

CLAY MINERALS IN RELATION TO DEPOSITIONAL ENVIRONMENTS AND WATER CHEMISTRY OF WADI EL NATRUN AREA, EGYPT.

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ABSTRACT

The objective of the current work is to study the mineralogy of the different water bearing formations in Wadi El Natrun and its vicinities in relation to depositional environments and water quality.

The presence of illite with biotite, muscovite and montmorillonite or kaolinite in Miocene aquifer, West of the studied area is an indication of typical fluviomarine and marine environments which reflects high water salinity. In contrast, the disappearance of illite with biotite and muscovite in both Pleistocene and Pliocene aquifers at east of the studied area reflect continental condition and low water salinity.

Several indices; an alkali number and cation exchange index, are used for the identification of water that undergone cation exchange processes.

All groundwater samples of fluviatile Pleistocene, Pliocene and Miocene aquifers in all hydrochemical profiles display an alkali number above 100 and negative values of cation exchange index. This means that the alkaline earths (Ca^{2+} and Mg^{2+}) in their sulfates and carbonates in groundwater replace alkalis on the surface of fluviatile clay minerals in aquifers matrices (equations 1 and 2). These groundwater types have the assemblage of salt combinations (I): NaCl , Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$, $\text{Ca}(\text{HCO}_3)_2$ and (III): NaCl , Na_2SO_4 , MgSO_4 , CaSO_4 and $\text{Ca}(\text{HCO}_3)_2$. These assemblages reflect the effect of leaching and dissolution of terrestrial salts (continental facies groundwater) with some contribution of cation exchange processes.

As a result of cation exchange processes, the increase of Na^+ and decrease in Ca^{2+} and Mg^{2+} concentrations in groundwater due east and northeast of the studied area, lead to the decrease in salts causing temporary hardness in the form of Ca , $\text{Mg}(\text{HCO}_3)_2$ salts, while causing an increase in water salinity but a considerable change in pH and HCO_3^- content. The loss of Ca^{2+} and Mg^{2+} decreases the degree of water saturation with respect to both carbonate and gypsum minerals. This case is quite coincident with the cation exchange equations, where the cation exchange process is related to clay minerals which are formed under fluviatile condition and intercalated with the investigated aquifers matrices.

This means the presence of $\text{CaMg}(\text{HCO}_3)_2$, NaHCO_3 , $\text{Ca}(\text{Mg})\text{SO}_4$ and Na_2SO_4 salts in groundwater of both fluviatile Pleistocene and Pliocene aquifers. Cation exchange reactions lead to relatively low increase in water salinity of continental groundwater facies. This is due to the relatively low values of cation exchange capacity which are related to mineralogical composition of fluviatile clay intercalation and impervious bed barriers of the Pleistocene, Pliocene and Miocene aquifers.

On the other hand, all groundwater samples of marine Pliocene and Miocene aquifers in all profiles (except D-D¹) display an alkali number up to 100 and positive values of cation exchange index. This means that the alkalis in their halogens in groundwater replace Ca^{2+} and Mg^{2+} on the surface of clay minerals in these aquifers matrices which are formed under marine condition. These groundwater types are

characterized by the assemblages (V) and (VI), (IV) of salt combinations that reflect the effect of leaching and dissolution of marine and fluviomarine salts, respectively, with some contribution of cation exchange phenomenon, forming the following hypothetical salt combinations;

V-NaCl, MgCl₂, MgSO₄, CaSO₄ and Ca(HCO₃)₂, marine condition.

VI-NaCl, MgCl₂, CaCl₂, CaSO₄ and Ca(HCO₃)₂, marine condition.

IV-NaCl, MgCl₂, MgSO₄, Mg(HCO₃)₂ and Ca(HCO₃)₂, fluviomarine condition.

In this case, the high concentration of alkalis (Na⁺ + K⁺) in their halogens in groundwater replace (Ca²⁺ + Mg²⁺) on the surface of clay minerals in aquifer matrix, where high concentration of monovalent ions can replace the divalent ions (El Sayed, 1977).

As a result of cation exchange processes, Na⁺ concentration decreases, this is accompanied by an increase of Ca²⁺ and Mg²⁺ concentrations in groundwater due to West and Northwest of the studied area, leading to a decrease in salts causing temporary and an increase in salts causing permanent hardness beside a decrease in expected water salinity of marine groundwater facies but actually a considerable increase in water salinity in the same time due to leaching and dissolution. This case is in agreement with the general reaction of cation exchange.

Noteworthy to mention that the effect of leaching and dissolution of terrestrial, fluviomarine and marine salts is more apparent rather than cation exchange and mixing processes in the different water bearing formations of three profiles. This is due to the relatively low values of cation exchange capacity which are related to mineralogical composition of clay intercalation and impervious bed barriers of the Pleistocene, Pliocene and Miocene aquifers.

INTRODUCTION

The area northwest of Wadi El Natrun is bounded by latitudes 30° 16' & 30° 44' N and longitudes 29° 50' & 30° 30' E and occupies an area of 2860 km².

The origin of clay minerals in sandstones is either detrital or authigenic detrital clays. The authigenic clays are formed in place and include both new and regenerated forms; mainly by intrastratal solutions. The formation of detrital clays is governed by several processes under the influence of many factors (Visher, 1969) such as climate, parent material, relief, organisms and time. The recorded clay mineral assemblage reflects that the climatic conditions during the deposition of these sediments as mentioned by Hamilton (1982).

According to Mackenzie (1965 b) and Millot (1970): clay minerals have three modes of origin:

i) Detrital inheritance from the parent material. ii) Transformation (or alteration). iii) Neof ormation (or synthesis) when physical weathering processes prevailed and chemical weathering is at minimum level. Where a hot or cold dry climate prevail (detrital clays are common) when chemical weathering processes are involved, transformation and neof ormation predominante. It is clear that the formation of clay minerals was controlled by the presence of alkali, alkaline earth elements, pH and Eh in their environment.

Most groundwater samples of the Pleistocene, Pliocene and Miocene aquifers lie in the fresh zone (up 1500 mg/l) which encountered at the eastern and northeastern parts of the study area, referring to the continental origin of

most water bearing formations. The presence of brackish water zones (1500-5000 mg/l) in the Pliocene and Miocene aquifers as well as saline water type (>5000 mg/l) in the Pliocene aquifer are due to the change of depositional environment from fluvial to fluvio-marine, shallow marine and marine at the western and northwestern parts of the Pliocene and western part of the Miocene aquifers, El Sayed and Mohamed (2007).

The objective of this paper is to study the clay minerals of the different water bearing formations in Wadi El Natrun and its vicinities as indicator to depositional environments and water quality.

MATERIALS AND METHODS

In the current work, 28 drilled wells tapping the Pleistocene (16 wells), Pliocene (8 wells) and Miocene (4 wells) aquifers in the studied area were investigated and hydrogeochemically evaluated through the following activities, Fig. (1).

- 1) The chemical analyses of 28 groundwater samples were carried according to the methods adopted by U.S. Geological survey, Rainwater and Thatcher (1960), Fishman and Friedman (1985) and ASTM (2002). These analyses include the determination of total dissolved solids (TDS), pH, EC, cations Ca^{2+} , Mg^{2+} , Na^+ , K^+ and anions CO_3^{2-} , HCO_3^- , SO_4^{2-} and Cl^- .
- 2) Separation of clay fraction (4 samples) representing the three water bearing formation of Pleistocene, Pliocene and Miocene was carried out according to Jackson (1965).
- 3) Mineralogical analysis of the clay fractions was carried out using X-ray diffraction analysis, Philips PW 1170 with Ni filter Cu radiation at 40Kv and SSMA. The samples were scanned at a speed of 2θ / minute, (Moore and Reynolds, 1989).
- 4) Cation exchange capacity of the clay sediments representing the investigated aquifers was carried out according to Jackson (1965).
- 5) Saturation indices of carbonates and non-carbonates minerals were computed for 28 groundwater samples by WATEQ program, (Plummer et al., 1984).

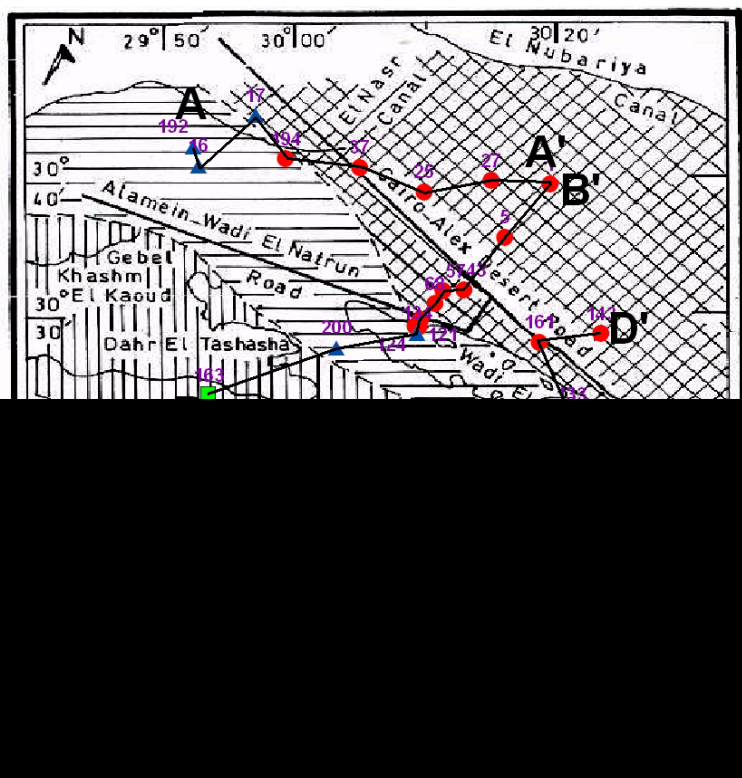


Fig. (1): Areal distribution of the different aquifers and hydrochemical profiles in Wadi El Natrun and its vicinities.

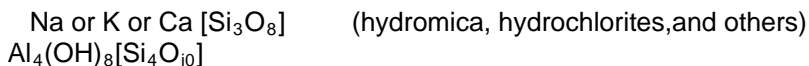
RESULTS AND DISCUSSION

This paper discusses the mineralogy of the clay fraction of different water bearing formations in relation to depositional environments beside water quality within the studied area:

I-Mineralogy of the clay fractions of water bearing formations.

X- ray diffraction patterns of the clay fraction separated from the sediments of Pleistocene aquifer, northeast Wadi El Natrun (sample No. 1A, at a depth of 57 m, well No. 113, depth to water 31m and water salinity 833 mg/l) are presented in Table (1) and Figure (2). They indicate that the clay fraction is dominated by kandite (Kaolinite) which represents more than 40%. The identified accessory minerals, X-ray patterns are quartz (5-15%), followed by both iron-bearing minerals (hematite and ilmenite,) and feldspars (5-15%) with traceable amounts of dolomite(< 5%) and few calcite (5-15%). With regard to the deficiency of the elements Na, K, Mg, Ca and Fe in mild acidic media; moderate Eh and fresh water (old rain and Nile water) products Kaolinite is authigenic clay that can be formed by the hydrothermal alteration or continental weathering of feldspars (orthoclase), feldsparthiods and other

silicates (Deer et al., 1976). The presence of detrital quartz, feldspars (plagioclase, orthoclase) of continental origin indicates that the depositional environment was close to the land mass. The disintegration pattern for feldspars, occurring under the action of water and PCO_2 may be presented as follows:



▲ Toward displacement of cations K, Na, Ca, while interacting with CO_2 , true carbonate and bicarbonate solutions (K_2CO_3 , $CaCO_3$, and others) are formed, Gorshkov and Yakushova (1977). The bicarbonates were precipitated as carbonates along the recharge route, where main sources of recharge to the Pleistocene aquifer are seepage from the irrigation system of Rosetta branch and its tributaries as well as from the old Nile Delta aquifer (Pavlov, 1962 & Atwa, 1968). The recharge to the Pleistocene aquifer, northwest Wadi El Natrun and south El Nasr canal is from the infiltrated water (El Nasr canal and its tributaries and drains) and return water after irrigation is about 87 million m^3 /year, (Diab et al., 2002). Carbonates may be carried away beyond the site of their formation or remain in situ with large quantity of water percolation (carbonatization), (Gorshkov, and Yakushova, , 1977). However; the presence of calcite and dolomite (little amount) are seemingly fluvial condition.

Iron oxides cement are authigenic in origin (continental) and precipitated by intrastratal moving solutions as a result of degradation of iron silicate minerals during chemical weathering (detrital replacement). The hydration reaction of hematite (Fe_2O_3) rapidly alters it to a more stable compound, limonite:

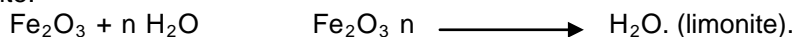


Table (1): Frequency distribution of minerals in the studied clay samples of Wadi El Natrun and its vicinities.

Clay No.	Clay minerals					Accessory minerals								
	Interst.	Smec.	Ill.	Kaol.	Feld.	Qz.	Calc.	Dolo.	Hem.	Goe.	Biot.	Musc.	Ilm.	Oliv.
1 A	-	-	-	++++ +	++	++	++	+	++	-	-	-	++	-
2 B	-	+++	-	++++ +	+	++	+	-	+	+	-	-	-	+
3 C	++	++++	++	++++	-	+	+++	+	+	+	+	+	+	++
4 D	-	++++	+++	+++	-	++	-	+	+	+	+	+	-	+

Notes: Interst. = Interstratified minerals (montmorillinite + mica or chlorite +vermiculite), Smec. = Smectite, Ill.= Illite, Kaol.= Kaolinite, Feld.=Feldspars, Qz.=Quartz, Calc.= Calcite, Dolo.= Dolomite, Hem.= Hematite, Goe.= Goethite, Biot.= Biotite, Musc.=Muscovite, Ilm.=Ilmenite, Oliv.=Olivine. None - : - Trace +: < 5%, Few ++ : 5-15%, Moderate +++: 15-25 %, Common ++++: 25-40 % and Dominant +++++ : >40%.

This mineralogical composition stands in agreement with CEC, where its values of clay of Pleistocene aquifer at different depths (57, 68, 71 and 79 m) are (6.1, 1.3, 1.5 and 1.7 meq/100g clay, respectively, with a mean of 2.7 meq/100g clay).

The basal reflections of the clay fraction of the sediments forming Pliocene aquifer at Wadi El Natrun (sample No. 2 B at a depth of 57 m, well No. 133, depth to water 30m and water salinity 614 mg/l, east W. El Natrun) are depicted in Figure (2). These reflections denote the dominance of kaolinite (kaolinite) which represents more than 40% together with moderate amounts of smectite (montmorillonite, 15-25 %). Accessory minerals are identified as quartz (5-15%) and feldspars (<5%), calcite (<5 %), hematite (<5%), goethite (<5%) and olivine (<5%). With regard to montmorillonite, it is formed by the alteration of basic rocks, or other silicates low in K, under alkaline condition, providing Ca, Mg. The montmorillonite (smectite) is important for base exchange properties and is relatively more stable than illite in the marine environments (Weaver, 1957 and Weaver and Pollard 1973). However, it seems to be the least stable clay mineral during transportation in fresh water. In early diagenetic stages, it can be changed either to Kaolinite or mixed layer montmorillonite and then to illite (Millot, 1970).

This confirms the presence of kaolinite more than montmorillonite with the absence of illite mineral in Pliocene aquifer while in Pleistocene aquifer montmorillonite mineral disappeared. On the other hand, basic or ultrabasic rocks such as olivine mineral are more susceptible to weathering and one can see on their surface agglomeration of the products of weathering at a different stages of disintegration. This mineralogical composition stands in agreement with CEC values of clay of Pliocene aquifer at different depths (45, 48, 57, 67, 76 and 83 m) are (0.4, 1.7, 2.2, 0.4, 1.3 and 5.4 meq/100g clay, respectively, with a mean 1.9 meq/100g clay).

Mineralogical composition of the sediments occupying the Miocene aquifer of west Wadi El Natrun and El -Farigh is represented by two clay samples (samples No. 3C and 4 D at 97 and 98 m depth, respectively, well No. 163, depth to water 95 m and water salinity 3775 mg/l) shows considerable variations with respect to aquifer depths. X-ray diffractograms of the first sample (at depth 97 m) (Fig. 2), indicate that the dominant clay minerals are abnormal montmorillonite (smectite) interstratified minerals (montmorillonite-chlorite, montmorillonite-vermiculite, mica-vermiculite, chlorite-vermiculite and illite-montmorillonite) and kaolinite (kaolinite group) with less considerable amounts of hydrous mica (illite). Quartz, layer silicates, calcite, dolomite, biotite, muscovite, olivine and iron-bearing minerals (ilmenite, goethite and haematite in a descending order) are detected in pronounced amounts. X-ray diffractograms of the second sample (at depth 98 m) (Fig. 2), reveal that the clay minerals assemblage is dominated with smectite (montmorillonite), kaolinite and illite. The accessory minerals are shown to be composed mainly of quartz, dolomite, biotite, muscovite, olivine and iron-bearing minerals (haematite and goethite) in a descending order). The cation exchange capacity at the two depths, 97 m and 98 m, is 6.7 and 6.5 meq/100g clay, respectively, with a mean CEC of 6.6 meq/100g clay.

Fig2

In general, the first sample contains considerably higher content of interstratified minerals which disappeared in the second sample while kaolinite and montmorillonite are seemingly the major clay minerals in both samples. Accessory minerals assemblage is also somewhat different where quartz is a major component of the clay fraction of both samples whereas, calcite is only detected in sample No. 97 and feldspars are entirely absent. Other accessory minerals showed great resemblance in the mineral assemblages of both clay samples.

Commenting on the abovementioned mineralogical composition of the clay fractions representing the sediments of the three aquifers matrices, the following conclusions could be worked out.

a) The presence of a mixture of clay mineral suggests that these sediments have been subjected to fresh water action for a long time, either during their deposition or transportation from the older sediments. Interstratified minerals are confirmative evidence for this suggestion. In this accord, interstratifications may take place either due to transformation of a principal mineral under water action, or a combination of two or more clay minerals. It is considered a transitional stage between the principal minerals and the end products of chemical weathering. The change in the depositional environments is indicated by the decrease in the relative proportions of smectites upwards in the studied Miocene and Pliocene sequence to the advantage of kaolinite and illite. Identification of abnormal montmorillonite is based essentially on the genesis of sediments and its formation from iron-rich materials.

b) The presence of illite with biotite, muscovite and montmorillonite or kaolinite in the Miocene aquifer, West of the studied area (samples 97 and 98) is an indication of a typical fluviomarine and marine environments. This is in agreement with Jackson et al. (1952) and Barashad (1964). In contrast, the disappearance of illite, biotite and muscovite in both Pleistocene and Pliocene aquifers at east of the studied area reflects continental condition.

c) With regard to the accessory minerals, one can figure out that iron-bearing minerals, particularly ilmenite (Fe- oxide) is a true reflection of sediments genesis as the relative enrichment of parent sediments in iron oxides has been manifested in the accessory minerals suite. In addition, the presence of feldspars which are known to be susceptible to weathering, indicates that the sediments are not drastically weathered under the prevailed conditions of the aquifers system.

d) In conclusion, the clay minerals of water bearing formations of both Pleistocene and Pliocene in the east of the studied area were formed under fluviatile condition. Consequently, this mineralogical composition reflects the environments of deposition (continental) and low water salinity (fresh water) of those aquifers. With regard to the clay minerals of water bearing formation of Miocene at the west of the studied area, they were formed under fluviomarine and shallow marine or marine conditions. Consequently, this mineralogical composition reflects the environments of deposition (marine) and high water salinity (brackish and saline waters) of such aquifer.

In brief, the coincidence with the paleo- hydrogeological aspects of the investigated aquifers in the studied can be summarized as follows, Fig. (2):

1-The fluvatile lower Miocene aquifer (El Moghra formation) is distributed south and southeast the study area while fluviomarine and shallow marine lower Miocene aquifer is restricted to the west.

2-The fluvatile lower Pliocene aquifer (wadi El Natrun water bearing formation) occurs at the east and central part of wadi El Natrun depression while fluviomarine and shallow marine middle Pliocene aquifer is distributed due west, North and northwest of the study area.

3-The fluvatile Pleistocene aquifer is distributed in the area between wadi El Natrun and Nile Delta. Noteworthy to mention that the lower Pliocene is directly underlain by the lower Miocene at wadi El Natrun depression and its vicinities. On the other hand, lower Pliocene is directly covered by the Pleistocene aquifer. This is noticed in the area east of Wadi El Natrun and southeast of Nasr canal especially at El Bustan locality.

The lithofacies changes of water bearing formation from continental to marine conditions have their impacts on the groundwater quality. Furthermore, the mutual relation between these aquifers and the adjacent aquifers has their distinct influence on the groundwater chemistry of these aquifers, Said (1962), Sanad (1973) and Abdullah (1996).

II.- The mineralogy of the clay fraction of different water bearing formations in relation to water quality (Spatial variations in groundwater chemistry):

1. Along the east- west direction (profile A-A\)

This profile passes through eight wells representing the fluvatile Pleistocene and marine Pliocene aquifers with a distance of about 41.5 km. The profile is hydrochemically developed in these aquifers along east-west direction (Figs. 1 and 3), where the general flow of groundwater is in the same direction of hydrochemical evolution of such profile. Consequently, the total mineralization (746 mg/l) and ionic concentrations of the groundwater are obviously low in Pleistocene aquifer which is considered as a main recharge area but increases rapidly in the western part to reach their maximum values (within the saline zone, 12492 mg/l at well No.192, marine Pliocene). The high water salinity in the western part reflects fluviomarine and shallow marine origin of Pliocene aquifer. Further northwest, a rapid drop in water salinity and ions concentration from 1370 mg/l at well No. 37 to 647 mg/l, at well No.194 within fluvatile Pleistocene aquifer (depth to water about 4m.) is noticed. This is due to the recharge from El Nasr canal which is considered as a local recharge area leading to an actual dilution of water salinity and ions concentrations. Thus, this canal has a direct effect on the recharge of Pleistocene aquifer through a difference in head ranging from +4.5 to +50 m., causing water-logging problem in some localities (depth to water less than 10 m.) as a result of impermeable clay intercalations with sand and gravels. Na⁺ content is generally higher than that of Cl⁻ along the fluvatile Pleistocene aquifer and vice versa in case of most marine Pliocene aquifer.

The ions concentrations behaviour from east to west direction follows, to a great extent, the geochemical evolution system given by Back and Hanshaw

(1979), where Ca^{2+} , Mg^{2+} and Cl^- ions concentrations go up as the down-gradient distance increases from east to west direction. This indicates the dissolution of salt evaporites along the recharge route and the aquifer matrices.

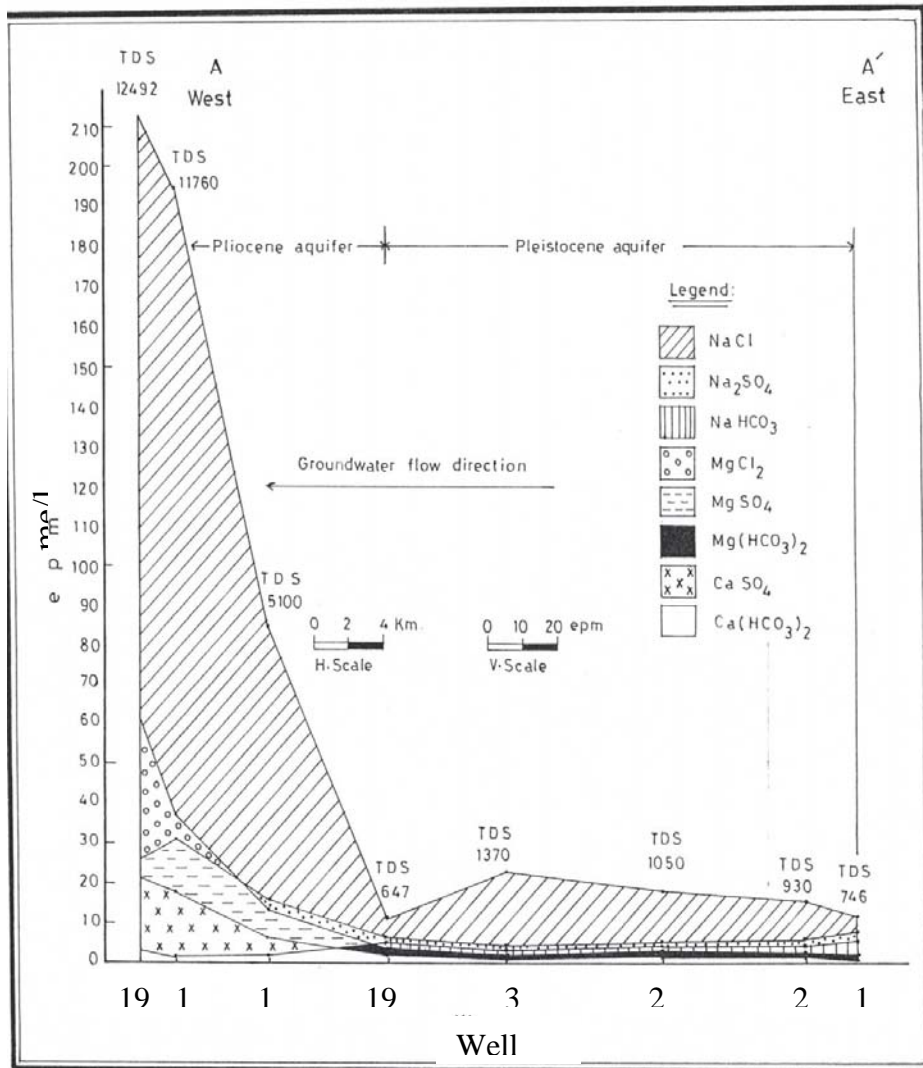


Fig. (3) Hydrochemical profile A – A' in West – East direction.

In contrast, Na^+ , SO_4^{2-} and HCO_3^- ions concentrations decreased in the same direction, indicating the contribution of cation exchange processes (Table 2). Therefore, there is a prominent change in water quality along east to west direction. Generally, in the main recharge area (R, fluvial)

Pleistocene aquifer) groundwater is typically of the Na-HCO₃ type. Such groundwater samples lie down reactions pathways for evolution which displays less mineralization, indicating an uncontaminated recharge source (pure meteoric water, continental facies waters, wells No. 1, 27, 25, 37 and 194). During the groundwater flow from fluvial Pleistocene to marine Pliocene aquifers, subsurface mixing occurred with marine water that has been encroached into deeper parts of the marine Pliocene aquifer (R----->D, down gradient movement----->M, marine facies, wells No. 16 and 192). Consequently, such ions behaviour is in harmony with reactions pathways R----->D----->M within marine Pliocene aquifer, as manifested by water salinity increase up to 12491 mg/l (well No. 192). In the marine Pliocene aquifer, pathway reaction M----->R, flushing took place by recharge from both Pleistocene aquifer and El Nasr canal which have a direct effect on the dilution of marine Pliocene aquifer. This leads to a decrease groundwater salinity of the marine Pliocene aquifer, e.g., few groundwater samples No.16 and No.17 of such profile are subdued under the heading (marine water) M----->R. Concisely, the dissolved solids content decreases (from 11700 mg/l in sample No. 16 to 5100 mg/l in sample No.17), with changing of groundwater facies from marine (assemblage V, rNa/rCl<1) to continental types (assemblage III, rNa/rCl>1) within such aquifer due to cation exchange process, Fig. (4).

The HCO₃⁻ content in forms of Ca and Mg makes about 14 % of TDS at the east (well No.1) then rapidly increases to a maximum of 37% of TDS at well No.194 with a mean of 19% along the fluvial Pleistocene aquifer (Table 3), indicating dissolution of carbonate minerals along the recharge route and aquifer matrix {ion activity products (IAP) < solubility product constant (KSP), (Freeze and Cherry 1979) as clarified in most groundwaters, which are unsaturated with carbonate minerals (Table 4). This indication confirms the feeding from Nile and El Nasr canal water to such groundwater in this particular area. Further west, a rapid decreases in HCO₃⁻ content (forms of Ca and Mg) from 37 % to a minimum of 1% of TDS (wells No.17) then remains constant along the marine Pliocene aquifer (wells No. 16 and 192 (Table 3). This is due to hydrochemical evolution of such groundwater from early (HCO₃⁻ dominant) to more advanced stage of mineralization (Cl⁻ dominant) as well as cation exchange process rather than bicarbonates precipitation as carbonates through groundwater movement from Pleistocene to Pliocene aquifers (most groundwaters are unsaturated with carbonate minerals (Table 4). Such behaviour of water salinity and ions concentrations especially bicarbonate ions which decrease from a mean of 27 at Pleistocene to 1% at marine Pliocene, confirms the general groundwater flow from east to west (Table 2).

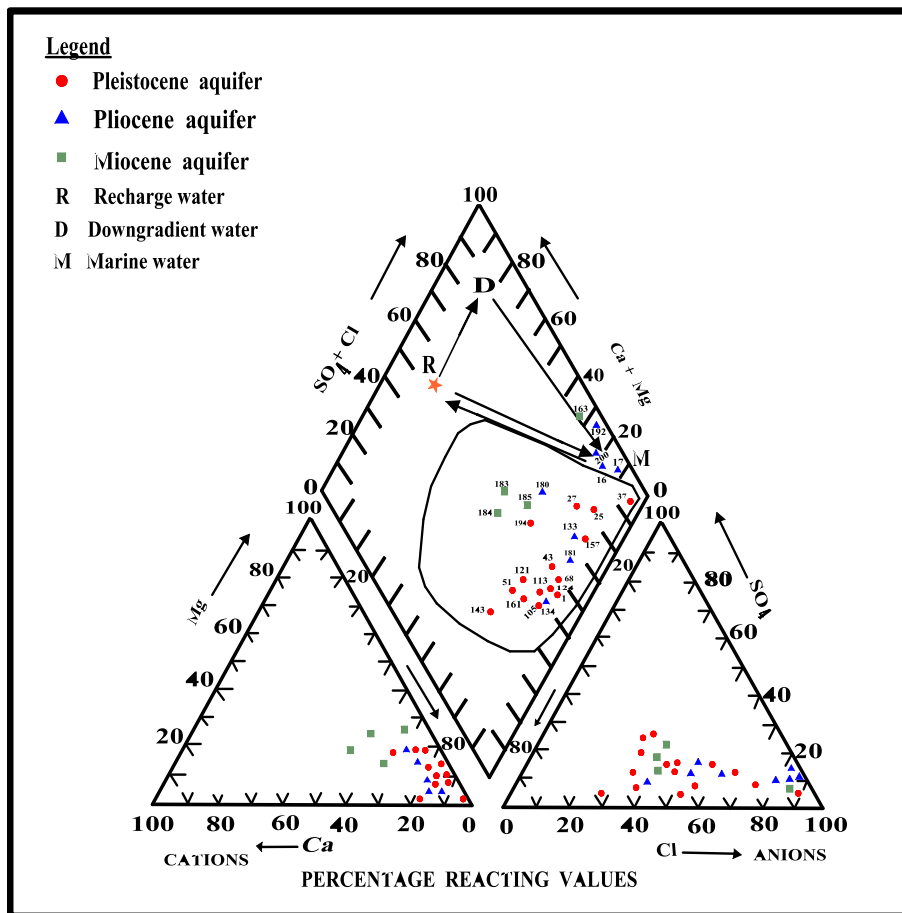


Fig. (4): Back and Hanshaw's evolution diagram (1979) of the different aquifers groundwater along three profiles in Wadi El Natrun area and its vicinities.

Along this profile, the Ca²⁺ and Mg²⁺ ions concentrations decrease while Na⁺ increases from east to west direction, reflecting the contribution of cation exchange beside leaching and dissolution processes. Several indices; an alkali number $[100 (rNa^+ + rK^+)/rCl^-]$ and cation exchange index $[rCl^- - r(Na^+ + K^+)/rCl^-]$, are used for the identification of water that have undergone cation exchange processes (Matthess, 1982). In this regard, all groundwater samples of fluvial Pleistocene aquifer (a main recharge area) in such profile display an alkali number above 100 and negative values of cation exchange index (Table 3). This means that the alkaline earths (Ca²⁺ and Mg²⁺) in their sulfates and carbonates in groundwater replace alkalis on the surface of clay minerals in aquifer matrix.

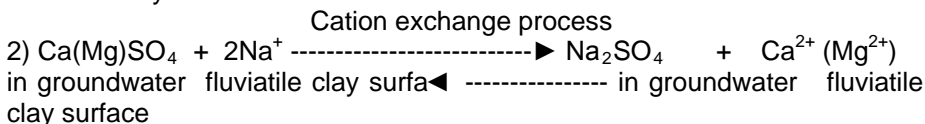
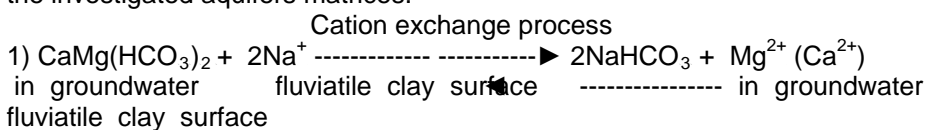
The groundwater types have the assemblage of salt combinations (I) and (III) which reflect the effect of leaching and dissolution of terrestrial salts

(continental facies groundwater) with some contribution of cation exchange processes, forming hypothetical salt combinations as follows:-

I-NaCl, Na₂SO₄, NaHCO₃, Mg(HCO₃)₂, Ca(HCO₃)₂, where the water samples have a temporary hardness {CaMg(HCO₃)₂salts}.

(III): NaCl, Na₂SO₄, MgSO₄, CaSO₄ and Ca(HCO₃)₂, where the water sample (well No. 17) has permanent hardness > temporary hardness.

As a result of cation exchange processes, the increase of Na⁺ concentration (78 to 97 %) and decrease in Ca²⁺ (5 to 2 %) and Mg²⁺ concentrations (9 to 1 %) in groundwater, lead to a decrease in salts causing temporary hardness in form of CaMg(HCO₃)₂ salts (14 to 3%), while causing an increase in water salinity (745 to 1370 mg/l) but change pH (7.5 to 9) and HCO₃⁻ content (45 to 4 %) because each mole of Ca²⁺ or Mg²⁺ adsorbed is replaced by 2 moles of Na⁺. The loss of Ca²⁺ and Mg²⁺ decreases the degree of water saturation with respect to both carbonate and gypsum minerals, (Tables 2 and 4) (Freeze and Cherry, 1979). This case is quite coincident with the following cation exchange equations (1 and 2), where cation exchange process is related to clay minerals which are formed under fluvial condition and intercalated with the investigated aquifers matrices.



Noteworthy to mention that the first and second reactions are reversible (equilibrium condition). This means the presence of CaMg(HCO₃)₂, NaHCO₃, Ca(Mg)SO₄ and Na₂SO₄ salts in groundwater of both fluvialite Pleistocene and Pliocene aquifers. Cation exchange reactions lead to relatively low increase in water salinity of continental groundwater facies. This is due to the relatively low values of cation exchange capacity (range 13 to 61 with a mean 27 me/100g clay) which are related to mineralogical composition of clay intercalation and impervious bed barriers of the Pleistocene aquifer. The Pleistocene aquifer is mainly composed of fluvialite graded sands and gravels with occasional thin lenses of clay, where the salinity of water extract of such aquifer matrices ranges from 353 to 690 mg/l. Consequently, the hydrochemical processes affecting water quality are mainly leaching and dissolution of continental salts, cation exchange and mixing processes which led to an increase in water salinity (745 to 1370 mg/l).

On the other hand, all groundwater samples (except well No. 17) of marine Pliocene aquifer in such profile display an alkali number up to 100 and positive values of cation exchange index. This means that the alkalis in their halogens in groundwater replace Ca²⁺ and Mg²⁺ on the surface of clay minerals in aquifer matrix which is formed under fluvio-marine and marine condition. These groundwater types are characterized by the assemblage (V) of salt combinations that reflect the effect of leaching and dissolution of

aquifers, while being replaced by $MgCl_2$ salt in marine Pliocene aquifer (wells No. 16 and 192). The presence of sodium sulfate salts in the whole Pleistocene and some Pliocene aquifers is mainly due to the dissolution of terrestrial salts rather than cation exchange process, where water bearing formation was formed under fluvial conditions, and the presence of the three bicarbonate salts $NaHCO_3$ and $Ca, Mg(HCO_3)_2$ is due to the effect of meteoric origin. On the other hand, the presence of $MgCl_2$ salt in groundwater samples of Pliocene aquifer is mainly due to the dissolution of marine salts rather than cation exchange process, where water bearing formation was formed under fluviomarine and marine conditions. The absence of both $NaHCO_3$ and $Mg(HCO_3)_2$ salts in the groundwater samples of marine Pliocene aquifer (wells No. 17, 16 and 192) may be due to base exchange after simple dissolution of $Ca(HCO_3)_2$ salt occurring in the aquifer matrices through fresh water of main recharge area (fluvial Pleistocene aquifer, about 47 million m^3 /year, Diab et al., 2002) beside seepage from El Nasr canal. This leads to the replacement of $NaHCO_3$ and $Mg(HCO_3)_2$ salts by $Ca(HCO_3)_2$ salt. This is confirmed by the decreasing of HCO_3^- ions from a mean of 27 % (a main recharge area, fluvial Pleistocene aquifer) to 1% (marine Pliocene aquifer), Table 2.

There are three assemblages of hypothetical salt combinations (I and III, continental facies groundwater and V, marine facies groundwater) dominating this profile while the assemblage of hypothetical salt combination III (continental facies groundwater) is recorded only at well No.17 within fluvial Pliocene aquifer, reflecting the fresh water recharge (about 47 million m^3 /year) from the adjacent Pleistocene aquifer (Diab et al., 2002) beside seepage from El Nasr canal. This confirms that these aquifers are hydraulically connected, as manifested by the previous hydrogeological and present hydrogeochemical evidences. The heterogeneity of the hypothetical salts combinations (I, III and V) along this profile in fluvial Pleistocene aquifer and marine Pliocene aquifers, indicates that the factors affecting groundwater quality are different, this heterogeneity is due to the different feeding from fresh water of Pleistocene aquifer and surface water as well as depositional environment (continental and marine conditions).

The geochemical evolution of the groundwater tapping the concerned aquifers along this profile changes from early advanced stage $HCO_3^- > SO_4^{2-} > Cl^-$ at the East to more advanced stage $Cl^- > SO_4^{2-} > HCO_3^-$ at the west. This prominent change follows the expected changes according to the general gradient evolution of Burdon (1958) due to the recharge by fresh water from the Pleistocene aquifer and El Nasr canal as well as depositional environment (continental and marine conditions). This confirms the expected groundwater flow from east to west that has been hydrogeologically provided (Saad, 1962, Atwa, 1968, , Abdel Baki, 1983, Gomaa 1995, Ahmed 1999, El Sheikh (2000), Diab *et al.*, 2002 and El Sayed and Mohamed, 2007).

Table (2). Chemical analysis of groundwater samples (% of TDS) of the Pleistocene and Pliocene aquifers along the East- West direction (profile A-A) in the studied area.

Aquifer type	Wells No.	TDS (mg/l)	pH (lab.)	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	CO ₃ ²⁻	HCO ₃ ⁻	SO ₄ ²⁻	Cl ⁻
Local recharge area (El Nasr canal)	1	745	9	5	9	85	1	10	36	24	30
						86		46			
	27	930	7.5	9	13	77	1	4	20	12	64
						78		24			
	25	1050	7.5	8	9	81	2	1	17	9	73
						83		18			
	37	1370	8.1	2	1	96	1	1	4	7	88
					97		5				
Local recharge area (El Nasr canal) Pliocene	194	647	8	18	19	55	8	8	33	26	33
						63		41			
	A mean	948	8	8	10	82		27		15	58
	17	5100	7.6	7	8	84	1	---	1	17	82
						85		1			
Local recharge area (El Nasr canal) Pliocene	16	11700	7.1	9	10	80	1	---	1	15	84
						81		1			
	192	12491	8.1	10	19	70	1	---	1	11	88
						71		1			
A mean	9764	7.6	9	12	79		1		14	85	

Table (3). Alkali number, cation exchange index (expressed as meq/l), soluble and hardness salts (%) in groundwater samples of the Pleistocene and Pliocene aquifers along the East- West direction (profile A-A) of the study area.

Aquifer Type	Well No.	Alkali No.	Cation exchange index	Salts	Soluble salts			Temporary hardness salts (CH)		Permanent hardness salts (NCH)		
					NaCl	Na ₂ SO ₄	Na(HCO ₃)	Mg(HCO ₃) ₂	Ca(HCO ₃) ₂	MgCl ₂	MgSO ₄	CaSO ₄
Pleistocene (A main recharge area).	1	297	-ve	I	30	25	31	8	6	----	----	----
								86	14		0	
	27	120	-ve	I	64	12	2	13	9	----	----	----
								78	22		0	
	25	109	-ve	I	73	9	1	9	8	----	----	----
								83	17		0	
	37	111	-ve	I	88	8	1	1	2	----	----	----
Local recharge area (El Nasr canal)	194	197	-ve	I	33	26	4	19	18	----	----	----
								63	37		0	
	A mean	167	-ve	I		81			19		0	
	17	102	-ve	III	82	3	-----	-----	1	-----	8	6
Local recharge (El Nasr canal)	16	94	+ve	V	81	----	----	----	1	3	7	8
								81	1		18	
	192	79	+ve	V	71	----	----	----	1	17	2	9
									71		28	
	A mean	87-102	+ve and -ve	III and V		79			1		20	
Profile A-A	A mean	297-79	-ve and +ve	I,III and V	65	11	5	6	6	2	2	3
						81			12		7	

Table (4). Saturation indices(S_i) of the groundwater samples along the East- West direction (profile A-A\) due to chemical equilibrium with Pleistocene and Pliocene aquifers matrices in the studied area.

Aquifer type	Wells No.	Aragonite CaCO ₃	Calcite CaCO ₃	Dolomite: CaMg(CO ₃) ₂	Huntite CaMg(CO ₃) ₄	Magnesite MgCO ₃	Gypsum CaSO ₄ ·2H ₂ O
Pleistocene (a main recharge area)	1	0.77	0.92	2.23	1.06	0.94	-2.29
	27	-1.01	-0.87	-1.41	-6.3	-0.91	-2.14
	25	-0.60	-0.55	-0.93	-5.49	-0.75	-2.17
	37	-3.14	-3.00	-6.71	-17.94	-4.08	-2.48
Local recharge area (El Nasr canal)	194	0.21	0.36	0.90	-1.80	0.18	-1.83
Local recharge area (El Nasr canal) Pliocene	17	-0.48	-0.33	-0.50	-4.64	-0.54	-1.69
	16	-0.81	-0.67	-1.13	-5.86	-0.83	-0.56
	192	0.65	0.79	2.04	0.74	0.88	-0.65

2. Along the northeast- southwest direction (profile B-B\)

Profile (B-B\) (Figs.1 and 5), is directed to northeast- southwest direction (from the Pleistocene, to both Pliocene and Miocene aquifers northwest of Wadi El Natrun), it passes through 12 wells along the three aquifers with a distance of about 50 km.

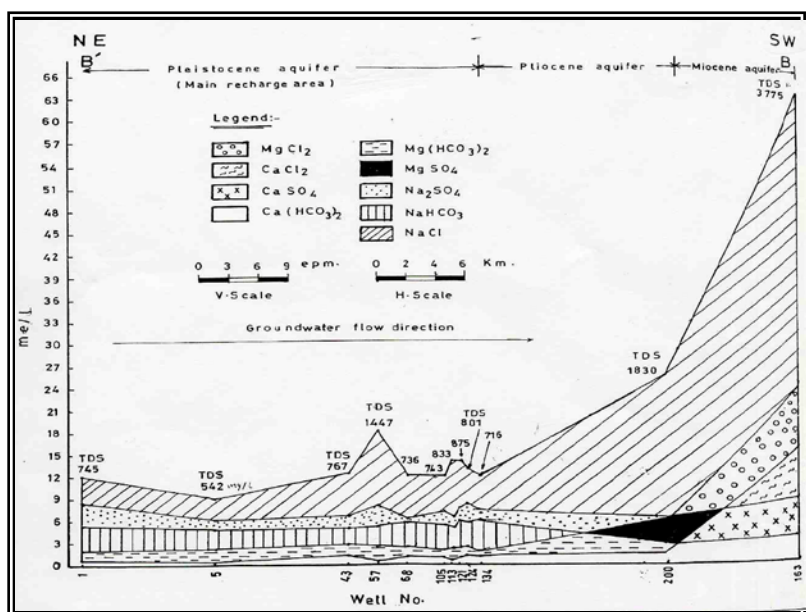


Fig.(5): Hydrochemical profile B – B' in SW – NE direction.

Generally, the hydrochemical properties along this profile, reveals that TDS increases remarkably from fresh (745 mg/l at northeast, fluvatile Pleistocene aquifer) to brackish water (3775 mg/l at southwest, shallow marine and fluviomarine Miocene aquifer) as shown by two peaks; the first (1447 mg/l) corresponds to over-pumping sites and the second (3775 mg/l) is attributed to the lateral facies changes from fluvatile to shallow marine and fluviomarine origin. Such increase in water salinity from northeast to southwest direction follows the same trend of water flow direction.

The low water salinity in both fluvatile Pleistocene and Pliocene aquifers is due to the flow of fresh water from the Nile Delta aquifer towards the Pleistocene and Pliocene aquifers where the Pleistocene aquifer is considered as a main recharge area leading to an actual dilution of water salinity and ions concentrations in both marine Pliocene and Miocene aquifers. The high water salinity is due to the leaching and dissolution of marine sediments, i.e., the water bearing formations of Pliocene and Miocene in the western parts formed under marine condition.

The ions concentration behaviour from northeast to southwest direction follows, to a great extent, the geochemical evolution system given by Back and Hanshaw's (1979) where Ca^{2+} , Mg^{2+} , SO_4^{2-} and Cl^- ions concentrations go up as the down-gradient distance increases from northeast to southwest direction, indicating the dissolution of sulfate minerals and salt evaporites along the recharge route and the aquifer matrices. On the other hand, Na^+ and HCO_3^- ions concentrations decreased in the same direction indicating the contribution of cation exchange processes and precipitation of bicarbonate as carbonate minerals (Tables 5 and 7). Therefore, there is a prominent change in water quality along this direction. Generally, in the main recharge area (R, both fluvatile Pleistocene and Pliocene aquifers) groundwater is typically of the Na- HCO_3 type. Such groundwater samples lie down reactions pathways for evolution, displays less mineralization, indicating an uncontaminated recharge source (pure meteoric water, continental facies waters, wells No.1, 5, 43, 57, 68, 105, 113, 121,124 and 134). During the groundwater flow in fluviomarine Pliocene (well No.200) and marine Miocene (well No.163) aquifers, subsurface mixing occurred with marine water that has been encroached into deeper parts of those aquifers (R----->D----->M). Consequently, such ions behaviour is in harmony with reactions pathways R----->D----->M within fluviomarine Pliocene and marine Miocene aquifers, as manifested by water salinity increase from 745 to 3775 mg/l, Fig. (4).

The HCO_3^- concentration in forms of Ca and Mg considerably decreases from 17% of the TDS(a mean value of the main recharge area) at northeast direction to 6 % of the TDS at southwest direction (marine Miocene aquifers, well No. 163) (Table 6). This is due to the bicarbonates precipitated as carbonates, along the recharge route from Pleistocene to Pliocene and Miocene aquifers as clarified in most groundwaters, which are supersaturated with carbonate minerals, i.e. the reversibility of bicarbonate to carbonate equilibrium due to CO_2 in the aquifer and factors controlling it, (Table 13). Such behaviour of water salinity and ions concentrations especially bicarbonate ions which decrease from a mean of 42 at Pleistocene to 31 at

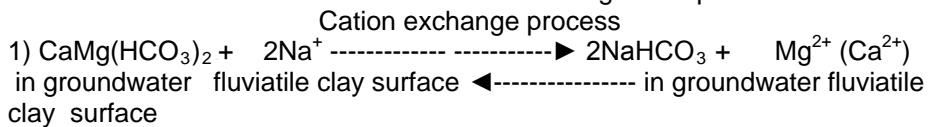
Pliocene and 6% at Miocene aquifers, confirming the general groundwater flow from northeast to southwest (Table 5).

In such profile, the Ca^{2+} and Mg^{2+} ions concentrations go up as the down-gradient distance increases from northeast to southwest direction, while Na^+ ions concentrations decreased in the same direction mainly reflecting the contribution of cation exchange processes.

All groundwater samples of both fluvial Pleistocene (the main recharge area) and Pliocene aquifers in such profile display an alkali number above 120 and negative values of cation exchange index (Table 6). This means that the alkaline earths (Ca^{2+} and Mg^{2+}) in their carbonate in groundwater replace alkalis on the clay minerals surface in aquifer matrix which is formed under continental condition. These groundwater types have the assemblages of salt combinations (I) which reflect the effect of leaching and dissolution of terrestrial salts (continental facies groundwater) with some contribution of cation exchange processes, forming hypothetical salt combinations as follows:-

I-NaCl, Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$, $\text{Ca}(\text{HCO}_3)_2$, where the water samples have temporary hardness $\{\text{CaMg}(\text{HCO}_3)_2$ salts} and have no permanent hardness.

As a result of cation exchange processes, the increase of Na^+ concentration (78 to 83%) and the decrease in Ca^{2+} (5 to 8 %) and Mg^{2+} (18 to 7%) concentrations in groundwater, lead to a decrease in salts causing temporary hardness in the form of Ca, $\text{Mg}(\text{HCO}_3)_2$ salts (22 to 17%), while causing an increase in water salinity (542 to 716 mg/l) but slight change in pH (9.0 to 8.4) and HCO_3^- content (53 to 52 %) because each mole of adsorbed Ca^{2+} or Mg^{2+} is replaced by 2 moles of Na^+ . The loss of Ca^{2+} and Mg^{2+} decreases the degree of water saturation with respect to both carbonate and gypsum minerals, (Table 7) (Freeze and Cherry, 1979). This case is quite coincident with the following cation exchange equation (1), where this cation exchange process is related to clay minerals which are formed under fluvial condition and intercalated with the investigated aquifers matrices.



Cation exchange process

Noteworthy to mention that the first reaction is reversible reaction (equilibrium condition). This dictates the presence of both $\text{CaMg}(\text{HCO}_3)_2$ and NaHCO_3 salts in groundwater of both fluvial Pleistocene and Pliocene aquifers. As a result of this cation exchange reaction, water salinity increases (542 to 716 mg/l) in continental groundwater facies. The salinity of water extract of both fluvial Pleistocene and Pliocene aquifer matrices ranges from 353 to 690 mg/l. Consequently, the hydrochemical processes affecting water quality is mainly leaching and dissolution of continental salts rather than cation exchange and mixing processes. This is due to the relatively low values of cation exchange capacity (range 13 to 61 with a mean 27 and 4 to

54 with a mean 19 meq/100g clay for both fluviatile Pleistocene and Pliocene aquifers, respectively).

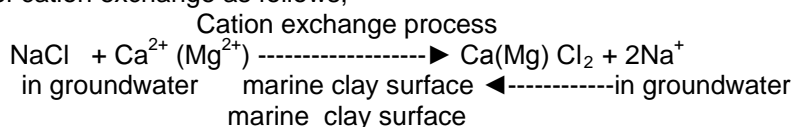
On the other hand, all groundwater samples of both marine Pliocene and Miocene aquifers in such profile display an alkali number up to 100 and positive values of cation exchange index. This means that the alkalis in their halogens in groundwater replace Ca^{2+} and Mg^{2+} on the surface of clay minerals in aquifer matrix which is formed under fluviomarine and marine conditions. These groundwater types are characterized by the assemblages IV and VI of salt combinations that reflect the effect of leaching and dissolution of fluviomarine and marine salts with some contribution of the cation exchange phenomenon, forming the following hypothetical salt combinations in such groundwater;

IV-NaCl, $MgCl_2$, $MgSO_4$, $Mg(HCO_3)_2$ and $Ca(HCO_3)_2$ where water sample NO.200 has the permanent hardness (16%) > temporary hardness (10%) { $MgCl_2$, $MgSO_4$ > Ca $Mg(HCO_3)_2$ }.

VI-NaCl, $MgCl_2$, $CaCl_2$, $CaSO_4$ and $Ca(HCO_3)_2$, where water sample No.163 have the permanent hardness(32%) > temporary hardness(6%) { $MgCl_2$, $CaCl_2$, $CaSO_4$ > $Ca(HCO_3)_2$ }.

In this case, the high concentration of alkalis ($Na^+ + K^+$) in their halogens in groundwater replace ($Ca^{2+} + Mg^{2+}$) on the surface of clay minerals in aquifer matrix, where high concentration of monovalent ions can replace the divalent ions, El Sayed (1977).

As a result of leaching, dissolution and cation exchange processes, Na^+ concentration decreases (83% at fluviatile Pliocene aquifer to 62% at marine Miocene aquifer, well No.163), this is accompanied by an increase of Ca^{2+} (10 at fluviatile Pliocene aquifer to 23% at marine Miocene aquifer, well No. 163) and Mg^{2+} concentrations in groundwater (7 at fluviatile Pliocene to 15% at marine Miocene aquifer, well No. 163). These changes lead to a decrease in salts causing temporary [$Mg(HCO_3)_2$ and $Ca(HCO_3)_2$, 17% at fluviatile Pliocene aquifer to 6% at marine Miocene aquifer, well No. 163] and an increase in salts causing permanent hardness [$MgCl_2$, $CaCl_2$, $MgSO_4$ and $CaSO_4$, 0 % at fluviatile Pliocene aquifer to 32% at marine Miocene aquifer, well No. 163] and a considerable increase in water salinity at the same time (716 mg/l at fluviatile Pliocene aquifer to 3775 mg/l at marine Miocene aquifer, well No.163). This case is in agreement with the general reaction of cation exchange as follows;



Noteworthy to mention that this reaction is reversible. This means the presence of both NaCl and $MgCl_2$ salts in groundwater of both marine Pliocene and Miocene aquifers in such profile. As a result of this cation exchange reaction, a decrease in water salinity of marine groundwater facies is expected but actually water salinity increases from a mean of 716 mg/l at fluviatile Pliocene aquifer to 3775 mg/l at marine Miocene aquifer, well No.163, where the salinity of water extract of fluviatile Pliocene and marine Miocene aquifers matrices ranges from 353 to 1121 and from 2000 to 4000

mg/l, respectively. Therefore, the hydrochemical processes affecting water quality are mainly leaching and dissolution of marine salts rather than cation exchange and mixing processes. Therefore, the increase in salinity of groundwater samples of such profile is due to increasing soluble salts (81% of TDS) that is more than that of temporary (15% of TDS) and permanent salts (4% of TDS) i.e., {NaCl and Na₂ SO₄, 81%} > {(Mg (HCO₃)₂ and Ca (HCO₃)₂, 15%}> (MgCl₂, CaCl₂, CaSO₄ and MgSO₄, 4%) }.

The heterogeneity of the hypothetical salts combinations (I, IV, and VI) along this hydrochemical profile in the three aquifers, indicates that the factors affecting groundwater quality are different, due to the magnitude of feeding by fresh water from a main recharge area (fluvial Pleistocene aquifer, assemblage I) to both marine Pliocene and Miocene aquifers (assemblage IV and VI) as well as seepage from Nile water and depositional environment (continental and marine conditions).

Table (5). Chemical analysis of groundwater samples (% of TDS) of the Pleistocene, Pliocene and Miocene aquifers along the northeast- southwest direction (profile B-B\) in the studied area.

Aquifer type	Wells No.	TDS (mg/l)	pH (lab.)	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	CO ₃ ²⁻	HCO ₃ ⁻	SO ₄ ²⁻	Cl ⁻
Pleistocene (a main recharge area)	1	745	9	5	9	85	1	9	36	25	30
	5	542	9	4	18	86		45		12	35
						77	1	15	38		
	43	767	8.6	11	10	78	1	3	38	11	48
						79		41			
	57	1447	8.5	4	10	85	1	7	21	16	56
						86		28			
	68	736	8.5	7	12	80	1	5	39	4	52
						81		44			
	105	743	8.2	8	9	82	1	3	39	14	44
						83		42			
	113	833	8.4	6	10	83	1	4	33	9	54
84							37				
121	875	8.8	7	8	84	1	7	32	14	47	
					85		39				
124	801	9.3	8	10	81	1	----	44	20	36	
					82		44				
A mean	832	8.7	7	10	83		42		14	44	
Pliocene	134	716	8.7	10	7	82	1	8	44	10	38
						83		52			
	200	1830	7.7	7	16	76	1	2	8	12	78
A mean	1273	8.05	9	11	80		31		11	58	
Miocene	163	3775	7.9	23	15	61	1	1	5	7	87
						62		6			

The chemical composition of groundwater as indicated by the hydrochemical profile displays more hydrochemical evolution ($\text{Cl}^- > \text{SO}_4^{2-} > \text{HCO}_3^-$, both marine Pliocene and Miocene aquifers) in the southwest direction while being less hydrochemically evolved in the northeast direction ($\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-}$, both fluviatile Pleistocene and Pliocene aquifers). Thus, the expected groundwater flow from the hydrochemical evolution, is from northeast to southwest direction. This trend coincides with the regional flow direction of groundwater in the studied area. Therefore, the three aquifers are hydraulically connected to each others. This is clarified from the groundwater samples No.124 and 134 which belong to both fluviatile Pleistocene and Pliocene, respectively, having the same salinity, early metasomatic sequence ($\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-}$) and assemblage of hypothetical salt combinations (I).

3. Along the northeast- southwest direction (profile D-D\).

This profile runs from northeast to southwest direction along the Pleistocene, Pliocene and Miocene aquifers, southwest of Wadi El Natrun, where hydrochemical profile passes through eight wells with a distance of about 30km (Figs. 1 and 6) The profile shows the total mineralization (TDS) is obviously low (303 mg/l, well No.143, Pleistocene aquifer) at northeast but gradually increases towards southwest to reach its maximum values (974mg/l) at well No.181, within the fresh zone (fluviatile Pliocene aquifer). This is indicated by one peak at well No. 181, then decreases rapidly southwestward along the fluviatile Miocene aquifer, where the Pleistocene and Miocene aquifers in such profile are considered as a main and local recharge sources, respectively, leading to an actual dilution of water salinity and ions concentrations. The fresh groundwater of both fluviatile Pleistocene and Miocene aquifers could be explained on basis of the presence of paleo-channels in case of Miocene aquifer. These channels facilitated the fluvial sediments to pass during the Miocene time (Nile sediments). Consequently, water from the Nile and its Delta branches gets its way to flow.

The continuity of this fresh water till now is due to expected hydraulic connection between the Pleistocene, Pliocene and Miocene aquifers in such profile. This agreed well with the movement of fresh groundwater flow from the Nile Delta aquifer towards the Pleistocene and from Pleistocene to Pliocene of wadi El Natrun aquifers and also from southeast parts of Miocene to adjacent Pliocene of wadi El Natrun aquifers, this is in agreement with El Sayed and Mohamed (2007). The relatively high water salinity in the Pliocene aquifer (974mg/l at well No.181) is due to the leaching and dissolution of terrestrial salts for a long distance movement rather than cation exchange process cation exchange.

Fig6

Fig. (6) Hydrochemical profile D – D' in SW – NE direction.

The behaviour of the different ions concentration from northeast (Pleistocene aquifer, wells No. 143 and 161, the main charge area) to southwest direction (Pliocene aquifer, wells No.133, 180 and 181) and from southwest (Miocene aquifer, wells No. 183, 184 and 185, a local recharge area) to northeast (Pliocene aquifer, 181, 180 and 133) in such profile follows, to a less extent, the geochemical evolution system given by Back and Hanshaw (1979), where such groundwater samples lie down reactions pathways for evolution, displays less mineralization, indicating an uncontaminated recharge source (pure meteoric water, continental facies

waters, Fig.(4). This is shown by Na^+ , Cl^- and SO_4^- ions concentrations increase while Ca^{2+} , Mg^{2+} and HCO_3^- decreased due Pliocene aquifer of wadi El Natrun owing to the probable dissolution of salt evaporites and sulfate minerals in the former case and precipitation of carbonate minerals in the latter case (Table 10). This does not exclude the contribution of cation exchange process.

The HCO_3^- concentration in forms of Ca and Mg considerably decreases from 26% of the TDS (a mean value, main recharge area, Pleistocene aquifer) at northeast direction to 20 % of the TDS at the southwest direction (Pliocene aquifer of wadi El Natrun, well No. 181) and also, from 46 % of the TDS at southwest direction (Miocene aquifer) to 20% of the TDS at northeast direction (Pliocene aquifer of wadi El Natrun) due to the bicarbonates precipitated as carbonates along the recharge route from Pleistocene to Pliocene and also, from Miocene to Pliocene aquifers. This is clarified in most groundwaters, which are supersaturated with carbonate minerals (Tables 9 and 10). Such behaviour of water salinity and ions concentrations especially bicarbonate ions which decreases from 70% of the TDS at northeast direction (Pleistocene aquifer) to 31% of the TDS at southwest direction (Pliocene aquifer of wadi El Natrun) and also, from 47% of the TDS at southwest direction (Miocene aquifer) to 31 % of the TDS at northeast direction (Pliocene aquifer of wadi El Natrun) (Table 8), confirms the general groundwater flow from northeast to southwest as well as the local groundwater flow from southwest to northeast towards the southeastern part of wadi El Natrun. These trends are in agreement with El Sheikh (2000) and El Sayed and Mohamed (2007). Generally, such ion behaviour of the groundwater samples in such profile which lie down reactions pathways for evolution system (Back and Hanshow, 1979) displays less mineralization, indicating an uncontaminated recharge source (continental facies waters). The ions behaviour changes from early to less mineralized water (pure meteoric water at two recharge areas to less advanced mineralized water along the recharge route and Pliocene aquifer ($\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^{2-}$ and $\text{Cl}^- > \text{HCO}_3^- > \text{SO}_4^{2-}$, respectively). In other words, the groundwater samples undergo, to a less extent, chemical evolution processes, where the metasomatic sequence did not reach the final sequence of more advanced stage ($\text{Cl}^- > \text{SO}_4^{2-} > \text{HCO}_3^-$). In such profile, aggradations in chemical development from early to less advanced stage in Pleistocene to Pliocene aquifers along northeast- southwest and also from Miocene to Pliocene aquifers along southwest- northeast directions is noticed. This suggests that the general groundwater flow is from Pleistocene to Pliocene aquifers along northeast- southwest and also a local flow from Miocene to Pliocene aquifers along southwest- northeast directions. Pleistocene and Pliocene aquifers along northeast- southwest and also, Miocene and Pliocene aquifers along southwest- northeast directions are hydraulically connected in such profile. This may be confirmed by the apparent homogeneity in the prevailing salt combinations (I) along the whole profile.

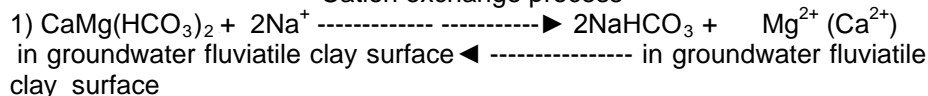
In such profile, the Na^+ concentrations increase while Ca^{2+} and Mg^{2+} decrease due Pliocene aquifer of wadi El Natrun, mainly reflecting the

contribution of cation exchange beside leaching and dissolution processes. All groundwater samples in such profile display an alkali number above 120 and negative values of cation exchange index (Table 9). This means that the alkaline earths (Ca^{2+} and Mg^{2+}) in their carbonate in groundwater replace alkalis on the surface of clay minerals in aquifer matrix. The groundwater types have a unique assemblage of salt combinations (I) which reflects the effect of leaching and dissolution of terrestrial salts (continental facies groundwater) with some contribution of cation exchange processes, as follows:-

I- NaCl , Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$, $\text{Ca}(\text{HCO}_3)_2$, where the water samples have temporary hardness { $\text{CaMg}(\text{HCO}_3)_2$ salts} and have no permanent hardness.

As a result of cation exchange processes, the increase of Na^+ concentration (69-80% and 53- 80%) and decrease in Ca^{2+} (11-7% and 24-7%) and Mg^{2+} (20-13% and 23-13%) concentration in groundwater due Pliocene aquifer of wadi El Natrun, lead to the decrease in salts causing temporary hardness (31-20% and 46-20%) in the form of Ca , $\text{Mg}(\text{HCO}_3)_2$ salts, while causing an increase in water salinity (303-974 mg/l and 494-974 mg/l) but a considerable change in pH (8-8.9) because each mole of Ca^{2+} or Mg^{2+} adsorbed is replaced by 2 moles of Na (Tables 8 and 9). The loss of Ca^{2+} and Mg^{2+} decreases the degree of water saturation with respect to both carbonate and gypsum minerals, (Freeze and Cherry, 1979), (Table 10). This case is quite coincident with the following cation exchange equations (1 and 2), where this cation exchange process is related to clay minerals which are formed under fluvial condition and intercalated with the investigated aquifers matrices.

Cation exchange process



Cation exchange process

Noteworthy to mention that the first reaction is reversible reaction (equilibrium condition). This dictates the presence of both $\text{CaMg}(\text{HCO}_3)_2$ and NaHCO_3 salts in groundwater of fluvial Pleistocene, Pliocene and Miocene aquifers. Therefore, the increase in salinity of groundwater samples from Pleistocene (a main recharge area) to Pliocene and from Miocene (a local recharge area) to Pliocene aquifers is due to increasing soluble salts (mean 74- 80 and 59-80% of TDS, respectively) and decreasing temporary salts (mean 26- 20 and 41-20 % of TDS, respectively, (Tables 8 and 9).

One assemblage of hypothetical salt combination [I: NaCl , Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$ and $\text{Ca}(\text{HCO}_3)_2$] dominates the whole profile. The apparent homogeneity in the prevailing salt combinations (I) along the profile is a good evidence that factors affecting groundwater quality in this particular area are mainly leaching and dissolution under continental condition rather than cation exchange process. This is due to the relatively low values of cation exchange capacity (range 6.5 to 6.7 with a mean of 6.6 and 1.3 to 6.1 with a mean 2.7 meq/100g clay for both Miocene and Pleistocene aquifers,

respectively) which are related to mineralogical composition of fluvial clay intercalation and impervious bed barriers of the Miocene and Pleistocene aquifers, respectively. The Pleistocene, Pliocene and Miocene aquifers in such profile are mainly composed of both fluvial coarse sands and sandy clay deposits and the salinity of water extract of Miocene and Pleistocene aquifer matrices ranges from 238 to 690 mg/l. . Furthermore, the presence of Na_2SO_4 salt reflects the effect of leaching and dissolution of terrestrial salts rather than cation exchange and mixing processes, where the terrestrial salts are transported along the recharge route from Pleistocene to Pliocene aquifers and also, from Miocene to Pliocene aquifers. Also, the presence of Na_2SO_4 , NaHCO_3 , $\text{Mg}(\text{HCO}_3)_2$ and $\text{Ca}(\text{HCO}_3)_2$ salts in the groundwater samples of such profile reflects the contribution of the recent recharge (Nile water) to these aquifers as well as cation exchange processes.

Table (8) Chemical analysis of groundwater samples (% of TDS) of the Pleistocene, Pliocene and Miocene aquifers along the northeast- southwest direction (profile D-D') in the studied area.

Aquifer type	Wells No.	TDS	pH (lab.)	Ca^{2+}	Mg^{2+}	Na^+	K^+	CO_3^{2-}	HCO_3^-	SO_4^{2-}	Cl^-
Pleistocene (a main recharge area)	143	303	8.9	11	20	67	2	10	60	3	27
	161	374	8.8	17	3	69		70		9	38
						80		53			
	A mean	339	8.85	14	12	75		62		6	32
Pliocene	133	614	8.7	6	10	83	1	2	27	12	59
	180	871	8.2	6	18	84		29		13	52
						75		6			
	181	974	8.2	7	13	76		35		14	55
						79		6			
A mean	753	8.4	6	14	80		32		13	55	
Miocene (a local recharge area)	185	670	8.2	11	27	66	1	7	30	21	42
	184	677	8.0	19	26	67		37		13	42
						54		7			
	183	494	8.4	24	23	55		45		14	39
						52		7			
A mean	614	8.2	18	25	58		43		16	41	

CONCLUSION

The objective of the current work is to study the mineralogy of the different water bearing formations in Wadi El Natrun and its vicinities in relation to depositional environments and water quality which is used for drinking, domestic and agricultural purposes.

The presence of illite with biotite, muscovite and montmorillonite or kaolinite in Miocene aquifer, West of the studied area is an indication of typical fluviomarine and marine environments which reflects high water salinity. In contrast, the disappearance of illite with biotite and muscovite in both Pleistocene and Pliocene aquifers at east of the studied area reflect continental condition and low water salinity.

The chemical composition of groundwater samples as indicated by the hydrochemical profiles (A-A), east -west) and (B'-B, northeast-southwest) displays less hydrochemical evolution in the East and northeast while being more hydrochemically evolved in the west and southwest directions, respectively. Thus the expected groundwater flow is from the less hydrochemical evolution at east and northeast to more hydrochemical evolution at west and southwest directions, respectively. This trend coincides with the regional flow direction of groundwater in the studied area. Finally, these hydrochemical profiles suggest a hydraulic connection between Pleistocene, Pliocene and Miocene aquifers. All groundwater samples of Pleistocene and some groundwater samples of Pliocene aquifers in those profiles display an alkali number above 120, negative values of cation exchange index and the assemblages of salt combinations (I) and (III), reflecting the effect of fluvial environments, where the clay minerals were formed under continental condition. On the other hand, some groundwater samples of both Pliocene (both profiles) and Miocene (profile B'-B) aquifers, display an alkali number up to 100, positive values of cation exchange index and the assemblages V (profile A-A), IV (profile B'-B) and VI (profile (B'-B) of salt combinations, respectively, reflecting the effect of fluviomarine and marine conditions, where the clay minerals were formed. Therefore, the less hydrochemical evolution reflects the effect of fluvial environment while the more hydrochemical evolution reflects the effect of fluviomarine and marine conditions.

The behaviour of different ions in the hydrochemical profiles (D-D) along northeast and southwest direction from Pleistocene to Pliocene and Miocene to Pliocene aquifers follows, to some extent, the geochemical evolution systems given by Burdon (1959) and (Back and Hanshaw, 1979). Therefore, there is a slight change in water quality along such direction. Aggradations in chemical development from early to less advanced stage in Pleistocene to Pliocene aquifers along northeast- southwest and also from Miocene to Pliocene aquifers along southwest- northeast directions as well as the apparent homogeneity in the prevailing salt combinations (I) along the profile suggests hydraulic connection between Pleistocene and Pliocene and also between Miocene to Pliocene aquifers. This also, confirms the occurrence of two directions of groundwater flow where the first is general

flow from northeast- southwest (Pleistocene to Pliocene aquifers) and the second is local flow from southwest- northeast directions (Miocene to Pliocene aquifers). All groundwater of Pleistocene, Pliocene and Miocene aquifers in such profile displays an alkali number above 120, negative values of cation exchange index and the assemblages of salt combinations (I), reflecting the effect of fluvial environment, where the clay minerals were formed under continental condition.

I-NaCl, Na₂SO₄, NaHCO₃, Mg(HCO₃)₂, Ca(HCO₃)₂, where the water samples have temporary hardness {CaMg(HCO₃)₂salts} and have no permanent hardness.

As a result of cation exchange processes, the increase of Na⁺ concentration and decrease in Ca²⁺ and Mg²⁺ concentrations in groundwater, lead to a decrease in salts causing temporary hardness in form of CaMg(HCO₃)₂ salts, while causing an increase in water salinity but change in pH and HCO₃⁻ content because each mole of Ca²⁺ or Mg²⁺ adsorbed is replaced by 2 moles of Na⁺.

All groundwater of Pleistocene and most groundwater samples of Pliocene and Miocene aquifers in all profiles lie down reactions pathways for chemical evolution, displays less mineralization, indicating an uncontaminated recharge source (pure meteoric water, continental facies waters). During the groundwater flow in fluvio-marine and both marine Pliocene and Miocene aquifers, subsurface mixing occurred with marine water that has been encroached into deeper parts of those aquifers (R----->D----->M,). Consequently, such ions behaviour of some groundwater samples is in harmony with reactions pathways R----->D----->M within fluvio-marine and marine Pliocene and marine Miocene aquifers, as manifested by water salinity increase. In the marine Pliocene aquifer, pathway reaction M----->R, flushing took place by recharge from both Pleistocene aquifer and El Nasr canal. This leads to decreasing salinity of groundwater in the marine Pliocene aquifer.

Noteworthy to mention that the effect of leaching and dissolution of terrestrial and marine salts is more apparent than cation exchange and mixing processes in the three profiles. Also, the clay minerals are an indicator to depositional environments and water quality of the different aquifers in the studied area.

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"معادن الطين وعلاقتها بظروف الترسيب وكيميائية المياه بمنطقة وادي النطرون- مصر".

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يهدف هذا البحث الى دراسة علاقة معادن الطين الموجودة في الطبقات الحاملة للمياه المختلفة ببيئات الترسيب وبنوعية المياه الجوفية التي تستخدم في أغراض الشرب والزراعة والأغراض المنزلية بوادي النطرون والمناطق المتاخمة.

ويدل وجود معادن الإليت مع البيوتاييت، ومعادن الموسكوفيت مع المونتمورلينييت أو الكولنيت في عينات الطين لخزان الميوسين بغرب منطقة الدراسة على بيئة الترسيب البحرية والنهرية والتي تعكس ارتفاع لموحة المياه. وفي المقابل، فإن اختفاء معادن الإليت مع البيوتاييت، ومعادن الموسكوفيت في عينات الطين في خزاني البليستوسين والبليوسين في شرق منطقة الدراسة يعكس ظروف الترسيب القارية وعذوبة المياه.

ويدل ايضا التركيب الكيميائي لعينات المياه الجوفية كما يتبين من القطاعات الهيدروكيميائية (A- A'، شرق- غرب) و (B-B' والشمال الشرقي – الجنوب الغربي) على أن هناك تطور هيدروكيميائي الأفل (بسيط) في الشرق والشمال الشرقي في حين أن هناك تطور هيدروكيميائي الأقصى (كبير) في اتجاهات الغرب والجنوب الغربي بمنطقة الدراسة، على التوالي طبقا لنظم التطور الجيو كيميائي بوردن (1958) و باك وهانشو (1979). هذا التغيير في التطور الهيدروكيميائي يتوافق مع الاتجاه العام لحركة المياه الجوفية في منطقة الدراسة من الشرق والشمال الشرقي إلى الغرب والجنوب الغربي كما تشير الى أن هناك اتصال هيدروليكي بين خزانات البليستوسين والبليوسين والميوسين. جميع المياه الجوفية لخزان البليستوسين وبعض عينات المياه الجوفية لخزان البليوسين في هذه القطاعات الهيدروكيميائية تعطى عدد قلوي أكبر من 120، وقيم سلبية لمعامل التبادل الكاتيوني، ومجموعات الأملاح القارية المتكونة تعكس تأثير البيئة النهرية (القارية) حيث التركيب المعدني لعينات الطين تكونت تحت ظروف نهرية قديمة. ومن ناحية أخرى، فإن بعض عينات المياه الجوفية لخزاني البليوسين والميوسين تعطى عدد قلوي يصل إلى 100، وقيم إيجابية لمعامل التبادل الكاتيوني ومجموعات الأملاح البحرية المتكونة تعكس تأثير البيئة النهرية أو البحرية حيث التركيب المعدني لعينات الطين تكونت تحت ظروف نهرية وبحرية.

وسلوك الأيونات المختلفة في القطاع الهيدروكيميائي (D-D') في اتجاه شمال شرق وجنوب غرب في خزانات البليستوسين إلى البليوسين ومن الميوسين إلى البليوسين يتبع إلى حد ما، نظم التطور الجيو كيميائي لبوردين (1958) و باك وهانشو (1979). ولذلك، هناك تغير طفيف في نوعية المياه على طول هذا الاتجاه. والتدرج في التطور الكيميائي نادرا أي غير متأثر بالعمليات الكيميائية (مياه بكر) إلى أقل مرحلة متطورة في خزانات البليستوسين إلى البليوسين في الاتجاهات الشمال الشرقي- الجنوب الغربي، وأيضا في خزانات الميوسين إلى البليوسين في الاتجاهات الجنوب الغربي - الشمال الشرقي وكذلك تجانس واضح في مجموعات الأملاح القارية السائدة (I) خلال القطاع الهيدروكيميائي يدل على أنه هناك اتصال هيدروليكي بين خزانات البليستوسين والبليوسين وكذلك بين خزانات الميوسين والبليوسين. هذا أيضا، يؤكد وجود اتجاهين لحركة المياه الجوفية الأول اتجاه عام من الشمال الشرقي الى الجنوب الغربي، والثاني ثانوي من الجنوب الغربي الى الشمال الشرقي. كل المياه الجوفية التابعة لخزانات البليستوسين والبليوسين والميوسين تعطى عدد قلوي فوق 120، وقيم سلبية لمعامل التبادل الكاتيوني ومجموعات الأملاح القارية، مما يعكس تأثير البيئة النهرية حيث التركيب المعدني لعينات الطين تكونت تحت ظروف نهرية.

ومما هو جدير بالذكر أن تأثير عمليات الغسيل والإذابة للأملاح القارية هو أكثر من تأثير عملية التبادل الأيوني والخلط خلال القطاعات الهيدروكيميائية الثلاثة.

قام بتحكيم البحث

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