Integrated Geochemical Indicators and Geostatistics to Assess Processes Governing Groundwater Quality in Principal Aquifers, South Sinai, Egypt

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ABSTRACT

The groundwater system in the principal aquifers situated in the Southern Sinai was regionally investigated, using hydrochemical tools and geostatistical technique to evaluate the recharge sources and salinization origins which consider the main constraint for sustainable development in such an arid region. The environmental stable isotopes (δ\textsubscript{18}O and δ\textsubscript{2}H), groundwater salinity, conservative ions (Cl and Br), ion ratios and the sea water mixing index (SWMI) were utilized to identify the salinization mechanism and to delineate the recharge for different aquifers (Quaternary, Miocene, U. Cretaceous, L. Cretaceous and Precambrian) situated in the upstream watersheds and along the coastal regions. The regional study depends on four hundred and sixty-eight groundwater samples tapping the main aquifers. The geochemical data have been analyzed statistically to estimate the seawater mixing index (SWMI) in order to delineate the deteriorated aquifer zones. The environmental stable isotopes confirm the upstream of Gharandal, Watir, Dahab basins and Saint Catharine areas receives considerable amount of the annual precipitation that could be managed for sustainable development. The hydrochemical ion ratios and the SWMI values give good insights for aquifer salinization, where; mixing with seawater intrusion in the downstream coastal aquifers, leaching processes of minerals in the aquifer matrix, evaporation processes are considered the main sources for aquifer deterioration. The hydrogeochemistry shows the injection of high saline water comes from the desalination plant deteriorates the coastal Miocene aquifer located in Sharm El Shiekh and Nabaq areas.

Keywords: Groundwater chemistry, Statistics, rCl/rBr, Stable isotopes, South Sinai Peninsula

Introduction

The South Sinai Peninsula is located in the coastal arid region, where the groundwater recharge is limited and seawater intrusion threatens the groundwater quality due to the over withdrawal. The groundwater is the main source of drinking and potable uses for about 177,900 inhabitants (South Sinai, 2012). Groundwater salinization is an environmental global phenomenon that affects many different aspects of our life (Williams, 2001). The groundwater quality deterioration is considered one of the most important challenges facing the sustainable development strategies in arid and semiarid areas. In the coastal arid aquifers, groundwater salinization processes mainly arisen from seawater mixing, geogenic sources, evaporation, and dissolution as well as the anthropogenic activity such as over-exploitation and rejecting brine deeper into aquifer saturated zone (S´anchez-Martos et al., 2002; Indu et al., 2013).

Recently, the environmental stable isotope in conjunction with conservative ions rCl/rBr as well as other geostatistical and geochemical tools have been used to explain the factors deteriorate the groundwater quality (Alcala and Custodio, 2008; DeMontety et al., 2008; Carol et al., 2009; Ben Hamouda et al., 2010; Han et al., 2011). The Cl/Br ratio gives good insights related to the groundwater contamination and mixing due to its constant value from different end members. The value of rCl/rBr can not significantly modified by the anthropogenic or the physical processes taking place in soil including dilution, evaporation, transpiration (Samantara et al., 2015). Stable isotopes (δ\textsubscript{18}O and δ\textsubscript{2}H) are used to identify recharge source of groundwater (Clark and Fritz, 1997; Mook, 2001) and the mixing with seawater in coastal aquifers (Leduc et al., 2007). The previous studies (Mazor et al.,1973; Gat and Issar, 1974; Tantawi et al. 1998; Abd El Samie and Sadek, 2001;
Saied, 2004; Zaghlool, 2006; Rosenthal et al., 2007; El-Sayed et al., 2011; Eissa et al., 2016; Isawi et al., 2016) have been conducted on the Southern Sinai aquifers using hydrochemical ratios and environmental stable isotopes as a tracer to identify the origin of salinity. However, no regional research has been conducted on the global aquifers located in the South Sinai governorate, in order to delineate the promising localities and aquifer zones that receive significant and sustainable recharge from annual precipitation. In this research the geochemical indicators, statistical analyses and isotopic tracers have been utilized to identify the main recharge sources for different aquifers and sources that cause groundwater deterioration for sustainable developmental strategies.

Study Area

The study area located in the southern part of Sinai Peninsula located in between longitude 32º 00’ to 35º 00’ (East) and latitude 28º 00’ to 30º 30’ (North). It covers an area about 31,272 Km² and bounded from the north by the North Sinai governorate, from the South by The Red Sea, from the East by the Gulf of Aqaba and from the East by the Gulf of Suez (Figure. 1). South Sinai generally characterized by the arid climate, where the annual mean temperatures range from 18 ºC to 33 ºC, with less than 50 mm of annual precipitation and the wind blows from the northwest in summer and from the southwest in winter (Caragnano et al., 2009; Hercher, 2015).

Fig. 1: Location map of the Southern Sinai, Egypt

The South Sinai region comprises four main principle aquifers; the Precambrian (basement rocks), the Miocene, Cretaceous, and the Quaternary aquifer. The Precambrian aquifer, exposed in
the upstream watershed areas and is mainly composed of granites, and gabbros. The groundwater occurs in the fractured media and in the weathered surface zone up to 5m depth. The Precambrian aquifer in South Sinai receives an annual recharge estimated 52,000 m³/day (Dames and Moore, 1985).

![Fig. 2: The main surface geologic setting of Southern Sinai, Egypt (Omara, 1972; Kora, 1995; Abdel Zaher et al., 2014).](image)

The Cretaceous aquifer are represented by the middle and the lower Cretaceous units. The lower Cretaceous unit (Nubian sandstone) consists of sandstone together overlayed with the Cenomanian sandstone that has been considered the upper unit of the Nubian sandstone (Dames and Moore, 1985; Sultan et al., 2011). The tertiary units serving as aquifers, belonging to the lower Miocene age and formed mainly of sandstone and grits, at the basal Miocene unit (Shata, 1992). The aquifer thickness together with the upper confining unit varies between 100 m near its outcrops along the eastern catchments and gradually increases westward to reach about 450 m thick along the Gulf of Suez. The aquifer discharges westward into the Gulf of Suez with minor discharge represented by natural springs along shoreline at Hammam Faroun and Hammam Mousa springs (El-Rayes et al., 2014). Quaternary aquifers, is mainly composed of sands, gravels, and silt mixed with rock fragments in wadi alluvial along many of the major wadis and at the downstream deltas. The aquifer thickness varies between 3 and 150 m and the depth to water level ranges between 2 m and 47 m (El-Rayes et al., 2014).

**Methodology**

The water-chemistry data analyzed in this paper include water samples collected from the previous publications in South Sinai during the last decades. These data included 468 groundwater samples from 5 principal aquifers, 3 seawater samples and 2 rain sample (Table, 1), (Figure. 3). The
chemistry data of all water samples are available, (Isawi et al., 2016; Eissa et al., 2016; Abouelmagd et al., 2014; Omran, 2013; El-Sayed et al., 2011; El-Fiky, 2010; El-Fiky, 2009; Rosenthal et al., 2007; Zaghlool, 2006; Saied, 2004; Abd El Samie and Sadek, 2001; Gorski and Ghodeif, 2000; Tantawi et al., 1998; Gat and Issar, 1974; Mazor et al., 1973). Seawater mixing index was used to estimate the relative degree of seawater mixing in the principal aquifers in the study area, this index is based on four major ionic constituents concentrations in seawater (Na, Mg, Cl, and SO_4) as follow.

\[
SWMI = a \frac{C(Na)}{T(Na)} + b \frac{C(Mg)}{T(Mg)} + c \frac{C(Cl)}{T(Cl)} + d \frac{C(SO_4)}{T(SO_4)}
\]

a, b, c, and d are constants denote the relative concentration proportion of Na, Mg, Cl, and SO_4 in seawater, respectively (a = 0.31, b = 0.04, c = 0.57, d = 0.08); C is the measured concentration of water samples in mg/L and T is the regional threshold values of the considered ions estimated from the interpretation of cumulative probability curves (Mondal and Singh, 2011).

**Fig. 3**: Map showing the location of groundwater samples of the main aquifers in Southern Sinai, Egypt.

**Results and Discussion**

1. **Groundwater Geochemistry:**

The study area characterized by a wide range of groundwater salinity. The groundwater salinity ranges between 211 mg/L at to 53347 mg/L. The groundwater in different aquifers has been classified according to the salinity into; fresh water (<1000), brackish water (1000 to 10,000), saline water (10,000 to 100,000) and brine water (>100,000) (Freeze and Cherry, 1979) (Table 1, Figures 4 and 5). In general the highest groundwater salinity in south Sinai aquifers has been recorded in the Miocene aquifer, while the most fresh groundwater in the upper cretaceous aquifer.
Fig. 4: Map showing groundwater classification based on the groundwater salinity (mg/L) according to Freeze and Cherry, (1979).

Fig. 5: Box plots showing the salinity (mg/L) of groundwater samples in the main aquifers located in Southern Sinai, Egypt.

The lower groundwater salinity is locally recorded at the upstream portion of watersheds basins draining toward the Gulf of Suez and Aqaba draining system. However, higher groundwater salinity is recorded locally along the coastal zones and close to the desalination plants in Sharm El Sheikh area. The most fresh groundwater samples represented by the U. Cretaceous, L. Cretaceous (Nubian sandstone) Quaternary, and Precambrian aquifers located in St. Catherine mountains, El-Qaa plain at El-wadi village, and in the upstreams of Dahab (Saal and Nassab), Surd and Gharandl basins. This
mainly attributed to the implications of recent recharge comes watershed of south Sinais' aquifers. The areas have lower groundwater salinity could be considered as promising localities for sustainable development. Brackish groundwater type is represented by the Quaternary, the Miocene, the Upper Cretaceous, the Lowe Cretaceous (Nubian sandstone), and the Precambrian aquifers located in the coastal plains, downstreams of Sudr, Warden and Watir basins as well as some localities of El-Qaa plain, and in the north of the study area at Nekhel and Hasana areas. The brackish groundwater zones are considered a favorable locality for low-cost groundwater desalination. The saline groundwater represented in the Quaternary, Miocene, and L.Cretaceous (Nubian sandstone) aquifers located along the coastal plain of the Suez gulf and Aqaba Gulf. The hypersaline groundwater has been recorded in Sharm El Shiekh and Nabaq areas are mainly due to the impact of brine disposal deeper into the coastal aquifer (Isawi et al., 2016).

Table 1: Water chemistry data for groundwater samples tapping different aquifers located Southern Sinai, Egypt.

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<th>PH</th>
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<th>Mg</th>
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<th>K</th>
<th>CO₂</th>
<th>HCO₃</th>
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2. Indicators of groundwater salinization

The rNa/rCl (meq/L) ratio is used to discriminate between the origin of groundwater, where meteoric water have a ratio more than unity while the rNa/rCl value less than unity indicates the contribution of marine sediments and/or seawater mixing (Figures 6). The ratio could reach unity due to the mixing of seawater and freshwater (Ivanov et al., 1968). The rNa/rCl ratio of the Quaternary, Eocene, Miocene, and L. Cretaceous aquifers in the coastal areas record values less than unity, reflecting the contribution of marine origin, while the all groundwater samples located in the inland areas have values of rNa/rCl more than unity reflecting meteoric origin and the contribution of recent recharge comes from precipitation.

In Figure 7, the graphical relation between chloride and electrical conductivity (EC) (kelly, 2005) shows three distinctive groundwater zones. The first zone is groundwater that has low salinity; the second zone characterizing groundwater that has relative mixing with seawater and the third zone has a groundwater salinity greater than seawater and reveal the impact of reject brine injection deeper into the groundwater aquifer. The Precambrian and the inland Quaternary aquifers has no great
influence of seawater mixing, while the Quaternary and the Miocene coastal aquifers are mostly influenced by seawater intrusion.

**Fig. 6:** Binary diagram of Na/Cl versus Cl (meq/L) for the groundwater samples in the study area.

**Fig. 7:** Chloride vs Electrical conductivity for the groundwater samples in the study area.
3. Statistical Analyses and Groundwater Mixing Sources

The seawater mixing index (SWMI) gives good indication of the mixing with different waters as well as the origin of groundwater. When the calculated SWMI value is greater than unity, the water may be considered to obviously record the effect of seawater mixing (Han et al., 2015). The estimation of the inflection points corresponding to regional threshold Ti values (Sinclair, 1974) for the major ions (Na, Mg, SO₄, and Cl) were calculated depending on the statistical cumulative probability curves for the principal aquifers in the study area (Figure, 8a). According to SWMI values (Table 2), it is found that the groundwater samples can be classified into three groups (Figure, 8b), group A which have SWMI values less than 1 and TDS less than 16800 mg/L. This values have been detected in the groundwater aquifers located mainly in the upstream portions of different basins including the Quaternary, Eocene, L. Cretaceous, U. Cretaceous, and Precambrian aquifers. Therefore, the main sources for the groundwater salinization in these aquifers are restricted to the water-rock interaction and leaching of marine origin. Group B; characterizing coastal groundwater aquifers located in the downstreams, that have SWMI values more than 1 and groundwater salinity ranges between 4360 to 38957 mg/L which is close to seawater reflecting possible seawater intrusion and/or leaching of marine deposits. Group C, characterizing groundwater aquifers located in Sharm El Sheikh area and Nabq area, that have SWMI values greater than 10 and groundwater salinity exceeding the seawater (more than 47000 mg/L), reflecting the impact of brine water disposal from desalination plants in the Miocene aquifer. According to the SWMI values we can identify the locations which can be influenced by seawater intrusion (Figure, 8.c), this map reflected the sea water intrusion is clearly appeared in the Quaternary aquifer at Nuwbiaa city and in the Miocene aquifer at Nabq area. The groundwater samples have SWMI greater than one and located in the upstream portion indicating the impact of leaching of marine deposits.

Table 2: Sea water mixing index (SWMI) and reflection point (R) values for groundwater samples tapping different aquifers located Southern Sinai, Egypt.

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<th>Item</th>
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<th>SWMI</th>
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</tr>
<tr>
<td>L. Cretaceous (No of samples: 44)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na</td>
<td>678</td>
<td>0.089</td>
</tr>
<tr>
<td>Mg</td>
<td>224</td>
<td></td>
</tr>
<tr>
<td>SO₄</td>
<td>1250</td>
<td></td>
</tr>
<tr>
<td>Cl</td>
<td>1450</td>
<td></td>
</tr>
<tr>
<td>Precambrian (No of samples: 18)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na</td>
<td>643</td>
<td>0.019</td>
</tr>
<tr>
<td>Mg</td>
<td>182</td>
<td></td>
</tr>
<tr>
<td>SO₄</td>
<td>1649</td>
<td></td>
</tr>
<tr>
<td>Cl</td>
<td>1449</td>
<td></td>
</tr>
</tbody>
</table>
Figure 8a. Statistical normalized probability curves for Na⁺, Mg²⁺, Cl⁻ and SO₄²⁻ for groundwater samples in the study area (S.W sea water, R.W rain water, R reflection point).
4. Endogenous Salinization Processes

This relation is one of the most important relations to illustrate the natural mechanism that change the groundwater chemistry, including rock weathering, evaporation and participation dominance. According to the relationship between the groundwater salinity and the (Na⁺/[Na⁺ + Ca²⁺]) ratio (Figure 9), the groundwater could be classified into four unique classes (Gibbs, 1970). The groundwater Class A; representing fresh groundwater sample of the Quaternary aquifer located in
the upstream of Wadi Dahab Basin at Saal Sub-Basin indicating the superior groundwater replenishment and the rock weathering is the main controlling factor affecting the groundwater salinity. The groundwater Class B; plotted into the marginal area dominating the rock weathering and closer to the zone dominating evaporation-precipitation. Class B represents the watershed aquifers of the Quaternary, U. Cretaceous, L. Cretaceous (Nubian sandstone), Carboniferous, Cambrian and Precambrian. These aquifers are mainly located in St. Catherine mountains, El-Qaa plain (El-wadi village), upstream of Sudr, Wadi and Gharandl basins. In Gibbs diagram (Figure. 9) the coastal groundwater samples of the Quaternary aquifer (Class C) are plotted close to the precipitation and evaporation dominance region. Class D; is representing the Miocene coastal groundwater aquifer in Sharm El Shiekh area, that has been plotted in the upper right side and has salinity exceeding the seawater samples indicating the impact of seawater intrusion and mixing with disposal of high saline reject water comes from the desalination plants.

![Gibbs plot](image)

**Fig. 9:** Gibbs plot explains groundwater chemistry and geochemical process in the study area.

The Cl and Br ions behave conservatively in potable groundwater which leads to that the Cl/Br ratios remain unaltered because these ions have varied abundances in natural fluids and solids (Davis et al., 1998). Recently, many studies used Cl/Br ratios to identify the groundwater flow, to determine the main source of groundwater salinization and to evaluate the seawater intrusion in coastal aquifers (Fontes et al., 1986; Cartwright et al., 2006; Han et al., 2011). Moreover, Cl/Br ratio has been used to distinguish the influence of groundwater contamination from wastewater sources and other anthropogenic impact (Davis et al., 1998; Katz et al., 2011; McArthur et al., 2012; Samantara et al., 2015) and identification of the origin of salinization in groundwater (Sánchez-Martos et al., 2002; Kim et al., 2003; Alcala and Custodio, 2004; Alcala and Custodio, 2008; Skrzypek et al., 2013; Li et al., 2016; Eissa, et al., 2016 and Ouhamdouch, et al., 2017).
In Figure 10a, the Quaternary, the L. Cretaceous and Precambrian aquifers show positive correlation coefficients (R ranges from 0.42 to 0.79) between the concentration of dissolved chloride and bromide ions in groundwater. In Figure 10b, the Quaternary, L. Cretaceous and Precambrian groundwater samples are plotted upper left side on the mass balance mixing line between the recharge and seawater, reflecting the main source of groundwater mineralization is leaching and dissolution of aquifer matrix (Alcala and Custodio., 2004). Additionally, that indicating insights for high potential recharge from the annual precipitation at El Wadi, St. Catherine, Saal, Nassab, Sudr, Gharandl basins and El-Qaa plain (El Wadi village). The high Cl concentrations with approximately low values of Cl/Br ratio (C) reflects the impact of marine origins (Alcala and Custodio, 2008) and/or relative percentages of seawater intrusion in the coastal aquifers with some agricultural activities. The Br versus Cl (meq/l) plot presents a positive correlation for the Miocene groundwater located in Sharm El Shiekh and Nabaq areas, has a correlation coefficient value close to 1 (R = 0.91), indicating the same origin of groundwater salinization, where the reject brine disposal is the main source for groundwater salinization.

**Fig. 10 a:** Br vs Cl (meq/l) Plot for principle aquifers in South Sinai.
5. Delineate Recharge Zones Using Environmental Isotopes:

The environmental stable isotopes have been used to delineate the groundwater recharge and salinization sources in coastal arid aquifers worldwide (Eissa et al., 2018). According to the isotopic content of the δD and δ18O, the groundwater in the investigated aquifer can be classified into four groups. The Group A; has δ18O ranges between -10 and -6 % VSMOW, which are more depleted compared with the isotopic print of the recent precipitation in South Sinai (Eissa et al., 2013). These water reflects paleo-groundwater where the groundwater samples have plotted close to the Global Meteoric Water Line (GMWL) (Craig, 1961). This group represent the U. Cretaceous and the L. Cretaceous (Nubian sandstone) aquifers. The Group B, have δ18O ranges between -6 and -2 %VSMOW, and plotted between Global Meteoric Water Line (GMWL) and the Mediterranean Meteoric Water Line (M.MWL) (Gat et al., 1969) closed to the recent precipitation. This group represent groundwater samples tapping the Quaternary, U. Cretaceous, L. Cretaceous, and Precambrian aquifers. The range of the δ18Oand δ2H indicate the contribution of the recent replenishments and sustainable recharge for these aquifers, Group C, characterized by the enrichment of δ18O and δ2H values and aligned on the evaporation trend line and along the sea water mixing line. This group is mainly represent by the coastal Quaternary and the inland Miocene aquifers. Group D, specified by high δ18O and δ2H values reflected the influence of disposal of reject of desalination plants on the Miocene aquifer in Nabaq area.

The relation between δ18O and d-excess was used to predict the effect of evaporation processes on the isotopic composition of groundwater samples as well as the paleo climatic conditions during the precipitation time. In Figure 12, the L. Cretaceous aquifer exhibits higher value of the d-excess and the most depleted δ18O content reflecting a paleo-water source with cooler climate and noneffect of evaporation during the recharge processes. The Quaternary and Miocene groundwater, exhibits lower d-excess values and enrichment with the δ18O content indicating the dominance of evaporation processes (El-Sayed et al., 2011).
Fig. 11: $\delta^2$H vs $\delta^{18}$O for the groundwater tapping the principle aquifers in Southern Sinai, Egypt.

Fig. 12: The d-excess vs $\delta^{18}$O for the groundwater tapping the principle aquifers in Southern Sinai, Egypt.
In Figure 13, the relation between $\delta^{18}$O and chloride (mg/L) indicates that the groundwater samples of the U. Cretaceous aquifer (group A) are not affected by evaporation processes where the main source of aquifer salinization is leaching and dissolution processes (water-rock interaction) of the aquifer matrix. The L. Cretaceous and Precambrian aquifers are moderately influenced by evaporation processes and the source of salinity of these aquifers is due to the evaporation, and dissolution processes inside the aquifer. The salinity source of the Quaternary aquifers in Watair basin (Group D) is due to the effect of strong evaporation and mixing with seawater. The Miocene aquifer in Nabq area are mostly influenced by the aquifer salinization due to the disposal reject of the desalination plants (Group D).

**Fig. 13:** $\delta^{18}$O vsCl (mg/L) the groundwater tapping the principle aquifers in Southern Sinai, Egypt.

**Conclusions**

The South Sinai's watersheds drain East toward the Gulf of Aqaba and West toward the Gulf of Suez. The area comprises five main aquifers which considered the main source for potable water. The groundwater geochemistry and isotopic data showed different salinization and recharge sources for different aquifers. The isotopic signature of different groundwater aquifers in the South Sinai's were utilized to divide the groundwater into four distinct groups. The group A represent the lower Cretaceous groundwater which has been replenished during paleo-geological Pleistocene time under cooler climatic condition and still receiving considerable amount of the recent recharge. The lower Cretaceous aquifer at the upstream portion of Watair basin is considered good aquifer for sustainable management and the aquifer could be managed sustainably. Group B and Group C are characterizing the Quaternary and the Basement aquifers, where the isotopic signature of $\delta^2$H and $\delta^{18}$O in most areas are close to the recent precipitation and have brackish water type indicating considerable mixing with seawater at the downstream portions of different basins and slight evaporation processes. These two groups comprises Saint Catherine, Upstream of Dahab Basin and El Tour areas, where the groundwater aquifers receives considerable amount of the annual precipitation and runoff water comes
from the upstream watershed of El Egma and El Tih Plateau. On the other hand, the coastal groundwater aquifers located in the downstream are characterized by high groundwater salinization due to mixing with the seawater. The groundwater geochemistry and isotopes indicated the impact of anthropological impact of injecting brine waste comes from the desalination plants at Sharm El Shikh and Nabaq area deteriorates the groundwater quality in the Miocene coastal aquifer. The regional studies in south Sinai's groundwater aquifers delineates the promising areas receives considerable and sustainable groundwater recharge and sound an alarm for aquifer deterioration due to over pumping and anthropological activities.

References


Alcalá, J and E. Custodio, 2004. Use of the Cl/Br ratio as a tracer to identify the origin of salinityin some coastal aquifers of Spain. 18 SWIM. Cartagena, Spain, 481 – 497.


