RESEARCH ARTICLE



Application of water quality index and chemometric methods on contamination assessment in the shallow aquifer, Ganges River basin, India

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Abstract

Water quality index and chemometric methods were employed to assess the groundwater quality and contamination sources in the upper Ganges basin (UGB) and lower Ganges basin (LGB) as groundwater is a sole source for drinking, domestic and agricultural uses. Groundwater samples were collected from UGB (n = 44) and LGB (n = 26) and analysed for physicochemical parameters. Groundwater in this basin is desirable (51%) to permissible (TDS < 1000 mg/l, 96%) classes for drinking. Chemical constituents in the groundwater are lower than the maximum allowable limit recommended by the WHO for drinking except K. Drinking water quality index (DWQI) values reveal that groundwater belongs to excellent (89%) and good (10%) classes. However, the high concentrations of Fe and Mn in 61 and 77% of samples, respectively, restrict the usage for drinking according to USEPA recommendations. Both LGB and UGB groundwater in shallow wells have elevated concentration of TDS, EC and other ions (Ca²⁺, Cl⁻ and SO4²⁻ in LGB; major ions, NO3⁻, PO4³⁻, F⁻, Fe and Mn in UGB) and imply the influences of anthropogenic activities. Principal component analysis and hierarchical cluster analysis reiterated that groundwater quality is affected by the anthropogenic activities as well as mineral dissolutions (carbonate and silicate minerals). This study highlighted that the infiltration of wastewater from various contamination sources likely triggered the dissolution of the minerals in the vadose zone that resulted in the accumulation of ions in the shallow aquifer. An effective management plan is essential to protect this shallow aquifer.

Keywords Groundwater, \cdot Contamination, \cdot Metals, \cdot Water quality index, \cdot Chemometric methods, \cdot Lower and upper Ganges basin \cdot India

Introduction

Ganges River basin (GRB) is the largest river basin (1.086 million km²) in Asia and it connects India, Bangladesh, Nepal and China (Amarasinghe et al. 2016). In the GRB, the majority of the populace depends on agriculture, which is a major source for food security and livelihood (Sharma et al. 2010). GRB is broadly classified into upper, middle and lower basins (HWRIS 2020). The water requirement of agriculture, domestic and industrial sectors is balanced by the groundwater

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Natarajan Rajmohan rnatarajan@kau.edu.sa; nrmohan 2000@yahoo.com due to poor quality and scarcity of surface water (Sinha et al. 2006; Chandra et al. 2011; CPCB 2013; CWC/NRSC 2014; Khan et al. 2015; Amarasinghe et al. 2016). GRB has multilayered highly potential aquifers. From the socio-economic point of view, shallow aquifers are less expensive precious water resources for people's livelihood compared to the deeper one.

In recent days, researchers concentrated more on shallow aquifers because shallow aquifers are mostly over-exploited and contaminated by various sources (Bu et al. 2020; Hajji et al. 2020; Houemenou et al. 2020; Hui et al. 2020; Quino Lima et al. 2020; Ranjbar et al. 2020). Alcaine et al. (2020) reported that shallow aquifer has high arsenic and fluoride content and evaporation enhanced trace metal concentrations during dry seasons in the discharge area. Manikandan et al. (2020) reported that monsoon recharge triggered the pollutant movement in the vadose zone and contaminated the shallow aquifer. Therefore, these studies emphasise that shallow

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aquifers are highly vulnerable to contamination and periodic monitoring is essential for systematic water distribution and management worldwide.

High permeability soil in the vadose zone and shallow water table also enhance the vulnerability of shallow aquifer to contamination (Nolan et al. 2002; Davraz et al. 2009; Jiang et al. 2009). The shallow aquifer in the GRB is predominantly affected by the surface and subsurface contamination sources, which resulted in high salinity, chloride, nitrate, ammonium, phosphate, heavy metals and bacteria in the groundwater (Somasundaram et al. 1993; Sinha and Saxena 2006; Saha et al. 2009; Chakraborti et al. 2011; Rajmohan and Prathapar 2014; Rajmohan and Amarasinghe 2016; Rajmohan et al. 2017; Rajmohan 2020). In cultivated areas, the application of fertilisers, organic manures and irrigation return flow are major pollution sources for groundwater. In the residential and urban regions, sewage disposal, dumping sites, pit latrines, septic tanks and accumulation of cattle droppings are important roots for groundwater contaminations. Besides, groundwater contamination by geogenic sources is also documented in the GRB, namely, arsenic, iron, manganese, fluoride, selenium, radon, etc. (Rajmohan and Prathapar 2014; Sankar et al. 2014; Mukherjee et al. 2018). Among these pollutants, arsenic, iron and fluoride are widely reported in GRB, mainly in Uttar Pradesh, Bihar, West Bengal, Nepal Terai and Bangladesh (Pandey et al. 2009; MDWS 2011; Rajmohan and Prathapar 2014; Sankar et al. 2014; Chakraborty et al. 2015; PHED 2015). Further, vertical flow/leakage from polluted shallow aquifer likely affects the deeper confined aquifer (Zhai et al. 2013; Qian et al. 2020). Earlier studies imply that protecting the shallow aquifer from contamination is a primary concern. Therefore, detailed knowledge about groundwater quality and pollution sources is essential for planning and aquifer management. Besides, earlier studies carried out in the GRB are site-specific. Hence, this study intends to compare the groundwater quality in different regions of the GRB. The primary objectives of this study are to compare the groundwater quality in the lower (LGB) and upper Ganges basin (UGB), to assess the groundwater suitability for drinking using drinking water quality index (DWQI) and to identify the source of contaminants in the shallow aquifer using chemometric methods. As the groundwater is an important source for drinking in the study site, this study will assist various stakeholders for planning and management and protect the shallow aquifer for future needs.

Materials and methods

Study area

The study was carried out in two regions in the Ganges basin: upper Ganges basin (UGB) and lower Ganges basin (LGB) (Fig. 1). In the UGB, groundwater samples were collected from selected villages in the Bareilly, Rampur and Shahiahanpur districts, Uttar Pradesh, India. In the LGB, sampling was carried out in the Vaishali district, Bihar, India. In the UGB, the temperature varies from 8.6 to 40.5 °C and it experiences a subtropical climate. The annual average rainfall is between 967 and 1088 mm and the major rainy period is from June to October. Ramganga River and its tributaries are the major surface water resources in the study site. Based on the geomorphology, UGB is classified into the lower piedmont plain of Tarai, older alluvial plain, younger alluvial plain and meander flood plain soils. The study site has Tarai (calcareous), Khadar (silty loamy sand or sandy) and Bangar soils (located at the upland tract of older alluvial plain). Older alluvium, younger alluvium and Tarai formations are major hydrogeological formations. Older alluvium is formed by the clay with Kankar and sand, while younger alluvium consists of fine sand and silt clay mixed with gravel. Likewise, the Tarai formation consists of sandy clay, sand, gravel and clay. Alluvial sediments are the important water-bearing formations (CGWB 2009; CGWB 2014). During pre-monsoon, the groundwater level varies from 2.45 to 14.88 metres below ground level (mbgl) whereas, during post-monsoon, it is between 1.95 and 14.65 mbgl (CGWB 2009).

In the LGB, the river Ganges and Gandak are major surface water resources. In this region, the temperature is from 4 to 40 °C and the average annual rainfall is 1168 mm. The south-west monsoon (June to September) has a major contribution (85%) to the total rainfall. The study site mainly consists of alluvium deposits formed by the Ganges, Gandak and their tributaries. According to geomorphology, the Vaishali district is covered by the Hazipur surface, Vaishali surface and Diara surface. Entisols (potash and lime) and inceptisols (potash, lime and kanker) are the major soil types in the Vaishali district. Ouaternary alluvial formations, comprised of alternating layers of sand, silt, clay and gravel, formed unconfined and semiconfined aquifer in this region (CGWB 2007). In the shallow unconfined aquifer, the transmissivity ranges from 1000 to 5000 m²/day, while specific yield is between 8 and 12%. Groundwater level ranges from 3 to 9 mbgl and from 1 to 5 mbgl during pre- and post-monsoons, respectively.

Groundwater sampling and analysis

Detailed fieldwork was performed in the upper Ganges basin (UGB) and lower Ganges basin (LGB) to locate the suitable wells for water sampling (Fig. 1). Groundwater samples were collected from 44 and 26 bore wells in the UGB and LGB, respectively, using pre-cleaned 500-ml HDPE bottles in duplicate after the stabilisation of field parameters and stored at 4 °C until analysis. Temperature, electrical conductivity (EC) and pH were estimated in the field using portable metres (SevenGo Duo SG23, Mettler Toledo). Well-depth details were received from owners and the local populace. Handheld Garmin Global Positioning System (GPS) was used to mark the coordinates of

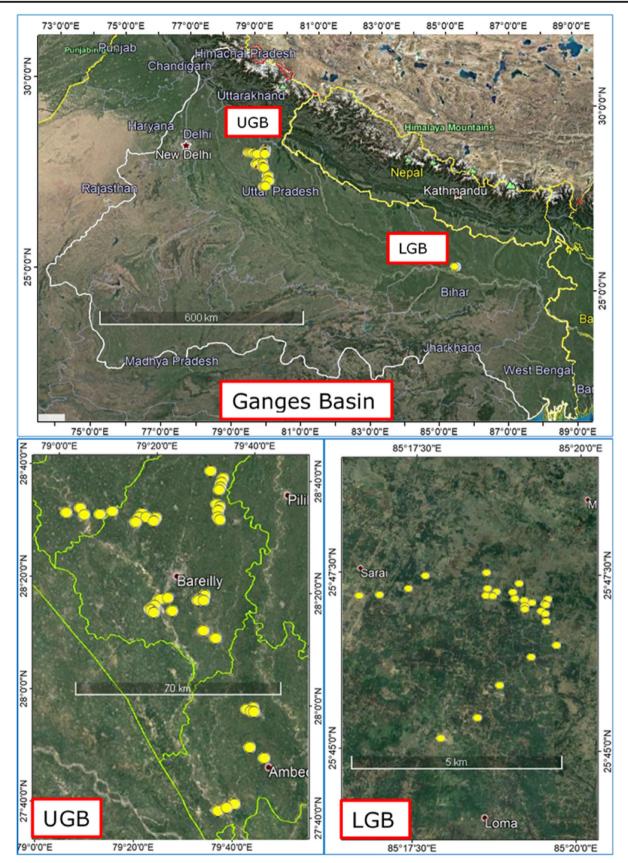


Fig. 1 Maps show the groundwater sampling locations in the upper (UGB) and lower (LGB) Ganges basins and other features

the wells, which were plotted on the location map (Fig. 1). Groundwater sampling, storage and analysis were performed based on the international standard procedure (Keefe et al. 2003; APHA 2017). Bicarbonate and carbonate were analysed using titration and major and minor ions were measured using ion chromatography (Thermo Scientific, ICS 5000+). The concentration of Fe, Mn and Zn was analysed by atomic absorption spectrophotometer (AAS4141, ECIL). The precision and measurement repeatability of each analysis was < 2% and the ion balance error calculated was within $\pm 5\%$.

Data analysis

Water quality data were utilised to calculate the drinking water quality index (DWQI) and were compared with international standards to assess the groundwater suitability for drinking uses. Further, data analysis was carried out using various software. SPSS (v16.0) and ArcGIS (v10.2) were employed for chemometric analysis and spatial maps preparation, respectively. Hydrochemical facies and water types were identified through Piper diagram using Rockworks (v15.0) software.

Drinking water quality index (DWQI)

DWQI is widely used by several researchers for appraising groundwater quality for drinking uses worldwide (Jasmin and Mallikarjuna 2014; Kumar et al. 2014; Zaidi et al. 2015; Khan and Jhariya 2017; Adimalla and Qian 2019; Muzenda et al. 2019). DWQI is a dimensionless single value, created using multiple variables analysed in the water samples. In the DWQI calculation, weight and rate have a major role. In the first step, weight (w_j) is assigned for each parameter based on its relative importance on water quality and human health (Zaidi et al. 2015; Adimalla 2020). The weight (w_j) is generally between 2 and 5 and Table 1 shows the assigned weight and relative weight of each parameter. Parameters, namely, TDS, F⁻ and NO₃⁻, are

| Table 1 DWQI—weight assigned and relative Image: Comparison of the second sec | Parameter | Weight | Relative |
|---|------------------------------|--------|----------|
| weight of each parameter | pН | 3 | 0.079 |
| | TDS | 5 | 0.132 |
| | TH | 3 | 0.079 |
| | Na ⁺ | 2 | 0.053 |
| | K^+ | 2 | 0.053 |
| | Ca ²⁺ | 3 | 0.079 |
| | Mg ²⁺ | 3 | 0.079 |
| | Cl | 4 | 0.105 |
| | SO_4^{2-} | 3 | 0.079 |
| | NO ₃ ⁻ | 5 | 0.132 |
| | F ⁻ | 5 | 0.132 |
| | | | |

assigned for a maximum weight of 5 and low weight is assigned to Na^+ and K^+ .

The relative weight (W_j) of the " j_{th} " parameter is calculated from its assigned weight (w_j) using the following Eq. (1).

$$W_j = \frac{w_j}{\sum_j^n w_j} \tag{1}$$

The rate of each parameter is estimated using the following relation (Eq. 2).

$$Q_{j} = \left\{ \frac{C_{j} - C_{jp}}{S_{j} - C_{jp}} \right\} *100$$
(2)

where Q_j is the rate of " j_{th} " parameter; C_j is the concentration of " j_{th} " parameter; S_j is permissible limit of " j_{th} " parameter in the drinking water recommended by the WHO (2011); C_{jp} is the ideal value of " j_{th} " parameter in pure water (equal to zero except pH = 7); and *n* is the number of parameters.

The sub-index (SI_j) is calculated using Eq. 3.

$$SI_j = W_j X Q_j \tag{3}$$

Finally, DWQI is obtained using Eq. 4.

$$DWQI = \sum_{j=1}^{n} SI_j \tag{4}$$

where SI_j is sub-index of " j_{th} " parameter and W_j and Q_j are the relative weight and rate of " i_{th} " parameter.

Chemometric methods

In this study, chemometric methods, namely, principal component analysis (PCA) and hierarchical cluster analysis (HCA), were used to assess the contamination sources. Initial data set obtained from chemical analysis were standardised using SPSS v16 (IBM Corporation, NY, USA) and transformed into z-score to remove the bias caused by the individual variables and to get a normal distribution. PCA was carried out using Varimax rotation with Kaiser normalisation. Factors with an eigenvalue greater than one are extracted and considered for discussion. HCA analysis was employed to identify the wells with similar water chemistry as well as to explore the relationship between the variables. In this study, R-mode (variables) and Q-mode (wells) HCA analysis were executed with standardised variables (z-score). Squared Euclidean distance with Ward's method was used.

Results and discussion

weight

Groundwater quality in the Ganges River basin (GRB)

Groundwater samples (n = 70) collected from both LGB and UGB are alkaline and the pH varies from 7 to 8.3 with a mean value of 7.5 (Table 2). The groundwater pH suggests that

Table 2 Descriptive statistics of physiochemical parameters analysed in the groundwater and suitability assessment for drinking

| Parameters | Min | Max | Average | STD | Kurtosis | Skewness | WHO (2011) | | | | | | |
|-------------------------------|------|------|---------|------|----------|----------|----------------------------------|-------------------------------------|------------------------------------|------------------------------------|--|--|--|
| | | | | | | | Highest desirable limit (HDL) | Maximum allowable limit (MAL) | No. of samples (%) exceeded HDL | No. of samples (%) exceeded MAL | | | |
| Depth | 6.1 | 53.3 | 21 | 14 | -0.7 | 1.0 | | | | | | | |
| рН | 7.0 | 8.3 | 7.5 | 0.27 | 0.9 | 0.9 | 6.5-7.5 | 6.5–9.2 | 24 (34) | 0 | | | |
| EC | 270 | 2000 | 843 | 338 | 1.6 | 1.1 | | | | | | | |
| TDS | 173 | 1280 | 540 | 216 | 1.6 | 1.1 | 500 | 1500 | 34 (49) | 0 | | | |
| TH | 108 | 536 | 297 | 100 | -0.49 | 0.33 | 100 | 500 | 70 (100) | 1 | | | |
| Na ⁺ | 11 | 233 | 52 | 36 | 9.5 | 2.6 | - | 200 | - | 1 | | | |
| K^+ | 1 | 173 | 16 | 33 | 10 | 3.2 | - | 12 | - | 14 (20) | | | |
| Ca ²⁺ | 21 | 101 | 49 | 19 | 0.0 | 0.7 | 75 | 200 | 7 (10) | 0 | | | |
| Mg ²⁺ | 13 | 95 | 42 | 23 | -0.5 | 0.8 | 50 | 150 | 21(30) | 0 | | | |
| Cl | 25 | 355 | 94 | 60 | 3.9 | 1.5 | 200 | 600 | 3 (4) | 0 | | | |
| HCO ₃ ⁻ | 97 | 651 | 325 | 123 | -0.6 | 0.2 | | | | | | | |
| $SO_4^{2^-}$ | 7 | 147 | 55 | 35.9 | -0.3 | 0.7 | 200 | 400 | 0 | 0 | | | |
| NO ₃ | BDL | 37.7 | 4.5 | 8.14 | 7.2 | 2.7 | 45 | - | 0 | - | | | |
| PO4 ³⁻ | BDL | 1.3 | 0.09 | 0.18 | 32 | 5.0 | | | | | | | |
| F ⁻ | 0.04 | 1.49 | 0.32 | 0.33 | 1.5 | 1.5 | - | 1.5 | - | 0 | | | |

Unit: mg/l except for pH, EC (μ S/cm). TH—mg/L as CaCO₃. Number of samples (*n*) = 70

dissolved carbonates are mostly present in the form of HCO₃⁻ (Adams et al. 2001). EC ranges from 270 to 2000 μ S/cm with an average of 843 μ S/cm and the total dissolved solids (TDS) is between 173 and 1280 mg/l with a mean value of 540 mg/l. Groundwater is fresh (TDS < 1000 mg/l; *n* = 67) and the TDS is less than 500 mg/l in 51% of wells. Groundwater hardness (TH) varies from 108 to 536 mg/l with an average of 297 mg/l. In this aquifer, Ca²⁺ + Mg²⁺ and HCO₃⁻ are dominated over Na⁺ + K⁺ and Cl⁻ + SO₄²⁻ in 91 and 71% of samples, respectively.

Groundwater quality in the UGB and LGB

The groundwater quality in the UGB and LGB is illustrated in the Figs. 2, 3 and 4. The average concentrations of analysed variables suggest that groundwater in the LGB (n = 26) has high EC (866 µS/cm), TDS (554 mg/l), TH (374 mg/l), HCO₃⁻ (407 mg/l), Mg²⁺ (62 mg/l) and Zn (614 µg/l) concentrations (Fig. 2). Likewise, the average concentration of Mn (604 µg/l), Fe (564 µg/l), SO₄²⁻ (65 mg/l), Cl⁻ (104 mg/l), K⁺ (24 mg/l), Na⁺ (59 mg/l), NO₃⁻ (5.76 mg/l) and Ca²⁺ (50 mg/l) in the groundwater is higher in the UGB (n = 44) compared to LGB (n = 26).

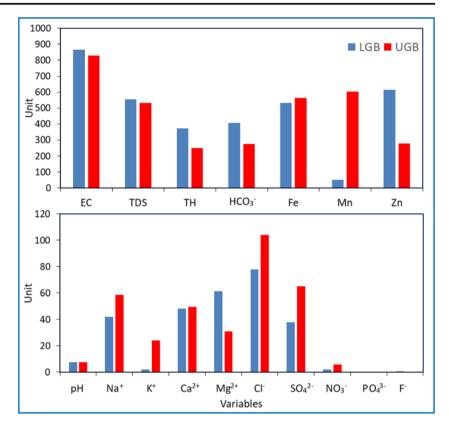
Figure 3 illustrates the groundwater chemistry in the shallow and deep wells in the UGB and LGB. Vertical distribution pattern depicts that the concentrations of Na⁺, K⁺, Mg²⁺, HCO₃⁻, NO₃⁻, F⁻, Fe, Mn and Zn in the groundwater are not varying with depth in the LGB. However, a high concentration of TDS, EC, Ca²⁺, Cl⁻ and SO₄²⁻ is observed in the shallow wells. A similar trend is also observed in the UGB. In the UGB, shallow wells have high concentrations of TDS, EC, major ion, NO₃⁻, PO₄³⁻, F⁻, Fe and Mn compared to the deeper one. These observations evidenced that shallow aquifer is highly vulnerable to contamination and affected by the surface pollution sources especially in the UGB.

Groundwater samples were plotted on the Piper diagram (Piper 1944) to identify the chemical facies and water types (Fig. 4). Figure 4 exhibits that groundwater samples fall on three facies, namely, CaHCO₃, CaMgCl and CaNaHCO₃; but, CaHCO₃ is dominated over CaMgCl water type. In the LGB, CaHCO₃ is a predominant water type, whereas all the three facies are observed in the UGB (Fig. 4). Groundwater with high fluoride and from deep wells is plotted on CaHCO₃ zone, which is predominantly regulated by the carbonate and silicate mineral dissolution (Fig. 4). Similarly, wells with high NO₃⁻, Cl⁻ and SO₄⁻ belong to the CaMgCl water type. Groundwater chemistry in the CaMgCl water type is likely affected by the infiltration of wastewater from the surface and subsurface contamination sources (Nagarajan et al. 2010).

Groundwater suitability assessment for drinking

As groundwater is a major source of water for drinking and domestic uses in the study regions, groundwater quality data were compared with international drinking water standards recommended by the WHO (2011) (Table 2). According to Davis and De Weist (1966) classification, groundwater in 51 and 96% of wells is desirable (TDS < 500 mg/l) to permissible (TDS < 1000 mg/l) classes for drinking, respectively. Table 2 shows that groundwater hardness exceeded the highest desirable limit (HDL) but within the maximum allowable limit (MAL) recommended by the WHO (2011) for drinking. Based on hardness classification (Sawyer and McCarty 1967), the groundwater samples are classified into moderately

Fig. 2 Average concentrations of the variables analysed in the groundwater. Unit—mg/l except for pH, EC (μ S/cm) and trace metals (Fe, Mn and Zn, μ g/l)



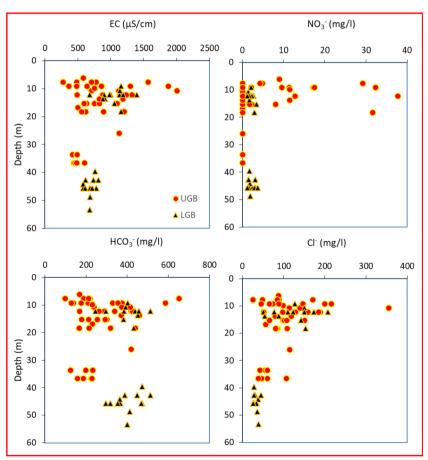
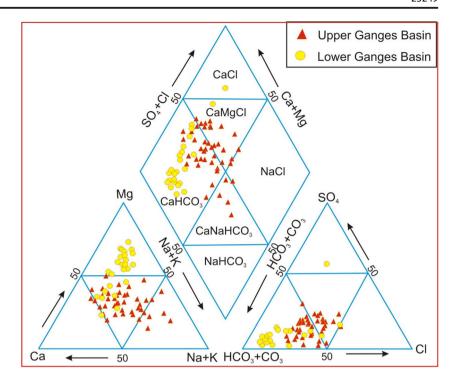


Fig. 3 Vertical variation of selected variables in these aquifers

Fig. 4 Piper plot explains the hydrochemical facies



hard (150<TH>75, 6%), hard (300<TH>150, 54%) and very hard (TH>300, 40%) classes. Hard water generally causes an unpleasant taste and reduces the lather-producing ability of soap and encrustation in the plumping fixtures and pipe network (Sawyer and McCarty 1967). Ca²⁺ and Mg²⁺ are the most essential elements of human health. The concentration of Ca²⁺ and Mg²⁺ in the groundwater is within the MAL; but 7 and 21% of samples, respectively, exceeded the HDL recommended for drinking. In these regions, groundwater Ca²⁺ ranges from 21 to 101 mg/l with an average of 49 mg/l, while the Mg²⁺ content varies from 13 to 95 mg/l with a mean value of 42 mg/l (Table 2). Groundwater with high Mg²⁺ may be laxative, cathartic and diuretic. Similarly, groundwater with high Ca²⁺ may cause irritation in urinary passage and kidney/bladder stones for consumers (Kaushik 2002).

The concentration of Na⁺ is between 11 and 233 mg/l with a mean value of 52 mg/l and is less than 200 mg/l in the groundwater except one well (Table 2). The groundwater K⁺ varies from 1 to 173 mg/l with a mean value of 16 mg/l. Table 2 shows that 20% samples exceeded the MAL (K^+ > 12 mg/l) and are unfit for human consumption. Generally, K^+ is an essential element for the biological cycle of human beings. However, high K⁺ in drinking water may cause a laxative effect for consumers. The concentration of Cl⁻ ranges from 25 to 355 mg/l with an average of 94 mg/l in the groundwater and it is lesser than the HDL except for three wells (Table 2). Table 2 depicts that SO_4^{2-} and NO_3^{-} concentrations in the groundwater are lower than the WHO-recommended HDL. Likewise, groundwater F⁻ concentration is also within the prescribed limit for drinking ($F \le 1.5 \text{ mg/l}$) and it is less than 1.3 mg/l except for one well. In this basin, F⁻ concentration varies from 0.04 to 1.49 mg/l with an average of 0.32 mg/l (Table 2). F⁻ is an essential element for human health; but, high concentration (F > 1.5 mg/l) leads to skeletal fluorosis, calcification of ligaments and endemic fluorosis for consumers (Furi et al. 2011; Morales-Arredondo et al. 2016; Olaka et al. 2016; Enalou et al. 2018; Elumalai et al. 2019).

Table 3 Descriptive statistics of metals analysed in the groundwater and suitability assessment for drinking

| Parameters | Min | Max | Average | STD | Kurt | Skew | WHO guideline value (GV) (µg/l) | USEPA maximum contaminant level (MCL) (µg/l) | No. of samples (%) exceeded GV | No. of samples (%) exceeded MCL |
|------------|-----|------|---------|-----|------|------|------------------------------------|--|-----------------------------------|------------------------------------|
| Fe | BDL | 2436 | 552 | 521 | 3.1 | 1.7 | 1000 | 300 | 13 (19) | 43 (61) |
| Mn | 20 | 3592 | 398 | 627 | 10.1 | 2.9 | 400 | 50 | 19 (27) | 54 (77) |
| Zn | BDL | 1905 | 403 | 376 | 3.0 | 1.5 | 3000 | 5000 | 0 | 0 |

Unit: $\mu g/l$. Number of samples (*n*) = 70

Trace metals content (average) in the groundwater indicates that Fe is dominating over Zn and Mn (Table 3). The concentration of Fe varies from BDL to 2436 µg/l with a mean value of 552 µg/l. Drinking water suitability assessment suggests that 19 and 61% of samples exceeded the guideline value (GV) (1000 µg/l) and maximum contaminant limit (MCL) (300 μ g/l) prescribed by the WHO (2011) and USEPA (2012), respectively, for drinking (Table 3). The groundwater Mn concentration ranges from 20 to $3592 \mu g/l$ with an average of $398 \mu g/l$. Like Fe, Mn content in 27 and 77% of samples exceeded the WHO GV and USEPA MCL, respectively, for drinking (Table 3). Table 3 depicts that Zn concentration is within the prescribed limit for drinking utilities. The concentration of Zn is between BDL and 1905 μ g/l with a mean value of 403 μ g/ 1 (Table 3). In both LGB and UGB, Zn concentration in the groundwater is not varying with depth.

Drinking water quality index (DWQI)

DWOI is frequently applied in several studies to assess the groundwater quality for drinking. Based on the DWQI values, drinking water is classified into five classes, namely, excellent (DWQI < 50), good (50–100), poor (100–200), very poor (200-300) and not suitable for consumption (> 300) (Adimalla et al. 2018; Jehan et al. 2020). In the study sites, DWQI ranges from 14 to 112 with an average of 34 (n = 70). According to DWQI classification, groundwater in these regions comes under excellent (89%) to good (10%) classes (Fig. 5). Moreover, groundwater in the LGB falls under excellent class (DWQI < 46). In the UGB, DWQI varies from 14 to 112 with a mean value of 35. Further, 82 and 16% of water samples are classified as excellent and good classes, respectively. However, the high concentrations of Fe, Mn and total hardness in the groundwater restrict the usage for drinking in the study sites (Tables 2 and 3).

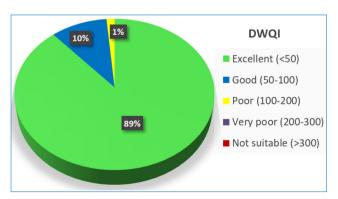


Fig. 5 Groundwater quality assessment using drinking water quality index (DWQI) in the study region

Chemometric methods

Principle component analysis

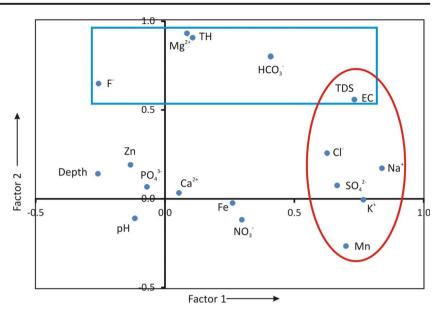
Principle component analysis (PCA) was performed in this study and six factors with eigenvalue > 1 were selected, which explains 79% of the total variance (Table 4). Factor 1 (23% of the total variance) has a high positive loading of EC, TDS, Na⁺, K⁺, Cl⁻, SO₄²⁻ and Mn (Fig. 6; Table 4). Aforementioned, shallow wells have high Cl^{-} and SO_4^{2-} compared to the deeper one (Fig. 3); thus, this factor explains the contribution of anthropogenic influences on water chemistry. A similar result is reported by Kumar et al. (2018). Further, positive loading of Mn in factor 1 suggests that Mn is partially/completely derived from surface contamination sources as fireworks, fertilisers, fungicides, varnish, bleaching and livestock food supplements are common sources for Mn (WHO 2011). The shallow aquifer in the UGB has a high concentration of Mn compared to the deeper one. Factor 2 (20% of total variance) is positively loaded by EC, TDS, TH, Mg^{2+} , HCO_3^- and F^- (Fig. 6; Table 4). This factor ascribes the role of carbonate and silicate mineral dissolution in the water chemistry especially in the LGB as HCO₃⁻ and F⁻ are not varying with depth. But, in the UGB, shallow wells also have elevated in HCO3⁻ and F⁻ concentrations, which seems to be derived by anthropogenic sources or infiltration of

 Table 4
 Results of principal component analysis

| | - | - | - | | | |
|-------------------------------|-------|-------|-------|-------|-------|-------|
| Variable | F1 | F2 | F3 | F4 | F5 | F6 |
| Depth | -0.26 | 0.14 | -0.67 | 0.38 | -0.04 | -0.05 |
| EC | 0.73 | 0.56 | 0.34 | -0.12 | -0.02 | -0.07 |
| TDS | 0.73 | 0.56 | 0.34 | -0.12 | -0.02 | -0.07 |
| TH | 0.11 | 0.91 | 0.33 | 0.00 | 0.04 | 0.03 |
| рН | -0.12 | -0.11 | -0.05 | 0.82 | 0.06 | -0.07 |
| Na ⁺ | 0.84 | 0.17 | 0.06 | -0.10 | 0.11 | -0.07 |
| K ⁺ | 0.77 | -0.01 | -0.04 | -0.17 | -0.37 | -0.03 |
| Ca ²⁺ | 0.05 | 0.03 | 0.86 | 0.10 | -0.12 | 0.09 |
| Mg ²⁺ | 0.09 | 0.93 | -0.09 | -0.05 | 0.10 | -0.01 |
| Cl | 0.63 | 0.26 | 0.59 | -0.17 | -0.03 | -0.15 |
| HCO ₃ - | 0.41 | 0.80 | -0.16 | 0.07 | 0.04 | 0.07 |
| SO4 ²⁻ | 0.67 | 0.07 | 0.32 | -0.39 | 0.09 | -0.07 |
| NO ₃ | 0.30 | -0.12 | 0.07 | -0.25 | -0.68 | 0.06 |
| PO ₄ ³⁻ | -0.07 | 0.07 | 0.06 | -0.03 | -0.05 | 0.96 |
| F- | -0.26 | 0.65 | -0.12 | 0.38 | -0.15 | -0.01 |
| Fe | 0.26 | -0.02 | -0.04 | -0.15 | 0.75 | 0.01 |
| Mn | 0.70 | -0.27 | 0.07 | 0.02 | 0.28 | 0.36 |
| Zn | -0.13 | 0.19 | -0.04 | 0.81 | 0.01 | 0.05 |
| Variance (%) | 23 | 20 | 12 | 11 | 7 | 6 |
| Cumulative variance (%) | 23 | 43 | 55 | 66 | 72 | 79 |
| | | | | | | |

Rotation method: Varimax rotation with Kaiser normalisation

Fig. 6 Result of principal component analysis and factor loading details



contaminated water that triggered the dissolution of respective minerals in the vadose zone (Rajmohan et al. 2017). Earlier studies also reported high fluoride in the shallow aquifer in the UGB (Sinha and Saxena 2006; CWC/NRSC 2014; Rajmohan and Amarasinghe 2016; Kumar et al. 2018). In the UGB, application of phosphate fertilisers and irrigation return flow are likely to promote the F⁻ enrichments in the shallow wells (Datta et al. 1996; Farooqi et al. 2007; Kundu and Mandal 2008; Brindha and Elango 2013).

Factor 3 explains 12% of the total variance and has high positive loading of Ca²⁺ and Cl⁻ and negative loading of depth. This factor justifies that enhancement of Ca²⁺ and Cl⁻ in the shallow wells is mainly due to the recharge of wastewater from the surface and subsurface sources (Rajmohan and Amarasinghe 2016). Factors 4 and 5 explain 11 and 7% of the total variance, respectively, and have positive loading of pH, Fe and Zn and negative loading of NO₃⁻. A similar observation is reported by Paul et al. (2019) and Palmucci et al. (2016) and suggested that metal mobility is governed by the redox process. Rajmohan et al. (2020) reported that the reductive dissolution of Fe-Mn oxyhydroxides in the reducing environment resulted in the accumulation of Fe, Mn and other trace metals in the vadose zone in the LGB. Sankar et al. (2014) also reported similar observations in the LGB. Besides, the dissolution of carbonates (smithsonite (ZnCO₃) and rhodochrosite (MnCO₃)) is likely to contribute Zn and Mn concentrations in the groundwater. Reductive dissolution process and denitrification in the aquifer will increase the pH because the denitrification process consumes acidic proton $(5CH_2O + 4NO_3^- +$ $4H^+ \Rightarrow 5CO_2 + 2N_2 + 7H_2O$), which enhances pH in the groundwater. Figure 3 depicts that groundwater in the shallow wells has high nitrate in comparison to the deeper one. Hence, denitrification seems to be possible only in the deep wells not in the shallow wells. Overall, groundwater NO3⁻ in the shallow wells is derived from pollution sources and the nitrification process, whereas low NO₃⁻ concentrations in the deep wells are due to the denitrification process. Factor 6 (only 6% of the total variance) has high loading of PO₄³⁻ and corresponds to agricultural source. Overall, factors 1, 3 and 6 suggest the role of wastewater recharge from surface and subsurface, whereas factors 2, 4 and 5 indicate the impact of mineral dissolution and denitrification on groundwater chemistry. Earlier studies in the UGB reported that excessive fertiliser application, irrigation return flow, improper disposal of domestic sewage water, leakage from septic tank and accumulation of cattle waste are common contamination sources that affected the groundwater quality (Somasundaram et al. 1993; Chakraborti et al. 2011; Khan et al. 2015; Rajmohan and Amarasinghe 2016).

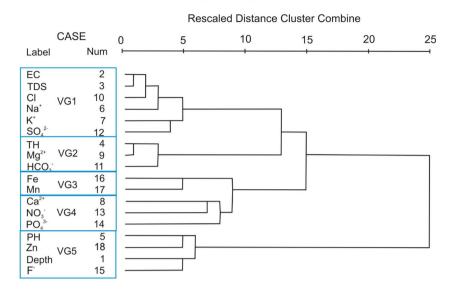
Hierarchical cluster analysis (HCA)

In this study, R-mode (variable) HCA gave five major clusters (Fig. 7). EC, TDS, Na⁺, K⁺, Cl⁻ and SO₄²⁻ are associated with cluster VG1, whereas Ca²⁺, NO₃⁻ and PO₄²⁻ are in cluster VG4. Both clusters VG1 and VG4 suggested the influences of anthropogenic sources on water chemistry. Similarly, clusters VG2 (TH, Mg²⁺ and HCO₃⁻) and VG3 (Fe and Mn) indicate the effect of mineral dissolution on water quality. The association of depth, pH, Zn and F⁻ in the cluster VG5 explains the relation between depth and pH and Zn and F⁻. Results obtained from PCA and HCA are consistent with the source of contamination identified.

Q-mode (wells) HCA also resulted in five clusters (Fig. 8). Depth-wise distribution and groundwater chemistry of each cluster are illustrated in Fig. 9. Table 5 shows the descriptive statistics of groundwater variables in each cluster group in the study region. In the five clusters, wells in the LGB are

Fig. 7 Result of R-mode (variable) HCA analysis in the study region

Dendrogram using Ward Method



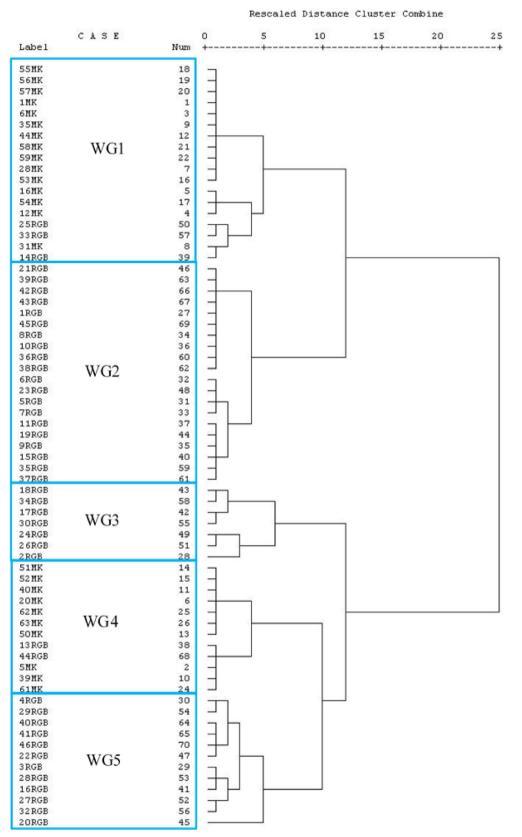
associated with WG1 and WG4, whereas wells in the UGB fall in WG2, WG3 and WG5. The average values of EC and TDS indicate that wells in WG3 are highly mineralised and the clusters are in the order of WG3 > WG4 > WG5 > WG1 > WG2 based on salinity. Pictorial representation of each cluster's water chemistry is given as a Stiff diagram (Fig. 9). Groundwater in the cluster WG2 has low ionic concentrations (Fig. 9, Table 5).

In the WG1 cluster (n = 18), the average concentrations of Zn and F⁻ are high and wells in this cluster are generally deep (Table 5; Fig. 9). Besides, the average concentrations of Cl⁻, SO₄²⁻ and Mn are low in the groundwater (Table 5). EC and TDS vary from 457 to 1186 μ S/cm and from 292 to 759 mg/l with an average of 710 μ S/cm and 454 mg/l, respectively. In WG1, groundwater in nine wells has elevated concentration of Fe and Mn and exceeded the safe limit (Fe > 300 μ g/l; Mn >

 Table 5
 Descriptive statistics of the cluster groups

| | Depth | EC | TDS | TH | pН | Na ⁺ | K^+ | Ca ²⁺ | Mg ²⁺ | Cl | HCO ₃ - | SO4 ²⁻ | NO ₃ - | PO4 ³⁻ | F | Fe | Mn | Zn |
|--------|---------|------|------|------|------|-----------------|-------|------------------|------------------|------|--------------------|-------------------|-------------------|-------------------|------|------|------|------|
| WG1 (| n = 18) | | | | | | - | _ | | | | | | | | | | |
| Mean | 36 | 710 | 454 | 307 | 7.7 | 41 | 3 | 45 | 47 | 54 | 371 | 23 | 1.8 | 0.1 | 0.6 | 343 | 87 | 770 |
| STD | 14 | 156 | 100 | 63 | 0.3 | 7 | 1 | 21 | 18 | 40 | 90 | 13 | 1.1 | 0.1 | 0.4 | 173 | 88 | 404 |
| Kurt | -0.1 | 4.6 | 4.6 | -0.8 | 0.3 | 0.7 | 5.4 | 1.0 | -0.7 | 0.9 | -1.0 | -0.2 | -0.2 | 3.7 | -0.5 | -0.2 | 1.8 | 2.6 |
| Skew | -1.27 | 1.62 | 1.62 | -0.4 | 1.03 | -0.6 | 2.42 | 1.20 | -0.1 | 1.52 | -0.26 | 0.96 | -0.28 | 2.09 | 0.36 | 0.59 | 1.80 | 1.6 |
| WG2 (| n = 20) | | | | | | | | | | | | | | | | | |
| Mean | 17 | 555 | 355 | 201 | 7.5 | 31 | 7 | 45 | 22 | 70 | 186 | 48 | 3.3 | 0.0 | 0.1 | 330 | 197 | 222 |
| STD | 11 | 157 | 100 | 54 | 0.3 | 16 | 8 | 16 | 6 | 29 | 47 | 29 | 5.2 | 0.0 | 0.1 | 312 | 109 | 191 |
| Kurt | -0.7 | 0.0 | 0.0 | -0.6 | -0.1 | -0.3 | 14.3 | -1.0 | -0.9 | 1.2 | 0.0 | -0.8 | 1.5 | 5.4 | 0.1 | 2.8 | 0.4 | 0.1 |
| Skew | 0.96 | 0.56 | 0.56 | -0.0 | 0.33 | 0.61 | 3.54 | 0.26 | 0.20 | 0.94 | 0.19 | 0.51 | 1.55 | 2.24 | 1.22 | 1.63 | 0.89 | 0.8 |
| WG3 (4 | n = 7) | | | | | | | | | | | | | | | | | |
| Mean | 11 | 1486 | 951 | 338 | 7.3 | 123 | 93 | 53 | 50 | 195 | 461 | 106 | 16.6 | 0.0 | 0.2 | 394 | 743 | 141 |
| STD | 4 | 272 | 174 | 72 | 0.2 | 66 | 62 | 20 | 20 | 42 | 123 | 42 | 16.6 | 0.1 | 0.1 | 539 | 453 | 162 |
| Kurt | 2.6 | -1.3 | -1.3 | -0.4 | -1.1 | 0.4 | -1.3 | -1.9 | -0.9 | 2.8 | -0.4 | 2.9 | -2.4 | 7.0 | 0.3 | 1.7 | -0.1 | -2.1 |
| Skew | 1.45 | 0.74 | 0.74 | 0.40 | -0.4 | 0.80 | -0.2 | -0.1 | 1.06 | 1.41 | 0.96 | -1.5 | 0.21 | 2.65 | 1.31 | 1.46 | 1.28 | 0.6 |
| WG4 (4 | n = 12) | | | | | | | | | | | | | | | | | |
| Mean | 13 | 1098 | 703 | 443 | 7.4 | 47 | 2 | 57 | 73 | 135 | 406 | 63 | 1.8 | 0.1 | 0.5 | 544 | 138 | 498 |
| STD | 2 | 143 | 91 | 48 | 0.3 | 14 | 1 | 27 | 20 | 46 | 79 | 23 | 1.0 | 0.1 | 0.3 | 248 | 206 | 281 |
| Kurt | 2.4 | 0.4 | 0.4 | -0.3 | 5.8 | 1.9 | 5.3 | -1.5 | -1.6 | -0.6 | 1.0 | 2.6 | -0.2 | 3.2 | -0.1 | -0.6 | 5.3 | -0.1 |
| Skew | 1.06 | 0.34 | 0.34 | 0.32 | 2.04 | 1.73 | 2.30 | 0.46 | -0.6 | -0.4 | -1.18 | -0.4 | -0.42 | 1.89 | 0.21 | 0.39 | 2.40 | -0.1 |
| WG5 (4 | n = 12) | | | | | | | | | | | | | | | | | |
| Mean | 14 | 905 | 579 | 272 | 7.4 | 70 | 21 | 51 | 35 | 102 | 321 | 79 | 6.0 | 0.2 | 0.1 | 1205 | 1291 | 225 |
| STD | 5 | 182 | 117 | 44 | 0.2 | 26 | 20 | 11 | 13 | 24 | 80 | 26 | 9.3 | 0.4 | 0.0 | 642 | 951 | 333 |
| Kurt | 0.9 | -1.1 | -1.1 | -0.9 | -1.2 | -0.9 | 1.2 | -1.2 | 1.4 | 0.0 | -0.7 | -1.2 | 2.5 | 9.5 | 6.1 | 0.5 | 2.0 | 7.1 |
| Skew | 0.93 | 0.22 | 0.22 | -0.1 | -0.1 | 0.21 | 1.39 | -0.3 | 1.10 | 1.06 | 0.00 | 0.71 | 1.68 | 2.97 | 2.20 | 0.42 | 1.31 | 2.5 |

Note: Unit-mg/l except depth (mbgl), pH, EC (µS/cm), Fe (µg/l), Mn (µg/l) and Zn (µg/l). Bold—high concentrations among the groups



Dendrogram using Ward Method

Fig. 8 Result of Q-mode (wells) HCA analysis of groundwater in the study region

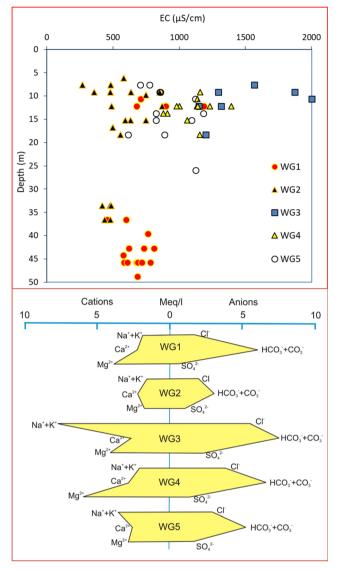


Fig. 9 Salinity distribution and groundwater chemistry (Stiff diagram) in the cluster groups

50 µg/l) recommended by the USEPA. Figure 9 illustrates that groundwater in the WG2 cluster (n = 20) is less mineralised and the average concentrations of most of the variables such as EC, TDS, TH, Ca²⁺, Mg²⁺, Na⁺, HCO₃⁻ and Fe are recorded low in comparison to other clusters (Table 5). Figure 9 also shows that most of the wells in this cluster are shallow and the depth ranges from 6 to 37 m with an average of 17 m. The EC varies from 270 to 872 µS/cm with a mean value of 555 µS/cm, whereas the TDS is between 173 and 558 mg/l with an average of 355 mg/l. The concentration of Mn is generally higher than the USEPA drinking water standards (Mn > 50 µg/l; n = 20). Likewise, eight wells show elevated concentration of Fe and exceeded the USEPA-recommended MCL. Groundwater chemistry in WG1 and WG2 clusters is governed by natural processes.

Among the five clusters, wells in WG3 (n = 7) have high concentrations of most of the parameters and the depth of

these wells is from 8 to 18 m. The EC and TDS range from 1150 to 2000 μ S/cm and from 736 to 1280 mg/l with a mean value of 1486 μ S/cm and 951 mg/l, respectively. The average concentration of Cl⁻, SO₄²⁻ and NO₃⁻ in the groundwater is higher than the other clusters and wells in the WG3 are strongly affected by the wastewater recharge from the surface and subsurface sources. Groundwater Mn content exceeded the safe limit recommended both by the WHO (Mn > 400 μ g/l) and USEPA (Mn > 50 μ g/l). Groundwater Fe concentration in three wells exceeded the USEPA-recommended MCL. However, groundwater Zn in these wells is low compared to other clusters.

In cluster WG4 (n = 12), the borewell depth varies from 9 to 18 m with a mean value of 13 m (Table 5, Fig. 9). The average TDS and EC in groundwater are 703 mg/l and 1098 µS/cm, respectively. In this cluster, groundwater has high TH, Ca^{2+} and Mg^{2+} and lowest K⁺ and NO_3^{-} compared to other clusters. The HCO₃⁻ concentration in groundwater is from 246 to 513 mg/l with a mean value of 406 mg/l, which is higher than other clusters except for WG3. Similarly, the average Cl concentration in this cluster is high next to WG3. Hence, groundwater chemistry in these wells is regulated by both mineral dissolution and anthropogenic sources. Moreover, the recharge of wastewater likely triggered the carbonate and silicate mineral dissolution occurred in the vadose zone. Among the 12 wells, groundwater in 10 and 6 wells shows a high concentration of Fe and Mn concentrations, respectively, which exceeded the USEPA safe limit for drinking.

Groundwater in the WG5 cluster (n = 12) is less mineralised compared to WG3 and WG4 and more mineralised compared to WG1 and WG2. The average concentration of PO₄³⁻, Fe, Mn Na⁺, K⁺, SO₄²⁻ and NO₃⁻ in the groundwater is higher than other clusters except WG3. Further, wells in this cluster existed in the UGB where shallow wells have a high concentration of Fe and Mn. Aforementioned, shallow wells in the UGB are affected by anthropogenic activities, which enhanced the solute load in the groundwater. Moreover, infiltration of wastewater likely facilitated the dissolution of Fe and Mn minerals in the vadose zone, which accumulated these metals in the groundwater. In this cluster, groundwater in 9 and 11 wells exceeded the recommended Fe and Mn concentrations, respectively, for drinking by the WHO. Likewise, groundwater in 11 and 12 wells exceeded the safe limit of Fe and Mn concentrations, respectively, recommended by the USEPA for drinking.

Conclusions

Drinking water quality index and chemometric methods were successfully employed to assess the groundwater quality and to identify the contamination sources in the upper (UGB) and lower Ganges basin (LGB). The key findings are summarised below.

- Groundwater is desirable (51%) to permissible (TDS < 1000 mg/l, 96%) classes for human consumption. According to DWQI values, groundwater is excellent (89%) to good (10%) classes. However, Fe and Mn concentrations in groundwater surpassed the safe limit for drinking recommended by the WHO (19% (Fe) and 27% (Mn)) and USEPA (61% (Fe) and 77% (Mn)).
- In both LGB and UGB, groundwater in the shallow wells has elevated concentration of TDS, EC, major and minor ions, Fe and Mn, which implies that shallow wells are affected by the pollution sources from the surface especially in the UGB.
- PCA and HCA analysis implied that groundwater chemistry is greatly influenced by both anthropogenic activities and natural processes. Weathering of carbonate and silicate minerals and reductive dissolution of Fe-Mn oxyhydroxides affected the groundwater chemistry.
- HCA (R-mode) resulted in five major clusters and clusters VG1 and VG4 suggested the impact of anthropogenic sources; but, clusters VG2 and VG3 indicated the effect of mineral dissolution on water chemistry. Cluster VG5 explained the relationship between well depth and pH and Zn and F.
- Q-mode HCA also resulted in five clusters and ensured that groundwater chemistry in WG1 and WG2 wells is governed by the natural processes whereas wells in WG3 and WG5 are strongly affected by the anthropogenic sources. Both mineral dissolution and anthropogenic sources greatly influenced the water quality in WG4 wells.
- This study highlighted that shallow wells are predominantly affected by the infiltration of wastewater from the surface and subsurface sources, which facilitated the mineral dissolution in the vadose zone followed by the accumulation of solutes in this aquifer, especially in the UGB.
- Excessive fertiliser application, irrigation return flow, leakage from the septic tank, domestic sewage water and accumulation of cattle waste are common contamination sources that affected the water quality in these regions. As the groundwater is an important source for drinking in the study regions, this study recommended periodic monitoring to protect the shallow aquifer.

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Authors Contributions Natarajan Rajmohan performed all the work in this paper.

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Compliance with ethical standards

Competing Interests None

Ethical Approval Not applicable

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