Identifying major factors controlling groundwater chemistry in predominantly agricultural area of Kattumannarkoil taluk, India, using the hydrochemical processes and GIS

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ABSTRACT

Hydrochemical investigation was carried out in predominantly agricultural area of Cuddalore district, South East India. The objective was to determine the potential factors controlling groundwater chemistry and their seasonal variation. Fifty samples obtained from 37 agricultural boreholes and 13 surface waters were used for analyses of 13 physico-chemical parameters. Qualitative and quantitative methods such as titration methods, flame photometry and Ultra-violet visible spectrometry were used to define the major ions; Na⁺, Ca²⁺, Mg²⁺, Cl⁻, K⁺, SO₄²⁻, PO₄³⁻, Si(OH)₄, and NO₃⁻. The results further were subjected to geographic information system (GIS), statistical analysis and geochemical plots. The evaluation of factors controlling water chemistry was accomplished by interpreting Gibb’s diagram, Piper diagram, Index of Base Exchange (IBE) and factor analysis. The interpretation proved that salts percolation, agriculture, weathering and dissolution as predominant factors controlling water quality of the study area. A Piper diagram classified the study site’s water types into Ca-HCO₃ water type, Ca-Cl water type and mixed Ca-Mg-Cl water type indicating freshwater recharge, mineral dissolution and reverse ion exchange, respectively. Factor analysis extracted four significant factors, namely leaching of secondary salts, weathering, and mineral dissolution as well as anthropogenic factors, that cumulatively responsible for 80.34% of the total variance of the taluk’s groundwater chemistry.

Introduction

Agriculture is a central sector in the economic growth of India, as it is the resource of victuals for the preponderance of inhabitants and contributes 18% of the gross national product (GDP) (Sushruth, 2018). More than 90% of rural and almost 30% of urban residents depend on groundwater for their drinking, domestic and irrigation uses (Srinivas, Aghil, Hudson Oliver, Nithya Nair, & Chandrasekar, 2017). Whereabouts 50–80% of the irrigated land use groundwater and put up near 38% of the world-wide food production (Selvakumar, Ramkumar, Chandrasekar, Magesh, & Kaliraj, 2017). However, utilization of excessive chemical products in agriculture has shown the negative effect on groundwater chemistry (Liu et al., 2015).

Generally, groundwater chemistry depends on the quality of recharge water and on geological as well as geographical placement in the area (Chandrasekar et al., 2014). Several factors such precipitation, groundwater flow patterns, rainfall patterns, infiltration rate, degree of chemical weathering of diverse rock types and inputs from sources other than water-rock interaction affect groundwater chemistry as well (Okiongbo & Douglas, 2015). Groundwater chemistry can also be influenced by another features, but also reflects inputs from the atmosphere, soil and also pollutant sources i.e.: saline intrusion, mining and excessive irrigation activities (Srinivas et al., 2017). The chemistry of groundwater is altered by hydrochemical processes through aquifer varying over time and space (Brindha, Neena Vaman, Srinivasan, Sathis Babu, & Elango, 2014).

As groundwater moves from recharge to discharge area there are significant temporal and spatial changes in the groundwater chemistry reflecting several hydrochemical processes in the aquifer systems. There are several important hydrochemical process that control its chemistry such evaporation, weathering, precipitation, mixing, ion exchange, redox and dissolution (Krishna Kumar et al., 2016). The comprehension of hydrochemical processes with GIS is very useful for characterization of groundwater chemistry, which determines utility purposes such as domestic, agricultural and industrial (Gnanachandrasamy et al., 2015).

Geographical information system (GIS) is used by geoscientists to interpret the hydrochemical process occurring within groundwater host rock, hydrogeological unit and the aquifers. Combination of GIS and multivariate statistical approaches have been used in groundwater study to identify factor controlling the
groundwater chemistry such as rock weathering, evaporation, precipitation and ion exchange (Singaraja, Chidambaram, Prasanna, Thivya, & Thilagavathi, 2014). GIS helps to make spatial distribution maps of different hydrochemical parameters and relate to the geology of the sampling locations (Remy, Mukesh, & Chidambaram, 2016).

The vagaries in monsoon and insufficient surface water flow conditions have led to increased agricultural and domestic extraction. The demand for large-scale supplies of freshwater from various competing end users has increased. In this scenario, 37 groundwater samples and 13 surface water samples have been collected from predominantly agricultural area, Kattumannarkoil taluk of Cuddalore district, South East India to understand the geochemical variation of water quality. This research was also focused on three main objectives: (1) understanding hydrochemical aspects of groundwater; (2) assessing the quality aspects of groundwater for domestic and irrigation purposes using a computer program WATCLAST written in C++; (3) to identifying the possible factors controlling groundwater chemistry using geochemical plot and multivariate statistical analysis.

**Depiction of study region**

**Location and climate**

Our study is Kattumannarkoil, a panchayat village and part of Cuddalore district, Tamil Nadu, India. It is located within southeast of India between 11°30' to 11°10' North latitude and 79°20' to 79°40' East longitude with a total area of 449.61 km² (Figure 1). It forms parts of Geological survey of India topographical maps Nos. S8M/7, M/8, 11 and M/12. It is characterized by sedimentary high grounds, elevation greater than 80 m of Cuddalore sandstone of Tertiary age (Central Ground Water Board 2009). Like most of coastal area of Tamil Nadu, the areas adjoining Veeranam Lake has a subtropical climate receiving most of the rainfall during the North-East monsoons from October to December. The average annual rainfall and temperature is 1273 mm and 38°C. Kattumannarkoil is largely devoted to agriculture land (Figure 2) and land use area which is surrounded by Veeranam Lake and Coleroon River. The major crops are paddy, sugarcane, cotton, groundnut, sunflower, banana and ginger. Canal irrigation, well irrigation and lake irrigation are under practice.

**Geology, geomorphology, and hydrogeology**

Geologically, the study area consists of four types of formations such as Coarse Sandstone, Clay Black Clay, and Clayey Sandstone (Figure 3). Therefore, it is one of the sedimentary terrains, which is mainly covered by Clay and Clayey Sandstone on its major part, whereas its small portion is covered by Black Clay. The soils of Cuddalore district are classified as black, red, ferruginous, and arenaceous. They are again subdivided into Clay, Loam, and Sand. Three major geomorphologic units are generally observed, namely (1) moderately buried pediplain, (2) shallow flood plains, and (3) shallow buried Pediplain. Groundwater occurs...
in these formations both under water table as well as under confined conditions and is developed through well, tube, and bore wells.

Materials and methods

Water sampling and physico-chemical analysis

Samples were picked up from surface water and groundwater; bore holes and water hand pumps between April and June 2015 (pre-monsoon season), to detect the factors influencing the chemistry of well water in predominantly agricultural areas of Kattumannarkoil taluk, Cuddalore district, Tamil Nadu, India. All samples were picked up and stored in clean polyethylene bottles as per sampling procedures. The digital apparatuses were utilized to record pH, total dissolved solids and conductivity immediately after sampling onsite. The latitude and longitude of sampling locations were identified by Global Positioning System (GPS, GARMAN 76CSx). Samples were filtered with 0.45 µm filter paper and kept at 4°C in plastic bottles (1000 ml) and conserved for later chemical analyses according to APHA (1998).

The concentrations of ions such as Ca$^{2+}$, Mg$^{2+}$, Na$^+$, and K$^+$ were determined by Titrimetry and Flame photometry (Figure 4), respectively. Concentrations of major anions (Cl$^-$, HCO$_3^-$, SO$_4^{2-}$, PO$_4^{3-}$, and H$_2$SiO$_4^-$) were determined by Titrimetry and UV/visible spectrophotometer respectively. The physico-chemical results were further processed by WATCLAST (Chidambaram, Ramanathan, Srinivasamoorthy, & Anandhan, 2003). The physico-chemical parameters of the analytical results of groundwater were compared with standard guideline values recommended by the WHO (2011)and BIS (2003). Sodium absorption ratio (SAR), sodium
percentage (Na %), residual sodium carbonate (RSC) were used to evaluate the suitability of water for irrigation purposes.

The Piper plot and Gibbs diagram illustrating the hydrochemical facies were determined using the computer programme AquaChem 4.0 to unravel the leading process overriding the groundwater chemistry. The trilinear (Piper, 1944) diagram is composed of triangular field and diamond shaped field. One triangular field consists of (Na\(^+\)) + (K\(^+\)), (Ca\(^{2+}\)) and (Mg\(^{2+}\)). The other triangular field characterizes (Cl\(^-\)),(SO\(_4^{2-}\)) and (CO\(_3^{2-}\)) + (HCO\(_3^{-}\)). These two fields are plotted separately with the value of EPM; (Na+ K) alkali and (Ca+Mg) alkaline earth. Whereas another triangle with weak acid (HCO\(_3^{-}\)+CO\(_3^{2-}\)); SO\(_4^{2-}\) and Cl strong acid. Overall, the properties of water in diamond field are represented by the triangular field’s position (Karanth, 1991).

Multivariate statistical techniques integrated with Geomatics (GIS) have been successfully adopted by many researchers to identify the factors controlling groundwater chemistry in the region of Tamil Nadu, India (Singaraja et al., 2014; Thilagavathi et al., 2017; Venkatramanan, Chung, Ramkumar, Rajesh, & Gnanachandrasamy, 2016). Factor analysis (FA) was implemented here as statistical method for identifying factors governing the chemistry of groundwater in the research site. The contribution of a factor is significant when the Eigen-value is greater than 1\(^{1}\). The factors that are significant with Eigen values (>1) and explaining for higher percentage of the variability of the data were noted. The factor loadings were sorted according to the criteria of Liu et al. (2015), i.e strong, moderate and weak, corresponding to absolute loading values of >0.75, 0.75–0.50, and 0.50–0.30, respectively. They
suggest the dominance of the corresponding variables which, when projected onto the data, give factor scores. The contour map of factor score was produced using Inverse Distance Weighted (IDW) raster interpolation and spatial analysis techniques built within ArcGIS® 9.3 software. The ionic balance errors (varies between 5 and 10%) were calculated to verify analytical precision for the measurements of anions and cations (Freeze & Cherry, 1979).

Results

Water chemistry

The summary statistics of the analytical results of the physicochemical parameters of groundwater and surface water samples collected from different sampling sites of predominantly agricultural area in the South East India is presented in Table 1. Comparative analyses of groundwater samples of the study area exceeding the desirable limits described by BIS (2003) for drinking purposes are furnished in Table 2. Hardness of water refers to soap neutralizing power. It also refers to reaction with soap and scale formation. It increases the boiling point of water. Table 3 shows the results based on USGS (United States Geological Survey) Hardness classification.

Classification for irrigation purpose

According to Wilcox (1955) and Richards (1954); EC, SAR, and Na% are major factors in classifying irrigation waters. Table 4 shows irrigational quality parameters results in the study area. The United States Salinity Laboratory Classification (USSL) Figure 5 is an informative diagram to finding out the groundwater samples suitability for irrigation in the study area. Figure 6 shows the results based on Doneen’s chart to find out suitability of groundwater for irrigation purpose. Four water types are clearly distinguishable to Doneen’s chart: Class I waters are the “Excellent” Types suitable for irrigation and are characterized by Low PIs; Class II waters are generally “Good” which is acceptable type Class III waters are ‘Fair “and Class IV if Poor which is Not Suitable” for irrigation.
Mechanism controlling groundwater chemistry

Groundwater chemical composition plays a vital role in setting the suitableness of water for different purposes.

Chemistry of groundwater alters from the point of its entry to exit from the aquifer, due to various parameters such as soil textures, structures, weathering, mining, contamination and other environmental factors. For a proper interpretation, it is fundamental to note that all changes in chemistry of aquifer systems. To find out the mechanisms controlling groundwater chemistry in the study site Piper (1944) and Gibbs boomerang diagram (Figures 7 and 8(a,b)) has been used.

Index of base exchange (IBE)

Schoeller (1967) described reactions occurring in groundwater using a measure called “Index of Base Exchange” (IBE).

Factor analysis

Factor analysis is a statistical method applied to investigate the general relationship between physico-chemical variables by indicating multivariate patterns for classification of the original data. Since groundwater chemistry is described through major inorganic compounds Ca, Cl, K, Mg, Na, HCO₃, NO₃, PO₄, and SO₄ is resulted from the natural or chemical weathering, human-induced and climatic inputs. For each compound, the concentration is carved up in two partial contributions, one associated to weathering reactions, and other connected to pollution. Therefore, factor was used to distinguish the partial contributions. Numerous variables were used in this present research for factor analysis such as EC, Ca, Cl, K, Mg, Na, HCO₃, NO₃, PO₄, SO₄ and H₄SiO₄. The factor analysis, loadings, eigen values and total data variability for groundwater is given in Table 6.

Factor scores

The factor scores were used to define the spatial illustration of each factor variation, and to find the zone of its representation in the study site. Two approaches such as regression and weighted least square technique
were applied in this current research. The positive zone of each factor shows the domination of hydrochemical process. Spatial variation of the first four factors representation of ground water samples are explained by their scores. Spatial representation of four factors of groundwater samples are represented by their scores Figure 9 (a, b, c & d).

Discussions

Classification for domestic purpose

The findings indicate that groundwater is mainly of alkaline nature (pH>7). The pH varies between 6.95 and 8.44, an average of 7.7, which clearly showed the alkali nature in majority of the groundwater samples (Table 1). Table 2 demonstrates the analytical results of collected samples in comparison with standard guidelines parameters of (BIS, 2003) and (WHO, 2011) for potable water quality. The groundwater of the study site was ascertained to be safe water for public use in 75–85% of area. The results classification based on USGS (United States Geological Survey) Hardness revealed that 8% of all groundwater samples fall under slightly hard category, whereas 41% falls under moderately hard category. The remaining 51% falls under very hard category. Similarly, 15% of samples from surface water indicate slightly hardness, while 85% falls under moderately and highly hardness. Therefore, groundwater can be considered suitable for domestic purpose. Whereas surface water is unfit for domestic use.

Classification for irrigation purpose

According to Wilcox (1955) in (Table 4), 67.57% of all sampled groundwater falls under permissible category, whereas 21.62% falls under good category. The remaining 10.81% falls in doubtful category due to husbandry activities in the region. However, 91% of collected samples from surface water are classified as good or excellent categories with minor (7.69%) falling in permissible. As a result groundwater and surface water in the study area are fit for irrigation purpose. Na% value indicates that bulk samples are included in excellent, good and permissible categories. While 15% of samples collected from surface water ranges doubtful zone due the farming activities in the region. Finally, wholly samples are classified as excellent and good categories, according to Richards (1954) (Table 4).

SAR and salinity of groundwater always determine its suitableness for agrarian usage (Chidambaram, 2010). The salinity of groundwater is usually originated from weathering of rocks, percolating from top soil, human-induced activities and climate alteration. For irrigation purpose, the high concentration level of HCO₃ and Na in groundwater can affects soil permeability and drainage of the agricultural area. Groundwater included in very high-salinity category (C4) is not recommended for irrigation use, but it may be utilized for salinity-tolerant crops on permeable soil with special treatment. However, groundwater that is included in the C3S1 and C2S1 categories is considered to be of moderate quality to irrigate semi-tolerant crops. It may have a little danger or harmful effect to soil by exchanging Na to soil (Karanth, 1989). A high level of RSC in groundwater results in increasing sodium adsorption in soil (Eaton, 1954). In view of these definitions, the USSL (1954) diagram (Figure 5) point out that most of groundwater samples fall into the C2S1 and C3S1 categories. This indicates that groundwater has high to low sodium hazard and medium salinity respectively. Though, few samples classified as C4S1 category indicates that groundwater has low sodium hazard and high salinity. Therefore, groundwater can be considered for irrigation purposes.
According to Richards (1954) when RSC value is <1.25 mg/l for groundwater samples, it proves that the water is safe for irrigation usage (Table 4). Relative to soil development, permeability index is a valuable factor which influences quality of well water for irrigation use. Doneen (1964) has revealed the suitableness for irrigation water deeply depend on permeability index, by classifying groundwater as class I, II and III. In this research, all collected groundwater samples come under Class I and infer water is permissible for irrigation use (Figure 6).

Giving to Stuyfzand (1989) chloride classification groups, most of groundwater samples come under fresh category (18.92%) and fresh brackish to brackish-salt (81.08%) (Table 4). Similarly, the findings show that 23.08% of all gathered samples from surface water are classified under fresh category, while 76.92% are included in fresh brackish to brackish water category. Corrosivity Ratio groups groundwater samples into safe (94.59%) and (5.41%) unsafe categories. In this case, the unsafe category cannot be transported through metal pipes distribution system. While all sampled surface water falls into safe category.

**Mechanism controlling groundwater chemistry**

Piper diagram (Figure 7) reveal that the cation is well-ordered by Na>Ca>Mg and anions by Cl>HCO$_3$>SO$_4$.
Three major hydrochemical facies were identified: Ca-HCO$_3$ water type, Mixed Ca-Mg-Cl water type and Ca-Cl water type. Ca-HCO$_3$ and Ca-Cl water types suggest mineral dissolution, an interaction between rock-water and recharge of freshwater (Mondal & Singh, 2012). Mixed Ca-Mg-Cl water type indicates, mixing of high salinity water caused from surface contamination sources, such as irrigation return flow with existing water followed ion exchange process (Ayuba, Tijani, & Omonona, 2017; Jeyaraj, Ramakrishan, Jai Anandhi, Arunchalam, & Magudeswaran, 2016; Ravikumar & Somashekar, 2017). While, Gibbs boomerang diagram (Figure 8(a,b)) designates that most of samples come under weathering dominant zone in all seasons. However, few groundwater samples are classified in boomerang zone, implying that secondary structure of saline precipitation and mining water control water chemistry.

**Index of base exchange (IBE)**

From the analysis, 5.41% of sampled groundwater, and 23.08% of all gathered surface water samples show negative index. The remaining 94.59% of all groundwater samples and 76.92% of gathered surface water samples show positive index. This finding reveals that the primary sources of dissolved salts are prevailed by the secondary percolating followed by ion exchange (Table 5). All groundwater and surface water samples come under type I signifying that the chemistry of water is largely controlled by residence time and extent of rock-sediment-water interactions.

**Factor analysis**

Four types of factors governing the chemistry of groundwater in study area were identified. For factor loadings, a strong loading was defined as greater than 0.75, a moderate (0.75–0.50) and (0.50–0.30) weak for interpretation. The proper values (Eigen values) among these four factors were shown in cumulative variance of 80.34% (Table 6).

**Factor I**

Factor I represents a total variance of 41.96% and is described by moderate to strong loadings. The associations of ions in factor I representing Ca, Mg, Na, K, Cl, PO$_4$, SO$_4$, EC, and TDS. Ca, Mg, Cl, EC, and TDS have
a strong loading while PO₄, SO₄, Na, and K have moderate loadings. This factor reflects the secondary salts leaching (Table 6). For this factor, Cl⁻ is the dominant anion. The higher value of Cl⁻ is observed in agricultural area adjoining Veeranam Lake along South Eastern part of the study area. Strong loading of Cl⁻, Ca, and Mg indicates the excessive interaction of groundwater with aquifer (Thilagavathi et al., 2016). Therefore, the representation of numerous ions in this factor also shows the mixing of diverse hydrogeochemical processes.

**Factor II**

Factor II, which explains a total variance of 17.54% with positive association Na, HCO₃, NO₃, and SO₄²⁻. This factor is associated with positive association of Na, HCO₃, NO₃, and SO₄, which is mainly due to weathering. Positive loading of NO₃ and HCO₃ may be resulted from applying the fertilizers in the agricultural fields. The source of SO₄ in groundwater might be derived to break down of organic substances from fertilizers (Thivya et al., 2014) Spatially, the higher concentration of factor 2 is observed at the Northern part of the study area where agriculture practice is dominant. Hence, the main source behind this is agricultural activities present in the study area.

**Factor III**

Factor III, relates to a total variance of 10.44% with strong loading of pH resulting from pH dissolution. Strong loading of pH shows groundwater is mostly polluted by discharge of waste water as regular source in the study site. This factor is named as degradation factor due to the organic material degradation by increased pH (Jacintha, Rawat, Mishra, & Singh, 2017). According to Rajesh, Elango, and Brindha (2019) strong loading indicates the major ions concentration are controlled by pH.

**Factor IV**

Factor IV, represents the total variance of 10.41% of the total variance with strong loading of H₂SiO₄, which is implying dissolution of silica. This factor reflects weathering of silica rock forming minerals i.e SiO₂ + 2 H₂O = H₄SiO₄ quartz (crystalline) and chalcedony (amorphous) (Ramachandran et al., 2012). Therefore, various processes such as; secondary salts leaching, weathering, and anthropogenic effects and mineral dissolution are believed to control the geochemistry of well water in the study site. Four factors are represented and reflecting the hydrogeochemical complexness during sampling in the study area. Factor analysis rendered four factors explaining 80.341% of total variance. This shows that are numerous factors with lesser eigen values contributing to the hydrochemistry of groundwater in the study area.

**Factor scores**

The spatial representation for Factor 1 (Figure 9(a)) fall between the Northern part of study area near and South eastern part of study area near the Coleroon River along the direction of groundwater flow. This arises as a consequence of secondary salt leaching from the surface water. The higher score is located in clay formation. Factor 2 (Figure 9(b)) is chiefly represented by predominance of weathering by the sewage water in the Northern part of the study area. This may be caused by utilization of fertilizers in the agricultural fields of study site (Thilagavathi et al., 2016). The spatial representation for Factor 3 (Figure 9(c)) falls between the North Eastern part of study area surrounding Veeranam Lake and Coleroon River. It is indicating the predominance ion exchange. Higher scores are located in clay and clay sandstone formation. Finally, the Factor 4 is represented (Figure 9(d)) along the Vellar area and upstream directions, with less impact of agriculture practice. The factor scores represent the main variations of chemical processes in the Northern, Eastern, Western, and South-Eastern of the study site. Factor scores show that secondary salt leaching, ion exchange and agriculture practices are major factors controlling groundwater chemistry in the study area.

**Conclusion**

This study was conducted to examine the factors controlling the groundwater chemistry of predominant agricultural areas of South India. The study reveals that groundwater in study site is alkaline in nature. Based on the Gibb’s boomerang diagram; groundwater chemistry of study site is governed by weathering dominant zone and secondary salt precipitation. The Index of Base
Exchange (IBE) classification shows that the chemistry of groundwater is largely controlled by water residence time and extent of rock-sediment water interactions. The results from factor analysis indicate that different processes, such as leaching of secondary salts, weathering, anthropogenic effects and mineral dissolution are the major processes controlling groundwater chemistry of the study site. Factor scores further explained the fact that natural and human-induce processes influence the groundwater chemistry in the study sites. It is concluded that the agricultural practices governed the major groundwater geochemistry in predominantly agricultural areas of the region. Further, the study reveals that there is a direct impact of surface water chemistry on groundwater. Since the study site is composed of porous and permeable sedimentary formation, there is a direct infiltration of the surface water to groundwater.

Acknowledgments

I am thankful to Professor and Head, for permitting to carry out the research project. I am very grateful also to my guide and research scholars for field and laboratory work.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

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