

Geochemical and Isotopic Evidence of Seawater Intrusion into the Shallow Pleistocene Coastal Aquifer, West Alexandria, Egypt

Magdy M.S. El Maghraby

Geology Department, Faculty of Science, Alexandria University, Egypt
magdy_elmaghraby@yahoo.com

Abstract: The geochemical and isotopic data were examined to identify the origins of salinization in the shallow groundwater aquifer of El Agamy area, northwestern coast of Egypt. The groundwater of the coastal aquifer showed paths of hydrochemical evolution, from Ca-HCO₃ type to Na-Cl or Mg-Cl type. Different molar ratios revealed the impact of seawater on the hydrochemical composition of groundwater of the aquifer to a great extent. The sewage effluents are expected to be important sources for increasing fluoride contents in the study area. Piper diagram showed that Cl and SO₄ are the dominant anions, whereas Na and Mg is the most dominant cation. Durov diagram plot revealed that the groundwater type has been evolved from Ca-HCO₃ type recharge water through to give mixed water of Mg-Cl and Na-Cl water type. The isotopic signature of groundwater shows that the stable isotope values of groundwater fall near the local meteoric water line and lie in the midway between the Mahmoudia Canal and the Mediterranean Sea indicating that major part of this groundwater originated from leakage of municipal wastewater, from improperly constructed septic tanks and directly from injection wells, and mixing with seawater underneath.

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1. Introduction

Groundwater quality is governed by lithology, groundwater flow, nature of geochemical reaction, dissolution/precipitation reactions, ion exchange, residence time, solubility of salts, sea-salt spray, seawater intrusion, sewage water from septic tanks or leakage of wastewater from uncontrolled industrial activities, anthropogenic activities and agriculture (Cruz and Silva 2000; Jordan and Smith 2005).

The salinization of groundwater is one of the most important causes of water-quality deterioration. The phenomenon is well identified in coastal areas, where the deterioration of water is caused by a variety of reasons, among which: subsurface migration of seawater; upward movement of seawater that has entered the aquifer during deposition or during a high stand of the sea in the past geologic times (connate water); seepage of highly saline water concentrated by evaporation in tidal lagoons, playas, or other enclosed areas; aquifer over-exploitation; and recycling of wastewater and agricultural activities and return flow from irrigated lands (Ahmed et al. 2013).

Salinization processes can be diagnosed using many techniques such as geochemical analysis of major and minor elements and isotopic tracers in the groundwater. Usually, major and minor elements concentration such as Br analysis coupled with $\delta^2\text{H}$ and $\delta^{18}\text{O}$ can be used to identify origins of aquifer water salinization due to single process (Martos et

al. 2002; Boluda-Botella et al. 2008). Chloride concentration and molar ratio such as Na/Cl and Br/Cl are used to determine the sources of salinity, especially when seawater intrusion occurs (Sanchez et al. 2002; Rao et al. 2005; El Moujabber et al. 2006; Park et al. 2012). When groundwater salinization results from a combination of several processes, isotopic analysis of $\delta^2\text{H}$ and $\delta^{18}\text{O}$ is more efficient to identify origins of saline groundwater (Jorgensen et al. 2008). Environmental isotopes have become important tools in studying the sources of deterioration of groundwater. Seawater intrusion phenomenon seems to be a multivariate problem. It can be governed by several variables such as hydrochemical properties, cationic exchanges and transport parameters (Boluda-Botella et al. 2008).

Due to rapidly increased urbanization, groundwater development for drinking, domestic and agricultural uses in the northwestern coastal zone of Egypt has drastically increased since 1970. Increased groundwater development has furthered depletion, deterioration of water quality, land subsidence and seawater intrusion throughout the area of study.

In the area of study, seawater intrusion and sewage disposal are the main deterioration sources of groundwater. Domestic and irrigation uses are largely dependent on groundwater in these areas and wells are located near the coast. The overpumping is the main cause of seawater intrusion in the study area.

The purpose of this study is to identify the origins of salinization in the shallow groundwater

aquifer of the El Agamy area, northwestern coast of Egypt, using geochemical and isotopic approaches.

2. Study area

The study area is suffering from lack of wastewater collecting network where sewage is disposed in septic tanks. This has led to leakage from improperly designed and unlined septic tanks to the groundwater. Consequently, groundwater level in El Agamy district has been increased dramatically in the last few years.

El Agamy district is currently experiencing rapid population increase, especially, in summer, since it is considered as one of the main summer resorts in Alexandria. The shallow groundwater aquifer in general and the drilled water wells in particular become susceptible to contamination by wastewater from septic tanks and urban runoff. Recently, those wells had to be abandoned by their owners and many are used for wastewater injection.

The study area lies within the semi-arid belt south of the Mediterranean Sea; its climate is

characterized by rainy unstable winter and a stable hot summer. The temperature reaches its minimum in January (9.1 °C), while its maximum value is recorded in August (30.1 °C). Generally, the Mediterranean coastal area of Egypt receives noticeable amounts of rainfall especially in the winter season. The rainy months are October, November, December and January; while May, June, July and August are dry ones. The rainy season begins in October with 7.76 mm/month, and reached 44.65 mm/month in December. The area extends from the shoreline southward with a maximum distance of about 5 km with latitudes ranging between 31° 03' 57" and 31° 07' 39" N and longitudes ranging between 29° 42' 09" and 29° 47' 33" E (Fig.1). The stable isotope ratios of H and O, and concentrations of major and minor ions, including bromide (Br) and fluoride (F) of groundwater are essential in understanding geochemical processes affecting water quality. The results of this study may contribute to the optimization of water resources management.

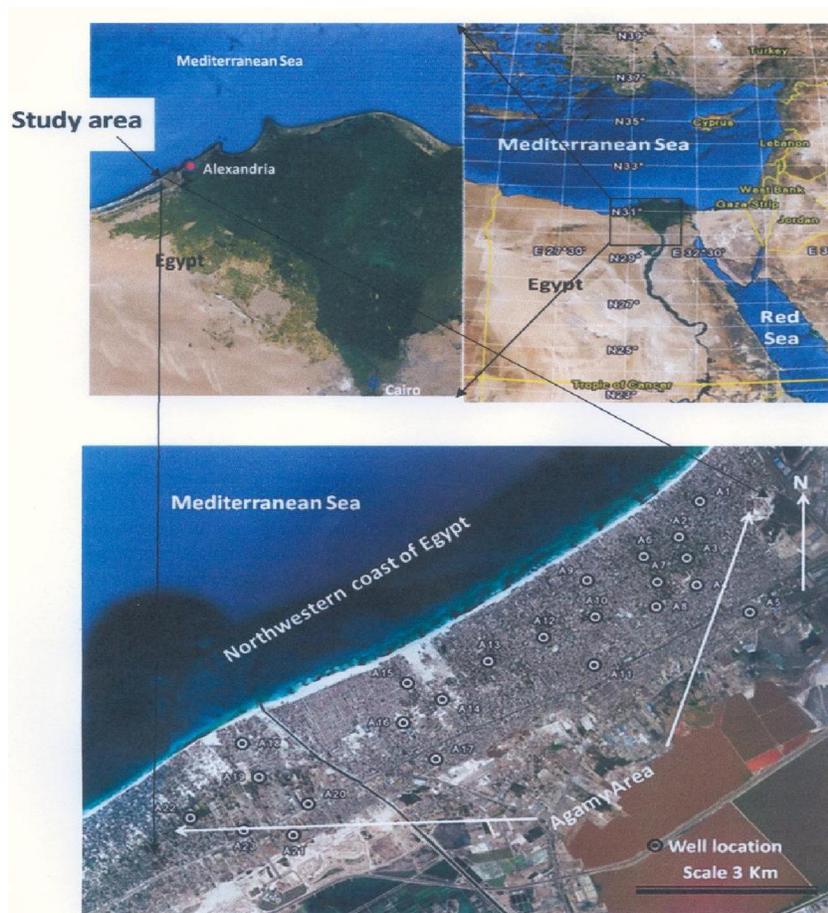


Fig.1 Location map of different wells the study area

3. Hydrogeological setting

The northwestern coastal zone of Egypt occupies a narrow strip lying between the Mediterranean Sea coast to the north and the tableland to the south. It extends westward from the Nile Delta to the Egyptian-Libyan frontiers. This area is divided into two geomorphologic units, namely, a low altitude northern-coastal plain with maximum elevation of about 40 m and characterized by the occurrence of a series of elongated ridges oriented parallel to the present coast. The second unit is alternating with shallow depression areas and the southern higher tableland extending from the northern Mediterranean coastal plain southward to the Qattara Depression with a maximum elevation of about 120 m. These two geomorphologic units are separated from each other by a narrow piedmont plain.

Many geomorphological, geological and hydrogeological studies have been carried out on the northwestern Mediterranean coastal zone of Egypt (e.g. **El Shamy et al. 1969; Rizzini et al. 1978; Hassouba 1995; El Asmar and Wood 2000; Yousif and Bubbenzer 2012; Ahmed et al. 2013**).

The northwestern area of Egypt is entirely composed of sedimentary rocks belonging to Quaternary and Late Tertiary (Fig.2). The Quaternary rocks are represented by both Holocene (beach deposits, dune sand accumulations, and alluvial deposits) and Pleistocene (oolitic limestone). The Tertiary rocks belong mainly to Middle Miocene (Vindobonian). These rocks are developed essentially into shallow marine limestone, which is rich in its faunal content. Pliocene rocks are not exposed, but are expected in the subsurface of the foreshore part of the area (**El Senussi and Shata 1969**).

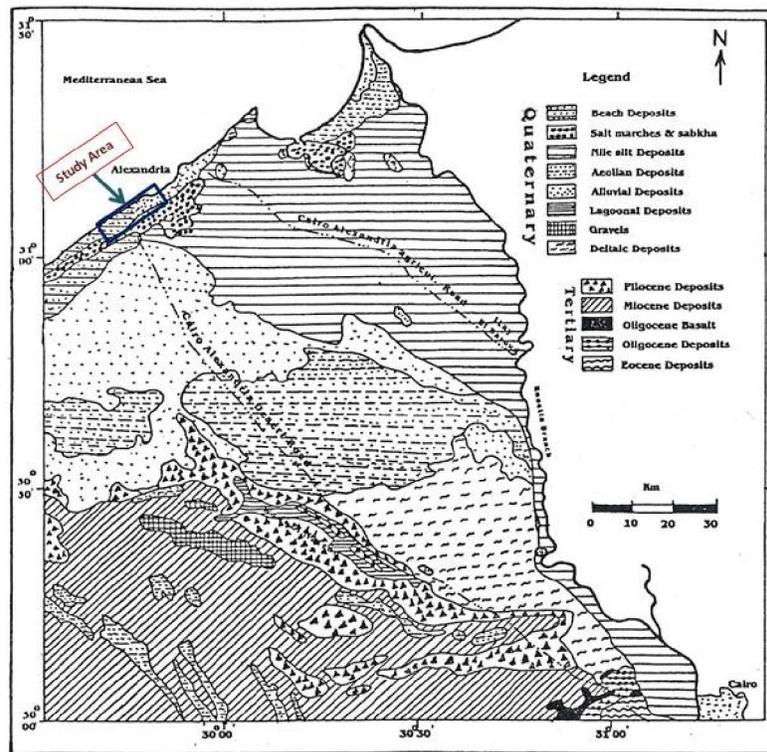


Fig.2 Geological map of northwestern coast of Egypt including the study area (CONOCO 1987)

The aquifers of northwestern coastal zone of Egypt are classified into three types (**Hilmy et al. 1977; El Maghraby and El Fiky 2002**): (i) The Holocene dune sand accumulations consist of unconsolidated calcareous sand of high porosity. Such accumulations act as good local reservoirs for rainfall. They are tapped by a number of wells to yield water of low salinity. (ii) The Pleistocene oolitic limestone; forms the most important aquifer in the region. It covers the whole coastal plain forming

elongate ridges. The oolitic limestone extends southward from the coastline to about 10 km. The foreshore oolitic limestone ridge is characterized by less cementing materials compared with the inland ridges and hence it has more porosity. The flanks of the foreshore ridge are covered by loose foreshore sand accumulations, which permit direct infiltration and percolation of rainwater. The groundwater found in the oolitic limestone ridges originates either from direct infiltration and percolation of annual rainfall

on the ridges or from the rainwater falling on the tableland located to the south; and (iii) The Middle Miocene fissured limestone form the underlying rocks of the whole area, including few clay intercalations.

4. Materials and methods

The present study is based on both field observations and laboratory investigations. A reconnaissance survey was carried out throughout the investigated area in September 2012.

Twenty three wells, representing different parts of the study area with an intensity is about 11 km and of shallow depths ranging between 3 and 22 m, were sampled from the El Agamy area, northwestern coast of Egypt (Fig.1). Hydrogen ion concentration (pH) was measured in the field using ORION Expandable Ion-Analyzer EA920, Electrical conductivity (EC) and Total Dissolved Solids (TDS) were measured at the wellhead using portable HI 991300 Hanna Instruments. Duplicate water samples were taken at each well. One duplicate sample was collected at its natural pH, in polyethylene bottles after rinsing several times with water from the point of collection and preserved (approximately at 4°C) for anion analysis. The second sample was also collected in polyethylene bottles and acidified using HNO₃ to pH < 2 for cation analysis. All the samples for laboratory analyses were filtered through 0.45 µm membrane filters. Chemical analyses of major ions were carried out at the laboratory of Chemistry Department, Faculty of Science, Alexandria University, Egypt, using standard analytical procedures (American Public Health Association APHA 1998). Calcium, Mg, Cl and Br were determined using Digital Titrator HACH. Bicarbonate concentration was determined using titration against HCl. PFP7 Flame photometry was used to measure the concentrations of Na and K. Sulfate, NO₃, NH₄, PO₄ and F concentrations were determined using DR2010 HACH Spectrophotometer. The analytical precision for measurement of ions was determined by calculating the ionic balance error, which falls within the acceptable limits of +5%. The total period of all analyses was taken about two weeks from the time of sampling.

The samples for isotope analysis were separately collected and sealed in 20 ml HDPE bottles. The bottles were completely filled and sealed with a cap fitted with an integral rubber seal to prevent liquid-vapour fractionation arising from evaporation into headspace. Once collected, the samples were kept cool until analysis.

Stable isotopes of oxygen and deuterium (¹⁸O, ²H or D) were performed at the laboratory of Middle Eastern Regional Radioisotope Center for the Arab

Countries (MERRCAC) in Cairo, Egypt, using Mass-Spectrometer Delta-S. Deuterium isotope ratio analyses have been performed using a hydrogen equilibration technique similar to **Coplen et al. (1991)**. For oxygen isotope analysis, about 2 ml of each water sample was equilibrated with CO₂ by shaking for 6 hrs at 25°C (**Epstein and Mayeda 1953**). Analytical precision was ±0.1‰ for ¹⁸O and ±1‰ for ²H. Isotopic compositions were reported in ‰ deviation from isotopic standard reference material, VSMOW (Vienna Standard Mean Ocean Water) using the conventional δ notation where:

$$\delta(\text{‰}) = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \times 10^3$$

where R is the ratio of the heavy to light isotope abundances (**Coplen 1996**).

Saturation indices of some common minerals were calculated using the program PHREEQC (**Parkhurst and Appelo 1999**) interfaced with AquaChem 2010.1 software.

5. Results and discussion

Groundwater in the northwestern coastal zone occurs mainly under main water-table conditions. The source of groundwater that recharge the main water-table in the northwestern coastal zone is through precipitation on the coastal plain and the southern tableland (**Yousif et al. 2013**).

The free surface of the main water-table has a level at or about the mean sea level up to about 20 km inland. The main fresh water-table forms a thin freshwater layer floating on the saline water. The water table is controlled by the bouncy effects and the saltwater intrusion into coastal aquifers. **Shata (1970)** pointed out that near the sea, the inflow of seawater maintains a dynamic equilibrium with a comparatively thin layer of fresh water existing on the upper surface of the salt water. Most of the wells and open trenches along the coastal zone depend on their supply from the main water table. A balance or equilibrium tends to be established between the fresh groundwater and the salt water pressing in from the sea. Where coastal aquifers are over pumped, lowered by natural drainage, or natural recharge is impeded by construction or other activities, the groundwater level is lowered, thereby reducing the fresh water flow to the sea. A reversal or reduction of freshwater flow allows the heavier salt water to advance inland, which can seriously threaten the continued use of an aquifer as a water supply source. Fresh water and seawater, being of different densities, when brought together under steady-state balance tend not to coalesce but to form a distinct interface (**Ghyben 1889; Herzberg 1901**). Because seawater intrusion, however, represents a

displacement of the miscible liquids in the porous media, the diffusion and hydrodynamic dispersion mix the two liquids and the interfacial surface becomes a transition zone of variable thickness (US EPA 1973). The position of the interface between freshwater and salt water depends on a number of factors; flow rates, nature of the flow paths, high tide and change in recharge. A change in any of these factors will cause the salt-water/fresh-water interface to migrate. Reduction of stream flow, over pumping or deepening of the open channels results in the landward migration of the seawater wedge (Cooper et al. 1964) whereas the increased stream flow result in a seaward migration.

5.1. General groundwater chemistry

The results of analyzed chemical and isotopic constituents of the El Agamy area were

presented in Table 1. Seawater data were also added as a possible source of salinization of the Pleistocene aquifer.

The groundwaters of the study area have pH values ranged from 7.25 to 7.89 indicating slightly alkaline water. The total dissolved solids (TDS) values ranged from 567.4 mg/l at well A14 to 1221 mg/l at well A13 indicating fresh to brackish water. Three water points (A6, A13, A20) showed TDS values more than 1000 mg/l, indicating mixing with the underlying dense salt water during upcoming resulted from high rate of discharge in these water points. Groundwater with high TDS is not suitable for both irrigation and drinking purposes (Fetter 1994). The lowest value of TDS concentration at well A14 indicates no impact of the seawater and sewage water on its salinity.

Table 1. Chemical compositions of the groundwaters sampled from the El Agamy area (concentrations are in mg/l).

No.	pH	EC	TDS	Ca	Mg	Na	K	HCO ₃	SO ₄	Cl	Br	F	NO ₃	NH ₄	PO ₄
A1	7.39	1168.73	747.9	70.14	47.40	80.00	10.00	284.34	72.00	184.10	ND	0.79	4.30	ND	0.50
A2	7.39	1162.66	744.1	86.17	49.23	71.00	10.00	275.34	68.00	184.36	ND	1.39	5.80	ND	1.20
A3	7.62	936.8	599.6	73.67	34.33	60.00	10.00	194.23	70.00	157.33	ND	0.84	6.60	ND	1.00
A4	7.62	1050.53	672.3	58.60	45.25	64.00	10.00	277.44	32.00	185.05	ND	1.44	5.30	ND	1.30
A5	7.32	1153.34	738.1	16.01	71.58	123.00	10.00	157.19	136.00	224.36	ND	4.80	11.30	4.20	0.50
A6	7.46	1801.18	1152.76	53.11	66.71	210.00	20.00	391.40	110.00	301.54	45.30	1.39	8.30	0.01	1.90
A7	7.89	1169.79	748.7	29.06	62.60	100.00	20.00	281.67	74.00	181.34	ND	0.51	6.20	ND	1.90
A8	7.51	1118.50	715.8	34.07	58.17	80.00	10.00	279.26	62.00	192.34	ND	1.31	5.00	ND	1.60
A9	7.64	1248.11	798.8	64.31	23.15	131.00	10.00	203.36	201.33	165.64	ND	5.60	13.20	5.60	1.04
A10	7.45	981.9	628.4	22.55	61.64	110.00	10.00	64.21	96.00	264.00	ND	6.60	12.30	7.80	1.01
A11	7.25	1139.95	729.6	61.88	42.17	100.00	10.00	220.87	102.65	192.00	ND	6.70	13.50	8.80	1.20
A12	7.34	886.6	567.4	76.12	30.49	51.00	3.5	159.45	112.54	134.32	ND	5.60	11.30	5.40	0.90
A13	7.75	1908.45	1221.41	51.48	77.14	210.00	10.00	387.42	124.23	361.14	47.40	1.46	4.20	ND	0.90
A14	7.75	897.2	574.2	64.12	31.14	65.00	3.5	249.45	47.00	114.02	ND	1.22	4.90	ND	0.50
A15	7.46	1189.03	760.9	14.03	44.09	162.00	10.00	108.66	124.87	297.33	ND	2.40	9.70	5.10	0.90
A16	7.42	1102.89	705.9	49.19	64.44	85.00	3.5	197.54	148.22	157.96	ND	4.90	12.30	5.40	0.90
A17	7.51	1273.39	814.9	15.12	36.31	180.00	10.00	239.77	152.41	181.36	ND	3.70	7.10	2.80	0.70
A18	7.32	975.9	624.6	29.34	61.88	65.00	3.5	190.16	140.14	134.54	ND	1.80	8.20	1.80	0.50
A19	7.41	958.5	613.4	49.71	62.14	55.00	10.00	143.00	69.23	224.33	ND	1.40	7.40	0.80	0.50
A20	7.42	1601.09	1024.70	55.27	18.65	232.00	20.00	284.22	134.25	280.31	ND	1.90	6.20	0.02	2.00
A21	7.53	1004.91	643.1	29.46	59.67	100.00	3.5	55.42	148.34	246.75	ND	1.60	4.50	1.60	0.10
A22	7.58	1023.31	654.9	39.88	22.48	125.00	3.5	186.67	147.92	129.47	ND	0.90	7.60	2.24	0.10
A23	7.36	1187.16	759.8	18.53	36.41	178.00	10.00	131.05	188.55	197.24	ND	3.10	7.60	3.70	0.50
SW ^a			35169.00	412.0	1284.00	10781.00	399.00	126.00	2712.00	19353.00	67	1			
			^b WHO (2011)	^c NGL		1.50	50	^c NGL	0.4-0.5 ^d						

ND: Not Determined

^a: Mediterranean Sea Water, Pilson 1998.

^bWHO (2011). World Health Organization (maximum limit for drinking)

^cNGL. No guideline (Not of health concern at levels found in drinking water)

^d(Carney 1991)

The brackish nature of groundwater may be attributed to mixing with the underlying dense saltwater and with wastewater. In addition the nature of the alluvial deposits rich in soluble minerals is another possible source of salinization of groundwaters from the coastal shallow aquifer.

Chloride is the major solute in the composition of seawater and its predominance in the coastal aquifer can normally be related to the presence of salt-water intrusion. Chloride can be used to distinguish between several mixing mechanisms of freshwater and saline water (Kim et al. 2003).

The chemical constituents of the groundwater of the study area showed the following ranges (Table 1): 14.03-86.17 mg/l for Ca, 18.65-77.14 mg/l for Mg, 51.00-232.00 mg/l for Na, 3.50-20.00 mg/l for K, 55.42-391.4 mg/l for HCO₃, 32.00-201.3 mg/l for SO₄ and 114-316.4 mg/l for Cl.

Dissolution of carbonate minerals is the main source of Ca, CO₃ and HCO₃ in groundwater. Ca and Mg from carbonate minerals are removed from solution by cation-exchange and replaced by Na. The sodium that occurs on cation-exchange minerals (primarily clay minerals) probably

originates from salt water or from weathered sodium-rich clay (**Petalas and Diamantis 1999**).

On the other hand, dominance of Na and Mg in a carbonate coastal aquifer is mainly caused by mixing with seawater, where these cations are the most dominant in seawater. High salinity values and the appearance of Na-Cl and Mg-Cl waters, which represents an advanced stage of geochemical evolution, strongly suggest mixing with seawater at the coastal plain of the study area.

The groundwater of the El Agamy shallow coastal aquifer (Table 1) showed paths of hydrochemical evolution, from Ca-HCO₃ type to Na-Cl or Mg-Cl type. The ionic dominance of the groundwater shows the predominance of chloride over bicarbonate and sulfate; except for sample A14, where HCO₃>Cl; indicating that this site was not affected by seawater intrusion as it is revealed by the TDS value is the lowest one and reaches 567.4 mg/l (Table 1). Whereas, the cations show two types of

predominance: Na>Mg>Ca and Mg>Na>Ca. These patterns indicate that the groundwater chemistries were changed by cation exchange reaction, as well as simple mixing (**Appelo and Postma 2005**). The prevalence of Na, Mg and Cl strongly suggests the effect of seawater intrusion on groundwater of the study area (**Pulido-Leboeuf 2004**).

Chemical analyses of the groundwater of the study area were plotted on Piper trilinear diagram (Fig. 3). The chemical evolution represents in general water quality changes due to the water-rock interaction in the regional aquifer system from recharge zone with different groundwater residence time. In this study, the water chemistry changed from Ca-HCO₃ type to Na-Cl or Mg-Cl type is mainly caused by groundwater contamination such as seawater intrusion, and thus it should not be dealt with in this study. In addition, the Piper (Fig.3) and Durov's (Fig.4) diagrams do not show the hydrochemical evolution path clearly.

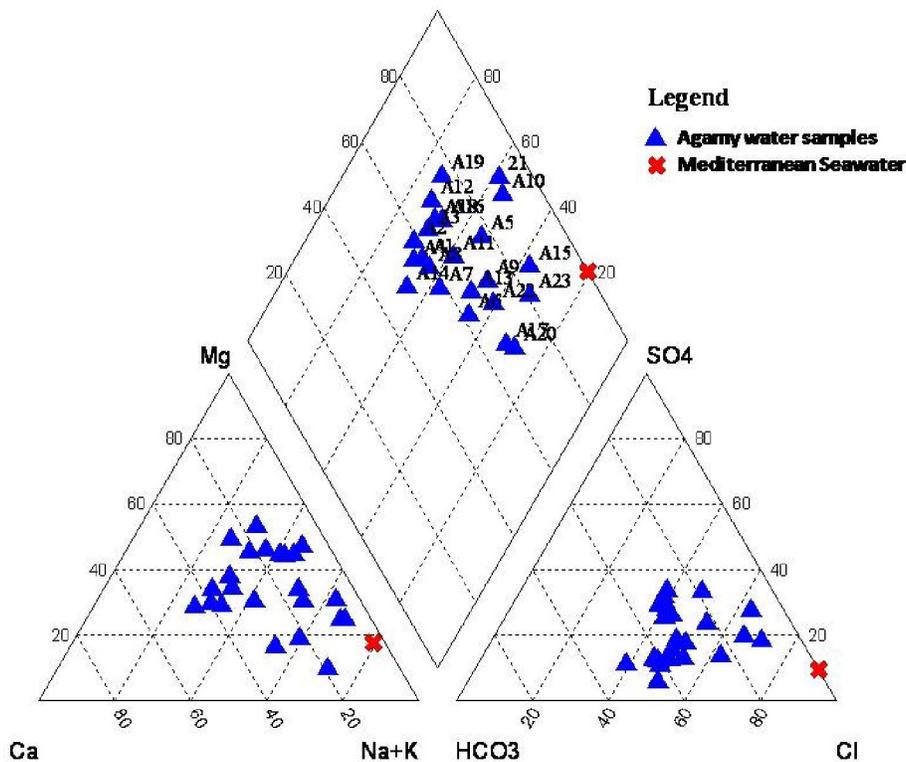


Fig.3 Piper diagram plot of groundwater samples of the study area

Durov diagram plot revealed that the groundwater type has been evolved from Ca-HCO₃

type recharge water through to give mixed water of Mg-Cl and Na-Cl water type.

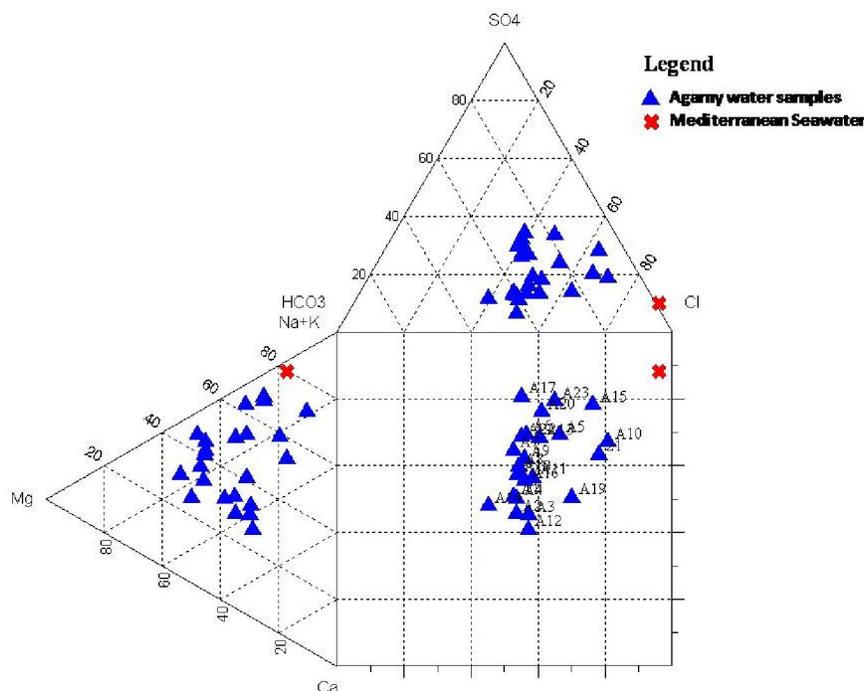


Fig.4 Durov's diagram plot of the groundwater samples of the study area.

5.2. Bromide

The concentration of some solutes such as Br and Cl species have often been used as a reliable indicator of the origin of salinity due to its specific composition in various saline sources (Ahmed et al. 2013). Bromine is one of the halogen elements found in trace amounts in all ground water (Davis et al. 2004). In nature, bromine occurs mainly in the inorganic form of bromide. Bromine, because of its high solubility, is found in seawater (65 mg/l) and in brines (Neal et al. 2007). Bromide can be used to separate salinity of marine and non-marine origins (Kharroubi et al. 2012). The level of bromide in groundwaters varies considerably and is usually linked with chloride concentration (Edmunds 1996).

Generally, the bromide content in groundwater is low (Hitchon et al. 1999). Although, high concentrations of this element sometimes occur in groundwater by another sources such as interaction with seawater, agriculture return flows, reaction with evaporite deposits or anthropogenic origin. The ratio of Br/Cl must be unchanged when salinization of freshwater is caused by mixing between seawater and groundwater.

Seawater has a Br/Cl molar ratio around 0.0015 in Mediterranean countries (Vengosh et al. 1999). A contribution of anthropogenic sources give a ratio around 0.0005 for wastewater seepages or around 0.02 for agriculture return flows (Vengosh et al. 1999; Ghabayen et al. 2006). For potable groundwaters of low salinity, bromide are typically

less than 0.4 mg/l (Edmunds 1996; Davis et al. 2004).

In the area of study, measurements of Br content were performed at two groundwater samples (A6 and A13) that have the highest TDS values (Table 1), and the Br content at these two sites was high (45.30, 47.40 mg/l, respectively). The calculated Br/Cl molar ratios for the analyzed two samples were 0.0661 and 0.0696 indicating seawater intrusion (Table 2).

5.3. Fluoride

Most of the fluoride found in groundwater is naturally occurring from the breakdown of crystalline rocks containing fluorine-rich minerals, especially granites and volcanic rocks and shallow aquifers in arid areas experiencing strong evaporation. Fluoride can also come from runoff and infiltration of chemical fertilizers in agricultural areas; septic and sewage treatment system discharges and from waste from industrial sources (Smedley et al. 2002; Edmunds and Smedley 2012).

Occurrence of fluorine in groundwater has drawn worldwide attention due to its considerable impact on human physiology. Fluorine occurs mainly as free fluoride ions in natural waters. The assimilation of fluorine by the human body from potable water at the level of 1 mg/l (WHO 2011) enhances bone development and prevents dental caries (Kundu et al. 2001). However, continuing consumption of higher concentrations can cause dental fluorosis and in extreme cases even skeletal

fluorosis. The guideline value for fluoride in drinking water is 1.50 mg/l (**WHO 2011**).

The effects on health depend on a number of factors including dose from other sources of fluoride (toothpastes, food), age, gender and general health status (**Feenstra et al. 2007**).

The fluoride concentration of 52.17% water samples was more than 1.5 mg/l (Table 1). The sewage effluents including septic tanks discharged to the unsaturated aquifers of the study area, fertilizers and rainfall are expected to be important sources for increasing fluoride contents in the study area.

5.4. Nutrients

The presence of nutrients in groundwater, such as nitrate, ammonia and phosphate can be attributed to anthropogenic sources such as sewage effluent through seepage from septic tanks and the few agricultural activities in the area (**Zanini et al. 1998; Rouabhia et al. 2009**).

The concentration of nitrate in the groundwater samples of the study area were below the maximum permissible limit for drinking (**WHO 2011**) (Table 1).

The decrease in ammonia concentrations may be due to adsorption of soil clay particles and rapid ammonia oxidation in water to nitrite and nitrate by bacterial action (**APHA 1998**). Ammonia in drinking-water is not of immediate health relevance, and therefore no health-based guideline value is proposed. The values of ammonia in the groundwater samples of the El Agamy area indicate low concentrations (Table 1).

The maximum permissible limit of phosphate in water for drinking purposes ranges between 0.4–0.5 mg/l (**Carney 1991**). A percent of 65.22% of the total samples have a phosphate concentration higher than 0.5 mg/l; a percent of 26.09% of the samples have a phosphate concentration equal to 0.5 mg/l; and the rest of samples 8.70% have a concentration below the maximum permissible limit for drinking purposes.

5.5. Molar ratios

Encroachment of seawater can be evaluated by studying a series of molar ratios (**Sanchez et al. 1999 and 2002; Vengosh et al. 2002; El Moujabber et al. 2006**). Studying the molar ratios of groundwater have been used to evaluate the influence of seawater intrusion on coastal groundwater (**Vengosh et al. 1999; Park et al. 2012**).

Most of the molar ratios of the groundwater samples of the study area, compared to the values of seawater molar ratios, show an effect of seawater intrusion to a great extent (Table 2).

Na/Cl values of groundwater less than the seawater one (0.86) (**Vengosh and Rosenthal 1994**)

indicate that fresh the groundwaters were contaminated with the saline waters. The values of Na/Cl of the groundwater of the El Agamy area were ranged from 0.38 to 1.53. For Na/Cl, 65.22% of the water samples were less than the seawater ratio. This indicates that the groundwaters were affected by seawater intrusion to some degree. Values close to the seawater ratio indicate recent simple mixing of groundwaters with seawater (**Mercado 1985**). Whereas the Na/Cl ratio is slightly high in 34.78% of the samples, this may be attributed to the cation-exchange process that releases Na to the aqueous phase leading to increase the value of Na/Cl ratio. High Na/Cl ratios may be indicative of anthropogenic contamination.

Groundwaters resulting from mixing between seawater and fresh groundwater are characterized by the decrease of K/Cl, Ca/Cl, Mg/Cl, HCO₃/Cl, and SO₄/Cl ratios. From Table 2, it is noticed that the values of these ratios are low indicating trend towards increasing of mixing with seawater to some extent. For example; it is noticed that the values of HCO₃/Cl ratio (95.65% of samples) are low.

Seawater is characterized by high Mg/Ca ratio (5.14) due to dominance of Mg in seawater and also due to the elimination of Ca ions through the precipitation of gypsum. In fresh water Mg/Ca ratio values are less than 1. From Table 2, it is noticed that 69.57% of the water samples of the study area have high values of Mg/Ca ratio more than 1 indicating the effect of mixing with seawater.

Furthermore, the Ca/SO₄ ratio of the groundwater samples are usually distinct from those of seawater (0.36) showing a higher calcium concentration with respect to sulfate (Table 2). Higher calcium content was found in 43.48% of the groundwater samples. Such a high Ca/SO₄ ratio suggests reverse cation-exchange processes as well as gypsum dissolution.

5.6. Geochemical modeling

Interactions between groundwater and surrounding host rocks are believed to be the main processes responsible for the observed chemical characteristics of the groundwater. The geochemical interactions lead to changes in water chemistry. The potential for a chemical reaction can be determined by calculating the chemical equilibrium of the water with the minerals phases. The equilibrium state of the water with respect to a mineral phase can be determined by calculating a saturation index (SI) using analytical data. Where, the reactive mineralogy of the subsurface can be predicted by using the saturation index approach from groundwater data without collecting the samples of the solid phase and

analyzing the mineralogy (Deutsch 1997). The potential for mineral precipitation or dissolution is assessed using the saturation index (SI), which is based on the relation between analytic activities (the ion activity product, IAP) and the thermodynamic calculation of the solubility product (K_{sp}). The SI of a

mineral is calculated based on the following equation (Parkhurst 1995):

$$SI = \log_{10} \frac{IAP}{K_{sp}}$$

Table 2. Results of molar ratios in the El Agamy area (meq/L).

No.	Na/Cl	K/Cl	Ca/Cl	Mg/Cl	HCO ₃ /Cl	SO ₄ /Cl	Mg/Ca	Ca/SO ₄	Br/Cl
A1	0.67	0.05	0.67	0.75	0.90	0.29	1.11	2.34	
A2	0.64	0.03	0.15	0.68	0.14	0.27	4.51	0.56	
A3	0.80	0.05	0.57	0.64	0.67	0.39	1.12	1.44	
A4	0.59	0.02	1.00	0.66	0.69	0.62	0.66	1.62	
A5	0.90	0.03	0.25	0.62	0.62	0.25	2.47	0.99	
A6	0.88	0.03	0.99	0.80	1.27	0.30	0.80	3.27	0.06956
A7	0.84	0.03	0.08	0.43	0.21	0.31	5.18	0.27	
A8	0.83	0.02	0.55	1.19	0.73	0.69	2.16	0.80	
A9	1.53	0.05	0.15	0.58	0.77	0.62	3.96	0.24	
A10	0.74	0.02	0.39	1.34	0.82	0.77	3.48	0.50	
A11	0.38	0.04	0.39	0.81	0.37	0.23	2.06	1.72	
A12	0.59	0.05	0.83	0.78	0.87	0.27	0.94	3.04	
A13	1.28	0.06	0.35	0.19	0.59	0.35	0.56	0.99	0.0661
A14	0.62	0.01	0.21	0.71	0.13	0.44	3.34	0.48	
A15	1.49	0.02	0.54	0.51	0.84	0.84	0.93	0.65	
A16	1.39	0.05	0.17	0.54	0.39	0.71	3.24	0.24	
A17	0.59	0.06	0.83	0.64	0.72	0.33	0.77	2.52	
A18	0.53	0.05	0.56	0.71	0.87	0.13	1.27	4.39	
A19	0.85	0.04	0.13	0.93	0.41	0.45	7.37	0.28	
A20	1.07	0.06	0.31	0.65	0.75	0.27	2.07	1.16	
A21	0.85	0.10	0.28	1.01	0.90	0.30	3.55	0.94	
A22	0.64	0.05	0.31	0.88	0.84	0.24	2.82	1.32	
A23	1.22	0.05	0.69	0.41	0.71	0.90	0.59	0.77	
SW ^a	0.86	0.02	0.04	0.19	0.004	0.10	5.14	0.36	0.00154

SW^a: Mediterranean Sea Water, Pilson 1998.

If $SI > 0$ the solution is theoretically oversaturated with respect to the mineral and precipitation may be expected. For $SI = 0$, the mineral and solution are in equilibrium and neither dissolution nor precipitation is predicted to occur. If $SI < 0$, the solution is theoretically undersaturated with respect to the mineral, and if present in the system, dissolution might be possible.

The geochemical modeling program PHREEQC (Parkhurst and Appelo 1999), interfaced with Aquachem 2010.1, was used to calculate saturation indices in the groundwater of the study area. The calculations of SI were depended on the results of chemical analysis of the analyzed ions. Table 3 shows the results of SI calculations for the groundwater with respect to various specific minerals.

The groundwaters of the study area show slightly to moderately undersaturation with respect to

carbonate minerals, where SI for calcite ranged between -0.12 - -1.44, aragonite -0.27 - -1.59 and dolomite -0.20 - -2.21. Whereas, calculations of SI values in the studied groundwaters indicate that these waters are moderately undersaturated with respect to sulfate minerals (gypsum and anhydrite) and strongly undersaturated with respect to halite, revealing that halite does not play an important role in the groundwater chemistry (-1.44 - -2.25 for gypsum, -1.67 - -2.48 for anhydrite, -5.78 - -7.00 for halite). Dissolution of carbonate is expected due to the aquifer material is mainly composed of carbonate minerals.

Figure 5 shows that the SI of groundwater samples are slightly to moderately undersaturation with respect to carbonate and sulfate minerals. All the groundwater samples are highly undersaturated with respect to halite.

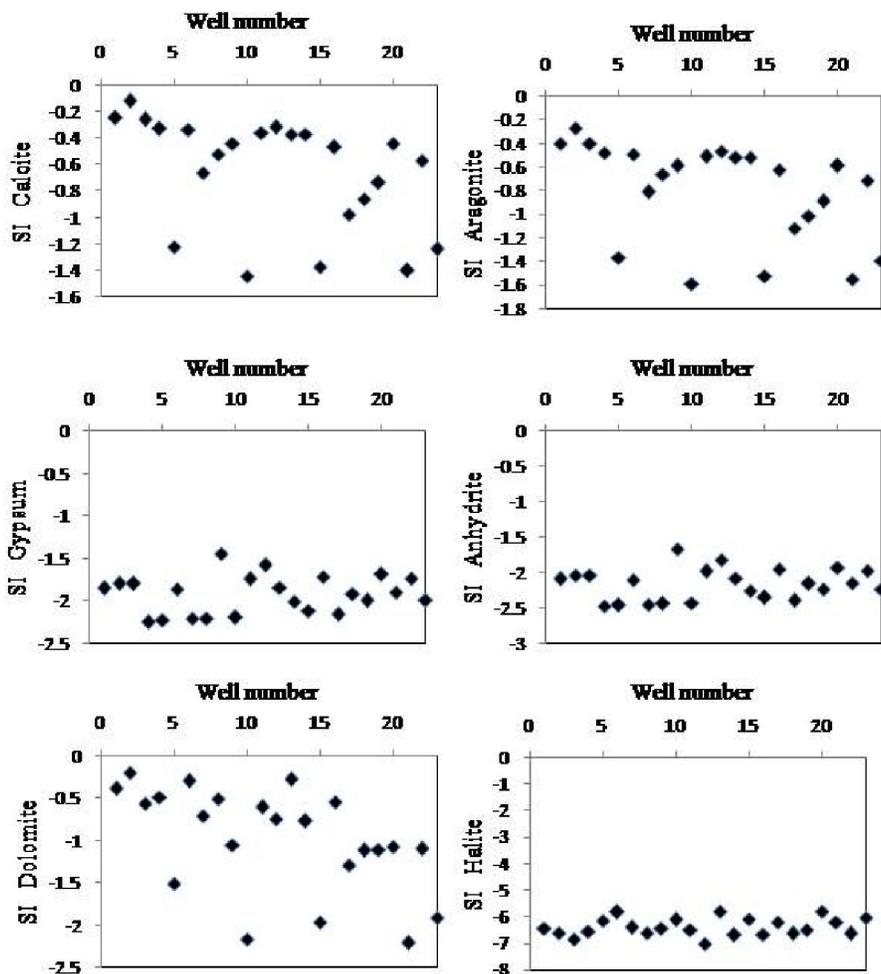


Fig.5 Plots of saturation indices of aragonite, calcite, dolomite, gypsum, anhydrite and halite in the groundwater samples of the study area

Table 3. Values of saturation indices of the common minerals for groundwater samples of the study area.

No.	Calcite	Aragonite	Dolomite	Gypsum	Anhydrite	Halite
A1	-0.25	-0.40	-0.39	-1.85	-2.09	-6.42
A2	-0.12	-0.27	-0.20	-1.80	-2.04	-6.61
A3	-0.26	-0.40	-0.56	-1.80	-2.04	-6.85
A4	-0.33	-0.48	-0.49	-2.25	-2.48	-6.56
A5	-1.22	-1.37	-1.51	-2.22	-2.46	-6.15
A6	-0.34	-0.49	-0.30	-1.86	-2.10	-5.82
A7	-0.66	-0.81	-0.71	-2.21	-2.45	-6.41
A8	-0.52	-0.66	-0.51	-2.21	-2.44	-6.59
A9	-0.44	-0.59	-1.05	-1.44	-1.67	-6.43
A10	-1.44	-1.59	-2.16	-2.20	-2.43	-6.12
A11	-0.36	-0.51	-0.61	-1.74	-1.98	-6.48
A12	-0.32	-0.46	-0.75	-1.58	-1.82	-7.00
A13	-0.37	-0.52	-0.28	-1.84	-2.08	-5.79
A14	-0.37	-0.52	-0.77	-2.01	-2.25	-6.70
A15	-1.37	-1.52	-1.96	-2.11	-2.35	-6.10
A16	-0.47	-0.62	-0.55	-1.71	-1.95	-6.65
A17	-0.98	-1.12	-1.29	-2.16	-2.40	-6.21
A18	-0.86	-1.01	-1.12	-1.92	-2.16	-6.64
A19	-0.74	-0.89	-1.11	-2.00	-2.24	-6.49
A20	-0.44	-0.59	-1.08	-1.69	-1.93	-5.78
A21	-1.40	-1.55	-2.21	-1.90	-2.14	-6.20
A22	-0.57	-0.71	-1.10	-1.73	-1.97	-6.64
A23	-1.24	-1.39	-1.91	-1.99	-2.23	-6.04

5.7. Isotopic investigation of groundwater

Environmental isotopes have much to offer where the contamination of groundwater is of concern. Their analysis can provide information on the origin of contaminants, geochemical reactions.

Most environmental isotope studies have focused on the light elements and their isotopes: hydrogen (^1H , ^2H , ^3H), carbon (^{12}C , ^{13}C , ^{14}C), nitrogen (^{14}N , ^{15}N), oxygen (^{16}O , ^{18}O) and sulfur (^{32}S , ^{34}S). These are the most important elements in biologic systems and also participate in most chemical reactions. Furthermore, they occur in relative great abundance and their contents in different terrestrial compounds can be determined with analytical uncertainties much smaller than natural variations (**Fritz and Fontes 1980**).

Some general rules can be recognized for the distribution of these isotopes in groundwater: if the isotope content does not change within the aquifer, it will be reflect the origin of the water. If the isotope content changes along groundwater paths, it will reflect the history of the water. Origin deals with location, period and processes of the recharge,

whereas, history deals with mixing, salinization and discharge processes. Stable isotope concentrations in waters are basically controlled by the number of condensation stages resulting in precipitations and by environmental conditions of any subsequent evaporation.

Representative eight water samples were analyzed for their ^{18}O and ^2H contents (Table 4). The results of isotopic contents of the Mediterranean Sea water and Mahmoudia Canal water (main source of drinking water in Alexandria) were also used and correlated with those of groundwater to reveal the extent of possible effects of both seawater intrusion and leakage of wastewater from improperly constructed septic tanks and injection wells.

The observed variation in stable isotope content in groundwater of the study area ranges from 2.56 to 3.12‰ for $\delta^{18}\text{O}$ and from 20.96 to 39.36‰ for $\delta^2\text{H}$ except for sample No. A14 where the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values were -0.55‰ 5.04‰, respectively (Table 4).

Table 4. Environmental isotopic data for the Agamy coastal area.

Water samples No.	$\delta^2\text{H}$ (‰) SMOW	$\delta^{18}\text{O}$ (‰) SMOW
Mediterranean Sea ^a	9.7	1.77
Mahmoudia canal ^b	32.6	4.55
A1	33.14	2.93
A4	38.57	3.01
A6	20.96	2.83
A7	22.82	3.12
A8	39.36	3.06
A13	23.67	2.87
A14	5.04	-0.55
A20	22.92	2.56

^a Shided and El-Sayed, 2003

^b Awad et al., 1994

The $\delta^{18}\text{O}$ - $\delta^2\text{H}$ diagram (Fig.6) shows that the stable isotope values of groundwater fall near the local meteoric water line. Precipitation lies on parallel straight lines with a slope close to 8. The value of the intercept depends on the origin of the condensing vapour (**Fritz and Fontes 1980**).

A typical interception value for oceanic precipitation is +10 and then the equation $\delta^2\text{H} = 8\delta^{18}\text{O} + 10$ (Craig, 1961) is generally used as a reference for precipitations when no local data are available. In the eastern Mediterranean, the value of the intercept is +22 (**Gat and Carmi 1970**).

Figure 6 also shows that the groundwater samples from the El Agamy coastal area lie in the

midway between the Mahmoudia Canal and the Mediterranean Sea plots indicating that major part of this groundwater originated from leakage of municipal wastewater, from improperly constructed septic tanks and directly from injection wells, and mixing with seawater underneath. However, sample no. A14 shows different isotopic signature which is much more depleted in heavy isotopes of oxygen and deuterium. This can be explained by minimum impact of both wastewater leakage and mixing with seawater.

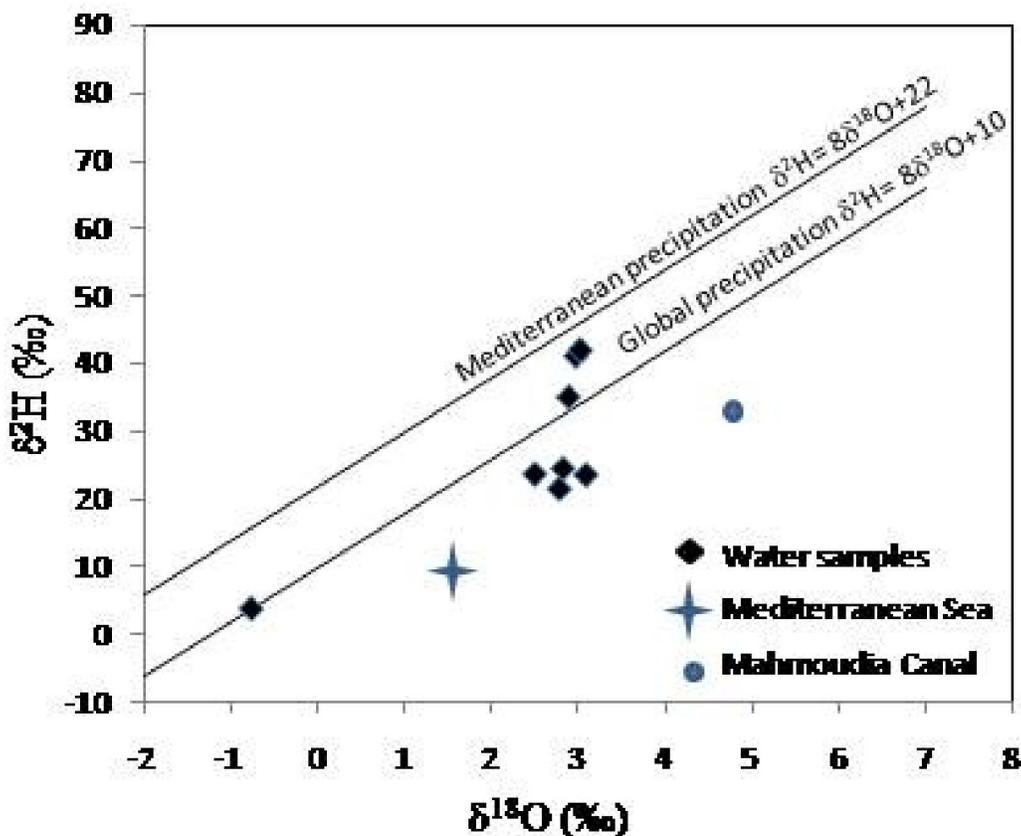


Fig.6 $\delta^2\text{H}$ - $\delta^{18}\text{O}$ relationship for the groundwater samples of the El Agamy area. The Global Meteoric Water Line (GMWL) and Mediterranean Meteoric Water Line (MMWL) are also shown.

6. Conclusions

Geochemical and isotopic investigations were conducted to determine the geochemical characteristics and to identify the origins of salinization in the shallow Pleistocene aquifer of the El Agamy area, northwestern coast of Egypt. The aquifer shows high salinity values and predominance of Na or Mg and Cl in the groundwater chemical facies suggesting the effect of sea water intrusion on groundwater of the study area. The groundwater of the coastal aquifer showed paths of hydrochemical evolution, from Ca-HCO₃ type to Na-Cl or Mg-Cl type. Most of the molar ratios of the groundwater samples of the study area, compared to the values of seawater molar ratios, show an effect of seawater intrusion to a great extent. High Ca/SO₄ ratio was found in 43.48% of the groundwater samples which suggests reverse cation-exchange processes as well as gypsum dissolution. The sewage effluents are expected to be important sources for increasing fluoride contents in the study area. The concentration of nitrate and ammonia in the groundwater samples of the study area were low. Whereas, 65.22% of the samples have a phosphate concentration higher than

0.5 mg/l indicating a possible sewage source of phosphate. The groundwaters of the study area are slightly to moderately undersaturation with respect to carbonate and sulfate minerals, and strongly undersaturated with respect to halite. Consequently, halite does not play an important role in the groundwater chemistry. Dissolution of carbonate minerals are expected due to the aquifer material is mainly composed of carbonate minerals. The isotopic signature of groundwater indicates that the stable isotope of the groundwater fall near the local meteoric water line and lie in the midway between the Mahmoudia Canal and the Mediterranean Sea. This indicates that major part of this groundwater originated from leakage of municipal wastewater, from improperly constructed septic tanks and directly from injection wells, and mixing with seawater underneath.

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