Hydrogeochemistry and groundwater quality assessment in Nour coastal plain, Mazandaran province, Iran

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Abstract

Groundwater quality in Nour coastal plain in Mazandaran province of Iran is assessed by measuring physicochemical parameters including total dissolved solid, electrical conductivity, pH, total hardness, major cation and anion compositions and Nitrate pollution. For this purpose, fifty groundwater samples were collected in July 2012. The hydrochemical types, Ca–HCO_3 and Mg–HCO_3 dominate in all samples of groundwater. Nearly 98% the groundwater samples fall in the very hard category**.** Mineral saturation index of groundwater were calculated by PHREEQC. The all groundwater samples are oversaturated with respect to dolomite and calcite. According to Wilcox classification diagram, 98% of groundwater samples classified as C2–S1and C3–S1. In 80% of the groundwater samples sodium hazard was classified as low. In comparison with the WHO's drinking water guideline of 50 mg l^{-1} for NO_3^-, the NO_3^- concentration in groundwater was lower than this limit. The Kruskal–Wallis test is used to compare the NO_3^- parameter between five land uses. The result of Kruskal–Wallis test shows that samples have similar NO_3^- parameter in different land use.

Keywords: Geochemistry, Groundwater, Nitrate, Water quality, Sodium absorption ratio, Iran.

1– Introduction

Groundwater is the major source of water for various purposes in many countries. The chemical characteristics of groundwater play an important role in assessing and classifying water quality. Each groundwater system in the area has a unique chemistry, acquired as a result of chemical alteration of meteoric water recharging the system. Agricultural activities have increased the demand of groundwater resources in Iran. Water quality is influenced by natural and anthropogenic effects including local climate and geology.

Hence, to utilize and protect valuable water sources effectively and predict chemical changes in groundwater environments, it is important to understand the hydrochemical parameters of groundwater (Guendouz *et al.* 2003; Edmunds *et al.* 2006; jalali 2007; Prasanna *et al.* 2010).

The quality of groundwater is an important criterion to decide the suitability of water for irrigation activities and various parameters such as sodium adsorption ratio (SAR) and residual sodium carbonate (RSC) have been used to assess the suitability of water for irrigation purposes by several researchers (Kumar *et al.*, 2007; Jalali, 2007; Nagarajan *et al.*, 2010; Reddy and Kumar, 2010, Aghazadeh and Mogaddam, 2010). Nitrate (NO_3^-) is very soluble and the most usable form of N in plants. The recovery of fertilizer N in global crop production is about 50% (Krupnik *et al.* 2004, Jalali 2011). The monitoring of water quality is one of the important tools for sustainable development and water management and provides critical information for adapting measures to control pollution of groundwater (Jalali 2011).

Therefore, a hydrogeochemical investigation was carried out in Nour area in Mazandaran province to identify groundwater geochemistry and its suitability for drinking and irrigation purposes.

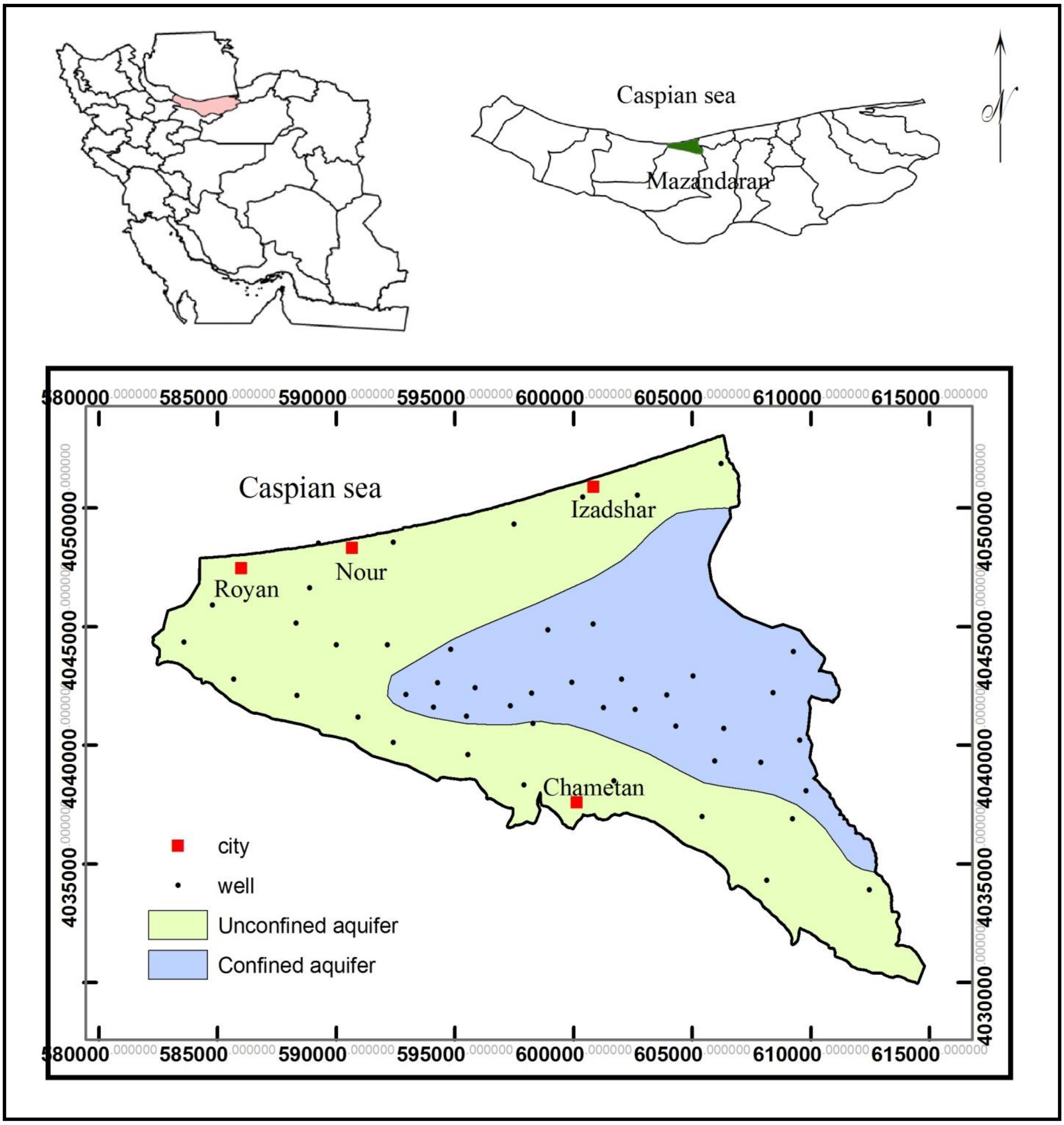

Figure 1) Study area showing location of wells sampled for groundwater analysis.

2– Study area

The study area was conducted in Nour township in Mazandaran province which is located on the Southern coast of the Caspian Sea, in the northern part of Iran. Nour township is divided into two parts: the coastal plain, and the mountainous areas. The Alborz Mountain Range surrounds the coastal plain of the Caspian Sea. The present research has been done on Nour coastal plain. It covers an area of 319 km^2 (Fig. 1) and lies between latitudes of 36°24′N to 36°37′N and longitudes of 51°55′E to 52°16′E.

Caspian Sea has experienced dramatic sea–level changes, ranging from lowstands probably several hundreds of metres below the present one in the Pliocene (Kakroodi, 2012) to 50 m above oceanic level in the Last Glacial. Thus, deposition in the Pleistocene–Holocene of the Caspian Sea had been controlled by the sea level fluctuations response to climatic changes in the Caspian region and adjacent areas (Ali zadeh and Aliyeva 2004). The most geological formation in this area includes marine sediments and alluvial deposit. The weathered sediments are derived from primary rocks including calcareous formations, the coastal plain has hot, humid summers, and mild, humid winters (Torabi *et al.*, 2011). The average temperature of the plain varies from about 8◦ C in winter to 25◦C in summer. The average annual rainfall of the area is about 1000 mm. The peak rain months include October, November and December. A considerable area of this plain is used for rice culture (Fig. 2).

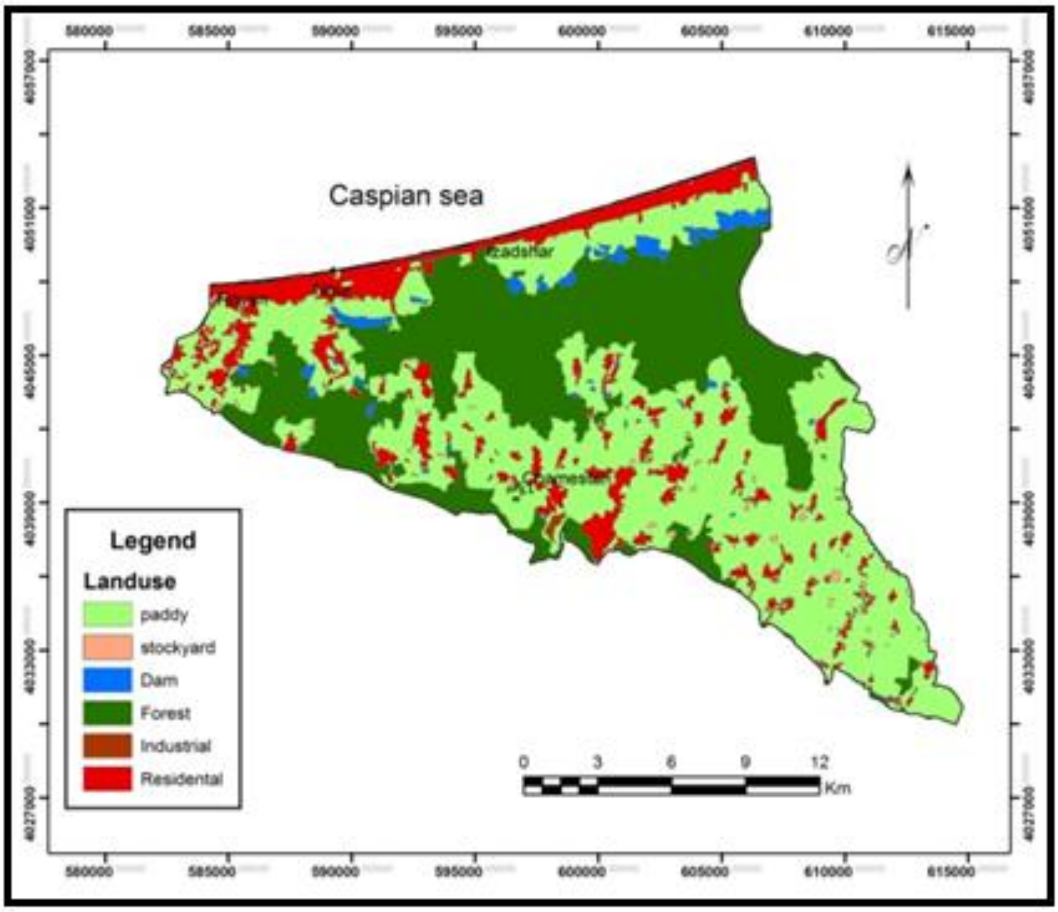

Figure 2) distribution of land use in study area.

3– Materials and methods

Fifty groundwater samples were collected from Nour coastal plain, during July 2012 (Fig. 1). Most of these wells supply water for irrigation, while some are exploited by industry, and some of them are used for drinking water. A total of 50 groundwater samples were taken from 25 unconfined Aquifer and 25 confined Aquifer. First a water level meter used to measure the water level in a well. Then, Water samples were collected after a pumping time of about 30 min. All water samples were collected in sterile polyethylene bottles. The collected samples were immediately stored in a field refrigerator on ice and transported to the laboratory where they were stored in the refrigerator at 4°C until parameter measurement. In all selected monitoring wells, 17 groundwater parameters including electrical conductivity (EC), total dissolved solids (TDS), total hardness (TH), pH, nitrate (NO_3^-), nitrite (NO_2^-), ammonium (NH_3), carbonate (CO_3^{2-}), dolomite and limestone bicarbonate (HCO_3^-), chlorine (Cl^-

),sulfate (SO_4^{2-}), phosphate (PO_4), calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), and (Fe) were measured. Samples were analyzed in the laboratory using standard methodologies. PH and EC were measured with a pH and EC meter (Mettler Toledo). Calcium, magnesium, chlorine, carbonate and bicarbonate were determined by titration (using standard EDTA, $AgNO_3$ and HCl, respectively). Sodium and potassium were measured by flame photometer (Jenway PFP7) and nitrate, nitrite, and sulfate were determined by colourimetry with an UV–visible recording spectrophotometer (Shimadzu UV–160A; Clesceri *et al*. 1989). The accuracy of the chemical analyses was verified by calculating ion–balance errors. The errors were generally around ±5%. An estimation of sodium adsorption ratio (SAR) in irrigation water was predicted using the following equation:

$$\mathrm{SAR} = \frac{\mathrm{Na}}{\sqrt{\frac{\mathrm{Ca+Mg}}{2}}} \qquad (1)$$

Where ionic concentrations were in milliequivalents per liter (meq l^{-1}). Residual sodium carbonate (RSC) value was calculated, using the relation, where ionic concentrations are expressed in meq l^{-1}.

$$RSC = (CO_3^{2-} + HCO_3^{-}) - (Ca^{2+} + Mg^{2+})$$

The data was statistically analyzed, Kruscal–Wallis test were determined to reveal the relationship between parameters, and a correlation matrix was carried out using the SPSS 18 software package. Pearson correlation coefficients were used to illustrate and measure associations between variables. Mineral saturation indices of the groundwater were calculated using PHREEQC software.

The maps of chemical parameters were prepared by GIS capability. Interpolation of spatial variables such as electrical conductivity and ion concentrations in groundwater are made by different kriging methods (Ahmad 2002, Kuswantoro *et al* 2013). Chemical analysis results of a certain chemical constituent were spatially interpolated by utilizing Kriging method available in ARC GIS software. In this method, the best semivariogram model for every parameter varies based on the root mean square error (RMSE) criterion.

4– Results and discussion

4.1– Groundwater level

There are two types of aquifers in the study area (Fig.1) namely unconfined and confined (Mazandaran regional water, 2009). The water level varied from 0 to 60 m below the ground level. The distribution pattern of groundwater levels is related to the general topography (Fig. 3). The water table depth is mostly less than 10 m in coastal plain (north of area) and greater than 20 m in alluvial fan (south of area).

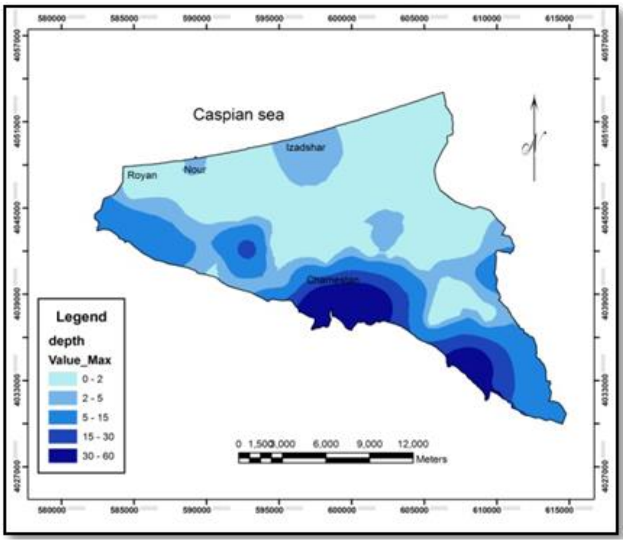

Figure 3) Distribution of groundwater level in study area.

4.2– Major ion chemistry

Quality of groundwater determines its suitability for different purposes depending upon the specific standards. The chemical compositions of the groundwater samples were statistically analyzed and the results are presented in Table 1. Chemically, the water used for drinking should be soft, low in dissolved salts and free from toxic constituents (Ramesh and Elango 2011). The pH values of the groundwater vary between 7 and 8, with an average of 7.5, which indicates the alkaline nature of groundwater of the study area. The EC values varied from 542 to 3,090 µS cm^{-1}, with an average of 833 µS cm^{-1}. EC was above the maximum permissible limit (1,500 µS cm^{-1}) in 5% of groundwater samples. Almost all the samples exceed the desirable limit for EC of 500 µS cm^{-1}. The TDS values are between 347 and 2,070 (mean 540.6

mg/L). TDS level of less than about 600 mg/L is generally considered to be good; drinking–water becomes significantly and increasingly unpalatable at TDS levels greater than about 1000 mg/L (WHO 2011).

Table 1) Chemical compositions (major elements) in the well waters (50 samples) Include WHO Standards 2011.

	Mean	Min	Max	Sd	WHO
TDS	540.6	347.0	2070.0	234.3	<1000
EC	833.0	542.0	3090.0	346.3	–
PH	7.5	7.0	8.0	0.3	6.5–8.5
Ca	114.9	80.0	294.0	30.3	75
Mg	15.8	9.6	34.8	4.0	30
Na	26.8	6.9	301.3	43.6	200
K	2.9	2.0	5.1	0.6	–
NO_3	5.0	1.3	16.7	3.3	<50
NO_2	0.02	0.02	0.1	0.02	<3
HCO_3	400.3	262.3	707.6	74.1	–
SO4	12.4	4.8	81.6	11.8	250
Cl	44.2	17.8	617.7	84.5	250
Fe	0.4	0.0	2.57	0.55	0.3
TH	352	284	877	88.4	100–300

The mean values of Ca^{2+}, Mg^{2+}, Na^+ and K^+ ions varied from 114.9, 15.8, 26.8 and 2.9 mg/L in water samples, respectively (Table 1). According to WHO (2011) guidelines, the maximum admissible limit these cations are acceptable.

TH as $CaCO_3$ in the study area ranges from 284 to 877 mg/L with an average value of 352 mg/L. Groundwater exceeding the limit of 300 mg/L $CaCO_3$ is considered to be very hard (Sawyer *et al*., 2003). The 98% of the groundwater samples fall in the very hard category. There is some suggestive evidence that long term consumption of extremely hard water might lead to an increased incidence of urolithiasis, anencephaly, pre–natal mortality, some types of cancer and cardiovascular disorders (Agrawal and Jagetai 1997, Ramesh and Elango 2011).

Among the anions, the concentrations of HCO_3^-, Cl^-, SO_4^{-2} and NO_3^- represent on average 400.3, 44.2, 12.4 and 5 mg/L respectively. The carbonate concentration (CO_3^{-2}) in all water samples was negligible. The desirable limit of Cl^- and SO_4^{-2} for drinking water is specified as 200 mg/L as per WHO and only 2% of the samples are above this limit.

The dominance of the major cations is as $Ca^{2+}>Na^+>Mg^{2+}>K^+$ and anion abundance is $HCO_3^->Cl^->SO_4^{2-}$. Based on dominant lithological formations in the study area (calcite and dolomite), the sources of Ca^{2+} and Mg^{2+}, are calcite ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$). The sources of Na^+ probably, is chloride salt in marine sediment of Caspian sea.

4.3– Saturation index

The saturation indices describe quantitatively the deviation of water from equilibrium with respect to dissolved minerals (Jalali, 2010). Mineral saturation index of groundwater was calculated by PHREEQC. Table 2 shows summary statistics of groundwater samples classified into three categories: saturated (SI>0), saturation (SI=0) and undersaturated (SI<0). All water samples are saturated with respect to dolomite, calcite and aragonite (SI>0). Therefore, water chemistry in the study area seems to be largely affected by the dissolution of carbonate minerals. All groundwater samples were undersaturated with respect halite, anhydrite and gypsum.

Table 2) Summary statistics of mineral saturation index of groundwater calculated by PHREEQC.

mineral	Anhydrite	Aragonite	Calcite	Dolomite	Gypsum	Halite
Maximum	–0.09	3.28	3.42	6.15	–0.18	–2.67
Minimum	–1.23	2.31	2.04	4.17	–1.2	–5.82
Average	–0.86	2.75	2.87	5.10	–0.66	–5.16
Standard deviation	0.26	0.24	0.26	0.46	0.24	0.56

4.4– Groundwater quality

The chemical composition of water samples is plotted in a Piper diagram (Fig. 4). HCO_3^- and Ca^{2+} were the dominant ions in water samples (Table 1). Therefore, most of the groundwater samples (98%) have Ca–HCO_3 types in the piper diagram.

Cl^- ion concentration normally arises from three sources; dissolution of halite (NaCl) and related minerals in evaporate deposits, ancient seawater entrapped sediments, and atmosphere deposit (Walker *et al*. 1991).

The EC and Na concentration are usually considered important in classifying irrigation water. High EC in groundwater leads to formation of saline soil, whereas high Na content causes alkaline soil (Nagarajan *et al*. 2010). According to the Wilcox classification diagram (1955), groundwater classes are mainly C2–S1 (40%), C3–S1 (58%), and only one sample is C4–S2 (Fig. 5). Nearly 60% samples fall in C3–S1 classes that salinity is high. Water cannot be used on soil with restricted drainage. Sodium in this water class is low. Water can be used for irrigation on almost all soils with little danger of the development of Na+. In C2–S1 classes, salinity is medium. Water can be used if a moderate amount of leaching occurs. Thus, the salinity hazard in groundwater samples is regarded as low to medium, but the Na+ hazard is regarded as low.

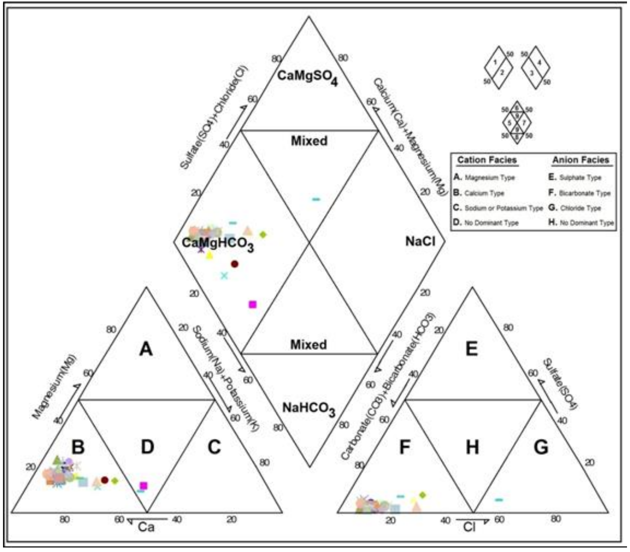

Figure 4) Piper diagram for groundwater samples in study area.

SAR is an important parameter for determining the suitability of groundwater for irrigation because it is a measure of alkali/sodium hazard to crops. The calculated SAR in this area ranged from 0.8 to 23.4. In coastal zone, the SAR values are higher than others (Fig. 5). Sodium hazard for most of the water samples (80%) was classified as low (SAR<3), 16% had medium (3 < SAR < 9), and 4% had high SAR values (SAR > 9). When SAR values are greater than 9, irrigation water will cause permeability problems on shrinking and swelling in clayey soils (Saleh *et al*. 1999). There was a high correlation between EC and SAR of groundwater in the study area (r=0.866), the spatial distributions of EC and SAR showed nearly a similar pattern.

Residual sodium carbonate (RSC) has been calculated to determine the hazardous effect of CO_3^{2-} and HCO_3^- on the quality of water for agricultural purpose (Eaton 1950).

The RSC<1.25 are safe for irrigation, it is considered unsuitable if it is greater than 2.5 (USEPA1999). In study area RSC in groundwater varies from −5.9 to 2.8 meq/L with an average of −0.47 meq/L. Negative RSC indicates that Na^+ buildup is unlikely since sufficient Ca^{2+} and Mg^{2+} are in excess of what can be precipitated as CO_3^{2-}. However, with respect to RSC, all samples are within the safe quality categories for irrigation. This indicates suitability of water for irrigation purposes.

4.5– Nitrate concentrations in groundwater

Marked increase in fertilizer N rates applied to agricultural soils causes N to be leached below the rooting zone (Pratt 1985; Hallberg 1989). Nitrate concentrations in the groundwater samples varied from 1.3 to 16.7 with an average of 5 mg/L. In comparison with the WHO’s (2011) drinking water guideline of 50 mg/L for Nitrate, NO_3^- concentration in groundwater less than permissible limit.

Groundwater with NO_3^- concentration exceeding the threshold of 13 mg/L NO_3^- is considered contaminated due to human activities (the so–called human affected value; Eckhardt and Stackelberg 1995; Jalali and

Kolahchi, 2007, Jalali 2011). In study area only 2% of the water samples showed NO_3^- concentrations above the human affected value and located near cities (Fig. 6). The distribution pattern of nitrate does not have any relationships with land use.

The Kruskal–Wallis test is a nonparametric test for deciding whether is significant between three or more samples (Ebdon 1992). It is a useful alternative to analysis of variance. This test was used to compare the NO_3^- parameter between five land uses. The result of Kruskal–Wallis test shows that nitrate in different land use have sig. >0.05 and thus have similar NO_3^- parameter in different land use.

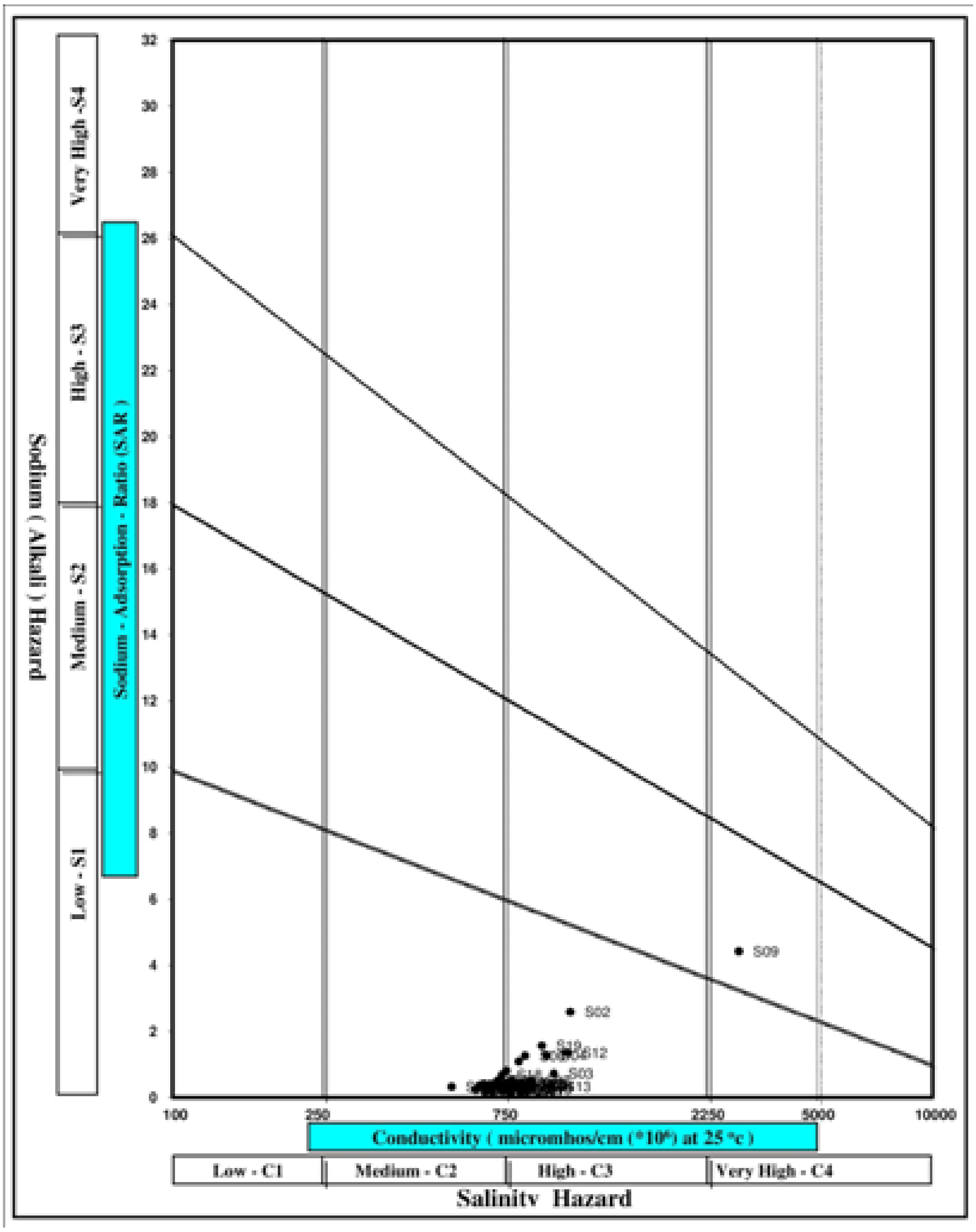

Figure 5) Diagram of sodium adsorption ratio (SAR) and salinity for the classification of groundwater for irrigation purposes in Nour coastal plain.

4.6– Correlation coefficient

The chemical composition of groundwater depends on the mineralogical composition, physical properties of rocks, duration of contact with the aquifer materials and the nature of recharge water (Born *et al*. 1987, Dudeja *et al*. 2011). In Nour coastal plain, groundwater chemistry is strongly influenced by the minerals of the limestone and dolomite rocks which recharge water had flowed.

Correlation coefficient is a statistical measure between any pair of variables. In this study, Pearson correlation is used to measure correlation between ions. The correlation coefficients of the analyzed parameters are presented by correlation matrix in Table 3.

Table 3 The matrix correlation among various water quality parameters

Variables	PO_4	NO_3	NO_2	NH_3	HCO_3	Cl	SO_4	Ca	Mg	EC	TDS	Na	K	Fe
PO_4	1.00													
NO_3	–0.18	1.00												
NO_2	–0.16	0.29	1.00											
NH_3	0.62	–0.25	–0.25	1.00										
HCO_3	0.22	–0.17	0.02	0.57	1.00									
Cl	0.01	–0.17	–0.10	0.35	0.59	1.00								
SO_4	–0.02	–0.19	0.00	0.21	0.55	0.87	1.00							
Ca	–0.07	–0.17	0.00	0.28	0.82	0.83	0.81	1.00						
Mg	–0.03	–0.42	0.06	0.21	0.68	0.66	0.66	0.72	1.00					
EC	0.08	–0.20	–0.06	0.45	0.79	0.96	0.87	0.92	0.74	1.00				
TDS	0.08	–0.20	–0.06	0.45	0.79	0.96	0.87	0.92	0.75	1.00	1.00			
Na	0.20	–0.16	–0.13	0.55	0.67	0.95	0.82	0.76	0.61	0.95	0.95	1.00		
K	0.27	–0.21	–0.01	0.56	0.86	0.63	0.66	0.72	0.61	0.79	0.79	0.75	1.00	
Fe	–0.11	–0.25	–0.20	0.10	–0.02	0.21	0.04	0.07	0.09	0.15	0.15	0.20	0.09	1.00

Ca^{2+} and Mg^{2+} presented a positive correlation ($r = 0.72$), indicating a common source. Also, they were positively and significantly correlated with Cl^-, HCO_3^-, SO_4^{2-} ،Na^+ and K^+**.** The dissolution of carbonate releases Ca^2+ into solution, yielding water of type Ca–HCO_3 as a final product**.**

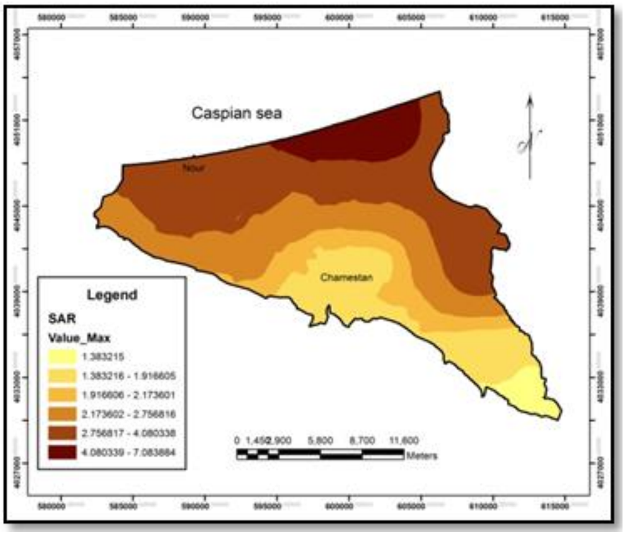

Figure 6) Sodium adsorption ratio (SAR) map of Nour coastal plain.

The plot of HCO_3 vs (Ca + Mg) shows that plotted points lie well above the 1:1 line (Fig. 7a). Concentrations of (Ca + Mg) exceeding HCO_3 suggests that a fraction of the (Ca + Mg) in these waters has to be balanced by other anions such as SO_4^{2-} and/ or Cl^-. The plot of (Ca +Mg) vs (HCO_3 + SO_4 + Cl) shows good correlation throughout the range of data (Fig. 7b). The presence of carbonate lithology in the surrounded mountain supports carbonate dissolution and the major fraction of Ca + Mg and HCO_3 is derived from the weathering of carbonates**.**

The correlation between SO_4^{2-}, Ca^{2+} and Mg^{2+} are 0.81 and 0.66, respectively. The presences of $MgSO_4$ and $CaSO_4$ in marine sediment likely are the major sources of sulphate in groundwater of study area.

Na^+ and Cl^- exhibit high correlation coefficient in p value <0.01 (0.954). The low concentration of Cl^- (mean 44), high Mg/Na ratio (2) and absence of evaporates formation indicate that the fraction of cations derived from evaporates accompanying Cl^- are likely to be negligible. But, the presence of NaCl, $MgCl_2$ and KCl in the marine sediment is the major sources of Cl^- in groundwater.

The Na+K/Cl ratio more than 0.55 (in the study area 1.02) is indicative of the source to be weathering of silicate mineral for the contribution of sodium and potassium (Zhang *et al*. 1995).

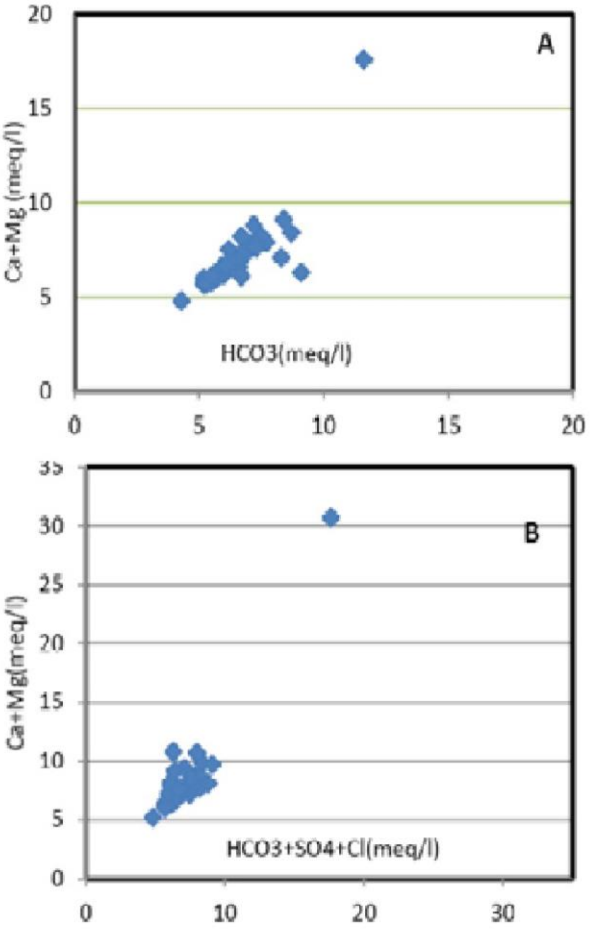

Figure 7) Scatter diagrams showing relationship between. ***A:*** *(Ca + Mg) vs HCO3;* ***B:*** *(Ca + Mg) vs HCO3 + SO4 + Cl.*

Factor analysis reduces the contribution of less significant variables by further simplifying data structure resulting from PCA (Nosrati and Eeckhaut, 2012).

FA was carried out using SPSS software with principal component analysis (PCA) method and varimax rotation in order to determine the optimum number of factors. The solution using the eigenvalue 1 in three components explain 78% of the total variance (Table 4). Component 1 indicate that TDS, EC, $Ca^{2}+$, Cl^{-}, $SO_4{}^{2-}$, Na^{-}, Mg^{2-}, $HCO_3{}^{-}$ and K^{+} explain 52.25% of the total variance. The constituents in component 2 are PO_4 and NH_3 explain 14.79% of total variance. Component 3 explains 10.99% of the total variance defined by Fe and probably indicates weathering of carbonate minerals (Fig. 8).

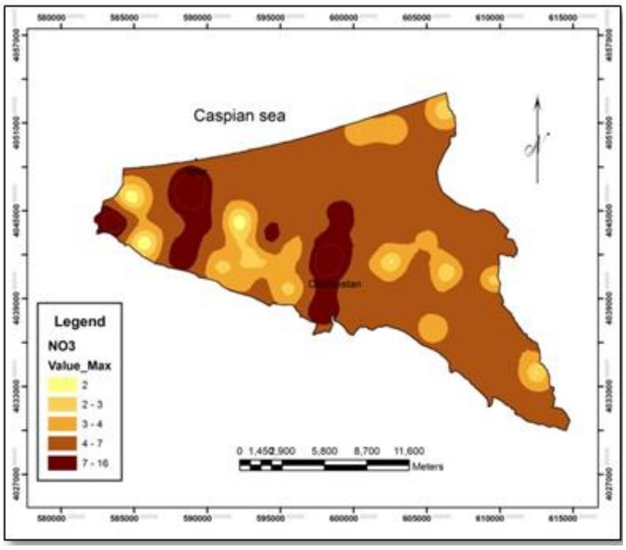

Figure 8) Spatial distribution of Nitrate (NO_3) in study area.

5– Conclusions

Ionic concentration, TDS, EC and other groundwater quality parameters in Nour coastal plain suggest that climate; lithological units and residence time are major controlling factors of groundwaters chemistry. The Piper diagram showed that the all water sample have CaMg–HCO_3 type**.** Mineral saturation index of groundwater calculated by PHREEQC, show that nearly all water samples are oversaturated with respect to dolomite and calcite. Also, correlation positive between Ca^{2+}, Mg^{2+}, Cl^{-} , $HCO_3{}^{-}$, $SO_4{}^{2-}$, Na^{+} and K^{+} show the nature of aquifer that includes marine sediment of Caspian Sea. The marine sediment consist NaCl, $MgSO_4$, $CaSO_4$, $MgCl_2$, $CaCO_3$ and KCl that have been deposited by Caspian sea level fluctuations response to climatic changes. Hence, According to Wilcox diagram, nearly 60% samples fall in C3–S1 classes that salinity is high. Sodium hazard for most of the water samples (80%) was classified as low value. In study area, according to WHO guidelines, the maximum admissible limit cations are acceptable. However all the groundwater samples fall in the very hard category. Notwithstanding, agriculture is the dominant land use in the studied area, $NO_3{}^{-}$ concentration in groundwater less than permissible limit. The

Kruskal–Wallis test shows that nitrate in different land use have similar value.

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