Using isotopes for dating and residence time of groundwater in an aquifer, a case study at Al-Najaf, Middle Iraq

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Abstract

Groundwater is the main source of sustaining life in the Najaf Desert that helps sustain human and animals' lives. The aquifer of the Dammam Formation is considered the main source of groundwater in the study area. The isotope elements have been used as a geochemical indicator to determine the age, direction of flow and the effect of rainwater of this area. The values of stable isotopes $\delta^{18}O$ and $\delta^{2}H$ range between -2.56‰ to -1.99‰, and between 7.12‰ to -4.84‰ with an average of -2.32‰ and -6.20‰ in rainfall respectively. Their values range from -3.26‰ to -2.01‰ for $\delta^{18}O$, and from -27.00‰ to -16.21‰ for $\delta^{2}H$ with an average of -2.70‰ and -22.27‰ in the Dammam groundwater respectively. The groundwater is old and does not mix with new water, because tritium has not been not detected. Furthermore, the $\delta^{18}O$ and $\delta^{2}H$ values show that the rainwater, which feeds the aquifer does not come from the continental lands. They represent a humid and cold climate, as well as a considerable amount of rainfall in the past. Analyses of ^{14}C indicate that the groundwater dates back to approximately 4176 years ago. The groundwater is moving from the west and southwest directions to the north and northeast directions.

Keywords: Dammam; groundwater; najaf; radioactive isotopes; stable isotopes.

1. Introduction

The groundwater is studied in Al-Najaf Governorate, middle Iraq where the desert covers most of its territory. The inhabitants of this area live in small towns and villages, and many of them are Bedouins leading a pastoral life. The study area is characterized by the absence of rivers, and during most months of the year there is no surface water except for a few rainy days. As such, the inhabitants of the area rely on groundwater for their whole needs. The study area contains a large reserve and significant quality of groundwater in the Dammam aquifer. It is a wonderful groundwater system which is not deep but is near to the surface (Al-Enezy, 2019 and Aladwani, 2022).

Many hydrological studies use environmental isotopes to provide evidence for the knowledge of water sources, quality, water age, groundwater recharge, and movement of groundwater (Izbicki *et al.*, 1998). Groundwater quality is affected by the interaction of rocks with various water bodies, geochemical evolution, and salinity, as well as polluting processes (Clark & Fritz, 1997, Wilopo & Putra, 2021). Isotopes are some particular elements that have the same atomic number but have a different atomic weight, including hydrogen atoms (^{1}H, ^{2}H, and ^{3}H). This means that they have the same atomic number; yet, they have different mass numbers greater than the atoms of the same chemical elements. This can create a tight bond which requires a high amount of energy to break the nucleus into its constituent nucleons (Kendall & Caldwell, 1998).

This study aims to determine the groundwater dating in the area under scrutiny. The aims can be fulfilled by using radioactive isotopes. Delineation of recharge and discharge zones and the movement paths of groundwater have been performed by identifying the newest groundwater age for each region. Stable isotope analysis of rainwater compositions aims to determine the origin of the air mass in the region. Stable isotope analysis for groundwater is deployed to check out the mixture of the new and old waters. As a result, the stable isotopes are used to determine the origin of salination. The coordinates of the study area are at the latitudes 31°8′43.74″- 31°26′8.38″ and longitudes 44°02′42.33″- 44°30′52.30″ which span approximately over 9630 km^2 (Figure 1).

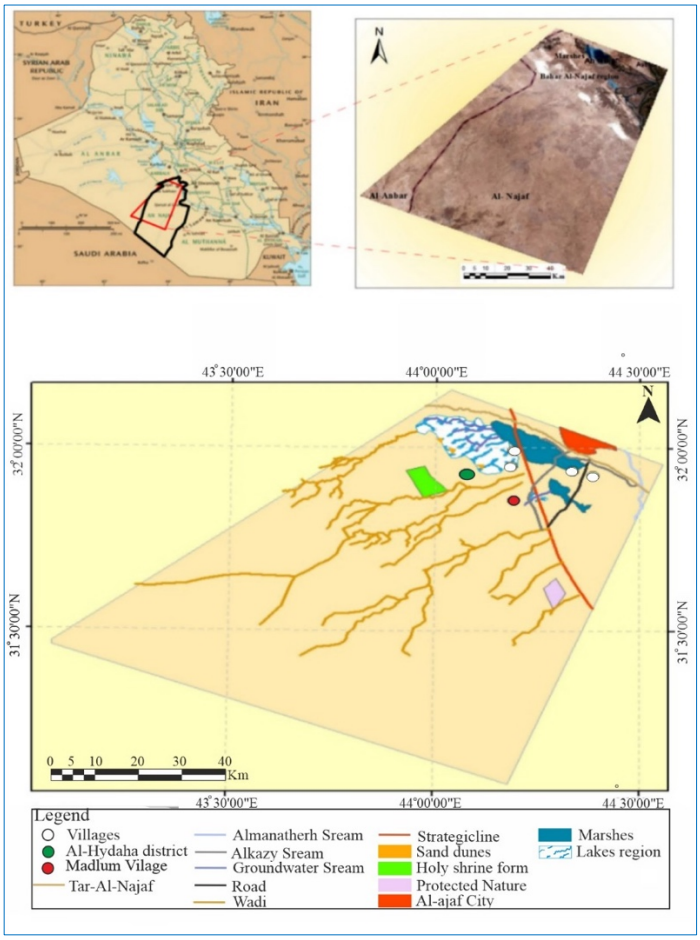

Fig. 1. Location and features map of the study area.

2. Geological setting

2.1 Geological description

The region is located within the boundaries of the Salman Subzone including the stable shelf which is characterized by its simple structures (Buday, 1980). From the newest to the oldest, the stratigraphic sequence arranges over Dibdibba, Injana, Nfayil, Euphrates, and Dammam formations (Al-Owaidi *et al.*, 2021).

Geomorphologically, Bahr-Al-Najaf depression is filled with water and many breeding fish lakes occupy the east side of the study area (Al-Owaidi *et al.*, 2021). A few reliefs are seen to rise gradually from the north and northeast to the south and southwest, approximately over 50m of the sea level for every 10-15 km (Jassim & Goff, 2006). The valleys Haussab, Al-Kharr, and other smaller ones including Al-Rhimawi and Abo Kumssat are subsumed under the study area. The Abu Jir Fault zone, extending to the eastern side, forms the stable shelf boundary. It has played an important role in controlling the groundwater movement in the study area.

2.2 Hydrological setting

Although the studied area is affected by a dry desert climate, yet; rainfall sometimes occurs in spring and summer seasons resulted from a severe intermittent rain once every several years. These rainfall events have caused a shallow flow of rainwater and created many temporary and ephemeral rivers in the desert, which all flood the large valleys downwards. The Dammam aquifer (Dammam Formation) is the main supplier of groundwater in the southern desert of Iraq and in Kuwait as well. (Al-Ruwaih, 1993).

3. Analysis Methods

Six samples of rainfall were collected in the time course of six months (January, February, March, April, May, and June/ 2020) to determine the stable isotopes $\delta^{18}O$ and $\delta^{2}H$ of rainwater. Twelve groundwater samples were tapped from wells (during January 2020), following the procedure of the International Atomic Energy Agency (IAEA) (2009) instructions applied to the analysis of ^{2}H and ^{18}O isotope concentration. The samples were collected in polyethylene (PE) containers (impermeable to light) each of 30 ml capacity. The stable isotope samples were analyzed at UC Davis Stable Isotope Facility, Department of Plant Sciences, on Shields Avenue, Davis, California, 95616, USA.

Three samples were analyzed for Tritium (^{3}H), and other four samples were analyzed for radiocarbon (^{14}C) which have been collected as radioactive isotopes in order to determine the groundwater age. For proper analysis of ^{14}C depending on IAEA, 2.5 gm of carbon must be obtained. Ordinarily, this amount can be precipitated from a total of 120 liters of water samples depending on the alkalinity of groundwater (Geyer *et al.*, 1993). Then, concentrated NaOH was added to raise the pH of the sample to about 11; then about 150 gm of $BaCl_2$ powder was added and the mixture was stirred well for 10 minutes leaving it to precipitate for an hour. Later, 1 liter of distilled water was pulled from the bottom of the container to precipitate. Finally, the samples were analyzed by using a Tri-Carb 3110TR along with low activity liquid scintillation analyzer (LSA) (PerkinElmer, Inc., Waltham, MA, USA), in the laboratory of the Department of Water Analysis/ Ministry of Science and Technology/ Iraq. The sample coordinates are shown in Table 1.

Table 1. Studied wells coordinate

Well No.	Longitude	Latitude	Elevation
W.1	44°15'47.506"E	31°55'13.294"N	6m
W.3	44°15'59.034"E	31°48'50.250"N	22m
W.6	44°18'46.106"E	31°50'30.144"N	21m
W.8	44°20'12.412"E	31°55'14.818"N	8m
W.9	44°14'07.575"E	31°51'15.225"N	30m
W.11	44°14'33.919"E	31°51'50.946"N	29m
W.13	44°16'22.212"E	31°45'34.115"N	39m
W.17	44°12'13.641"E	31°52'13.636"N	35m
W.20	44°08'55.239"E	32°00'46.465"N	17m
W.22	44°05'37.221"E	32°01'56.412"N	23m
W.27	44°05'15.475"E	32°00'25.549"N	29m
W.31	44°08'43.012"E	31°57'07.715"N	32m
W.1	44°15'47.506"E	31°55'13.294"N	6m
W.22	44°06'02.202"E	32°01'08.589"N	21m
W.12	44°14'33.919"E	31°51'50.946"N	25m
W.39	44°5'09.765"E	31°13'00.082"N	26.5m

4. Results

4.1 Stable isotopes

After analyses, the stable isotopes in rainfall water and groundwater samples are described below.

4.1.1 Stable isotopes in rainfall samples

The stable isotope composition ($\delta 1^8O$ and δ^2H) in rainwater indicated that the lowest value was measured in January and the highest value in June. The isotopic composition values were recorded between -2.56‰ and -1.99‰ and between -7.12‰ and -4.84‰ for $\delta^{18}O$ and ^2H with an average of -2.32‰ and -6.3‰ respectively (Table 2).

Table 2. Rainwater values of $\delta^{18}O$ and δ^2H during 2020

Stable Isotope	Jan.	Feb.	Mar.	Apr.	May	Jun	Min.	Max.	Average
δ^2H	-7.12	-7.03	-6.91	-6.89	-4.91	-4.84	-7.12	-4.84	-6.3
$\delta^{18}O$	-2.56	-2.52	-2.45	-2.41	-2.1	-1.99	-2.56	-1.99	-2.32

4.1.1.1 Deuterium-excess

D-excess values are an index that shows the physical-chemical characteristics of water if it is evaporated (Tsujimura *et al.*, 2007). A typical character that shows the origin of air masses from which the precipitation is formed is a deuterium excess. D-excess reflects the prevailing conditions during the evolution and the interaction or the mixing up of air mass with the

locations of precipitations. However, the value d defines a slope of eight and is calculated for any precipitation sample as the equation (1) below can show (Faucher *et al.*, 2020):

$$d = \delta^2 H - 8 \times \delta^{18} O \quad (1)$$

Under conditions of 100% humidity, the vapor will be in isotopic equilibrium with seawater, but when the humidity is lower than 100%, excess deuterium can be found in the rain. Thus, the increase of deuterium is attributed to isotopic exchange, moisture, and lower humidity conditions. The values signal the importance of recycled continental vapors, if they are higher than 100.5 (Table 3) (Clark & Fritz,1997).

Table 3. Values of D-excess in precipitation

Months	d-excess values
January	13.04
February	13.45
Mar	12.69
April	12.39
May	11.89
Jun	11.08
Min	11.08
Max	13.45
Average	12.23

4.1.2 Stable isotopes in groundwater

The stable isotope composition of groundwater samples ranged from -3.26‰ to -2.01‰ for $\delta^{18}O$ and from -27.00‰ to -16.21‰ for $\delta^{2}H$ with an average of -2.70‰ and -22.27‰ respectively (Table 4).

Table 4. Values of $\delta^{18}O$ and $\delta^{2}H$ in the groundwater during 2020

Sample ID	$\delta^{2}H$	$\delta^{18}O$
W.1	-25.65	-3.14
W.3	-21.64	-2.67
W.6	-26.16	-3.14
W.8	-27.00	-3.26
W.9	-25.80	-2.99
W.11	-22.42	-2.70
W.13	-26.47	-3.17
W.17	-25.67	-3.02
W.20	-17.91	-2.22
W.22	-16.21	-2.01
W.27	-17.23	-2.16
W.31	-16.41	-2.10
min	-27.00	-3.26
max	-16.21	-2.01
average	-22.27	-2.70

4.1.2.1 D-excess in the groundwater

Deuterium excess is a measurement of non-equilibrium conditions during the evaporation of water in a relatively humid conditions (Sebastian *et al.*, 2010). Groundwater D-excess values ranged from -1.91‰ to -0.39‰, with an average of -0.66‰ (Table 5).

Table 5. d-excess of groundwater samples

Wells ID	**W.1**	**W.3**	**W.6**	**W.8**	**W.9**	**W.11**	**W.13**	**W.17**	**W.20**	**W.22**	**W.27**	**W.30**
D-excess	-0.5	-0.26	-1.02	-0.92	-1.91	-0.82	-1.13	-1.49	-0.19	-0.13	0.05	0.39
Min.	-1.91					Max.	0.39				Avg.	-0.66

4.1.2.2 Origin salinity

The origin of salinity and mechanisms of groundwater salinization can be determined successfully by deploying stable isotopes (IAEA, 2009). The relationship between $\delta^{18}O$ and Ec can be used to detect groundwater salinity. Salinization originates from the dissolution of the salts. This means that there are no accompanying notable changes in the stable isotopic composition. Yet, the existing of sensitive changes in the stable isotopic composition are due to the presence of the mixing or the evaporating processes (IAEA, 2001).

4.2 Radioactive isotopes in groundwater

Radioactive isotopes ^{3}H and ^{14}C can be used to measure the time and the environmental radionuclides used to estimate the age or the circulation of groundwater (Mook, 2000).

4.2.1 Tritium (^{3}H)

Tritium, the radioisotope of hydrogen (^{3}H), here had been relatively abundant: (0.5×10-5%). Tritium is continual and can be produced in small quantities in the upper atmosphere by the interaction of cosmic rays with gaseous nitrogen and thermonuclear weapons testing (Clark and Fritz, 1997). It can be used to provide information about the residence time and groundwater movement. Tritium has a short-live reaching to no more than 12.32 years, and its concentration is realized as tritium units TU (Fetter, 2001). In this study, tritium had not been detected in the samples (TU= zero) due to its short half-life.

4.2.2 Carbone-14

Carbone-14 exists in several naturally occurring isotopes, ^{12}C, ^{13}C, and ^{14}C. It is formed in the atmosphere by the bombardment of ^{14}N cosmic radiation (Faure & Mensing, 2005). The long half-life, 5730 years, of ^{14}C makes it useful for the late Quaternary period (Godwin, 1962). Carbon-14 activities are expressed as present and modern carbon (pMC): the fractions of CO_2, and dissolved inorganic carbon (DIC). Dissolved organic carbon (DOC) is derived from the living biomass record of ^{14}C activities, which is close to the modern atmospheric level of ~100 (pMC) (Faure & Mensing, 2005).

Radiocarbon dating of DIC in groundwater was based on the radioactive decay of ^{14}C. Rainwater infiltrated the soil whereby the DOC contained much ^{14}C during groundwater

recharge. The carbonic acid dissolved the old soil carbonate devoid of the ^{14}C (DIC). Other physical and chemical processes in the aquifer affected the ^{14}C activity of DIC. The radiocarbon age of DIC must be converted into actual age.

The second step involves the correction of the ^{14}C value controlled by geochemical reactions in the aquifer (Clark & Fritz, 1997).

5. Discussion

Depending on the values of stable isotopes $\delta^{18}O$ and $\delta^{2}H$ for rainwater (Table 2), the equation (2) of the Najaf Meteoric Water Line (NMWL) of the study area was linear

$$\delta^2 H = 4.5\delta^{18}O + 4.32 \quad (2)$$

The slope 4.5 pointed to the enrichment process of isotope composition which was more than the Global Meteoric Water Line (GMWL) due to the fractionation process, where the NMWL could be governed by the local climate factors such as a temporal change in the moisture source and the seasonal variations (Durowoju *et al.*, 2019) (Figure 2).

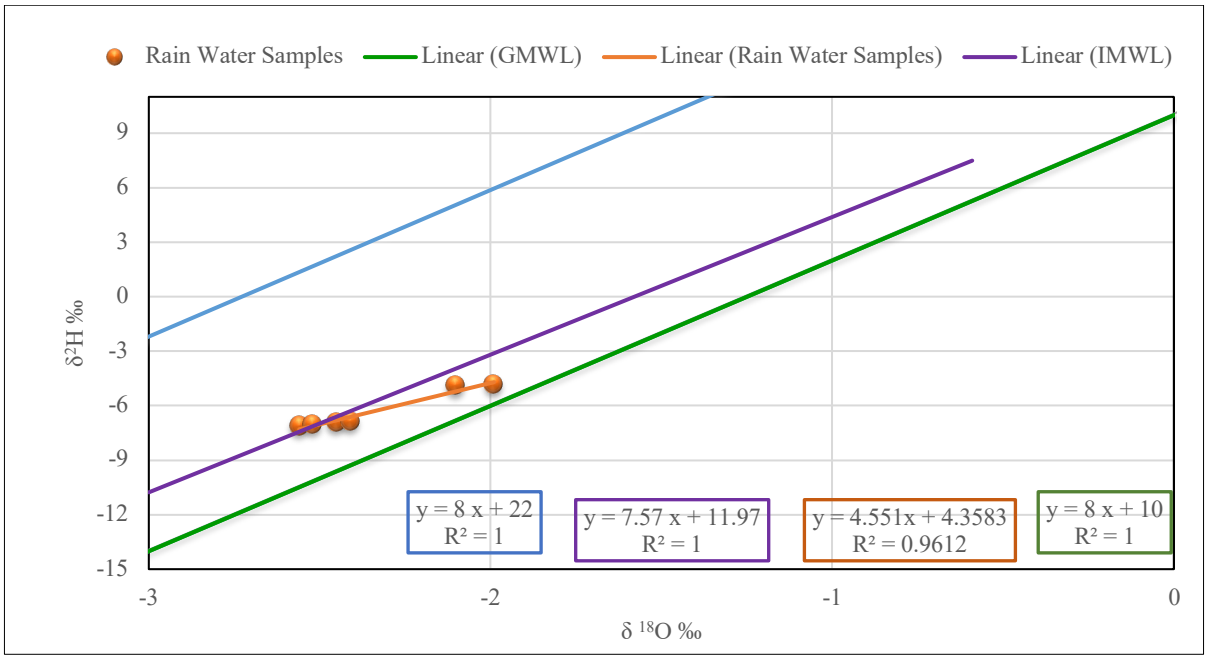

Fig. 2. NMWL of rainwater samples with GMWL, Eastern Mediterranean Meteoric Water Line (EMWL) and Iraqi Meteoric Water Line (IMWL)

The enrichment of stable isotopes can increase along with the increasing temperature degrees (Positive relationship) (Clark & Fritz, 1997). Furthermore, the variations of the values of $\delta^{18}O$ and $\delta^{2}H$ can be attributed to the passage of air mass from the Mediterranean Sea loaded with humidity to affect the line slope (Kattan, 1997). The NMWL was shifted to the GMWL and approached IMWL due to the influence of Mediterranean air masses.

All d-excess values of rainwater were less than 10.5 (Table 3). This pinpointed the source of air masses, which was not coming from the continent lands (Figure 3). The Mediterranean region is characterized by a relatively low humidity and distinctive vapor formation conditions (Gat & Carmi, 1970).

Figures 4 and 5 show the positions and distribution of $\delta^{18}O$ and $\delta^{2}H$ of wells respectively, and the interpolations of enrichment of $\delta^{18}O$ and $\delta^{2}H$ within the study area respectively.

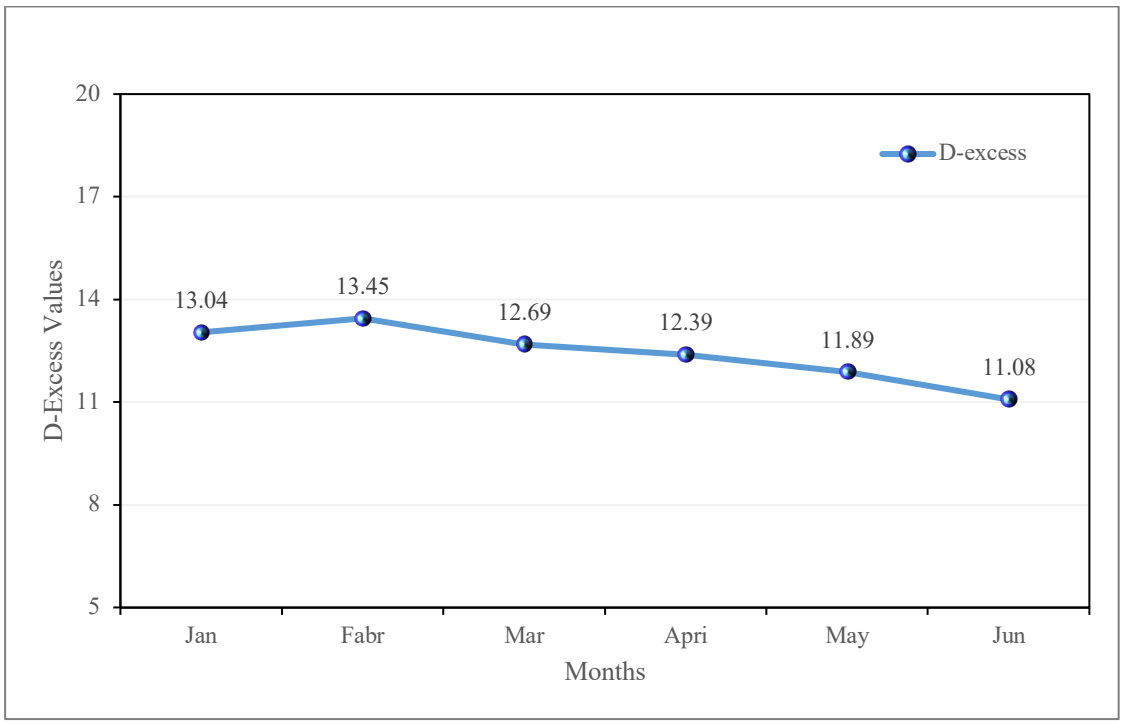

Fig. 3. D-excess variation versus monthly precipitation

Groundwater stable isotope values were compared with GMWL, IMWL and NMWL (Figure 6) where the slope and the regression interception line of groundwater was: $\delta^{2}H = 9.09\ \delta^{18}O + 2.305‰$.

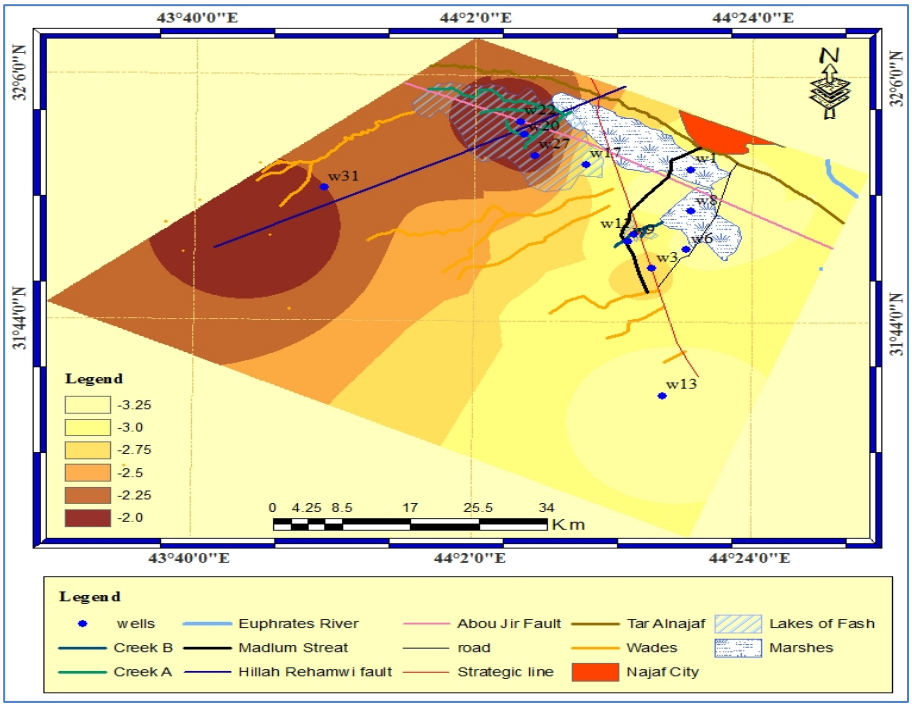

Fig. 4. Stable isotopes wells positions and the interpolations of enrichment for $\delta^{18}O‰$

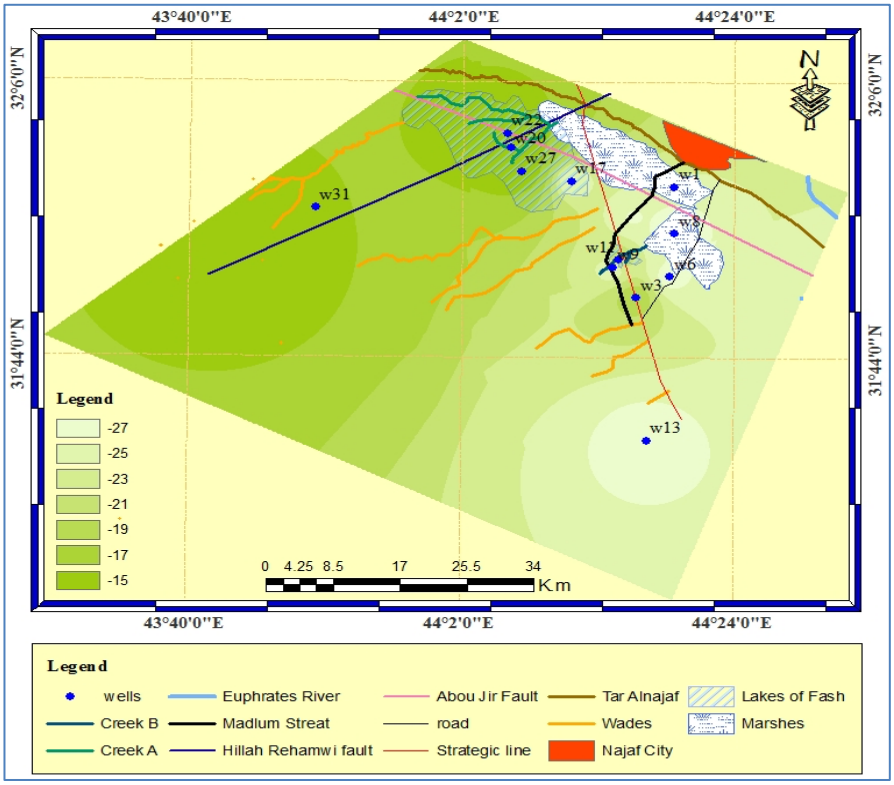

Fig. 5. Stable isotopes and the interpolations of enrichment for δ^2H‰

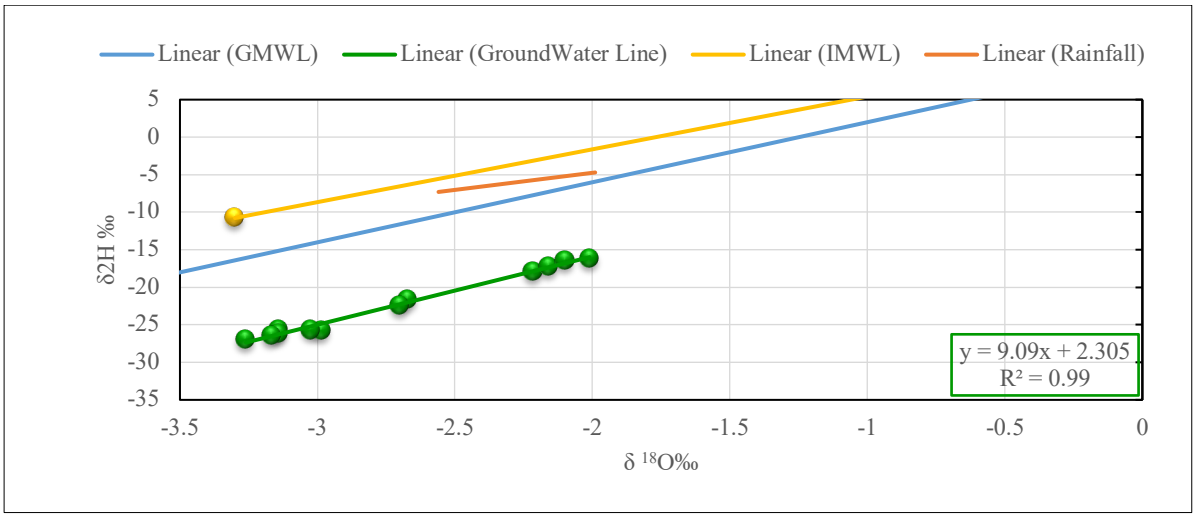

Fig. 6. δ^2H and $\delta^{18}O$ relation of groundwater samples compared to the GMWL and the IMWL

The source and evolution process of groundwater in a specific location can be determined based on the groundwater's relationship with hydrogen and oxygen isotopes in meteoric water (Liu & Yamanaka, 2012).

All groundwater samples were located beneath the three lines IMWL, GMWL and NMWL. This indicated that there was no intermixing between them recently. Consequently, the results suggested that the groundwater came from higher regions. In addition, the $\delta^{18}O$ and δ^2H depletion in the groundwater referred to a humid, cold climate and a high amount of rainfall in the past. Therefore, the values of $\delta^{18}O$ and δ^2H in the groundwater samples were significant as being lower than the values of $\delta^{18}O$ and δ^2H in the rainwater samples due to the natural

evaporation and variation of stable isotope content in the rainwater. Based on the distribution positions of data points of δ^2H and $\delta^{18}O$ diagram, three groups were distinguished from stable isotope components (Table 6 and Figure 7).

Table 6. Mean values of $\delta^{18}O$ and δ^2H for groundwater groups

Groups	$\delta^{18}O$‰			δ^2H‰		
	Min	Max	Average	Min	Max	Average
Group A	-3.26	-2.99	-3.12	-27.00	-25.65	-26.18
Group B	-2.70	-2.67	-2.69	-22.42	-21.64	-22.03
Group C	-2.22	-2.01	-2.12	-17.91	-16.21	-16.98

Group A included six groundwater samples with a ratio of 50%, withdrawn from wells W.1, W.6, W.8, W.9, W.13, W.17 (Figure 8).

Group B contained two samples of groundwater with a ratio of 16.60%, within wells W.3 and W.11. Group C had four samples with a ratio of 33.33%, within wells W.20, W.22, W.27, and W.30. As a result, these groups created a movement of groundwater from the west and southwest to north and northeast affected by the regional Abu-Jir fault. Notable changes between $\delta^{18}O$ and Ec (Figure 9) were detected, where the salinity origin resulted from the dissolution processes. This revealed that a groundwater-confined aquifer did not witness a mixing with modern water recently. As a consequence, this was an indicator of very old age. The age corrections of ^{14}C in the studied samples based on radioactive decay (t) was carried out by using the equation (3) (Kalin, 2000):

$$t = -8267 \times ln\,(at^{14}C/ao^{14}C) \quad (3)$$

Where: $a_t{}^{14}C$: the measured value of (pMC), a_o ^{14}C: initial activity (100%).

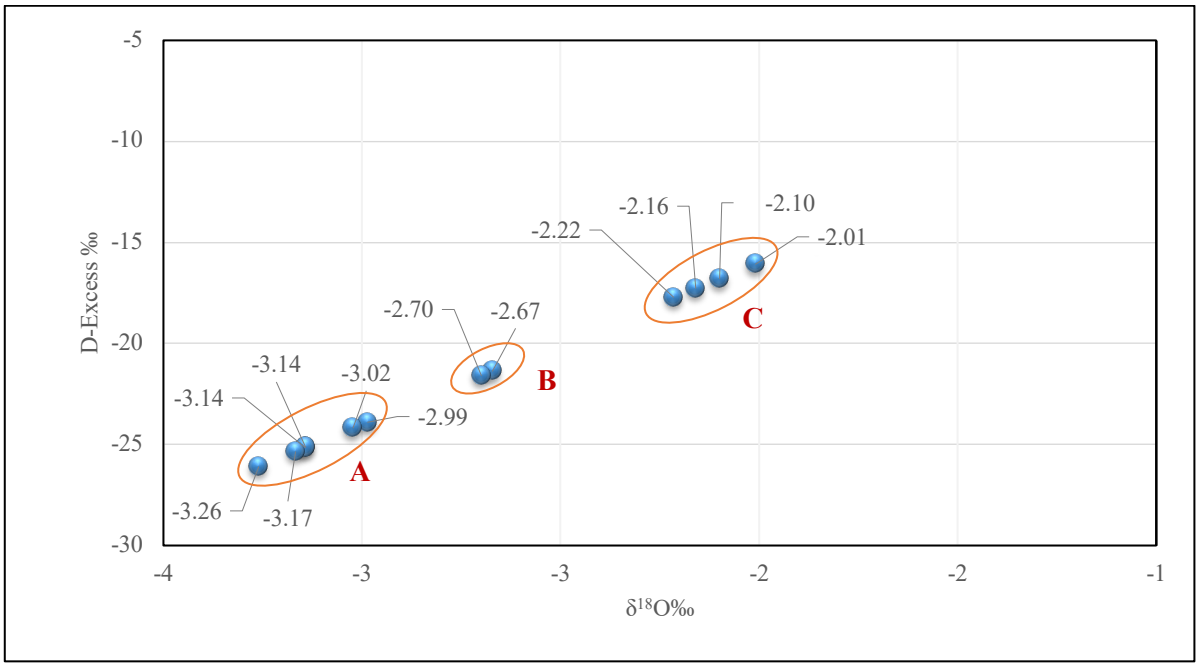

Fig. 7. Groundwater groups depending on $\delta^{18}O$ and δ^2H diagram

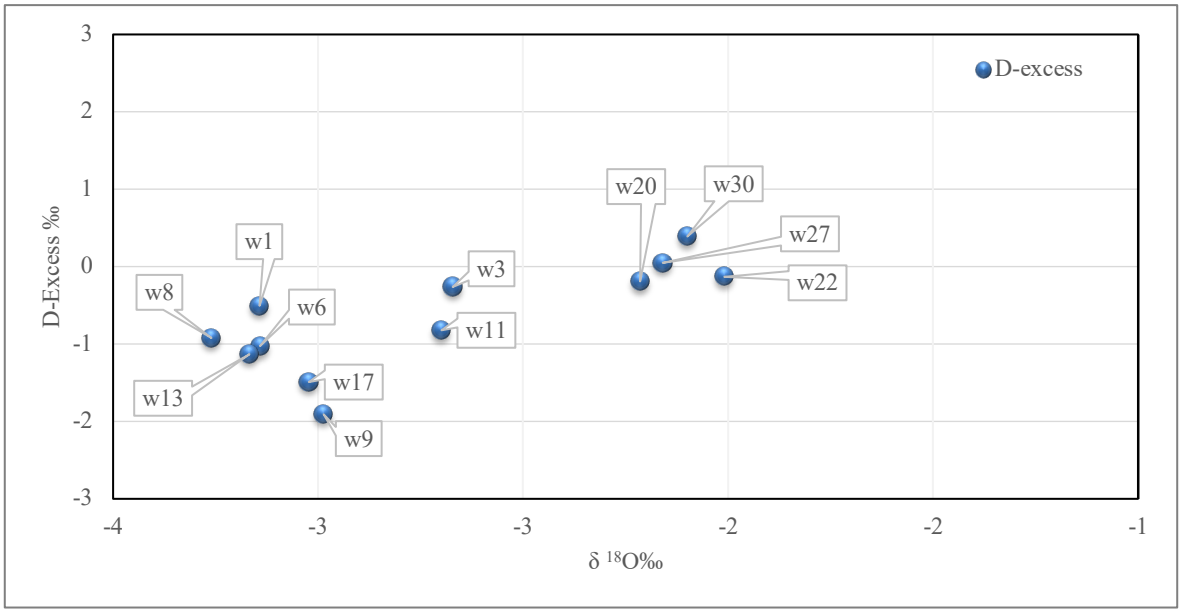

Fig. 8. Relationship between d-excess and $\delta^{18}O‰$ in the groundwater samples

The correction of the $\delta\,^{14}C$ age, and the carbonate dissolutions was fulfilled by using ^{13}C mixing model which must be accounted for to get q (corrections of carbonate dissolution) equation (4) (Pearson & Hanshaw, 1970):

$$q = (\delta^{13}C_{DIC} - \delta^{13}C_{carb})/(\delta^{13}C_{soil} - \delta^{13}C_{carb}) \quad (4)$$

Where: $\delta^{13}C_{DIC}$: measured ^{13}C in the groundwater (DIC). $\delta^{13}C_{soil}$: $\delta^{13}C$ of the soil CO_2 (usually close to -20‰). $\delta^{13}C_{carb}$: $\delta^{13}C$ of the carbonate dissolution (usually close to 0.0‰). The results pointed to the average residence time of groundwater that had come up to 4176 years (Table 7).

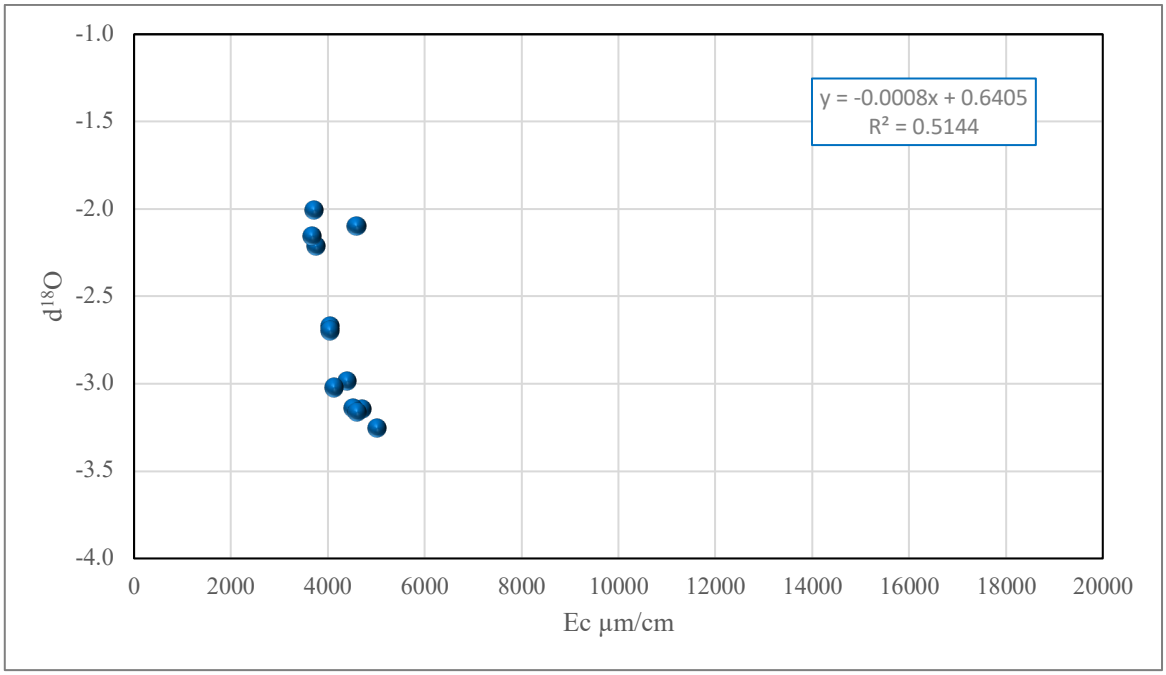

Fig. 9. Relationship between mean δ^{18} O with Ec

Table 7. Groundwater samples ages

No.	Well no.	pMC	t. year
1	W.1	50	5726
2	W.22	70	3603
3	W.12	60	4877
4	W.39	80	2498
Average		-	4176

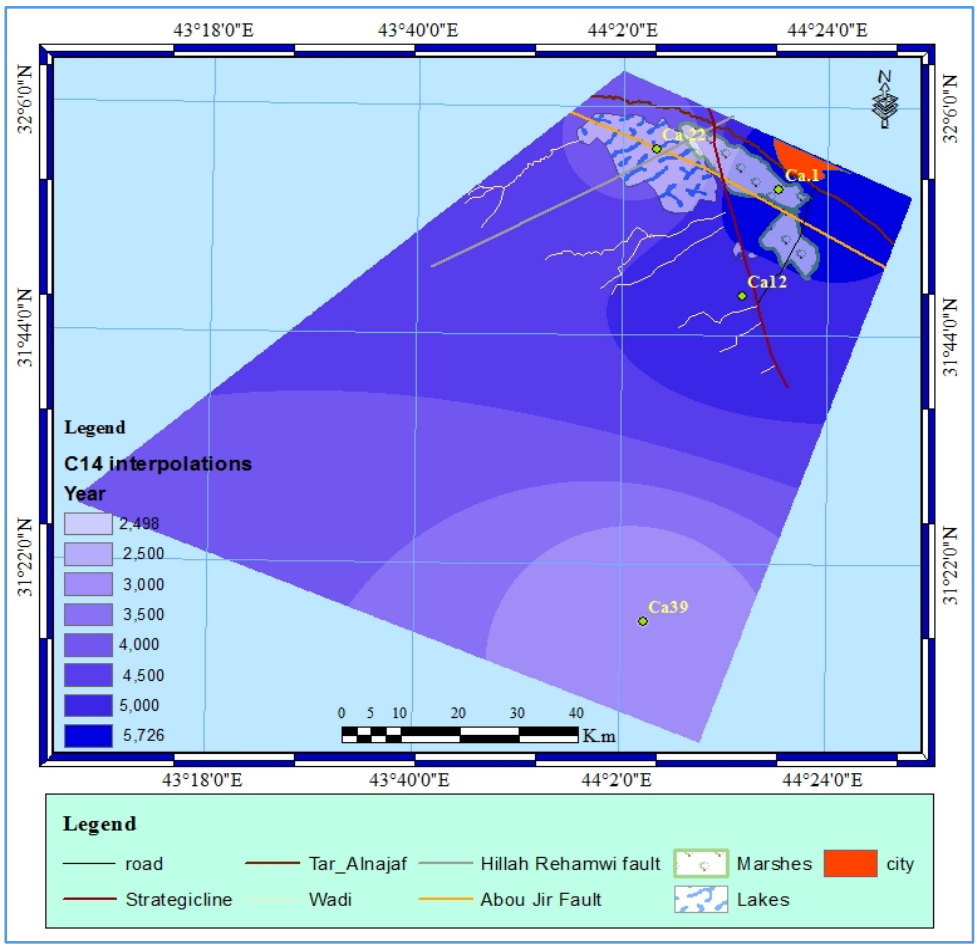

Fig. 10. Groundwater distribution of age

A contrast in groundwater ages was observed, depending on the sites of sampling ^{14}C, where the well W.39 contained a groundwater age reaching to 2498 years. It could have an age of less than the age of other samples in the study area, due to its location at the beginning of recharging area (Figure 10).

Well W.1 contained a ground water aging 5726 years due to its location in a confined aquifer as being far from the recharging area, and because of its location in the west of the Abu-Jir fault. Well W.22 contained a water age of 3602 years, due to its location in the high discharging zone (flowing well with high discharge), being distinguished by fractures facilitating the renewal of the water process where the increased pumping can increase the proportion of modern contribution (Clark & Fritz, 1997).

6. Conclusions

The stable isotope composition of rainwater indicated that the lowest values were measured in January and the highest value was in June. The values were recorded between -2.56‰ and -1.99‰ and between 7.12‰ and -4.84‰ for $\delta^{18}O$ and ^{2}H respectively, with an average of -2.32‰ and -6.20‰ respectively.

The linear equation of the NMWL was $\delta^{2}H= 4.5\ \delta^{18}O+ 4.32$ with a slope of 4.5. It reflected the enrichment process of the isotope composition due to the fractionation process. The NMWL was governed by the local climate factors. The sources of air masses were not coming from continental lands, depending on d-excess values (less than 10.5).

Groundwater stable isotope composition samples of the Dammam aquifer ranged between -3.26‰ and -2.01‰ of $\delta^{18}O$ and between -27.00‰ and -16.21‰ of $\delta^{2}H$ with an average of -2.70‰ and -22.27‰ respectively. The slope and the interception of the regression line of groundwater was $\delta^{2}H= 9.09\delta^{18}O+ 2.305$‰.

All samples of groundwater were located beneath the two lines GMWL and NMWL, and they did not match with the precipitation line NMWL. However, NMWL located above the line of GMWL indicated that there was not an intermixing between them recently. The results indicated that the groundwater was imported from a high elevation above the sea level with a more altitude than its current location. Furthermore, the depletion of $\delta^{18}O$ and $\delta^{2}H$ in the groundwater reflected a humid and cold climate and a high amount of rainfall in the past.

Based on the distribution of data point positions for the $\delta^{2}H$ and $\delta^{18}O$ diagram, three groups were distinguished from the stable isotope components of groundwater. They were: Group A, Group B and Group C. The relationship between $\delta^{18}O$ and Ec showed that the origin of salinity resulted from processes of dissolution.

The age of groundwater was determined via an existing ratio of ^{14}C in the groundwater of the study area. The mean age of groundwater was approximately 4176 years. The groundwater movement reflected that there was a difference in the age of groundwater of many places in the study area.

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