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Geology and mineralogy of the radioactive Quaternary sediments of the North Fayium depression, Western Desert, Egypt

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ABSTRACT

Quaternary sediments of the North Fayium depression, are mainly represented by residual soil and calcrete, and have been found to be anomalously radioactive. These sediments are deposited on the irregular surface of the carbonaceous shale of the middle member of the Oligocene Qatrani Formation in the northern part of the Fayium depression. A mineralogical investigation involving a preliminary microscopic examination and detailed Environmental Scanning Electron Microscope (ESEM) has revealed the presence of several heavy accessory minerals. These include zircon, apatite, ilmenite, rutile and garnet as persistent detrital minerals. In addition, a highly suspected secondary uranium mineral has been identified between some mica flakes.

While uranium in the bed rock does not exceed 3.7 ppm, its assay in the residual soil varies from 20 to 200 ppm with an average of 94 ppm, whereas the analyzed two samples of calcrete assay 85 and 122 ppm. Trace element analysis in the studied rock types indicates enrichment in Zn, Zr, Y, Sr and V.

Keywords: radioactive Quaternary sediments, residual soil, calcrete, zircon, apatite, Western Desert, Egypt

1. INTRODUCTION

Uranium occurrence in the Quaternary surficial sediments of the North Fayium depression (residual soil and calcrete) has only recently been discovered and classified. The occurrence is typically young and has been developed when uranium was extracted or precipitated from shallow ground or surface waters. On the surface of bench scarps that form the Fayium depression, the Quaternary sediments which represent the host rock of the radioactive anomalies are indeed arid land deposits (residual soil and calcrete) that have been laid upon the eroded surface of the carbonaceous shale of the middle member of the Oligocene Qatrani Formation.

Carbonaceous sediments denote rocks that contain original organic tissues and/or their decomposition products and their fluid derivatives and extracts have been deposited under anaerobic conditions (PETTIJOHN, 1975). The carbonaceous shale of the Fayium depression has been the point of interest of many studies from ZEID & MONEM (1957), CAMERON (1967), MOSTAFA (1972), MORSY (1978), EL SHAZLY et al. (1974) to HAROUN (2001) and more recently to MOUSA & HAROUN (2005).

The arid climate of the Fayium depression (HAROUN, 2001) has helped in the formation of the residual soil because this climate accelerates the weathering processes on the Qatrani Formation involving sandstone, clays and limestone beds. In addition, the arid conditions have also facilitated formation of the duricrust sediments (calcrete) associated with the residual soil.

The calcrete sediments occur in general in the form of a thin crust of sand cemented by calcite. Morphologically, this crust is cavernous, spongy, semi friable and pale white in colour, besides being characterized by a highly irregular surface. CARLISLE (1983) mentioned that deposition of calcrete results from common ion precipitation when Ca/Mg sulfate or chloride brines are enriched in salt lakes or in local highly evaporative domains. In the study area, calcrete is of a non pedogenic type and has resulted by lateral transport of soluble ions towards favourable sites of deposition. This also makes it a favourable site for uranium concentration from the different poor source areas into the relatively compact duricrust body, (EL-SAYED et al., 2000).

The present work is mainly concerned with both the geological and mineralogical characteristics of the Quaternary sediments of the North Fayium depression that have been deposited on the irregular surface of the carbonaceous shale. Besides the necessary field geological studies, mineralogical investigation of some mineralized samples has involved proper mineral separation and ESEM analysis of separated pure minerals. In addition, the uranium occurrence in these sediments would be identified as well as the general genetic aspects of this mineralization. This involved the study of detrital heavy accessory minerals as another source for uranium in carbonaceous shale.

2. GEOLOGY OF THE EL FAYIUM DEPRESSION

The study area is located at 30º 35′ long E and 29º 37′ lat., and is considered as part of the Fayium depression the general geology of which is illustrated in Figure 1. It consists mainly of sedimentary rocks of upper Eocene, Oligocene, Early Miocene and Quaternary ages. The oldest rocks exposed in the study area belong to the upper Eocene and mainly include the Qasr El Sagha Formation which covers the southern part of the study area (Fig. 1). This formation is composed of sand with some gypsiferous clay, and interbedded with fossiliferous limestone in the middle part of the section. The subsequent Oligocene rocks are represented by the Qatrani Formation which covers the middle part of the study area and is mainly composed of a succession of variegated sands and sandstones with alternating beds of clays, shale and limestone. Carbonaceous shale occurs in the form of lenticular masses within the sand and sandstone beds of the Qatrani Formation.

2.1. Bed rock geology

The study area is considered as a locally closed swamped area and is stratigraphically composed from the base upwards of fine-grained sandstone, silt, sandy clay and clay. This sequence is topped by a carbonaceous shale bed that occurs in the form of lenses of variable thickness ranging from 2 to 7 m, and is characterized by lateral transition into green shale and clay.

The sandstone bed covers a great part of the study area, and extends to the south and south west, while the radioactive carbonaceous bed is confined to certain parts of the upper red fine grained sandstone (Fig. 2). The sandstone bed is underlain by siltstone and limestone beds of fluvio-marine origin. PEJATOVIC (1968) stated that this red sandstone sequence was deposited under arid conditions where lithogenesis occurred under conditions where evaporation was more rapid than water percolation. This sandstone is enriched in carbonate and sulphate minerals including gypsum.

The carbonaceous shale covers an area of about 35 km^2 in the northeastern part of the study area and forms a locally enclosed swamp area (HAROUN, 2001; MOUSA & HAROUN, 2006). Lithologically, the carbonaceous shale bed was deposited within the upper member of the Qatrani Formation of Oligocene age. As shown in Figure 3, the carbonaceous shale bed is occasionally recorded within the sequence composed of sandy clay, silt, clay and fine grained sandstone, which represents flood plain fluorine sediment (REINECK & SINGH, 1973). The radioactive shale is purple, black, brown, ferruginous, highly enriched in gypsum, and 5–7 m thick.

The depositional environment of the radioactive carbonaceous shale must have involved a stagnant small basin, characterized by the following criteria: 1 – a highly reduc-

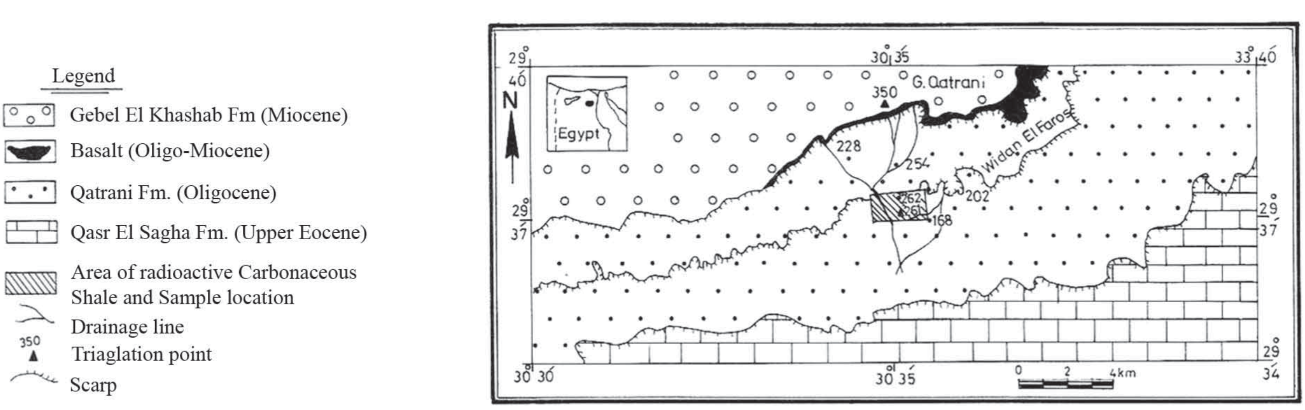

Figure 1: Geologic map of Fayium Depression.

ing environment enriched in anaerobic bacteria, organic matter, pyrite and iron content; 2 – a low carbonate content; 3 – an unusual concentration of some heavy metals including V, Zn, Cu and Cr; 4 – absence of currents (PETTIJOHN, 1975).

2.2. Geology of the radioactive Quaternary sediments

The Quaternary deposits which are represented in the study area by residual soil and calcrete, cover the middle part of the study area and extend southward for about 2km (Fig. 2). These deposits cut the Oligocene red sandstone sediments of the Qatrani Formation at a topographic elevation of 194 m a.s.l. and are about 2 m thick. Towards the southeast, the Quaternary deposits are at a topographic elevation of 168 m (Fig. 2).

2.2.1. Residual soil

In the study area, the residual soil is associated with a duricrust (locally present in the central part of the deposit) and occupies an area of about 4 km^2 in the north eastern part of the mapped area (Fig. 2). It is bounded to the south by bedrock composed of fine grained reddish silty sandstone that belongs to the upper part of the Qatrani Formation.

The residual soil is indeed the product of in situ weathering of some parent rocks that mainly included red sandstone, clay, limestone, gravelly sandstone and basalt.

The surficial solutions involving meteoric water, surface ground waters as well as hydrothermal solutions have laterally moved across the drainage patterns in the investigated area. These drainage patterns dissect the investigated area and extend northerly into low lying areas (geomorphologically local basin) where the residual soils have been deposited. The drainage lines have thus acted as channels through which the surficial solutions were laterally transported. In the local basin, chemical weathering must have been active (oxidation, reduction and ion exchanges) during formation of the residual soils that cover the carbonaceous shale bed. The residual soils are friable and brownish black in colour and are actually rich in roots, rock debris and plant remains.

2.2.2. Duricrust (calcrete)

The duricrust is characterized by a hard, roughly horizontal and highly irregular surface. It is the product of precipitation of different dissolved or airborne salts and other minerals within the existing soils, unconsolidated sediments as well as in the decomposed or permeable rocks. The calcrete crust is actually the major type of duricrust present in the study area and is considered as a crustation that has been precipitated in the pore space and on the eroded surface of the lower member of the Qatrani sandstone. Generally, the calcrete crust deposits consist of abundant soil, fine grained alluvium, angular sand grains and angular clay fragments, soft sediments or weathered bed rock variably cemented and replaced by authigenic carbonate. Many compositional varieties of the calcrete crust exist, and their textures range from banded in the lower part to nodular or cavernous in the upper part of the calcrete crustation. The transition from calcrete to the underlying pelitic detritus (the so-called red sandstone) is general.

Formation of the calcrete deposits in the study area probably occurred as follows: 1 – Leaching of the carbonate and fine grained sediments from the limestone and clays of the country rock by ground and/or surface water. 2 – Oscillating ground water levels resulted in redeposition or precipitation of the suspended micro-matter, and minerals at different levels within the alluvium and unconsolidated sediments present in the study area.

3. MINERALOGICAL AND GEOCHEMICAL INVESTIGATION

Radiometric studies of fossil trees which grew in the residual soil of the North Fayium scarp indicated the presence of anomalous uranium values (HAROUN & RASLAN, 2007), which supports the radioactivity of the residual soils and calcrete. Uranium from the trees attained 35 ppm while the background of the residual clay and soil in the area does not exceed 3.5 ppm. MOHAMED (1965) concluded that Oligocene rocks of the Qatrani Formation, which constitute the north Fayium succession did not show any uranium anom-

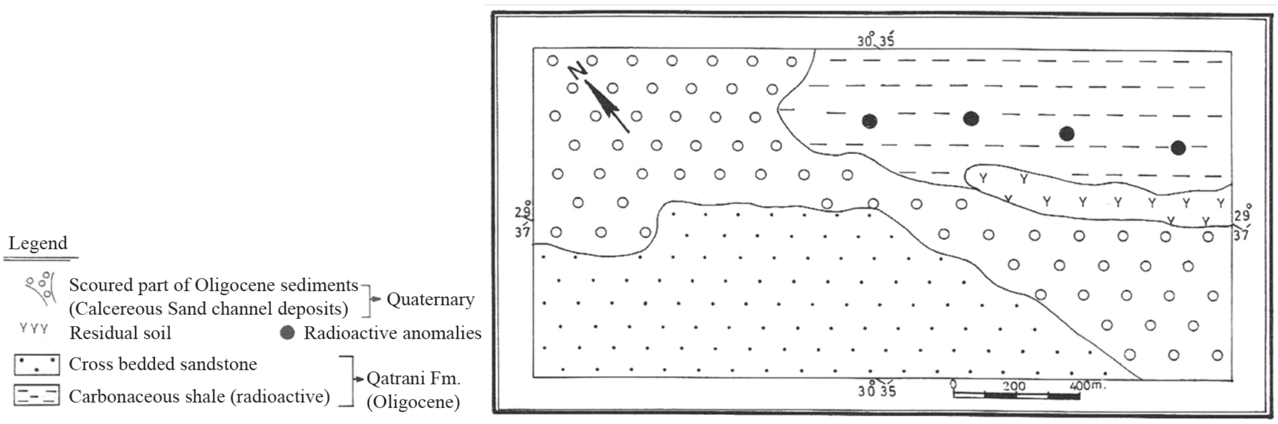

Figure 2: Geologic map of detailed study area (rectangular area in Fig. 1).

aly. Therefore, it was necessary to study both the mineralogy and the radioactivity of both the residual soil and calcrete sediments of the Quaternary rocks in this area.

3.1. Sampling and analytical techniques

In order to determine the minerals responsible for the high radioactivity in the Quaternary deposits of the North Fayium depression, two bulk composite samples representing the residual soil and calcrete were prepared and subjected to various mineral separation techniques. These included disintegration (crushing and grinding), followed by heavy liquid separation using bromoform (sp. gr. 2.85 gm/cm^3), to estimate and identify the heavy mineral content. Monomineralic fractions were prepared from the heavy fraction for preliminary mineralogical investigation and identification using the binocular microscope followed, by detailed examinations using environmental scanning electron microscope (ESEM) techniques. For this purpose, a Philips Model XL 30 supported by an energy dispersive X- ray unit (EDAX) was used.

Seven rock samples from the residual soil and two samples from the calcrete sediments in the study area were collected, representing the highest values of anomalous field radioactivity. These were prepared for geochemical analysis using the LASER method as well as for the determination of some trace elements using X-ray fluorescence.

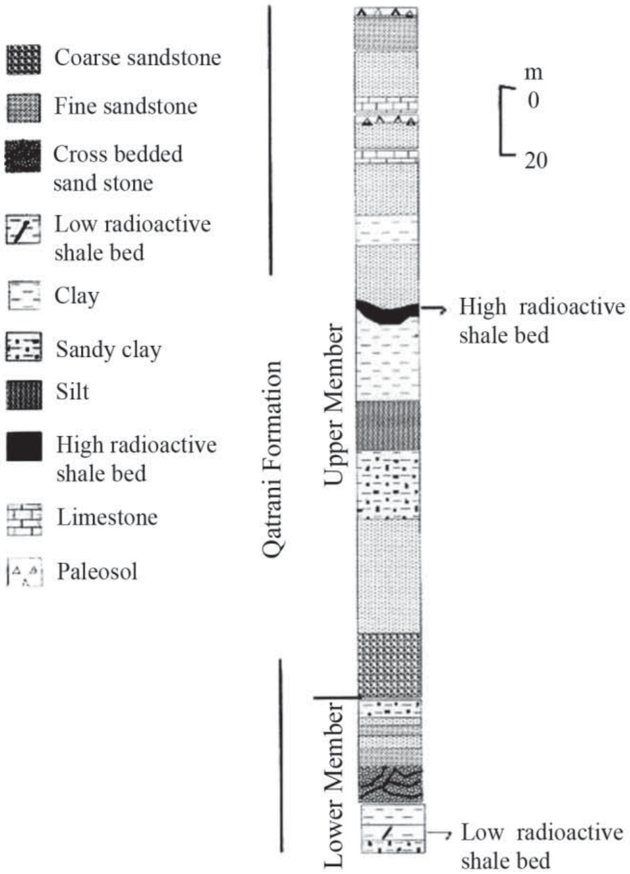

Figure 3: Composite stratigraphic cross section of Qatrani Formation including both low and high radioactive shale beds, after (MOUSA & HAROUN, 2006).

Both the mineralogical investigation and the geochemical analyses were carried out in the laboratories of the Egyptian Nuclear Materials Authority.

3.2. Mineralogical investigation

The accessory heavy minerals indicated that zircon is the most dominant mineral in all the heavy fractions of different studied size classes of the residual soil samples, together with lesser amounts of apatite, garnet, rutile and mica. The heavy fractions of the calcrete crustation sample revealed the presence of zircon, apatite, ilmenite, rutile and garnet. The content of these minerals generally increases with decreasing grain size.

The detailed mineralogical and chemical characteristics of the separated heavy accessory minerals from the studied residual soil and calcrete samples of the Quaternary sediments are briefly discussed below.

3.2.1. Zircon

Zircon crystals have actually been detected in both the residual soil and the calcrete crustation samples. These crystals are characterized by considerable variation in shape and size. They are generally colourless, translucent, and are mainly found as persistent heavy accessory detrital mineral species. Some grains are elongated with bipyramidal form, while others occur without distinct crystal faces. Others are generally subrounded to well round in shape. However, broken, zoned, outgrowths and other parallel aggregate-bearing varieties are generally absent. The back scatter electron images (BSE images) show the characteristic habit of the zircon (Figs. 4A–C). Alternatively, the EDAX spectrum for the separated zircon grains presented in Fig. 5A reflects the chemical composition of the mineral in addition to minor amounts of Hf, U and Th. Finally, the presence of subrounded to well rounded detrital zircon in the studied sediments reflects the effect of erosional weathering and transportation, indicating probable derivation from both rivers and beaches.

3.2.2. Apatite

The majority of the separated apatite grains vary in colour from pale to deep brown, and are mainly massive and translucent besides showing a well rounded to subrounded shape. The SEM photomicrographs and EDAX spectra for the apatite grains are presented in Figs. 4E&5B for the residual soil sample and Figs. 6C & 7B for the calcrete sample. The data reflects the chemical composition of apatite (Ca 59.73%, P 30.85% and Si 3.70%), with a relatively appreciable amount of uranium and thorium attaining 1.60 and 1.40% respectively. According to HEINRICH (1958), uranium usually substitutes for Ca in the apatite structure due to the similarity of the ionic radii (Ca = 1.06A° and U = 1.05A°).

3.2.3. Garnet, rutile and ilmenite

The EDAX scanned spots of a garnet crystal from a residual soil sample shown in Figs. 4F and 5C indicate a Si content

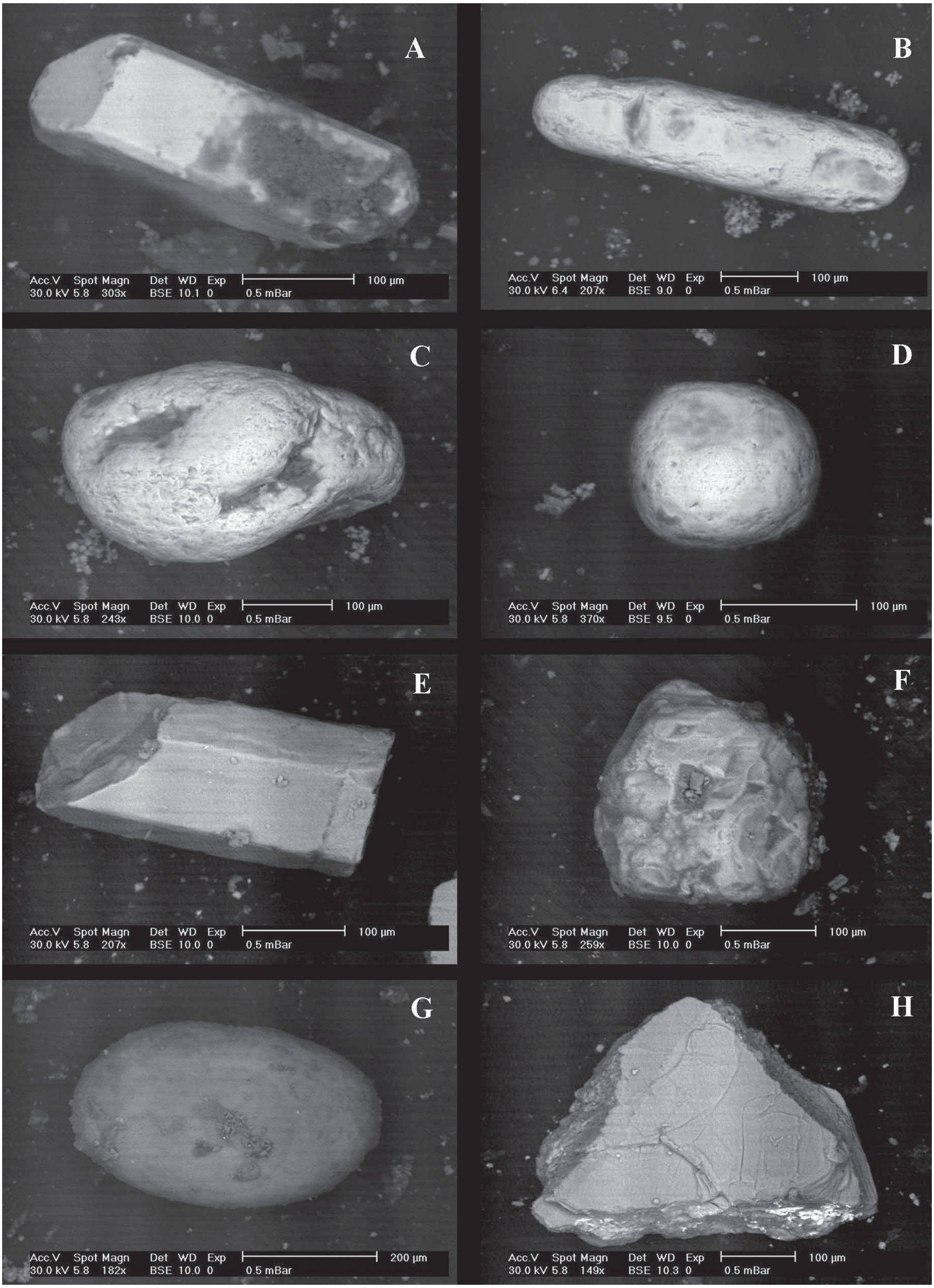

Figure 4: SEM backscattered electron images for the studied heavy minerals separated fro residual soil sample. A, B, C & D - Zircon crystalls of various habits, E - Apatite, F - Garnet, G - Rutile, H - Mica flak with interstitial uranium.

of 23.26%, 12.65% Al. Fe (48.03%), 11.44% Mn, Ca (1.54) and a Mg content of 1.22%. The SEM data for a garnet crystal picked from a calcrete sample shown in Figs. 6F &7E suggests a Si content of 39.22%, Al (19.25%), Fe (16.90%), Mn (15.14%), Ca (5.16%), and Mg content of 1.73%.

Rutile is actually found in both analyzed samples and assumes different reddish brown and straw-yellow colours. Rutile grains vary from highly rounded to sub angular in shape. According to Deer et al. (1962), the colour differences in the rutile grains generally correspond to variation in their chemical composition. SEM data confirm the chemical composition of rutile (Figs 4G& 6D) for the residual soil sample and the calcrete sample (Figs. 6D and 7E). Ilmenite crystals have only been detected in the calcrete sample, being completely absent in the residual soil sample. They occur as black metallic crystals characterized by their angular to irregular shape. The corresponding SEM microphotograph (Fig. 6E) and the EDAX spectra (Fig. 7C) for a separated ilmenite mineral grain reflect both the morphological features and the chemical composition of this mineral.

3.2.4. Mica

Mica grains occur in minor amounts in the residual soil sample in the form of soft flexible flakes, varying in colour from dark brown to black (mainly phlogopite). The corresponding SEM microphotograph (Fig. 4H) and the EDAX spectra (Fig. 5E) for the separated mica flakes reflect the morphological features and chemical composition. A bright yellow secondary uranium mineral was discovered interstitially between the mica flakes (Fig. 4H), semiquantitative analysis of which suggests that U attains up to 76.41% with an Fe content of 14.15% and a Si content of 2.77 (Fig. 5F).

3.3. Geochemical investigation

The country rocks of the studied residual soil and calcrete are actually represented by the clastic and non clastic rocks of the upper member of the Qatrani Formation. These rocks are generally non-radioactive (MOHAMED, 1965) however; some uranium anomalies are associated with the carbonaceous shale where it forms a ridge or a cliff, whereas when this shale is interbedded within the succession, it only shows uranium background levels (HAROUN, 2001). According to MOUSA & HAROUN (2006), the uranium concentration of some carbonaceous shale sites ranges from 17 to 34 ppm while the average universal background of the carbonaceous shale is 3.5 ppm (TUREKIAN & WEDEPOHL, 1961). Radiometric analyses of the collected representative samples from the residual soil and calcrete deposits indicated that the uranium concentration in the former ranges from 20 to 200 ppm with an average of 94 ppm while it assays at 121 and 85 ppm in the two studied calcrete samples (Table 1). These results indicate that both the residual soil and calcrete sediments assay a uranium content that is generally higher than that of the country rocks, and the fossil tree trunks, which are scattered within the studied residual soil in some sites.

Table 1: Uranium content in residual soil (R.S.) and calcrete (C.) as Quaternary sediment

Sample No.	U (ppm)
R.S.1	50
R.S.2	123
R.S.3	200
R.S.4	99
R.S.5	133
R.S.6	20
R.S.7	32
Average	94
C.1	121
C.2	85

The trace elementscontent of the study rock facies was evaluated by XRF analysis, performed on 14 trace elements in some representative samples of the residual soil and calcrete. The results are summarized in Table 2, in comparison with the data of TUREKIAN & WEDEPOHL (1961) from shale. This comparison indicates quite clearly that the studied sediments are enriched in Zr, Y, Zn, Sr and V. However, it has been discovered that while Sr in the residual soil is lower than TUREKIAN & WEDEPOHL'S (1961) average, it is much higher than the latter in the calcrete samples. This is most probably due to the presence of the carbonate cement. The genesis of uranium and the analyzed trace elements in both the studied residual soil and calcrete facies is discussed below.

4. MINERAL GENESIS

In their study of Oligocene rocks surrounding the local depression and sub basin of the radioactive Quaternary sediments, EL KAMMAR et al. (2000) reported that both the sandstones and the siltstones are the most common lithologies in the Oligocene Qatrani Formation. The sandstone can be described as a quartz – arenite where the detrital quartz constitutes >90% of the framework material. The heavy mineral fraction of these sandstones and siltstones indicates that hematite and goethite are the main opaque constituents as they represent about 50% of the total heavy fraction. The detected non-opaque heavy minerals include zircon, rutile, epidote, tourmaline, garnet, kyanite, biotite, staurolite and amphiboles.

Field investigation revealed that both the clastic sandstone and siltstone facies of the Oligocene Qatrani Formation are the main bed rocks and country rocks of the studied residual soil and calcrete deposits, and all the heavy detrital minerals separated from these deposits are actually derived by mechanical weathering of the country rocks. Weathering was by the action of both wind and water currents where the resistant heavy minerals such as zircon, garnet, apatite and rutile were deposited in the local low lying area of the residual soil and calcrete.

Table 2: Trace element content (ppm) of the studied residual soil (R.S) and calcrete (C) as Quaternary sediments compared with shale of major unit of earth crust (T&W), after TUREKIAN & WEDEPOHLE, 1961.

Nb	V	Ga	Sr	Pb	Ba	Y	Rb	Zr	Zn	Cu	Ni	Co	Cr	Sample No.
2	167	10	132	22	98	250	98	380	111	45	50	50	102	R.S.1
9	170	16	102	23	51	240	51	437	211	62	86	52	86	R.S.2
5	8	u.d	1381	34	261	u.d	u.d	295	174	13	19	3	85	C.1
u.d	6	u.d	1192	42	236	u.d	5	266	259	21	42	3	74	C.2
		25	400	12	600	180	140	-	90	50	80	20	100	T&W

u.d. undetected

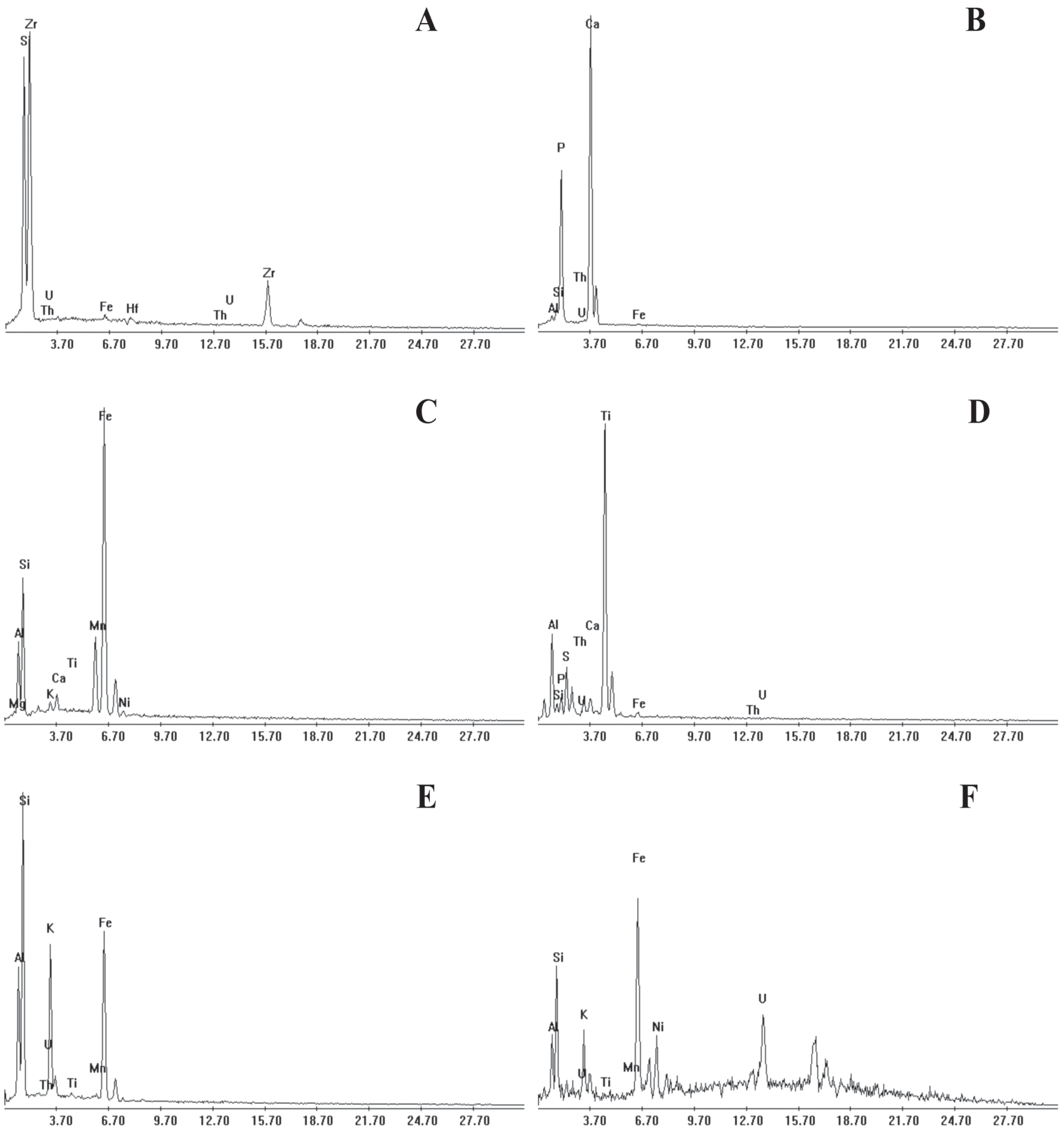

Figure 5: EDX analysis for the studied minerals of residual soil A - Zircon; B - Apatite; C - Garnet; D - Rutile; E - Mica and F - uranium mineral.

4.1. Genetic aspects of uranium mineralization

The possible mechanism of uranium accumulation in both the residual soil and the calcrete can be described according to the following sequence of operations;

1. The area of the residual soil is topographically considered as a local sub basin which has received all the detrital sediments and surficial fluids that have been derived from the bed rocks forming the upper successive scarps. These different types of surficial fluids involved both hydrothermal solutions that accompanied the Tertiary volcanicity as well as ground and meteoric waters.
2. The fluids leached uranium from the Oligocene bones formed mainly of apatite and which were enriched in uranium (HAROUN, 2001), ranging from 113 up to 930 ppm with an average of 394 ppm. However, the Th content in these bones is below the limit of detection (EL KAMMAR et al, 2000).
3. The leached uranium remained in a soluble form in the acid waters (mixed descending and ascending waters), probably as the complex uranyl ion, to be secondarily enriched in the porous sediments of the residual soil and calcrete via different adsorbents; namely, clays, organic matter, carbonate etc.

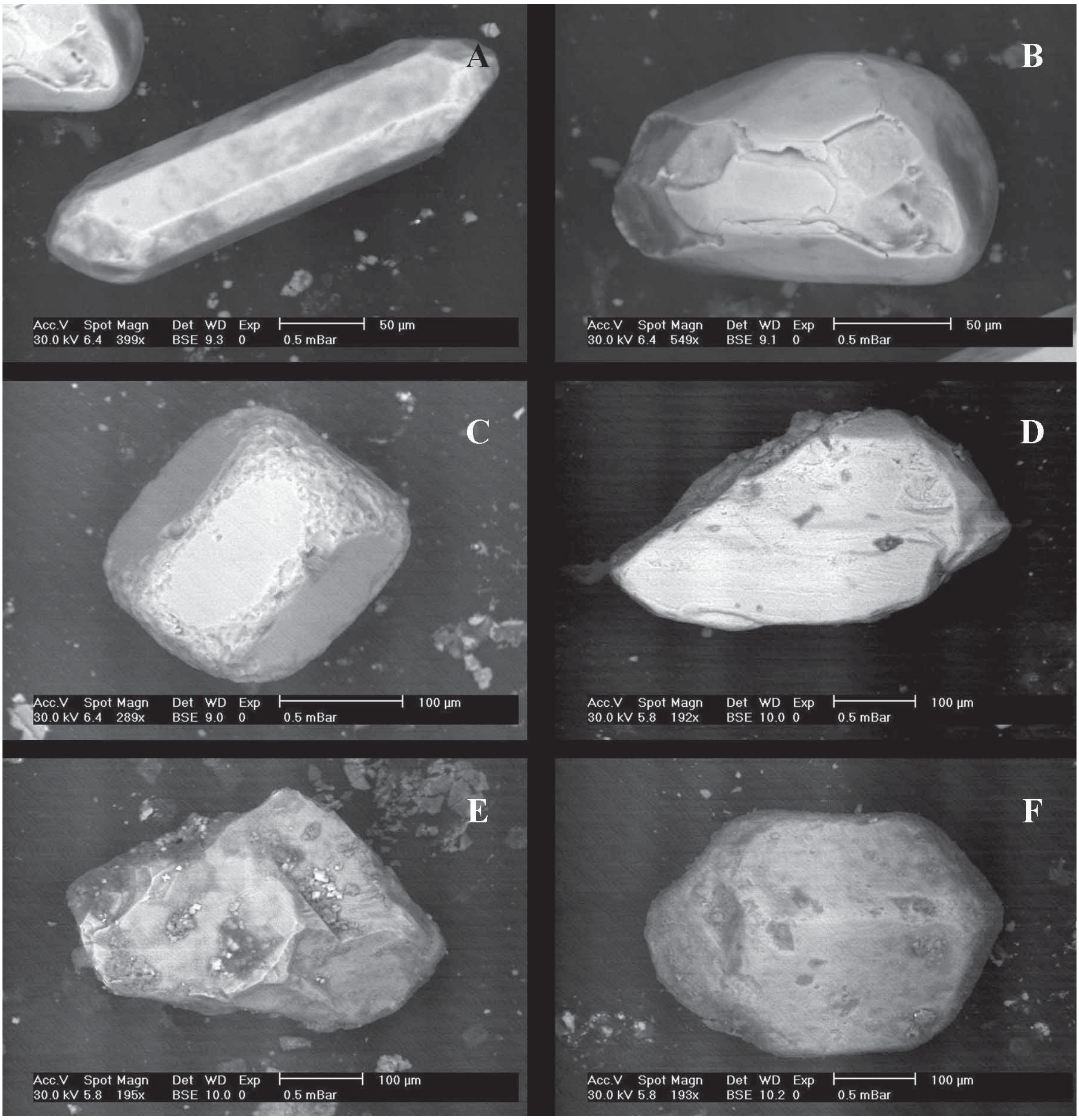

Figure 6: SEM backscattered electron images for the studied heavy minerals separated from calcrete sample. A & B - Zircon crystals, C - Apatite, D - Ilmenite, E - Rutile, F - Garnet

4.2. Trace element distribution

The relative enrichment of some trace elements can be determined by comparing the trace element content of both the residual soil and calcrete with their average values in the average crustal shale of TUREKIAN & WEDEPOHL (1961), (Table 2 and Fig. 8). From this comparison, the following can be concluded;

1. The enrichment of Zn and Zr in the residual soil and calcrete may be attributed to the following:
   - According to El Kammar and Basta, (1983), Zn is considered one of the elements enriched in the weathering zone. The highest enrichment factor is for Zn. The authors believe that Zn (0.74 Aº in Zn=6) which was substituting for Fe (0.74 Aº in Zn =6) in pyrite was liberated during oxidation.
   - High iron content is present in both the basalt and the red bed forming the bed rock surrounding the local basin of the residual soil and calcrete.
   - The increased Zr assay may be attributed to the presence of zircon in the residual soil as confirmed by the SEM analyses.
2. The enrichment of V in the residual soil as indicated by XRF analyses (Table 2 and Fig. 8) is mostly due to the weathering processes of the basaltic bed rock (ABDEL

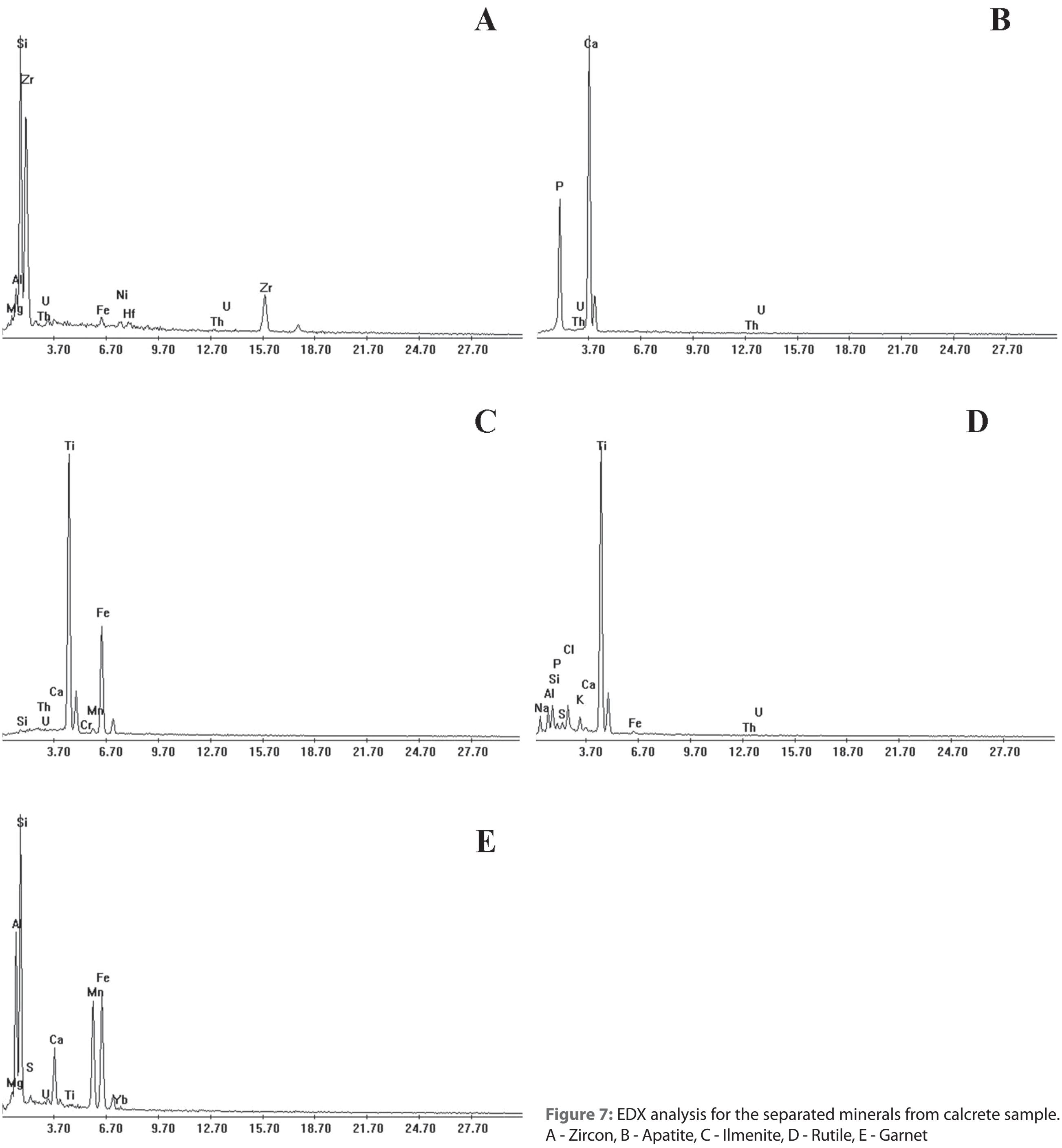

Figure 7: EDX analysis for the separated minerals from calcrete sample. A - Zircon, B - Apatite, C - Ilmenite, D - Rutile, E - Garnet

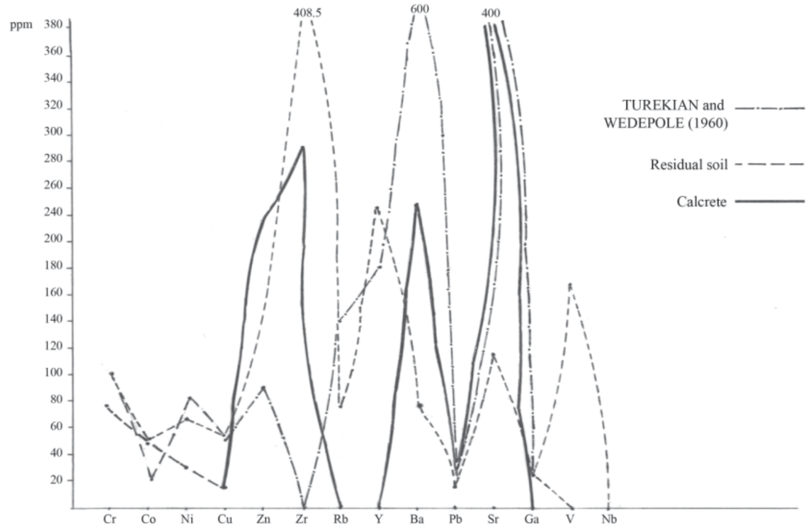

Figure 8: Variation diagram of trace elements in the studied Quaternary deposits.

MAGID et al., 1992), which contains a relatively high content of mafic minerals. Also, the increased V content may be due to the presence of iron minerals such as hematite, goethite and limonite, which represent the main opaque grains in the heavy mineral fraction of the sandstone bed rock (EL KAMMAR et al., 2000).

3. The enrichment of Y in the residual soil may be attributed to the weathering of Y- rich vertebral bone embedded in the Oligocene rocks. According to El Kammar et al, (2000), Y concentration in the vertebral bone can reach up to 583 ppm. These authors further mentioned that the accumulation of Sr+Y+REE in the rocks would follow in decreasing order from bone > shale > siltstone > sandstone > duricrust.
4. Enrichment of Sr in the calcrete deposits (Table 2 and Fig. 8) is most probably due to its liberation during diagenetic dolomitization of the calcite cement (EL HASHIMY, 1976). Such released Sr would readily be accommodated together with the sulphate of the weathering zone (EL KAMMAR & BASTA, 1983). The high concentration of Sr in the calcrete can also be attributed to its leaching from Sr francolite and phosphate minerals (EL AGAMI et al., 2005) and/or its leaching from the petrified Oligocene bones randomly scattered upon the surface of the bench scarps of the study area. According to El KAMMAR et al., (2000), the vertebrate bones could contain a high concentration of Sr reaching up to 1940 ppm.

5. CONCLUSION

Field relationships, SEM and chemical analyses revealed the relative enrichments of uranium and some trace elements such as Sr, Zr and Y in the studied residual soil and calcrete deposited on the eroded surface of Oligocene rocks. Mineralogical investigation of the studied sediments reveals the presence of zircon, apatite, ilmenite, rutile and garnet. Additionally, a secondary uranium mineral was discovered interstitially between the mica flakes. Weathering, erosion and transportation processes led to the presence of zircon, apatite, ilmenite, rutile and garnet as persistent heavy accessory detrital minerals in the studied radioactive sediments. The U, Sr, Zr, Y and V budget of these sediments are potentially interesting.

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