

Hydrochemical Characteristics of Groundwater in Siwa Oasis, Egypt. Part I. Significance of the Situation Groundwater Resources and Future Outlook

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Abstract: *This research, carried out with the objective of developing water management strategies in Siwa Oasis to ensure its sustainable development. Siwa Oasis (1000 km²) is today suffering an excessive rise in the subsoil water levels. The rate of groundwater rises from 1.33 to 4.6 cm/ year. Consequently, the fertile soils are subjected to deterioration and salinization. This is proposing and testing chain water management scenarios that makes use of suitable water (multiple reuses) before it reaches the last disposal point to increase the cultivated area (17000 feddans). So, this paper carried out the hydrogeochemical groundwater study based on the results of the chemical analysis of groundwater samples collected. To determine the evolution of groundwater resources through different analysis of water, such as total dissolved salts (TDS) and total hardness in two aquifers, Nubian sandstone and fractured dolomite limestone. The results show, the salinity of groundwater representing the area changed from fresh to saline groundwater, 168.8 to 7472.8 mg/l and total hardness from soft to hard, 30.7 to 1829.7mg/l as CaCO₃.*

Keywords: Groundwater, Hydrochemical, salinity, Siwa Oasis, water resources

1. Introduction

Groundwater is a very important source of water, especially in arid and semi-arid regions. It is considered as the only source of water in the Eastern and Western Deserts of Egypt. In Africa, groundwater is a vital resource with nearly 80% of the continent's population relying on it as a main source of drinking water. However, in many parts of the continent, reaching basic health requirements is still a concern [1].

Now, the drinking and irrigation water in Egypt is one of the biggest problems. Egypt is endowed with numerous water resources, but an assessment of these resources, including their reliability, quality and sustainability has not been systematically carried out. With the increasing demands for water due to increasing population, urbanization and agricultural expansion, groundwater resources are gaining much attention, particularly in arid and semi-arid regions.

In Egypt, in the recent years a great deal of attention has been directed toward expanding agricultural project developed on groundwater. One of the major regions qualified for this activity was west of the Nile Delta region and the New Valley including Siwa Oasis due to its soil and groundwater potentialities [2]. Siwa Oasis is a natural depression about 18m below sea level. It is located in the northern part of the Western Desert of Egypt (about 90 kilometers east of the Libyan border) and 300 kilometers south of the Mediterranean Sea and occur mostly to the east of Agormy [3]. Groundwater is the only source of drinking and irrigation water in Siwa Oasis. Old artesian wells originated from the top shallow aquifer are the traditional source of irrigation water in Siwa Oasis; the numbers of these wells are 220. As

the need for more land aired, the upper limestone aquifer has been tapped by hand-drilled and boreholes as a new source of irrigation water.

For historical, demographic, and economic reasons as well as its high potential for agricultural development, the Siwa Oasis is considered the most important oasis in the western desert of Egypt. In addition to the existing cultivated area, there are more than 17,000 feddans that were determined to be suitable for agricultural development. The area under cultivation has been gradually increased in recent years as the population of the oasis is on the rise. New land reclamation development projects aimed at the exploitation of water resources have been in progress. Therefore, there is an urgent need for monitoring water quality in Siwa Oasis [4,5].

It is noted that the water quality in Siwa Oasis is deteriorating over time, and there is an urgent need for long term monitoring of water quality of the available water resources in the Siwa Oasis. The current research study was set to do just that. The present paper presents the results of this investigation and the future outlook for the situation of the limited water resources of the oasis. Scientific study concluded that the Siwa Oasis is suffering from high salinity, soil as a result of the exchange, which will reflect negatively on agricultural areas where irrigation water is misused, leading to lower productivity of crops [6,7]. In order to achieve sustainable development in these regions an integrated, complete, and accurate monitoring information system regarding the status of the groundwater quality is indispensable. Groundwater and surface water geochemical studies can provide a better understanding of potential water quality variations due to geology and land use practices [8,9].

In Siwa Oasis the strata of hydrogeological interest are composed of 450-600 m thick of fractured dolomite limestone overlying the Nubian sandstone aquifer. The fractured carbonate zones are separated from the underlying Nubian sandstone by the low permeability shale and clay layer. This layer acts as acting and its thickness varies between 60 in the west and 250 m in the east [10].

The Nubian aquifer has a thickness of about 2600 m and belongs to the Mesozoic and Paleozoic age. The upper zone of the Nubian Sandstone aquifer in Siwa Oasis has a thickness of about 500m and is saturated with fresh water with a salinity less than 500 ppm. The net sand thickness of this aquifer decreases northward and westward. The water pressure head of the Nubian Sandstone aquifer ranges between 80m in the western part of the Siwa area (where shallow wells and springs exist) and 120 meters in the east [11].

The aim of the present work is to study the hydrochemical assessment of the aquifer in Siwa Oasis, Northwestern Desert, Egypt. In continuation of our previous work [12,13] were studied of different localities for water resources quality with monitor overall objectives. In the Siwa Oasis study way to remove unwanted ions by safe and environmentally friendly methods in the second stage in our searches.

2. Experimental Method

Groundwater samples were collected from 24 different locations that cover Siwa Oasis water resources through September 2014. These water samples were collected in clean polyethylene bottles. At the time of sampling, bottles were thoroughly rinsed 2–3 times with groundwater to be sampled. In the case of bore holes and hand pumps, the water samples were collected after pumping for 10 min. This was done to remove groundwater stored in the well. These water samples carried out chemical analysis to assess the water quality. These samples were analyzed at Water Chemistry Department, Desert Research Center (DRC). Laboratory measurements were carried out by an EC meter (Orion model 150A+), pH meter (Jenway model 3510), flame photometer (Jenway model PFP7) and UV/Visible spectrophotometer (Thermo-Spectronic model 300). The analysis includes the determination of the different properties of water, such as TDS (Total Dissolved Salts), Major ions as Na^+ , K^+ , Ca^{2+} , Mg^{2+} , CO_3^{2-} , HCO_3^- , SO_4^{2-} and Cl^- , Table (1). The concentrations of these constituents are expressed in milligrams per liter (mg/l, ppm), milli equivalents per liter (me/l) and percentage (%), [14].

3. Results and Discussion

The study is mainly based on the results of the chemical analysis for the collected groundwater samples (24 samples) in the Siwa Oasis. Most of the samples (79% of total samples) are located in the fracture dolomite limestone aquifer and the rest of the samples (21% of total samples) are in the Nubian sandstone aquifer, Fig. (1).

The study of the groundwater chemistry in Siwa Oasis and its relation to the prevailing hydrogeological and environmental conditions are the main target of the present study. The chemical composition of groundwater in Siwa Oasis is the combined result of the water constituents that passes into the groundwater and reacts with main channels that may modify the water composition, Table (1).

3.1 Chemistry of Ground Water

The chemistry of the groundwater includes the following topics such as:

3.1.1. Water salinity (TDS, Total dissolved solids)

Water salinity is detected from the chemical analysis can be expressed by TDS which is widely used in evaluating water quality and convenient mean used for comparing water sample with another one. Total dissolved solid (TDS) is the sum of all cations and anions which is widely used in evaluating water quality and convenient mean used for comparing water samples Twenty four groundwater samples were collected from the Siwa Oasis area to interpret the variations in total salinity and ion behaviors. The chemical data of groundwater reveal that water salinity of the Siwa Oasis area varies from 168.8 mg/l to 7472.8 mg/l as in Table (2).

According to Chebotarev classification [15], the groundwater water can be classified into three main categories, as follows:

- 1) Fresh groundwater type: fresh groundwater samples lie in the Nubian sandstone aquifer. The low salinity is attributed to low leaching and dissolution processes due to direct recharge from the local rainfall in the catchment area, low evaporation rate where the depth to water (1000 m).
- 2) Brackish groundwater type: the brackish groundwater samples are located in the fractured dolomite limestone aquifer
- 3) Saline groundwater type: the saline groundwater samples are located in the fractured dolomite limestone aquifer

The two types of groundwater brackish and saline are due to the stagnation of the groundwater due to the well abandonment, the shallow depth of the wells which activates the role of evaporation and hence, the concentration of salts, the scarcity of rainfall (45 mm/year) which is considered the main source of the aquifer recharge, the marine origin of the exposed leached sediments dominating the recharge area which are mostly composed of limestone and shale [16].

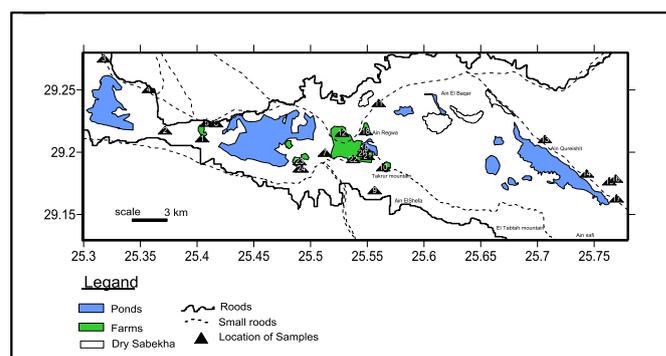


Figure 1: Location map of water samples in Siwa Oasis

Table 1: The hydrochemical results of groundwater samples of Siwa Oasis

well	pH	TDS/ppm	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	CO ₃ ²⁻	HCO ₃ ⁻	SO ₄ ²⁻	Cl ⁻
1	6.9	2226.22	126.4	62.00	600	28	0.00	286.70	295.48	971.09
2	7.5	276.98	21.8	7.04	65	15	0.00	195.20	20.11	50.33
3	7.3	324.49	23.2	11.06	70	21	0.00	201.30	20.50	77.30
4	6.9	2574.69	139.4	82.04	652	40	0.00	170.80	376.77	1199.44
5	6.7	7472.84	317.6	203.64	2097	89	0.00	140.30	704.14	3992.09
6	6.8	3360.29	190.0	84.58	900	34	0.00	170.80	430.54	1635.66
7	6.7	4797.59	184.8	193.91	1300	51	0.00	152.50	600.00	2391.63
8	7.3	3082.96	169.7	92.26	800	38	0.00	109.80	359.96	1568.42
9	7.1	271.08	9.2	7.14	71	19	0.00	128.10	21.53	78.71
10	6.8	168.85	6.3	5.77	40	17	0.00	115.90	14.38	28.18
11	6.7	2148.82	141.1	74.50	523	42	0.00	152.50	356.26	935.73
12	6.8	2327.00	154.0	76.80	560	33	0.00	152.50	340.94	1086.32
13	6.9	170.34	4.0	5.05	47	13	0.00	128.10	13.54	23.97
14	6.8	6229.20	349.8	232.29	1594	97	0.00	195.20	893.68	2964.58
15	6.7	6605.26	315.8	239.21	1700	69	0.00	195.20	971.23	3212.03
16	6.9	6627.23	324.7	240.70	1678	87	0.00	189.10	973.36	3228.72
17	7.5	5606.59	305.0	215.78	1393	64	0.00	158.60	823.23	2726.10
18	7	6287.16	294.0	178.61	1700	55	0.00	158.60	1211.00	2769.25
19	7	3945.58	191.9	118.12	1055	54	0.00	152.50	500.00	1950.00
20	7.1	2724.96	151.9	83.45	700	37	0.00	152.50	384.00	1292.16
21	7.1	2506.00	136.5	72.67	660	35	0.00	152.50	362.12	1163.10
22	7	1794.53	122.0	61.35	420	24	0.00	152.50	333.19	757.92
23	6.9	4130.50	217.2	164.00	1024	46	0.00	158.60	400.00	2200.00
24	7	2537.07	175.2	81.48	604	35	0.00	152.50	345.15	1220.00

Table 2: Frequency distribution of water salinity of the groundwater samples of the studied aquifers in Siwa Oasis

Aquifer type	No of samples	%	Frequency distribution			Water salinity (mg/l)		
			Fresh water (T.D.S <1500 mg/l)	Brackish water (T.D.S 1500-5000 mg/l)	Saline to extremely saline water (T.D.S>5000 mg/l)	Min.	Max.	Avg.
Fractured dolomite limestone aquifer	19	79%	0	54%	25%	1795	7473	4052
Nubian sandstone	5	21%	21%	0	0	168.8	324.5	242.3

3.1.2. Distribution of total water hardness

According to the chemical analysis of the groundwater samples of Nubian sand stone aquifer (fresh water) and Fractured dolomite limestone aquifer (brackish and saline water) it is clear that the mean value of total, temporary and permanent hardness reaches (61.9, 126.1, 0), and (1080.7, 136.2, 944.5) mg/l as CaCO₃, respectively. Noteworthy to mention that the groundwater samples nearly have the same values of total, temporary and permanent hardness also, the temporary hardness relative to total hardness in the fresh water (Nubian sandstone aquifer) is more than that brackish and saline water (fractured dolomite limestone aquifer) and vice versa in permanent hardness relative to total hardness.

The hardness data indicate an increase in total, temporary and permanent hardness with increasing water salinity in all groundwater, according to the change of water type from

fresh to saline water as in Table (3). This is mainly attributed to the effect of leaching and dissolution of soluble salts leading to the increase of hardness with particular importance to the effect of NaCl concentration (effect of ionic strength) on increasing solubility of Ca²⁺ and Mg²⁺ in water [17,18]. This does not exclude the contribution of CO₂, longer residence time, influence of salty water and cation exchange process.

With regard to total, permanent and temporary hardness relative to water salinity (TH/TDS, NCH/TDS and CH/TDS %) in the Nubian sandstone aquifer (fresh water) and Fractured dolomite limestone aquifer (brackish and saline water) samples, the obtained ratios are (25%, 0% and 53%) and (27%, 23% and 4%), respectively in the fresh to highly saline water type.

Table 3: Average and relative values of total, temporary and permanent hardness compared to the water salinity of groundwater.

Nubian sandstone (fresh water)								
TDS mg/l	TH mg/l	CH mg/l	NCH mg/l	(TH/TDS)%	(CH/TDS)%	(CH/TH)%	(NCH/TDS)%	(NCH/TH)%
242.3	61.93	126.1	0	25	53	100	0	0
A. Fractured dolomite limestone aquifer (brackish and saline water)								
TDS mg/l	TH mg/l	CH mg/l	NCH mg/l	(TH/TDS)%	(CH/TDS)%	(CH/TH)%	(NCH/TDS)%	(NCH/TH)%
4051.8	1080.74	136.2	944.58	27	4	15	23	85

3.1.3. Frequency distribution of major ion concentration. The major ions Na⁺, Ca²⁺, Mg²⁺, HCO₃⁻, Cl⁻ and SO₄²⁻ constitute about 98% of total mineralization of most groundwater.

All samples of groundwater chemistry in Siwa Oasis can distribute into histograms as cations and anions for fresh, brackish and saline as in Table (4) and Fig. (2):

Table 4: Range and mean values of the major dissolved ions of groundwater, mg/l.

Ground water	Ca ²⁺	Mg ²⁺	Na ⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ²⁻	
Nubian sand stone aquifer (Fresh water)	4-23.2	5.05-11.06	56-92	115.9-201.3	23.97-78.71	13.54-20.5	Range
	12.9	7.21	76	153.72	51.7	18	Mean
Fractured dolomite limestone (brackish and saline water)	122-349.8	61.35-240.7	444-2185	109.8-286.7	757.92-3992	295.48-1211	Range
	210.9	134.6	1101	165.98	1961.28	561.1	Mean

3.2 Nubian sandstone aquifer (Fresh water type, 21% of the total samples)

The frequency distribution of Ca²⁺ patterns shows three unequal categories; where the majority of fresh groundwater samples (40%) has Ca²⁺ within the range 5-10 mg/l and 20-25 mg/l respectively. However, the mean value for Ca²⁺ of fresh groundwater samples is 12.9 mg/l. The frequency distribution of Mg²⁺ patterns shows three unequal categories; where the majority of fresh groundwater samples (40%) has Mg²⁺ within the range 3-6 mg/l and 6-9 mg/l respectively. However, the mean value for Mg²⁺ of fresh groundwater samples is 7.21 mg/l. The frequency distribution of (Na⁺ and K⁺) patterns shows three categories; where the majority of fresh groundwater samples (40%) has (Na⁺ and K⁺) within the range 40-60 mg/l and 80-100 mg/l. However, the mean value for (Na⁺ and K⁺) of groundwater samples is 76 mg/l.

The frequency distribution of (Cl⁻) patterns show four categories; where the majority of fresh groundwater samples (40%) has (Cl⁻) within the range 20-40, 60-80 mg/l. However, the mean value for (Cl⁻) in fresh groundwater samples is 51.7 mg/l.

The (HCO₃⁻ and CO₃²⁻) distributions show four unequal categories. The majority of samples (40%) have (HCO₃⁻ and CO₃²⁻) concentration ranges between 120 to 160 mg/l, the mean value for (HCO₃⁻ and CO₃²⁻) in fresh groundwater samples is 153.72mg/l. The (SO₄²⁻) distributions show two unequal categories from 13.54 to 20.5 mg/l. The majority of samples have the concentration range between 20-25mg/l (60% of the total fresh samples), the mean value for (SO₄²⁻) in fresh groundwater samples is 18mg/l.

3.3 Fractured dolomite limestone aquifer (brackish and saline water, 79% of the total samples)

From the distribution histograms as in Table (5) and Fig. (3), the following are the main findings. The frequency distribution of Ca²⁺ patterns shows three unequal categories; where the majority of groundwater samples (47%) have Ca²⁺

within the range 150-300 mg/l. However, the mean value for Ca²⁺ of groundwater samples is 210.9 mg/l. The frequency distribution of Mg²⁺ patterns shows four unequal categories; where the majority of groundwater samples (53%) have Mg²⁺ within the range 50-100 mg/l. However, the mean value for Mg²⁺ of groundwater samples is 134.6 mg/l. The frequency distribution of (Na⁺ and K⁺) patterns shows five categories; where the majority of groundwater samples (47%) have (Na⁺ and K⁺) within the range 400-800 mg/l. However, the mean value for (Na⁺ and K⁺) of groundwater samples is 1101 mg/l.

The frequency distribution of (Cl⁻) patterns show five categories; where the majority of groundwater samples (42%) has (Cl⁻) within the range 750-15000mg/l, (21.37-34.4 me/l). However, the mean value for (Cl⁻) of groundwater samples is 1961.28 mg/l. The (HCO₃⁻ and CO₃²⁻) distributions show four unequal categories, the majority of samples (53%) have (HCO₃⁻ and CO₃²⁻) concentration ranges between 120 to 160 mg/l, the mean value for (HCO₃⁻ and CO₃²⁻) of groundwater samples is 165.98 mg/l. The (SO₄²⁻) distributions show four unequal categories from 295.5 to 1211 mg/l. The majority of samples has the concentration range between 350-700 mg/l (47% of the samples), the mean value for (SO₄²⁻) in brackish groundwater samples is 561.1mg/l.

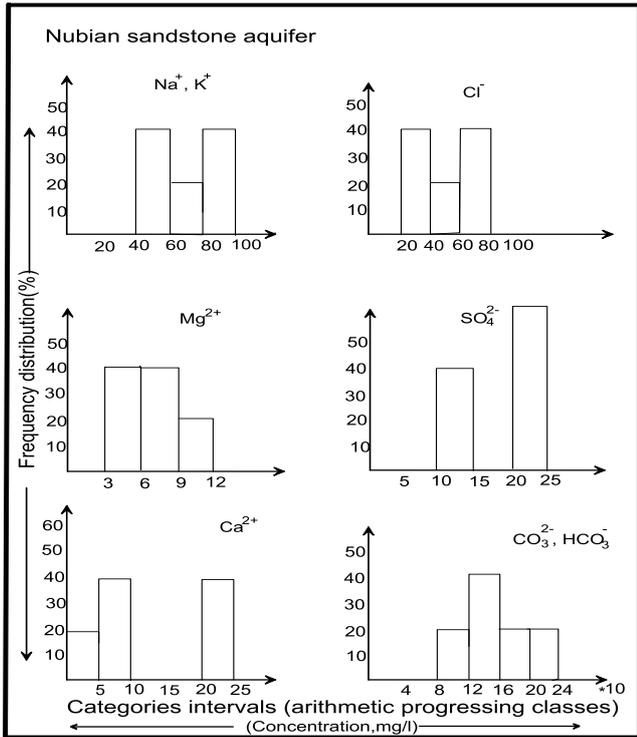


Figure 2: Frequency distribution histograms of major ions of fresh ground water samples, Siwa Oasis

3.4 Ion dominance

Nubian sandstone aquifer

The mean percent for cations and anions of Nubian sandstone aquifer (fresh water) of Siwa Oasis samples compared with those of both rain and sea waters, it is clear that the mineralization of cations and anions are of mixed sources where the meteoric origin (rain fall) of such aquifer. The obtained data show that the effective ions that cause an increase of water salinity are in a descending order: As cations; $\text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+}$ (40%) and $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+}$ (60%) in fresh water. While anions $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-}$ (80%) and $\text{Cl}^- > \text{HCO}_3^- > \text{SO}_4^{2-}$ (20%) in fresh water. The stage, $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-}$ reflects the effect of direct recharge by fresh water from rainwater.

3.5. Fractured dolomite limestone aquifer

The mean percent for cations and anions of Fractured dolomite limestone aquifer (brackish and saline waters) of Siwa Oasis samples compared with those of both rain and sea waters, it is clear that the mineralization of cations and anions are of mixed sources where the meteoric origin (rain fall) of such aquifer is affected by continental and marine conditions.

Thus, leaching and dissolution processes of terrestrial and marine salts in the aquifer matrix play an important role in water mineralization. From the arithmetic mean values of different ions that predict the most effective ions affecting the salinity level, the obtained data show that the effective ions that cause an increase of water salinity are in a descending order: As cations; $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+}$ (53%) and $\text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+}$ (47%). While anions $\text{Cl}^- > \text{SO}_4^{2-} > \text{HCO}_3^-$ (100%) in both brackish and saline to highly saline waters. The stage Cl^-

$> \text{SO}_4^{2-} > \text{HCO}_3^-$ reflects the contribution of leaching and dissolution of marine salts.

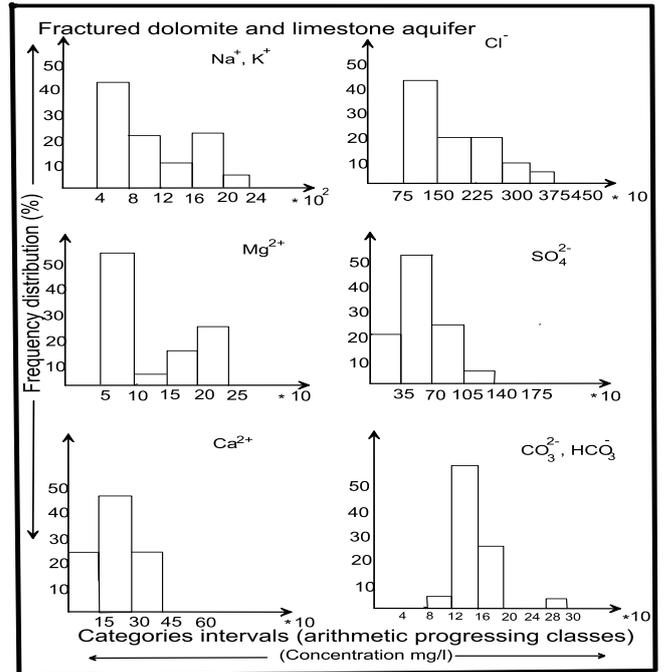


Figure 3: Frequency distribution histograms of major ions in Brackish water, Siwa Oasis.

3.6 Stiff diagram

The cation and anion concentrations are connected to form an asymmetric polygon known as a stiff diagram, where the size is a relative indication of the dissolved-solids concentration [19]. Fig. (4) reflects that ground water in the study area can be represented and divided into three main groups as follows:

- Group I: Samples characterized by Na-HCO₃ water type. Such type is represented (17%) all water samples as Nubian sandstone aquifer (fresh water) and is an indication of rainwater origin of groundwater.
- Group II: Samples of Na-Cl water type (83% of total samples) represented by all samples of dolomite and limestone aquifer with one sample from Nubian sandstone aquifer this indicates that the ground water affected by marine deposits.

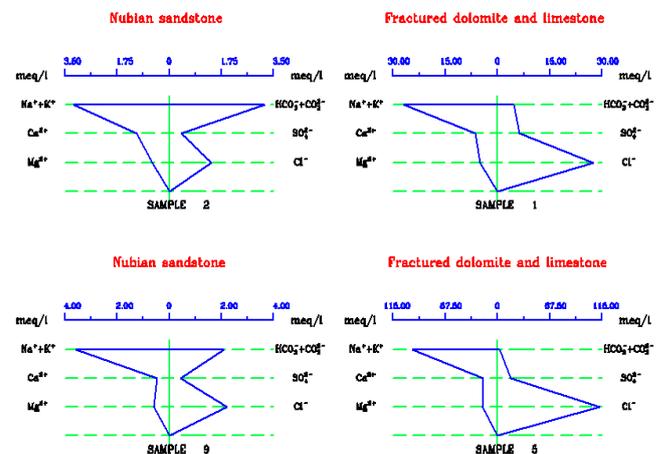


Figure 4: Stiff classification diagrams of four samples represent groundwater types.

3.7. The hydrochemical coefficients (ion ratios)

The relationships between the different major components could be studied through the determination of the ionic ratios. The expression of the ionic relationship in terms of mathematical ratios is quite helpful in establishing chemical similarities among the waters. Ions ratios expressed as me/l is also useful in detecting groundwater contamination (pollution) or mixing with other water resources. The ion ratios are important for possible determination of the origin of groundwater and the different processes contributing to water quality development. The significance of these ratios in

the concerned area, as in Table (6), can be evaluated as follows:

3.7.1. Sodium-chloride ratio (rNa^+/rCl^-)

The variation in (rNa^+/rCl^-) ratio is used for differentiating fresh and saline water. The values of this ratio are always higher than unity in fresh and meteoric water, while it is less than unity in sea water or saline water. All Nubian sandstone aquifer (fresh water) samples have (rNa^+/rCl^-) values greater than unity ($Na^+ > Cl^-$), where the (rNa^+/rCl^-) ratio ranges from (1.62 to 3.5) with a mean value 2.35 as in Table (5).

Table 5: Ranges and mean values of hydrochemical ratios of the different groundwater samples, Siwa Oasis in Egypt.

Hydrochemical Ratios	Nubian sandstone aquifer (Fresh water)		Fractured dolomite limestone aquifer (Brackish and Saline waters)		Rain water	Sea water
	rang	mean	rang	mean	mean	mean
rNa^+/rCl^-	1.62-3.5	2.35	0.74-0.98	0.86	0.65	0.84
rSO_4^{2-}/rCl^-	0.20-0.42	0.3	0.13-.032	0.22	0.86	0.1
$rCl^-/r(HCO_3^-+CO_3^{2-})$	0.3-1.1	0.6	5.8-48.9	20.9	0.65	189.9
rCa^{2+}/rMg^{2+}	0.47-1.88	1.02	0.58-1.36	1.03	7.22	0.17

The increasing ratio in Nubian sandstone aquifer of Siwa Oasis may be attributed to the cation exchange process and leaching of terrestrial salts, groundwater samples reflect meteoric water recharges, this refers to this group of water samples (21%) of total samples are fresh and meteoric waters. While in Fractured dolomite limestone aquifer (brackish and saline waters) less than unity ($Cl^- > Na^+$) where NaCl represent the effects of marine deposits [20]. The increasing Na^+ ions may have theoretically originated by dissolution of sodium bearing silicates from the country rocks. The mean values of rNa^+/rCl^- increase as the total salinity increases. This is due to the increases of soluble NaCl and Na_2SO_4 salts as total salinity increases. From these values of rNa^+/rCl^- one can conclude that 79% of groundwater samples of the concerned aquifer are affected by marine deposits (marine facies groundwater type) and the ratios of Fractured dolomite limestone aquifer (brackish and saline waters) ranges from (0.74-0.98) with a mean value (0.86) as in Table (5).

3.7.2. Sulfate -chloride ratio (rSO_4^{2-}/rCl^-)

Sulfate-chloride ratio could be taken as a guide for detecting any excess of sulphate in the surface water due to ($CaSO_4$) dissolution or ($CaCO_3$) precipitation. The solubility of sulfate mineral components gradually increased in the hydrochemical composition by depth and distance from the recharge to discharge area.

Consequently, the percolation of meteoric water and the greater distance of recharge area are the main factors of increasing the sulfate concentration by direct action of leaching processes. With respect to the groundwater samples collected from Siwa Oasis water have the values of (rSO_4^{2-}/rCl^-) of Nubian sandstone aquifer (fresh water) samples ranges from (0.2 to 0.42) with a mean value (0.3) and the Fractured dolomite limestone aquifer (brackish and saline waters) have the values of rSO_4^{2-}/rCl^- ranges from (0.13 to 0.32) with a mean value (0.22).

All ground water of Siwa Oasis samples has values of rSO_4^{2-}/rCl^- more than that in seawater (0.1). This indicates the solution of a local terrestrial source of sulfate such as gypsum, anhydrite, glauberite ($Na_2SO_4 \cdot 10H_2O$) and epsomite ($MgSO_4 \cdot 7H_2O$).

The non-cyclic sulfate (NCS) was calculated from this equation [21].

$$NCS = (Cl/7.2) - (SO_4^{2-})$$

Where the ions will be taken in mg/l, the data show that all groundwater samples have been negative values of NCS indicates that the sulfate is terrestrial origin.

3.7.3. Chloride-Carbonate and bicarbonate ratio $rCl^-/r(HCO_3^-+CO_3^{2-})$:

The Nubian sandstone aquifer (fresh water) samples of Siwa Oasis have values of $rCl^-/r(HCO_3^-+CO_3^{2-})$ ranges from (0.3 to 1.1) with a mean value (0.6) indicating normal good water [22]. While the values of Fractured dolomite limestone aquifer (brackish and saline waters) ranges from (5.8-48.9) with a mean value (20.9) closed to sea water.

3.7.4. Calcium/magnesium ratio (rCa^{2+}/rMg^{2+})

The ratio rCa^{2+}/rMg^{2+} gives certain indications with regard to the nature of the carbonate aquifer. When this ratio has a value close unity, it is concluded that groundwater is flowing entirely through dolomite terrain. However, when the rCa^{2+}/rMg^{2+} ratio is equal to or greater than four the groundwater is likely have flowed in pure limestone [23]. The values of (rCa^{2+}/rMg^{2+}) for Nubian sandstone and fractured dolomite limestone aquifers range from (0.47 to 1.88) and (0.58 to 1.36) with the main values (1.02 and 1.03) respectively. These values are more than seawater (0.19me/l) and less than the rainwater (7.14me/l). This is due to the presence of $CaCO_3$ materials (aragonite, calcite and dolomite) and gypsum within a dominant aquifer material leading to CO_2-CaCO_3 , $CO_2-CaMg(CO_3)_2$ interactions supported by weathering of dolomite, limestone and gypsum deposits.

3.8 The assemblages of hypothetical salt combinations

Hypothetically, the ions of the strong acids (Cl^- and SO_4^{2-}) form chemical combination with alkalis (Na^+ and K^+) and the rest of the acid radicals combine with the alkaline earths (Ca^{2+} and Mg^{2+}). If the cations of alkalis and alkaline earths are surplus in the water, they will combine with the weak acids (CO_3^{2-} and HCO_3^-). To clarify such combinations, the relations between cations and anions in the investigated waters, are illustrated in the form of bar graphs [24].

In this method of representation, the concentrations of dissolved cations and anions, expressed in me/l, are represented by a vertical bar graph, the height of which is equivalent to the total concentration of either cations or anions. Each bar is subdivided into two longitudinal halves; the cations are plotted on the left half while the anions are plotted on the right one. Vertically, each bar is subdivided into several sections representing the present concentrations of the major cations and anions.

The deduced salt combinations of qualitative and quantitative bases are appended. The combination between major anions and cations reveals the formation of three main groups of hypothetical salts combinations in the groundwater, Table (6) and Figs. (5,6).

In the groundwater, three main groups (assemblages) of salt combinations are distinguished as [24]:

(I) NaCl , Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$ and $\text{Ca}(\text{HCO}_3)_2$.

(II) NaCl , MgCl_2 , MgSO_4 , CaSO_4 and $\text{Ca}(\text{HCO}_3)_2$.

(III) NaCl , MgCl_2 , CaCl_2 , CaSO_4 and $\text{Ca}(\text{HCO}_3)_2$.

Regarding the hypothetical salt combination of Siwa Oasis water samples. Two main assemblages is detected: Assemblage (I) in Nubian sandstone aquifer, five samples represent (21%), as in Table (4) and Fig (7). The presence of Na_2SO_4 salt in this assemblage is a true indication of dissolution of terrestrial salts from continental deposits. Also, the presence of NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$ and $\text{Ca}(\text{HCO}_3)_2$ salts indicates the origin of a meteoric water. Therefore, such water acquires its chemical composition from leaching and the dissolution of terrestrial salt contents, another main assemblage detected in Siwa Oasis samples is assembled (II) in eighteen water samples of the fractured dolomite limestone aquifer, representatives (75%). The assemblage (II) (intermediate stage of chemical development), where two chlorides and two sulfates salts are found. This is affected by leaching and the dissolution of chlorides and sulphate minerals from channel matrices.

The third assemblage detected is: assemblage (III) present one sample of the fractured dolomite limestone aquifer, representatives (4%), as in Table (7) and Fig (6). The presence of three chlorides represents a more advanced stage of chemical development. In general, graduation in chemical evolution starts from dominant HCO_3^- salts (I) which changed to dominant Cl^- and SO_4^{2-} salts (II) and ended by dominant chloride salts (III).

Table 6: Assemblages of the hypothetical salt combinations of Siwa Oasis samples, Egypt

Hypothetical salt combinations	Assemblage	No of Samples	%	Type of water
I- NaCl , Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$ and $\text{Ca}(\text{HCO}_3)_2$.	I	5	21	Nubiansandstoneaquifer (Freshwater)
II- NaCl , MgCl_2 , MgSO_4 , CaSO_4 and $\text{Ca}(\text{HCO}_3)_2$.	II	18	75	Fractured dolomite limestone aquifer (Brackish and saline waters)
(III)- NaCl , MgCl_2 , CaCl_2 , CaSO_4 and $\text{Ca}(\text{HCO}_3)_2$.	III	1	4	Fractured dolomite limestone aquifer (Brackish water)

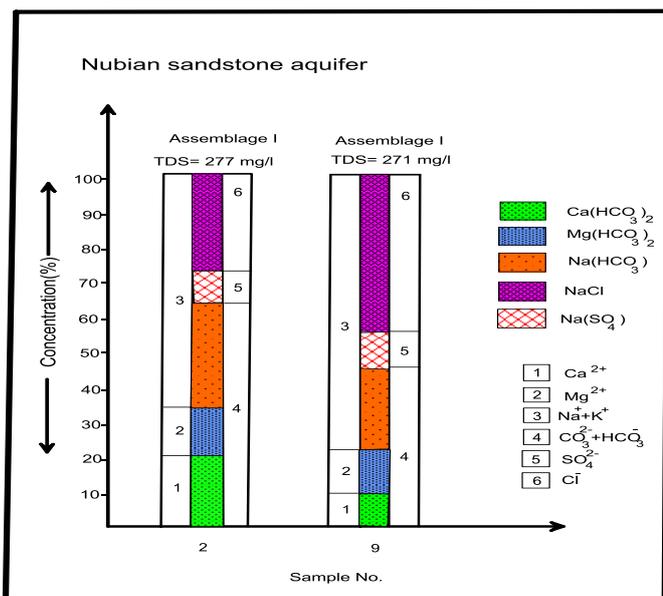


Figure 5: Bar graphs representing the Nubian sandstone aquifer of Siwa Oasis samples

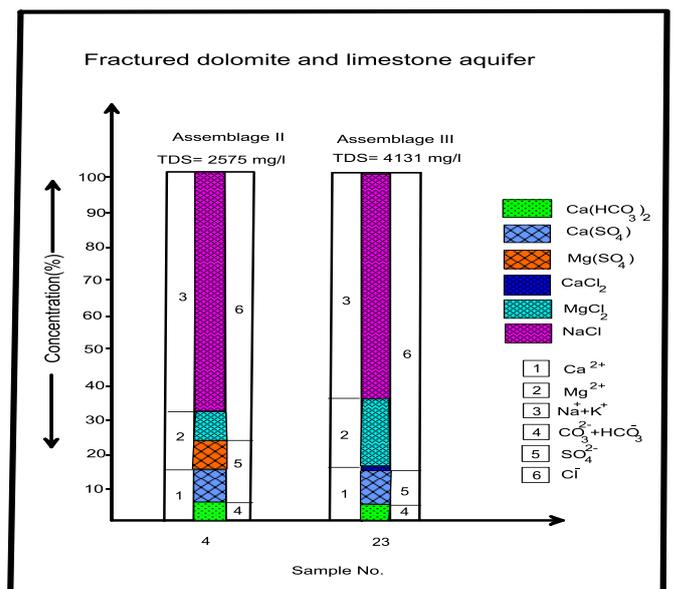


Figure 6: Bar graphs representing the Fractured dolomite limestone aquifer of Siwa Oasis samples

3.9. Genesis and mineralization of ground water

Piper's Diagram

Piper diagram [25] as in Fig. (7), provides a convenient method to classify and compare water types based on the ionic composition of different water samples. Piper trilinear diagram shows that the groundwater is characterized by Na-Cl (83% of total samples) type. These results reveal the relatively polluted nature of groundwater recharged from surface runoff through leaching and dissolution processes. The rest of the samples (17%) are characterized by Na-HCO₃ type. This indicates that the origin of water is meteoric water.

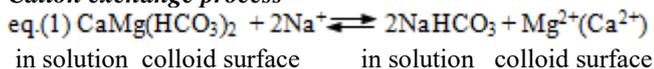
The chemical data are plotted on Piper's diagram of Fractured dolomite limestone aquifer (brackish and saline water) of Siwa Oasis samples are located in sub-area 7. Non-carbonate alkali "Primary salinity" exceeds 50 percent, that is, chemical properties of the water are dominated by alkalies and strong acids. While all samples of Nubian sandstone aquifer (fresh water) are located in sub-area 9, one cation-anion pair exceeds 50 percent.

3.10. Geochemical evolution of ground water in Siwa Oasis

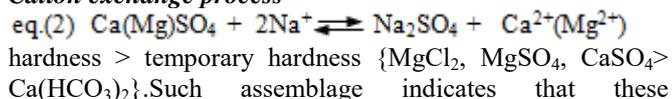
3.10.1. Cation Exchange processes.

It is evident that the groundwater types, affected by leaching and the dissolution of terrestrial salts (assemblage of hypothetical salt combinations I, Nubian sandstone aquifer), are possibly accompanied by a cation exchange process. This process is related to clay minerals where the aquifer matrix is composed of loose pebbles, cobbles and gravels mixed together with fine sand, clay, silt and weathered limestone. The cation exchange process leads to an increase in the water salinity, temporary hardness; equations (1 and 2) as follows:

Cation exchange process



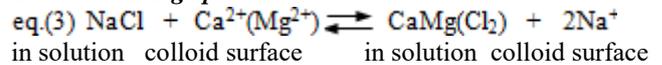
Cation exchange process



in solution colloid surface in solution colloid surface

On the other hand, the groundwater types affected by leaching and dissolution of marine salts (assemblages of marine salts combination II and III, Fractured dolomite limestone aquifer) are possibly accompanied by cation exchange processes that lead to decrease in the water salinity and permanent hardness (Equation 3) as follows:

Cation exchange process



The presence of NaHCO₃, Mg(HCO₃)₂, Ca(HCO₃)₂, Na₂SO₄, MgCl₂ and CaCl₂ salts gives an evidence of cation exchange processes in groundwater.

Several indices are used for the identification of water that has undergone cation exchange processes. Among them the alkali number is expressed as 100 (Na+K)/Cl in me/l, an increase or decrease of the alkali number is mainly attributed to cation exchange that takes place under two conditions [26].

- 1) Up to alkali number 100, alkalis (Na⁺+K⁺) in their halogens in solution replace Ca²⁺ and Mg²⁺ on the surface of clay minerals in the aquifer matrix.
- 2) Above 100, alkaline earths (Ca²⁺ and Mg²⁺) in their sulfates and carbonates in solution replace alkalis (Na⁺+K⁺) on the surface of clay minerals in the aquifer matrix.

The alkali number of brackish and saline groundwater samples (79%) in the fractured dolomite limestone aquifer ranges from 73.66 to 97.86. These groundwater types are characterized by the assemblages (II), (III), of salt combinations which reflect the effect of leaching and dissolution of marine salts with some contribution of the cation exchange phenomenon, forming the following hypothetical salt combinations in such groundwater.

Regarding II {NaCl, MgCl₂, MgSO₄, CaSO₄ and Ca(HCO₃)₂} 75% of water samples have the permanent groundwater was formed under marine conditions affected by dilution of rainfall. Regarding III {NaCl, MgCl₂, CaCl₂,

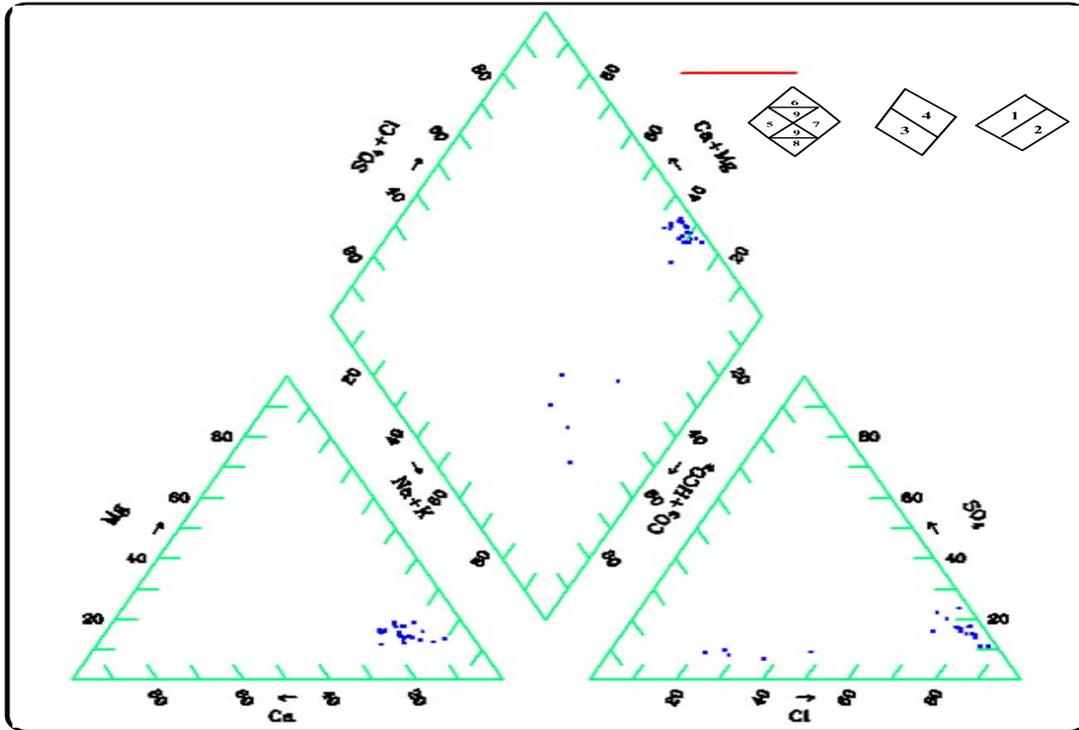
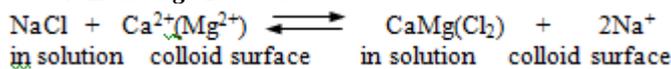


Figure 7: Piper's classification and Back and Hanshaw's evolution diagrams for all groundwater samples.

CaSO_4 and $\text{Ca}(\text{HCO}_3)_2$ 4% of water samples have the permanent hardness > temporary hardness $\{\text{MgCl}_2, \text{CaCl}_2, \text{CaSO}_4 > \text{Ca}(\text{HCO}_3)_2\}$.

As a result of such cation exchange processes, Na^+ concentration decreases, this is accompanied by an increase of Ca^{2+} and Mg^{2+} concentrations in solution, leading to an increase in salts causing permanent hardness (MgCl_2 and CaCl_2) and considerable decrease in water salinity in the same time. This case is in agreement with the general reaction of cation exchange as follows:

Cation Exchange Process

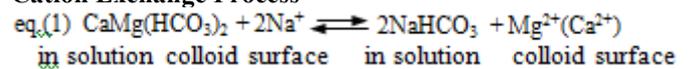


All of the fresh groundwater samples (21%) of the Nubian sandstone aquifer have alkali number values more than 100. This groundwater type belongs to the salt combination assemblages (I) which reflect the effect of leaching and dissolution of terrestrial salt (continental facies groundwater) with some contribution of cation exchange processes, forming hypothetical salt combinations as follows: NaCl , Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$ and $\text{Ca}(\text{HCO}_3)_2$ where 21% of samples have temporary hardness > permanent hardness ($\text{CaMg}(\text{HCO}_3)_2 > \text{MgSO}_4$ salts). Also, this assemblage reflects the recharge from local precipitation and drainage of wadi streams. In this case, alkaline earths (Ca^{2+} and Mg^{2+}) in their sulfates and carbonates in solution replace alkalis ($\text{Na}^+ + \text{K}^+$) on the surface of clay minerals in the aquifer matrix.

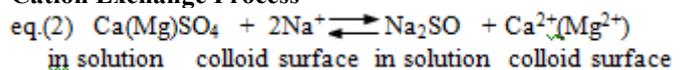
As a result of cation exchange processes, the increase of Na^+ concentration and in contrast to the decrease in Ca^{2+} and Mg^{2+} concentration in solution, lead to a considerable decrease in salts causing temporary and permanent hardness in form of $\text{MgCa}(\text{HCO}_3)_2$ salts and CaMgSO_4 salts, respectively, while causing a considerable increase in water

salinity but no change in pH and HCO_3^- content because each mole of Ca^{2+} or Mg^{2+} adsorbed is replaced by 2 moles of Na^+ . The loss of Ca^{2+} and Mg^{2+} decrease the degree of water saturation with respect to both carbonate and gypsum minerals leading to ion activity products (IAP) of carbonate and gypsum minerals which became less than that of the solubility product constant (K_{sp}). This is supported by some groundwater samples that are unsaturated with respect to carbonate minerals and all groundwater samples that are unsaturated with respect to gypsum minerals. This case is quite coincident with the following cation exchange equations (1 and 2).

Cation Exchange Process



Cation Exchange Process



Combination of cation exchange index and alkali number reveals that, most groundwater samples (79%) of the Fractured dolomite limestone aquifer (marine facies groundwater) are characterized by an alkali number less than 100, positive values of the cation exchange index and marine salts (II and III). The cation exchange process leads to a considerable increase in permanent hardness while leading to a considerable decrease in water salinity. On the contrary, few groundwater samples, (21%) of the Nubian sandstone aquifer, are characterized by an alkali number up to 100, the negative value of the cation exchange index and terrestrial salts combinations (I). Cation exchange processes lead to a considerable decrease in the temporary and permanent hardness, which is accompanied by a considerable increase in water salinity.

3.10.2. Mixing process as a Controlling Factor of Water Quality.

Generally, the mixing process is the mixing of the two water bodies (fresh and salt water) is either through sea water encroachment, or simply through diffusion process, or both. The intermingling of fresh and salty waters results in a transitional zone of mixed water having a composition ranging between the two principal fluids. This zone, termed the zone of diffusion, reflects the chemical and hydraulic properties of each of the parent fluids in varying degrees. Seawater encroachment is defined as the landward movement of seawater as a body, so as to displace fresh water. In normal conditions, freshwater discharge into the sea.

When mixing takes place between different water types of different qualities and in case of absence of any influence other than the mixing effect, the resultant water type should attain such a quality where the relation between salinity and the concentration of different ions is linear as in the Fig. (8). The relationships between total salinity (TDS) and the concentration of different ions (Ca^{2+} , Mg^{2+} , $\text{Na}^+ + \text{K}^+$, $\text{HCO}_3^- + \text{CO}_3^{2-}$, SO_4^{2-} and Cl^-) are plotted for groundwater are discussed as follows:

3.11 Nubian Sandstone Aquifer (Fresh Water):

For the fresh water of Siwa Oasis samples, the mixing curve [27], Fig. (9) reflects the following results. The data obtained from the samples revealed that all plotted points of Cl^- concentration in fresh water lie below the normal mixing line (rain and sea). This means that seawater is not the source of Cl^- for the groundwater, but the source of Cl^- is of meteoric origin. This agreed well with the log normal distribution of Cl^- in the groundwater of the aquifer, indicating that the origin of Cl^- is meteoric water [28]. The plotted points of SO_4^{2-} concentration of samples are located on the normal trend of the mixing curve, indicating a marine source of the mineralization of SO_4^{2-} ions. The HCO_3^- occupies a rather high position relative to the normal mixing trend. The increase of bicarbonate ions is related to the dominant carbonate materials of aquifer matrices and the effect of dilution by rainfall. In general, all the plotted anions increase steadily with increasing TDS of fresh groundwater.

The relation between the cations and total mineralization is not a simple mixing process, where the majority of plotted points of Na^+ , Ca^{2+} and Mg^{2+} concentrations in the fresh Siwa Oasis water samples Ca^{2+} are located at the highest of the mixing line between meteoric and sea water indicating that the origin of calcium ion is the meteoric water, Na^+ and Mg^{2+} are located around the mixing line indicating that the origin of these ions from meteoric water affecting by marine deposits. This is resulted from the cation exchange process between Na^+ and Ca^{2+} or Mg^{2+} ions. This general behavior of cations is mainly explained on the basis of meteoric water of fresh Siwa Oasis water samples as well as other secondary processes such as leaching of salts in sediments, cation exchange, concentration and oxidation-reduction processes.

Fractured dolomite limestone aquifer (brackish and saline waters):

All points of Cl^- in the brackish and saline waters were plotted. The concentration of Cl^- in groundwater of the concerned aquifers lies below the normal mixing line as in the Figs. (10, 11). This means that seawater is not the source of Cl^- for the groundwater, but the source of Cl^- is of meteoric origin. This agreed well with the log normal distribution of Cl^- in the groundwater of the aquifer, indicating that the origin of Cl^- is meteoric water. But the locations near the normal mixing line between rain and sea due to impact of marine deposits affected by meteoric water.

All plotted points of SO_4^{2-} concentration of the brackish and saline water samples are located higher the mixing line between rain and sea; this indicates a continental source of the mineralization for SO_4^{2-} which moved into the solution through leaching and dissolution processes, this indicates the marine and terrestrial origin in the SO_4^{2-} . All HCO_3^- concentration points are plotted slightly high the normal mixing trend between rain and sea, indicating the dominant is NaCl due to marine deposits and meteoric water (continental and marine sources) affected by local deposits.

The relation between the cations and total mineralization all plotted points of Na^+ and Ca^{2+} or Mg^{2+} in brackish and saline waters, water samples the Ca^{2+} plotted at the highest of the mixing line indicating that the origin is meteoric water, the Mg^{2+} lies in mixing line indicating that the origin is meteoric water and marine deposits and the Na^+ lie below the mixing line between rain and sea, indicating the mixed sources of marine deposits. This general behavior of cations is mainly explained on the basis of the mixing process between different meteoric and marine waters as well as other secondary processes such as leaching of salts in sediments, cation exchange and oxidation-reduction reactions. In general, the effect of mixing process between meteoric and marine waters is ill defined as a result of flushing by groundwater. On the other hand, the mixed water samples are affected by marine condition.

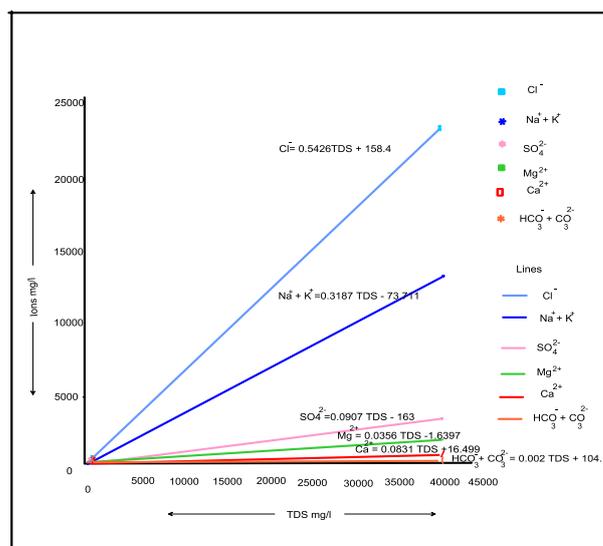


Figure 8: Graphical representation showing the ion concentration of Rain and Sea waters.

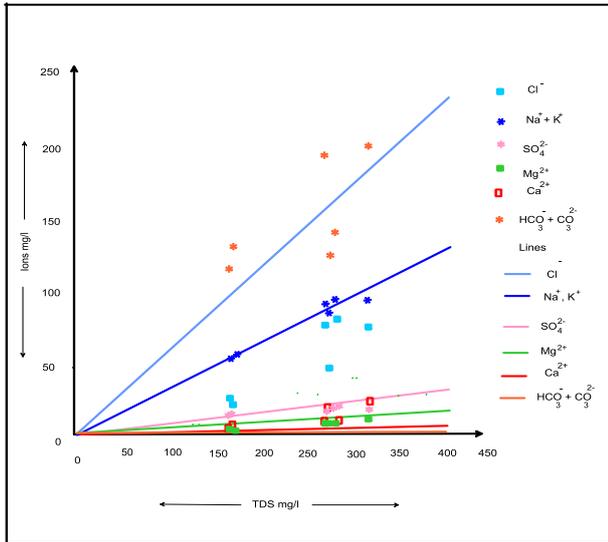


Figure 9: Graphical representation showing the ion concentration of fresh water samples of Siwa Oasis, Egypt.

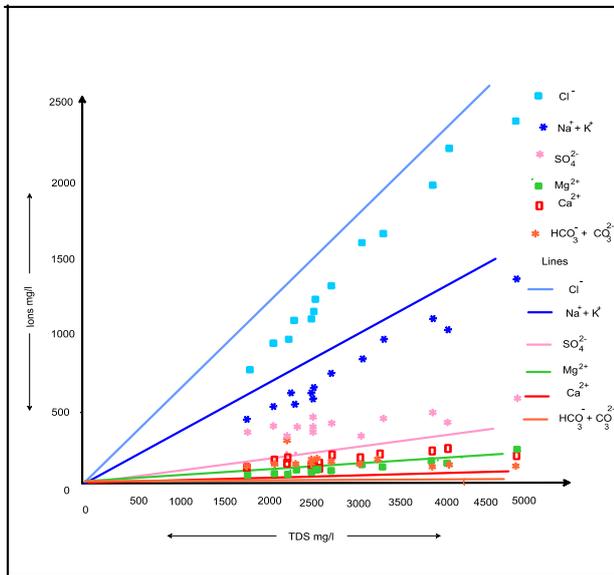


Figure 10: Graphical representation showing the ion concentration of brackish water samples of Siwa Oasis, Egypt.

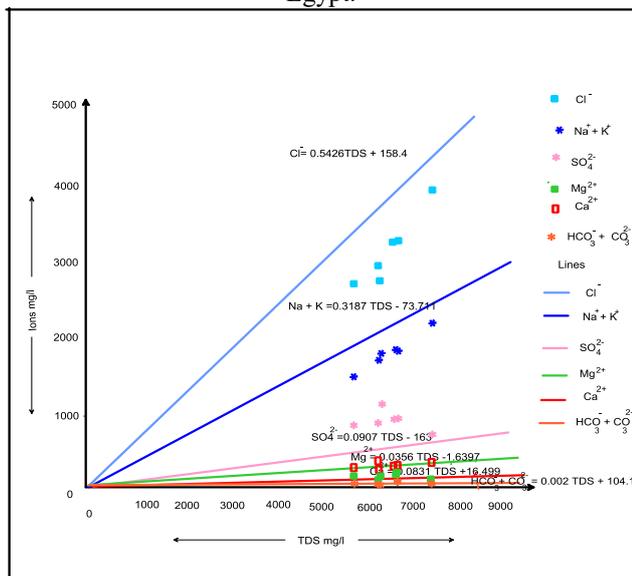


Figure 11: Graphical representation showing the ion concentration of saline water samples of Siwa Oasis, Egypt.

4. Conclusion

The groundwater samples in Siwa Oasis lie in two different aquifers, Nubian sandstone and fractured dolomite limestone. The fresh groundwater samples and brackish & saline are in Nubian sandstone and fractured dolomite limestone, respectively. The obtained data show that the TDS changed from fresh to saline 168.8 to 7472 mg/l, total hardness ranges from soft to hard, 30.7 to 1829.7mg/l as CaCO₃. The effective ions that cause an increase of water salinity are in descending order: As cations; Na⁺ > Mg²⁺ > Ca²⁺ while the anions Cl⁻ > SO₄²⁻ > HCO₃⁻ in both brackish and saline groundwater samples and HCO₃⁻ > Cl⁻ > SO₄²⁻ in fresh. Also, the ion ratios indicated that, the groundwater samples have a mixed mineralization that is possibly pure meteoric water affected by leaching and dissolution and cation exchange of both terrestrial and marine salts. Besides that, the hypothetical salts of groundwater samples show three main groups. With continued research, we will evaluate the groundwater samples for drinking, domestic and irrigation uses with solving problem of water samples to be valid for drinking and agriculture uses.

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