Hydrochemical and stable isotopes indicators for detecting sources of groundwater contamination close to Bahr El-Baqar drain, eastern Nile Delta, Egypt

Mustafa Eissa a, Mohamed Ali b, Ehab Zaghlool c and Orfan Shouakar Stash h

aHydrogeochemistry Department, Desert research Center, Cairo, Egypt; bIsotope Tracer Technologies Inc., Waterloo, ON, Canada

ABSTRACT
Groundwater pollution through the hydraulic connection with wastewater and agricultural drains is a crisis problem facing the developed countries worldwide. Shallow groundwater in the Quaternary aquifer in the eastern Nile delta close to Bahr El-Baqar drain is considered the main source for irrigation and potable water. Geochemistry and stable isotopes (δ 18O, δ 2H and δ 15N) have been utilized to detect the main sources for groundwater contamination. The groundwater samples were classified into two main groups; fresh and brackish water types. The δ 18O and δ 2H elucidate that the fresh groundwater source is mainly from the recently infiltrated drainage water, while brackish groundwater experience extensive evaporation processes and leaching from the aquifer matrix. High ammonia concentration (4.3 to 37.5 mg/l) indicates the influence of anthropogenic activities and seepage from the sewage drains. The δ 15N isotopic content indicates the main groundwater contaminant sources are; sewage disposal, mineral fertilizer, and open drains.

Introduction
In Egypt, groundwater is considered one of the main sources for rural agriculture communities, where it contribute about 7% of water resources, which is restricted to 4.8 billion m³/year (Graveline, 2013). The quality of groundwater depends mainly on the recharge sources, water-bearing formation, surface water seepage, and human activities. In Bahr El-Baqar area, the Nile water is not sufficient to meet demands for the agricultural activities. Therefore, the shallow groundwater in the Quaternary is important for potable and irrigation uses. In the study area, there are two main sources threaten the groundwater quality in the study area; Bahr El-Baqar drain and the agriculture networks drainage that extends from east to west and from north to south (Figure 1). Bahr El-Baqar drain serves an agricultural area of about 119 km²; where it receives an annual amount of 300 million m³ of untreated sewage water that comes from Cairo mixed with different ratios of Nile water (Zahran, El-Amier, Elnaggar, Abd El-Azim, & El-Alfy, 2015). The previous studies indicated hydraulic connections between the Nile water and the shallow Quaternary aquifer in the investigated area (Salem, 1996; Sallouma, 1983). The excessive use of both surface and groundwater in the study area is due to rapid development and population growth (Khalil, Diab., & Atta, 1985). The nitrate (NO₃⁻) and ammonium (NH₄⁺) are common nitrogen compounds dissolved in the polluted groundwater zones in the study area (Mc Mahon and Böhlke, 2006). Nitrate and ammonium are commonly contained in leachate that causes a great threat to shallow groundwater (Koda, Szieczka, & Osinski, 2016). The NH₄⁺ is commonly found in groundwater as a result of anthropogenic activities, leaching from fertilizers, organic waste disposal and leaking sewage systems. The natural level of ammonium in both surface and groundwater should be below 0.2 mg/l, while the NO₃⁻ concentrations should not exceed 2 mg/l in the uncontaminated groundwater (Kresic, 2007; WHO, 2008).

Tracing the contaminant and solute transport in groundwater by geochemical indicators and environmental isotopes offers unique and valuable insights regarding the origin and groundwater movements. Additionally, they allow quantitative evaluation of mixing and other physical processes such as evaporation and isotopic exchange in hydrogeologic systems (Mook, 2000). Stable nitrogen isotopes have been largely used around the world to identify the sources of water pollution. The Nitrogen isotopes can differentiate between the origin of nitrogen source either from wastewater or fertilizers using the definite isotopic signature. On the other side, it is difficult to distinguish between fertilizer nitrate and soil nitrate sources (Lindembaum, 2012). Many authors have used stable nitrogen isotopes for samples containing nitrogen from different sources to identify sources of nitrate and ammonium in groundwater (Jiang, Wu, & Yuan, 2009; Li et al., 2010; Lindembaum, 2012;
Moore, Ekwurzel, Esser, Hudson, & Moran, 2006; Widory et al., 2004; Widory, Petelet-Giraud, Negrel, & Ladouche, 2005).

The main goal of this work is to investigate the influences of Bahr El-Baqar drainage and the anthropogenic activities on the groundwater quality of the Quaternary aquifer. Also, to illustrate the efficiency of using multi-stable isotopes and major ions to improve our understanding of the groundwater flow system in order to achieve sustainability of water resources.

Study area description

Bahr El-Baqar study area lies in the North Eastern portion of Egypt, between longitudes 31º 48’00”\;31º 52’48” East and latitudes 30º 42’36”; 30º 46’12” North. It is located 100 kilometers from Cairo closed to Faqous city and dissected by many open drains (Figure 1).

The study area is characterized by a warm winter, hot summer, and moderate humidity. The annual precipitation ranges from 20 to about 100 mm/year and the daily evapotranspiration varies from 1.5 mm in November to 7.5 mm in July (RIGW-IWACO, 1992). Geomorphologically, the eastern Nile Delta is commonly flat and slopes regionally toward the north and east (El-Shazly et al., 1975). The ground elevations range from the sea level close to Manzala Lake from the northern side to + 35 m a.s.l in the southern side (Said, 1962). The study area is characterized by low relief bounded from the southern side by a moderately elevated plateau essentially occupied by sedimentary rocks belonging to the Quaternary. The northern side is bounded by a landscape with the flat floodplains of the Nile Delta (Gerles, 1994) (Figure 2(a)).

Figure 2(b) shows that the thickness of the Quaternary groundwater aquifer gradually decreases southward (100 m) and increases toward the north close to the sea (900 m). The groundwater level slope generally from south to north (El-Fayounny, 1968). The aquifer is overlain by a silty clay layer, which displays semi-confined conditions, and in some localities, the groundwater
occurs under unconfined conditions. The body of groundwater constitutes huge convex lenses that occupying the entire delta region. The thickness of the freshwater body is directly related to the water-bearing thickness and generally decreased north and eastwards due to saltwater intrusion (Tantawi, 1998). The main source of recharge of the aquifer is the vertical leakage downward from Nile River water, subsurface flow from Ismailia Canal, Damietta branch, irrigation canals and drains, and infiltration of the occasional rainfall. Whereas the discharge of the aquifer is due to over-pumping of groundwater wells for different uses such as irrigation and domestic, and seaward seepage into Manzala Lake and the Mediterranean Sea (Ahmed, AbdSamie, & Badawy, 2013).

Materials and methods

Two field trips were conducted during January 2017, where nineteen groundwater and three surface water samples were collected. The total salinity has been measured in the field using the electrical conductivity meter model GLF-100RW. The pH was measured in the field using portable pH meter model HANNA.
HI8424. The water samples were collected in polyethylene bottles for major ions and heavy metals analysis, which acidified with concentrated HNO₃ to pH < 2 in case of heavy metals. Also twenty-two groundwater and surface water samples were collected in airtight bottles with no headspace for δ¹⁸O and δ²H analyses, and seven groundwater samples were frozen in an ice box for the Nitrogen isotope analysis. These samples run water chemistry for nitrate, sulfate, and chloride, then the amount of water to be used (dependent on nitrate conc.) and the amount of barium chloride to add to each sample (dependent on sulfate concentration) were calculated. Ultrapure barium chloride powder has been used to precipitate the BaSO₄ and then samples have been centrifuged and Filtered out then run samples through the resin (IT₂ laboratory).

The analyses of major and minor ions (Calcium, Magnesium, Sodium, Potassium, Ammonium, Sulfate, Chloride, Bromide, Nitrate and Nitrite) were determined using ion chromatography (Dionex, ICS-1100). Carbonate and bicarbonate were measured titrimetrically against sulphuric acid by neutralization method using phenolphthalein and methyl orange as indicators (Table 1).

Total dissolved solids (TDS) in mg/l were determined according to the following equation:

\[
\text{TDS} = (\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+ + \text{CO}_3^{-} + (\text{HCO}_3^{-}/2) + \text{SO}_4^{2-} + \text{Cl}^{-})
\]

For quality assurance the major ions were estimated from the electrical balance (E.B.) according to the following equation (Appelo and Postma, 2005):

\[
\text{E.B.} = \left(\frac{\text{Sum Total Cations}}{\text{Total Anions}}\right) 
\]

\[
\times 100
\]

Where the concentrations of total cations and total anions are represented in milliequivalent per liter. To confirm the water the quality assurance, the acceptable range for (E.B., %) is within ±2%. The Environmental stable isotopes (δ¹⁸O, δ²H, and δ¹⁵N) were carried out at the IT₂ laboratory in Waterloo, Canada (Table 2). The δ¹⁸O and δ²H samples were carried out using Cavity Ring-Down Spectroscopy the CRDS Model LI230-i, Picarro. The standard used was IT₂-10B/IT₂-13A/ IT₂-00 Calibrated with IAEA Standards (V-SMOW, SLAP, and GISP). The δ¹⁵N samples were carried out using Delta Plus Isotope Ratio Mass Spectrometry (IRMS), Finnegan MAT, Germany, coupled with an Elemental Analyzer EA 1110 CHN, CE Instruments, Italy stander used was IT₂-601/IAEA-N1/IAEA-N2/ Acetanilide (B-2000).

### Results and discussion

#### Hydrochemical characteristics

The chemical analyses of the surface water and groundwater show that pH values are ranged between 7.5 and 8.2, reflecting mild alkaline water (Table 1). The surface water salinity is varying from 200 to 736 mg/l, reflecting the effect of ecological activities on drain water samples (S₁ and S₂). The water type of two surface water samples (S₁ and S₂) is Na-HCO₃, O and

#### Table 1. Chemical, isotopic composition and saturation index data of groundwater and surface water samples collected in January 2017.

| No | PH | EC µmols/cm | TDS mg/l | Mg mg/l | Ca mg/l | Na mg/l | K mg/l | CO₃²⁻ mg/l | HCO₃⁻ mg/l | SO₄²⁻ mg/l | Cl mg/l | NO₃⁻ mg/l | NH₄⁺ mg/l | E.B.% |
|----|----|-------------|----------|----------|---------|---------|--------|-----------|-----------|-----------|---------|-----------|----------|-------|
| 1  | 8.0| 2790        | 1643     | 36.8     | 100.7   | 248.9   | 6.9    | 0.0       | 628.3     | 337.0     | 355.5    | 40.41    | 37.5     | 0.02  |
| 7  | 7.9| 1350        | 883      | 34.1     | 90.2    | 173.1   | 7.4    | 0.0       | 445.3     | 166.3     | 107.2    | 82.08    | 10.6     | 0.02  |
| 8  | 8.0| 1185        | 696      | 16.7     | 279.0   | 221.5   | 3.8    | 0.0       | 506.3     | 103.2     | 68.6     | 1.91     | 19.6     | 0.04  |
| 9  | 8.0| 1100        | 661      | 25.1     | 55.8    | 158.3   | 6.7    | 12.0      | 331.9     | 120.5     | 87.7     | 29.86    | 17.1     | 0.00  |
| 10 | 8.0| 1010        | 598      | 24.5     | 55.1    | 140.5   | 7.4    | 0.0       | 341.6     | 113.9     | 77.7     | 9.01     | 9.9      | -0.01 |
| 11 | 8.0| 930         | 531      | 21.5     | 80.9    | 80.8    | 6.4    | 0.0       | 390.4     | 71.9      | 38.6     | 36.26    | 14.3     | 0.04  |
| 12 | 7.9| 933         | 534      | 19.8     | 74.2    | 97.0    | 6.1    | 0.0       | 305.0     | 93.9      | 83.9     | 5.65     | 10.2     | -0.01 |
| 13 | 7.9| 935         | 572      | 25.3     | 75.3    | 94.9    | 6.5    | 6.0       | 353.8     | 84.4      | 4.8      | 47.94    | 14.8     | 0.03  |
| 14 | 7.9| 1305        | 883      | 34.1     | 90.2    | 173.1   | 7.4    | 0.0       | 445.3     | 166.3     | 107.2    | 82.08    | 10.6     | 0.02  |
| 15 | 7.9| 1315        | 736      | 24.9     | 74.9    | 156.2   | 17.9   | 0.0       | 335.5     | 127.4     | 167.6    | 0.02     | 28.0     | 0.05  |
| 16 | 7.9| 492         | 200      | 13.62    | 50.4    | 80.8    | 6.4    | 0.0       | 390.4     | 71.9      | 38.6     | 36.26    | 14.3     | 0.04  |
| 17 | 7.9| 1185        | 534      | 18.1     | 65.3    | 104.6   | 21.0   | 0.0       | 372.1     | 43.2      | 9.66     | 33.2     | 0.08  |

S₁: Bahr El-BAṣar Drain S₂: Irrigation canal S₃: Developed irrigation project canal N: Not detected - Not measured
while S₂ sample has an Mg-SO₄ type. According to the recorded total salinity of the investigated groundwater samples, two main categories were observed. The first category is the freshwater type (TDS < 1000 mg/l) representing 68% of the groundwater samples, while the second category is the brackish water type (1000 < TDS > 3000 mg/l) representing 32% of all the groundwater samples. The total groundwater salinity can be classified into three groups (Figure 3). The groundwater Group I (samples 9, 10, 16 and 17) with salinity less than 500 mg/l, reflecting the direct influence of fresh water

\[
\begin{array}{cccccc}
\text{Model} & \text{Type} & \text{Group} & \text{Initial Water} & \text{Final Water} & \text{Mixing Ratio (%)} \\
\text{Reaction} & I & S_2 & 9 & - & - \\
\text{Model} & & & 10 & - & - \\
\text{Mixing} & II & S_1 & S_2 & - & 7 \\
\text{Model} & & & 8 & 87.3 & 12.7 \\
\text{III} & S_1 & S_2 & 5 & 52.0 & 48.0 \\
\end{array}
\]

Table 3. NETPATH reaction and mixing model results for groundwater close to Bahr El Baqar.
from the surface water drainage. The groundwater Group II (samples 7, 8, 11, 12, 13, 14, 15, 18 and 19) with salinity between 500 to 1000 mg/l, reflecting mixing with surface water and the evolution of groundwater due to leaching and dissolution of aquifer matrix.

Finally, Group III (samples 1, 2, 3, 4, 5 and 6) of salinity values more than 1000 mg/L indicating the final stage of groundwater evolution.

The nitrate and ammonium are dissolved constituents give valuable insights of the anthropological impact and connection between surface and groundwater. In Figure 4, the distribution contour map of nitrate concentration increases toward the northwest with a maximum concentration of 82.1 mg/l. The ammonium concentration increases towards the northern and the eastern sides (Figure 5) with a maximum value of 37.49 mg/l, indicating possible contamination with bacteria, sewage and animal waste (WHO, 2008).

### Water classification and evolution diagrams

Piper’s diagram (Piper, 1945) shows the average compositions of fresh water and sea-water and factors affecting the groundwater salinization (Figure 6). Generally; the surplus of Na⁺ of all collected groundwater samples indicate the dilution and freshening the shallow Quaternary aquifer due to direct seepage of surface water. The groundwater could be classified into main three groups. The Group I; located in sub-areas 2, 3 and 6 having Ca-HCO₃ water type which indicates the fresh water is mainly recharged almost exclusively from irrigation canals (Appelo and Postma, 2005).

**Figure 4.** Distribution of Nitrate (mg/l) in the groundwater samples tapping the Quaternary aquifer.

**Figure 5.** Distribution of Ammonia (mg/l) in the groundwater samples tapping the Quaternary aquifer.
The Group II; plot in the sub-areas 3 and 6 and having Na – HCO₃ and Ca – Na – HCO₃ water types, respectively; reflecting the recharge from the surface water system and leaching of soil zone and aquifer matrix through the infiltration paths. The enrichment of calcium in these groundwater samples with depletion of Na suggested reverse cation exchange processes, where calcium in the aquifer system would be exchanged by sodium from groundwater. Group III; plotted in the sub-area 4 and having Na-Cl type, indicating increasing of chloride concentrations as a result of evapotranspiration process (Appelo and Postma, 2005). The results show the relevance of Na⁺ concentration than Cl⁻, while Ca²⁺ ions relatively decrease, suggesting cation exchange of Ca²⁺ for Na⁺ through the soil zone.

Durov diagram (1948), has been used to indicate the mixing between the surface water and groundwater (Figure 7). The majority of groundwater samples are plotted in 2, 4 and 5 fields, indicating direct recharge of surface water, otherwise; mixed water or simple dissolution with reverse cation exchange process.

Stable isotopes

Oxygen-18 and δ²H isotope
The variation of δ¹⁸O and δ²H values in the groundwater (Table 2) are mainly depending on the source of recharge and consequent geochemical processes dominated in the aquifer. The groundwater in the Quaternary aquifer had a wide range of δ¹⁸O (+4.06 to +5.56 %) and δ²H (+19.4 to +30.8 %) values (Table 2). The δ¹⁸O and δ²H relationships are consistent with the former geochemical classifications, where, it reveals three groundwater groups (Figure 8). The Groups I and II have isotopic signature close to the surface water samples and plot under the Global Meteoric Water Line (Craig, 1961), on the mixing trend line between the surface water samples (S1, S2, and S3) and the recent Nile water of Ismailia Canal. This demonstrates the recent replenishment of the shallow Quaternary aquifer from the irrigation canals and wastes drainage with insignificant evaporation processes prior to infiltration. The groundwater Group III is characterized by the enrichment of δ¹⁸O and plot on the evaporation trend line indicates that these waters were affected by evaporation processes prior seeping from the recent and the paleo Nile waters.

The relation between the groundwater salinity and δ¹⁸O (Figure 9) confirms the origin groundwater Group I and Group II are from the infiltration of irrigation water, where they are mostly plot on the mixing trend line between the Ismailia Canal and the low saline surface water. The groundwater Group III has high groundwater salinity which exceeds the surface water and enriched with the isotopic signature of δ¹⁸O due to extensive evaporation (Figure 9).
Legend

- Green circle: Group I (TDS < 500 mg/l)
- Yellow circle: Group II (500 < TDS < 1000 mg/l)
- Red circle: Group III (TDS > 1000 mg/l)

Figure 7. Durov diagram (Durov, 1948) for the collected groundwater and surface water samples.

Figure 8. $\delta^{18}$O versus $\delta^{2}$H content of the surface drains and groundwater in the shallow Quaternary aquifer.
Nitrogen isotopes

Groundwater contamination by nitrate represents a widespread problem around the world (Foster, Hirata, Gomes, D’Elia, & Paris, 2002). The sources of groundwater pollution by nitrate are mainly from sewer and septic tank leakage, the leaching of nitrogen-based chemicals and fertilizers, and polluted surface water infiltration. Additionally, the nitrification of soil-borne ammonium and from nitrification of mineralized organic matter (Ford & Tellam, 1994; Wakida & Lerner, 2005). Stable nitrogen isotopes have been used largely around the world to identify the source of pollution in surface and groundwater (Lindenbaum, 2012). The variation of the nitrogen stable isotopes values (Table 2) reflected the source of contamination and identify sources of nitrate and ammonium in groundwater. The isotopic signature of the $\delta^{15}N$ range between 5.5 to 34.8 % at site 14 and

Figure 9. $\delta^{18}O$ versus groundwater salinity of the surface drains and groundwater in the shallow Quaternary aquifer.

Figure 10. $\delta^{18}O$ versus $\delta^{15}N$ content of the groundwater in the shallow Quaternary aquifer, Bahr El-Baqar area, east Nile delta.
site 15, respectively. Figure 10 shows the plot diagram of $\delta^{15}N$ versus $\delta^{18}O$ for selected seven groundwater samples according to previous works (Clark & Fritz, 1997; Finlay & Kendall, 2007). The isotopic signature suggests that either the manure or sewage in addition to the chemical fertilizers are the main sources of nitrate in the study area, nevertheless some samples show denitrification trend, this process was significant in wells 1, 12 and 15, which indicated mixing with surface water (Mariotti, Landreau, & Simon, 1988). These results compatible the previous hydrochemical interpretation which confirms the interaction between surface water (Bahr El-Baqar and agricultural drains) and groundwater in the study area and either the probability of septic tank influence on the groundwater in the study area.

**Geochemical modeling**

The inverse geochemical model NETPATH 2.0 (Plummer, Prestemon, & Parkhurst, 1994) was used to interpret the geochemical processes and to compute the mixing proportions of different water resources according to changes of chemical composition in groundwater along a flow path or the effect of surface water recharge. In this study, the major ions concentration and the mineral phases in the quaternary aquifer were adopted in the water–rock reaction models. The relation between TDS (mg/l) and $\delta^{18}O$ %, Figure 9, shows that the water samples in the investigated area can be classified into three groups (I, II and III). The NETPATH model results show the groundwater samples group I are mainly originated from the infiltration of irrigation water (Initial water, $S_2$) that has been evolved due to dissolution of halite while gypsum while precipitation of calcite and dolomite minerals (Final water, sites 9, 10, 16 and 17) (Tables 2 and 3). Whereas groundwater group II are mainly originated from mixing and evolution of both Bahr-El-Baqar drain drain ($S_1$) and irrigation canal ($S_2$) (Initial Waters), though infiltration and dissolution of aquifer matrix (Table 3). The mixing ratios between 24.5 % to 87.3 % from Bahr El-Baqar drain ($S_1$) to 12.7 % to 75.5 % from the irrigation canal ($S_2$) (Table 3).

Finally, the origin of Group III samples is mainly from two types of mixing processes: 1) mixing between both agricultural drainages ($S_3$) and the original groundwater represented by sites 3 and 2) mixing between agricultural drainages ($S_3$), irrigation canal ($S_2$) and the original groundwater represented by site 5 with different mixing ratios (Table 3). The estimated NETPATH mixing ratios ranges between 31.7 % to 37.1 % from the agriculture drains ($S_3$), 4.6 % to 7.3 % from the irrigation canals and 55.6 % to 68.3 % from the original groundwater.

**Conclusion and recommendations**

The groundwater geochemistry and environmental stable isotopes approach were used to investigate the contaminant sources that deteriorate the groundwater quality in the shallow Quaternary aquifer at Bahr El-Baqar area. The geochemistry reveals three main groundwater groups. The groundwater Group I, have groundwater salinity less than 500 mg/l. The groundwater Group II has groundwater salinity ranges between 500 to 1000 mg/l. The Group III has groundwater salinity ranges between 1000 to 5000 mg/l. The Piper and Durov’s diagrams indicated that the main factors controlling the groundwater quality are mixing with surface water drains, evaporation, and ion exchange processes. High concentration of ammonium in the groundwater samples have been detected which indicated the influence of anthropogenic activities, fertilizers, organic waste disposal and leaking sewage systems on groundwater aquifer. The relation between $\delta^{18}O$ and $\delta^2H$ indicated that the groundwater source in the study area is mainly originated from direct infiltration of the surface water and the mixing between Pleistocene aquifer water with the surface water (Irrigation canals, agricultural drains and Bahr-El Baqar sewage drain). The $\delta^{15}N$ isotopic content in the groundwater samples demonstrates the contamination in the study area are coming from sewage disposal, mineral fertilizer, septic tanks and Bahr El-Baqar sewage drain. Finally, in order to stop the contamination sources in the study area, the subsequent basis should be taken. Routine checks of the quality of the groundwater and establishment of environmental monitoring stations in the study area are necessary to ascertain its pollution status. The Egyptian government, non-governmental agencies and international organizations should join hands together to ensure the adequate and safe drinking water provided for inhabitants. It is recommended also to control and restrict the agricultural activities in the study area to prevent the leachate from reaching the groundwater. Enforcement of suitable verdicts and regulations in order to manage pollutants that may affect groundwater quality. Sewage disposal and septic tank systems are considered one of the sources of groundwater pollution in the area, so healthy drainage system for sewage must be constructed.

**Highlights**

1. Groundwater Geochemistry Eastern Nile Delta, Egypt
2. Utilizing N-15 isotopes for detecting Quaternary groundwater pollution
3. Utilizing Isotopes (18O and 2H) for determination of aquifer recharge sources
4. Sewage drains and fertilizers are the main sources for groundwater pollution
Disclosure statement

No potential conflict of interest was reported by the authors.

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