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Use of Isotope and Solute Chemistry to Define the Sources of Pollution in Mawqaq Groundwater, Hail, Saudi Arabia

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إستعمال النظائر المشعةِ والكيمياءِ التحليلية فى التعرّفَ على مصادر التّلوثِ فى المياه الجوفية فى ماوقاق، حايل، بالمملكة العربية السعودية

إبراهيم عبدالعزيز الساجابي ومحمد علي معلم

خلاصة: تستعملُ المياه الجوفية فى المنطقة للأغراضِ المختلفةِ مثل الاستعمال المنزلى و الصناعي و الزراعي والجمالى. أكثر من ستين عينة من عينات المياه الجوفية قدْ جُمِعتْ فى حقل ضحل في مدينةِ ماوقاق والمناطق المحيطة بها. وقد كان الهدف هو دراسة نوعية المياه الجوفية وتحديد مصادر النترات (إن أو 03) والتلوث البكتريولوجي في المياه الجوفية باستعمال النظائر المشعة. وقد اتضح من نتائج الدراسة أن متوسط تركيز الملوحةِ أعلى من الحدِ المسموح به فى مياه الشرب (500 ملغرام/لتر). وقد تم تحديد ثلاث مجموعاتِ من المياه المالحة كما صودفَ أيضا كميات عالية من النّتراتِ كنتروجين (إن أو 03 إن) في هذه المنطقة ، وقد تم العثور أيضا على جراثيم الكوليفورم في بعض الآبار. وباستخدام النظائر المشعة المستقرّة أمكن تحديد عُمر ومصدر التّلوثِ في المياه الجوفية فى منطقة ماوقاق. وقد أتخذت السلطات الصحية المختصة والبلديات إجراءات علاجية وبرنامج مُرَاقَبة يطبقان بالتّنسيق بين هذه السلطات.

ABSTRACT: Groundwater in the area is used for different purposes like domestic, agricultural, industrial and landscaping. More than sixty groundwater samples were collected in an alluvial shallow aquifer in and around Mawqaq Town. The objective was to study groundwater quality and identify the sources of Nitrate (NO_3) and bacteriological pollution in groundwater using environmental isotopes (^2H, ^3H and ^{18}O). The results of the study have revealed the presence of average salinity concentration higher than the permissible limits (500 mg/l) for drinking water. Three groups of water saline boundaries were identified. The presence of high nitrate as nitrogen (NO_3-N) was also encountered in the study area. Coliform bacteria have also been identified in some wells. Application of stable isotope has shown the age, origin and the sources of the pollution in the groundwater of Mawqaq area. Appropriate health and municipal authorities of the area have initiated remedial measures and a monitoring program is to be implemented in coordination with these authorities.

KEYWARDS: Salinity, Bacteriological, Pollution, Septic Tanks, Nitrate, Saudi and Remedial.

1. Introduction

The shallow groundwater aquifers have been increasingly used to meet the different uses resulting from urban expansion and agricultural activities. Constraints imposed by quality and growing problems of contamination has become very important issues. Water supply requires an understanding of groundwater flow and the chemical processes involved in the water bearing systems in order to develop, augment and present a solution to any problem in the water supply system. Shallow aquifers in the Kingdom are the main water supply for rural areas and are prone to chemical change and contamination from many factors such as formational change, high pumping, salt intrusion, septic tanks and agricultural activities. Concerns for water resources shortage and augmentation demands were received from the inhabitants of Mawqaq town, in central Saudi

Arabia. The research presented in this paper is part of a study that covered hydrogeology, hydrogeochemical and geophysical investigations to develop water resources and to find adequate water supply in an acceptable quantity and quality for the town. The nearest similar groundwater quality studies were done in Qasim and Hail. Few detailed studies can be found in the literature about the chemistry and quality change (Alsagaby and Moallim, 1996; Sharaf and Hussein, 1996; Sowayan and Allayla 1989; Hussein, *et. al*. 1992; Segar, 1988; Edgell, 1989; Jerias, 1986; BRGM, 1985 and Watban, 1976). Various ideas in different topics were achieved and presented in these studies. High saline groundwater, elevated nitrate concentration and bacteriological contamination were encountered in the supply system of Mowqaq town. The objective of this study is to apply solute chemistry and isotopes to clarify the sources of water quality change and biological contamination in the shallow aquifer and present a solution to the contaminated groundwater supply system.

2. Description and Geological Setting of the Study Area

Mawqaq Town lies in the middle of a Wadi alluvial deposit surrounded by mountains, at an elevation of 1200 m above sea level. It lies 65 km southwest of Hail the capital of the area. It is elongated in an alluvial strip towards northwest direction and depicted between longitude 41^{0} 00’ 00” and 41^{0} 20’ 00” and latitude 27^{0} 15’ 00” and 27^{0} 30’ 00”. The relevant physiographics structures (Figure 1) around the town can be described as dunes in the north, mountains in the east and south and wadis in the west. Three alluvial Wadi trends are identified to be the main watercourses, which enter the town from the south and passes towards the north as observed from the satellite image and verified in the field. These trends were also observed from the preliminary analysis of water samples. Three distinctive water types were defined from the preliminary chemical data analysis.

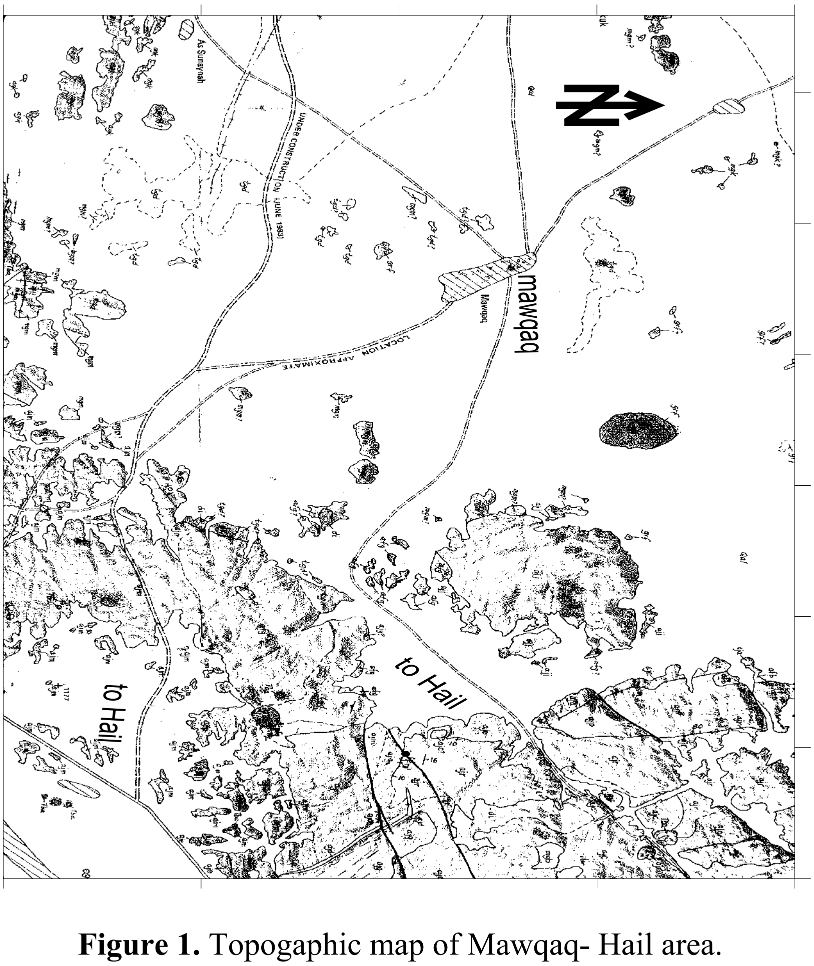

Figure 1. Topogaphic map of Mawqaq- Hail area.

Geologically, the study area is located in the middle of the northern part of the Arabian shield in central Saudi Arabia. It is composed of a low lying plain composed of sand and gravel of various grain sizes with low strength of consolidated beds intercalated in the alluvial material deposited originally by floods from the mountains. A complex of crystalline rocks of various nature and origin surrounds this low land filled by alluvial fans. Buried channels of various extensions are located. The forms and directions of the channels were controlled by structural phenomenon. Small mountains of crystalline rocks are scattered in the plain. The major crystalline rocks surrounding the study area can be texturally classified into porphyry and aphanitic Granite. The age of the recognized rock range from Precambrian to Tertiary followed by Quaternary alluvial deposits in stratigraphical sequence, (Figure 2).

The Precambrian units in the area are divided into two groups: Sedimentary and volcanic (layered) rocks and plutonic rocks. The layered rocks are classified as Banana Formation (volcanic and hypabysal rocks) and Hadn Formation of silicic volcanic intercalated with fieldpathic sedimentary rocks. The rocks covering this area are characterized by the presence of medium to coarse Granite, monzogranite and Grandiorate with Biotite and Hornblende. The Mawqaq complex is, in general, made of deformed, metamorphosed and foliated Biotite with or without hornblende, Granodiorite, Tonalite, Monzogranite and quartz Diorite. The overall intrusive relationship of all rock types in the area is not clear, but it appears that these quartz Diorites and Tonalites are closely related and Granodiorite and Monzograinte appears to be somewhat younger. These rocks are cut by series of Basalt dikes, which also appears to be related to the intrusive complex. Olivine Basalt of Tertiary age occupies, as a field of pipeline and necks, throughout the area. Quaternary alluvial and colluvim deposits of unconsolidated Wadi material and pediment silt, sand and gravel occur in the study area. Deposits of alluvial fan originating from the Wadi drainage system were located to the south and west of Jabal Aja. Moderate to well sort sand sized particles, but includes silt, and pebbles in the wadi channels dominate these sediments. Complicated tectonic history and formation of structural diversity in consequence of tectonic events have affected the area. Metamorphism of various degrees has been reported and intrusion of molten material, resulting in the creation of dikes. Lineaments and faults, which control the pattern and formation of the alluvial aquifer and groundwater flow, were also found in the study area (Stoesser and Elliot, 1985).

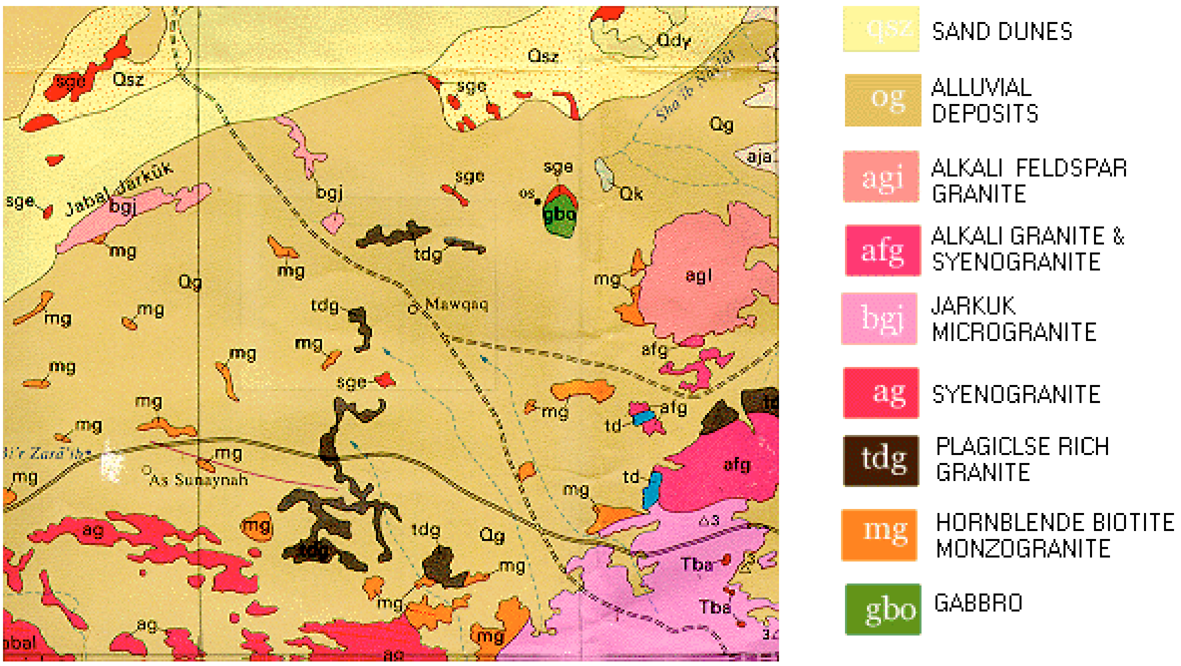

Figure 2. Simplified geologic map of Mawqaq (Hail area).

3. Methods and Material

More than 60 samples from water wells in the study area for water quality assessment were collected in order to achieve reasonable interpretation, evaluate the sources of groundwater salinity and delineate any possible pollution. Representative chemical and bacteriological groundwater samples in the study area were used in order to assess water quality. Measurements of pH, temperature, and EC were done in the field. The analyses conducted in the laboratory included major anions like Ca, Mg, Na, K, CL, SO_4, HCO_3, NO_3, and hardness as $CaCO_3$ and CO_3. Sampling has been conducted with time difference of about three month. The first sampling (55 samples) was started in March 1998 and second part (11 samples) was in July 1998.

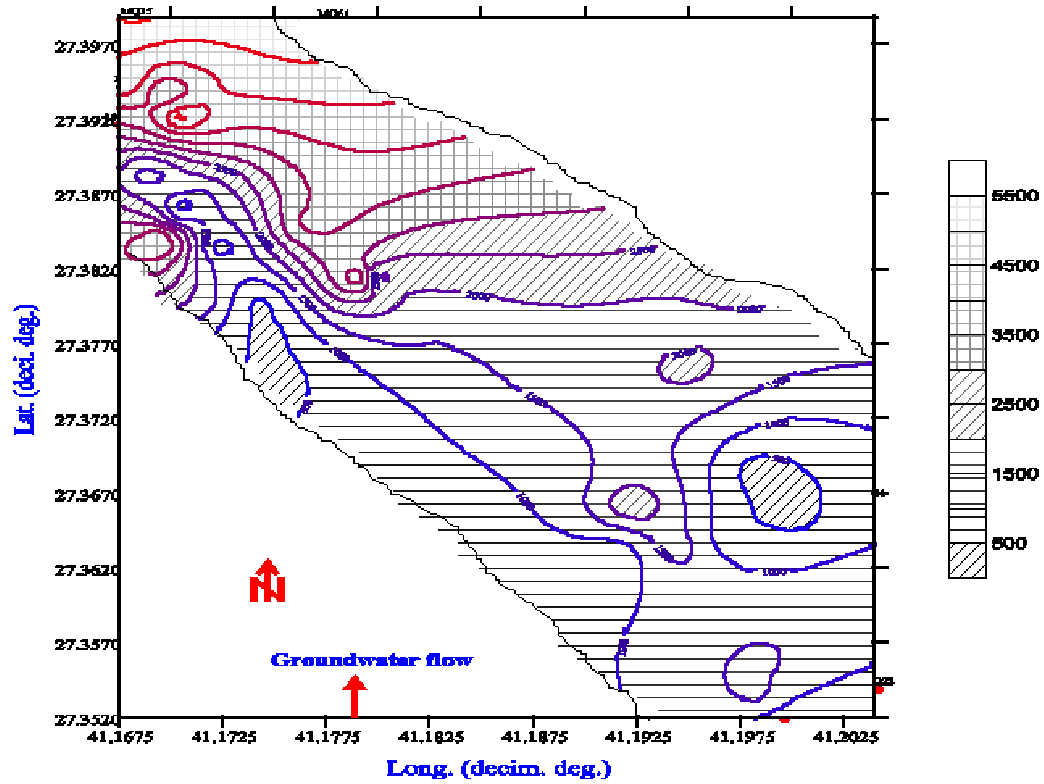

Figure 3. Total dissolved solids in Mawqaq wells, Hail area.

4. Results and Discussion

4.1 Water Quality

The results of the data collected from the field have shown field pH variation ranging from 7.86, slightly alkaline water, to 5.67, slightly acidic water.

Groundwater salinity in the aquifer changes from low to high saline along the down gradient. The maximum TDS (5786 mg/l) in the aquifer is found in the north and the minimum (265 mg/l) in the south with average of about 1960 mg/l. The average salinity is higher than the permissible limit (500 mg/l) for drinking water (WHO, 1984). This increase of TDS coincides with the decrease of well production in the area. The result also revealed an abrupt salinity change within a perimeter in some locations coupled with water well depth change in some parts of the study area, especially in the northern part. Structural complexity, which controlled the depositional environment of the alluvial material of the aquifer, may have contributed to the salinity increase.

Figure 3 presents the pattern and distribution of salinity in the aquifer. Three salinity trends and boundaries were observed from the Figure. The first saline condition extends from the southern boundary to 1000 mg/l salinity (low salinity condition) and the second lies between 1000 mg/l-salinity boundary and 3000 mg/l-salinity boundary (moderate salinity condition). The third system of salinity is seen above 3000 mg/l salinity boundary (high salinity condition). The chemical data have also revealed that about 50% of the sampled wells high chloride concentrations above the acceptable limits for drinking water (CL>250 mg/l). About 43 % of the samples have a sulfate concentration of more than the acceptable limits for drinking water (SO_4 >250 mg/l). High sodium, calcium and bicarbonate concentrations were also observed (Table1). Geologically, the aquifer is a recent alluvial aquifer and has characteristically high saline water.

4.2 Ion Relationship and Water Composition

The quality of water generally expected from an alluvial aquifer is an access of Ca-HCO_3 water type unless influences by other materials are involved in the system. A plot of chloride versus sodium (Figure 4) for the study area shows nearly a 1:1 ratio in the beginning of the graph and chloride starts increasing reaching a concentration of about 1400 mg/l (39.5 meq/l), while maintaining the same sodium concentration of up to 200 mg/l (8.7 meq/l). Scattered points, were observed at the end of the graph. Figure 5 displays a water type of high chloride with proportional sodium in the beginning and in the middle part, while at the end of the graph the ratio is nearly maintained constant proportion. The line nearly extrapolates to the sodium axis, indicating that high concentration of sodium is present in the system. Moreover, significant concentration of sulfate scattered at three different groups with different concentrations and positive correlation between TDS and sulfate were observed. Figure 6 presents bicarbonate distribution as a function of total dissolved solids in the aquifer and displays three distinctive constant concentrations of bicarbonate. The possible interpretation is mixing between fresh water and saline water. This mixing could be the result of modern day recharged waters (high bicarbonate).

Table 1: Mawqaq isotopic data

Serial #	Name	Delta 0-18	Delta D
1.	Mq1	-1.72	-0.4
2.	Mq2	-1.95	-3
3.	Mq3	-0.35	-0.8
4.	Mq7	9.53	
5.	Mq8	0.03	1.6
6.	Mq9	0.01	1.7
7.	Mq10	-1.38	-3
8.	Mq13	-0.93	-2.2
9.	Mq15	-0.77	
10.	Mq17	-1.35	-1.9
11.	Mq19	-0.98	-4
12.	Mq20	-2	-7.1
13.	Mq22	0.46	-0.3
14.	Mq29	-2.21	-2.4
15.	Mq32	5.67	
16.	Mq36	6.45	15.7
17.	Mq42	0.5	-4.6
18.	Mq43	-2.14	-2.3
19.	Mq44	-1.59	-2.9
20.	Mq50	-2.27	-4.5
21.	Mq53	-1.06	-5.2
22.	Mq55	-0.32	-2

Hem (1970) has reported that nearly all-natural waters, in terms of chemical classification, lie between 0.55 to 0.75 (correlation coefficients for EC and TDS). A mixture of water types represented by NaCL, $CaSO_4$ and $NaSO_4$ range between 0.65 to 0.75. The relationship between EC and TDS is shown in Figure 7.

The coexistence of a good relationship is evident from the correlation coefficients (R^2 = 1). The conversion factor (0.65) for the samples falls in the range designated for NaCL, Mg-$CaSO_4$, $NaSO_4$ and Ca-HCO_3 water type by Hem (1970). Therefore, a mixing system of waters is prevalent in the aquifer.

4.3 Groundwater Pollution

Certain environmental conditions may have resulted in a widespread distribution of groundwater pollution in Mawqaq town. In the study area, it is manifested by the way in which septic tanks are constructed. The presence of harmful contaminants indicates that environmental controls should be applied in the area. Biochemical contaminant presence in the groundwater of the study area makes it necessary to initiate protection of the groundwater for the inhabitants of the town. The sources of the pollution, its impact and solutions will be considered in the following paragraphs.

4.4 Nitrate Pollution (NO_3-N)

In addition to high salinity presence, very significant high nitrate concentrations, which increases towards down gradient of the aquifer, have been recorded in the study area. The nitrate concentration recorded in the water samples range from 2.8 mg/l (minimum) to 497 mg/l (maximum) with an average of 111.53 mg/l. The geographical distribution of nitrate concentration in groundwater of the alluvial aquifer is shown in Figure 8. Elevated concentrations are found in the northern part of the aquifer although local variations are common in some parts. Artificial fertilizers for farming purposes were not used in the area (personal contact with inhabitants). The presence of nitrate as nitrogen in Mawqaq groundwater is higher than the permissible limit for the drinking water (10 mg/l), which was defined by World Health Organization in 1981 and 1993 (Edmunds and gaye, 1997).

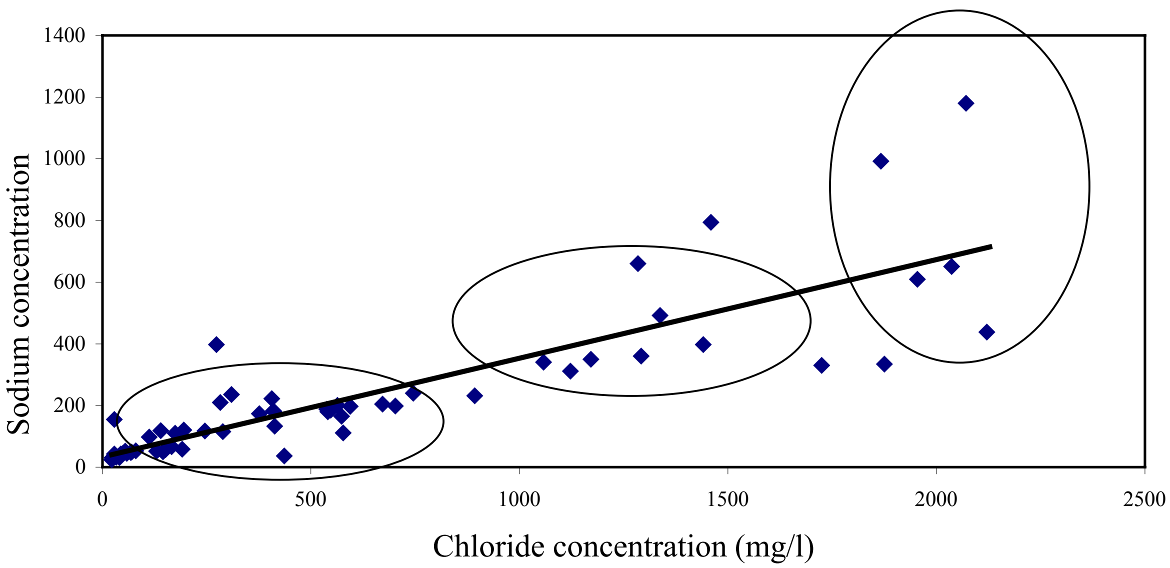

Figure 4. A plot of chloride versus sodium for Mawqaq wells, Hail area

High nitrate concentrations are generally assumed to be the results of human activities, but occurrences of high NO_3 groundwater far from man made activities have been recorded in arid and semiarid regions, especially in North Africa. Up to 2800 mg/l of NO_3-N were recorded in Sudan, near Khartoum. This was attributed to very low recharge rates and the accumulations from vegetation over many centuries. In Kalahari and Libya similar problems were encountered (Edmunds *et. al*., 1992). Excess of Nitrogen concentrations in groundwater, leaching from the domestic septic systems, (residential area) and wastewater disposals were also recorded in Australia (Gerriste, *et. al*. 1995) and in Ontario, Canada (Aravena, *et. al*, 1993).

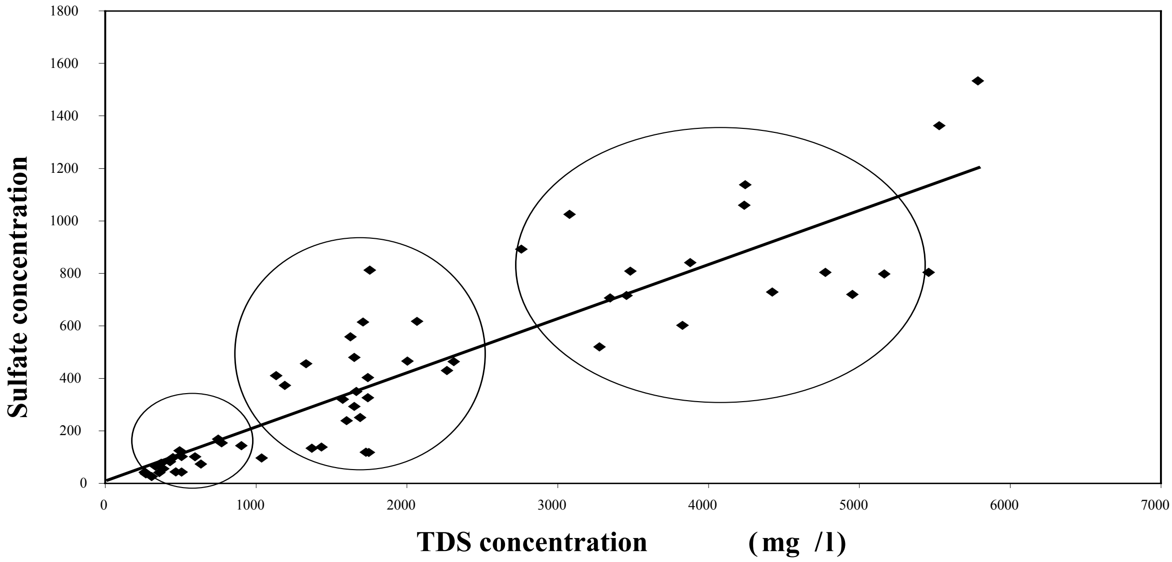

Figure 5. A plot of TDS versus sulfate in the study area.

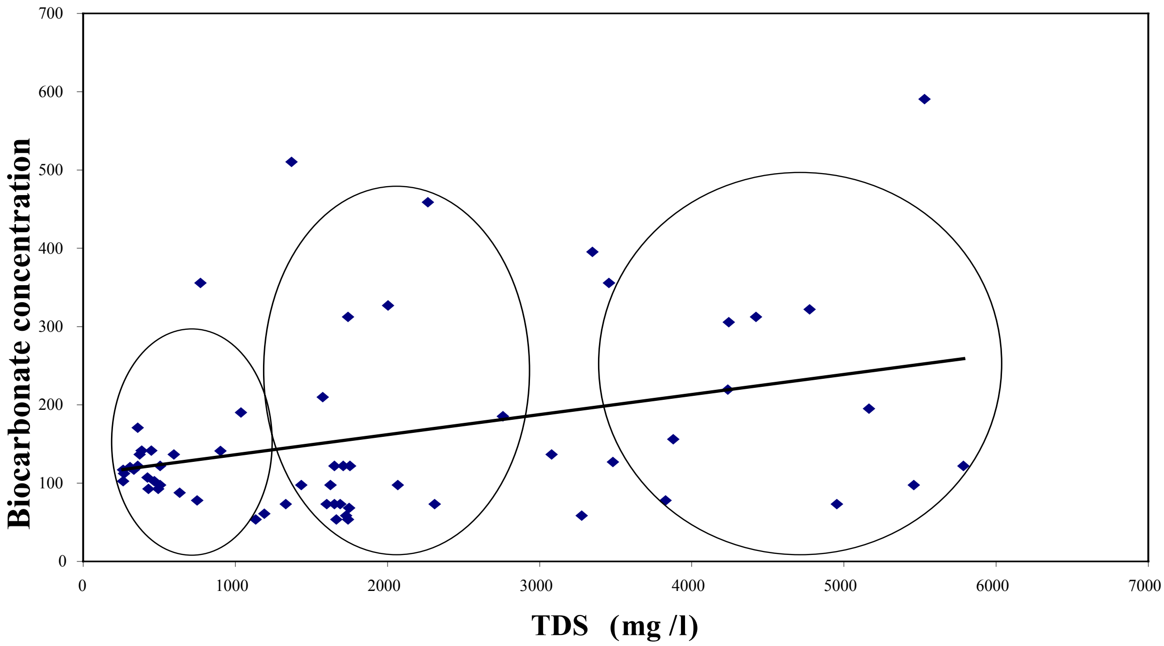

Figure 6. A plot of TDS vs biocarbonate in the study area.

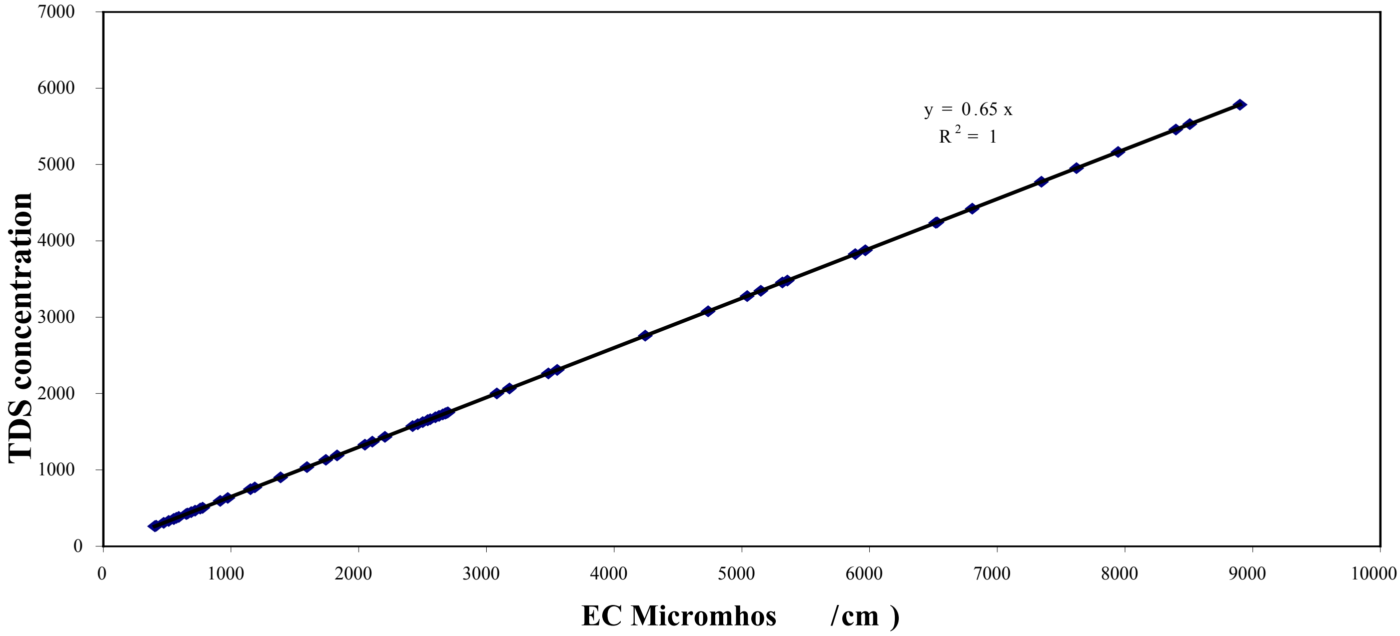

Figure 7. Electric conductivity vs total dissolved solids (TDS) in the study area.

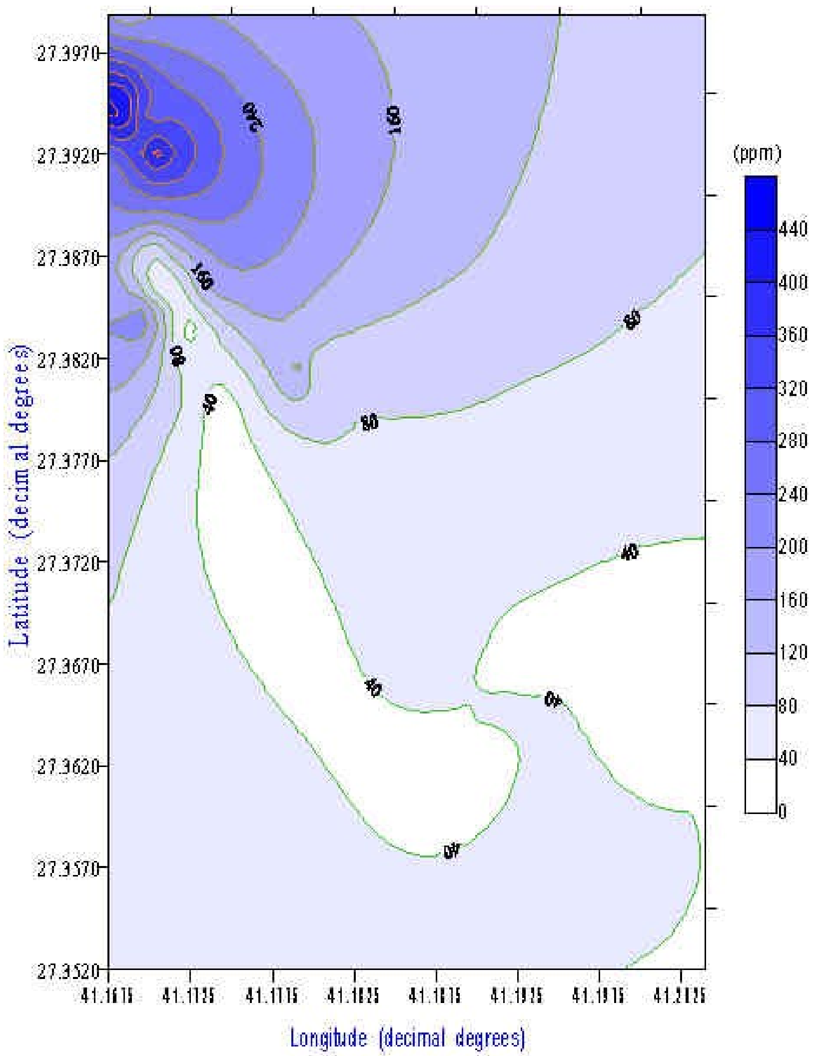

Fig. 8: Nitrate Distribution in Mawqaq, Hail Area(ppm)

Figure 9 presents a pattern of NO_3 /Cl ratio. The nitrate enrichment is a wide variation of wetter and drier periods in the unsaturated soils. Therefore, the mechanism of enrichment of NO_3–N in the system is likely to be due to the presence of leguminous plants or nitrogen fixation of indigenous plants (vegetation). It could come as a loss of nitrogen from the domestic septic tanks systems since there are no applications of fertilizers in the agricultural system or manure from the animals. The latter hypothesis (septic tanks) is more acceptable because the concentration of nitrate as nitrogen in groundwater is relatively very high in the inhabited area while nitrate concentration in the outside of the town (Mq1 and Mq2) is very low.

4.5 Bacteriological Pollution

The significance of the various coliform organisms in Mawqaq groundwater was considered in order to determine the nature of the pollution, detect and estimate the coliform group of bacteria. Almost all the extension of aquifer was not covered for bacteriological sampling but random sampling has been conducted for this matter, especially in the northern part, where some people are utilizing water for various usage. The Directorate General of Health in the Region has sampled about 10 wells and reported the presence of harmful bacteria in some wells. The septic tank systems are parallel with the wells in some areas, especially in the north of the town with slight difference in depth. Aquifer material is mainly channeling deposits, which helps seepage, infiltration, and easy flow in the fractures and faults available in the area.

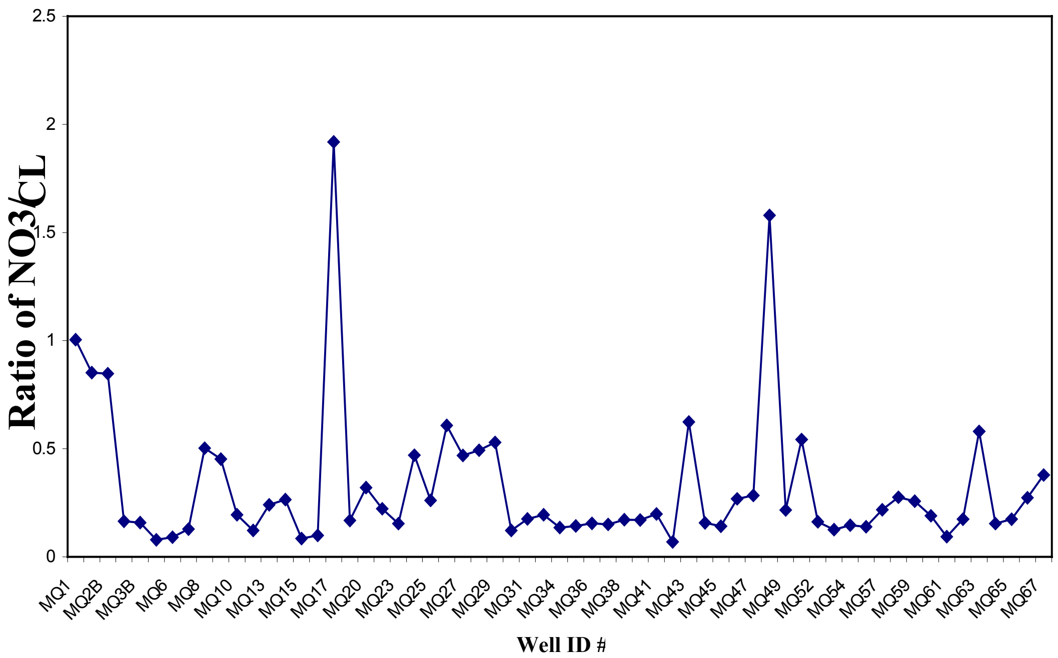

Figure 9. Plot of NO3/CL for Mawqaq groundwater.

The presence of bacteria, which lives in the intestine of human being and animals, is suspected in the groundwater system. The main point source of contaminants in shallow groundwater systems in the area and the growth of bacteria in these systems is mainly attributed to the processes, which occur in the septic systems for household wastewater disposals. The impact of septic tank systems on groundwater contamination and its consequence to the environment have been addressed and disinfection of the wells has been made with constant monitoring of the concerned authorities.

4.6 Isotopic Application

4.6.1 Tritium

Tritium data, which are useful for providing information for modern recharge, have been collected randomly in the study area. The tritium result has shown that the maximum content of tritium in Mawqaq groundwater is about 8.7TU and the minimum is about 1TU and the average is 5.33TU. These values present a picture of a pre-and post-pump replenishment and characterization of old and young recharge in the alluvial aquifer. Figure 10 shows a linear plot of tritium versus TDS and emphasizes the relationship between the salinity and tritium in Mawqaq groundwater. Sample Mq16 is spring water, which flows from the mountains, with low TDS concentration and relatively moderate content of tritium. This plot shows three grouping patterns of samples in relation to the salinity of the area. First group shows low TDS concentration, which represents juvenile water, and relatively high content of tritium. The second group has moderate TDS concentration and relatively low tritium (transitional of two mixing system) while the third group is characterized by very high salinity and moderate tritium content. Thus, this is the condition of Post pump groundwater with the exceptional salinity elevation presence cases. There are rare cases in which fresh groundwater is characterized by high salinity. The attribute to the presence of high saline groundwater is due to the vicinity of the source of sabkha deposits in Mawqaq area.

4.7 Oxygen-18 and Deuterium

Deep groundwater usually reflects the long-term average isotopic composition recharge waters while shallow groundwater reflects more closely short-term isotope variability in precipitation and change in the ratio of winter and summer. Table 1 shows the oxygen-18 and deuterium data for the study area. Figure 11 reflects the pattern of deuterium and Oxygen-18 in Mawqaq groundwater. The picture presents mainly three types of distribution of isotope, which has a relation with the origin of water, and replenishment of groundwater. These groups present a picture of old and young groundwater. These groupings are also mentioned earlier in Figure 10.

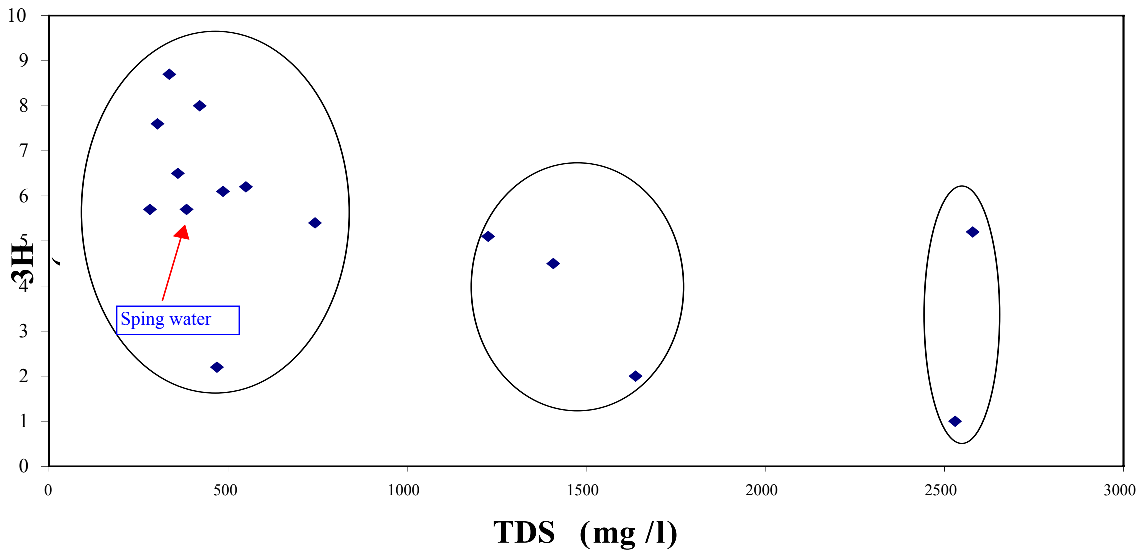

Figure 10. TDS concentration vs tritium content in the study area.

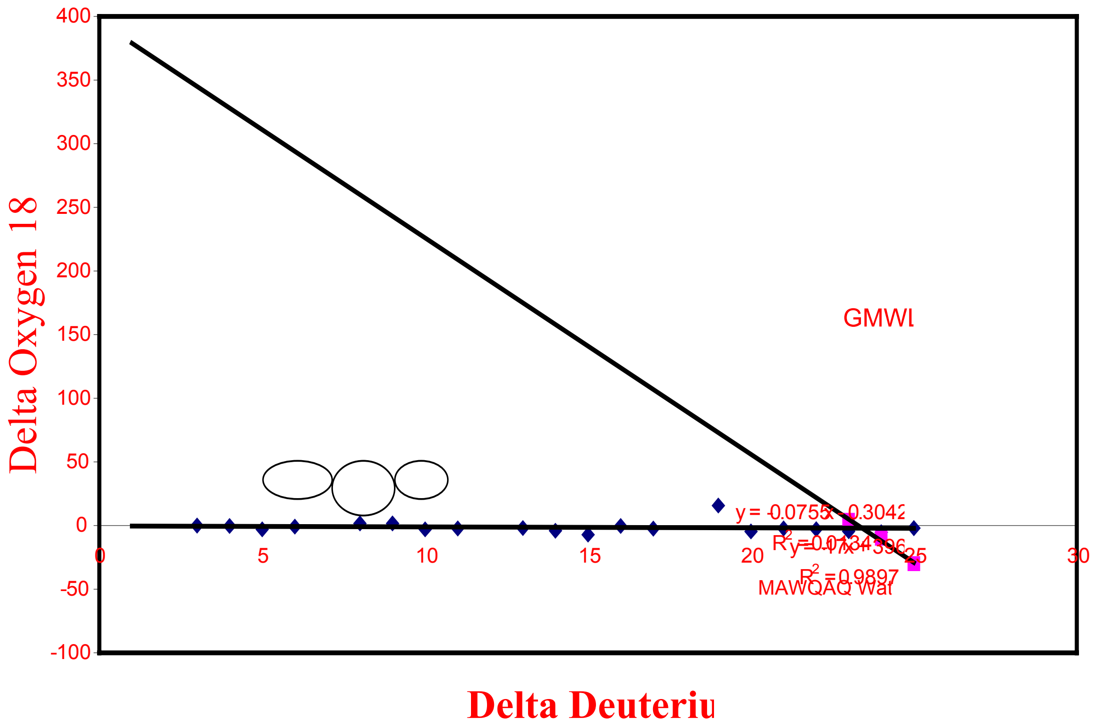

Figure 11. Istopic composition Mawqaq groundwater.

5. Conclusions

This study has revealed progressive groundwater quality deterioration towards the central and northern part of the town (in the alluvial aquifer). Groundwater salinity in the aquifer increases from low to high along the down gradient with maximum TDS (5786 mg/l) is found in the north and minimum TDS (265 mg/l) in the south. Ion relationship sequence, water chemistry and application of stable isotopes have also shown water grouping in the study area. Transitional mixing system was deducted from these results. Structural constraints, improper distribution of water wells and uncontrolled pumping are the other main causes of the aquifer deterioration, especially in the central and northern part.

Presence of high-elevated NO_3-N concentration was encountered in the study area. The existence of hydraulic continuity between septic tank systems and groundwater may be the cause of the elevated nitrate concentration. Some harmful bacteria have also been identified in some wells.

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