Geochemical modeling and isotopic approach for delineating water resources evolution in El Fayoum depression, Egypt

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Abstract
In this study, an attempt was made to understand the hydrogeochemical processes controlling water resources (surface and groundwater) evolution in El Fayoum depression, Egypt. Hydrochemical, inverse geochemical modeling integrated with environmental isotopes was applied. Forty-three surface and groundwater samples were collected and analyzed for major chemistry (cations, anions) and environmental stable isotopes ($\delta^{18}$O, $\delta^{D}$). The ionic sequence based on relative molar proportions for cations was $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$ and $\text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+} > \text{K}^+$ for surface and groundwater, respectively and for anions was $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-}$ for all water samples. $\delta^{18}$O and $\delta^{D}$ relationship plot on a typical evaporation line, enhance potential evaporation of recharging water prior to infiltration. Saturation index estimation revealed that water resources were affected by carbonate and evaporites minerals dissolution and supersaturated with Goethite, Gibbsite and Barite minerals. Inverse geochemical modeling using PHREEQC was used to identify water resources evolution in the study area through two flow paths A, B. The inverse model was constrained so the primary mineral phase including calcite, Aragonite, Anhydrite and Gypsum with Carbon dioxide (gas) are constrained to dissolve until they reach saturation, Whereas $\text{H}_2\text{S}$, hydrogen sulphide (gas) and $\text{O}_2$ (g) are constrained to out gas in all models for flow path (A). For flow path (B), Dolomite, Anhydrite and Gypsum are tend to dissolve with carbon dioxide (gas), $\text{H}_2\text{S}$ and $\text{O}_2$ (g) gases tend to outgassed. Finally, two processes controlling water resource evolution; carbonate and evaporites dissolution and development of cation exchange process through the aquifer system.

Keywords Hydrogeochemical processes · Environmental stable isotopes · Geochemical modeling · El Fayoum depression · Egypt

Introduction
El Fayoum area is one of most important agricultural lands of Egypt for a long time due to good soil cover and high crop productivity. In addition to it has a beauty nature represented by natural springs, waterfalls and the great natural lake (Qaroun Lake). El Fayoum’s water enters through two main gated intakes, one being the natural channel Bahr Youssef, the other Bahr Hassan Wasef a canal dug in 1905 (Wolters et al. 1987). There are two main drains; The Bats in the east and the Wadi in the west. All drainage water discharged into Qaroun Lake. Population growth and increase of human activities cause several problems in El Fayoum area such as water logging and soil salinization; these problems have already emerged after High Dam construction. Mismanagement of agricultural fertilizers and pesticides, reuse of drainage water for irrigation, wastewater disposal and inadequate land use planning had a major impact on groundwater degradation and soil quality in the studied area. Qaroun Lake, a closed saline lake acts as a natural discharging system for agricultural drainage water in El Fayoum depression. Previous studies focused mainly on this lake (Mansour and Sidky 2002, 2003; Gupta and Abd El-Hamid 2003) to investigate the influence of those waters on the lake water quality. As the groundwater resource in El Fayoum depression is a Quaternary aquifer that shows increasing signs of quality deterioration (Abdel Kawy and Belal 2012; Gad and El-Zeiny 2016; Gad and Hattab 2019) attributed to irrigation return and anthropogenic activities. The quality of water is a function of its physiochemical parameters that reflect groundwater characteristics and factors that controlling the quality of water (Hussein et al. 2017; Mgbenu and Egbueri
Environmental stable isotopes (δO 18, δD) modified by (Slimani et al. 2015; Berihu et al. 2017; Liu et al. 2020). The use of geochemical models is increasing in addressing groundwater quality problems involving geochemistry. Chemical modeling attempts to account for the chemical changes that occur as water evolves along a flow path. Assuming two water analyses, that represent starting and final water composition along the flow path within a groundwater system, inverse modeling is used to calculate the moles of minerals and gases that must enter or leave solution to account for the differences in composition. The use of geochemical models is increasing in addressing groundwater quality problems involving geochemistry (Slimani et al. 2015; Berihu et al. 2017; Liu et al. 2020). Environmental stable isotopes (δO 18, δD) modified by meteoric processes provide relevant and useful tracers to identify flow regimes and sources of groundwater recharge (Leontiadias et al. 1988). Frequently, groundwater retains its stable isotopic signatures unless diluted or mixed with waters of different isotopic compositions (Fontes 1980; Gat 1981). Therefore, water from different sources or those exposed to different processes such as evaporation and/or mixing often acquire identifiable isotopic composition which can serve as natural tracers. In this paper, physical, hydrogeochemical information integrated with isotopic data were incorporated into the inverse geochemical modeling to delineate water evolution in El Fayoum depression. Hence, the objectives of this study were: (1) identify various factors affecting water (surface and groundwater) hydrogeochemical processes to understand and distinguish between the water–rock interaction and anthropogenic influences (2) characterize and assess recharge areas and the origin of surface and groundwater in the study area (3) Understand the geochemical evolution of water resources as an essential part in water management and remediation processes with a better knowledge of how the groundwater system functions.

Geology and hydrogeology

El Fayoum area is a green, fertile, near-circular depression and it has a unique physical and environmental setting. El Fayoum area occupies one of the spectacular depressions in Western Desert of Egypt, about 90 km southwest of Cairo between latitudes 29° 00’ and 29° 40’ N and longitudes 30° 20’ and 31° 05’ E (Fig. 1b). It is surrounded by desert terrain except for the southeastern side, where it is connected to the Nile Valley by the Bahr Youssef canal which derives its water from Al Ibrahimiya Canal. The depression is limited at the north-western side by a shallow brackish lake, called Birket Qarun (or Lake Qarun), with an area of about 200 km² and forming the deepest part at about 45 m below the mean sea level (msl). The total surface area of El Fayoum depression is approximately 6000 km². About 1800 km² of the depression is filled with rich alluvial sediments and more than 1580 km² is cultivated. This area represents more than 5% of the total cultivated area of Egypt. It is an area of intensive agriculture, reliant solely on irrigation from the Nile River. El Fayoum depression has a special irrigation system due to the nature of its land which slopes downward from south to north. The wastewater flows from south to north towards the slope where it pours into Qarun and Wadi Al Rayan Lakes, the main sources of water drainage for the agricultural lands in the depression (UNDP 2003). Population of El Fayoum is about 2 million and the density of population is about 1000 people per km². The overall climate of this area is characterized by hot, long and dry summers and warm, short winters with scarce precipitation. Also, great temperature differences between summer and winter and between day and night characterize this belt. The temperature ranges between 46.7 and 35.1 °C in summer months and from 20 to 21.5 °C during winter months. The average annual rainfall is around 10.3 mm which is relatively low. The total annual evaporation intensity reaches 2296 mm/year, and the annual mean of relative humidity is 51.6%.

El Fayoum basin was formed initially during the Jurassic Period, probably by wind erosion in the desert, and its current shape created from tectonic subsidence that terminated in the late Eocene Epoch. The basin then subsided relative to the Nile, allowing the river to break through in flood and deposit fertile alluvial sediments. Rock units that exposed in El-Fayoum region range from Eocene to Quaternary Ages as shown in Fig. 2c. In the study area, the subsurface stratigraphic column is capped by the Pleistocene deposits that are widely distributed over the entire area of El Fayoum area. These deposits are mainly of fluvo-lacustrine origin. The Quaternary lacustrine deposits dominate in the south of Qaroun Lake, which consist of clay stone, gypsum and calcareous materials intercalated with ferruginous sandy silt (Redwan and Elhaddad 2017). The Quaternary sediments
overlay the Middle Eocene deposits, which are composed of limestone, marl, sand, and gypsiferous shale that outcrop around the depression (Metwaly et al. 2010). The Middle Eocene deposits overlay the Oligocene deposits, which are composed of fluvio-marine variegated sands and sandstone, with alternating shale, marls, and calcareous beds. The Lower Miocene sediments are composed of continental sandstones and conglomerates, while sediments of Pliocene age are composed of calcareous sandstones and limestones. Several distortion lines, such as faults, breaks, and some folds striking N–S and NW–SE, that affect El Fayoum Depression are dominant at the peripheries of the depression (El-Sayed et al. 2015). Based on a detailed analysis of the available lithological data, geological map and lithological succession were constructed (Fig. 2b, c).

The surface water hydrology system is represented by the Nile River, irrigation canals, and drains. The area gets irrigation water from the River Nile and the main irrigation branches Bahr Youssef and Bahr Hassan Wassef canals to the depression. Generally, three groundwater aquifers were distinguished in El Fayoum Depression (Mohamed et al. 2015) namely, the Quaternary aquifer, Eocene aquifer, and Nubian sandstone aquifer. Quaternary aquifer is a perched water table aquifer that is maintained by infiltration from canals, drains, and irrigated fields. Many of the springs in El Fayoum depression have their sources from the Quaternary aquifer. Beneath this aquifer, the Eocene aquifer, that is composed of limestone and marls, has high salinity and is therefore unsuitable for potable or irrigation use. The Nubian aquifer is composed of sandstones and shales of Cretaceous
and it was very deep aquifer (> 1000 m), not exposed in the area of study but reported in the subsurface at Wadi El Rayan and in numbers of oil wells drilled in some localities close to the study area. The Nubian formation is generally characterized by relatively high hydrostatic pressure (Himida and Abdel Baki 1983). The Quaternary aquifer has its maximum thickness (about 60 m) at the center of the depression while, the minimum thickness is less than 1 m along the depression peripheries due to local faulting (Ahmed, 2012). The groundwater depths varied from few centimeters to 9.1 m, with groundwater level ranges from +20 m (amsl) in the area near Nile River to—50 m (bmsl) near Qarun Lake. In addition, the ground-water flows from SE to N and NW in concordance with the general slope of the depression topography in Fig. 1c that shows the land slopes from Lahun to Lake Qarun which occupies the deepest part in El Fayoum depression (Gad and El Hattab 2019). The Quaternary aquifer is thought to be recharged by the seepage from surface irrigation canals, drains, and Nile River. In addition percolation from the underlying fractured limestone of the Eocene aquifer through hydraulic connection (Doerfliger et al. 1999). This study focused on the shallow water-bearing formations of the Quaternary aquifer that directly related to the objective of this investigation.

**Materials and methods**

**Sample collection and analysis**

A set of representative surface water samples (19 drainage water and 13 canals) and 12 groundwater samples were analyzed for physical measurements including electrical conductivity (EC), pH, temperature, total dissolved solids (TDS), and dissolved oxygen (DO) using portable probes. Chemical parameters comprising major cations (Ca, Mg, Na, K) and anions (Cl, SO\(_4\), HCO\(_3\)) concentrations were also analyzed. All the wells located in El Fayoum depression had been in use during the time of sampling (Fig. 1b). The depths of the selected wells ranged from 8 to 13 m below the surface. The samples were collected in 1-L narrow neck pre-washed polyethylene bottles. Analysis of the water samples was carried out following the methods described in APHA (1995). Total hardness (TH) as CaCO\(_3\) and Ca\(^{2+}\) were analyzed using standard EDTA. Mg\(^{2+}\) was calculated by taking the differential value between TH and Ca\(^{2+}\) concentrations. Na\(^+\) and K\(^+\) were measured using a flame photometer. Total alkalinity and CaCO\(_3\), CO\(_3\)^{2−}, and HCO\(_3\)^{−} were estimated by titrating with HCl. Cl\(^−\) was determined by standard Hg (NO\(_3\))\(_2\) titration. SO\(_4\)^{2−} and NO\(_3\)^{−} were analyzed using

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**Fig. 2**  
**a** Location map of El Fayoum depression, Egypt.  
**b** Geological map of El Fayoum and its adjacent area (Nile valley and northward to the greater Cairo), compiled and modified after Beadnell (1905) and Said (1962).  
**c** Lithological succession with the location of proposed shallow aquifer at the center of the depression. Redrawn and modified after Metwaly et al. (2010)
UV/visible spectrophotometer. All parameters are expressed in milligrams per liter and milliequivalents per liter. Data quality was assessed using the charge balance between the difference of cations and anions (expressed in meq/l) divided by their summation according to the following equation:

\[
\Sigma (\text{Cations} - \text{Anions})/\Sigma (\text{Cations} + \text{Anions}) \times 100.
\]  

(1)

With an acceptable range of ±5 (Hem 1991) that confirm the water quality assessment. Environmental stable isotope \((\delta^{18}O\text{ and } \delta D \text{ in } \%e)\) samples were collected according to the procedure described in Clark and Fritz (1997). The analysis was done using Picarro L2120-I Isotopic Water Liquid and Vapour Analyzer. Both results were expressed in \(\delta^{18}O\) (IAEA 1995) with analytical error of ±0.1 and ±1‰ for \(\delta^{18}O\) and \(\delta D\) respectively. The stable isotope data are reported in Vienna-Standard Mean Ocean Water (V-SMOW) in %e using δ notation according to the equation:

\[
\delta = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1\right) \times 1000.
\]  

(2)

\(R_{\text{sample}}\) and \(R_{\text{standard}}\) are the measured isotopic ratios \((^{18}O/^{16}O)\) and \((^{2}H/^{1}H)\) of sample and standard material.

Ground surface elevation and the location of the sampling sites were recorded using a global positioning system (GPS) manufactured by GARMIN. This was supported by a topographic sheet made available by the Egyptian Survey Department. Measured and estimated groundwater variables were analyzed within ArcGIS 10.2.2 package. All the chemical and Environmental isotope analyses were carried out in the Central Laboratory of Stable Isotope Hydrology at Egyptian Nuclear and Radiological Regulatory Authority (ENRRA).

**Geochemical modeling**

The saturation index is a vital geochemical parameter in the fields of hydrogeology and geochemistry, often useful for identifying the existence of some common minerals in the groundwater system (Deutsch 1997). In this present study, saturation indices (SIs) was calculated in terms of the following equation (Lloyd and Heathcote 1985):

\[
\text{SI} = \log(\text{IAP}/K_i(T)),
\]  

(3)

where IAP is the relevant ion activity product, which can be calculated by multiplying the ion activity coefficient \(\gamma_i\) and the composition concentration \(m_i\), and \(K_i(T)\) is the equilibrium constant of the reaction using the measured sample temperature. When the groundwater is saturated with some minerals, SI equals zero; positive values of SI represent oversaturation, and negative values show undersaturation (Appelo and Postma 1994; Dreyer 1997). Inverse geochemical modeling with the thermodynamic program PHREEQC (Parkhurst and Appelo 1999) was used to calculate the geochemical processes responsible for groundwater evolution or changes in quality. To deduce geochemical reactions that accounts for the change in chemical composition of groundwater along the flow path, both aqueous species and saturation indices were calculated. The flow paths were selected on the basis of hydrogeology, hydraulic head variation within the flow system and geographic location in the study area. Minerals used in inverse geochemical modeling are limited to those known to occur in the study area. The mole transfer of minerals and gases reported in mmol kg\(^{-1}\) of water. Positive values of phase mole transfers indicate dissolution of minerals or gases, and negative values represent precipitation or outgassing.

**Results**

**General hydrochemistry**

Table 1 presents the statistical summary of all the parameters analyzed. The minimum, maximum, mean and standard deviation of the major ions in the Quaternary aquifer of El Fayoum depression compared with WHO (2017) guidelines for drinking water. Temperature of surface water samples (canals, drains) ranged from 20.8 to 26.13 °C with mean of 23.6 °C and 19.46–27.24 °C with mean value of 22.8 °C respectively, while the groundwater ranged between 21.7 and 31.8 °C with mean value of 24.7 °C. The pH values of surface water samples (canals, drains) ranged from 6.16 to 6.7 with mean value of 6.38 and 5.91–7.12 respectively. For groundwater samples the pH values ranged from 5.65 to 6.51 with mean value of 6.09 the data were reflective of slightly acidic to slightly neutral conditions due to that the selected wells were shallow with depth to water ranged from 8 to 13 m, the dissolved carbonates is predominately in \(\text{HCO}_3\) form. In surface water, EC and TDS values exceeded those of groundwater may be due to anthropogenic pollution inputs. The EC of surface water (canals and drains) varied from 606 to 2636 μs/cm, with a mean value of 1108 μs/cm and 565 to 3756 μs/cm with a mean value of 1660 μs/cm respectively. In groundwater samples EC values ranged from 162 to 4426 μs/cm, with a mean value of 1582 μs/cm and 565 to 3756 μs/cm with a mean value of 1660 μs/cm respectively.
as fresh respectively. The relative abundance of major cations in shallow groundwater is \( \text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+} > \text{K}^+ \) (on a molar basis) and \( \text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} \) for anions. For surface water samples (canals and drains) the predominant cations are \( \text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+ \) (on molar basis) and \( \text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-} \) for anions. The maximum \( \text{Ca}^{2+} \) and \( \text{Mg}^{2+} \) concentrations of 144 mg/l and 168 mg/l respectively in groundwater samples are however, higher than WHO (2017) standards limits by 50% and 58% respectively. The prevailing common lithology is the old lacustrine deposits which are present within the depression and consist mainly of clay, fine to medium coarse sand with considerable amounts of calcareous and gypsiferous materials. The mean sodium concentrations is 159 mg/l is within the standard limits but potassium concentration is 17.6 mg/l in groundwater samples that exceeds WHO (2004) standards while, for irrigation canals the mean sodium and potassium concentrations are within WHO standards limits. While, for drains Na, K exceeds the international standard as illustrated in Table 2. The presence of \( \text{HCO}_3^- \) ions in shallow groundwater samples is derived from carbon dioxide in the atmosphere, soils and by dissolution of carbonate rocks. Bicarbonate ion represents the first dominant anion in the study area. Most \( \text{Cl}^- \) in the groundwater related to lacustrine deposits found in quaternary aquifer of El Fayoum depression. The most extensive and important occurrences of sulphate ions in the investigated area are sedimentary rocks such as gypsum (\( \text{CaSO}_4\cdot2\text{H}_2\text{O} \)) and anhydrite (\( \text{CaSO}_4 \)) that give the chance for active dissolution, leaching and ion-exchange processes. This dominates in the central and western parts of the depression. Further addition of sulphate to groundwater comes from breakdown of organic matter in the soil, 

| Parameters | WHO (2017) | Groundwater (N=12) | Irrigation canals (N=13) | Drainage water (N=19) |
|------------|------------|---------------------|-------------------------|-----------------------|
| TDS        | 1000       | 6 50                | 3 23                    | 8 42                  |
| Na         | 200        | 2 17                | 2 15                    | 8 42                  |
| K          | 10         | 3 25                | 4 31                    | 7 37                  |
| Ca         | 75         | 6 50                | 3 23                    | 7 37                  |
| Mg         | 30         | 7 58                | 5 38                    | 12 63                 |
| Cl         | 250        | 1 8                 | 2 15                    | 8 42                  |
| SO4        | 500        | 2 17                | 1 8                     | 3 16                  |
| HCO3       | 300        | 7 58                | 2 15                    | 10 53                 |

*Based on WHO guidelines (2004)
and from addition of leachable sulphate in fertilizers of intensively cultivated area in the depression.

Hypothetical salt combination could classify the hydrochemical data into three groups as follow:

Group I include salts of [(NaCl, Na₂SO₄, MgSO₄, Mg (HCO₃)₂ and Ca (HCO₃)₂] most of water samples belong to this group (92% groundwater, 85% Irrigation canals and 89% of drainage water).

Group II include salts of [(NaCl, Na₂SO₄, NaHCO₃, Mg (HCO₃)₂ and Ca (HCO₃)₂] some of water samples belong to this group (17% groundwater and 5% of drainage water).

Group III include salts of [(NaCl, Na₂SO₄, MgSO₄, CaSO₄ and Ca (HCO₃)₂] lower numbers of water samples belong to this group (8% of Irrigation canals and 5% of drainage water).

As indicated by the spatial distribution maps of salinity (Fig. 3), The TDS increases from the peripheries that dissected by drains towards the central part of the study area, where the seepage from drains leads to groundwater salinization. Higher TDS values may be related to water–rock interaction, upward recharge from the underlying fracture limestone of Eocene aquifer, irrigation return flow and seepage from the surrounded drains (El-Sheikh 2004). Low TDS values may be attributed to refreshment from irrigation canals.

Discussion

Processes controlling major solutes distribution

The rock dominance of the major ion chemistry in the study area provides an insight of chemical weathering in the aquifer, since weathering of different parent rocks
(e.g., carbonates, silicates and evaporites) yield different combinations of dissolved cations and anions to solution. The Gibbs diagram is widely used to study the relationship between water chemistry and aquifer lithology (Gibbs 1970). The functional sources of the dissolved chemical constituents can be divided into three distinct fields in this diagram, namely rainfall dominance, evaporation dominance and rock-weathering dominance areas. The ratios of Cl/ (Cl + HCO₃) and (Na + K)/(Na + K + Ca) as a function of TDS were drawn in Fig. 4. The majority of the samples fell in evaporation-dominated field demonstrating the

Fig. 4  Gibbs diagram for cations and anions in (meq/l) of collected water samples

| Sample no. | Na/Cl | Ca/Mg | Ca/SO₄ | Sample no. | Na/Cl | Ca/Mg | Ca/SO₄ |
|------------|-------|-------|--------|------------|-------|-------|--------|
| Groundwater|       |       |        | Drainage water|       |       |        |
| 2          | 1.207 | 1.706 | 1.325  | 3           | 1.589 | 1.200 | 1.326  |
| 10         | 1.227 | 0.486 | 0.563  | 4           | 1.195 | 1.255 | 0.680  |
| 13         | 1.457 | 0.542 | 0.963  | 6           | 3.394 | 0.567 | 0.178  |
| 15         | 1.749 | 0.538 | 1.261  | 8           | 1.550 | 0.667 | 1.168  |
| 19         | 1.342 | 0.800 | 1.194  | 9           | 1.486 | 1.500 | 2.069  |
| 21         | 1.444 | 1.000 | 1.127  | 12          | 1.829 | 0.462 | 0.676  |
| 22         | 2.013 | 1.352 | 1.063  | 24          | 1.661 | 0.778 | 0.883  |
| 28         | 3.280 | 1.200 | 0.267  | 25          | 1.486 | 1.000 | 0.964  |
| 29         | 1.417 | 0.771 | 1.095  | 26          | 1.888 | 1.599 | 1.257  |
| 30         | 1.500 | 0.800 | 1.053  | 27          | 1.271 | 0.571 | 1.036  |
| 31         | 1.155 | 1.000 | 1.274  | 32          | 1.386 | 1.400 | 0.918  |
| 42         | 1.805 | 1.023 | 1.157  | 33          | 1.408 | 1.167 | 0.805  |
| 44         | 1.000 | 0.560 | 1.028  | 34          | 1.772 | 1.273 | 0.579  |
| Irrigation canals |       |       |        |             |       |       |        |
| 1          | 2.191 | 1.259 | 1.138  | 37          | 2.081 | 0.750 | 0.258  |
| 7          | 1.432 | 1.102 | 0.784  | 40          | 1.570 | 0.875 | 0.638  |
| 14         | 1.431 | 0.556 | 1.031  | 43          | 1.664 | 0.769 | 0.909  |
| 16         | 1.300 | 0.667 | 1.000  | 46          | 1.984 | 0.840 | 0.676  |
| 20         | 1.275 | 1.500 | 1.983  | 47          | 1.569 | 0.700 | 1.407  |
| 23         | 1.477 | 1.000 | 1.290  | 35          | 1.231 | 1.111 | 0.701  |
| 38         | 1.608 | 1.500 | 1.290  | 39          | 1.864 | 1.444 | 0.432  |
| 41         | 1.591 | 1.061 | 1.489  | 45          | 1.444 | 1.000 | 1.515  |
| 5          | 1.356 | 1.652 | 1.487  | 11          | 1.178 | 1.045 | 1.267  |

Bold values were < 1
importance of irrigation return flow on collected surface and groundwater samples chemistry.

According to (Table 3) for calculated ionic ratios and (Fig 5a) the Na+/Cl− showed clear enrichment of Na+ in the surface (canals and drains) and groundwater samples. The Na+/Cl− ratios in the study area varied from 1 to 3.3 with an average value of 1.6 in groundwater and for surface water, it varied from 1.2 to 2.2 in canals and 1.2 to 3.4 in drains. In general, the Na+/Cl− molar ratio should be approximately equal to 1, if the halite dissolution is the only source. The majority of the samples (Fig 5a) having a higher Na+/Cl− ratio suggested another possible source of Na+ may be ion-exchange or silicate weathering be responsible for the exceeding of Na+ concentration in collected water samples. The Ca2+/Mg2+ ratio suggests that dissolution of calcite and dolomite in the aquifer. Results of surface and groundwater samples ratios (Fig 5b) ranged between 0.49 and

![Fig. 5](image-url) Relationships of a Na+ vs. Cl−, b Ca2+ vs. Mg2+, c Ca2+ vs. SO42−, d Cl− vs. (Ca2+ + Mg2+), e Chadha’s plot, f (Ca2+ + Mg2+) − (SO42− + HCO3−) vs. (Na+ − Cl−) in (meq/l) for collected water samples in the study area.
1.71 with an average value of 0.93 for groundwater and from 0.56 to 1.65 with an average value of 1.02 for canal and drain water samples respectively. That is, if the ratio Ca/Mg=1, dissolution of dolomite should occur, whereas a higher ratio is indicative of greater calcite contribution (Maya and Loucks 1995). For groundwater samples, about 54% have Mg>Ca and 46% have Ca>Mg according to (Table 3) indicative of dissolution of calcite/dolomite minerals that largely controlled by CO2 fugacity and pH according to the reactions:

\[
\text{CaCO}_3 + \text{CO}_2(g) + \text{H}_2\text{O} \rightarrow \text{Ca}^{2+} + 2\text{HCO}_3^-,
\]

\[
\text{CaMg(CO}_3)_2 + 2\text{CO}_2(g) + 2\text{H}_2\text{O} \rightarrow \text{Ca}^{2+} + \text{Mg}^{2+} + 4\text{HCO}_3^-.
\]

Drainage water samples show more abundance of Mg>Ca by 53% than only 15% for canals water. Ca\(^{2+}\)/SO\(_4\)^{2−} ratio illustrated in Fig 5c showed that drainage water samples have an excess SO\(_4\)^{2−} over Ca\(^{2+}\) in about 68% of samples compared with 23% and 24% in groundwater and canals water, respectively that may be due to gypsum dissolution effect. The plot of (Ca\(^{2+}\) + Mg\(^{2+}\)) vs. (SO\(_4\)^{2−} + HCO\(_3\)^{−}) (Fig.5d) shows that all surface and groundwater samples are distributed close to or below 1:1 line due to an excess of (SO\(_4\)^{2−} + HCO\(_3\)^{−}) over (Ca\(^{2+}\) + Mg\(^{2+}\)) an indication of that the dissolution of calcite, dolomite, anhydrite and gypsum are the dominant reactions and ion exchange tends to be predominant process in the system (Mclean and Jankowski 2000).

Chadha’s diagram can be used to study the various hydrochemical processes responsible for water system evolution such as base cation exchange, mixing of natural waters and saline water (Chadha 1999). Results of hydrochemical analysis plotted on this diagram (Fig 5e) show that some groundwater samples can be related to field 1 of (Ca-Mg-HCO\(_3\) water type) and drains are related to field 2 of (Ca-Mg-SO\(_4\)/Cl mixed water) and the rest of samples related to field 3 (Na-Cl water type). To check the hypothesis of cation exchange is one of geochemical process that may control water chemistry. The relation of (Ca\(^{2+}\) + Mg\(^{2+}\)) − (SO\(_4\)^{2−} + HCO\(_3\)^{−}) vs. (Na\(^{+}\) - Cl\(^{−}\)) meq/l was illustrated in Fig 5f can better indicate the involvement of cation exchange between groundwater and its host environment. Negative values of (Ca\(^{2+}\) + Mg\(^{2+}\)) − (SO\(_4\)^{2−} + HCO\(_3\)^{−}) are usually used to demonstrate the involvement of cation exchange, inducing the ions absorbed on the surface of fine-grained materials of the aquifer to be replaced by the ions in the solutions. The ratio of this trend in surface and groundwater samples was linear with a slope of –1.33 and the correlation coefficient was 0.97. As the cation exchange between Na\(^{+}\) and Ca\(^{2+}\) or Mg\(^{2+}\) will lower the concentration of Ca\(^{2+}\) dissolved in water, this was illustrated in the relation between Ca\(^{2+}\) and SO\(_4\)^{2−} in Fig 5c.

### Geochemical modeling

In order to evaluate possible hydrochemical reactions along the flow paths, inverse mass-balance models were developed using the PHREEQC program (Parkhurst and Appelo 1999). Three geochemical processes may contribute to generation of solutes in groundwater including evaporation, carbonate dissolution/precipitation and silicate weathering (Garrels and MacKenzie 1971). Statistical summary of thermodynamic speciation calculations for surface and groundwater samples using PHREEQC program were illustrated in Table 4. The results indicated that the surface (canals, drains) and groundwater samples of the Quaternary aquifer

| Table 4 Statistical summary of saturation indices calculation using PHREEQC |
|-------------------------------------------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
|                  Al(OH)\(_3\)   | Anhydrite | Aragonite | Barite | BaSO\(_4\) | Calcite | CaCO\(_3\) | CO\(_2\)(g) | Dolomite | CaMg(CO\(_3\))\(_2\) | Gibbsite | Goethite | Gypsum | CaSO\(_4\)2H\(_2\)O | Hematite | Fe\(_2\)O\(_3\) |
|-------------------------------------------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| **Groundwater** |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Min               | −1             | −2.82           | −2.28           | 0.76            | −2.14           | −1.21           | −4.28           | 1.69            | 2.83            | −2.12           | 7.02            |
| Max               | 0.88           | −1.1            | −0.79           | 1.8             | −0.65           | 0.06            | −1.07           | 3.58            | 6.23            | −0.91           | 14.98           |
| Mean              | −0.093         | −1.91           | −1.390          | 1.141           | −1.245          | −0.664          | −2.269          | 2.637           | 4.506           | −1.647          | 10.951          |
| SD                | 0.529          | 0.500           | 0.435           | 0.361           | 0.436           | 0.468           | 0.967           | 0.514           | 0.895           | 0.426           | 1.994           |
| **Irrigation canal** |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Min               | 0.11           | −2.28           | −1.58           | 0.77            | −1.44           | −1.47           | −2.75           | 2.69            | 4.33            | −2.05           | 10.69           |
| Max               | 1.98           | −1.21           | 1.2             | 1.9             | −0.67           | −0.8            | −1.3            | 4.66            | 7.26            | −0.98           | 16.28           |
| Mean              | 0.97           | −2.11           | −1.39           | 1.05            | −1.245          | −1.025          | −2.37           | 3.67            | 6.025           | −1.89           | 14.05           |
| SD                | 0.428          | 0.344           | 0.765           | 0.309           | 0.258           | 0.187           | 0.503           | 0.447           | 0.775           | 0.346           | 1.540           |
| **Drainage water** |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| Min               | 0.49           | −2.34           | −1.53           | 1.01            | −1.38           | −1.42           | −2.87           | 3.2             | 4.98            | −2.11           | 11.81           |
| Max               | 1.85           | 2.24            | −0.2            | 1.75            | −0.05           | 1.27            | 0.22            | 4.52            | 8.6             | −0.45           | 18.97           |
| Mean              | 1.113          | −1.244          | −1.103          | 1.276           | −0.941          | −0.748          | −1.781          | 3.823           | 6.335           | −1.482          | 14.519          |
| SD                | 0.321          | 1.276           | 0.337           | 0.211           | 0.342           | 0.582           | 0.710           | 0.319           | 0.880           | 0.425           | 1.758           |

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were undersaturated (dissolution is required to achieve equilibrium) with minerals and gases in order of Al(OH)₃ > partial pressure of CO₂(g) > Calcite CaCO₃ > Aragonite CaCO₃ > Gypsum CaSO₄·2H₂O > Anhydrite CaSO₄ > Dolomite CaMg(CO₃)₂ with the mean SI values of (−0.093, −0.664, −1.245, −1.390, −1.647, −1.911 and −2.269 respectively. Negative values of PCO₂(g) may reflect the recharge from underlying aquifer and seepage from drains. Surface and groundwater samples have a tendency to precipitate the minerals/gases in order of Hematite Fe₂O₃ > Goethite FeOOH > Gibbsite Al(OH)₃ > Barite BaSO₄ with the mean values of SI of 10.951, 4.506, 2.637 and 1.141 respectively.

Two flow paths were modeled based on Hydrochemical data as illustrated in Fig. 1b, the flow path (A) at which the less mineralized (recharge water) was canal water (sample 20) which has TDS value of 388 mg/l and the moderate mineralized (transition water) one was drainage water (sample 6) which has TDS value of 2431 mg/l. The flow path (B) at the moderate mineralized drainage water (sample 6) also modeled with the more mineralized (Final water) groundwater sample (sample 10) which has TDS value of 2833 mg/l.

The inverse modeling in PHREEQC takes into account uncertainty limits that are constrained to satisfy the mole balance for each element and valence state as well as the charge balance for each solution within the simulation. The simulations were constrained within the specified uncertainty limits (0.025 or 0.05) as default for all model runs. Potential phases in the inverse modeling were selected based on precipitation/dissolution results from hydrochemical data and saturation index calculations for the modeled samples (Table 5). The results of inverse geochemical modeling were illustrated in Table 5 as shown below: 26 and 14 models were resulted for flow paths A, B respectively with selection done based on the lowest sum of residuals and uncertainty limits. The inverse model was constrained so the primary mineral phase including calcite, aragonite, anhydrite and gypsum with carbon dioxide (gas) are constrained to dissolve until they reach saturation, Whereas H₂(g), hydrogen sulphide (gas) and O₂(g) are constrained to out gassed in all models for flow path (A). For flow

Table 5 Inverse modeling along the flow paths A, B (Inverse model calculations were conducted using PHREEQC v.214.03 computer program (Parkhurst and Appelo 1999). Thermodynamic database used: phreq.dat values are in mol/kg H₂O. Positive (mass entering water) and negative (mass leaving water) phase mole transfers indicate dissolution and precipitation, respectively, as mmol kg⁻¹)

| Mineral | Flow path A: phase mole transition (mol/kg H₂O) |
|---------|-----------------------------------------------|
| Phases  |                                               |
| Canal water (20) → Drainage water (6) |                                               |
| Calcite |                                               |
| Aragonite | – 1.70E−03 – – – |
| Anhydrite | – 1.70E−03 – 1.70E−03 – |
| Gypsum |                                               |
| CO₂(g) | 9.59E−03 – 9.59E−03 – 1.13E−02 |
| H₂(g) | – 7.03E+01 – 7.03E+01 – 7.03E+01 |
| H₂S(g) | – 3.24E−03 – 3.24E−03 – 4.94E−03 |
| O₂(g) | – 3.52E+01 – 3.52E+01 – 3.52E+01 |

| Mineral | Flow path B: phase mole transition (mol/kg H₂O) |
|---------|-----------------------------------------------|
| Phases  |                                               |
| Drainage water (6) → groundwater (10) |                                               |
| Calcite |                                               |
| Dolomite | – 2.70E−03 – – – 2.70E−03 |
| Aragonite | – 2.70E−03 – – – |
| Anhydrite | – 2.70E−03 – 2.70E−03 – |
| Gypsum |                                               |
| CO₂(g) | – 1.28E−02 – 1.28E−02 – 1.01E−02 |
| H₂(g) | – 1.80E+01 – 1.80E+01 – 1.80E+01 |
| H₂S(g) | 7.13E−03 – 7.13E−03 4.43E−03 – 4.43E−03 |
| O₂(g) | – 8.97E+00 – 8.97E+00 – 8.97E+00 |

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The Quaternary deposits cover wide stretches of the investigated area are distinguished into Holocene deposits and Pleistocene deposits as mentioned in El-Sheikh (2004). The old lacustrine deposits (45 m thick) formed the Pleistocene deposits composed mainly of clay fine to medium coarse sand with considerable amounts of calcareous and gypsiferous materials. As carbonate minerals (calcite, dolomite) dissolution for both flow paths A, B contribute to Ca²⁺, Mg²⁺ and HCO₃⁻ and Gypsum dissolution leads an increase in Ca²⁺ and SO₄²⁻ to the system. Additional increase in Ca²⁺ leads to dissolution of more calcite that increase CO₂, which leads to slightly lower pH as illustrated in Table 1. The out gassing of hydrogen sulphide is occurring in flow path A, which an indication of SO₄²⁻ reduction. Sulphate reduction occurs extensively in natural systems that involves the consumption of substantial amounts of hydrogen ions and production of HS⁻ at certain pH. As drainage water contains organic carbon or H₂ that is oxidized while sulphate is reduced (Canfield 2001) according to the following equations:

\[ \text{SO}_4^{2-} + 2\text{CH}_2\text{O} \rightarrow \text{H}_2\text{S} + 2\text{HCO}_3^- , \]

\[ 2\text{H}^+ + \text{SO}_4^{2-} + 4\text{H}_2 \rightarrow \text{H}_2\text{S} + 4\text{H}_2\text{O} . \]

The hydrogen ions are consumed and bicarbonate is produced, which leads to an increase in pH as shown in Table 1 for drainage water samples. Field measured ORP (oxidation – reduction potential) for drainage water have ranged between (−64 to 611 mv) while it was ranged between (227–767 mv) and (125–551 mv) for groundwater and irrigation canals respectively. Field measured ORP (oxidation – reduction potential) for drainage water have ranged between (−64 to 611 mv) while it was ranged between (227–767 mv) and (125–551 mv) for groundwater and irrigation canals respectively. (El Mahllawy et al. 2013) studied the composition and activation aspects of El Fayoum clays for use as a drilling fluid, the mineralogical analysis of the studied samples using XRD (X-Ray Diffraction) were essentially montmorillonite, kaolinite and illite as clay minerals in a descending order of abundance. Quartz and calcite minerals were also found in minor amounts as non-clay components. Inverse model calculations were conducted using PHREEQC v.214.03 computer program (Parkhurst and Appelo 1999). Thermodynamic database used: phreeq.dat values are in mol/kg H₂O. Positive (mass entering water) and negative (mass leaving water) phase mole transfers indicate dissolution and precipitation, respectively, as mmol kg⁻¹.

### Environmental isotope studies

#### Isotopic composition of surface and groundwater samples

Stable isotopes composition (δ¹⁸O and δD) of 44 surface and groundwater samples were measured to provide basic information on the origin and sources of recharge for the Quaternary aquifer in the studied area. The stable isotope composition of surface and groundwater samples collected together with those of d-excess values is compiled in Table 6. Groundwater values of δ¹⁸O range between (+3.17 and +4.21 ‰) with an average value of 3.84‰ and δD values fall or lie between (+25.41 and +32.31 ‰) with an average value of 29.53 ‰ and d-excess values range from (−2.15 to 0.14 ‰).

For surface water (canals, drains) samples, δ¹⁸O values range from +4.03 to +4.83‰ and +4 to +5.19‰, while δD values range from +30.75 to +34.91‰ and +28.6 to +35.57‰ with d-excess values fall between (−3.73 to −0.95‰) and (−5.95 to −2.18‰) respectively. The plot of δ¹⁸O and δD (Fig. 6) offers a complementary insight into the origin of groundwater in El Fayoum depression according to their isotopic compositions and to their position regarding GMWL (Global Meteoric Water Line) (Craig 1961). The global meteoric water line (GMWL) represents the regression line of global continental precipitation samples with the following equation (Craig 1961):

\[ \delta D = 8 \delta^{18}O + 10, \]

with a slope of 8 and intercept 10. The plot of δ¹⁸O versus δD in ‰ surface and groundwater samples (Fig. 6) illustrate that all collected water samples are located within an evaporation line given by a regression equation of slope 6.63 and intercept of 3.98. An enrichment occurs for both δ¹⁸O and δD values compared with Nile River revealing the occurrence of intensive evaporation process in addition to irrigation return phenomena with an increase of salinity as shown in Fig. 7 for TDS in (mg/l) and δ¹⁸O in ‰.

TDS values in Fig. 7 show an increment compared with slightly constant values of δ¹⁸O for collected water samples. This may be due to evaporation and dissolution processes that affect surface and groundwater samples in El Fayoum depression.

### Conclusions

The results of this study showed that integration of hydrochemical data, inverse geochemical modeling coupled with isotopic data can help to understand the hydrogeochemical processes controlling water chemistry evolution in El Fayoum depression. The relative abundance of major cations in shallow groundwater is Na⁺ > Mg²⁺ > Ca²⁺ > K⁺ (on molar basis) and HCO₃⁻ > Cl⁻ > SO₄²⁻ for anions. For surface water samples (canals and drains) the predominant cations are Na⁺ > Ca²⁺ > Mg²⁺ > K⁺ (on molar basis) and HCO₃⁻ > Cl⁻ > SO₄²⁻ for anions. Gibbs plot that illustrate Rock-water interaction which affects Quaternary aquifer system indicates that most water samples fall in evaporation...
dominant zone. As Nile River being the only recharge source for the depression via Bahr Youssef, the groundwater in the study area is characterized by Na-Ca-Mg/HCO₃ type in 92%, while irrigation canals and drainage water have this type 77%, 58% respectively. The concentration of \((\text{SO}_4^{2-} + \text{HCO}_3^-)\) exceeds \((\text{Ca}^{2+} + \text{Mg}^{2+})\), which is the result of calcite, dolomite in the system. Gypsum dissolution contributes to increase of both \(\text{Ca}^{2+}\) and \(\text{SO}_4^{2-}\) in the aquifer, which leads to a slightly low PH and supersaturation or near equilibrium of calcite. \(\text{Na}^+\) is comparatively higher than \(\text{Cl}^-\) for surface and groundwater samples suggested cation-exchange process that

| Sample no. | δO-18 | δD  | d-excess | Sample no. | δO-18 | δD  | d-excess |
|------------|-------|-----|----------|------------|-------|-----|----------|
| Groundwater|       |     |          | Drainage water|     |     |          |
| 2          | 4.05  | 30.25 | -2.15    | 3          | 4.34  | 31.81 | -2.91    |
| 10         | 4     | 4.47  | -2.94    |
| 13         | 3.17  | 25.5  | 0.14     |
| 15         | 3.68  | 28.46 | -0.98    |
| 19         | 4.15  | 32.15 | -1.05    |
| 21         | 4.21  | 32.31 | -1.37    |
| 22         | 4.03  | 30.69 | -1.55    |
| 28         | 3.71  | 28.56 | -1.12    |
| 29         | 4.13  | 31.14 | -1.9     |
| 30         | 4.14  | 31.43 | -1.69    |
| 31         | 4.08  | 30.94 | -1.7     |
| 42         | 3.73  | 28.89 | -0.95    |
| 44         | 3.3   | 25.41 | -0.99    |
| Irrigation canals|       |     |          | Nile River| 2.3   | 23.4 | 5        |
| 1          | 4.32  | 31.45 | -3.11    |
| 7          | 4.37  | 31.93 | -3.03    |
| 14         | 4.03  | 31.29 | -0.95    |
| 16         | 4.23  | 31.77 | -2.07    |
| 20         | 4.2   | 32.27 | -1.33    |
| 23         | 4.26  | 30.75 | -3.33    |
| 35         | 4.65  | 33.92 | -3.28    |
| 38         | 4.35  | 32.64 | -2.16    |
| 39         | 4.83  | 34.91 | -3.73    |
| 41         | 4.25  | 32.05 | -1.95    |
| 45         | 4.51  | 32.4  | -3.68    |

Fig. 6 δ¹⁸O versus δD in ‰ for collected water samples

Fig. 7 TDS in (mg/l) vs. δ¹⁸O in ‰ for collected water samples
greatly affects groundwater ion compositions through interaction between water and aquifer materials. Saturation Index using PHREEQC software results indicated that the surface (canals, drains) and groundwater samples were undersaturated (dissolution is required to achieve equilibrium) with minerals and gases in order of Al(OH)3 > partial pressure of CO2(g) > Calcite CaCO3 > Aragonite CaCO3 > Gypsum CaSO4·2H2O > Anhydrite CaSO4 > Dolomite CaMg(CO3)2. Negative values of PCO2(g) may reflect the recharge from underlying aquifer and seepage from drains. Surface and groundwater samples have a tendency to precipitate the minerals/gases in order of Hematite Fe2O3 > Goethite FeOOH > Gibbsite Al(OH)3 > Barite BaSO4. The inverse geochemical modeling demonstrated that dissolution of carbonate (calcite, dolomite) and evaporites (gypsum, Anhydrite) minerals with outgassing of hydrogen sulphide as a result of organic matter reduction and microbial processes. Cation-exchange of Ca2+ over Na+ also important geochemical processes in the aquifer system of the area. Environmental stable isotope data revealed that all surface and groundwater samples have enriched values of δ18O and δD compared with stable isotope data revealed that all surface and groundwater cal processes in the aquifer system of the area. Environmental Earth Sciences (2022) 81:105

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