Environ Monit Assess (2009) 159:341–351
DOI 10.1007/s10661-008-0633-7

Assessment of groundwater quality and hydrogeochemistry of Manimuktha River basin, Tamil Nadu, India

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Received: 28 February 2008 / Accepted: 5 November 2008 / Published online: 17 December 2008

Abstract Groundwater quality assessment study was carried out around Manimuktha river basin, Tamil Nadu, India. Twenty six bore well samples were analyzed for geochemical variations and quality of groundwater. Four major hydrochemical facies ($Ca–HCO_3$, Na–Cl, Mixed $CaNaHCO_3$, and mixed CaMgCl) were identified using a Piper trilinear diagram. Comparison of geochemical results with World Health Organization, United States Environmental Protection Agency, and Indian Standard Institution drinking water standards shows that all groundwater samples except few are suitable for drinking and irrigation purposes. The major groundwater pollutions are nitrate and phosphate ions due to sewage effluents and fertilizer applications. The study reveals that the groundwater quality changed due to anthropogenic and natural influence such as agricultural, natural weathering process.

Keywords Groundwater quality assessment · Geochemical process · Hydrogeochemistry · Major ions · Manimuktha River basin

Introduction

Groundwater is one of the important sources of water used for domestic and industrial purpose. In India, sedimentary aquifers are the important source of groundwater. Changes in groundwater quality are due to rock–water interaction and oxidation–reduction reactions during the percolation of water through the aquifers. In addition to these processes, water-born pathogens, toxic and nontoxic pollutants are the major water quality degradation parameters which are transported from recharge area to discharge area through aquifers by groundwater motion. Undesirable and soluble constituents in the water cannot be controlled after entering the ground (Johnson 1979; Sastri 1994). Sami (1992) has explained that leaching of surficial salts, ion-exchange processes, and residential time of groundwater in the aquifer causes the hydrogeochemical variations in the groundwater. In India and various parts of the

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world, numerous studies have been carried out to assess the geochemical characteristics of groundwater (Graniel et al. 1999; Umar and Sami Ahmad 2000). Several comprehensive studies are also carried out in south India (Subba Rao et al. 1998; Elango et al. 2003; Krishnakumar 2004; Jeevanandam et al. 2006).

Tertiary sandstone aquifer of Manimuktha River Basin is well known for groundwater potential and its artesian condition. Confining pressure was reduced due to overexploitation of groundwater for agricultural and industrial purposes and frequent failure of monsoon. The sandstone formation is separated from recent alluvium by impervious clay layers. The overexploitation of groundwater for domestic, industrial activities and anthropogenic pollution-induced factors reduced the groundwater quality. Under these circumstances, the aim of the present study is to identify the hydrogeochemical characteristics and quality factors of the Manimuktha river basin, Tamil Nadu, India.

Geological and hydrogeological settings of the study area

The Manimuktha river basin is located in the parts of Cuddalore and Viluppuram districts (11°40′00″ to 12°00′00″ and 79°00′00″ to 79°20′00″) in northern Tamil Nadu (Fig. 1). The river originates from the Kalrayan hills, and two tributaries Mayuranathi and Gomuhanathi join the river near

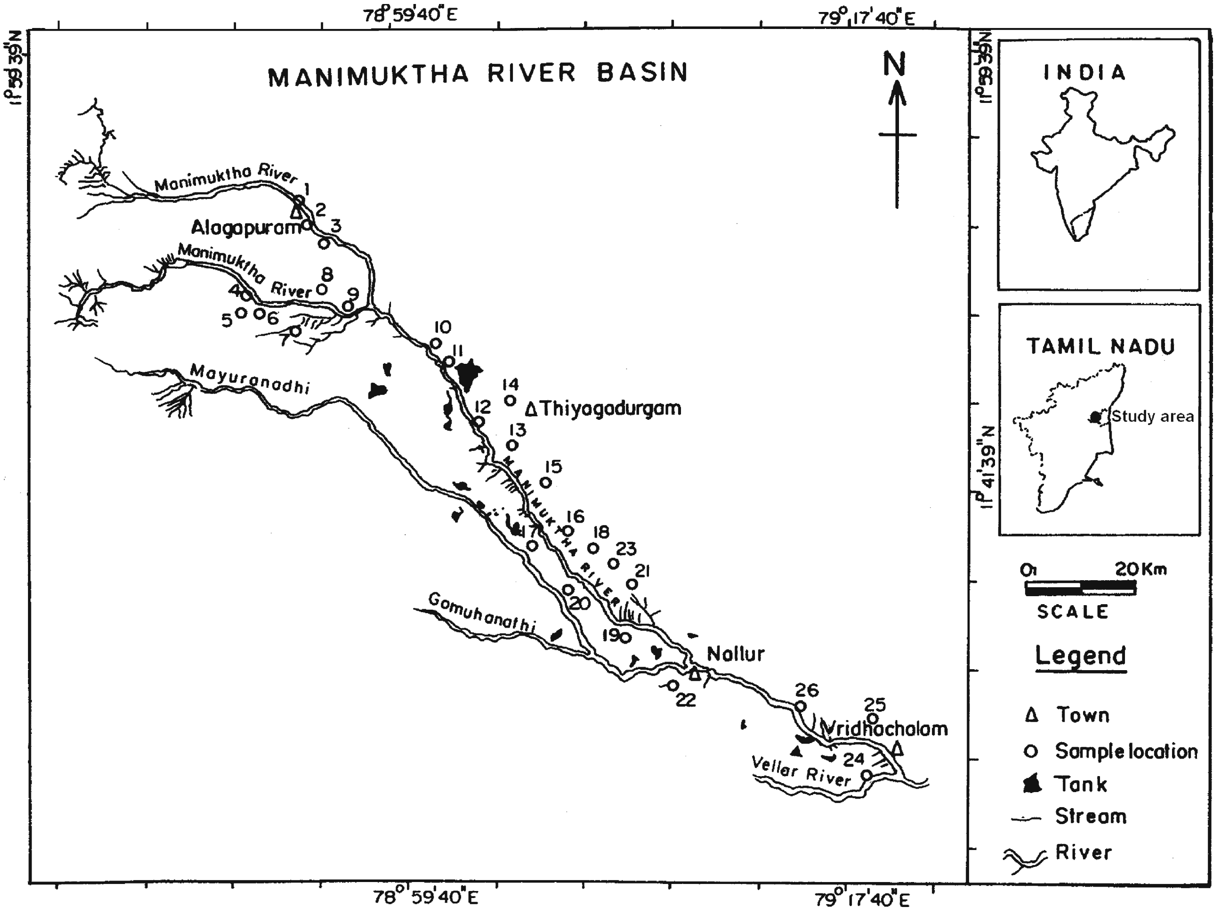


Fig. 1 Sample location map of the study area

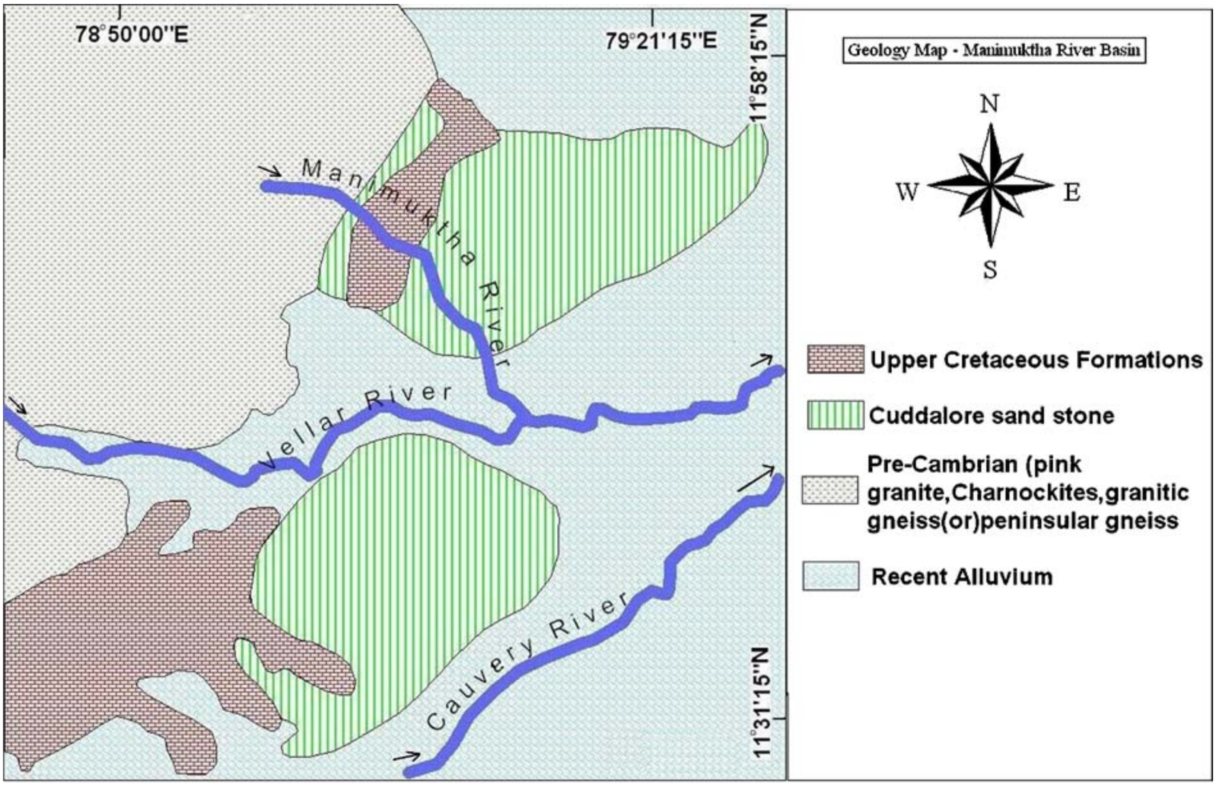


Fig. 2 Geology map of the study area

Nallur village before its confluence with Vellar River. The average annual rainfall in the river basin is 1,080 mm with about 70% of the rainfall received during the northeast monsoon from October to December. The flow of water in the river is reduced during the period from February to June, and as a result, the community in the region depends on groundwater for their use. A major part of the study area fall in the agricultural activities, where sugarcane, paddy, and groundnut are being cultivated.

The upper reaches of the river basin comprises the Precambrian Peninsular Gneiss and its retrograded products. The lower reaches of the basin is constituted by Tertiary sandstone, which include white clay, sand, sandy clays, and unconsolidated sand stone. At some locations, small patches of Cretaceous formations are exposed, and it is overlain by recent alluvium (Fig. 2). Weathered, fractured/jointed charnockites, gneisses, cuddalore sandstone, and recent alluvium act as good aquifers in this study area. The depth of bore holes in upper and lower reaches ranges from 90 to 150 and 50 to 70 ft, respectively.

Materials and methods

Twenty-six groundwater samples were collected from upper reaches of the basin to confluence point of the river with Vellar (near Vridhachalam town); the polythene containers were washed to avoid contamination. The pH, electrical conductivity (EC), total dissolved solids (TDS) were measured at the sample site using handheld analyzing kits. Groundwater samples were collected, and the samples were kept in a polyethylene bottle at 10°C for further laboratory analysis. The extrapure analytical reagents and chemical standards (Merck Grade) were used for the groundwater quality assessment. The analytical procedures are suggested by the American Public Health Association (APHA 1995). Carbonate (CO_3) and bicarbonate (HCO_3) were determined using acid titration method; chloride (Cl) concentration was measured by $AgNO_3$ titration method; sulfate measured by $BaCl_3$ method using spectrophotometer. Orthophosphate (PO_4) analysis was carried out using spectrophotometer–ascorbic acid method; nitrate was analyzed using cadmium column reduction method; sodium (Na) and potassium (K) were analyzed using flame photometer. calcium (Ca) and magnesium (Mg) were determined using the titration method.

Results and discussion

General hydrogeochemistry

The major ion concentrations, physical parameters and drinking water standards are presented in Table 1. The temperature variation of the groundwater in the study area ranges from 30°C to 32°C. The majority of the groundwater samples are colorless and a few samples were brown in color due to dissolved iron contents. The turbid nature of groundwater is due to the presence of suspended solids and silt. The total dissolved solids (TDS) in the groundwater ranges from 400 to 3,400 mg/l, with a mean value of 1,596 mg/l, if the TDS values of samples exceed the permissible limit recommended by World Health Organization (WHO), United States Environmental Protection Agency (USEPA) and Indian Standard Institution (ISI) standards making the groundwater unsuitable for various domestic and drinking purposes. Based on this values, groundwater samples in the study area varied from fresh (TDS $<$ 1,000 mg/l) to brackish (TDS $>$ 1,000 mg/l) in nature (Freeze and Cherry 1979). The pH of the groundwater in the study area ranges from 7.2 to 8.4, with an average value of 8.01. The pH values of groundwater samples are within the permissible limit suggested by WHO, USEPA, and ISI.

Major cation chemistry

The high concentration of Ca, Na, and Mg in groundwater is due to clay minerals such as montmorillonite, illite and chlorite (Garrels 1976).

Table 1 Drinking water specifications of the study area in comparison with WHO (1971), USEPA (1992), ISI (1983), minimum, maximum, and mean ion concentration

Parameters	This study			WHO standards (1971)	U.S Environmental Protection Agency Secondary drinking water standards	Indian Standard Institution (ISI 1983)
	Minimum	Maximum	Mean			
PH	7.2	8.4	8.01	6.5–8.5 on scale	6.5–8.5 on scale	6.5–8.5 on scale
TDS (mg/l)	400	3400	1596	500	500	500
CO_3^- (mg/l)	6	36	19.38	–	–	–
HCO_3^- (mg/l)	85.4	414.8	259.95	–	–	–
Cl^- (mg/l)	44.3	514	184.7	200	250	250
SO_4^- (mg/l)	2.1	33.9	14.7	200	250	150
PO_4^- (mg/l)	6.8		6.8	–	0.1	–
NO_2(as N) (mg/l)	0.4	12.2	4.9	45	10	45
NO_3(as Nitrite) (mg/l)	1.5	1717.9	465.51	$<$ 0.1	1	–
Ca^+ (mg/l)	2	72	11.38	75	–	75
Mg^+ (mg/l)	2.4	97.2	27.44	$<$ 30 if SO_4 is 250 mg/l, up to 150 mg/l if SO_4 is less than 250 mg/l	–	30
Na^+ (mg/l)	32	170	98.88	200	–	–
K^+ (mg/l)	1	153	15.07	12	–	–

WHO World Health Organization, *USEPA* United States Environmental Protection Agency

The major cation concentrations (Ca^+, Mg^+, Na^+, K^+) in the groundwater are below the WHO standards. Calcium and magnesium ions present in groundwater is particularly derived from leaching of limestone, dolomites, gypsum and anhydrites, whereas the calcium ions is also derived from cation exchange process (Garrels 1976). The concentration of calcium ion ranges from 2 to 72 mg/l, with an average concentration of 11.38 mg/l. The concentration of magnesium ranges from 2.4 to 97.2 mg/l, with an average value of 27.44 mg/l. The concentration of magnesium ion in this groundwater samples is relatively high when compared to calcium ion concentration and the magnesium concentration is mostly due to weathering of magnesium minerals and leaching of dolomites (Eq. 1).

$$(\mathrm{Ca, Mg})\,\mathrm{CO_3}+\mathrm{CO_2}+\mathrm{H_2O}=2\mathrm{HCO_3^-}+\mathrm{Ca^{2+}}+\mathrm{Mg^{2+}} \tag{1}$$

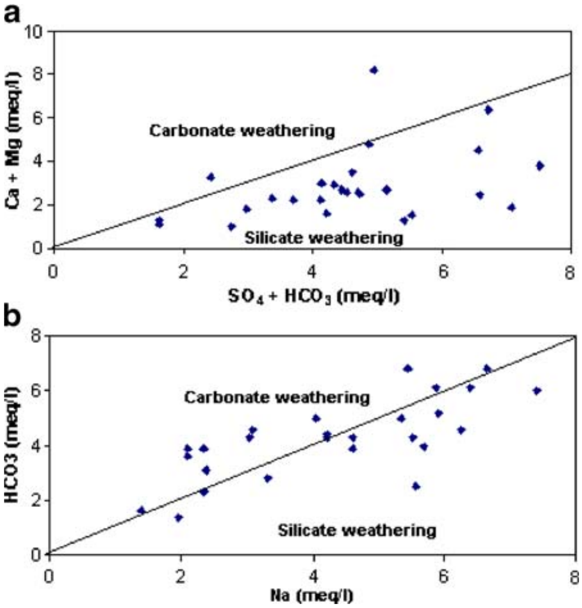


Fig. 3 **a**, **b** Scatter diagram for carbonate weathering *vs* silicate weathering processes

The $(\mathrm{Ca^{2+}}+\mathrm{Mg^{2+}})$ vs $(\mathrm{HCO_3^-}+\mathrm{SO_4^-})$ scatter diagram (Datta and Tyagi 1996) shows that most of the samples are falling below the equiline. It is indicating that the silicate weathering is the dominant process for supply of the calcium ions to the groundwater (Fig. 3a). In addition to silicate weathering, the carbonate weathering process is also a contributor for increasing of calcium ions in this groundwater. The possible source of sodium concentration in groundwater is due to dissolution of rock salts and weathering of sodium-bearing minerals. The concentration of sodium and potassium ranges from 32–170 to 1–153 mg/l, with a mean value of 98.88 and 15.07 mg/l. If the halite dissolution process is responsible for the sodium, Na/Cl ratio should be approximately 1, whereas the Na/Cl ratio greater than 1 typically indicates that the sodium was released from silicate weathering (Meyback 1987). In this present study, Na/Cl ratio is less than 1 in the predominant groundwater samples, making one to conclude

Table 2 Correlation coefficient matrix (R2) of major cations and anions of the study area

Correlation	pH	TDS	CO_3	HCO_3	Cl	SO_4	PO_4	NO_2	NO_3	Ca	Mg	Na	K
pH	1.000												
TDS	−0.410	1.000											
CO_3	0.171	−0.261	1.000										
HCO_3	−0.010	0.014	0.296	1.000									
Cl	−0.348	−0.029	0.189	0.664	1.000								
SO_4	−0.315	−0.048	0.233	0.693	0.729	1.000							
PO_4	0.217	−0.132	−0.030	0.105	−0.161	0.150	1.000						
NO_2	0.204	−0.132	0.212	−0.032	0.070	−0.132	−0.048	1.000					
NO_3	−0.364	0.259	−0.156	0.232	0.519	0.387	0.014	−0.111	1.000				
Ca	−0.322	−0.062	0.156	0.335	0.491	0.639	0.095	−0.125	0.358	1.000			
Mg	−0.032	0.048	−0.050	0.244	0.301	0.113	−0.226	−0.060	0.248	−0.286	1.000		
Na	−0.012	−0.036	0.408	0.732	0.551	0.619	0.123	0.194	0.332	0.102	0.068	1.000	
K	−0.038	−0.226	0.157	0.404	0.380	0.539	0.532	−0.058	0.266	0.795	−0.318	0.218	1.000

that the significant reduction of Na concentration may be due to ion exchange process (Fig. 3b). The increase in HCO_3^- concentration compared to Na^+ concentration in the groundwater indicates the dominancy of silicate weathering process; it is well supported by a high concentration of HCO_3^- (Elango et al. 2003). Potassium ion concentration in the groundwater also comes from the above said process. The dominancy of major cations and anions are shown as $Na > Mg > Ca > K = Cl > HCO_3 > CO_3 > SO_4$.

Major anion chemistry

The carbonate and bicarbonate concentration in groundwater is derived from carbonate weathering as well as dissolution of carbonic acid in the aquifers (Jeevanandam et al. 2006; Eq. 2).

$$CaCO_3 + CO_2 + H_2O \rightarrow Ca^{2+} + 2HCO_3^- \text{ and}$$
$$CO_2 + H_2O \rightarrow H^+ + HCO_3^- \qquad (2)$$

The concentration of carbonate and bicarbonate value ranges from 6 to 36 and 85.4 to 414.8 mg/l, with a mean value of 19.38 and 259.95 mg/l, respectively. The bicarbonate is dominant anion among the other anions of this study area. The availability of carbonate minerals in the recharge areas and silicate weathering are the main factors, which led to the increase of carbonates and bicarbonate concentration in the groundwater (Elango et al. 2003). The chloride concentration ranges from 44.3 to 514 mg/l, with a mean value of 184.7 mg/l. High chloride content has a deleterious effect on metallic pipes, structures, and agriculture crops. The natural process such as weathering, dissolution of salt deposits, and irrigation drainage return flow are responsible for chloride content in the groundwater, which is supported by Cl/HCO_3 ratio of 0.4 to 3.0 (Lusczynski and Swarzenski 1996). Chloride concentration in the groundwater is exceeding the limits of WHO and ISI standards (Table 1).

Table 3 Factor analysis of major cations and anions of the study area

Factor	1	2	3
PH	−0.075	0.300	0.181
TDS	−0.008	−0.300	−0.092
CO_3	0.074	0.210	0.235
HCO_3	0.192	0.022	0.181
Cl	0.204	−0.113	0.118
SO4	0.220	−0.012	−0.004
PO4	0.040	0.245	−0.195
NO2	−0.012	0.135	0.234
NO3	0.134	−0.221	−0.076
Ca	0.172	0.054	−0.278
Mg	0.021	−0.228	0.288
Na	0.168	0.050	0.257
K	0.165	0.208	−0.263

The concentrations of sulfate ranges from 2.1 to 33.9 mg/l, with a mean value of 14.7 mg/l. Sulfate ion concentrations are probably derived from weathering of sulfate and gypsum-bearing sedimentary rocks (Elango et al. 2003; Jeevanandam et al. 2006). The sulfate ion concentration is higher in predominant samples when compared with WHO standard. The phosphate ion concentration is low in all locations except one (well no. 26, 6.8 mg/l) because the location is in the vicinity of the paddy field and can be due to usage of phosphate-bearing fertilizers. The concentration of nitrate ranges from 0.4 to 12.2 mg/l, with a mean value of 4.9 mg/l. WHO prescribed maximum

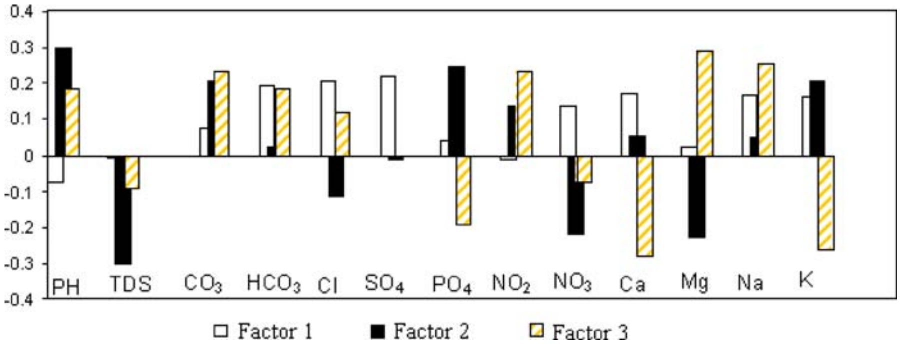
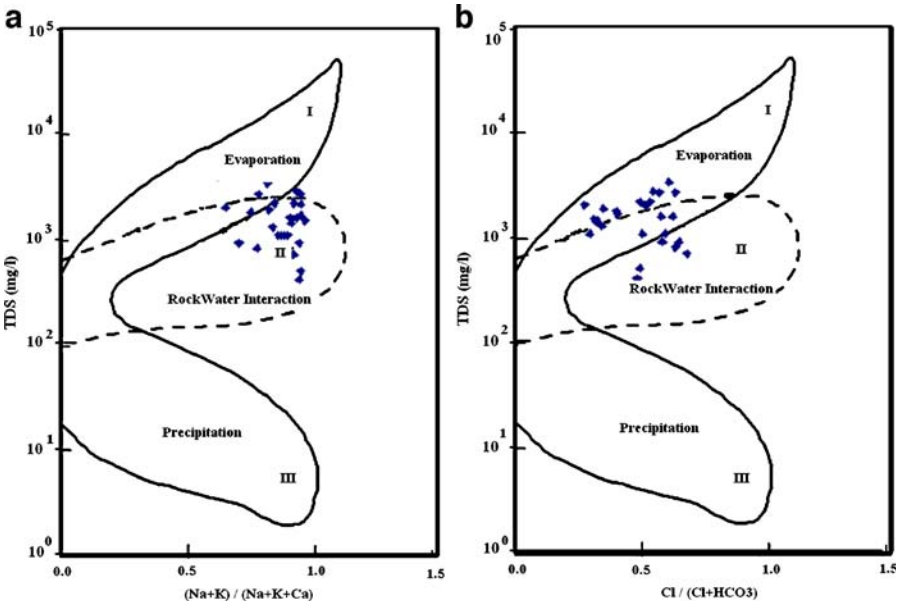


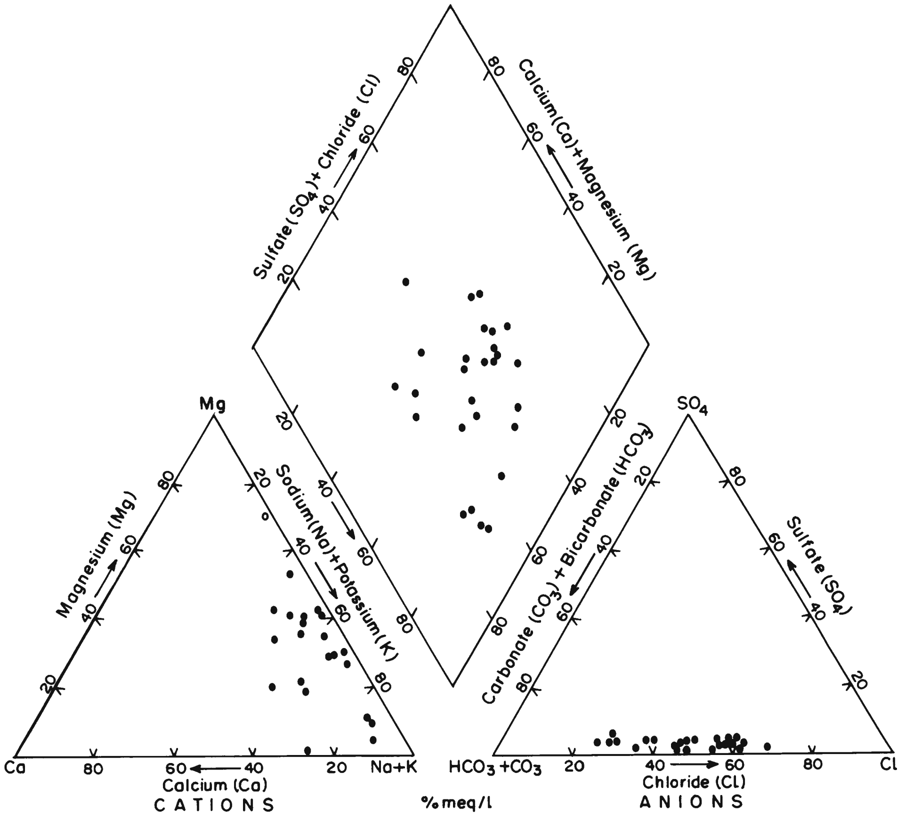
Fig. 4 Factor analysis results (R-mode) for primary associated variables of groundwater

Fig. 5 Gibbs diagram for controlling factor of groundwater quality



permissible concentration for nitrate as 45 mg/l. The concentration of nitrite ranges from 1.5 to 1717.9 mg/l, with a mean value of 465.51 mg/l. Nitrite was detected in a few groundwater samples. The consumption of water with high nitrate concentration causes blue babies or methe-

Fig. 6 Piper trilinear diagram for hydrogeochemical facies of study area groundwater



moglobinemia disease in infants, gastric carcinomas, abnormal pain, central nervous system birth defects and diabetes (Saba et al. 2006). Nitrogen compounds are present in groundwater in the form of nitrate (NO_3) and nitrite (NO_2) ions. Nitrite is more toxic to animal and human health than nitrate. Nitrates are extremely soluble in water and can move easily through soil into the drinking water supply (Saba et al. 2006). The fertilizers and domestic wastes are main sources of nitrogen-containing compounds and they are converted to nitrates in the soil.

Correlation matrix and factor analysis

The chemical composition of the groundwater is characterized by major cations and anions such as Ca, Mg, Na, K, Cl, SO_4, CO_3, HCO_3, NO_3, NO_2, and PO_4. The correlation matrix of physicochemical parameters was tabulated in Table 2. Weathering processes and anthropogenic inputs are the two main contributors for changing the geochemical composition of the groundwater (Chan 2001). R-mode factor analysis for extraction was carried out using SPSS software, and all the factors were plotted (Table 3 and Fig. 4). The extracted factor results were applied to calculate the primary associated factors. Table 3 shows that the variables HCO_3, Cl, SO_4, NO_3, Ca, Na, K have high positive factor loadings in factor 1, whereas pH, CO_3, PO_4, NO_2, K have high positive factor loadings in factor 2. Moreover, pH, CO_3, HCO_3, Cl, NO_2, Mg, Na have high positive factor loadings in factor 3. Three factor variables with different factor loadings explain the reason for variation of geochemical composition of groundwater. CO_3, HCO_3, Na have good positive factor loadings and indicates that these ions are derived from the same source such as weathering, carbonate and gypsum dissolution processes. The positive loadings of pH values suggested that the major ion concentration is controlled by pH variations in the study area, whereas pH is a controlling factor rather than TDS. Cl, Ca, Mg, K have moderate positive loadings, which indicates that they may be derived from rock–water interaction processes. The NO_3, PO_4, NO_2 are significantly derived from anthropogenic-induced pollution sources rather than natural processes.

Groundwater quality classification

Gibbs and piper diagram

The groundwater quality for drinking and irrigation purposes was assessed based on WHO (1971), USEPA (1992), and ISI (1983) standards. The quality of groundwater is significantly changed by the influence of weathering and anthropogenic inputs. The Gibbs diagram is widely used to establish the relationship of water composition and aquifer lithological characteristics (Gibbs 1970, Eq. 3). Three distinct fields such as precipitation dominance, evaporation dominance and rock–water interaction dominance areas are shown in the Gibbs diagram (Fig. 5). The predominant samples fall in the rock–water interaction dominance and evaporation dominance field of the Gibbs diagram. The rock–water interaction dominance field indicates the interaction between rock chemistry and the chemistry of the percolation waters under the subsurface.

$$\text{Gibbs ratio I (for anion)} = \text{Cl}^- \big/ \left(\text{Cl}^- + \text{HCO}_3^-\right)$$
$$\text{Gibbs ratio II (for cation)} = \text{Na}^+ + \text{K}^{2+} \big/ \left(\text{Na}^+ + \text{K}^{2+} + \text{Ca}^{2+}\right) \tag{3}$$

Table 4 Water quality classification based on SAR, RSC and Na% values of the study area

SAR (sodium absorption ratio)	Water class	Sample location numbers
<10	Excellent	All samples
RSC(Residual carbonate Content)		
<<1.25	Excellent	5, 9, 12, 13, 20
<1.25	Good	7
1.25–2.5	Fair	2, 3, 4, 16, 17, 18, 19, 23, 24
>2.5	Poor	1, 6, 8, 10, 11, 14, 15, 21, 22, 25, 26
Based on Na %		
20	Excellent	4, 5, 9, 20
21–40	Good	1, 2, 7, 13, 14, 17, 18, 19, 23, 26
41–55	Poor	3, 21, 24
50–70	Very poor	6, 8, 10, 11, 12, 15, 16, 22, 25

whereas all the ionic concentration is expressed in meq/l.

Piper trilinear diagram (Piper 1944; Fig. 6) shows that Ca–HCO_3, mixed $CaNaHCO_3$ and NaCl type domination hydrogeochemical facies. It is also suggested that silicate weathering domination and rock–water interaction are the primary factors in increasing the major ion concentration in the groundwater. Ca–HCO_3 and $CaNaHCO_3$ combination fields in the piper diagram are indicative of anthropogenic influences and irrigation return flow (Jeevanandam et al. 2006).

Residual sodium carbonate

The residual sodium carbonate (RSC) was calculated to determine the hazard effects of carbonate and bicarbonate on the quality of groundwater for agricultural and irrigation purposes (Eaton 1950, Eq. 4). The calculated RSC value was compared with the quality classification of groundwater in Table 4. The calculated values reveal that all the sampling sites are good for irrigation purpose except few. The increase of RSC in groundwater is significantly harmful for plant growth.

$$RSC= \left(CO^{3-}+HCO3^{-}\right) - \left(Ca^{2+}+Mg^{2+}\right) \tag{4}$$

Sodium absorption ratio and sodium percentage

The groundwater quality was classified and compared with sodium absorption ratio (SAR) and sodium percentage (Na %). Sodium concentration is important in classifying the water for irrigation purposes because sodium concentration can reduce the soil permeability and soil structure (Todd 1980; Domenico and Schwartz 1990). The calculated SAR values show that the groundwater samples are excellent for irrigation purposes (Table 4, Eq. 5). Sodium hazards are very low,

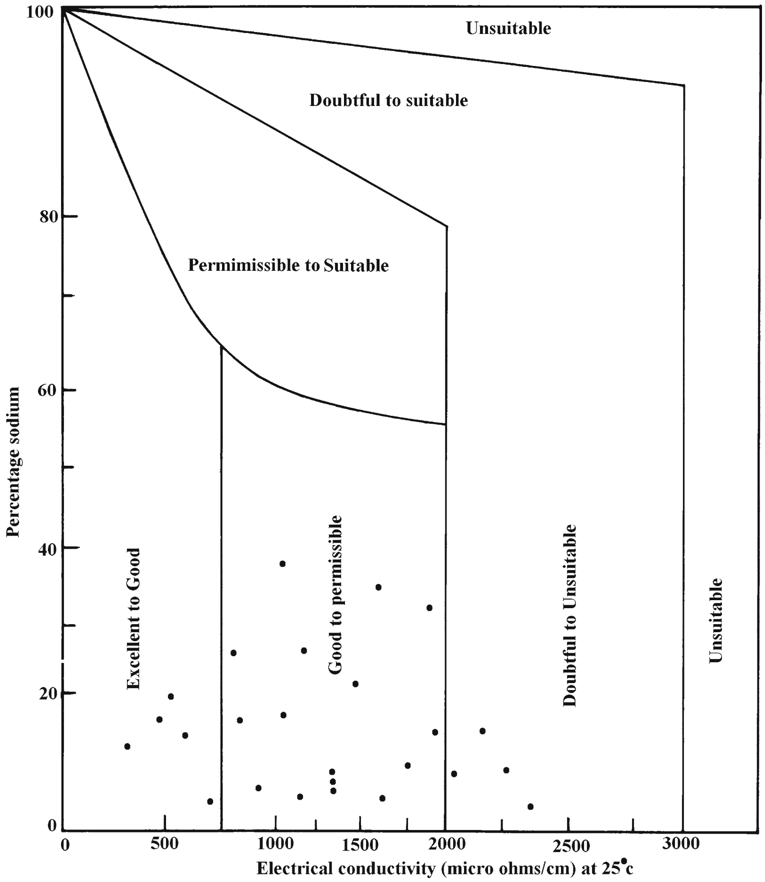


Fig. 7 Sodium percentage Vs EC values plot for water quality classification (Wilcox diagram 1955)

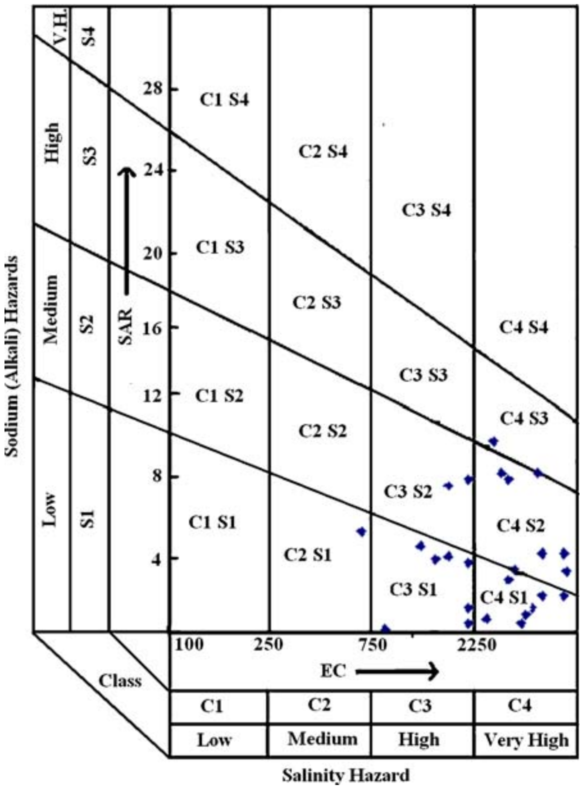


Fig. 8 USSL diagram for irrigation water quality classification (USSL Diagram 1954)

and the groundwater can be used on most crops for irrigation purposes. Generally high concentrations of bicarbonate and carbonate are predominant anion in the alkali soils, and chloride and sulfate are the predominant anion in the saline soils. Based on sodium percentage, the prominent groundwater samples are suitable for irrigation except a few samples (Table 4).

$$\mathrm{SAR}=\left(\mathrm{Na}^{+}\Big/\left(\sqrt{\mathrm{Ca}^{2+}+\mathrm{Mg}^{2+}/2}\right)\right) \qquad (5)$$

WILCOX and USSL classification

According to Wilcox (1955), the water quality diagram for irrigation and domestic purposes shows that 17 samples are falling under good to permissible, four samples are doubtful to unsuitable, and five samples are excellent to good (Fig. 7). The SAR *vs* EC values for groundwater samples of the study area were plotted in the USSL graphical diagram of irrigation water (Fig. 8). Based on USSL diagram (US Salinity Laboratory 1954), the water quality shows that majority of the samples fall in the C4-S1 (very high salinity with low sodium), C3-S1 (high salinity with low sodium) categories, a few samples fall in the C4-S2 (very high salinity with medium sodium) and C3-S2 (high salinity with medium sodium) categories. These groundwater samples show high to very high salinity hazard with low to medium alkali hazards. Based on the USSL diagram, the groundwater samples are satisfactory for irrigation use in almost all soil types.

Conclusion

The groundwater samples are dominated by Na, K, Cl, and HCO_3 ions. pH values reveal that the groundwater is slightly basic in nature. The excess amount of total dissolved solids in the groundwater is due to anthropogenic factors and geological characteristics of the aquifer. Based on the Gibbs diagram, rock–water interaction and evaporation are two main responsible processes for changing the chemical composition of groundwater. The carbonate and bicarbonates are mainly derived from carbonate mineral and silicate weathering. Chloride and sulfate concentration in groundwater is due to weathering, dissolution of rock formations, and irrigation drainage return flow. Na, K, Ca, Mg were derived from leaching and weathering of rock formations, anthropogenic activities. NO_3, NO_2, PO_4 are mainly derived from various agricultural and anthropogenic activities. The weathering process of rock formations, water dissolved natural organic, inorganic compounds, high consumption of fertilizer chemicals and dramatic development of urbanization are main factors for groundwater quality degradation. The pretreatment process on sewage before draining in to adjoining rivers, irrigation channels and the reduction of anthropogenic pollution stress on groundwater aquifer are good solutions to preserve and improve the groundwater quality.

Acknowledgements KK is thankful to Prof. S.P Mohan, Professor and Head, Department of Geology, University of Madras Chennai for his constant encouragement and providing laboratory facilities to carryout this work.

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