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USE OF ENVIRONMENTAL ISOTOPES TO EVALUATE THE SOURCES OF SUBMARINE SPRINGS ALONG THE SOUTH COAST OF LEBANON

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Geochemical and isotopic techniques are applied to evaluate the origin of submarine freshwater along the southern coast of Lebanon and to compare it with the northern coast. Results indicate that submarine springs in the southern and northern coasts have different geochemistries reflecting a difference in the specific geology of the regions. Environmental isotopes including ^2H , $\delta^{18}\text{O}$, and $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ in sulfate have confirmed the different geological conditions. A more enriched isotopic composition of submarine groundwater occurs in the south where submarine springs are primarily artesian flows that occur in confined aquifers. The deep circulation of groundwater is affected by a high geothermal gradient. Also high sulfate content is related to a fractionation in $\delta^{34}\text{S}$ isotopic composition. This is due to the dissolution of gypsum minerals and reduction of the Upper Cretaceous and Lower Eocene marly limestones in the confined aquifers. At the northern coast, a slightly depleted isotopic composition is found for submarine groundwater. The highly karstified northern region allows rapid infiltration of groundwater and discharge as submarine springs. Tritium results for all submarine waters indicate a short subsurface residence time in the aquifers when compared to rainwater in Lebanon.

INTRODUCTION

Geochemical parameters are important tools used to define groundwater quality and its origin and to characterize its flowpaths in aquifers. During hydrochemical evolution, the concentration of individual ionic species can increase, remains constant, or decrease. In the case of water-rock interaction in submarine freshwater, changes in calcium, magnesium and sulfate are due to anhydrite dissolution and dedolomitization (Plummer et al., 1994). Sodium increase along the aquifer flowpath is attributed to ion exchange with clay minerals (Clark and Fritz, 1997). In submarine freshwater, sulfur sources can be divided into mineral or rock contributions, marine and volcanic sources (Gislason and Eugster, 1987). Ionic molar ratios would distinguish between the different sources of sulfate in water. Environmental stable isotopes (oxygen-18 and deuterium) in combination with the geochemical parameters are used to determine the recharge origin and mixing of groundwater in submarine aquifers. The isotopic compositions of groundwater are governed by recharge from precipitation and infiltration from surface waters.

In karst systems, groundwater mixing is high where connected porosity occurs in small-scale fissures and the porous matrix that provides much of the storage in the system, and which contribute to base flow. Stable isotopes of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ can serve to distinguish flow through different systems. In deep and confined aquifers, fractures and faults allow groundwater to discharge as artesian flows into the sea giving submarine freshwater springs. The geothermal gradient within this deep flowpath increases the temperature of water. On the other hand, the isotopic ratio of sulfur in combination with $\delta^{18}\text{O}$ can be applied to observe special chemical processes in surface and groundwater. The stable isotopic ratio of the sulphur isotopes ($\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ in sulfate) allows differentiating between marine, evaporitic and volcanic sources of dissolved sulfate in groundwater (Krouse, 1980). The isotopic composition of sulfur in these sources, with the exception of seawater, varies in a wide range. When sulfur from various natural and anthropogenic sources dissolves in cold groundwater, no appreciable fractionation takes place unless dissolved sulfate is microbially reduced. The exchange of oxygen between SO_4^{2-} and H_2O is extremely slow at the low temperature of groundwater (Chiba and Sakai, 1985). Therefore, the isotopic composition of oxygen in sulfate reflects either mixing or reduction to sulfides.

These geochemical and isotopic techniques will be applied to delineate the origin of submarine freshwater in different coastal regions in Lebanon. In Lebanon, the heavily uplifted and dissected terrain displays extensive outcrops of an almost complete stratigraphic sequence of Early Jurassic to Quaternary age, mostly in conditions of only minimal tectonic deformation (Walley, 1998; Dubertret, 1975). The Jurassic period includes mainly dolomite and dolomitic limestone. The Cretaceous period consists of limestone, marly limestone and dolomite. With respect to groundwater, these are the most important sediments and their widespread and interconnected karstic nature makes them the source of major springs in Lebanon (Mijatovic et al., 1967). Some of these springs extend some distance into the sea such as the submarine spring at the Litani river mouth located 3 km offshore (FAO, 1972). In coastal Lebanon, the conditions most influencing water flows from land into the sea are geologic faults and karstic galleries. Around 54 submarine springs are identified throughout along the coast of Lebanon from the north to the south (NCRS, 2002). It was found that 52% of these springs are issuing from karstic rock formations with high flow rates; 22% are issuing along fault alignments with high flow rates; and 18% are offshore springs with low to high interrupted flow rates (Shaban et al., 2005; Saad et al., 2005). It is clear that the discharge of submarine freshwater flows is strongly influenced by the specific geological features at different locations.

This study gives an important insight into the geological differences in submarine springs between the northern and the southern coasts in Lebanon, by using hydrochemical and isotopic tools. The origin of recharge in the submarine springs is determined as a function of the variation of water quality through water-rock interactions.

MATERIALS AND METHODS

The first submarine springs studied are located along the northern coast in Lebanon near Chekka bay and they flow to the sea via a complex of fault and karstic zones. These submarine springs are estimated to flow from the Turonian-Cenomanian limestones at a mean depth of 120 m below mean sea level and at an average distance from the shoreline of 700 m (Qareh, 1966; Shaban et al., 2005). The second set of submarine springs are located along the south coast where hydraulic parameters are found to be more developed than in the north. These springs show longer lineament extents, dense fracturing systems and conduit extension offshore (Shaban et al., 2005). Aquifers in the coastal plateau of south Lebanon are primarily in Cenomanian-Turonian strata. In this region, artesian aquifers can extend for considerable distances offshore due to confining of the aquifer by the cover of marls and marly limestones of the Upper Cretaceous and Lower Eocene. The combined hydrochemical and environmental isotope survey ($\delta^{18}\text{O}$, $\delta^2\text{H}$, $\delta^{34}\text{S}$) was conducted for the following samples - along the north coast, sampling sites are 1 coastal groundwater, 2 submarine springs, and 1 sea water, and in the south, sampling sites are 3 coastal surface freshwater, 2 coastal groundwater, 4 submarine springs, and 1 sea water.

Locations of samples are presented in Table 1. All samples were preserved before analysis. Aliquots of water were stored in polyethylene bottles. The rest of the samples were passed through 0.45 μm Millipore filters and then refrigerated for analysis. The pH, electrical conductivity (EC), and total dissolved solids (TDS) were measured in the field. The laboratory chemical analyses of the water samples were conducted immediately after their collection. Anions and cations were analyzed with atomic absorption spectroscopy (Perkin Elmer 100) and an ion chromatograph (Dionex IC25). Each sample was analyzed for $\delta^{18}\text{O}$, $\delta^2\text{H}$, $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ in sulfate.

RESULTS AND DISCUSSION

Table 2 presents general physical characterization of all water samples. Mean value for TDS of coastal freshwater is 750 mg/L whereas mean TDS for submarine water is 7800 mg/L. This higher value in submarine freshwater is due either to mixing with sea water or to the elevated residence time in aquifers that promotes chemical dissolution (Freeze and Cherry, 1979). A similar trend of variation is observed with the conductivity parameter. The pH values range from 7 to 8.5. These values indicate a slight trend of alkaline chemical reaction within the aquifers in the study area.

Hydrochemical Characterization of Water Samples

Groundwater contains dissolved minerals derived from the geologic materials through which it moves. Factors affecting groundwater chemistry include residence time, length of flow path, initial water chemistry, chemical reactions, and land use. The mineral quality of water can be classified based on the predominant cations and anions. Major cations include calcium, magnesium, sodium and potassium. Major anions include bicarbonate, chloride, and sulfate.

Mean concentrations of calcium and bicarbonate are respectively 250 mg/L and 110 mg/L. The converted values of these parameters in milliequivalents per liter represent more than 50 percent of the cations and anions respectively in groundwater. This result indicates that coastal groundwater

Table 1. Location of sampling sites.

Site name and locality	Coordinates
1- Surface water Brak Ras el Ain, South	N33°28'59.1'' E35°19'28''
2- Surface water El saide, South	N33°26'18.1'' E35°17'25.5''
3- Surface water Kasmiyeh, South	N33°22'02'' E35°16'4.4''
4- Groundwater 1 Tyre, South	N33°18'15.1'' E35°14'31.4''
5- Groundwater 2 Tyre, South	N33°18'15.5'' E35°14'32''
6- Submarine Tyre, South	N33°20'7.92'' E35°10'46.7''
7- Submarine Borgolieh, South	N33°19'33.7'' E35°10'6.76''
8- Submarine Nsariyeh, South	N33°10'10.1'' E35°10'0.23''
9- Submarine Abou Asswad, South	N33°22'19'' E35°15'03''
10- Seawater Tyre	
11- Surface water Chekka	N34°16'5.4'' E35°39'38.1''
12- Groundwater Chekka, North	N34°2'40.8'' E35°37'51.1''
13- Submarine Chekka, North	N34°20'22.4'' E35°43'25.3''
14- Submarine Chekka, North	N34°20'23'' E35°43'26''
15- Seawater Chekka	

Table 2. Characteristics of sampling sites.

Site name and locality	pH	Conductivity mS	TDS mg/L
1- Surface water Brak Ras el Ain, South	7.01	0.6	420
2- Surface water El saide, South	7.6	0.64	434
3- Surface water Kasmiyeh, South	8.25	1.58	1062
4- groundwater 1 Tyre, South	8.37	1.35	935
5- groundwater 2 Tyre, South	7.6	1.46	990
6- Submarine Tyre, South	7.47	52.2	7920
7- Submarine Borgolieh, South	7.34	18.3	10900
8- Submarine Nsariyeh, South	7.16	39	2290
9- Submarine Abou Asswad, South	7.4	23.7	13400
10- Seawater Tyre	8.5	53.5	15000
11- Surface water Chekka	6.7	1.27	885
12- groundwater Chekka, North	7.2	0.66	709
13- Submarine Chekka, North	8.3	34.8	5090
14- Submarine Chekka, North	7.2	13.4	7100
15- Seawater Chekka	8.5	52.5	15000

is classified as calcium bicarbonate type (Hem, 1985). This is typical of regional groundwater chemistry within the limestone aquifer system in the area.

In order to evaluate the origin of water solutes in coastal and submarine groundwater, variations of chloride versus Na, K and Mg are plotted (Figure 1). If samples plot near the seawater composition, their solutes may represent simple dilution of seawater or the dissolution of marine

aerosols in the recharge area along the coast. Deviations from the sea water composition indicate potential solute sources contributing to the chemistry of water such as subsurface reaction with regional sedimentary rocks including the aquifer rocks (Struchio et al., 1996).

Variation of sodium against chlorine (Figure 1) shows a high correlation coefficient between coastal and submarine freshwater. This indicates that Na and Cl have the same origin in both types of water. Also, these points lie along the sea water dilution line indicating that the major origin of Na and Cl solutes in submarine freshwater is sea water. On the other hand Na and Cl concentrations in freshwater are 10 fold less that the sea water, indicating a low mixing with sea.

Figure 1 also shows that concentrations of K and Mg versus chloride deviate from the sea water dilution line. Possible sources of magnesium could be the Upper Cretaceous and Eocene carbonate rocks overlying much of the region. The enrichment in Mg may indicate that some of the excess of Ca was derived from the dissolution of dolomite.

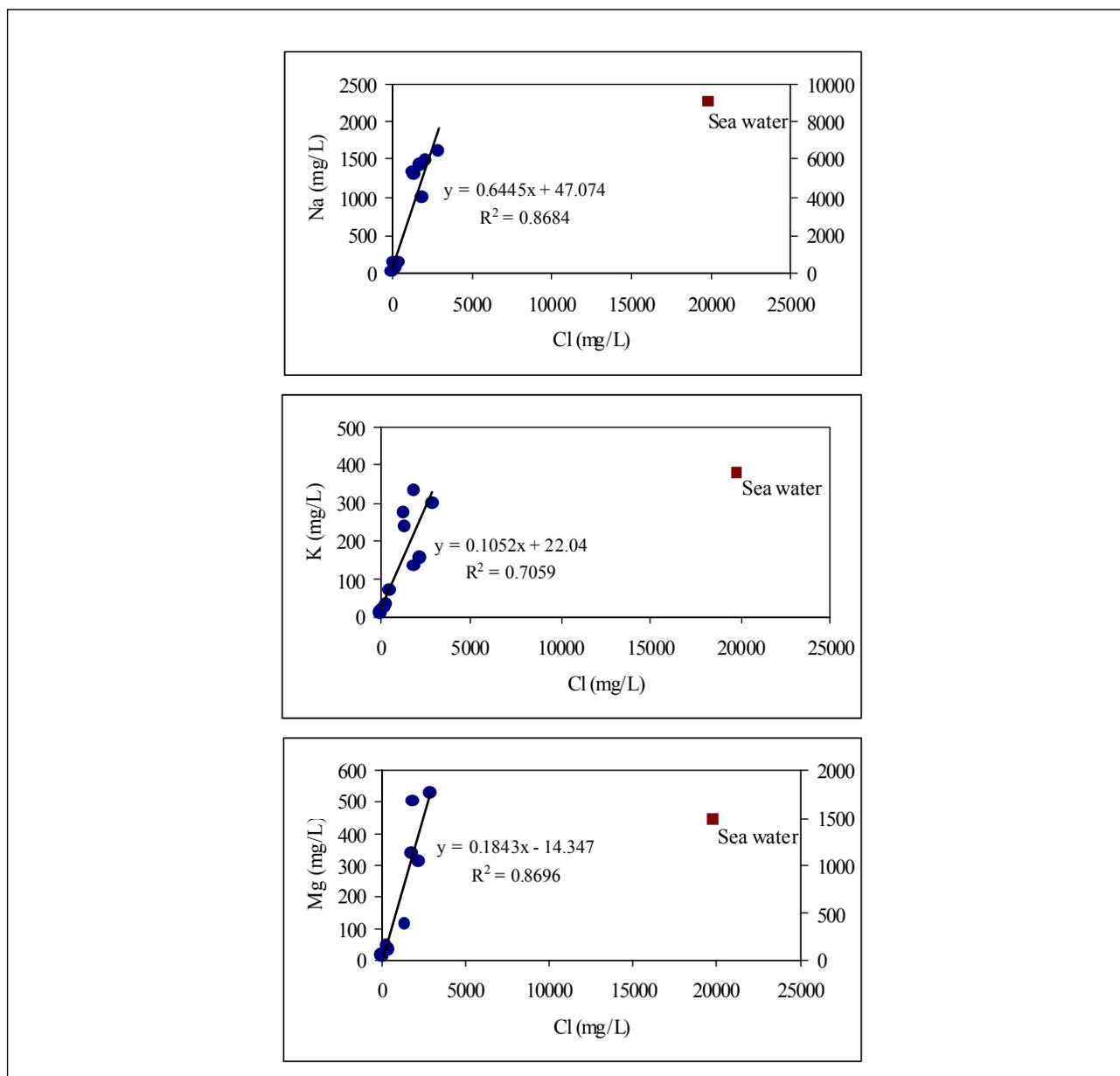


Figure 1. Plot of chlorine concentration vs. the concentrations of Mg, Na and K in coastal and submarine springs.

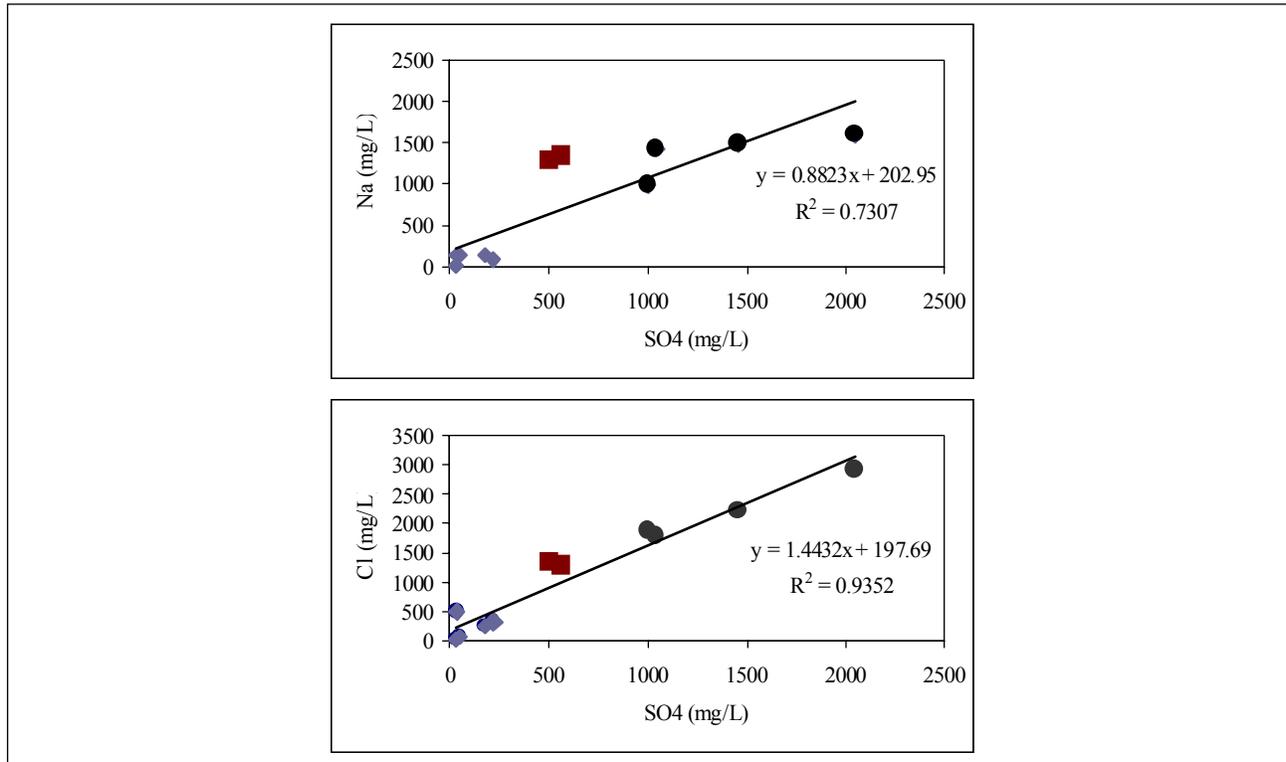


Figure 2. Plot of sulfate concentration vs. the concentrations of Na and Cl in coastal and submarine springs.

Figure 2 shows the variation of sodium and chlorine as a function of sulfate concentration. The plots show a distribution of points into three groups. The first group (◆) with relatively low concentrations in both Na and Cl is relative to coastal freshwaters. The second group (■) with medium concentrations is relative to submarine water of Chekka (north coast). The third group (●) with higher concentrations in sodium and anions is assigned to submarine freshwater in the south. This specific distribution of the points indicates that the characteristics of submarine water in the Chekka region is close to that of coastal groundwater, whereas different hydrogeologic conditions control submarine water in the south. The different geochemistry of submarine springs in the north and the south may also reflect the difference in the hydrogeology of the two regions. This specificity must be considered in the environmental isotope analysis in the next part.

Environmental Isotope Analysis of Submarine and Coastal Freshwater

Samples of rainwater from coastal and highland regions were taken for environmental isotope analysis of deuterium and oxygen-18. The stable isotopic composition of precipitation is explained by applying the Rayleigh model including the progressive condensation of vapor during transport to higher latitudes with lower temperatures. In addition, the local Lebanese meteoric water line provides a baseline for tracing groundwater recharge (Barnes and Allison, 1988; Saad et al., 2005). The position of meteoric waters on this line is controlled by a series of temperature-based mechanisms that drive the rainout process. These include vapor mass trajectories over the country, rising over topographic features, moving to high altitudes and seasonal effects. Each has a characteristic effect on the stable isotopic composition of precipitation (Clark and Fritz, 1997). In the Lebanon and Anti-Lebanon mountains where the average temperatures are lower than the coastal areas, precipitation will be isotopically depleted. This altitude effect is useful to distinguish groundwater recharged at high altitudes from that recharged at low altitude. The Lebanese

Meteoric water line LMWL is defined by the following equation (Saad et al., 2005):

$$\delta^2\text{H} = 7.13 \delta^{18}\text{O} + 15.98$$

To establish the origin of water recharge in the different reservoirs (surface water, groundwater, submarine water) the isotopic composition of deuterium and oxygen-18 was determined and compared to the LMWL (Table 3).

Figure 3 shows the variation of deuterium against oxygen-18 of the submarine water, surface water and groundwater in the north (Chekka) and the south (Tyre). All points lie between the Lebanese Meteoric Water Line (LMWL) and the Global Meteoric Water Line (GMWL). Furthermore, the distribution of points delineates two groups of waters. The first one includes surface water and groundwater with submarine springs in Chekka region that lie near the Lebanese Meteoric water line. The highly karstified region of Chekka induces a fast infiltration of groundwater in its galleries until they submerge as submarine springs in the sea (Shaban et al., 2005). The submarine springs of Chekka, estimated to flow at 10 m³/sec, are related to the Turonian-Cenomanian limestone aquifer. Tritium values of submarine water and groundwater (Table 3) are close to the mean values of rainwater in the region (mean ³H = 3) (Saad et al., 2005). This also indicates that groundwater is constantly renewed from fast rainwater infiltration.

The more enriched isotopic composition of submarine groundwater in the Tyre region could be explained by either a mixing with sea water or a geothermal effect within the aquifer (Figure 4). The mixing possibility is less likely because the isotopic composition of submarine springs is closer to rainwater in this region. On the other hand, a specific hydraulic feature exists in this region, which is a dense fracturing system and conduit extension offshore. Aquifers in the coastal plateau of south Lebanon are primarily in Cenomanian-Turonian strata and the submarine springs are primarily artesian flows that occur in confined aquifers. These aquifers are deep and groundwater reaches great depth before discharging into the sea. Fractures and fault systems result in deep circulation of groundwater and gains in temperature because of the geothermal gradient.

Table 3. Isotopic composition of submarine springs and coastal groundwater.

Site name and locality	$\delta^{18}\text{O}$	$\delta^2\text{H}$	Tritium
1- Surface water Brak Ras el Ain, South	-6.17	-27.87	3.94
2- Surface water El saide, South	-6.17	-27.87	3.5
3- Surface water Kasmiyeh, South	-5.77	-27.68	3.27
4- groundwater 1 Tyre, South	-6.2	-25.1	4.1
5- groundwater 2 Tyre, South	-6.2	-25.1	3.49
6- Submarine Tyre, South	-3.8	-17	1.54
7- Submarine Borgolieh, South	-3.85	-16.83	2.98
8- Submarine Nsariyeh, South	-3.92	-18.02	3.77
9- Submarine Abou Asswad, South	-3.36	-15.33	3.37
10- Seawater Tyre	1.65	11.76	1.53
11- Surface water Chekka	-6.17	-30.879	2.42
12- groundwater Chekka, North	-6.35	-29.76	3.4
13- Submarine Chekka, North	-5.6	-27	1.97
14- Submarine Chekka, North	-5.51	-27.38	2.95
15- Seawater Chekka	1.65	11.76	

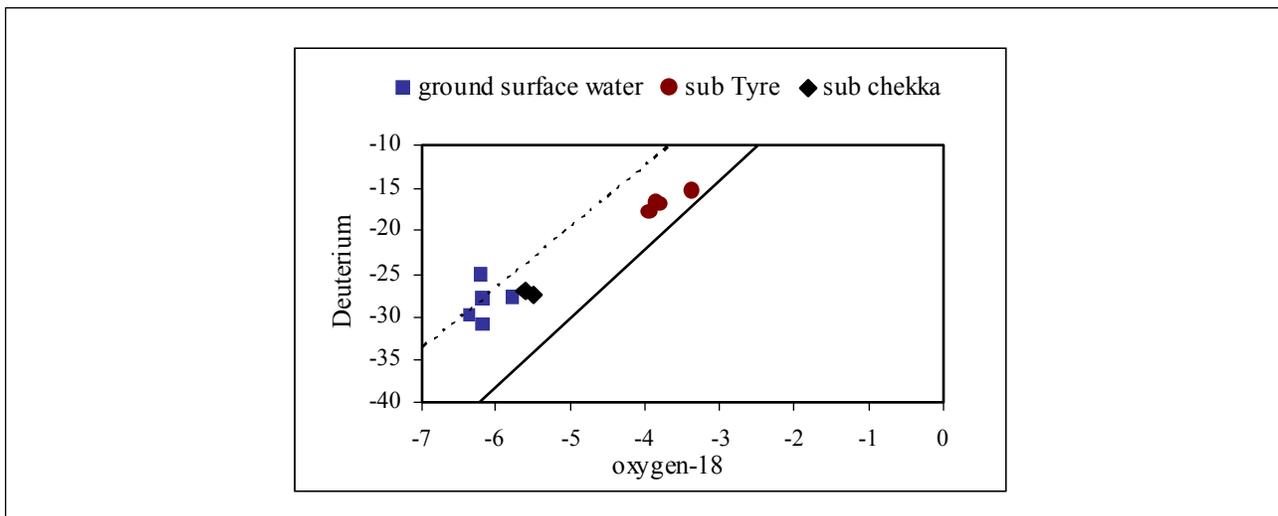
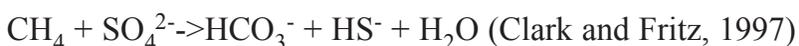


Figure 3. Isotopic composition of surface, groundwater and submarine springs in different regions.

In fact, the temperature of submarine springs in the south is about 42°C which confirms the deep circulation of groundwater before its discharge as submarine spring. Tritium content of submarine water is close to the rainwater values in the south, indicating a short subsurface residence time and a high mixing with young meteoric water (Sturchio et al., 1996).

In the confined aquifer, groundwater passes through a layer of marls and marly limestones of the Upper Cretaceous and Lower Eocene and is charged with gypsum minerals ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) when passing through the marl. This can be seen in Figure 5, which shows that submarine samples in the south (square form), enriched with $\delta^{18}\text{O}$, are the most concentrated in sulfate. The deep circulation of groundwater at high temperature promotes the dissolution of gypsum minerals.

At depth usually a certain amount of sulfate is usually reduced to sulfur via the following equation:



During this complex geochemical transformation of sulfate, fractionation processes affect the stable isotopic compositions of sulfur ($^{34}\text{S}/^{32}\text{S}$) and oxygen ($^{18}\text{O}/^{16}\text{O}$) in the sulfate mineral. The temperature of groundwater prevents most of the isotopic fractionation process to reach isotopic equilibrium and significant isotopic exchange of oxygen between sulfate ions and water (Krause,

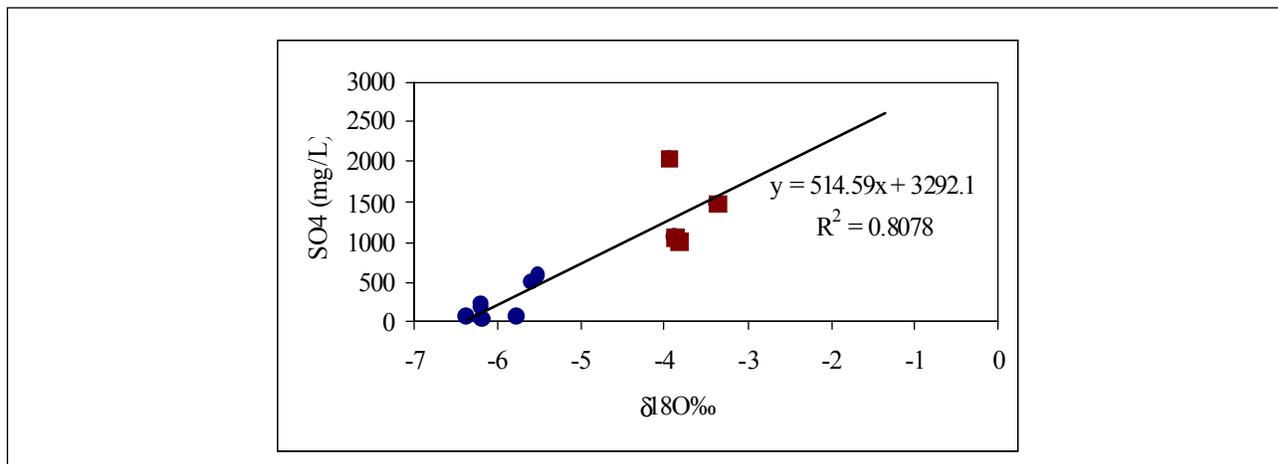


Figure 4. Variation of sulfate against $\delta^{18}\text{O}$ in coastal groundwaters and submarine springs.

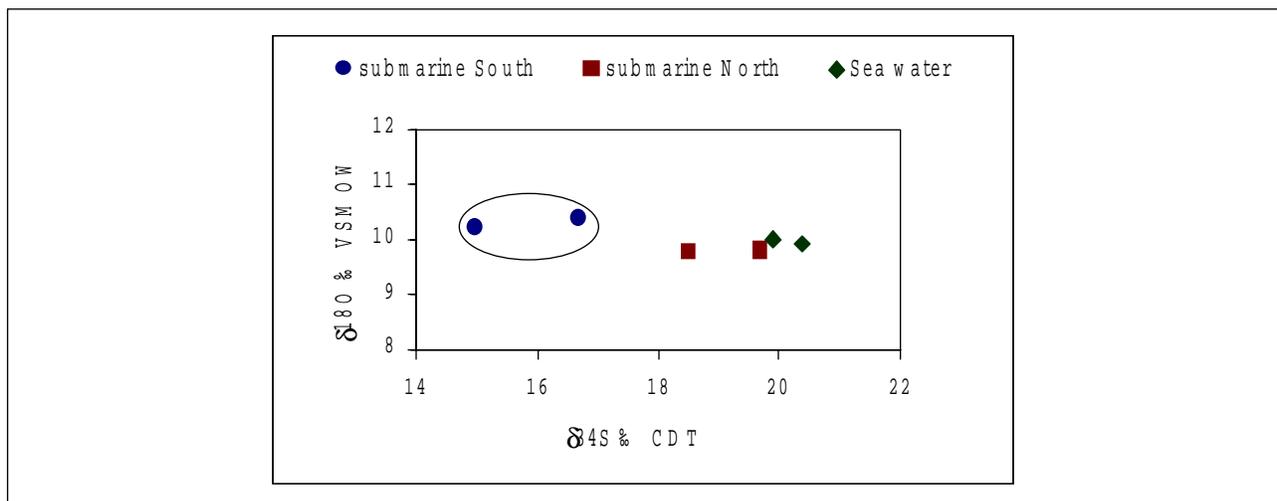


Figure 5. Ranges of variation of $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ in sulfate of submarine springs (north and south regions) and sea water.

1987). In the present study, the isotopic composition of sulfur in dissolved sulfate in sea water is around 20‰ (Figure 5). This value is concordant with recent sea water defined by $\delta^{34}\text{S} = 21\text{‰}$.

Figure 5 does not show important fractionation in $\delta^{18}\text{O}$ of sulfate for all submarine samples. Whereas submarine groundwater in the south has a lighter isotopic composition of $\delta^{34}\text{S}$ that reaches 15‰. This light fractionation of sulfur isotope composition tends to verify the specific hydrogeology in the Tyre region that is associated with the elevated geothermal gradient.

CONCLUSIONS

This study gives an important insight into the geological differences between the northern and the southern coastal regions in Lebanon, by using hydrochemical and isotopic tools. Submarine springs along the southern and northern coasts are characterized by different hydrochemical and isotopic parameters. The different characteristics imply a difference in hydrogeological features in the two regions. The northern coast is affected by a highly karstified areas while the south exhibits long lineaments and a dense fracture system leading to the formation of artesian submarine springs. The elevated temperature of springs in the south associated with isotopic fractionation of sulfur in dissolved sulfate is related to the geothermal gradient at depth.

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