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Identification and evaluation of hydrogeochemical processes on river Cooum, South India

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Abstract The salient features of the river water chemistry and the seasonal variations on the individual chemical parameter were evaluated and characterized. The order of abundance of the ions in the water is determined for both seasons. The contribution of chemical weathering to the water chemistry has been determined using the $(Ca^{2+} + Mg^{2+})/(Na^{+} + K^{+})$ ratio, ternary, and Gibb's diagrams. The results show that the chemical composition of river water during premonsoon is controlled mainly by evaporation–crystallization, while in the postmonsoon, the rock–water interaction dominates. The unique characteristic of the river water is the linear relationship among the principal ions. Hydrochemical characteristics of ions in the water were studied using 1:1 equiline diagrams. The nature of the water samples was determined using the piper diagram. The influence of trace metals on the chemical composition and the quality of the river water in the study area has been assessed using Wilcox and US Salinity Laboratory diagrams.

Keywords River Cooum · Geochemistry · Seasonal variations · Water quality

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Introduction

Surface water studies have received wide attention due to the need to understand the quality and chemistry of water, which is a cumulative reflection of the geology of the catchment area, precipitation, weathering processes, and anthropogenic activities. Exploitation of surface water has increased greatly, particularly for agricultural purposes. Poor quality of water adversely affects plant growth and human health. Urban rivers have been associated with water quality problems and the practice of discharging untreated domestic and industrial waste into the water course has emerged (Hall 1984). High population density in Chennai increases the pressure on the drainage system and sewage treatment plants. The existing system does not cope with the heavy domestic discharge, and at many points; domestic effluents are directly directed into the river water, which leads to the present level of degradation of Cooum river.

The geochemical study of river basins reveals the nature of the geochemical factors, which helps us to understand the exogenic cycles of elements in the continent–river–ocean system. Many authors made detailed geochemical studies on some of the world's large- and medium-sized river systems, including the Amazon (Gibbs 1972; Stallard and Edmond 1983, 1987), the Mackenzie (Reeder et al. 1972), the Mekong (Carbonnel and Meybeck 1975), and the Changjiang and Huanghe (Hu et al.

1982; Zhang et al. 1990, 1995). In India, various authors studied the geochemistry of the river water to understand the weathering, hydrological, seasonal, and anthropogenic factors that influence water quality (Dey 1981; Tiwari and Dhar 1994; Sarin et al. 1989; Biksham and Subramanian 1988; Ramesh et al. 1995).

In previous studies, the assessment of the seasonal variation of river Cooum had not been carried out in detail. Hence, the river water samples were collected for premonsoon and postmonsoon periods in order to study the seasonal variation in the geochemistry of river water, and also to evaluate the interdependencies and interrelationships of various ions present in this water. Gibbs diagrams are used to determine the source of chemical budget of the ions in the river water. Moreover, the suitability of the river water for irrigation was studied using various tools, viz., Wilcox diagram, US Salinity Laboratory (USSL), sodium adsorption ratio (SAR), Kelly's ratio, etc.

From the point of view of water quality and pollution, critical evaluation of the above factors becomes pertinent, so that specific sources of pollution can be identified. In this work, in order to achieve this objective, the geochemistry of river Cooum is studied in detail. Such studies contribute to effective management and utilization of the surface water resources.

Study area

In this study, the water samples of river Cooum were collected at 30 stations in order to evaluate the nature and quality of water (Fig. 1). River Cooum originates from Kesevaram dam, Kesavaram Village, at about 48 km west of Chennai. Though river Cooum originates from this dam, the excess water from the Cooum tank (79.82° latitude and 13.02° longitude) joins this course at about 8 km, and this is considered as the head of the river Cooum. In the upper part of the

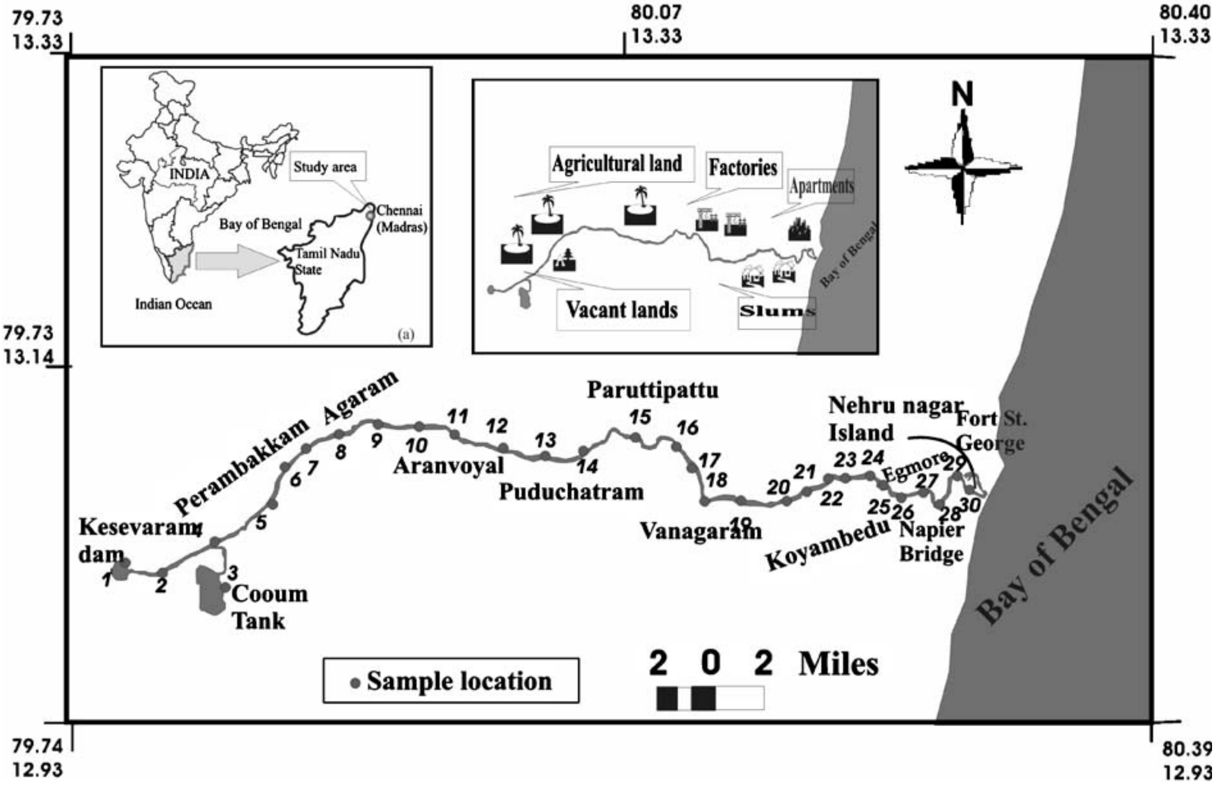


Fig. 1 Base map of Cooum river depicting sample locations and pollution sources

river stretch, it was found that many agricultural activities are being carried out. The river receives a sizeable quantity of sewage from its neighborhood after it reaches Vanagaram near Chennai. It flows through Kancheepuram, Tiruvallur, and Chennai districts for a distance of about 68 km, and after flowing through the heart of Chennai City, it enters into the Bay of Bengal. This river is almost stagnant and do not carry enough water except during the rainy season (NE monsoon). The period from October to December is referred to as Northeast Monsoon season over Tamil Nadu, and Chennai City receives the bulk of the rainfall from this monsoon.

It has been observed that, in the upper part of the river, there is no settlement along its bank, and hence, this part is not polluted by domestic effluents. However, due to intense agricultural activities, the chance of pollution due to fertilizers and pesticides is highly expected. In the middle and lower stretches of the river Cooum, the domestic sewage water was directed into the river, and it appears as a sewage water stream (Ramesh et al. 1995).

Geology of the river basin

The major part of the river basin of Cooum is covered by alluvium soil with tertiary and Gondwana rocks at depth. The alluvium soil varies from 10 to 20 m thickness and is mostly granular. In the downstream area of the river basin, coastal sand predominates, while the middle part and upper parts are dominated by sand and silt. Charnockites are also found in some part of the basins. The geological succession of the basin is given as follows:

Group	Age	Lithology
Quaternary	Subrecent	Soil, alluvium (sand and silt)
Tertiary	Pleistocene	Black clay
	Eocene to Pliocene	Sand stone and shale fossil ferrous
Mesozoic	Lower Cretaceous	Brown sand stone
	to Lower Jurassic	Silt, stone gray, and black shale
Azoic	Precambrian	Charnockite, granite, gneiss

Methodology

River water samples were collected during September 2005 and February 2006, representing pre- and postmonsoon seasons to evaluate the seasonal variations in chemical compositions. The samples were collected in new 1-l high-density polyethylene bottles prewashed with dilute hydrochloric acid and rinsed three to four times with the water sample before filling them to the required capacity. The samples were stored at a temperature below 4°C prior to analysis in the laboratory. For collection, preservation, and analysis of the samples, the standard methods (Rainwater and Thatcher 1960; Brown et al. 1970; AWWA 1971; APHA 1995) were followed. EC and pH of water samples were measured in the field immediately after the collection of the samples using pH and conductivity meters. Before each measurement, the pH meter was calibrated with reference buffer solution of $pH = 4$ and 7. Na^+ and K^+ were measured by using a flame photometer (model: Systronics Flame Photometer 128). Silica content was determined by molybdate blue method using an ultraviolet (UV)-visible spectrophotometer. Total dissolved solids (TDS) were measured by evaporation and calculation methods (Hem 1991). Ca^{2+} and Mg^{2+} were determined titrimetrically using standard EDTA. Chloride was estimated by $AgNO_3$ titration. Sulfate was analyzed using the turbidimetric method (Clesceri et al. 1998). Nitrate, nitrite, phosphate, and fluoride were analyzed using UV-visible spectrophotometer (Rowell 1994). Trace metals were determined by graphite furnace atomic absorption spectrophotometer (Perkin-Elmer AAnalyst 700). Multielement Perkin-Elmer standard solutions were used for the estimation of trace metals. The flow in river Cooum was found to be between 266.45 and 709.34 × 106 l/day (Gowri et al. 2008).

Results and discussion

Chemistry of water

The salient features of the hydrochemistry of the river water are as follows: During premonsoon, the pH of river water ranges from 6.4 to 6.9

with an average of 6.7. All the stations demonstrate slightly acidic behavior, but the values are within the World Health Organization (WHO)-allowed limit. However, during the postmonsoon, pH of water in all the stations was found to be alkaline, ranging from 7.3 to 8.9, with a mean of 8.2. Concentration of salinity (TDS) ranges from 588 to 3,274 mg/l, with a mean of 1,906 mg/l in premonsoon and 474 to 1,872 mg/l with a mean of 964 in postmonsoon. There is a considerable amount of dilution of concentration of ions during the postmonsoon due to heavy precipitation during this period. Higher concentration of TDS is observed in the middle and lower parts of the river. In the middle part of the area, both industrial and domestic effluents are directed into the river course, which would have increased the concentration of the ions in the water. In the lower part of the river, domestic effluents and the saline water intrusion influences the water chemistry of the river (Ramesh et al. 1995).

During premonsoon, the concentration of cations Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+} ions ranged from 24 to 265; 9.6 to 76.8; 79 to 1,069, and 5.5 to 71.8 mg/l with means of 152, 38.9, 377.1, and 31.3 mg/l, respectively. The ionic concentrations (based on millimoles per liter) are 16.22%, 6.41%, 73.37%, and 4.00%, and the order of abundance is $Na^{+} > Ca^{2+} > Mg^{2+} > K^{+}$. During postmonsoon, the concentration of cations Ca^{2+}, Mg^{2+}, Na^{+}, and K^{+} ions ranged from 56 to 159; 4.8 to 59; 65 to 356, and 2 to 33 mg/l, with means of 99.8, 25.5, 166, and 12.2 mg/l, respectively. The ionic concentrations (based on millimoles per liter) are 32.84%, 8.4%, 54.72%, and 4.0%. The order of abundance among the cations is $Na^{+} > Ca^{2+} > Mg^{2+} > K^{+}$. Similarly, in the case of anions during premonsoon, HCO_3^-, SO_4^{2-}, and Cl^- range from 85 to 640, 211 to 354, and 90 to 1,365 mg/l, with means of 321, 269, and 566 mg/l, respectively. The ionic concentrations (based on millimoles per liter) are 27.8%, 23.3%, and 48.9%. The order of abundance of anions is $Cl^- > HCO_3^- > SO_4^{2-}$. During postmonsoon, HCO_3^-, SO_4^{2-}, and Cl^- ranged from 87.6 to 388, 58 to 212, and 118 to 588 mg/l, with means of 204, 118, and 272 mg/l, respectively. The ionic concentrations (based on millimoles per liter) are 34.4%, 19.9%, and 45.8%. The order of abundance is $Cl^- > HCO_3^- > SO_4^{2-}$. It is observed that, even though there is a change in the concentration of ions, the order of abundance of ions during both seasons does not change.

Chemical weathering of the river basin

The ratio $(Ca^{2+} + Mg^{2+})/(Na^{+} + K^{+})$ has been used as an index to understand carbonate and silicate weathering in a river basin (Sarin and Krishnaswamy 1984). Most of the world's rivers and many Indian rivers have high ratios, suggesting the weathering of carbonate rocks in the catchment area (Subramanian 1979). The milliequivalent ratio of $(Ca^{2+} + Mg^{2+})/(Na^{+} + K^{+})$ for premonsoon is 0.84, and that of postmonsoon is 1.13, lower than the world's average of 2.2 and the Indian average of 2.5 (Singh and Hasnain 1999), suggesting that the contribution of weathering to the chemical composition of the river Cooum is not predominant. The higher ratio of postmonsoon in contrast with premonsoon reflects that the high precipitation during this period increases the weathering and dissolution of the rocks in the catchment area of the river basin. Gibb's diagram (Fig. 2) demonstrates that the chemical composition of the river water during premonsoon is controlled by evaporation–crystallization dominance. In the case of postmonsoon, rock–water interaction dominates the water chemistry, which

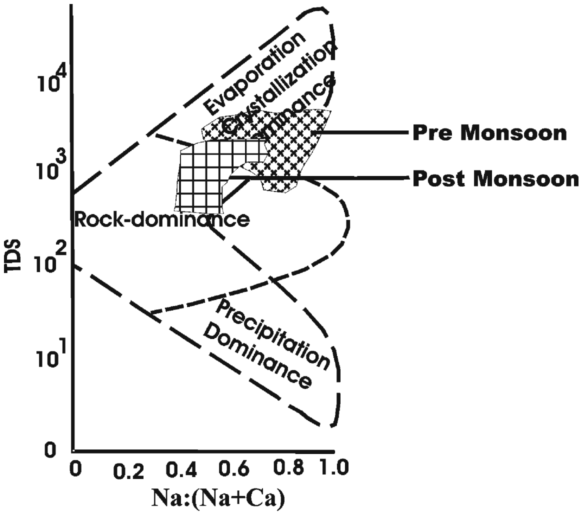


Fig. 2 Mechanism controlling groundwater quality—Gibb's diagram

supports the results of $(\mathrm{Ca}^{2+} + \mathrm{Mg}^{2+})/(\mathrm{Na}^{+} + \mathrm{K}^{+})$ ratio.

The ternary diagram plotted after Stallard and Edmond (1983) can also be used to characterize and evaluate the weathering reactions and the resultant chemical composition of the river water. The ternary diagram (Fig. 3) relating Na^{+} + K^{+}, Ca^{2+} + Mg^{2+}, and SiO_2 also reveals the lower contribution of silica in relation to other ions. Na^{+} predominates and the contribution of Ca^{2+} + Mg^{2+} is moderate. In the case of postmonsoon, the ternary diagrams show a slightly higher contribution of silica, and as far as other ions are concerned, it follows the same trend. The results suggest that the contribution of chemical weathering is only moderate and the chemical composition of the river water is highly influenced by anthropogenic activities.

Chemical characteristics of major ions

The bonding affinity of the alkali and alkaline earth metal ions towards the anions in the water is presented in the equiline diagrams (Fig. 4). In premonsoon, the plot of Na^{+} and Cl^{-} vs Cl^{-} shows that most of the values fall above and on the equiline, suggesting that alkalis are balanced by the chloride ions. The ratio between the Na^{+} + K^{+} and total cation is 0.56, and that of Cl^{-} and total anion is also found to be 0.55, indicating that most of the alkali ions are balanced by the chlorides. Among the alkalis, Na^{+} is dominant, and the concentration of potassium is apparently low. The concentration of K^{+} in global river is primarily from leaching of silicate minerals and in small amounts from other sources, such as evaporated minerals, fertilizers, rain water, and plant materials. Living plants, which acquire significant amounts of K^{+} primarily from weathered silicate materials, may also be considered as an intermediate storage of potassium. Part of this plant potassium is transferred to streams and rivers, as dead vegetation on the ground surface was leached by surface water. The study area does not contain potassium-bearing minerals, and the low contribution of K^{+} may also be due to the greater resistance of K^{+} to weathering and its fixation in the formation of clay minerals (Sarin et al. 1989). It has been observed that the Na^{+} + K^{+} and Cl^{-} values increase gradually from west to east in the direction of the river flow. The contribution of Na from domestic effluents occurs in the middle and

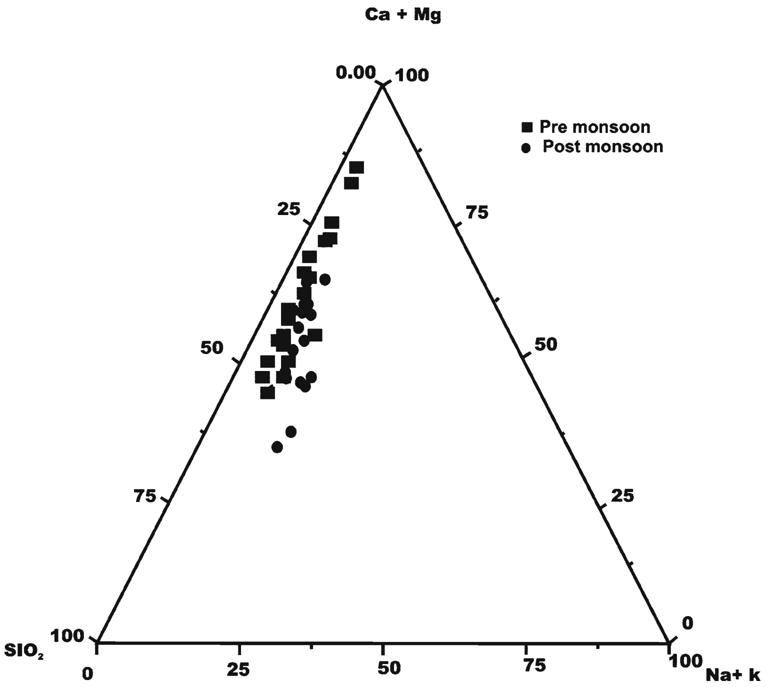


Fig. 3 Ternary diagram relating relative proportions of silica, alkali and alkaline earth metals, drawn after Stallard and Edmond (1983)

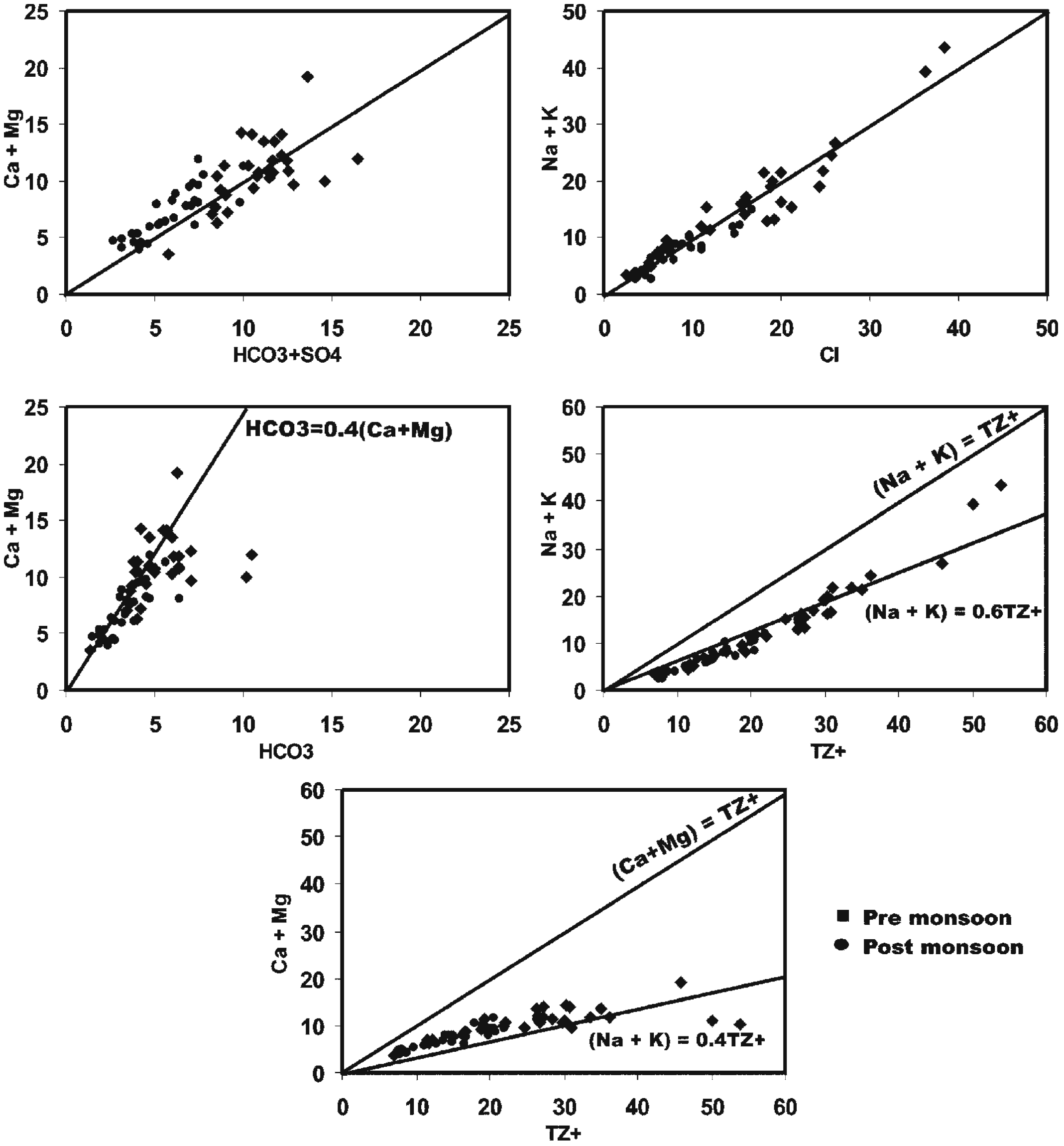


Fig. 4 Equiline diagrams

lower parts, and also from the saline water intrusion near the confluence point; the concentration of alkali and chloride ions increases towards the eastern side.

In postmonsoon, the plot of $Na^+ + K^+$ vs Cl^- shows that many points (about 60% of the values) fall just below the 1:1 lines, suggesting that there is an excess of Cl^- over $Na^+ + K^+$. However, in the plot of $Ca^{2+} + Mg^{2+}$ vs $HCO_3^{2-} + SO_4^{2-}$, a reverse case has been observed. In this plot, many points lie above the 1:1 equiline, suggesting that the excess chloride ions are balanced by the alkaline earth metal ions. The ratio of Na + K with TC is 0.48, and that of $Ca^{2+} + Mg^{2+}$ with TC is 0.52, which indicates that there is an inversion of ratio during postmonsoon clearly showing the dominance of alkaline earth metal ions over the alkalis. As in premonsoon, the concentration of sodium predominates over that of potassium. The ratio of Cl^- with TA is 0.55, indicating that there is no change with regard to this ratio. Cl^- ion concentration in the water bodies normally arises from three sources, viz., ancient seawater entrapped sediments, solution of halite and related minerals in evaporate deposits, and solution of dry fallout from the atmosphere, especially in the arid region (Walker and others 1991). The high concentration of chloride in the study area does not seem to arise from the above factors, but it may be caused by anthropogenic activities and due to the percolation of the saline water near estuary, where the stagnant backwater facilitates the seepage, thereby increasing the chloride content in the groundwater near the downstream.

Hydrochemical facies

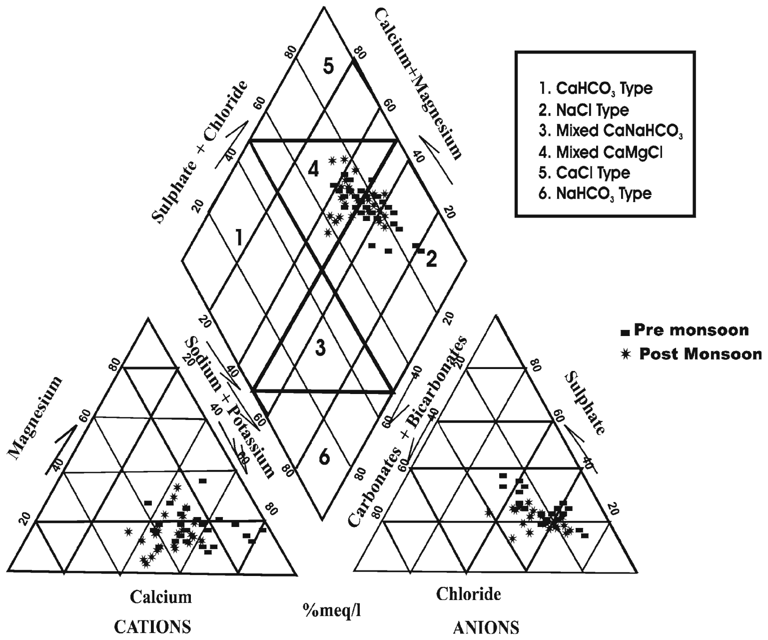
In the Piper diagram, major ions are plotted in the two base triangles as major cation (Ca^{2+}, Mg^{2+}, and Na^{+} + K^{+}) and major anion (Cl^{-}, SO$_4^{2-}$, and CO$_3^{2-}$ + HCO$_3^{2-}$) in milliequivalent percentages, and the overall information from the two triangles are combined together on a quadrilateral. The position of this plotting indicates the relative composition of river water in terms of the cation–anion pairs that correspond to four vertices of the field. The Piper diagram has been widely used to study the similarities and differences in the composition of waters and to classify them into certain chemical types. The plot (Fig. 5) shows that most of the river water samples analyzed during the premonsoon fall in the field of NaCl and mixed CaMgCl types. In the case of postmonsoon, a clear shift has been observed from NaCl to mixed CaMgCl type. From the plot, it is observed that alkalis (Na^{+} and K^{+}) exceed the alkaline earths (Ca^{2+} and Mg^{2+}) and Cl^{-} exceeds the other anions. The higher concentration of alkali ions present in the water may influence ion-exchange reactions between the solid-aquous phase, resulting in sodium-enriched sediments.

Trace metal chemistry

Water bodies get polluted with trace metals from a variety of sources, such as chemical weathering of rocks and soils, dead and decomposing vegetation and animal matter, wet and dry fallout of atmospheric particulate matter and humanity's activities including the discharge of various domestic and industrial effluents (Förstner and Wittmann 1983; Enguix González et al. 2000). Though trace metals, such as zinc, chromium, manganese, cadmium, cobalt, etc., play a biochemical role in the aquatic life, their excess presence is toxic and nonbiodegradable (Nurnberg 1982).

River Cooum demonstrates its nature due to the amalgamation of the factors such as return

Fig. 5 Piper diagram representing hydrochemical facies



flow of agricultural drainage water, industrial and domestic waste discharges directly directed into the river water, and decline of river velocity to almost stagnation in urban areas. The quality of water with regard to the concentration of trace metals (Cu, Co, Zn, Fe, Pb, and Cr) is assessed and the seasonal variation of the metals was evaluated in the river water samples (Fig. 6). The Cu values range from 0.046 to 0.099 mg/l, with an average value of 0.07 mg/l during premonsoon.

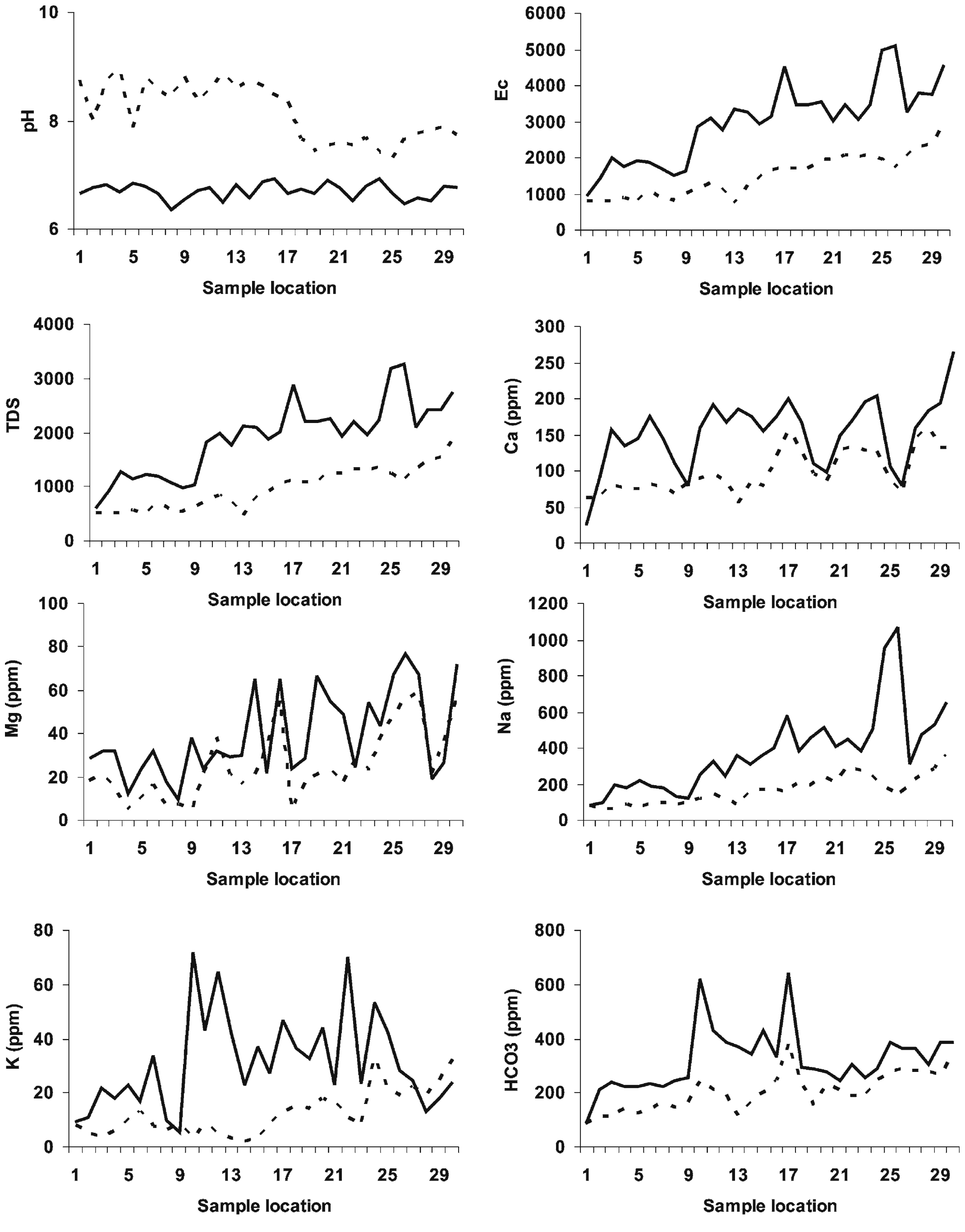


Fig. 6 Seasonal variation of the chemical parameters (**a**–**c**)

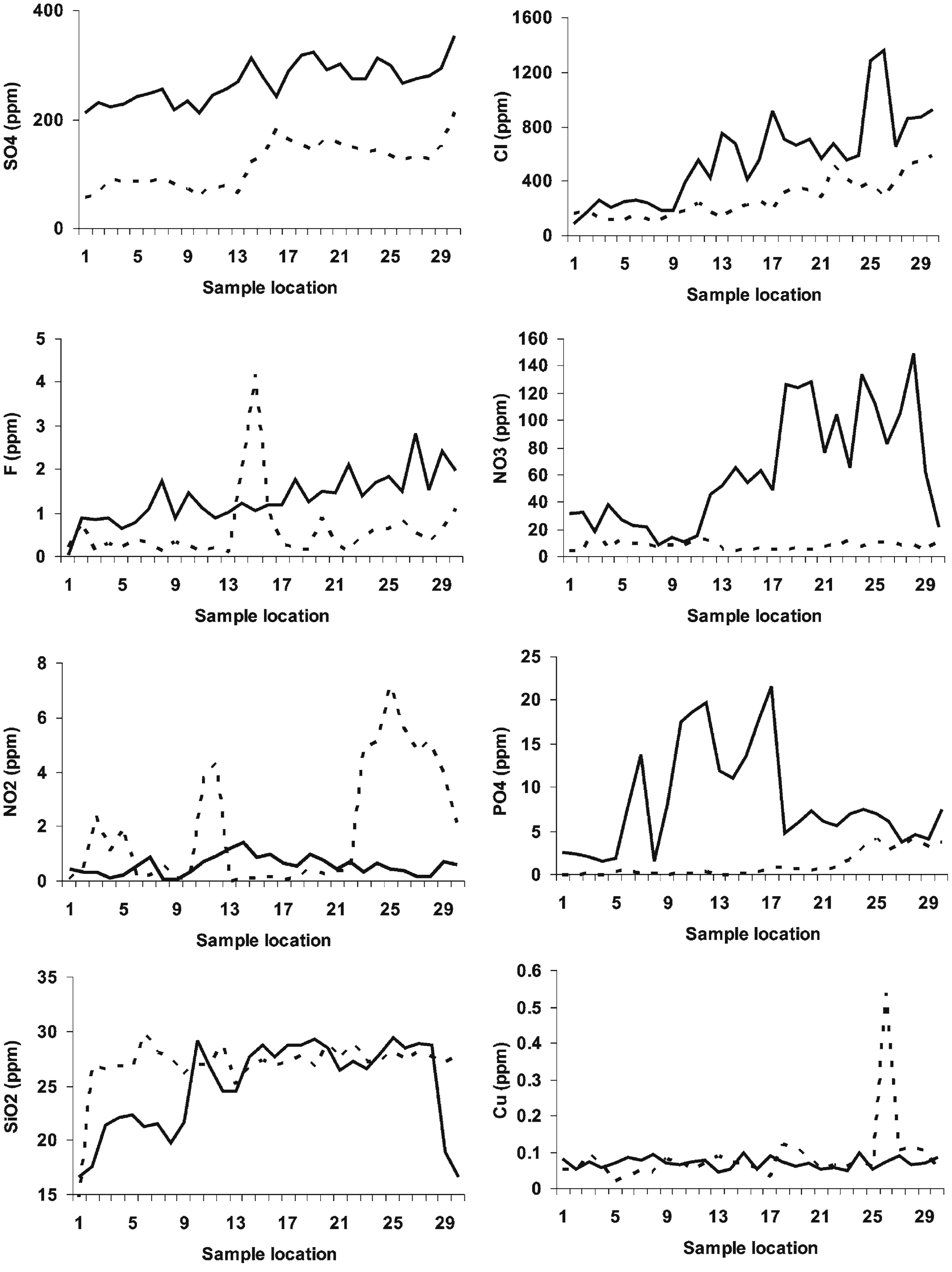


Fig. 6 (continued)

However, the postmonsoon values range from 0.02 to 0.54 mg/l, with a mean of 0.09 mg/l. Both in pre- and postmonsoon, the concentration of Cu is well within the WHO allowed limit of 2 mg/l. The range obtained was lower than the permissible value, and hence, adverse effects from domestic use are not expected as far as this metal is concerned. The concentration of cobalt during

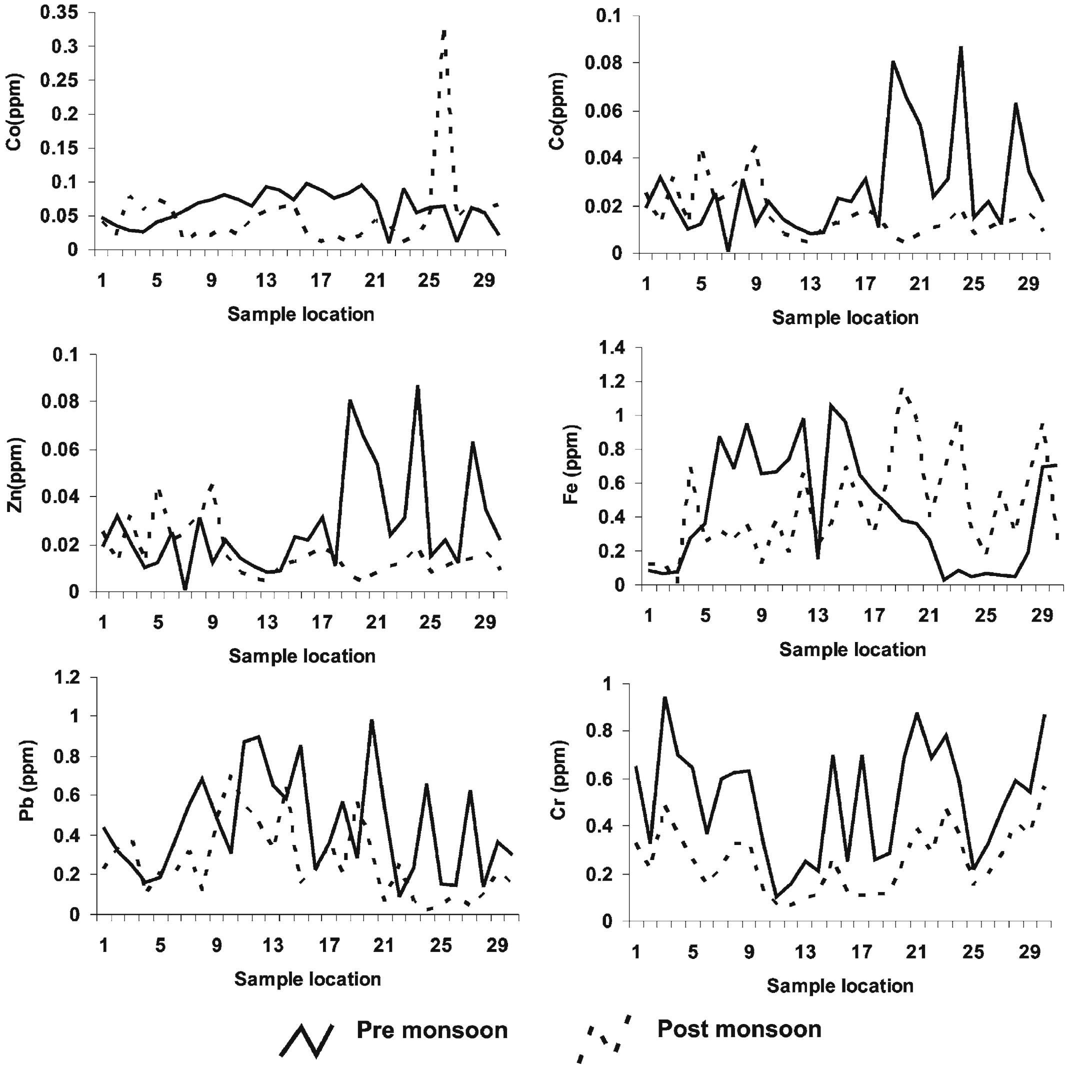


Fig. 6 (continued)

premonsoon ranges from 0.009 to 0.098 mg/l with a mean of 0.062, and the postmonsoon shows 0.01 to 0.33 mg/l with a mean of 0.050 mg/l. The maximum permissible limit of Co is 0.05 mg/l. The results show that many of the stations cross the WHO-allowed limit but none of the stations show extreme values. Zinc ranges from 0.001 to 0.087 mg/l, with a mean of 0.03 mg/l during premonsoon and postmonsoon, demonstrating 0.004 to 0.045 mg/l with a mean of 0.016 mg/l. The concentration of zinc in these waters during both seasons has not crossed the prescribed limit of 3 mg/l. The relatively higher concentrations of zinc in some of the stations of the study area may be attributed to the presence of unused remains of zinc sulfate, which is an important constituent of fertilizers used in the region. The concentration of Fe in many of the stations is higher than the WHO-permitted limit of 0.3 mg/l. The average values during pre- and postmonsoon are 0.44 and 0.45 mg/l, respectively. The high Fe concentration in these waters may be assigned to the soil–water interaction, especially in the middle part and in the lower part of the river stretch, where it is observed that the water is almost stagnant, facilitating the dissolution and concentration of the ions in the river water. Pb values during premonsoon range from 0.092 to

0.985 mg/l, with a mean of 0.44 mg/l, and the values during postmonsoon range from 0.03 to 0.70 mg/l, with a mean of 0.27. Lead is usually found in low concentration in natural waters because Pb-containing minerals are less soluble in water. Concentration of lead in natural water increases mainly through anthropogenic activities. Almost all stations show the concentration of Pb higher than the WHO-allowed limit of 0.01 mg/l, which may create health hazards. The concentration of various industries along the middle part of the river course and their activities should have increased the higher concentration of lead in the river. Moreover, the atmospheric depositions resulting from the automobile pollution and the urban run-off due to precipitation may also lead to the increase in the concentration of Pb in the aquifers (Varrica et al. 2003; Rajnikant and Pervez 2003). The value of Cr ranges from 0.102 to 0.947 mg/l, with an average value of 0.512 mg/l during premonsoon and 0.07 to 0.56 mg/l with an average value of 0.25 mg/l during postmonsoon. The concentration of various industries such as tanneries, chrome plating, and dyeing industries near the river stretch, especially in the middle part of the river, contributes to the river chemistry.

Water quality

In order to identify the suitability of the water for drinking and irrigation, the quality of the water was evaluated using various methods. The major factors that decide the quality of the water in the study area are agricultural activities near the upper stretch of the river, rock–water interaction, anthropogenic activities at the middle and lower parts of the river, and saline water intrusion near the confluence point. Except at a few stations, the concentration of TDS (588 to 3,276 mg/l) is more than the recommended limit of 1,000 mg/l allowed (WHO 1984) in the water samples during both seasons, especially in the case of premonsoon, which may cause gastrointestinal irritation in consumers.

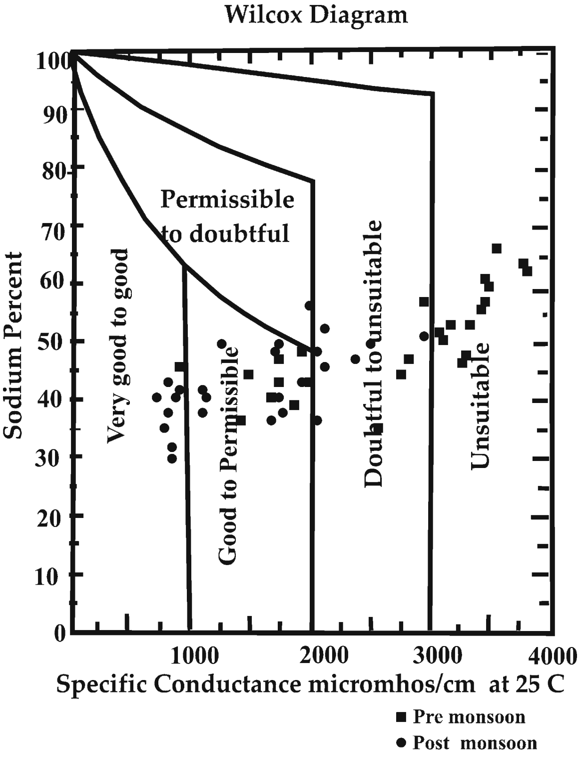


Fig. 7 Specific conductance and percent sodium relation for rating irrigation water (Wilcox 1955)

In order to assess the water suitability to irrigation, percent sodium, SAR, Wilcox, and USSL diagrams were evaluated. Wilcox (1955) used percent sodium and specific conductance in evaluating the suitability of water to irrigation (Fig. 7). Sodium-percentage determines the ratio of sodium to the total cations, viz., sodium, potassium, calcium, and magnesium. The results show that the water near the upstream is good for irrigation, and the contamination is found to be high near the middle and lower stretches of the river. During premonsoon, only 30% of water samples are shown to be good to permissible for irrigation; 13% fall in the region of doubtful to unsuitable; and 57% fall under the unsuitable region. This may be due to the increased anthropogenic activities near the middle- and downstream, owing to intense settlements along the bank of the river, directing the domestic sewage into the river and saline water intrusion. The results of postmonsoon show that 57% of the water values are good to permissible; 30% fall in the region of doubtful to unsuitable; and 13% of the water is unsuitable for irrigation. Postmonsoon demonstrates values favorable to irrigation, indicating that there is an extensive dilution of the river water during this period.

Electrical conductivity is a good measure of salinity hazard to crops, as it reflects the TDS

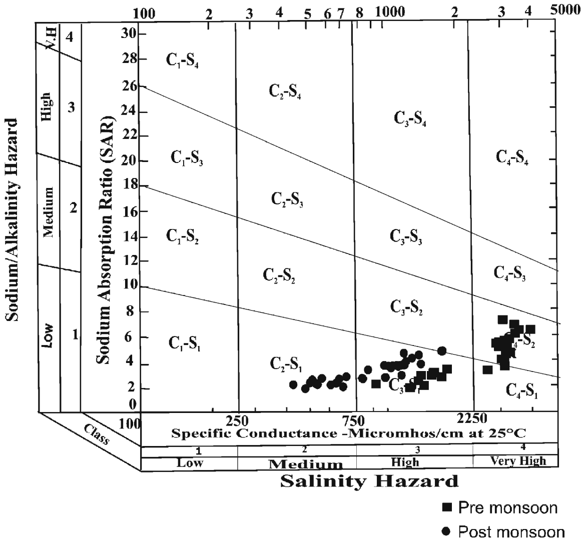


Fig. 8 Rating of water samples in relation to salinity and sodium hazard (USSL 1954)

in groundwater. Graphical representation (Fig. 8) of the chemical data on the irrigation suitability diagram (Richards 1954) clearly brings out the seasonal variation and the suitability of the water for irrigation. During premonsoon, the US salinity diagram demonstrates that the river water samples fall in the fields of C_3S_1, C_4S_1, and C_4S_2. About 37% of the samples fall in the field of C_3S_1 and C_4S_1, reflecting high salinity and low-sodium water, which can be used for irrigation on almost all types of soil, with only a minimum risk of exchangeable sodium. The water of this type can be used for plants having good salt tolerance (Karanth 1989). Remaining samples fall in the range of the high-salinity–medium-sodium (C_4S_2) region, which needs adequate drainage to overcome the salinity problem. In the case of postmonsoon, the US salinity diagram illustrates that most of the samples fall in the fields of C_2S_1 and C_3S1. Almost all the samples show medium to high salinity–low sodium, clearly demonstrating the seasonal variation and the requirement of adequate drainage for irrigation.

Conclusion

The present hydrogeochemical study of the river Cooum provides the following conclusions: The seasonal variation of the chemical parameters was evaluated and the order of abundance of the ions was determined. The $(Ca^{2+} + Mg^{2+})/(Na^{+} + K)$ ratio suggests that the composition of the water is not dominated by rock–water interaction. The ternary diagrams demonstrate low contribution of silica, suggesting that the contribution of silicate weathering to the water chemistry is low. The Gibb's diagram shows that the composition of water during premonsoon is dominated by evaporation–crystallization. In the case of postmonsoon, rock–water interaction predominates. One-to-one equiline diagrams represent the unique characteristic of the bonding affinity of ions. The diagram depicts that the chloride ions are found to balance the alkali ions and HCO_3^{2-} + SO_4^{2-} ions balance the alkaline earth metal ions. Piper diagram characterizes the water types of both premonsoon and postmonsoon. Premonsoon was found to be of NaCl and mixed CaMgCl type, whereas there is a shift from NaCl to mixed CaMgCl in the case of postmonsoon. Trace metal study reveals that the concentration of Fe is high in many stations. Pb and Cr are found to be high in certain stations due to anthropogenic activities. Wilcox and USSL diagrams suggest that the suitability of the water for irrigation is higher in the case of postmonsoon than in that of premonsoon.

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