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Geochemical Characteristics of Upper Cretaceous Dolomite in Northwest Libya: Implications for Dolomitization and Diagenesis; El Zintansection as a Case Study

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Abstract: ***Lithostratigraphically, the Sidi As sid Formation (Upper Cretaceous) in the El Zintan section (JabalNafusah, NW Libya) consists of three units: upper marl, middle marl with intercalations of dolostone, and lower dolostone. Based on crystal size and shape, three types of dolomite have been classified. Fine crystalline dolomite (D1) consists of nonplanar dolomites, fine to medium crystalline dolomite (D2), and very coarse, consisting of mostly planar dolomites (D - 3). The Cathodoluminescence technique also showed that most of these dolomites have bright yellow to red and dull orange luminescence and zoning. Based on X - ray diffraction (XRD), all dolomites are relatively well ordered and non to nearly stoichiometric dolomite. The stable isotopic studies and element analysis show that the major elements such as sodium (D1: 412.166 ppm; D2: 175 ppm; D3: 420 ppm), strontium (D1: 107 ppm; D2: 85 ppm; D3: 81.2 ppm), manganese (D1: 271 ppm; D2: 91 ppm; V3: 242 ppm), and iron (D1: 4856.66 ppm; D2: 373 ppm; D3: 3287 ppm) and the values of the stable isotope of oxygen (D1: - 2.202‰; D2: - 2.131; D3: - 4.359‰) and carbon (D1: 2.080; D2: 2.076 ‰; D3: 1.581 ‰). The values of major elements and depletion of the carbon and oxygen isotopes can be related to the effects of temperature diagenetic, and mesosaline reflux of dolomitizing fluids during the early diagenesis. These dolomites originated in a tidal environment in this Formation. The major sources of magnesium for the dolomites are seawater and hypersaline fluids.***

Keywords: SidiAs Sid Formation, Dolostone, Tidal Environment, Hypersaline brines, Mesosaline

1. Introduction

In Libya, the Early - Middle Cenomanian carbonates are mainly deposited in the north - western part of Libya, along a roughly SW - NE trending belt (Fig.1), and are characterised by variations in thickness and facies. These variations, which clearly appear along transects, mainly oriented north - south and northeast - southwest, could be controlled by local (tectonic) and global (eustatic) factors. According to a combination of field, petrographical, and geochemical evidence from the Upper Cretaceous SidiAs Sid Formation in northwest Libya, dolomitization occurred during the Cenomanian as a result of near - surface mesosaline reflux of dolomitizing fluids. Despite what appears to be a global shortage of platform - scale dolomitization throughout the Upper Cretaceous, the Jeffara Escarpment in northwestern Libya has significant and widespread dolomitization. This is thought to mostly reflect the climate of the region. The Formation was situated within a hot and arid climate area (31.9557'N and 12.2249' E), resulting in sufficient evaporation of seawater to produce mesosaline brines. This appeared along a distinct climate zone north of the Arabian Plate's hot, humid equatorial temperatures, and explains the abundance of dolomitized platforms restricted to the circum - Mediterranean region at the time. When sea - level rise caused by a second - order maximum flooding event occurred in the deposition of relatively shallow water at the Cenomanian–Turonian boundary, dolomitization was significantly less widespread. Reflux of hypersaline brines and mixed marine - meteoric fluids have been suggested as possible causes of dolomitization within laterally equivalent strata (M'rabet, 1981; Badalini et al., 2002; Bodin et al., 2010; Abdallah, 2003; Touir et al., 2009).

The Upper Cenomanian SidiAs Sid Formation in El Zintan is heavily dolomitized. The Upper Cenomanian Sidi As Sid Formation in El Zintan, provides an excellent opportunity to study the processes and controls of massive dolomitization of marine carbonates on the northern margin of the northwestern part of Libya during the upper Cretaceous. Previous research (Christie 1955; Burollet 1960; Desio et al., 1963; Conant and Goudarzi1967; Busson 1967b; Assereto and Benelli 1971; Evan 1977; El - Bakai; 1991; 2003; El Bakai et al., 2010) have documented the diagenesis and dolomitization of Cenomanian strata in northwestern Libya, but no previous work has been done on the El Zintan section. The El Zintan section is an excellent exposure of the Sidi As Sid Formation and provides a high - quality outcrop analogue for the region. This study will present a multidisciplinary and multi - scale evaluation, using petrographical, geochemical, and field evidence in order to fully evaluate the controls on dolomitization. This study presents a multidisciplinary and multi - scale evaluation of dolomitization controls using petrographical, geochemical, and field evidence.

Geological setting

During the Upper Cretaceous, regional sea transgression created a broad and shallow carbonate platform that extended through North Africa (late Cenomanian). Continued sea - level rise across a flat area resulted in the deposition of marine carbonate layers in shallow subtidal facies (Wood et al., 2014), which is represented in the Jiffarah Escarpment by the Sidi As Sid Formation (Wood et al., 2014), defined in the Jiffarah Escarpment by the Sidi As Sid Formation (Fig.1). The Formation is classified as the Ain Tobi and Yefrenmarl Member and is defined by a clay - rich marlfacies (Fig.2A). The Ain Tobi Member in the El Zintan section is defined by an upward decrease in clay concentration and widespread dolomitization, resulting in metre - scale, upward - shallow successions (Fig.2B). Within the Sidi As Sid Formation, an entirely upward - shallow succession is visible, culminating in the development of tidal flats within troughs in Northwest Libya (Köhler 1982). Cenomanian strata in this study show a high concentration of bivalves, cephalopods, gastropods, Peloids, thalassinoides, and rudist (Fig.2 C, D &E), indicating that the relative sea level has continued to rise and open marine conditions have persisted. As the rate of relative sea - level rise moderated, the Ain Tobi Member developed enormous rudist build - ups that prograded northward. . It has an average thickness of 20 meters in the field region (Fig.2B), but reaches a maximum of 50 meters in the escarpment's northernmost section (Fig.2B). The SidiAs Sid Formation is marked at its upper part by a thin, yellow marl layer (Yefrenmarl) and ischaracterised by a ca.4 m thick marl horizon containing open marine fauna such as echinoids and bivalves, indicating that this member gradually becomes more subtidal as it progresses upward.

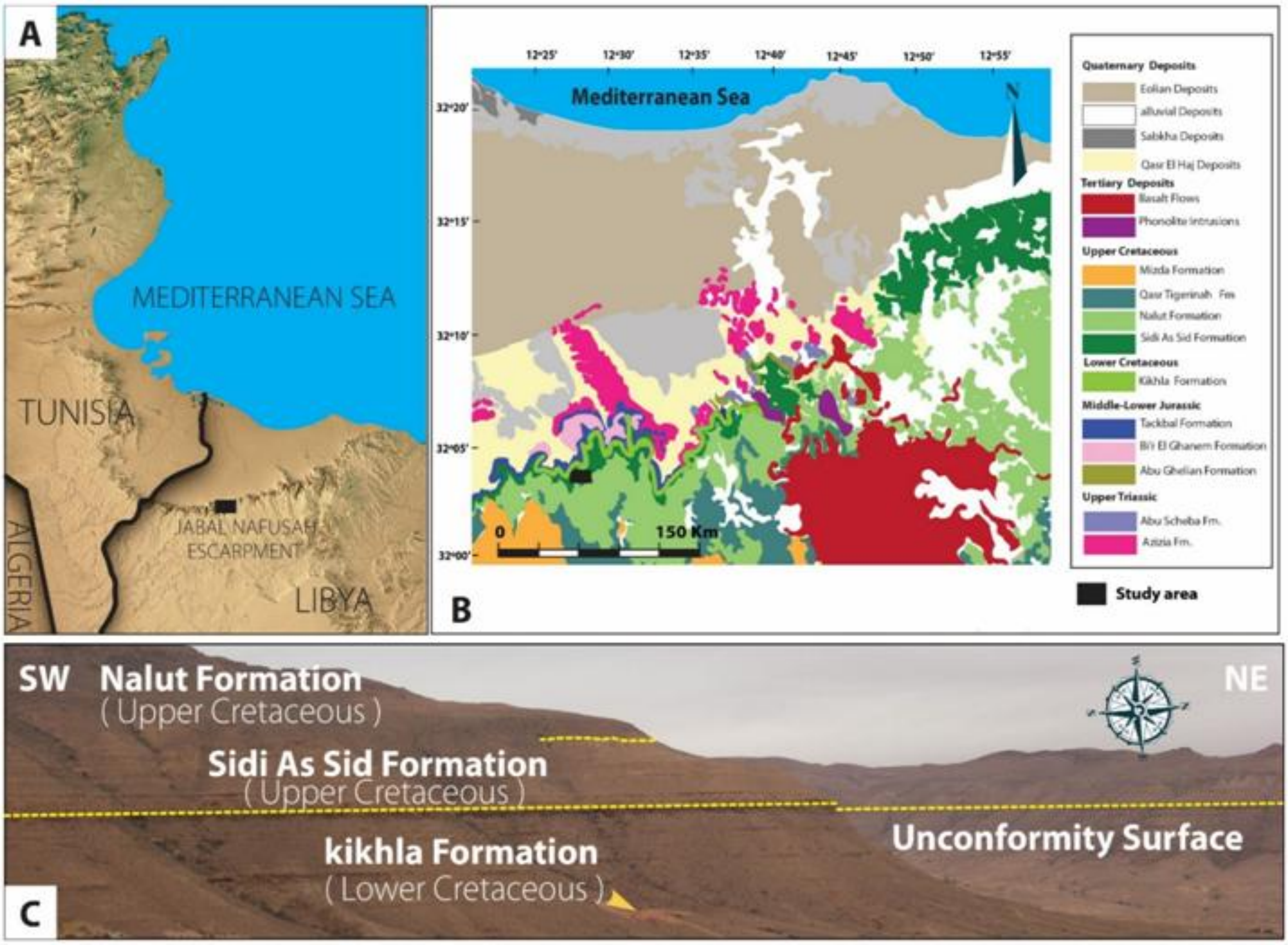

Figure 1: a) Shows the 3D render topographic map of the location of the study area and b) the geological map of the studied area. (Source; digital artwork by Frank Ramspott)

The transition between the Ain Tobi and Yefren Members is gradual and is characterised by an increase in the thickness and frequency of carbonate marl units (Fig.3A). Yefrenmarl Member is a calcareous shale to argillaceous limestone that weathers to a tan, yellow, or green powder. Beds of crystalline gypsum with white and green internal laminations are also present. This marl unit is only partially dolomitized, as well. The tubular construction, diameter, and branching patterns of these structures indicate that they belong to the ichnogenus Thalassinoides, which is characterized by the basal unit by highly bioturbated (Thalassonoides; Fig.3B), and quartz grains with abundant rounded to subrounded, moderately sorted detrital quartz grains (Fig.3 D). The siliciclastic content of the dolomitic quartz arenites, and dolomitic shales is estimated to be slightly greater than 5% in abundance. In the middle part, the Ain Tobi Member is 10 m thick and has a rather constant thickness and high concentration of dolomite in the studied area. However, certain broad trends and facies are recognized. The lower member's bottom section is characterized by a well - defined rudist (ca 2 m) (Fig.3C). The Rudists are commonly preserved (Fig.3 E) and are often only identifiable through calcite - cemented biomoulds. Moldicrudist debris may have a maximum dimension of up to 5 cm, such molds include bivalve, gastropod and ooids.

Petrography of the diagenetic process

The Ain Tobi Member (late Cenomanian) was deposited upon the Kikhla Formation, extending northwestward into a tidal flat deposit. This platform has restored the flow of dolomitizing fluids along the depositional dip. Bioturbation, small scale scours, lamination, and massive bedding are most of the sedimentary structures that characterized the lower Ain Tobi Member in the El Zintan section. Dolomite crystals are subhedral to euhedral with aggregate interlocking to sucrosic textures. Individual dolomite crystals may be clear, uniformly clouded by inclusions, or have cloudy cores and clear rims. The common occurrence of quartz grains in the dolomite at or near the base of the eastern section and the gradual shift of quartz grains (Fig.4A) to higher stratigraphic levels further west. The external morphology of these grains is easily visible with a hand lens, whereas their internal structures in thin section have been obscured by alteration to aphanitic or very fine crystalline dolomite. However, texture varies stratigraphically, within the Ain Tobi and Yefrenmarl Member. The upper parts of the Ain Tobi Member, which are controlled by peritidalfacies, dolomite is finely crystalline (D - 1) (Fig.4 B). Muddy sediments in packstones and wackestones produced a massive area of dolomitization, and nucleation rates exceeded growth rates in this area (Sibley & Gregg, 1987) and was characterised by coarsely crystalline and having a cloudy core in the middle and lower sections of the Ain Tobi's El Zintan section (Fig.4 C), clear rim morphology (D - 2). Coarse sediments in rudistgrainstones are eventually replaced by calcite (Fig.4 D) thisresults in the formation of coarser dolomite rhombs (D - 3).

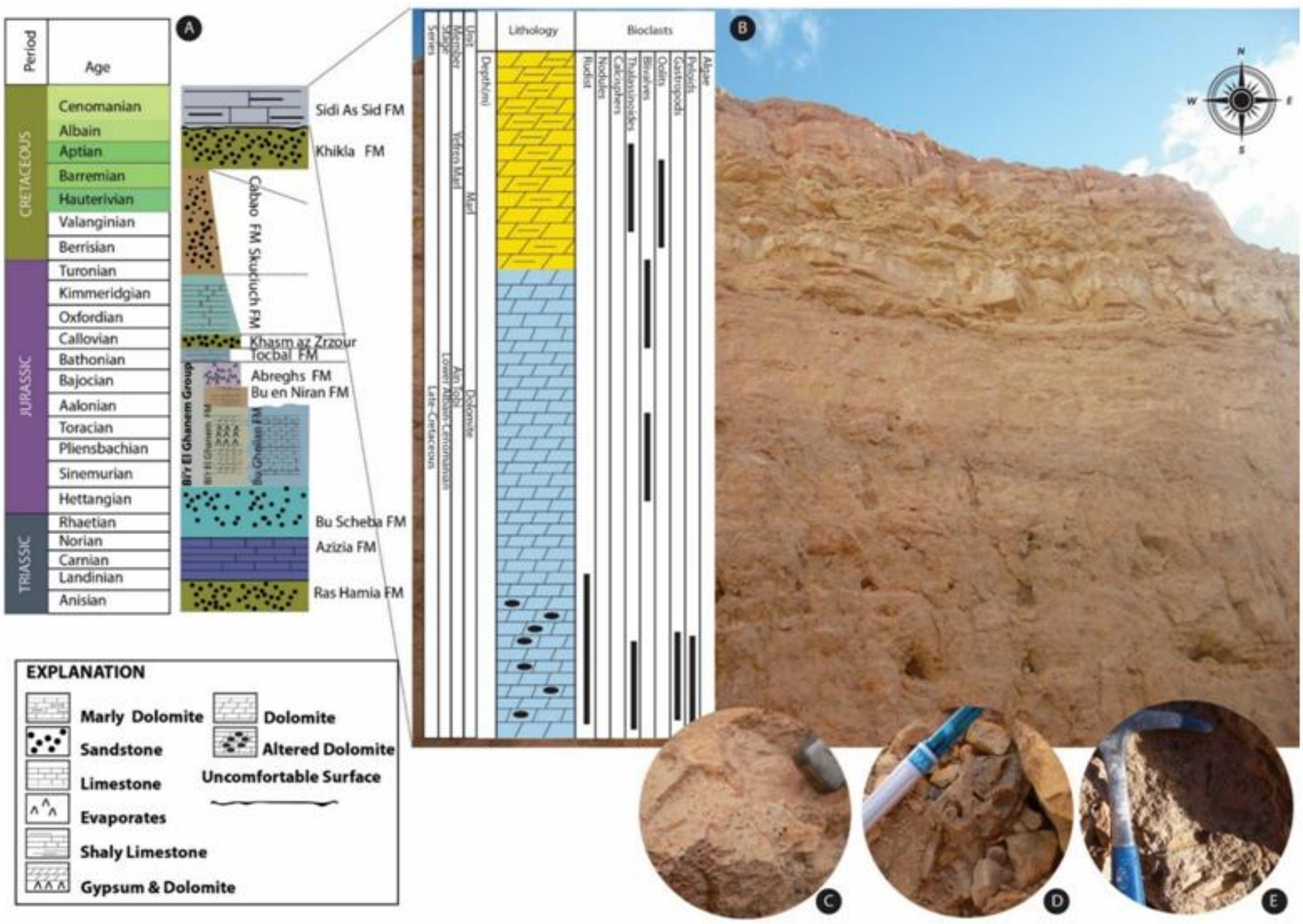

Figure 2: a) Stratigraphic Nomenclature of NW Libya b) aLithostratigraphic map of the study area showing the variouslithologic units. c) fossilized shells d) gastropods shell e) phylum Mollusca (bivalves) assemblages from the upper Cretaceous (Ain Tobi Mem.).

Figure 3: a) showing the stratigraphy of the Ain Tobi and Yefrenmarl Member b) bioturbated (Thalassonoides) c) rudist fragments are well preserved d) dolomite with quartz grains is thin section of detrital quartz grain with overgrowth. e) single rudist fragments are common constituents in Ain Tobi Member in the El Zintan section.

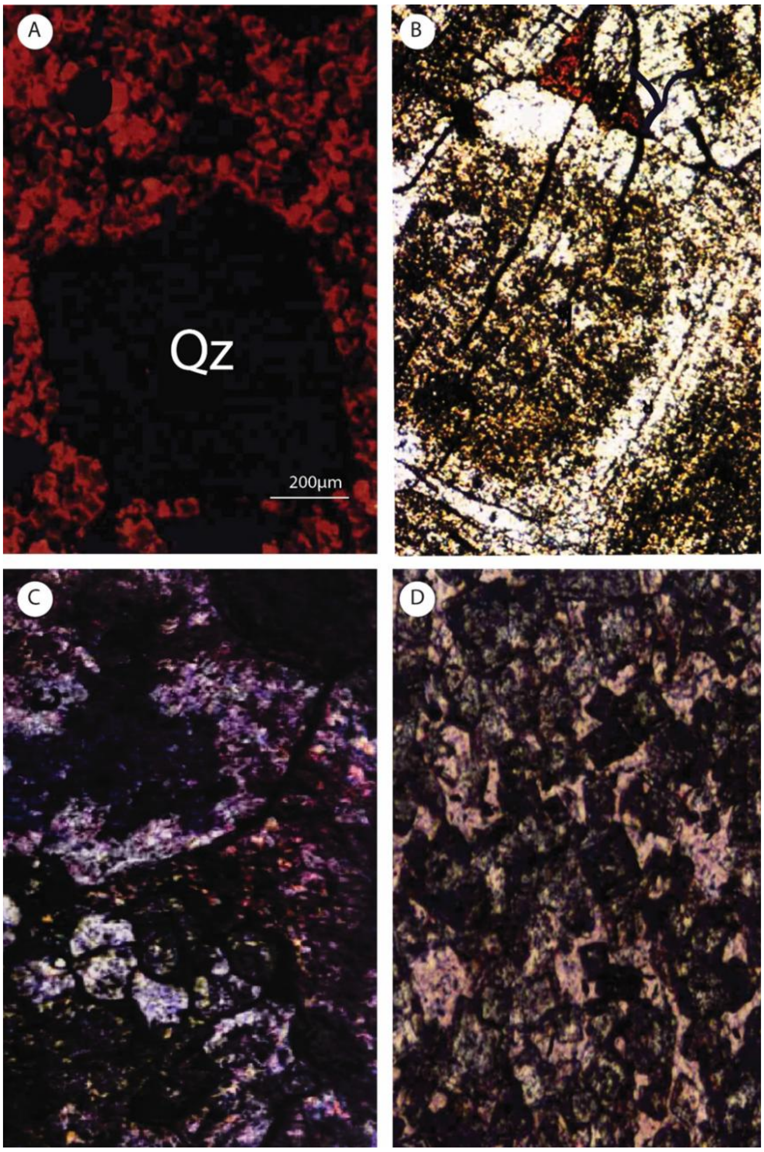

Figure 4: a) CL image of quartz from Ain Tobi Member b) Thin - section photomicrograph of fine dolomite crystalline (D - 1) with cloudy cores and clean rims d) Thin - section photomicrograph from the El Zintan section of dolomite crystalline (D - 2) shows the contact (arrow) between very coarse d) dolomite crystals are replaced by calcite (D - 3)

2. Methods

Elemental analysis was performed on 10 samples using an XRF (X - ray fluorescence spectroscopy) instrument. In this method, the samples are crushed and powdered by grinding processes. Major elements (Fe, Mn, Na and Sr) were analyzed in terms of weight level (ppm). The sectionis sampled in a systematic manner to cover the full range of facies and textures observed within Sidi as Sid Formation. X - ray diffraction was used to determine the mineralogy of samples.

Geochemistry

The results of stable isotope analysis and concentrations of major elements were used to constrain the origin of dolomitizing fluids are summarized in Table 1.

Major element

Overall, there is a clear variable in the concentration of major elements of the Ain Tobi Member in the El Zintan section, with the dolomite showing distinctive elementalconcentrations. Theiron concentration in the Ain TobiMember is the highest for the entire Ain Tobi Member in the El Zintan section, with a mean = 3685.88ppm. The Ain Tobi Member in El Zintan sectionalso has the highest Na (mean = 360.33 ppm, 100 to 550ppm.), Mn concentrationfor the whole Ain Tobi Member (mean = 227.78ppm) and Sr concentration for the whole Ain Tobi Member (mean = 99.14 ppm, 70 to 119 ppm).

Carbon, oxygen isotopes

The mean value of the $\delta^{18}O$ signature of dolomite in the Ain Tobi Member of the El Zintan section is (- 1.95) and has a slightly more negative oxygen isotope signature and an average of carbon isotope ratio of (2.024) (0.787 to 4.843). The Ain Tobi Member shows the slightest positive trend (Fig.5). The Ain Tobi Member has a low degree crystal order (Fig 6), which reduces the order degree of dolomite crystal to a non - stoichiometric level. The very low - order degree of the dolomite represents a high speed of dolomitization during the diagenetic stages (Luis A., et al,.2004). The existence of typical marine dolomite major element signatures and the presence of only slightly more positive oxygen isotope values all imply that mesosaline

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fluids (Fig.7) were used to dolomitize the rock (El Bakai2003; Richard et al., 2017).

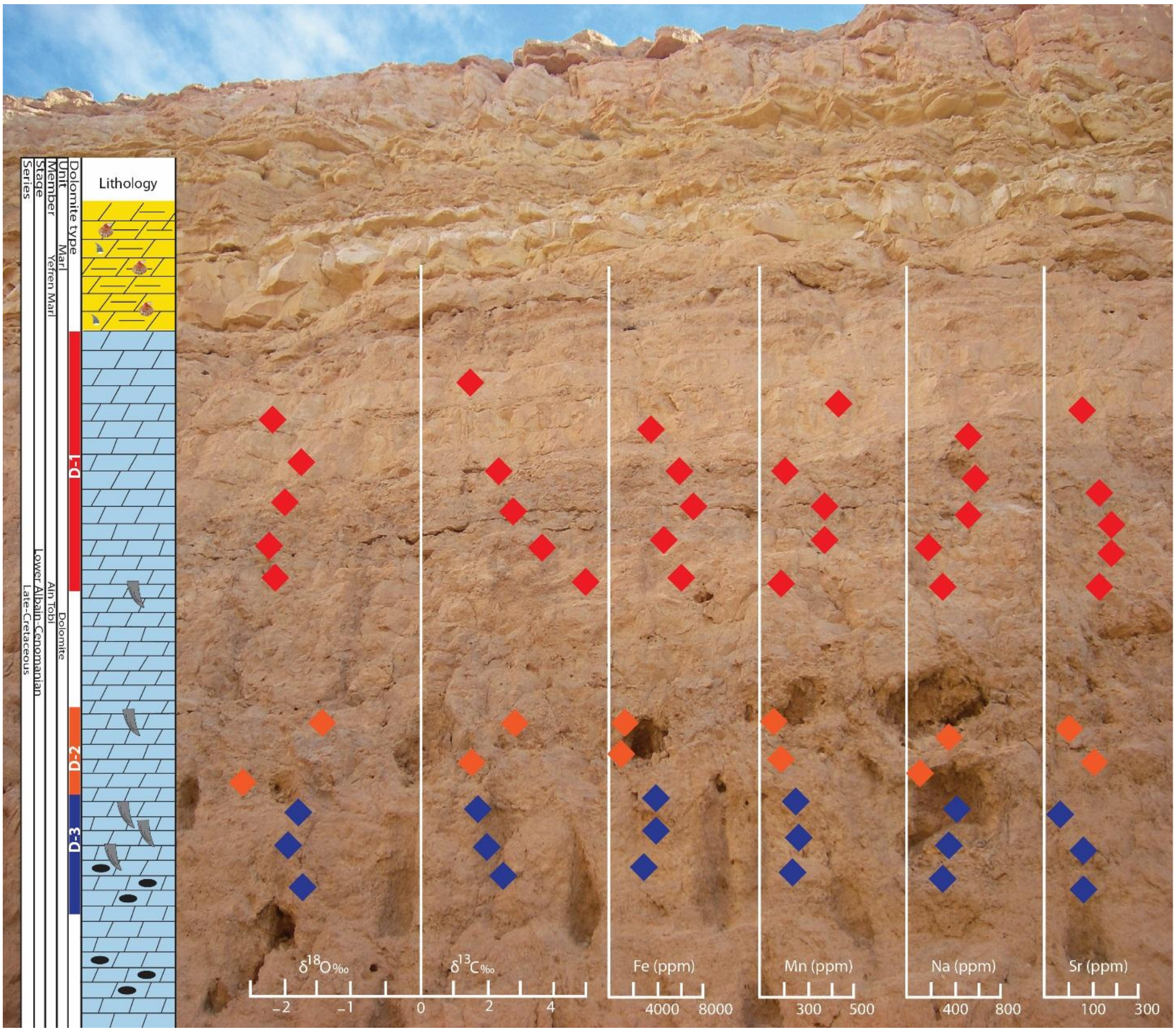

Figure 5: Stable isotope, trace element and dolomite type distribution of the El Zintan section

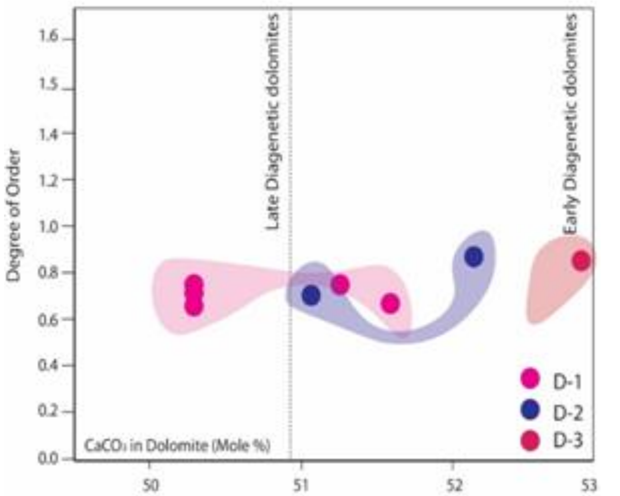

Figure 6: Differentiation of the Ain Tobi Member in the El Zintan section shows the samples are non - to near stoichiometric dolomite.

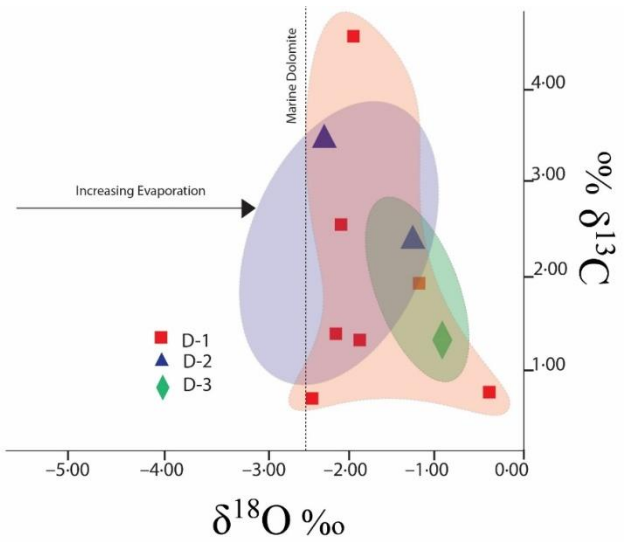

Figure 7: Carbon ($\delta^{13}C$) and oxygen ($\delta^{18}O$) and stable isotope cross plot of dolomite samples from the Ain Tobi Member. (based on data from El Bakai 2003).

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Table 1: Shows the Ain Tobi Member'saverage major element composition and endmember stable isotope signatures.

Sample	Fe	Mn	Na	Sr	$\delta^{18}O$*	$\delta^{13}O$*
D - 1	4226	274	466	96	- 2.597	0.787
D - 1	3545	415	460	98	- 2.027	1.197
D - 1	5243	179	550	103	- 1.879	1.9
D - 1	7620	326	470	119	- 2.17	1.25
D - 1	3875	320	370	115	- 2.225	2.508
D - 1	4631	112	157	110	- 2.315	4.843
D - 2	345	102	134	110	- 2.15	3.435
D - 2	181	33	250	70	- 1.318	2.86
D - 2	565	149	100	100	- 2.945	1.292
D - 3	3287	242	420	81.2	3.44	1.58

* Based on data from El Bakai 2003.

3. Discussion

Origin of dolomite

Dolomitization in the Sidi As Sid Formation is composed ofstratabound dolomite bodies, which resulted by mixed meteoric - marine fluids, as previously proposed (Köhler, 1982; Abdallah, 2003). Ain Tobi Member also shows normal marine $\delta^{13}C$ signatures (average = 2.024‰), most likely as a result of Upper Cretaceous limestone buffering the carbon isotopic composition during recrystallization (Land 1980; Rott& Qing 2013). At lower temperatures, the measured 18O value based on dolomite is slightly more negative (Wilson et al., 2002; Voigt et al., 2004). The slight increase in the $\delta^{18}O$ values of brines in the Ain Tobi Member ($\delta^{18}O$ = - 2.050 ‰) is consistent with deposition of these facies in very shallow water, implying that the platform was spatially closer to the brine pool and that hypersaline fluids were available to the platform later in the Cenomanian - Turonian. The Ain Tobi Member in the El Zintan section has an average Sr concentration of 99.13 ppm, more typical of normal seawater. According to oxygen isotope ratios and facies analysis, dolomite precipitated from seawater. . If the partition coefficient of iron and manganese is greater than one, both of these elements are preferentially incorporated into the dolomite crystal under reducing conditions (Machel, 1988). The measured values of Fe (181 to 7620 ppm) and Mn (33 to 415 ppm) for the Ain Tobi Member, imply precipitation under at least moderately reducing conditions.

Model of dolomitization

Since the late Cenomanian, carbonate sediments have dominated northwestern Libya. This sequence begins with the SidiAs Sid Formation, a dolomitized shallow platform deposite, which grades up within the intertidal lagnoonal (Evans., 1977) as a result of global sea level rise and the resulting global climate change, altering drainage patterns during the deposition of the Ain Tobi Member (Fig.8). The Cenomanian depositional environments occurred in the JabalNafusah escarpment as a part of restricted or lagoon and open to semi - restricted ramp (Burchette, T. P. and Wright, V. P.1992; Elrick and Read 1991). However, the ramp margin is inferred on the basis of Cenomanian rudist in the Ain Tobi Member in the El Zintan section. The Ain Tobi Member's base is dominated in the El Zintan section by rudistbioherms, indicating a return to relatively shallow water and greater energy conditions. It is probable that these facies indicate a low - relief shelf break, although widespread dolomitization has made specific stratigraphic relationships difficult to establish (Fig.8).

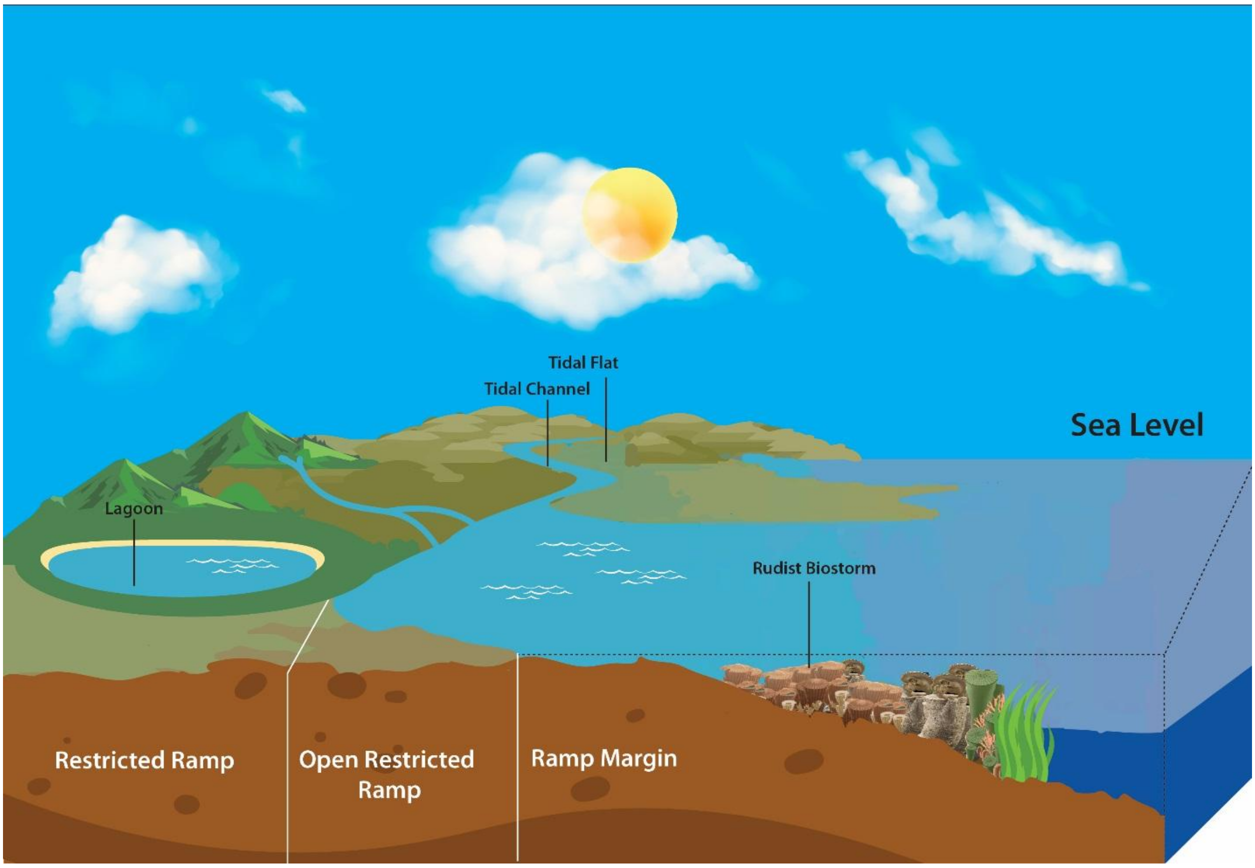

Figure 8: Block diagram of thedepositional environment of SidiAs Sid Formation. NW Libya. Modifies after (Köhler 1982).

The texture, distribution, and geochemistry of dolomite are generally compatible with dolomitization, slightly reduced, mesosaline fluids at low temperatures. Although conceptual models of reflux dolomitization commonly use penesaline fluids, close to gypsum saturation there is compelling theoretical evidence that ubiquitous dolomitization can occur using mesosaline brines. Many authors have proposed that dolomitization is associated with a relative sea - level fall (Köhler, 1982; Touir et al., 2009).

This study demonstrates unequivocally that when the amplitude of the sea - level shift is small, dolomitization can continue to proceed throughout sea - level rise, terminating only during periods of maximum flood. The Fe and Mn contents in the Ain Tobi Member imply a flow route that has been widened laterally as a result of relative sea - level rise.

Penesaline reflux modeling and subsurface investigations have revealed that the amount of dolomitization decreases with distance from the brine source (Saller and Henderson, 1998). However, Seepage reflux dolomitization occurs when hypersaline brines with high Mg^{2+}/Ca^{2+} ratios percolate through undolomitized sediment in the shallow subsurface, whereas the brines are generated by the evaporation of sea water in lagoons. The high Mg^{2+}/Ca^{2+} ratio occurs because evaporation results in the precipitation of gypsum or anhydrite which removes Ca^{2+} from the fluid and the evaporation raises the density of the brine. At this point, the brine can percolate into the underlying sediment and begin to dolomitize it. The operation of the seepage reflux mechanism is maintained if the lagoon from which the brine originates is replenished from the sea (Fig.9).

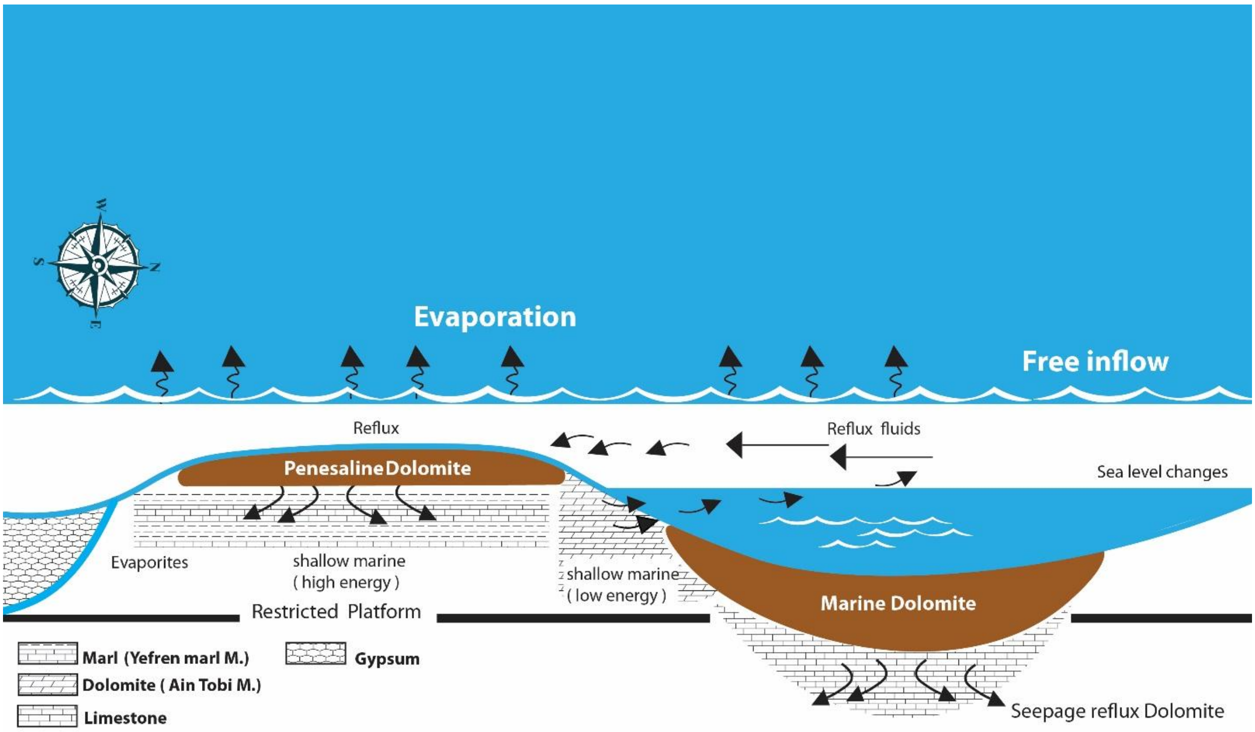

Figure 9: A conceptual model of dolomitization for the Ain Tobi Memebr in northwestern Libya. Under changing sea - level, northwards progradation of the platform ensured laterally extensive dolomitization. The mechanism involves the evaporation of sea water in an area of supratidal deposition until the point of gypsum precipitation. Removal of calcium leads to formation the brines with high Mg^{2+} content. Modified after (Tucker & Wright, 1990)

4. Conclusion

Dolomitization occurred during the Cenomanian and Turonian based on the integration of field, petrographical, and geochemical data from the Upper Cretaceous SidiAs Sid Formation in northeastern Libya. During the Cenomanian–Turonian, the area studied was tectonically stable, allowing reflux to persist for long enough for strata to become extensively dolomitized. This was most likely caused by a series of simultaneous passages of seawater. Due to sea level rise and a second - order maximum flooding event, very deep water marls were deposited at the Cenoman–Turonian border, where dolomitizing fluids were probably far away, resulting in considerably less widespread dolomitization.

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