Trends and Probabilistic Stability Index for Evaluating Groundwater Quality: the Case of Quaternary Alluvial and Quartzite Aquifer System of India

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Abstract

This study proposed a novel groundwater quality stability index (GQSI), which considers probabilistic estimate of reliability and resilience based on multi-year dataset. The developed index is validated and optimized adopting optimum index factor approach. The vulnerabilities of different groundwater quality parameters were also computed to provide an insight about the deviations of their concentrations from the safe drinking water limits. The application of the developed stability index is demonstrated through a case study in quaternary alluvial and quartzite aquifer system of India. In addition, trends in the groundwater quality parameters are identified by using variance-corrected Mann-Kendall test, and trends are quantified by using Sen's slope estimation test. Box-whisker plots revealed that EC and TDS mostly exceed their maximum permissible limits prescribed for drinking water in the southern and southwest hard-rock formations. Whereas, most parameters do not cross their maximum desirable limits under in the central and northern alluvial formations. Increasing trends of potassium and bicarbonate, and decreasing trends of carbonate, calcium, sulfate, and fluoride are found prominent. The GQSI values indicated high stability of groundwater quality under older alluvium geology and low stability under the gneiss and mica-schist. Results of the GQSI are found in agreement with that of groundwater quality index (GQI) at 84% sites, which proved adequacy of the developed GQSI. Also, three classes ('low'/'poor', 'moderate', and 'high'/'good') of both the GQSI and GQI showed a good coherence at 83, 78, and 87% sites. However, GQSI is more advantageous than GQI due to former's statistical framework, consistency and comparability over different areas. Three optimum index factors, i.e., TDS, pH and nitrate, are found to have the maximum impact on overall groundwater quality with their largest variations. Results of the optimum groundwater quality stability index (OGQSI) and GQSI closely matched with each other, and a significant linear relationship (R^2=0.70) exists between them. Therefore, OGQSI is a cost-effective approach for adequate monitoring and satisfactory evaluation of the groundwater quality in low-income nations.

Keywords: Groundwater quality stability index; Mann-Kendall test; Optimum index factor; Reliability; Resilience; Vulnerability.

1. INTRODUCTION

Groundwater is a ubiquitous source of freshwater, which supports human health, socio-economic development and functioning of ecosystems (Humphreys, 2009; Steube et al., 2009). It fulfills drinking water needs of more than half of the global population and 43% of all the irrigation water requirements (FAO, 2010). Basic daily water needs of about 2.5 billion people in the world are solely met by the groundwater resources (UNESCO, 2012). The global groundwater use, mainly for agriculture, amounts to 800 km^3 in the year 2010, and 67% of this estimate is extracted in India, the United States of America (USA), China, Iran and Pakistan in descending order (Burek et al., 2016). At present, a third of the world's major groundwater systems are reported to be in distress (Richey et al., 2015). In addition, the contemporary global water demand presently estimated at 4600 km^3 per year is projected to increase to 5500-6000 km^3 per year by 2050 (Burek et al., 2016). Therefore, sustainability of this vital resource is threatened due to combined effects of rising population, overexploitation, urbanization, and developmental activities, which have resulted in increasing depletion and degradation of the groundwater resources. The groundwater quality may further be degraded due to the impacts of climate change and global warming (Bondu et al., 2016; Foster and Gun, 2016).

It is imperative to comprehensively evaluate and characterize the groundwater quality in order to check its degradation and manage groundwater resources sustainably (Machiwal et al., 2011). A variety of tools and techniques exist in literature ranging from graphical to statistical that have been used by the researchers to interpret the groundwater quality (Machiwal et al., 2018). Few of the classical as well as modern techniques include Piper diagram, USSL diagram, Wilcox diagram, principal component analysis, cluster analysis, geostatistical modeling, among others (e.g., Wunderlin et al., 2001; Kumar et al., 2007; Cloutier et al., 2008; Güler et al., 2012; Machiwal and Jha, 2015; Davies and Crosbie, 2018). Another important technique is the water quality index (WQI), which is considered as a simplified way to aggregate and communicate the geochemical knowledge to groundwater managers for integrating the groundwater quality issues within the groundwater sustainability framework (Machiwal et al., 2018). In their pioneering work, Horton (1965) defined a WQI based on eight water quality parameters weighted according to their relative importance. An improved version (NSF-WQI) was proposed by the National Sanitation Foundation (NSF) of USA (Brown et al., 1970; Deininger and Maciunas, 1971) where parameter selection was based on the Rand Corporation's Delphi technique (Linstone and Murray, 1975). Thereafter, many different kinds of the WQIs were proposed and an excellent review of the same is presented by Lumb et al. (2011). All the WQIs evolved before 1998 were mainly used for the surface water quality assessments although few of them were later on used for evaluating the groundwater quality.

The WQI specific to the groundwater quality, i.e., groundwater quality index (GWQI), was derived for the first time by Backman et al. (1998) and Melloul and Collin (1998).

Subsequently, a large number of the GWQIs have been developed and used by the researchers from different parts of the world (Machiwal et al., 2018), and some salient studies are enlisted in Table 1. In general, computation of all such indices follows four basic steps: (i) selection of parameters, (ii) transformation of parameters to bring to a common scale (obtaining sub-index values), (iii) attributing the weights, and (iv) aggregation of sub-indices (Machiwal et al., 2018). These indices use water quality data collected through a monitoring network at a given point of time. These indices do not account for temporal variations of the groundwater quality occurring within the aquifer system over different years due to their inadequacy to deal with multi-year datasets. The multi-time datasets may provide an opportunity to estimate stability or sustainability of the groundwater quality (Babiker et al., 2007). Also, none of the indices are capable of providing the probabilistic estimate of the groundwater quality after aggravation of the considered parameter concentrations.

Trend identification is a useful tool for exploring temporal patterns in the groundwater quality. In groundwater quality literature, trend analysis is very rarely performed (Taylor and Loftis, 1989; Loftis, 1996). However, trend detection in groundwater quality parameters has been receiving increasing attention over the past one decade (Machiwal et al., 2018). Trend analysis plays a major role in providing useful information on possible water quality changes over a period of time. Apparently, different researchers have used different statistical tests for trend detection, which can be categorized as parametric and non-parametric tests (e.g., Visser et al., 2009; Machiwal and Jha, 2015; Koh et al., 2017). Mann-Kendal (M-K) test is the most-widely used nonparametric test for detecting trends in water quality data but its results are reported not to be true when serial correlation is present in the dataset (Yue et al., 2002). The effect of serial correlation on robustness of the M-K test may be avoided by adopting variance-correction (VC) approaches (Lettenmaier, 1976; Hamed and Rao, 1998; Yue and Wang, 2004).

This study proposed a novel probability-based groundwater-quality stability index considering temporal variability of the groundwater quality. The proposed stability index is developed based on the ’system-robustness’ and ‘system sustainability’ criteria (Hashimoto et al., 1982; Loucks, 1997), and it integrates two important probabilistic characteristics of the groundwater quality, i.e., reliability and resilience. The ‘system robustness’ of water resources systems in economic terms is defined as possible deviation between the actual costs of a proposed project and those of the least cost project design (Hashimoto et al., 1982). On the other hand, ‘system sustainability’ of water resource systems is examined by their designs and management that contribute to the objectives of society, now and in the future, while maintaining their ecological, environmental and hydrological integrity (Loucks, 1997). Furthermore, application of the developed groundwater quality stability index is demonstrated through a case study in a quaternary alluvial and quartzite aquifer system of India. Moreover, trends in the groundwater quality parameters are also identified by applying the variance-corrected M-K test and trends are quantified by using Sen’s slope estimation method.

2. STUDY AREA AND DATA DESCRIPTION

2.1 Location and Geography

The study area, Jaipur district, is situated in northeast part of Rajasthan State of India (Fig. 1). The district lies between 26°25′ and 27°51′ North latitude and 74°55′and 76°10′ East longitude covering an area of about 10878 km^2 that is 3.23% of total State's area. For administrative purpose, the entire district is divided into 13 blocks or sub-divisions (Fig. 1). Major landscapes of the area include hillocks, pediments, undulating fluvial plains, Aeolian dune fields, ravines, and palaeo-channels. Structural hills, trending NNE-SSW, mainly exist in north and northeast parts, and are generally composed of Delhi quartzite. Pediments with thin to thick soil cover have a spread around Dudu, Phagi and Chaksu blocks that form flat gneissic outcrops. Aeolian sand dunes are mainly found in the western parts, i.e., Sambhar, Jobner, and Renwal. The area is drained by ephemeral rivers, such as Banganga, Bandi, Dhund, Mendha, Mashi, Sota, and Sabi, and also through their tributaries. Large surface water reservoirs of the area are Ramgarh, Champarwara, Kalakh, Hingonia, Buchara, and Mansagar. Soils are classified as loamy sand to sandy loam, sandy clay loam, sandy clay, wind-blown sand and river sand.

2.2 Climatic Conditions and Groundwater Scenario

Climate of the study area is dry and semi-arid, and is subjected to extremes of cold and heat at distinct places. The minimum and maximum temperatures are 3°C and 45°C, respectively, with the mean temperature of 24°C. The mean annual rainfall is 548 mm, and total annual potential evapotranspiration is 1745mm (CGWB, 2007). Groundwater occurs both in unconsolidated quaternary formations and consolidated formations of Bhilwara and Delhi supergroups, and also post-Delhi granites. Talus and scree deposits at foothills form potential aquifer at places including Bassi block and parts of Amber, Jamwaramgarh and Govindgarh blocks. Well yield in the aquifer ranges from 100 to 500 m^3 d^{-1}. In southern and southwest areas situated in Dudu, Phagi and Chaksu blocks, hard-rocks of Bhilwara supergroup form the main aquifers comprising of granulitic gneisses, quartz mica-schist, phyllite along with granite and pegmatite intrusive. Similarly, quartzite, schist and phyllite of Delhi supergroup form the aquifers in Jamwaramgarh, Bairath, Kotputli, Shahpura, Amber and Bassi blocks. Depth of wells generally varies from 50 to 100 m in alluvium and 50 to 200 m in consolidated formations. Specific capacity of wells varies from 58 to 500 lpm m^{-1}. Transmissivity and storage coefficient values vary from 10 to 850 m^2 d^{-1} and 4.70×10^{-5} to 1.05×10^{-3}, respectively (CGWB, 2017). About 35.27% area of the district has irrigated agriculture, and groundwater is the dominant source for providing irrigation water supplies. Currently, groundwater is the major source of water in the area supplying 1178.92×10^6 m^3 for irrigation and 315.96×10^6 m^3 for drinking and industrial purposes (CGWB, 2017). In Jaipur district, 120,471 dugwells and dug-cum-tubewells are in use for irrigation purpose and 27,378 hand-pumps and dug-cum-tubewells are operational for domestic and industrial uses (CGWB, 2013).

2.3 Data Collection

This study utilized fourteen groundwater quality parameters, i.e., pH, electrical conductivity (EC), total dissolved solids (TDS), calcium (Ca), magnesium (Mg), sodium (Na), potassium (K), chloride (Cl), sulfate (SO_4), carbonate (CO_3), bicarbonate (HCO_3), nitrate (NO_3), fluoride (F), and total hardness (TH), monitored at 250 sites over the area. The data are procured from the State Ground Water Department, Jaipur, Rajasthan, India. These groundwater samples are collected from the wells during pre-monsoon season (April to May) for twelve years (2001-2012). The data are checked for regularity and presence of any anomalies, the anomalies are screened out and error-free groundwater quality parameters of 196 sites are used for subsequent analyses.

3. MATERIALS AND METHODS

3.1 Exploring Groundwater Quality Variations

Box-whisker plots, depicting five important statistical properties, i.e., 25th percentile, median or 50th percentile, 75th percentile, range, and outlier or extreme (Machiwal and Jha, 2012), of 14 groundwater quality parameters are drawn for 13 blocks of the study area to understand temporal variations over the given data period.

3.2 Computing Autocorrelations of Groundwater Quality Parameters

Autocorrelation or serial correlation, if present in a data series, causes inflation of the variance that may increase statistical significance of the trends detected by the nonparametric M-K test (Yue and Wang, 2002). Hence, presence of serial correlation at one-year time lag is determined in data series of every groundwater quality parameter by computing autocorrelation coefficient (ACF) using following expression (Haan, 2002):

$$\mathrm{ACF} = \frac{\sum_{t=0}^{n-1}(x_t \cdot x_{t+1}) - 1/(n-1)\sum_{t=0}^{n-1} x_t \cdot \sum_{t=0}^{n-1} x_{t+1}}{\left[\sum_{t=0}^{n-1} x_t^2 - 1/(n-1)\left(\sum_{t=0}^{n-1} x_t\right)^2\right]^{1/2}\left[\sum_{t=0}^{n-1} x_{t+1}^2 - 1/(n-1)\left(\sum_{t=0}^{n-1} x_{t+1}\right)^2\right]^{1/2}} \tag{1}$$

where, x_t = value of a parameter at time t, x_{t+1} = value of parameter at time t+1, and n = number of data in series.

The ACF is considered statistically significant at α level of significance, if its computed value crosses the critical limits (Anderson, 1942) as given below:

$$(r_1)_{upper} = \{1/(n-1)\}(-1 + z_{1-\alpha/2}\sqrt{n-1-1}) \tag{2}$$

$$(r_1)_{lower} = \{1/(n-1)\}(-1 - z_{1-\alpha/2}\sqrt{n-1-1}) \tag{3}$$

where, $z_{1-\alpha/2}$ = standard normal variate.

3.3 Evaluating Significance of Trends

The nonparametric M-K test is used for evaluating significance of trends in groundwater quality parameters. Details of the original M-K test may be found in standard textbooks, e.g., Salas (1993); Machiwal and Jha (2012). The original M-K test-statistic (z_k) is modified to remove the effect of serial correlation by applying the variance-correction following the effective sample-size approach of Lettenmaier (1976), as shown below:

$$z_k{}^* = z_k\sqrt{\frac{n^*}{n}} \tag{4}$$

where, $z_k{}^*$ = variance-corrected M-K test-statistic, and n^*/n = correction factor, which is computed using the formula given for lag-1 autoregressive process (Matalas and Langbein, 1962):

$$\frac{n^*}{n} = \frac{1}{1 + 2\dfrac{r_1^{n+1} - n\,r_1^2 + (n-1)\,r_1}{n\,(r_1 - 1)^2}} \tag{5}$$

3.4 Quantifying Trends by Sen’s Slope Estimation Test

Groundwater quality trends are quantified using Sen’s slope estimation test, which considers all combinations of data pairs, such as x_{ik} and x_{jk}, for a k^{th} site (j>i). Then, slope (β_k) of the fitted straight line for every paired data is computed for a given k^{th} site. Finally, the test-statistic is computed as shown below (Sen, 1968; Hirsch et al., 1982):

$$\beta_k = \text{median}\left[\frac{(x_{ik} - x_{jk})}{(i - j)}\right] \text{for all } i<j \tag{6}$$

The positive (or negative) β-values indicate the increasing (or decreasing) trends.

3.5 Developing Probability-Based Groundwater Quality Stability Index

Stability of a groundwater quality parameter is a measure of its variability over time. This study utilized the concepts of ‘system robustness’ (Hashimoto et al., 1982) and ‘system sustainability’ (Loucks, 1997), based on three probabilistic terms, such as reliability (R_y), resilience (R_e) and vulnerability (V_y), to define a probability-based groundwater-quality stability index. This is helpful to understand the changes occurring in concentration of a particular groundwater quality parameter over a given period of time. Terms R_y, R_e and V_y, require a threshold value of the parameter to decide successful and failure events, and also to take a difference of the observed and threshold values for that parameter. Thus, the desirable and/or permissible limits suggested for drinking water, prescribed by the World Health Organization (WHO), Geneva, are used as threshold values for the parameters while computing the stability index (Table 2). The groundwater-quality stability index (GQSI) developed in this study tells about the probability of groundwater quality being stable or not over the years with respect to the WHO-prescribed drinking water standards. The WHO standards prescribed for drinking water quality are used in this study because when water is used both for irrigation and drinking purposes, irrigation water should ideally meet the drinking water limits (Jensen et al., 2001). The stability criterion, used in this study, is based on the idea that sustainability is related to a high degree of reliability and resilience and a low degree of vulnerability (Duckstein and Parent, 1994). The terms, R_y, R_e, and V_y along with development of groundwater-quality stability index and composite stability index are discussed below.

3.5.1 Reliability

Reliability of a water quality parameter is the probability of having that parameter (concentration or value) within the safe drinking water limits (desirable or permissible) prescribed by the WHO. In other words, it is defined as the ratio of number of successful events to the total number of data in series. Here, number of successful events is computed by counting number of years when concentration of the given parameter does not exceed a threshold x^T, defined by the prescribed WHO limit. Thus, reliability (R_y) for a groundwater quality data series, containing n values of a parameter, is expressed as:

$$R_y = f_{SE} / n \tag{7}$$

where, f_{SE} = number of successful events in data series of given parameter.

The value of R_y ranges from 0 (no reliability when concentration of a parameter always exceeds the safe limits) to 1 (maximum reliability when parameter concentration never exceeds the safe limits).

3.5.2 Resilience

Resilience of a water quality parameter is defined as its capacity to adapt to changing conditions over time such as climate variability and change (WHO, 2009). In case of groundwater quality parameter, the R_e value suggests ability or power of a parameter to come within its acceptable range defined for the safe drinking water after some adverse condition when its value exceeds the desirable or permissible limit. Resilience is the probability that a parameter, having failure event at a time t-1, will have the next event successful at time t.

Thus, resilience (R_e) is ratio of number of times a successful event follows a failure event to number of total failure events in data series, and is shown by following expression:

$$R_e = f_{FE-SE} / f_{FE} \tag{8}$$

where, f_{FE-SE} = number of times a successful event follows a failure event, and f_{FE} = number of times a failure event occurs in data series.

Similar to the R_y, the value of R_e varies between 0 (no resilience when a parameter never returns to safe limits once it crosses it) and 1 (maximum resilience when a parameter not at all exceeds its limits of safe drinking water).

3.5.3 Vulnerability

Vulnerability of a parameter indicates the severity of the 'deviation' of the groundwater quality from stability state during the years of failure events, and is defined by extent of differences between the observed data during failure events and threshold values, x^T. Vulnerability (V_y) is defined as the probability of exceedance to vulnerability, and is computed by adding differences between data value and x^T, as shown below:

$$V_y = \sum_{i=1}^{n} \text{difference}\left(x^T - x_t\right) \Big/ f_{FE} \quad ; t = 1, 2, \dots n \tag{9}$$

where, $\sum_{i=1}^{n} \text{difference}\left(x^T - x_t\right)$ = sum of absolute values of ($x^T - x_t$) for failure events.

As quantification of trend 'magnitude' is equally-important in trend 'identification' studies, 'vulnerability' of the groundwater quality is very much relevant in studies dealing with computation of groundwater quality 'stability'. In this study, vulnerability of the groundwater quality parameters is computed by adding differences between their observed values and desirable/permissible safe drinking water limits (threshold values). The V_y values obviously have the unit of the parameters, i.e., mg l^{-1} for the major cations and anions.

3.5.4 Groundwater Quality Stability Index

In this study, a probability-based stability index is developed to adjudge steadiness of the groundwater quality parameters over time with respect to the drinking water standards prescribed by the WHO. The stability index has resemblance to the sustainability index that quantifies sustainability of water resources systems to facilitate the evaluation and comparison of water management policies (Loucks, 1997). Loucks (1997) defined the sustainability index (SI) by combining R_y, R_e and V_y in a multiplicative form. Later on, various alternative forms of the SI have been proposed in the literature. In this study, the

Groundwater Quality Stability Index (GQSI) is computed by modifying the original SI and considering only two terms, i.e., R_y and R_e in multiplicative combination, as expressed below:

$$\mathrm{GQSI} = R_y \times R_e \tag{10}$$

Positives linear relationship between R_y and R_e for individual nine groundwater quality parameters has been observed for 196 sites, indicating that small value of one parameter is likely to be accompanied with small value of other parameter, and vice-versa. Thus, value of GQSI ranges from 0 (no stability) to 1 (maximum stability). The GQSI approach uses an implicit weighting, because it adds the worst weight to the criteria having the worst performance. Term V_y is not considered in the development of GQSI in this study as its value depends upon the scale of measurements. However, the V_y values are computed for different groundwater quality parameters to provide an insight about the deviations of their concentrations from the safe drinking water limits.

Finally, a composite stability index ($\mathrm{GQSI}_{\mathrm{composite}}$) is computed by taking mean of the stability indexes for individual parameters, as defined below:

$$\mathrm{GQSI}_{\mathrm{composite}} = \frac{\sum_{i=1}^{n} \mathrm{GQSI}_i}{n} \tag{11}$$

where, GQSI_i = groundwater quality stability index for ^{ith} parameter.

3.6 Comparing GQSI with Groundwater Quality Index

The developed GQSI is compared with a rating-based average groundwater quality index (GQI). Computation of GQI, as defined by Babiker et al. (2007), involves three steps: (i) calculating contamination index (C) for the annual mean values of every parameter by relating its value with WHO prescribed limit, (ii) rating C between 1 and 10 by generating rank (R), and (iii) computing GQI using ranks (R) and relative weights (W) of every parameter using the following equation.

$$\mathrm{GQI} = 100 - \left[\frac{(R_1 W_1 + R_2 W_2 + \ldots + R_n W_n}{n}\right] \tag{12}$$

Detailed methodology for computing GQI may be found in Babiker et al. (2007) and Machiwal et al. (2011).

3.7 Developing Optimum Probabilistic Stability Index

In the GQSI, some of the parameters may be spatially invariable or having similar spatial patterns or correlations, and thus, may be repetitive or having little contribution to variation of GQSI. Therefore, an Optimum Index Factor (OIF) is chosen to remove redundancy by identifying a combination of three parameters providing the highest information (or standard deviations) with least duplication (or lowest correlation). The OIF is given by the following expression (Machiwal et al., 2011):

$$\mathrm{OIF} = \frac{\mathrm{SD}_1 + \mathrm{SD}_2 + \mathrm{SD}_3}{\left|\mathrm{R}_{1,2}\right| + \left|\mathrm{R}_{2,3}\right| + \left|\mathrm{R}_{3,1}\right|} \tag{13}$$

where, SD = standard deviation, and |R| = absolute value of correlation coefficient between two parameter pairs.

The OIF is computed for all three-parameter combinations among nine parameters, i.e., TDS, pH, Na, Ca, Mg, Cl, SO_4, NO_3 and hardness, and the best combination is selected based on the highest OIF value. Thereafter, the selected parameters are used to compute the optimum groundwater quality stability index (OGQSI) as follows:

$$\mathrm{OGQSI} = \frac{\mathrm{GQSI}_1 + \mathrm{GQSI}_2 + \mathrm{GQSI}_3}{3} \tag{14}$$

3.8 Validating Optimum Groundwater Quality Stability Index

Values of OGQSI for 196 sites are plotted against the corresponding GQSI values on scatter plot. This study performed regression analysis for observed versus predicted values (OP) instead of predicted versus observed values (PO) as suggested by Piñeiro et al. (2008), drawing scatter plot by placing original GQSI values in ordinate (in y-axis) and OGQSI values in abscissa (in x-axis). A straight line is fitted to the scatter plot, and linear equation and coefficient of determination (R^2) value are determined. Furthermore, the fitted line is compared with 1:1 line. A close proximity of the fitted line with 1:1 line validates the estimated values of the OGQSI.

4. RESULTS AND DISCUSSION

4.1 Spatial and Temporal Variations of Groundwater Quality Parameters

Box-whisker plots of 14 groundwater quality parameters for 13 blocks are shown in Figs. 2(a-n). The median pH value of groundwater exceeds the maximum permissible limit (MPL) of 8.5 prescribed for drinking water for all the blocks, except three blocks situated in the northern and northeast directions, i.e., Amber, Shahpura and Viratnagar (Fig. 2a). This finding suggests that the groundwater in most of the study area is under alkaline condition, which agrees with findings of Tatawat and Chandel (2008) for Jaipur city. The median EC value of groundwater is more than the maximum desirable limits (MDL) of the drinking water (750 μS cm^{-1}) in all the blocks, which further exceeds the MPL (2250 μS cm^{-1}) in four blocks, i.e., Chaksu, Dudu, Phagi, and Sambhar (Fig. 2b). Box-whisker plots of TDS are exactly similar to those of EC in each block, and the median TDS values exceed the MPL (1500 mg l^{-1}) in four blocks where EC also exceeded the MPL (Fig. 2c). Calcium concentrations in all blocks are found within the MDL of 75 mg l^{-1} (Fig. 2d). However, the median concentrations of magnesium crossed the MDL, i.e., 30 mg l^{-1} in 12 blocks (Fig. 2e). The median concentrations of both sodium and chloride go beyond the MDL (200 mg l^{-1}) in 7 blocks, i.e., Bassi, Chaksu, Dudu, Kotputli, Phagi, Sambhar, and Sanganer (Figs. 2f,h). The median of bicarbonate and nitrate exceeds the MDL in six blocks each (Figs. 2k,l). Total hardness in all the blocks crosses the MDL although it does not exceed the MPL (Fig. 2n). Hard to very hard groundwater quality for Jaipur city is also reported in some earlier studies (Tatawat and Chandel, 2008; Tank and Chandel, 2010). It is seen that the median concentration value of the most parameters is relatively high in four blocks, i.e., Chaksu, Dudu, Phagi and Sambhar, which are situated under gneiss and mica-schist type of hard-rock geology lying in southern and southwest portions of the area. On the other hand, the median concentration values of the most parameters are the lowest in three blocks, i.e., Shahpura, Viratnagar and Jhotwara, where mainly older alluvium exists. This indicates that the groundwater under hard-rock geology is contaminated more as compared to alluvium geology. Low vulnerability to groundwater pollution under alluvial formations of Jaipur district in comparison to fractured hard-rock formations has been also reported by Chintalapudi et al. (2017). It is further observed from Figs. 2(a-n) that box size and whiskers' length of the most parameters are larger for four blocks, i.e., Chaksu, Dudu, Phagi, and Sambhar. This suggests relatively large spatial variations of the groundwater quality parameters in these blocks. Therefore, blocks situated in the southern and southwest portions of the area depict higher parameter concentration exceeding the MDL and MPL, with larger spatial variations.

4.2 Presence of Serial Correlation in Parameter Series

Presence of the significant serial correlation at different sites of 13 blocks for 14 parameters is presented in Table 3. For individual parameters and blocks, number of sites with presence of serial correlation varies from 0 to 6. The total number of sites with serial correlation ($p>0.05$) is found to vary from 10 (fluoride) to 38 (pH) of the total 196 sites, which indicates that a large number of the sites are free from serial correlation for all parameters.

4.3 Trends in Groundwater Quality Parameters

Trends identified in 14 groundwater-quality parameters at three significance levels, i.e., 1, 5 and 10%, are found to vary spatially [Figs. 3(a-n)]. Of the total 196 sites, positive trends are dominant for potassium and bicarbonate parameters at 161 and 153 sites, respectively. However, significantly increasing trends at 10% level of significance are found to be present for potassium at 25 sites, followed by TDS, EC and bicarbonate at 12, 11 and 11 sites, respectively. The increasing potassium concentration in the groundwater may be attributed to the leaching of fertilizers through the subsurface (Sethy et al., 2017). Likewise, bicarbonates are probably derived from weathering of silicate rocks, dissolution of carbonate precipitates, atmospheric and soil carbon-dioxide (CO_2) gas (Jeong, 2001; Krishna Kumar et al., 2011). Furthermore, increased bicarbonate may be due to increased percolation from the recently-recharged surface water in the area (Deshmukh and Aher, 2016). The level of significance for most significant trends is higher for TDS and EC in comparison to potassium. On the contrary, prominent declining trends are visible for carbonate, calcium, sulfate, fluoride, and pH at 160, 156, 151, 142, and 130 sites, respectively. Of the total declining trends, statistically-significant trends are revealed mostly for carbonate (30 sites), followed by calcium (26 sites), sulfate (26 sites) and fluoride (26 sites). The statistical significance of identified trends is higher (level of significance is 5% or more) for calcium and sulfate. The declining trends of calcium and sulfate result in poor concentration of these ions in groundwater over the years, which indicate freshening of the groundwater resources (Appelo and Postma, 2005). Besides, decreasing sulfate-trends are most-likely due to less sulfur deposition and natural dilution processes occurring in the area (Wahlin and Grimvall, 2010). The partial pressures of CO_2 may also be responsible for diminishing trends of carbonates, which also decreases sulfur content under the presence of sulfur-reducing bacteria (Toscani et al., 2001). Decreasing fluoride content in the groundwater depends on pH, groundwater temperature, intensity of weathering processes of fluorine-bearing minerals and type of geology (Dar et al., 2011). It is observed that there is no definite spatial pattern of increasing/decreasing trends for all the groundwater quality parameters.

Trend magnitudes of 14 groundwater-quality parameters at 196 sites are estimated by using the Sen's slope estimator, and the mean values of the trend magnitudes for 13 blocks are shown in Figs. 4(a-n). The mean trend magnitudes of pH, sodium, calcium, sulfate, carbonate, nitrate, and fluoride are negative in all the blocks except 2 or 3 blocks, whereas potassium and bicarbonate have positive trend magnitudes over almost all the blocks. The mean trend magnitudes for EC, TDS and pH range from 82.2 to -83.1 $\mu S\ cm^{-1}\ year^{-1}$, from 29.6 to -51.0 $mg\ l^{-1}\ year^{-1}$, and from -0.024 to -0.001 $year^{-1}$, respectively. Similarly, the mean trend rates of sodium, potassium, calcium, magnesium, chloride, sulfate, carbonate, bicarbonate, nitrate, fluoride, and hardness vary from -18.8 to 2.9, -0.1 to 4, -0.9 to 0.2, -1.9 to 1.8, -17.8 to 19.7, -10.2 to 0.2, -2.8 to 0.6, 2.6 to 12.5, -2.3 to 1.2, -0.2 to 0.03, -10.5 to 7.1 $mg\ l^{-1}\ year^{-1}$, respectively. Of the total 14 parameters, 8 parameters (i.e., EC, TDS, sodium, magnesium, chloride, sulfate, fluoride, and hardness) depict the fast-declining trends in four blocks, i.e., Chaksu, Dudu, Kotputli, and Phagi. Concentration of EC and TDS are found fast-increasing in Jamwaramgarh (44.9 and 24.5 $mg\ l^{-1}\ year^{-1}$, respectively) and Sambhar blocks (82.2 and 29.6 $mg\ l^{-1}\ year^{-1}$, respectively). Likewise, potassium has an increasing trend magnitude of more than 0.1 $mg\ l^{-1}\ year^{-1}$ in six blocks including four blocks where EC and

TDS are found to have fast-increasing trends. In addition, trend magnitudes are observed to be considerably inclining for magnesium in Jamwaramgarh (1.8 mg l^{-1} $year^{-1}$), and for chloride in Sambhar block (19.7 mg l^{-1} $year^{-1}$). Presence of a salty lake in Sambhar block may be a likely cause for the increasing trend magnitudes there (Yadav et al., 2007; Joshi and Seth, 2011). Nitrate concentration is found to be increasing at mean rate of 1.2 mg l^{-1} $year^{-1}$.

4.4 Probabilistic Stability of Groundwater Quality over the Space

In this study, R_y, R_e and V_y values along with the groundwater-quality stability index are computed for nine parameters, i.e., pH, TDS, Ca, Mg, Na, Cl, SO_4, NO_3 and TH over 196 sampling sites. Out of total 14 parameters, we selected nine parameters as the WHO-prescribed (WHO, 2017) desirable and/or permissible limits for drinking water are available only for these nine parameters.

4.4.1 Reliability Measure of Parameters

Reliability (R_y) values are classified into three groups of equal interval, i.e., 0-0.33 ('low' reliability), 0.33-0.66 ('moderate' reliability), and 0.66-1.0 ('high' reliability). Distribution of the classified R_y values over 196 sites in the study area are shown in Figs. 5(a-i) for nine groundwater quality parameters. It is evident from Fig. 5 that calcium, sulfate and nitrate possess more reliability in comparison to other parameters as these three parameters have 'high' reliability at 100, 97, and 70% of the total sites, respectively. On the other hand, TH and TDS have 'low' reliability at majority of the sites, i.e., 95 and 47%, respectively. It is further revealed that 'low' reliability of TDS, sodium and chloride is mostly dominating in five blocks of Chaksu, Dudu, Kotputli, Phagi, and Sambhar mainly situated under gneiss and mica-schist geology in the southern, southwest, and southeast portions of the area. The low reliability of these parameters is most-likely due to geogenic contamination occurring from the hard-rock geologic terrain (Machiwal and Jha, 2015; Chintalapudi et al., 2017). The 'moderate' reliability for pH and magnesium is prominent over 66 and 63% sites over the area, respectively.

4.4.2 Resilience Measure of Parameters

The R_e values of the parameters are classified into three equal-interval probability classes: (i) 0-0.33 ('low' resilience), (ii) 0.33-0.66 ('moderate' resilience), and (iii) 0.66-1.0 ('high' resilience). All the sampling sites grouped under three probability classes of R_e are depicted in Figs. 6(a-i) for the nine groundwater quality parameters. It is revealed that calcium has 'high' resilience at all the sites followed by sulfate and nitrate at 95 and 64% sites, respectively. Likewise, resilience is prominently 'high' to 'moderate' at more than 73% sites for sodium and chloride. On the contrary, three parameters, i.e., pH, TDS and magnesium, showed 'moderate' to 'low' resilience at majority of the sites, i.e. 73% or more. Similar to the results of reliability, resilience of sodium and chloride is apparently 'low' in five blocks having gneiss and mica-schist rocky terrain and situated in the southern, southeast and southwest portions. The low resilience of sodium and chloride in the hard-rock terrain indicates that these constituents mostly remain under failure state (unacceptable range) and do not easily return to success state (acceptable/permissible range), which may be attributed to water-rock interactions and geogenic contamination occurring in the aquifer system (Chintalapudi et al., 2017).

A positive linear relationship between R_y and R_e indices is obtained for eight individual groundwater quality parameters, i.e., pH, TDS, Ca, Mg, Na, Cl, SO_4, and NO_3, as revealed from the correlation-coefficient (R) values of 0.71, 0.33, 0.44, 0.73, 0.82, 0.86, 0.67, and 0.66, respectively. In case of TH, relationship is found negative (R=-0.49); otherwise, most of the positive R_y-R_e relationships are statistically-significant (R>0.50), which suggest that small value of one parameter will likely be accompanied with small value of other parameter, and vice-versa. The similar observation of a monotonic relationship between R_y and R_e indices is reported by Maity et al. (2013), and also agrees with findings of Hashimoto et al. (1982). This finding justifies the consideration of product of two indices (Eq. 10) in this study to define the groundwater-quality stability index.

4.4.3 Vulnerability Measure of Parameters

Vulnerability (V_y), in statistical perspective, is the likelihood or measure of degree that a failure event will become a loss. The V_y value of a groundwater quality parameter is looked at as probability of exceedance that describes the magnitude of the average deviation of the parameter value or concentration during the failure events from its safe limit or threshold value set by the WHO for drinking water. The value of V_y depends upon the unit of measurement of the parameter, and therefore, its range is observed to be the smallest for pH and the largest for TDS. Hence, class values for three groups (i.e., 'low', 'moderate' and 'high' vulnerability) of V_y are not kept the similar for all nine parameters but the classes are chosen depending upon their threshold values (Table 4). The sites are grouped under the three vulnerability classes for all the nine parameters (Figs. 7a-i). It is apparent from Fig. 7 that sulfate and chloride have 'low' vulnerability at 95 and 91% sites followed by magnesium with 62% sites having 'low' vulnerability. Furthermore, TDS, sodium, nitrate, and TH have 'low' to 'moderate' vulnerability over 75% or more sites. In contrast, pH is highly vulnerable at 54% sites and moderately vulnerable at 46% sites. Also, it is observed that sites depicting 'high' vulnerability of six parameters, i.e., pH, TDS, sodium, chloride, nitrate and TH, are dominating in four blocks, i.e., Chaksu, Dudu, Phagi and Sambhar, which are mainly overlain by the gneiss and mica-schist geology in southern, southwest and western parts. These results are in agreement with findings of box-whisker plots, which revealed relatively high median concentration and a large temporal variation among the most of the groundwater quality parameters in these four blocks. Therefore, it is emphasized that vulnerability measure is an important indicator for understanding temporal dynamics of the groundwater quality.

4.4.4 Groundwater Stability Index

Composite probability-based stability index of the groundwater quality is computed based on reliability and resilience indicators. Spatial distribution of groundwater-quality stability index (GQSI), classified into three categories, i.e., 'low stability', 'moderate stability' and 'high stability', is shown in Fig. 8(a). The GQSI values indicate 'low', 'moderate', and 'high' stability at 92 (47%), 80 (41%) and 24 (12%) sites, respectively. It is apparent that stability of the groundwater quality is generally 'low' at almost all sites located in four blocks, i.e., Chaksu, Dudu, Phagi, and Sambhar having gneiss and mica-schist type of geology. Presence of hard-rocks in these blocks may be responsible for geogenic contamination of the groundwater resources, and degradation of the groundwater quality (Machiwal and Jha, 2015; Chintalapudi et al., 2017). In addition, the low GQSI values are found at 3, 6, 2, 9, 6, 1, and 2 sites in Amber, Bassi, Jamwaramgarh, Kotputli, Sanganer, Shahpura, and Viratnagar, respectively. On the other hand, groundwater quality is found stable mainly in Shahpura (7 sites) and Viratnagar (6 sites) along with Govindgarh (4 sites) and Jhotwara (4 sites) blocks. Spatially, the low stability of the groundwater quality is observed in the southern, southwest and northern parts, whereas the moderately stable groundwater quality is seen towards the central, eastern and northeast parts. The groundwater quality in the central and north-central parts having older alluvium geology is found to be highly stable.

4.5 Groundwater Quality Index

The mean groundwater quality index (GQI) map, shown in Fig. 8(b), depicts spatial distribution of 196 sites into three classes of 'good', 'moderate' and 'poor' groundwater quality. Groundwater quality at 84 (43%) and 89 (45%) sites is 'poor' and 'moderate', respectively. However, 'good' quality groundwater exists at 23 (12%) sites only. Overall, the groundwater quality of the study area is good as revealed from the mean GQI value of 73.93 (maximum GQI=100). On computing the mean GQI for 14 individual blocks, it is found that relatively poor (GWQI<72) groundwater quality exists in four blocks, i.e., Dudu, Phagi, Chaksu, and Sambhar, where almost all the sites are classified under 'poor' quality category. On the other hand, most of the 'good' quality sites exist in four blocks, i.e., Govindgarh, Jhotwara, Shahpura, and Viratnagar, which suggests that the groundwater quality is relatively good (GWQI>76) under alluvium geology. Existence of gradients of groundwater quality, in the study area (Fig. 8b), indicate that the groundwater quality gets deteriorated while moving from the northwest to southern, southwest, southeast, and northern directions.

4.6 Comparison of the Probabilistic GQSI and GQI

A spatial comparison of both GQSI and GQI maps (Fig. 9) clearly suggests that 165 (84%) sites, having 'good', 'moderate', and 'poor' groundwater quality, correspond to similar category of 'high', 'moderate', and 'low' stability of the groundwater quality. Dissimilar results are found for only 31 (16%) sites. Thus, stability of the groundwater quality is more for a site where groundwater quality is relatively good, and vice-versa. Furthermore, it is apparent from Fig. 10 that 83% of the 'high', 78% of the 'moderate', and 87% of the 'low' GQSI sites are grouped under 'good', 'moderate', and 'poor' classes of GQI, respectively. It is apparent from the above discussion that the sites having 'good' quality groundwater showed 'high' stability of the groundwater quality, whereas the sites with 'poor' quality groundwater depicted 'low' stability of the groundwater quality. Thus, GQSI has a good agreement with GQI over most of the sites in the area. However, GQSI in contrast to GQI is more advantageous as it is a probability-based indicator of groundwater quality with statistical framework. The probability indicators R_y, R_e and V_y, used in this study, have also been used to assess watershed health propensity through a drought index by some researchers (Sadeghi and Hazbavi, 2017; Hazbavi et al., 2018). However, none of these studies considered other indices available in literature for comparison and validation of the developed drought index. The GQSI reveals the probability of groundwater quality being in a successful or satisfactory state and how quickly it returns to a satisfactory state after falling below the satisfactory threshold. This study integrates two important probabilistic terms, i.e., reliability, and resilience to estimate stability index (Hashimoto et al., 1982), and the third probability indicator, i.e., vulnerability calculates the contrast between groundwater quality and acceptable range of water quality standards. This type of integration is suggested in literature for the water quality parameters over a range of water quality constituents at watershed-scale (Hoque et al., 2012, 2014). In addition, the proposed GQSI is superior to the existing water quality indices as it provides an estimate of temporal stability of the groundwater quality by amalgamating temporal variation as well as discrepancy from the water quality standards prescribed by the WHO. Various GQIs available in the literature are mostly developed for assessing water quality at some static-point of time, and are not apt to capture temporal variations of the dynamic water quality. Hence, studies dealing with dynamic water quality are compelled to adopt other measures to understand temporal variations of the water quality; for example the coefficient of variation (CV) as is used in Babiker et al. (2007), Khan et al. (2011), etc. Also, the CV values in such studies could not reveal an overall variability of the groundwater quality rather variations were obtained for the individual water quality parameters (e.g., Machiwal et al., 2011). On the contrary, the developed GQSI has the competency to precisely account for the temporal variability of the composite groundwater quality. This characteristic of the GQSI enables its applicability for the multi-season or multi-year groundwater quality dataset. Moreover, formulation of the proposed GQSI uses implicit weighting through reliability and resilience, and explicit assignment of weights to different parameters considered for the analysis are not needed. Hence, the GQSI is free from subjectivity, and thus, is more convenient, usable and preferable than other water quality indices where certain weights are associated with parameters, e.g., WQI (Boateng et al., 2016), modified DWQI (Abtahi et al., 2015), IWQI (Singh et al., 2018), etc. This merit of the GQSI qualifies its applicability over different aquifer systems under varying hydrogeologic conditions where the GQSI values may be easily compared with each other. Therefore, the GQSI is an excellent probabilistic indicator of groundwater-quality variations, which may have potential usability in different parts of the world.

4.7 Optimum Stability Index of Groundwater Quality

This study computed optimum index factor (OIF) for all possible three-parameter combinations of total nine groundwater quality parameters to reveal the largest variation among the parameters with the least duplication of them. These nine parameters are considered for developing the GQSI. Thus, in total, 84 combinations of nine parameters are used to find out OIF values, and the best five combinations having the largest OIF values are presented in Table 5. It is worth mentioning that two parameters, i.e., TDS and pH, are the similar in each of five combinations, which had the high vulnerability in four blocks, i.e., Chaksu, Dudu, Phagi and Sambhar. Thus, TDS and pH are the two groundwater quality parameters that depict the largest variation in comparison to other parameters. Similar to this study, pH depicted the largest variation in comparison to other parameters in a hard-rock aquifer system of India (Machiwal et al., 2011). The highest OIF value of 718.43 is obtained in case of TDS, pH, and nitrate, and hence, these three optimum parameters are further utilized to compute the stability of the groundwater quality.

4.8 Verification of the Optimum Probabilistic Stability Index

The optimum probabilistic estimates of the stability index are classified into three categories, i.e., low, moderate and high, and spatial distribution of the same over 196 sites is depicted in Fig. 11a. Similar to the findings of the GQSI, results of the optimum GQSI indicated that the groundwater quality at few sites in the central and north-central portions is highly stable. On the other hand, low stability of the groundwater quality is found towards the southern, southwest and northern portions. A good concurrence in spatial distribution of the GQSI (Fig. 8a) and optimum GQSI (Fig. 11a) is evidenced, which verifies capability of the optimum GQSI in revealing exact stability of the groundwater quality. In order to further verify the accuracy of the optimum GQSI, the estimates of the optimum GQSI are scatter plotted on 1:1 line along with the corresponding estimates of the GQSI for 196 sites in the study area, and the same is shown in Fig. 11b. The fitted linear trend line on upperside of the 1:1 line over a major portion further suggests that the optimum GQSI values are slightly over-estimated than the original GQSI values at most sites. However, an apparent linear pattern with statistical significance (R^2 value = 0.70) in the relationship between the two stability indices and scattering of the data points closed to the 1:1 line suggest competence in the optimum GQSI approach. The OIF criterion chosen in developing optimum GQSI has been successfully utilized for the cost-effective and long-term groundwater quality monitoring by Babiket et al. (2007) and Machiwal et al. (2011). Thus, the optimum GQSI provides satisfactory estimates of the probabilistic stability of the groundwater quality, and can be used to reveal the groundwater quality under economic perspectives of cost-effective monitoring of only critical groundwater quality parameters especially in the developing nations.

5. CONCLUSIONS

In this study, a novel probabilistic index is proposed to evaluate temporal stability of the groundwater quality. Application of the developed index is demonstrated through a case study in quaternary alluvial and quartzite aquifer system of Jaipur district of Rajasthan, India.

The vulnerabilities of different groundwater quality parameters, describing deviations of their concentrations from the safe drinking water limits, are also analyzed. Furthermore, trends in groundwater quality parameters are identified by using variance-corrected Mann-Kendall test, and quantified using Sen's slope estimator. Box-whisker plots revealed that most of the groundwater quality parameters including EC and TDS exceed their maximum permissible limits in the southern and southwest portions having gneiss and mica-schist type of geology. On the contrary, groundwater quality parameters remain within the maximum desirable limits prescribed by the WHO in older alluvium geology existing towards the central and northern portions. Autocorrelation analysis suggested that the serial correlation in the groundwater quality parameters is not present at most of the sites in the area. In the study area, the increasing trends of potassium and bicarbonate parameters are prevailing at most sites. On the other hand, the decreasing trends are prominent in carbonate, calcium, sulfate, and fluoride. The fast-inclining trends in TDS and EC are depicted in Jamwaramgarh and Sambhar blocks, and presence of a salty water lake in Sambhar block may be attributed to increase in both the parameters. Stability of the groundwater quality is found generally low under the gneiss and mica-schist geology, which may be due to geogenic contamination of the groundwater occurring in the hard-rock aquifer system. In contrast, the groundwater quality underlying the older alluvium geology is found to be highly stable. Results of the groundwater quality index (GQI) supported the findings of the groundwater-quality stability index (GQSI). However, GQSI, proposed in this study, has the added advantages of having statistical framework, consistency (free from subjectivity) and comparability over different parts of the world. Three classes of the stability index, i.e., 'high', 'moderate', and 'low', fairly matches with other three categories of the water quality index, i.e., 'good', 'moderate', and 'poor' at 84% sites with dissimilar results at only 16% sites. Also, 'high', 'moderate', and 'low' stability sites are found in perfect agreement with 'good', 'moderate', and 'poor' classes of the water quality index at 83, 78, and 87% sites, respectively, which clearly verifies the adequacy of the developed stability index in evaluating the groundwater quality probabilistically. It is worth mentioning here that the proposed GQSI provides a statistical framework to assess steadiness of the groundwater quality parameters over time and does not identify factors responsible for changes in the groundwater quality. The optimum index factor criterion indicated that three parameters, i.e., TDS, pH and nitrate, have the great impact on the overall groundwater quality. Thus, these parameters need to be monitored accurately and regularly in the study area. The stability index of the groundwater quality based on the three optimum index factors shows a good coherence with the findings of the original stability index with strong statistical significance (R^2=0.70) of the linear relationship between the two estimates. Thus, the optimum stability index is very valuable approach for cost-effective monitoring and evaluation of the groundwater quality in low-income nations.

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Table 1. Salient groundwater quality indices developed and used (in chronological order) in different parts of the world

Year	Country	Name of Index[1]	Parameters[2]	Source[3]
1998	Finland	GWQI (C_d)	pH, Na, Cl, SO_4, F, NO_3, UO_2, Ag, Al, As, B, Ba, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Rn, Se, Zn, $KMnO_4$ consumption	Backman et al. (1998)
1998	Slovakia	GWCI (C_d)	TDS, Cl, SO_4, F, NO_3, NH_4, Al, As, Ba, Cd, Cr, Cu, Fe, Hg, Mn, Pb, Sb, Se, Zn	Backman et al. (1998)
1998	Israel	GWQI (IAWQ)	Cl, NO_3	Melloul and Collin (1998)
1999	Egypt	GWQI (WQI)	TDS, BOD, NO_3, Cl, PO_4, Cd, Cr, Ni, Pb	Soltan (1999)
1999	Croatia	GWQI/SWQI (WQI)	Temperature, DO, BOD, Mineralization, Corrosion coefficient, Total N, Protein N, Total P, TC	Stambuk-Giljanovic (1999)
2004	Mexico	GWQI (ICA)/GWCI (C_d)	Temperature, EC, pH, Major ions	Ramos Leal et al. (2004)
2006	Portugal	GWQI / GWCI	Ca, Cl, SO_4, NO_3	Stigter et al. (2006)
2007	Brazil	GWQI	Cl, NO_3	Nobre et al. (2007)
2007	Japan	GWQI (GQI)	TDS, Ca, Mg, Na, Cl, SO_4, NO_3	Babiker et al. (2007)
2009	Ghana	GWQI (WQI)	EC, Ca, Mg, Na, Cl, F, NO_3	Banoeng-Yakubo et al. (2009)
2009	India	GWQI (WQI)	TDS, pH, Hardness, Ca, Mg, Cl, SO_4, HCO_3, F, NO_3, Fe, Mn	Ramakrishnaiah et al. (2009)
2010	India	MPI	Cu, Fe, Mn, Ni, Pb, Zn	Giri et al. (2010)
2010	India	DWQI	EC, pH, Hardness, Ca, Mg, Na, Cl, SO_4, Alkalinity, F, NO_3, NO_2, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn, TC, Salmonella	Ramesh et al. (2010)
2010	India	GWQI	TDS, Ca, Mg, Na, K, Cl, SO_4, HCO_3, F, NO_3, PO_4, Si	Vasanthavigar et al. (2010)
2010	Iran	GWQI	TDS, pH, Ca, Mg, Na, K, Cl, SO_4	Saeedi et al. (2010)
2011	India	GWQI	EC, TDS, pH, Hardness, Ca, Mg, Na, Cl, SO_4, HCO_3, NO_3	Machiwal et al. (2011)
2011	India	GWQI (GQI)	TDS, Ca, Mg, Na, Cl, SO_4	Khan et al. (2011)
2012	Tunisia	GWQI	EC, TDS, pH, Ca, Mg, Na, K, Cl, SO_4, NO_3	Ketata et al. (2012)
2013	Iran	Modified DWQI	Al, NH_3-N, Ca, Cl, Hardness, Fe, Mg, pH, Na, SO_4, TDS, Zn, As, Cd, Cr, Cu, FC, F, Pb, Mn, Hg, NO_3, NO_2, Turbidity	Mohebbi et al. (2013)
2014	Iran	GWQI	TDS, pH, Ca, Mg, Na, K, Cl, SO_4, HCO_3	Sadat-Noori et al. (2014)
2014	Lebanon	GWQI	TDS, Ca, Mg, Na, Cl, SO_4, HCO_3, F, NO_3, NO_2, FC, TC	El-Fadel et al. (2014)
2014	China	Entropy-weighted	TDS, Ca, Mg, Na, K, Cl, SO_4, HCO_3, F,NO_3-N, NO_2-N, NH_4-N, Al, As,	Li et al. (2014)

		fuzzy WQI	Cr, Cu, Hg, Mn, Zn	
2015	Syria	HPI	Cd, Cu, Pb, Zn	Zakhem and Hafez (2015)
2015	Iran	Modified DWQI	Al, NH_3-N, Ca, Cl, Hardness, Fe, Mg, pH, Ryznar index, Na, SO_4, TDS, Zn, As, Cr, Cu, FC, F, Mn, NO_3, NO_2, Turbidity	Abtahi et al. (2015)
2016	Ghana	WQI	EC, TDS, pH, Ca, Mg, Cl, SO_4, HCO_3, NO_3, PO_4	Boateng et al. (2016)
2016	Iran	FGQI/FWQI / FGWQI	TDS, Ca, Mg, Na, Cl, SO_4, NO_3	Vadiati et al. (2016)
2017	Egypt	WQI	EC, TDS, pH, Na, K, Hardness, SAR	El-Shahat et al. (2017)
2017	India	GWQI (GQI)	TDS, pH, Ca, Mg, Na, K, Cl, SO_4, HCO_3, NO_3	Sethy et al. (2017)
2017	Iran	EWQI	EC, Ca, Mg, Na, K, Cl, SO_4, HCO_3, F, NO_3, Al, As, Fe, Mn, Pb	Gorgij et al. (2017)
2018	Iran	PWQI	EC, TDS, pH, Hardness, Ca, Mg, Na, K, Cl, SO_4, HCO_3	Jamshidzadeh and Barzi (2018)
2018	Brazil	IQNAS WQI	TDS, pH, Hardness, Cl, F, NO_3	Leite et al. (2018)
2018	India	IWQI	EC, pH, Na, SAR, RSC, Cl, NO_3, B, As, Cd, F, Fe	Singh et al. (2018)

[1] GWQI/GQI: Groundwater Quality Index; IQNAS WQI: Groundwater Natural Quality Index; SWQI: Surface Water Quality Index; WQI: Water Quality Index; MPI: Metal Pollution Index; Cd: Contamination Index; FGWQI: Fuzzy Ground Water Quality Index; EWQI: Entropy Weighted Water Quality Index; PWQI: Potability Water Quality Index; IAWQ: Index of Aquifer Water Quality; GWCI: Groundwater Composition Index; HPI: Heavy Metal Pollution Index.

[2]TDS: Total Dissolved Solids; DO: Dissolved Oxygen; BOD: Biochemical Oxygen Demand; EC: Electrical Conductivity; FC: Fecal Coliform; TC: Total Coliform; SAR: Sodium Adsorption Ratio.

Table 2. Safe drinking water limits of the parameters prescribed the World Health Organization

S. No.	Parameter	Desirable Limit	Permissible Limit
1	pH	7-8.5	n.a.
2	Total Dissolved Solids	500 mg l^{-1}	1500 mg l^{-1}
3	Calcium	75 mg l^{-1}	200 mg l^{-1}
4	Magnesium	30 mg l^{-1}	150 mg l^{-1}
5	Sodium	200 mg l^{-1}	n.a.
6	Chloride	200 mg l^{-1}	600 mg l^{-1}
7	Sulfate	200 mg l^{-1}	400 mg l^{-1}
8	Nitrate	45 mg l^{-1}	n.a.
9	Total Hardness	100 mg l^{-1}	500 mg l^{-1}

Source: WHO (2017); n.a.: not available

Table 3. Number of sites with presence of significant serial correlation

S. No.	Block	Sites in Individual Block	Number of Sites with Presence of Serial Correlation in Groundwater Quality Parameters													
			EC	TDS	pH	Na	K	Ca	Mg	Cl	SO_4	CO_3	HCO_3	NO_3	F	TH
1	Amber	11	1	1	0	1	1	1	0	0	0	2	2	2	1	0
2	Bassi	14	2	4	4	1	1	0	3	1	2	3	0	1	0	1
3	Chaksu	14	0	0	0	0	2	0	1	1	3	1	2	0	0	1
4	Dudu	22	1	1	4	3	5	2	2	3	4	3	5	4	1	1
5	Govindgarh	12	2	1	2	1	2	1	1	2	0	2	0	3	0	1
6	Jamwaramgarh	14	0	0	4	0	1	0	2	0	1	1	4	1	0	2
7	Jhotwara	14	1	1	4	0	3	2	1	2	1	1	4	1	3	2
8	Kotputli	19	2	2	6	2	2	3	0	3	5	2	2	0	1	0
9	Phagi	17	3	1	2	2	5	2	2	1	0	0	3	2	3	2
10	Sambhar	13	1	1	2	0	0	0	1	0	2	3	0	2	0	1
11	Sanganer	14	1	1	2	1	2	2	0	2	1	2	1	0	0	0
12	Shahpura	13	1	1	4	0	2	0	1	2	0	2	0	1	0	2
13	Viratnagar	19	2	2	4	2	2	6	1	2	0	3	3	1	1	1
Total			17	16	38	13	28	19	15	19	19	25	26	18	10	14

Table 4. Range of three groups of vulnerability classes for nine groundwater quality parameters considered in this study

S. No.	Parameter	Vulnerability Class		
		Low	**Moderate**	**High**
1	pH	0-1.5	1.5-3.0	>3.0
2	Total Dissolved Solids (mg l^{-1})	0-500	500-1000	>1000
3	Calcium (mg l^{-1})	0-75	75-150	>150
4	Magnesium (mg l^{-1})	0-30	30-60	>60
5	Sodium (mg l^{-1})	0-200	200-400	>400
6	Chloride (mg l^{-1})	0-200	200-400	>400
7	Sulfate (mg l^{-1})	0-200	200-400	>400
8	Nitrate (mg l^{-1})	0-45	45-90	>90
9	Total Hardness (mg l^{-1})	0-100	100-200	>200

Table 5. Values of optimum index factors for five best combinations of three groundwater quality parameters

S. No.	Three Parameters			Optimum Index Factor
1	TDS	pH	NO_3	718.43
2	TDS	pH	Hardness	642.01
3	TDS	pH	Mg	589.74
4	TDS	pH	Cl	583.37
5	TDS	pH	Ca	575.07

Figure Caption

Fig. 1. Location map of study area showing groundwater monitoring sites

Fig. 2. Box-whisker plots of 14 groundwater quality parameters for 13 blocks (AM – Amber, BS – Bassi, CH – Chaksu, DU – Dudu, GG – Govindgarh, JR – Jamwaramgarh, JW – Jhotwara, KT – Kotputli, PH – Phagi, SB – Sambhar, SN – Sanganer, SP – Shahpura, VN – Viratnagar)

Fig. 3. Spatially-distributed results of the Mann-Kendall test showing presence of non-significant (n.s.) and significant increasing/decreasing trends at 1, 5 and 10% level of significance (l.s.) for 14 groundwater quality parameters

Fig. 4. Barcharts illustrating trend magnitudes of 14 groundwater quality parameters through Sen's slope estimates over 13 blocks

Fig. 5. Sites classified into three groups according to 'low', 'moderate' and 'high' reliability (R_y) of nine groundwater quality parameters

Fig. 6. Sites classified into three groups according to 'low', 'moderate' and 'high' resilience (R_e) of nine groundwater quality parameters

Fig. 7. Sites classified into three groups according to 'low', 'moderate' and 'high' vulnerability (V_y) of nine groundwater quality parameters

Fig. 8. Spatial distribution maps of (a) probabilistic groundwater quality stability index (GQSI) and (b) groundwater quality index (GQI) classified into three classes

Fig. 9. Spatial comparison of groundwater quality stability index (GQSI) and groundwater quality index (GQI) illustrating their similar/contrasting results

Fig. 10. Barcharts classifying sites, having good, moderate and poor groundwater quality, into groups of good, moderate and poor groundwater quality stability index

Fig. 11. (a) Spatial distribution of optimum groundwater quality stability index (GQSI) classifying sites into three groups, and (b) scatter plot of GQSI versus optimum GQSI (OGQSI) along with fitted straight line, 1:1 line, and coefficient of determination (R^2)

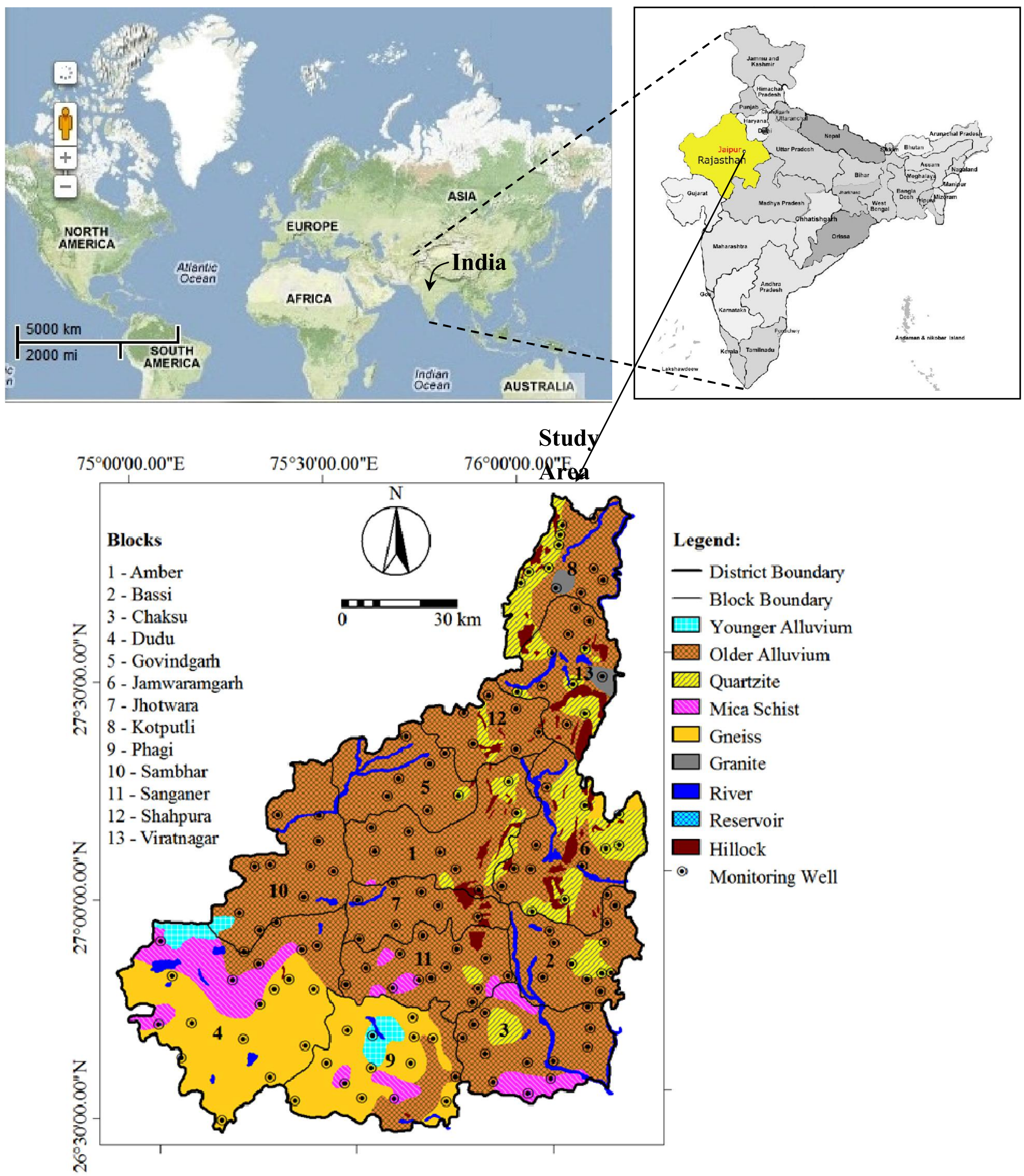

NORTH AMERICA
EUROPE
ASIA
AFRICA
SOUTH AMERICA
AUSTRALIA
Atlantic Ocean
Indian Ocean
5000 km
2000 mi
India
Jaipur
Rajasthan
Study Area
75°00'00.00"E
75°30'00.00"E
76°00'00.00"E
27°30'00.00"N
27°00'00.00"N
26°30'00.00"N
26°30'00.00"N
N
0
30 km
Blocks
1 - Amber
2 - Bassi
3 - Chaksu
4 - Dudu
5 - Govindgarh
6 - Jamwaramgarh
7 - Jhotwara
8 - Kotputli
9 - Phagi
10 - Sambhar
11 - Sanganer
12 - Shahpura
13 - Viratnagar
Legend:
District Boundary
Block Boundary
Younger Alluvium
Older Alluvium
Quartzite
Mica Schist
Gneiss
Granite
River
Reservoir
Hillock
Monitoring Well

Figure 1

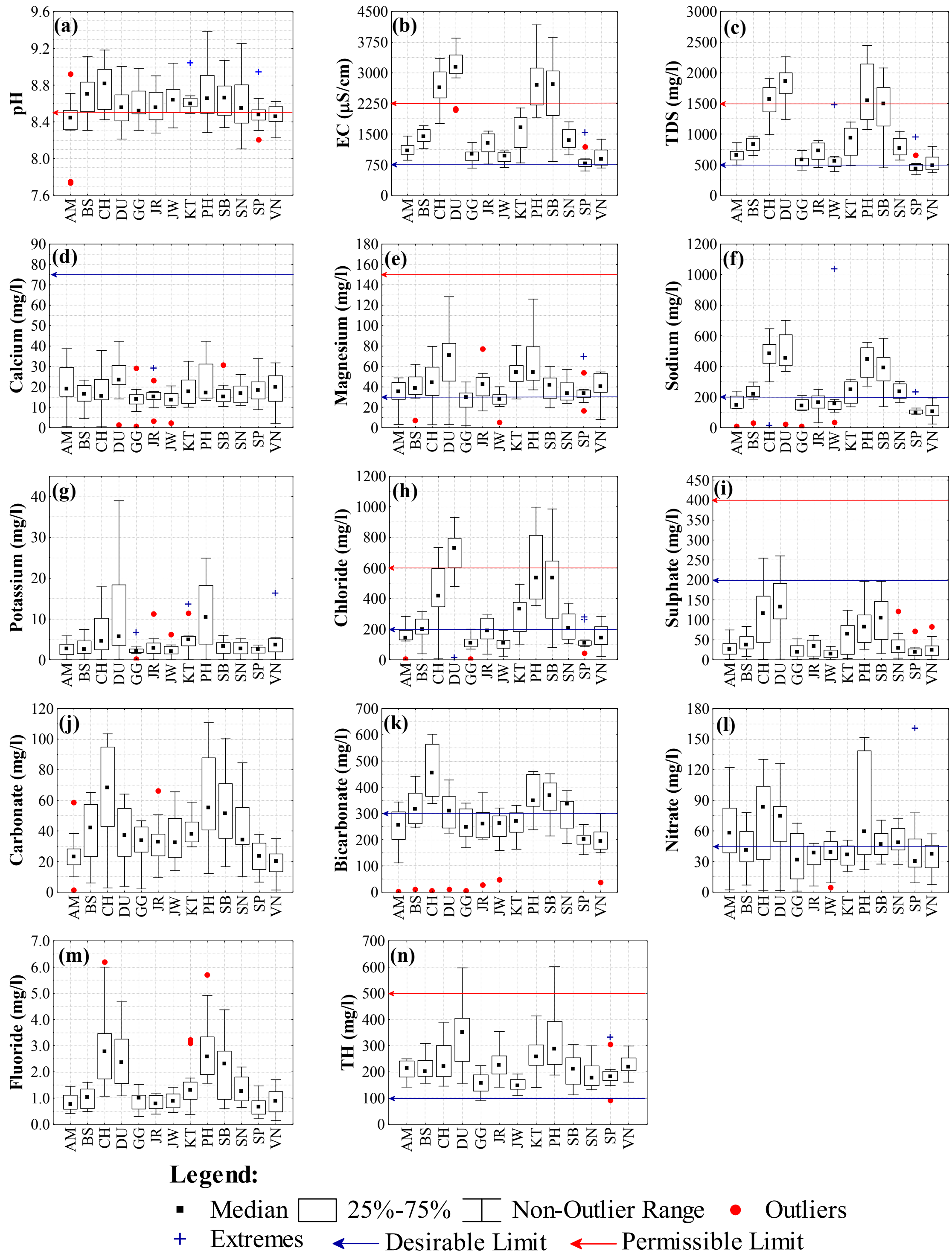
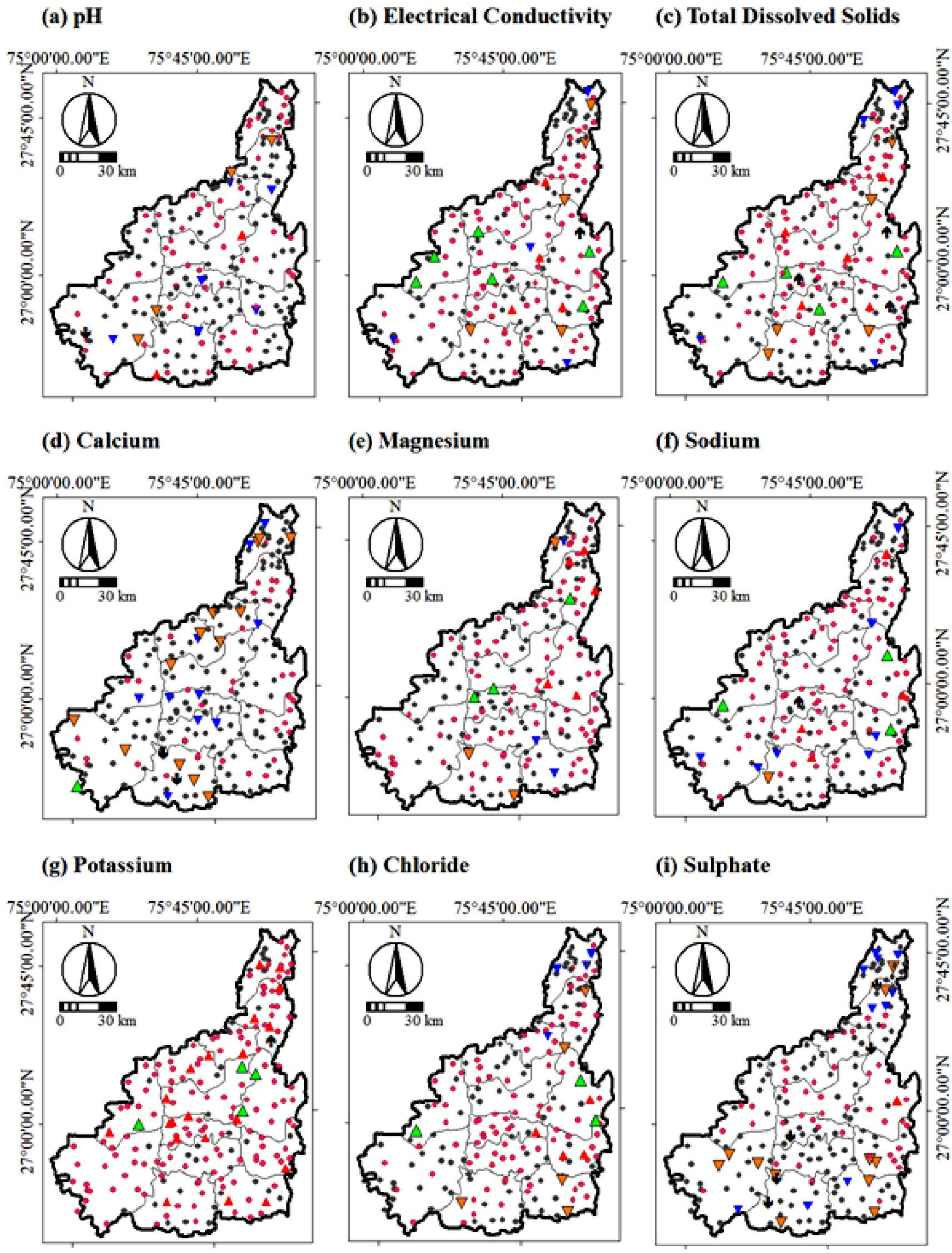

(a)
pH
(b)
EC (µS/cm)
(c)
TDS (mg/l)
(d)
Calcium (mg/l)
(e)
Magnesium (mg/l)
(f)
Sodium (mg/l)
(g)
Potassium (mg/l)
(h)
Chloride (mg/l)
(i)
Sulphate (mg/l)
(j)
Carbonate (mg/l)
(k)
Bicarbonate (mg/l)
(l)
Nitrate (mg/l)
(m)
Fluoride (mg/l)
(n)
TH (mg/l)
AM BS CH DU GG JR JW KT PH SB SN SP VN
Legend:
Median
25%-75%
Non-Outlier Range
Outliers
Extremes
Desirable Limit
Permissible Limit

Figure 2


(a) pH
(b) Electrical Conductivity
(c) Total Dissolved Solids
(d) Calcium
(e) Magnesium
(f) Sodium
(g) Potassium
(h) Chloride
(i) Sulphate
75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0 30 km

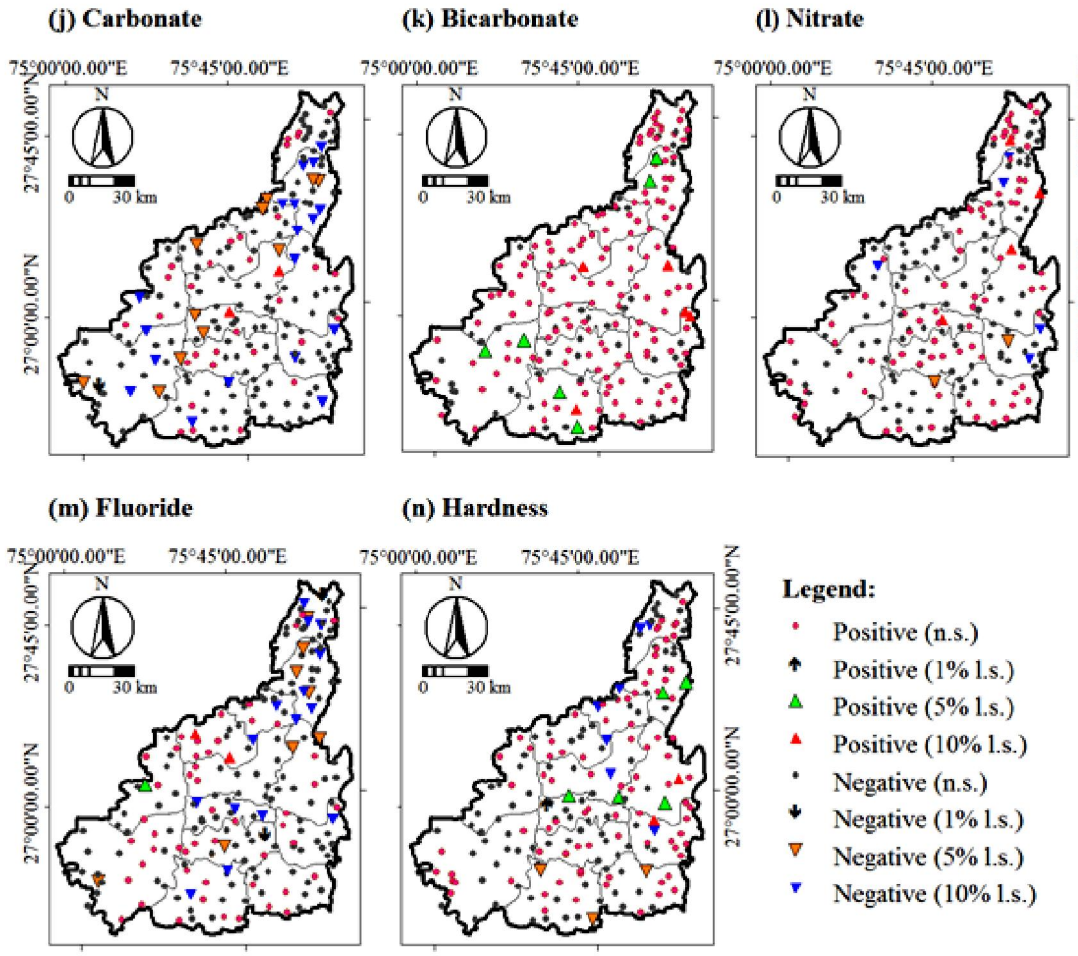

(j) Carbonate
(k) Bicarbonate
(l) Nitrate
(m) Fluoride
(n) Hardness
75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0 30 km
Legend:
Positive (n.s.)
Positive (1% l.s.)
Positive (5% l.s.)
Positive (10% l.s.)
Negative (n.s.)
Negative (1% l.s.)
Negative (5% l.s.)
Negative (10% l.s.)

Figure 3

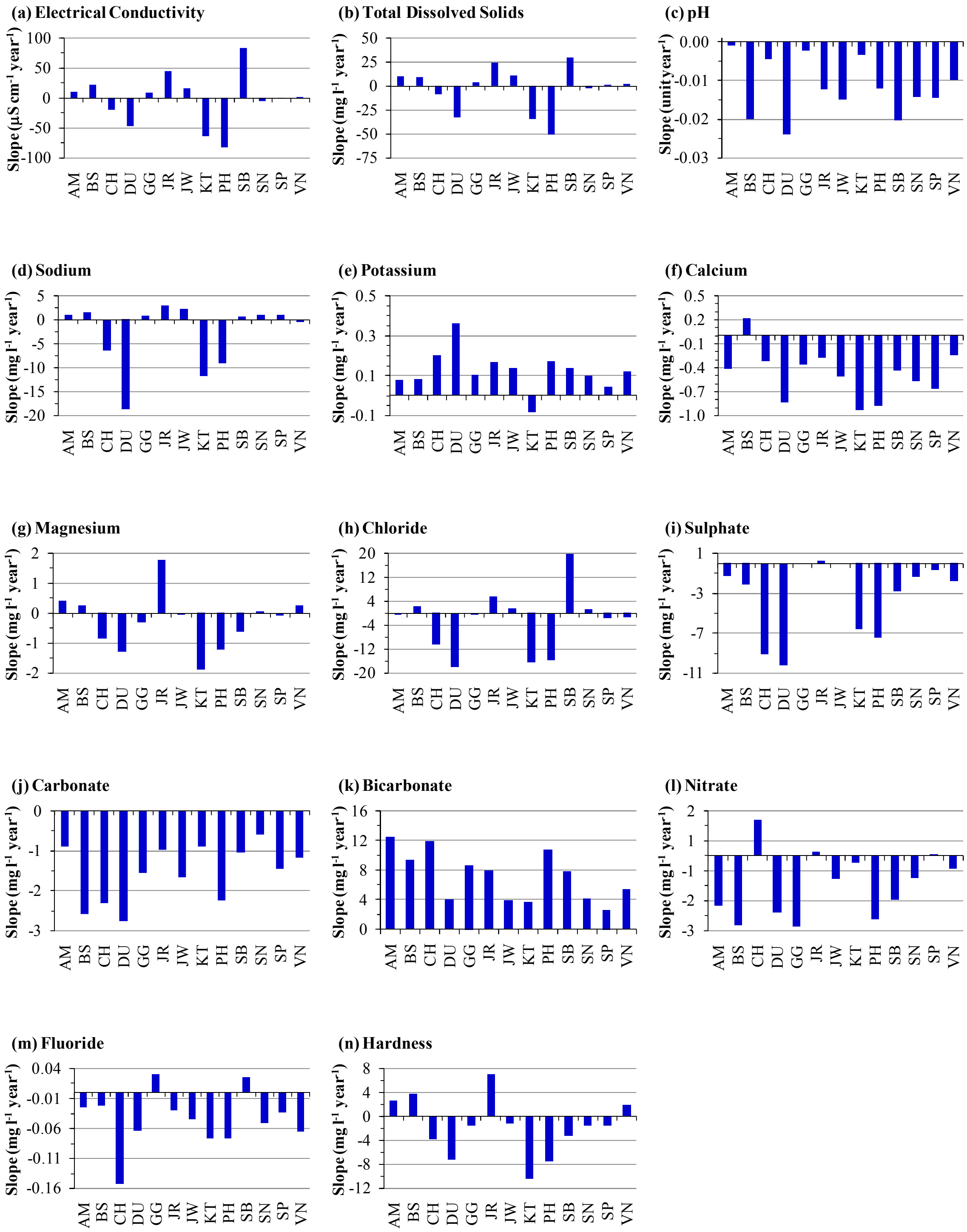

(a) Electrical Conductivity
Slope (µS cm-1 year-1)
(b) Total Dissolved Solids
Slope (mg l-1 year-1)
(c) pH
Slope (unit year-1)
(d) Sodium
(e) Potassium
(f) Calcium
(g) Magnesium
(h) Chloride
(i) Sulphate
(j) Carbonate
(k) Bicarbonate
(l) Nitrate
(m) Fluoride
(n) Hardness
Slope (mg l-1 year-1)
AM BS CH DU GG JR JW KT PH SB SN SP VN

Figure 4

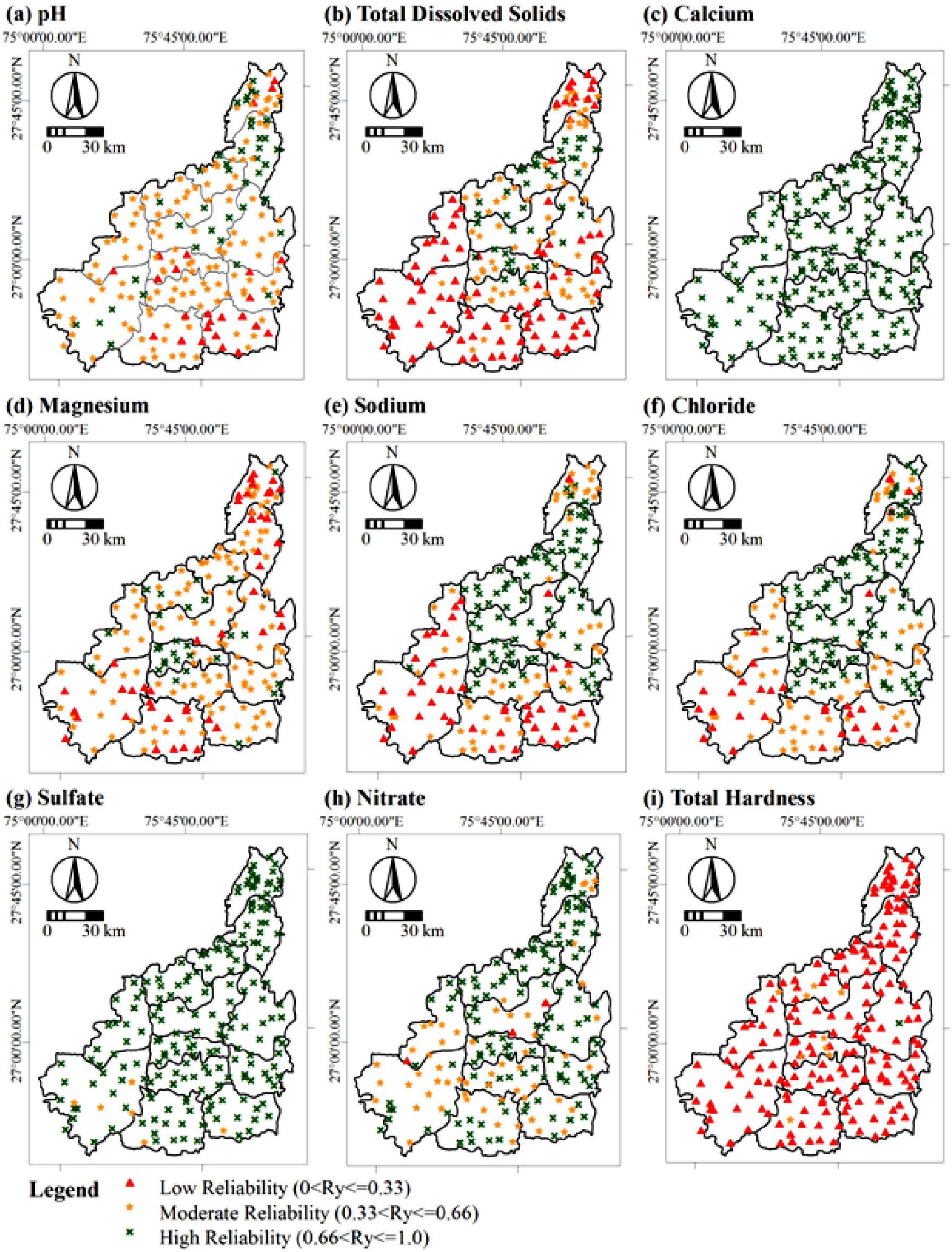

(a) pH
(b) Total Dissolved Solids
(c) Calcium
(d) Magnesium
(e) Sodium
(f) Chloride
(g) Sulfate
(h) Nitrate
(i) Total Hardness
75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0 30 km
Legend
Low Reliability (0<Ry<=0.33)
Moderate Reliability (0.33<Ry<=0.66)
High Reliability (0.66<Ry<=1.0)

Figure 5

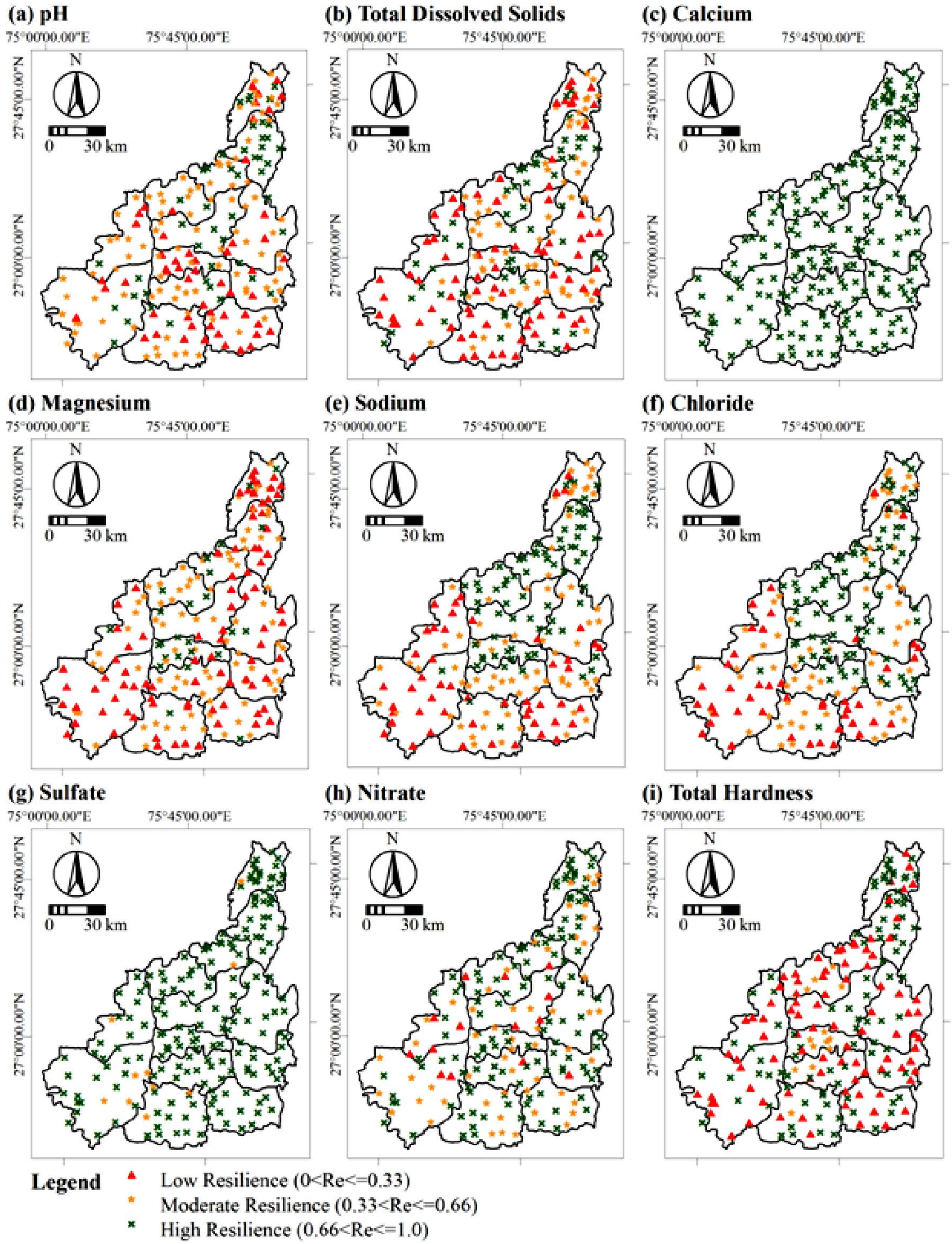

(a) pH
(b) Total Dissolved Solids
(c) Calcium
(d) Magnesium
(e) Sodium
(f) Chloride
(g) Sulfate
(h) Nitrate
(i) Total Hardness
75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0 30 km
Legend
Low Resilience (0<Re<=0.33)
Moderate Resilience (0.33<Re<=0.66)
High Resilience (0.66<Re<=1.0)

Figure 6

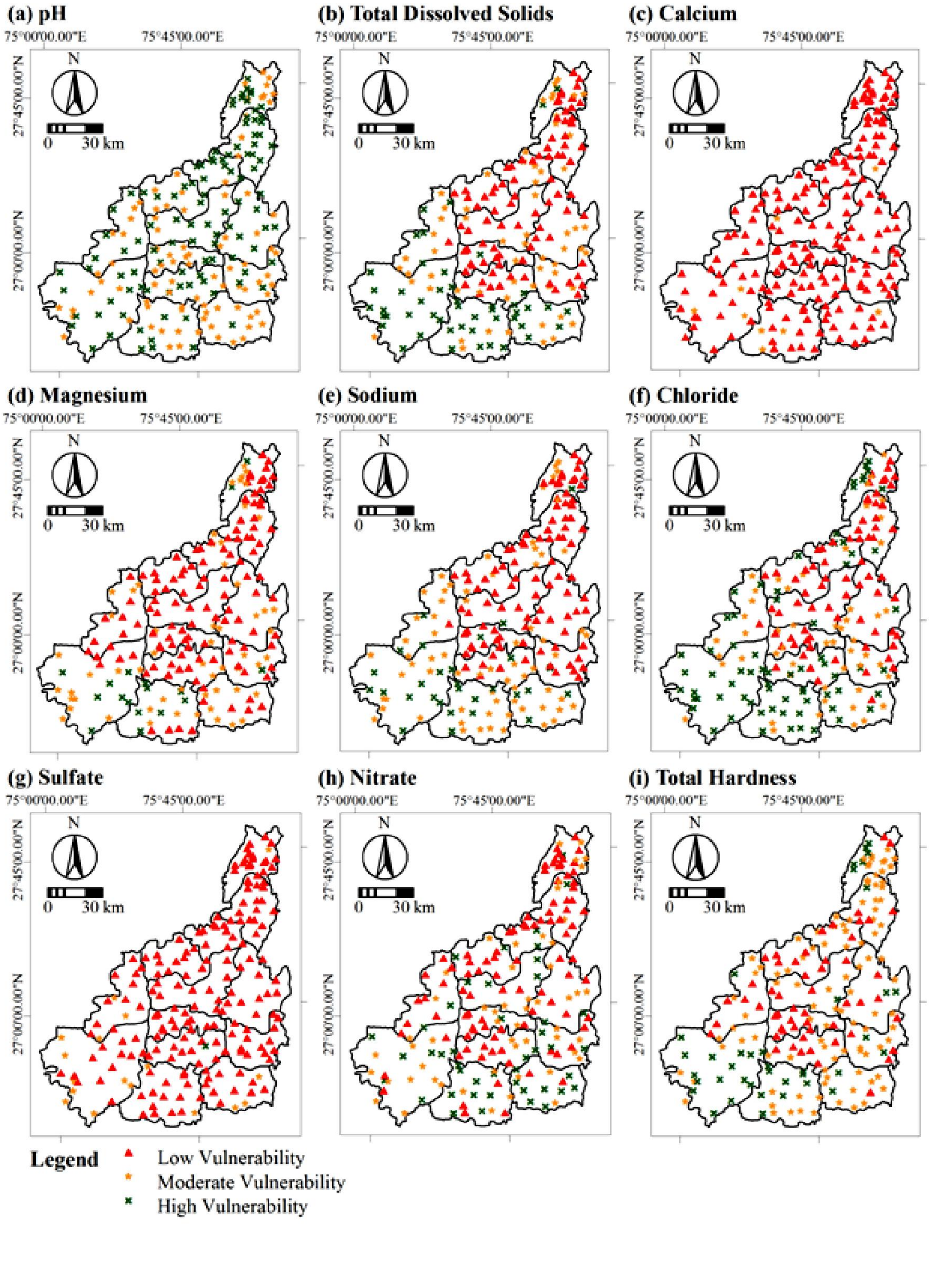

(a) pH
(b) Total Dissolved Solids
(c) Calcium
(d) Magnesium
(e) Sodium
(f) Chloride
(g) Sulfate
(h) Nitrate
(i) Total Hardness
75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0 30 km
Legend
Low Vulnerability
Moderate Vulnerability
High Vulnerability

Figure 7

(a) GQSI Map **(b) GQI Map**

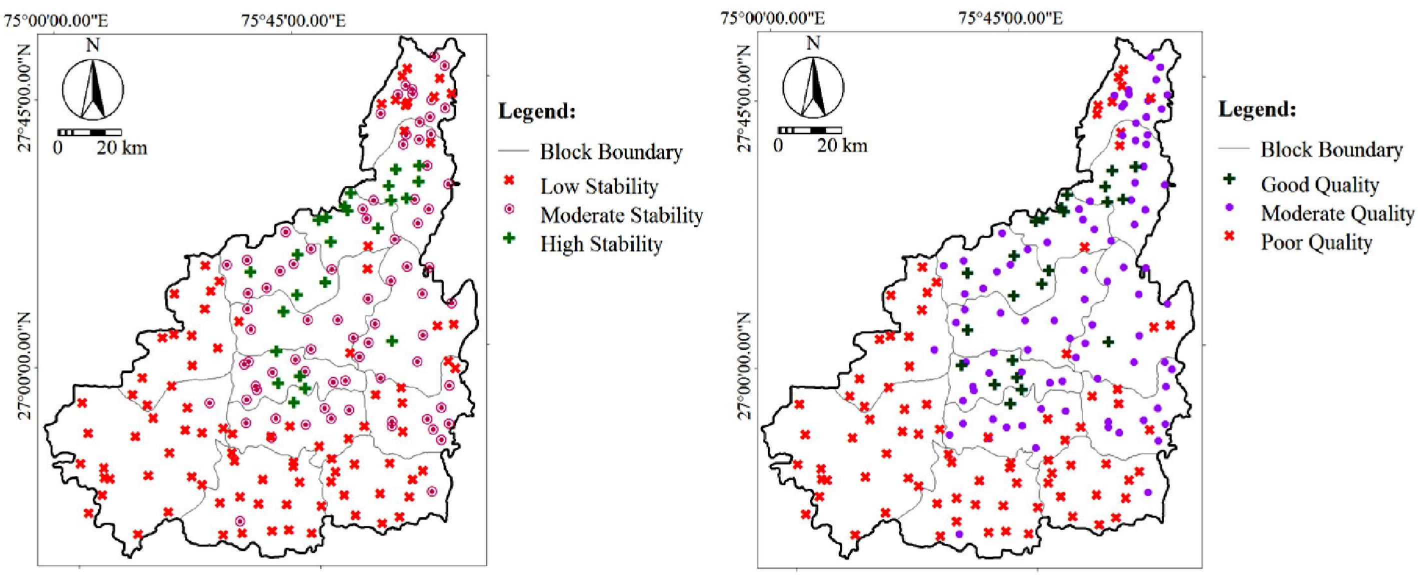


Figure 8

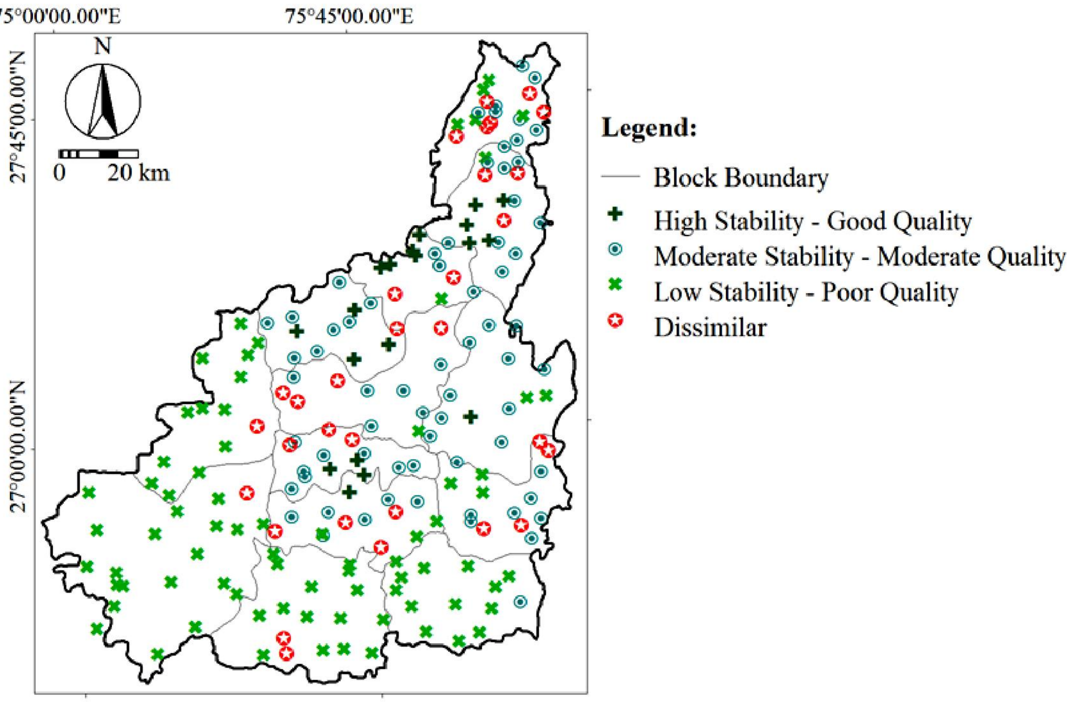

75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0
20 km
Legend:
Block Boundary
High Stability - Good Quality
Moderate Stability - Moderate Quality
Low Stability - Poor Quality
Dissimilar

Figure 9

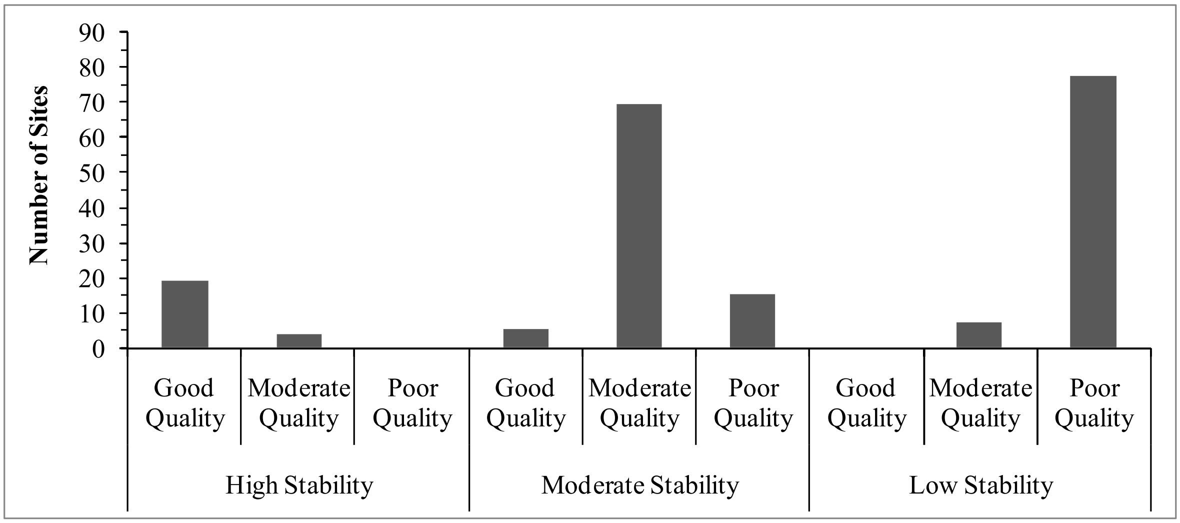
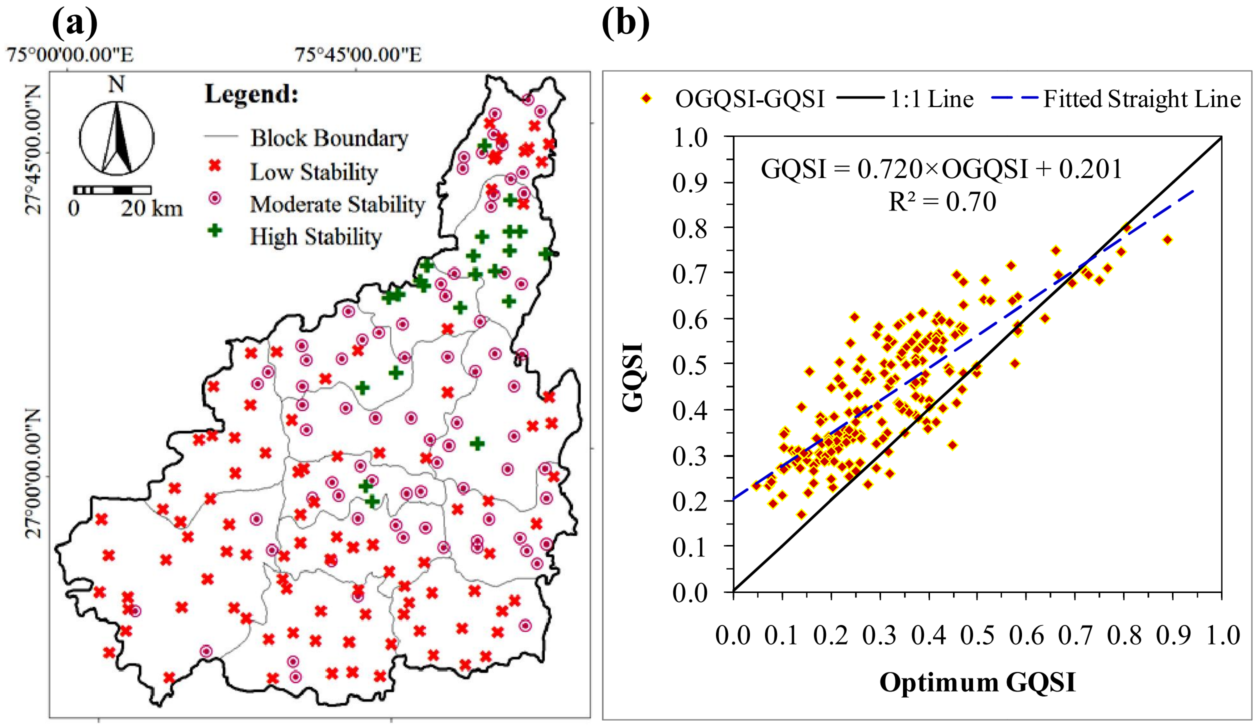

Number of Sites
90
80
70
60
50
40
30
20
10
0
Good Quality
Moderate Quality
Poor Quality
Good Quality
Moderate Quality
Poor Quality
Good Quality
Moderate Quality
Poor Quality
High Stability
Moderate Stability
Low Stability

Figure 10



(a)
(b)
75°00'00.00"E
75°45'00.00"E
27°45'00.00"N
27°00'00.00"N
N
0 20 km
Legend:
Block Boundary
Low Stability
Moderate Stability
High Stability
OGQSI-GQSI
1:1 Line
Fitted Straight Line
GQSI = 0.720×OGQSI + 0.201
R² = 0.70
GQSI
Optimum GQSI
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0

Figure 11