JOURNAL OF MARINE SCIENCES & ENVIRONMENTAL TECHNOLOGIES
Vol. 3, Issue No. 1 (June-2017)

ISSN: 2413-5267

Geochemical Characterization of The Wadies (Al Hash, Al Shaigh, and Rahib), Tobruq-Burdi Area, NE Libya

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التوصيف الجيوكيميائي لوديان (الهش، الشيخ والراهب)، بالمنطقة الواقعة بين طبرق–والبردي، شمال شرق ليبيا

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Abstract

This work was primarily focused on the chemical analysis (major and trace *wt. %*) of the Tobruq - Burdi area in the northeastern part of Libya, on the geological formations ranging in age from Late Cretaceous to Late Miocene at Wadies; Al Hash, Al Shaigh and Al Rahib. The chemical analysis data showed that the Al Faidiyah Formation is richer in SiO_2, TiO_2, $A1_2O_3$, Fe_2O_3, MgO, Na_2O, K_2O, and Cl than the other studied formations. In the Al Faidiyah and Al Jaghboub formations, the Na/Cl ratio is about 1 and Na2O is strongly correlated with Cl (r = 0.98) which indicates their accommodation in the form of halite. The positive correlations between Na_2O and SO_3 (r = 0.71) and between Cl and MgO and K_2O (r = 0.98 and 0.77, respectively) indicate the possibility of other evaporate. However, from the viewpoint of commercial the formations vary in grade from high purity to impure limestone rocks. In addition to calcite, limestone contains other carbonates such as dolomite, and non-carbonate includes quartz, clay, halite, hematite, and glauconite minerals. The possible applications of limestones are; the cement industry, animal feedstuffs, agriculture, construction as aggregates, weighting agents in drilling mud fluids, plastics, and glass. Generally, the studied sediments contain low concentrations of high-field strength element oxides and low concentrations of heavy metal oxides.

***Keywords**: Wadi Al Hash, Wadi Al Shaigh, Wadi Al Rahib, Tobruq, Burdi, NE Libya.*

الملخص

ركز هذا العمل في المقام الأول على التحليل الكيميائي (للأكاسيد بالنسبة المئوية للوزن ٪) من منطقة طبرق وحتى البردي في الجزء الشمالي الشرقي من ليبيا، تتراوح التشكيلات الجيولوجية في العمر من أواخر العصر الطباشيري إلى الميوسين المتأخر في الوادي؛ الهش، الشيخ، الراهب. وأظهرت بيانات التحليل الكيميائي أن تكوين الفايدية غنى في ثاني أكسيد الكبريت و TiO_2 و $A1_2O_3$ و Fe_2O_3 و MgO و Na_2O و K_2O و كل من التشكيلات الأخرى المدروسة. في تكوينات الفايدية والجغبوب، تبلغ نسبة Na/Cl حوالي 1، ويرتبط Na_2O بقوة مع Cl (r = 0.98) التي يشير وجودها على شكل أملاح (هاليت). وتوجد علاقة طردية ما بين Na_2O و SO_3 (r = 0.71) وبين Cl و MgO وK2O (r = 0.98 و0.77 على التوالي) وتدل على وجود تبخر مرة أخرى. ومع ذلك، من وجهة الناحية الاقتصادية التشكيلات تختلف في المواصفات الفيزيائية والمعدنية

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والكيميائية من درجة عالية من النقاء إلى صخور الحجر الجيري الملوثة. بالإضافة إلى الحجر الجيري تحتوي هذه الصخور على كربونات أخرى مثل الدولوميت، وغير كربونات وتشمل الكوارتز والطين، الهاليت، الهيماتيت والمعادن الجلوكونيت. ويمكن إستغلال هذه الصخور في الصناعات التالية: صناعة الإسمنت، الأعلاف الحيوانية، الزراعة، البناء وأهمها الطرق الديكورات والبلاط، الوزن في الحفر لطينة سوائل الحفر، والبلاستيك، والزجاج. وبصفة عامة، تحتوي الرواسب المدروسة على تركيزات منخفضة من أكاسيد المعادن الثقيلة.

الكلمات الدالة: وادي الهش، وادي الشيخ، وادي الراهب، طبرق، بردي، شمال شرق ليبيا.

1. Introduction

The study area is located in the Tobroq - Alburdi area, in the northeastern part of Al Jabal al Akhdar. The area included three studied wadies are indicated by the following coordination's: i) Wadi Al Shaigh Lat. 32°00’17.7” N and Long. 24°08’33.7” E; ii) Wadi Al Hash Lat. 32º 00' 23" N and Long. 24º 07' 32" E; and iii) Wadi Al Rahib Lat. 31° 47’ 56.7” N and Long. 25°03’44.5” E (Figure 1). This area with 2500 km^2 is part of in the northeastern part of Marmarica region, which represents the northern portion of the 1:250,000 geological map of Al Burdia sheet NH 35-1. The exposed stratigraphic sections in the study area consist of sedimentary rocks, ranging in age from Late Cretaceous to Late Miocene includes; Al Majahir, Darnah, Al Abraq, Al Faidiyah and Al Jaghboub formations. The most abundant sedimentary facies are carbonates with lesser amounts of clays. This study presents the first comprehensive study based on major oxides and trace elements in this area.

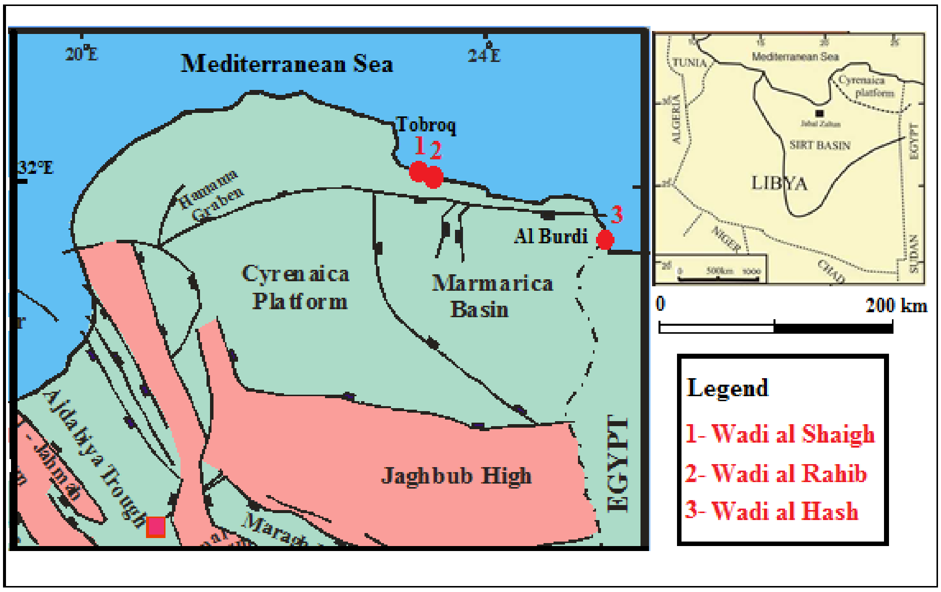

Figure 1. Location map of the study area

2. Geological Settings

Cyrenaica region occupies an area of 150,000 km^2 extending from Benghazi to the Egyptian border and from the coastline in the north to the Hameimat Trough in the south (Figure 2).

ISSN: 2413-5267

The stratigraphy and structure of Cyrenaica differ in several important respects from the rest of onshore Libya. It consists of two pronounced tectonic provinces, the Cyrenaica Platform in the south and the Jabal al Akhdar folded uplift which is called "Al Jabal al Akhdar anticlinorium" in the north, it is separated by the Cyrenaica fault system. However, Al Jaghbub High that forms the southern extension of Cyrenaica Platform is separated from the eastern extension arm of Sirt Basin that is called Al Hameimat (El Hawat and Abdulsamad, 2004). The Cyrenaica Platform was tectonically stable during Mesozoic-Cenozoic time (Hallett, 2002). It yields a thick Palaeozoic succession that was partly folded and locally eroded during the Hercynian Orogeny, and p. Al Jabal al Akhdar and Marmarica exposed rocks ranging in age from Late Cretaceous- Late Miocene. These rocks mainly made of Carbonates (mainly limestone with minor dolostone) with subordinates of clay and gypsum locally. More detailed information in stratigraphy and structure can be obtained from (Hallett, 2002; El-Arnauti and Shelmani, 1985; El-Hawat and Shelmani, 1993; Elwerfalli *et al.*, 2000; El Hawat and Abdulsamad, 2004; and El Amawy *et al.*, 2010).

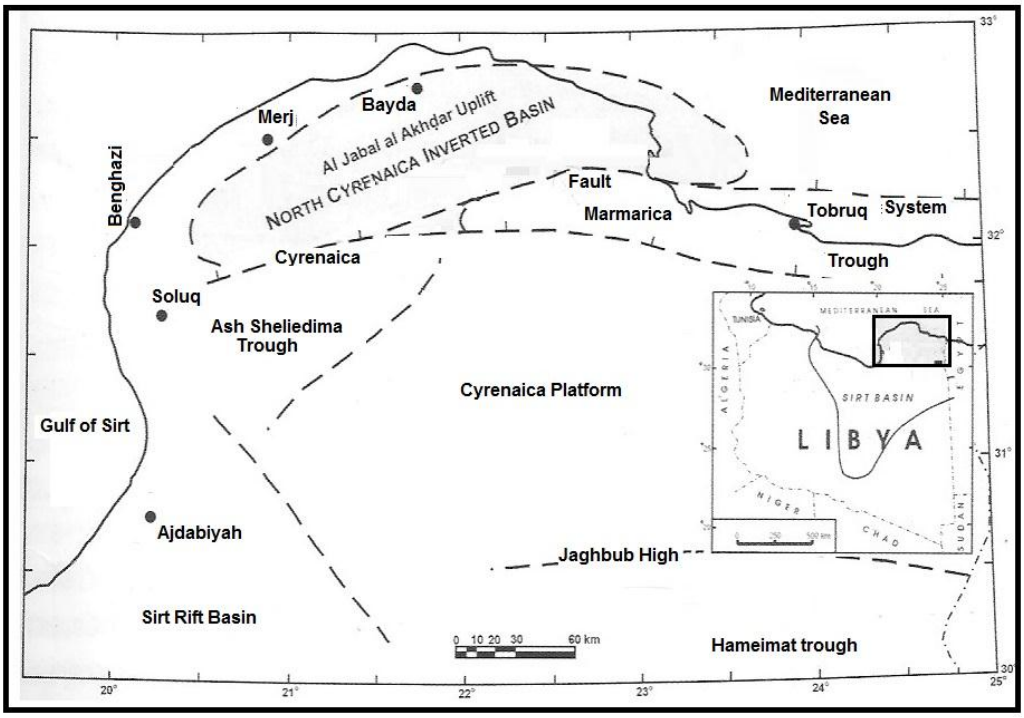

Figure 2. Tectonic map of NE Libya, Cyrenaica (El Hawat and Abulsamad, 2004)

3. Objectives

Determine the chemical composition and the distribution of elements (major and trace elements) for all formations in the study area, in order to identify the percentage of heavy metals in this region as well as classify the grade of limestone purity for the purpose of end-use in industrial fields.

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4. Methodology and Analytical Procedures

XRF analysis was used to determine the major and trace (oxides *wt.* %) of bulk chemical composition for the carbonate and the clay materials of the study area. Eight homogenous fine powder samples were selected from different areas and the result are summarized in (Tables 1 and 2). All these samples were carried out at Metallurgical Research Institute Helwan Laboratory, in Egypt.

Table 1. Chemical analysis data (major and trace oxides in *wt.* %) of the Darnah, Al Abraq and Al Faidiyah formations.

Formation	**Darnah**	**Al Abraq**	**Al Faidiyah**	
Location	**Wadi Al Hash**	**Wadi Rahib**	**Wadi Al Hash**	**Wadi Al Hash**
Sample No.	**SD2**	**R4**	**FS1**	**FS5**
SiO_2	0.17	2.46	10.87	7.24
TiO_2	0.00	0.14	0.38	0.32
Al_2O_3	0.06	1.04	2.88	2.78
Fe_2O_3	0.07	1.25	3.85	2.33
MnO	0.01	0.02	0.03	0.02
MgO	0.59	0.93	8.17	1.88
CaO	54.33	50.05	29.35	43.00
Na_2O	0.17	0.45	1.00	0.83
K_2O	0.22	0.22	0.55	0.63
P_2O_5	0.02	0.02	0.11	0.03
SO_3	0.10	0.08	0.36	0.13
Cl	0.22	0.61	0.95	0.79
LOI	44.00	42.60	41.30	39.90
Total	99.96	99.87	99.80	99.88
F	0.00	0.00	0.08	0.04
Br	0.00	0.00	0.01	0.002
SrO	0.03	0.08	0.03	0.06
Cr_2O_3	0.01	0.00	0.00	0.01
ZnO	0.00	0.01	0.01	0.004
NiO	0.00	0.00	0.01	0.00
CuO	0.00	0.01	0.00	0.00
Au	0.00	0.02	0.00	0.00
ZrO_2	0.00	0.00	0.03	0.02
Nb_2O_5	0.00	0.00	0.002	0.002
Y_2O_3	0.00	0.002	0.00	0.00

Table 2. Chemical analysis data (major and trace oxides in *wt.*%) of the Al Faidiyah and Al Jaghboub formations.

Formation	Al Faidiyah		Al Jaghboub	
Location	**Wadi Al Shaigh**	**Wadi Rahib**	**Wadi Al Shaigh**	**Wadi Al Hash**
Sample No.	**SG11**	**R10**	**SG15**	**HG4**
SiO_2	0.52	0.34	0.59	0.87
TiO_2	0.03	0.02	0.00	0.00
Al_2O_3	0.18	0.11	0.07	0.12
Fe_2O_3	0.23	0.16	0.21	0.16
MnO	0.01	0.00	0.00	0.00
MgO	0.42	0.25	0.47	0.56
CaO	54.76	55.17	55.34	54.40
Na_2O	0.02	0.06	0.14	0.03
K_2O	0.04	0.19	0.02	0.21
P_2O_5	0.01	0.19	0.17	0.05
SO_3	0.05	0.04	0.21	0.06
Cl	0.02	0.04	0.14	0.03
LOI	43.60	43.40	42.60	43.50
Total	99.89	99.97	99.96	99.99
F	0.00	0.00	0.00	0.00
Br	0.00	0.00	0.00	0.00
SrO	0.07	0.02	0.04	0.03
Cr_2O_3	0.02	0.01	0.01	0.00
ZnO	0.003	0.003	0.004	0.002
NiO	0.00	0.00	0.000	0.000
CuO	0.00	0.00	0.00	0.00
Au	0.00	0.00	0.00	0.00
ZrO_2	0.00	0.00	0.00	0.00
Nb_2O_5	0.00	0.00	0.00	0.00
Y_2O_3	0.001	0.00	0.000	0.000

5. Geochemistry

From a commercial viewpoint, the main impurities such as dolomite, quartz and clay minerals throughout the limestone deposits were found in some formations in variable amounts. Their distributions are very important factors in grade control for production of very high or impure quality limestone. The formation in the study area varies from high purity to impure limestone (Tables 1 and 2). Harries (1979) classified the limestone's according to purity as shown in (Table 3).

Table 3. Classification of limestone in the formation in the study area

Categories of limestone	Composition % $CaCO_3$
very high purity	>98.5
High purity	97-98.5
Medium purity	93.5- 97
Low purity	85-93.5
Impurity	<85.0

5.1 Statistical Treatment

The statistical treatment of the obtained data involves correlation matrix (Table 4). The graphical presentation of the correlation coefficients among the analyzed major oxides points to the intimate coherence among then, except for lime (Figure 3). Silica, alumina, iron oxide, titanium dioxide, potash, soda and chlorine.

Table 4. Correlation matrix of the studied samples

Oxides	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	SO_3	Cl	SrO	Cr_2O_3	ZnO
SiO_2	1.00														
TiO_2	0.98	1.00													
Al_2O_3	0.97	0.99	1.00												
Fe_2O_3	0.99	0.98	0.96	1.00											
MnO	0.85	0.89	0.86	0.89	1.00										
MgO	0.90	0.82	0.78	0.91	0.77	1.00									
CaO	-0.98	-0.95	-0.92	-0.99	-0.87	-0.96	1.00								
Na_2O	0.97	0.98	0.98	0.97	0.90	0.82	-0.94	1.00							
K_2O	0.88	0.90	0.92	0.85	0.73	0.67	-0.83	0.89	1.00						
P_2O_5	-0.02	-0.11	-0.13	-0.03	-0.38	0.11	0.01	-0.09	-0.16	1.00					
SO_3	0.76	0.66	0.63	0.77	0.58	0.11	-0.81	0.71	0.47	0.30	1.00				
Cl	0.92	0.95	0.95	0.94	0.92	0.77	-0.90	0.98	0.85	-0.17	0.66	1.00			
SrO	0.03	0.18	0.18	0.09	0.37	-0.19	0.63	0.14	-0.04	-0.62	-0.23	0.24	1.00		
Cr_2O_3	-0.40	-0.35	-0.34	-0.43	-0.31	-0.45	0.44	-0.41	-0.39	-0.06	-0.36	-0.47	0.18	1.00	
ZnO	0.64	0.67	0.61	0.72	0.72	0.64	-0.68	0.67	0.37	0.05	0.55	0.73	0.38	-0.56	1.00

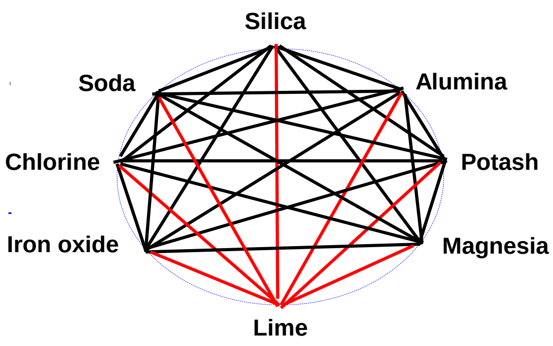

Figure 3. Correlations among the major oxides in the studied samples (intensity of lines corresponds to the strength of the correlation coefficient (< 0.4 to > 0.8) (red line means inverse relation).

ISSN: 2413-5267

5.2 Major Oxides

The major elements are generally considered somewhat mobile during weathering, transportation, and post-depositional processes (McLennan *et al.*, 1993). The geochemistry of the major oxides is essentially controlled by the mineral composition of the sediments which, in turn, roles the mutual abundance and distribution of the trace elements. Tables (1 and 2) showed that Al Faidiyah Formation is richer in SiO_2, TiO_2, $A1_2O_3$, Fe_2O_3, MgO, Na_2O, K_2O and Cl than the other studied formations. Silica and alumina are positively correlated in the studied samples (r = 0.97; Figure 4). Preda and Cox (2005) found that quartz and shell-rich sediments tend to have smaller amounts of Al in the shallow marine sediments in the Gulf of Carpentaria, Northern Australia.

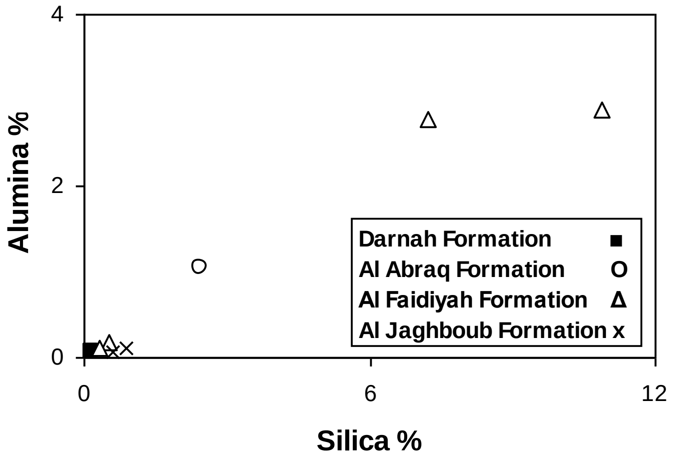

Figure. 4 Relationship between silica and alumina in the study area

Aluminum concentration is a reasonably good measure of detrital flux. The studied limestone samples (except Al Faidiyah Formation) show lower concentrations of alumina (Al_2O_3) than the siliciclastic contaminated carbonates (Al_2O_3 concentration of 1.59 %, as mentioned in Veizer, 1983).

Na_2O and K_2O are strongly correlated positively with Al_2O_3 (r = 0.98 and 0.92, for Figures (5 and 6) respectively) suggesting that in agreement with Zhang (2004) and Nagarajan *et al.* (2007), that these elements are almost entirely associated with detrital admixture.

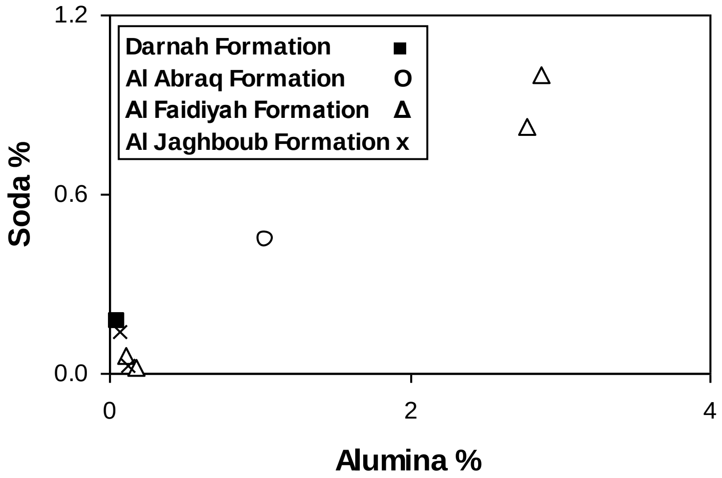

Figure 5. Relationship between alumina and soda in the study area

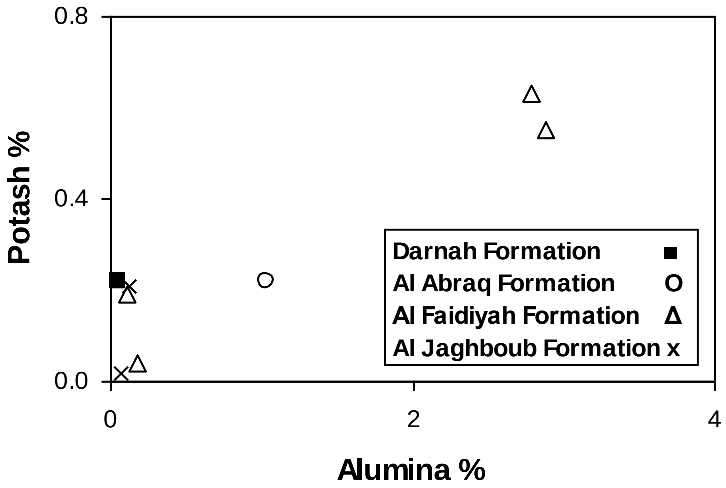

Figure 6. Relationship between alumina and potash in the study area

The K_2O/Al_2O_3 ratio is important in sedimentary rocks in understanding the source of aluminum and its distribution between clay and feldspars minerals (Katongo *et al.*, 2004). The K_2O/Al_2O_3 ratios for clay minerals and feldspars are different (0.0 to 0.3, 0.3 to 0.9, respectively (Cox *et al.*, 1995). In the studied samples, the K_2O/Al_2O_3 ratio ranges from 0.19 to 3.6, indicating that clay minerals have the major role in the distribution of aluminum in the study area. The distribution of CaO is clearly opposite to that of SiO_2 (r = -0.98, for Figure 7). Silica is likely to represent mineral components especially quartz and clay minerals, while lime may be mainly derived from carbonate mud and shell fragments.

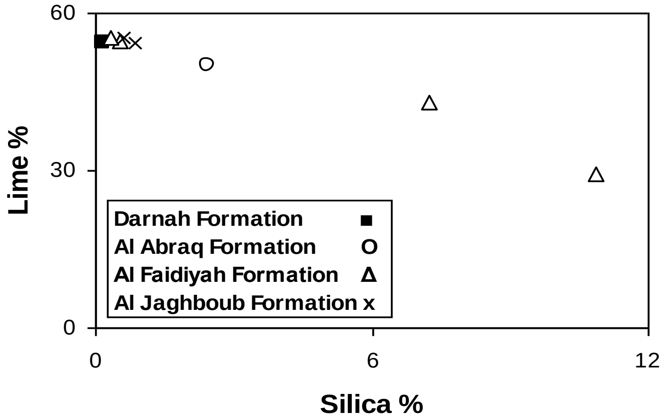

Figure 7. Relationship between silica and lime in the study area

In the present study, CaO is negatively with MgO (r = -0.96, for Figure 8). The relationship means that calcite is the sole carrier of MgO. The positive correlation between MgO and Al_2O_3 (r = 0.78, for Figure 9) suggests, in agreement with Zhang (2004) that magnesia is bound to alumino-silicate minerals and associated phases and, therefore, it has been concentrated during weathering processes. The microscopic investigation showed that glauconite is the only detected clay mineral in the studied formations (Figure 10 a and b).

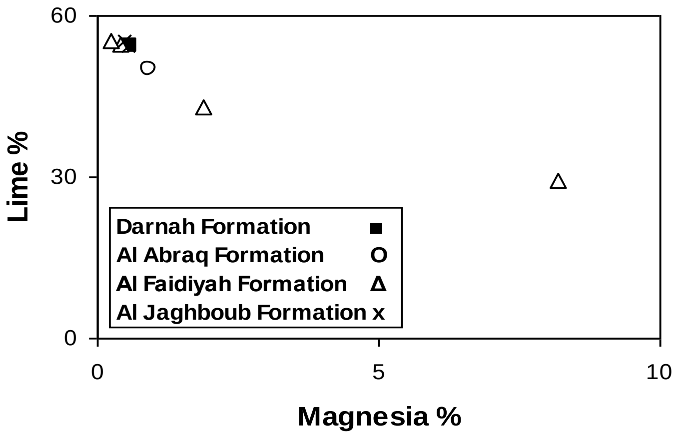

Figure 8. Relationship between magnesia and lime in the study area

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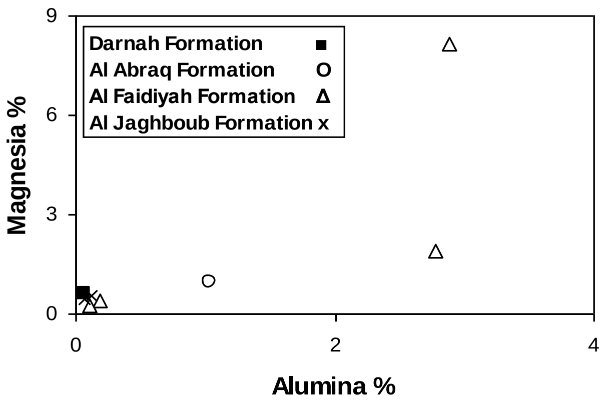

Figure 9. Relationship between alumina and magnesia in the study area

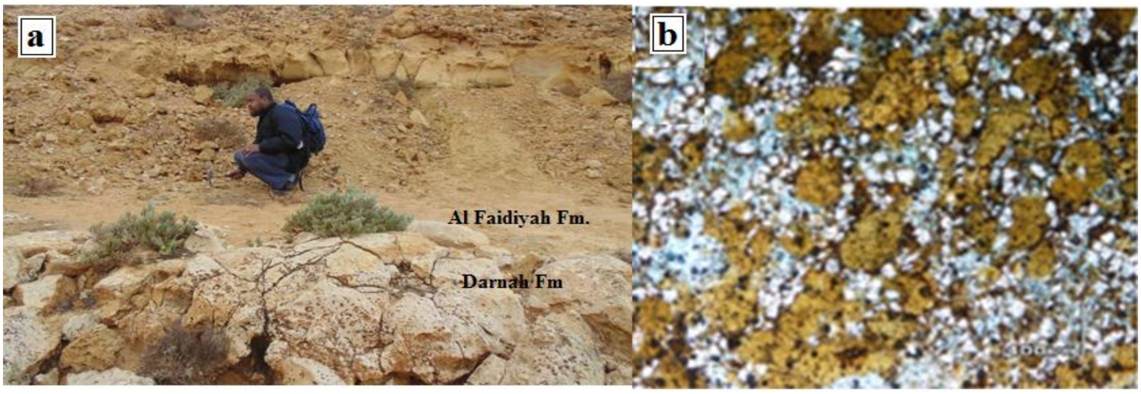

Figure 10. Photomicrograph of; ***a)*** unconformity surface between Middle Eocene Darnah Formation and Late Oligocene Al Faidiyah Formation at Wadi Al Hash area; ***b)*** glauconitic dolostone unit of the Al Faidiyah Formation, showed common glauconites, Quartz, and dolomite grains in grainstone texture

Titanium is relatively immobile compared to other elements during various sedimentary processes and may strongly represent the source rocks (McLennan *et al.*, 1993). TiO_2 is only detected in Al Abraq and Al Faidiyah formations. TiO_2 is strongly correlated to Al_2O_3 and Fe_2O_3 (r = 0.99 and 0.98 for Figures (11 and 12) respectively) suggesting, in agreement with (Basu and Molinaroli, 1989; Condie *et al.*, 1992; Carranza-Edwards *et al.*, 2001; and Carranza-Edwards *et al.* 2009) that Ti is contained in both iron minerals and alumino-silicates. These correlations may be a result of sorting under control of the depositional environments (Abu El-Ella, 2006). Chemically, the main result of the alteration of ilmenite mineral is an increase in (formation of co-existing different phases) TiO_2, Fe_2O_3 and a decrease in FeO. When the ilmenite is oxidized at a certain temperature the structure breaks down into rutile/anatase and hematite. With increasing degrees of alteration ilmenite disappears and the ratio of TiO_2:Fe_2O_3 increases until the altered grain becomes rutile/anatase (El Ebaidi, 2015).

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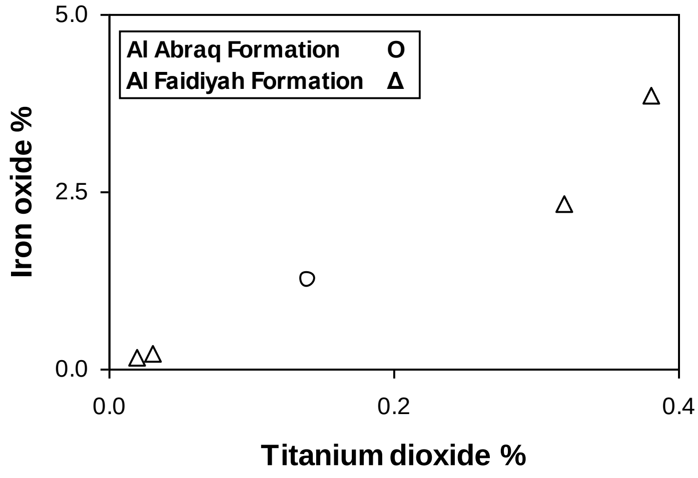

Figure 11. Relationship between titanium dioxide and iron oxide in the study area

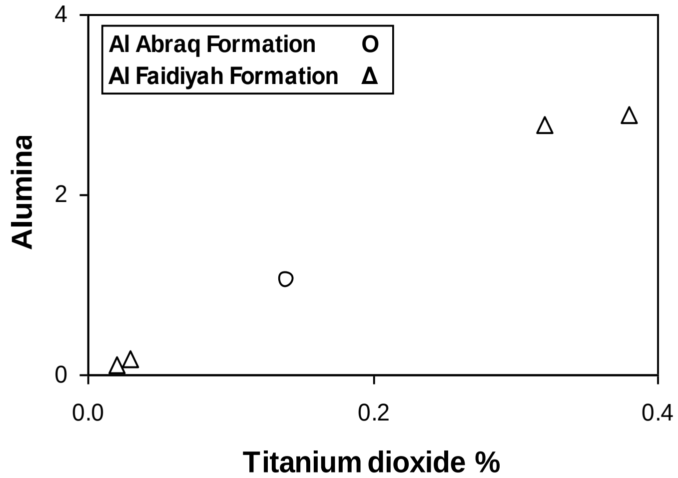

Figure 12. Relationship between titanium dioxide and alumina in the study area

Many authors (e.g. Al Shariani, 2006; and Shaltami, 2012) suggest that Na present in the original carbonate sediments can be modified greatly during diagenesis. In the Al Faidiyah and Al Jaghboub formations, the Na/Cl ratio is about 1 and Na_2O is strongly correlated with Cl (r = 0.98, for Figure 13) which indicate their accommodation in the form of halite. The writer believes that the positive correlations between Na_2O and SO_3 (r = 0.71, for Figure 14) and between Cl and both MgO and K_2O (r = 0.98 and 0.77, for Figures 15 and 16) indicate the possibility of other evaporate salts (e.g. thenardite, carnallite, and sylvite).

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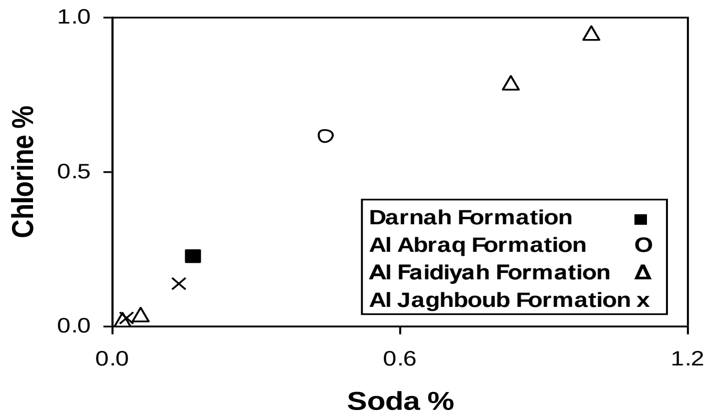

Figure 13. Relationship between soda and chlorine in the study area

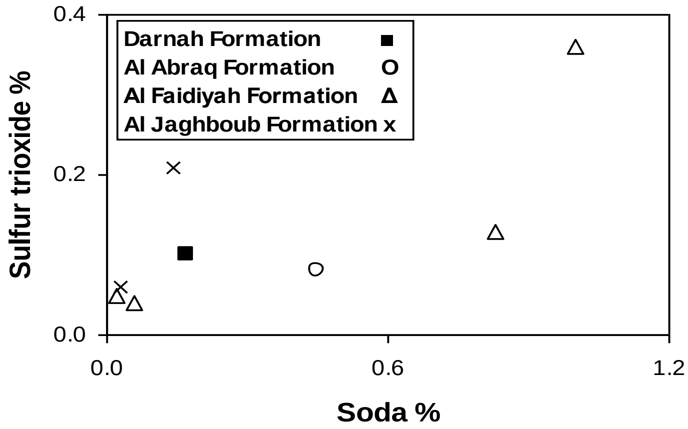

Figure 14. Relationship between soda and sulfur trioxide in the study area

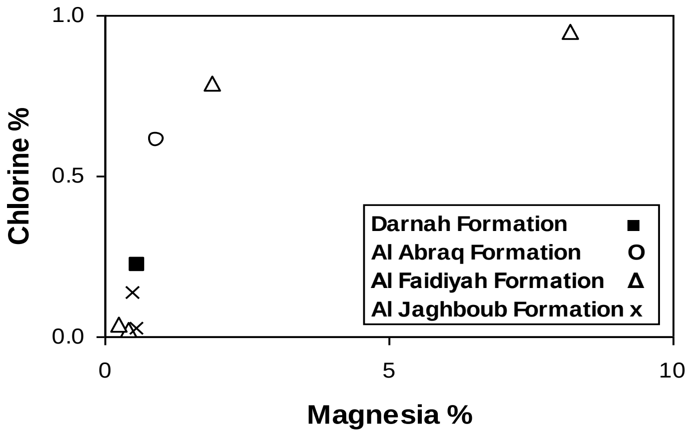

Figure 15. Relationship between magnesia and chlorine in the study area

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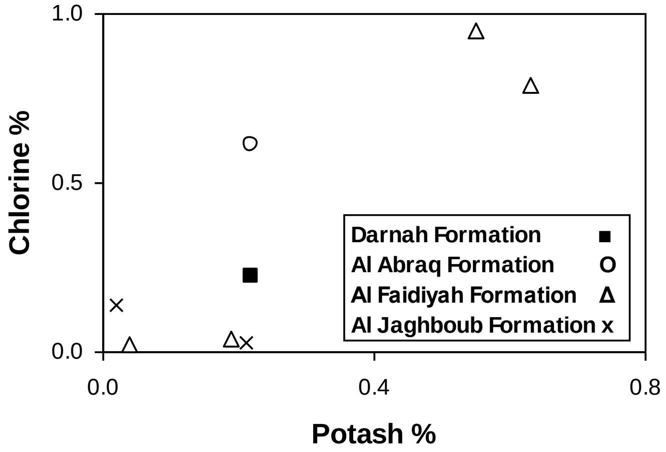

Figure 16. Relationship between potash and chlorine in the study area

5.3. Trace Elements

The study of trace elements have become a vital part in modern petrology and more capable for discrimination between petrological processes than the major elements. The behavior of trace elements during sedimentary processes is complex due to many factors including weathering, physical sorting, adsorption, provenance, digenesis, and metamorphism (e.g., Nebsitt *et al*., 1980; Wronkiewicz and Condie, 1987).

5.4. Low Field Strength Elements (LFSE)

They are large cations of small charge and tend to be compatible with major elements. The low ionic potential (ratio of charge to ionic radius) makes these elements relatively soluble in aqueous solution. Because of their solubility, they are quite mobile during metamorphism and weathering (White, 2001). LFSE include four elements namely, Rb, Cs, Ba and Sr. The Sr content in carbonate minerals may be used as an important chemical characteristic to identify their genesis (Thomson *et al.*, 2004). SrO is positively correlated with CaO (r = 0.63, for Figure 17) suggesting that Sr is contained in calcite. The SrO/CaO ratio ranges from 3.63×10^{-4} to 15.98×10^{-4}. These ratios coincide with the strontium calcitic carbonates (Reitz and de Lange, 2005).

5.5. Heavy Metals

Heavy metals are stable metals or metalloids and cannot be degraded or destroyed. Therefore, they tend to accumulate in soils and sediments. However, anthropogenic activities have drastically altered the biochemical and geochemical cycles and the balance of some heavy metals. The analyzed heavy metal oxides in the studied samples are Cr_2O_3, ZnO, NiO and CuO. Generally, the studied sediments contain low concentrations of heavy metal oxides.

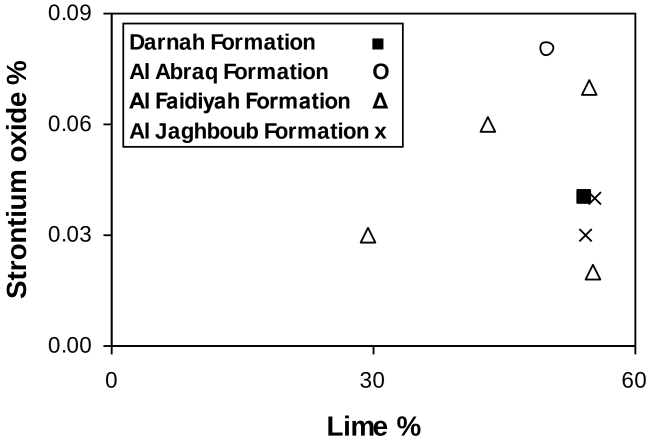

Figure 17. Relationship between lime and strontium oxide in the study area

5.6. High Field Strength Elements (HFSE)

They are highly charged cations and often have appropriate size for many cation sites in minerals. Their charge is too high and requires one or more coupled substitution to maintain charge balance; this is generally energetically unfavorable (White, 2001). Thus HFSE are incompatible elements as they have high ionic potential, they are insoluble and tend to be very immobile during weathering and metamorphism. In the studied samples, two high field strength element oxides, namely, ZrO_2 and Nb_2O_5 are analyzed. Generally, the studied sediments contain low concentrations of high field strength element oxides.

6. Conclusion

The present work describes the geochemistry of Darnah, Al Abraq, Al Faidiyah and Al Jaghboub formations at Wadi Al Hash, Wadi Rahib and Wadi Al Shaigh, Al Jabal Al Akhdar, NE Libya. Analysis of major and trace oxides was done by X-ray fluorescence (XRF) technique. The graphical presentation of the correlation coefficients among the analyzed major oxides points to the intimate coherence among then, except for lime. Silica, alumina, iron oxide, titanium dioxide, potash, soda, chlorine and magnesia are most probably accommodated within terrestrial admixture. The chemical analysis data show that the Al Faidiyah Formation is richer in SiO_2, TiO_2, $A1_2O_3$, Fe_2O_3, MgO, Na_2O, K_2O and Cl than the other studied formations. Silica and alumina are positively correlated (r = 0.97). This reflects the occurrence of silica in both silicate and free silica modes. The K_2O/Al_2O_3 ratio ranges from 0.19 to 3.6, indicating that clay minerals have a major role in the distribution of aluminum in the study area. In the Al Faidiyah and Al Jaghboub formations, the Na/Cl ratio is about 1 and Na_2O is strongly correlated with Cl (r = 0.98) which indicate their accommodation in the form of halite. The positive correlations between Na_2O and SO_3 (r = 0.71) and between Cl and both MgO and K_2O (r = 0.98 and 0.77, respectively) indicate the

ISSN: 2413-5267

possibility of other evaporite salts. SrO is positively correlated with CaO (r = 0.63) suggesting that Sr is contained in calcite. The studied formations contain low concentrations of Cr_2O_3, ZnO, NiO, CuO, ZrO_2 and Nb_2O_5. Generally, the studied sediments contain low concentrations of high field strength element oxides and contain low concentrations of heavy metal oxides.The formations are vary from high purity to impure limestones. In addition to calcite, limestone in the study area contains other carbonates such as dolomite, and non-carbonate included quartz, clay, halite, hematite and glauconite minerals. The possible applications of limestones are; cement industry, animal feedstuffs, agricultures, construction as aggregates, weighting agent in drilling mud fluids, plastics, and glass.

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