288 ± 20 , to account for analytical uncertainty; Banerjee and Sikdar 2022b; McArthur et al. 2018), with organic degradation being the cause of the surplus Br (Fig. 12). Around 30% of the samples had $Cl/Br > 308$ (upper limit of 288 ± 20 to account for analytical uncertainty; Banerjee and Sikdar 2022b; McArthur et al. 2018) with the majority of them congregating adjacent to the *Tolly's canal*, signifying wastewater contamination (Fig. 12). Furthermore, several samples had $NO_3/Cl > 0.0002$ and $SO_4/Cl > 0.14$, indicating that wastewater from sewage and septic tank leaks had contaminated groundwater in the area (Banerjee and Sikdar 2022b).

Out of 12 groundwater samples (Figs. 1 and 10 inset), four samples (SB3, SB21, SB45, SB66) have $Cl/Br < 268$ but tritium

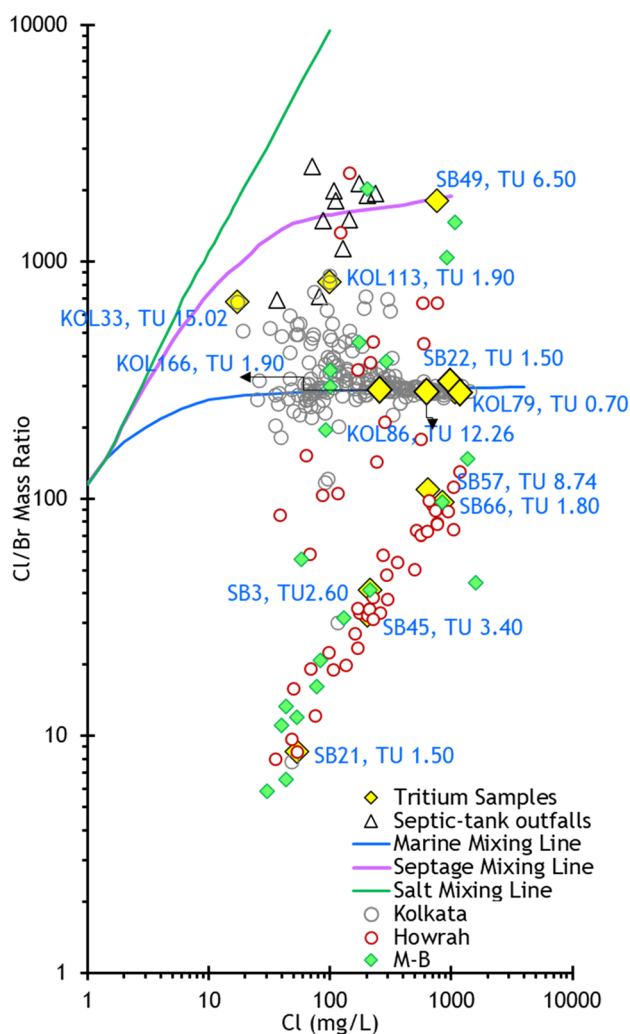


Fig. 12 Cross-plot diagram of Cl and Cl/Br in groundwater. Septic-tank outfall samples are sampled and analysed by McArthur et al. 2012 from Moyna, Barasat, North 24 Parganas, West Bengal (after Banerjee and Sikdar 2022b)

concentration between 0.8 and 5 TU indicating mixing of modern and sub-modern water but no infiltration of wastewater (Fig. 12). SB57 has $Cl/Br < 268$ and 8.74 TU of tritium, indicating infiltration of modern freshwater. Samples KOL33 and KOL113, which are located adjacent to *Tolly's canal* (Figs. 1 and 10 inset), have $Cl/Br > 308$ and tritium content of 15.02 and 1.90 TU respectively. Groundwater sample KOL86 that is also located adjacent to *Tolly's canal* has Cl/Br ratio of 284.5 and tritium content of 13.45 TU. Therefore, the tritium values and Cl/Br ratio of groundwater samples adjacent to *Tolly's canal* indicate infiltration of modern wastewater/freshwater into the aquifer system which mixes with the sub-modern groundwater. This is further corroborated by the presence of high concentrations of arsenic (10–79 $\mu\text{g/L}$) in groundwater of few wells adjacent to *Tolly's canal* as a result of a pollution plume emerging from a plant site where Paris Green, an arsenic bearing pesticide, was produced between 1965 and 1985 (Banerjee and Sikdar 2022a; McArthur et al. 2018).

The current work indicates that in certain pockets, the upper clay/silty clay is absent or its thickness is < 10 m (Figs. 1 and SI-2). These pockets serve as ‘stratigraphic shortcut’ for pollution plumes, including wastewater ($Cl/Br > 308$), to enter the aquifer and deteriorate groundwater quality (Banerjee and Sikdar 2022b; McArthur et al. 2018). In other areas, such as Park Street-Park Circus, where the upper aquitard exists, the presence of wastewater in the confined aquifer may be due to vertical leakage of wastewater through the aquitard caused by a rise in lithostatic pressure in the aquitard as well as in the aquifer due to heavy groundwater pumping.

SB49, which is on the ‘Septage mixing line’ (Fig. 12), contains 6.5 TU of tritium and is located adjacent to Dhulagarh and Sankaril Industrial Estate (Figs. 1 and 10 inset), where the upper aquitard is either < 10 -m thick or non-existent (Figs. 1 and SI-2). Therefore, the wastewater has infiltrated the aquifer over a decadal time span. Furthermore, if left untreated, industrial waste containing heavy metals may eventually reach the groundwater pumping level in the future. In the contemporary context, the implication is that if the sewage water includes PTEs, some of them may migrate to Kolkata’s well intakes. This is consistent with the findings of Banerjee and Sikdar (2022a, 2021), Sahu and Sikdar (2011), Sikdar and Banerjee (2003), Ghosh et al. (2001) and Sikdar et al. (2001), who identified the presence of PTEs such as chromium, cadmium, lead, copper and nickel in Kolkata’s groundwater.

Conclusions

The groundwater chemistry of the Quaternary aquifer under the Kolkata-Howrah twin-city in the South Bengal basin is primarily controlled by silicate weathering, evaporation, ion

exchange and to some extent mineral dissolution. The Cl/Br ratios of groundwater samples show that surface contamination from anthropogenic Cl is present up to 140 m bgl while elevated Cl at deeper level is obtained from marine water, rather than wastewater. Approximately 30% of the samples with Cl/Br > 308 (upper limit of marine mixing water) were polluted by sewage effluent and septic tank leaks. For the deep groundwater samples with elevated Cl, it is likely that flushing of freshwater from the recharge zones diluted the high-Cl marine water trapped during syn-sedimentation through heterogenous mixing. The presence of high Cl concentration in ‘brackish’ groundwater may indicate the presence of a remnant of seawater that underwent chemical alterations during its confinement period. This suggests that groundwater flow is restricted, and flushing is minimal.

The stable isotope ($\delta^{18}\text{O}$ and δD) data of groundwater samples show a slope lower than the LMWL and the GMWL, indicating that evaporation occurs before or during the recharge process or that there was some intermixing of the invading water with the pre-existing soil moisture, which had experienced a number of rounds of alternate evaporation and wetting. The varying values of groundwater $\delta^{18}\text{O}$ suggest that these groundwaters were not properly mixed to blend differences in the isotopic composition of the recharge water. This also demonstrates that many groundwaters are produced by mixing the most recent recharge with an earlier unit recharged under colder climatic conditions than the present day.

The depth plots versus the concentration of stable isotopes reveal that the shallower level groundwater samples are more depleted of $\delta^{18}\text{O}$ than the deeper level samples. The wide range of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values indicates groundwater recharged from multiple recharge sources with different residence times, resulting in different isotopic fingerprints. About 12.80% groundwater samples having depleted $\delta^{18}\text{O}$ mainly cluster on either side of *Tolly's canal* where the aquifer is locally unconfined. The tritium levels of groundwater in the study area vary from 0.70 to 15.02 TU, indicating the presence of ‘sub-modern’, ‘mix of modern and sub-modern water’ and ‘modern water’. This shows a connection between infiltrating isotopically depleted water from the Hugli river via *Tolly's canal* and less-deficient groundwater from Kolkata's late Pleistocene aquifer. The tritium values and Cl/Br ratios of groundwater samples adjacent to *Tolly's canal* and elsewhere indicate direct infiltration of ‘modern wastewater and freshwater’ into the aquifer system where it mixes with the ‘sub-modern water’. In the context of the current investigation, the assumption is that if the wastewater contains PTEs, some of it may end up in city-well intakes.

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Author contribution Data generation and processing, interpretation and writing of the paper have been carried out by Sumanta Banerjee. However, Prof. Sikdar (present guide of my ongoing PhD) has reviewed the paper critically at interpretation stage, presentation of the figures and several other aspects. Prof. S Kumar, as specialist of isotope studies, has also carried out the technical review of this research paper.

Data availability Data provided in the supplementary material.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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