

THERMAL CHARACTERIZATION OF LACUSTRINE DOLOMITES FROM THE SAMBHAR LAKE PLAYA, THAR DESERT, INDIA

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Abstract

The Sambhar Lake playa sediments in Thar desert, western India, host a variety of evaporite minerals. Carbonates form a major proportion of the evaporites in a 23 m deep core and several dolomite-rich horizons are recorded. Our XRD and DTA based investigations show that these dolomites are non-stoichiometric and disordered (proto-dolomites) except at one level (4.08 m) from where a well-ordered dolomite is reported. We also record frequent variation in the characteristics of dolomites with depth in terms of structural ordering, Ca/Mg ratio and Fe content. Dolomites reported from the Sambhar Lake playa are interpreted to be authigenic due to the absence of any carbonate rock in the catchment and seem to be derived from a precursor calcite under (sub-) evaporitic conditions.

Keywords: DTA, lacustrine carbonates, playa sedimentology, Sambhar Lake, Thar desert

Introduction

The occurrence and origin of dolomites has been a subject of interest for over six decades now and probably no other mineral has attracted as much speculation regarding its origin as dolomite. The so-called ‘dolomite problem’ has been discussed and reviewed by many workers (Usdowski [1], Lippmann [2], Zenger and Dunham [3], Morrow [4], Last [5], Warren [6, 7]), and the main facets of the problem are: (a) in spite of a simple composition, $\text{CaMg}(\text{CO}_3)_2$, it has not been possible to synthesize dolomite under normal sedimentary conditions, (b) despite its abundance in ancient rocks, dolomite is relatively rare in modern sediments, and (c) the understanding of the inorganic and organic geochemical factors conducive to form dolomite are still largely unresolved.

Recent formation of dolomite in coastal sabkhas and in inland lakes and playas has received a fair attention since the discovery and documentation of Coorong dolomites in southern Australia (Skinner [8], von der Borch *et al.* [9]; Rosen *et al.* [10, 11]). Since then a number of occurrences of dolomites have been reported and discussed from lacustrine

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or playa settings e.g. Deep Springs Lake, California (Jones [12]), Searles Lake, California (Smith *et al.* [13]; Smith [14]), Great Salt Lake, Utah (Graf *et al.* [15]), Waldesa and Deadmoose lakes in western Canada (Last and Schweyen [16]; Last and Slezak [17]), Spanish playa lakes (Comin and Alonso [18]); Didwana Lake, India (Singh *et al.* [19]; Wasson *et al.* [20]), Lake Bosumtwi, Ghana (Talbot and Kelts [21]), Namib desert lake deposits (Teller *et al.* [22]) and many others. Most of these occurrences are from modern or Pleistocene settings and dolomites have been interpreted to have formed in playa environments.

In recent years, lacustrine (playa) dolomites have received a fair amount of attention due to their sensitivity to lake level fluctuations and climate change. The current theories of formation of lacustrine dolomites can be grouped as (a) loss of CO₂ as a result of change in P-T conditions or plant assimilation, (b) evaporitic concentration and (c) 'mixing water' model (see reviews by Last [5], Tucker and Wright [23], Warren [7]). It has been argued that the dolomite formation is influenced by the interplay of a variety of geochemical factors viz. Mg/Ca ratio, carbonate alkalinity, sulfate concentration, temperature and total salinity. Much of the discussion has been centered on the distinction of 'primary' (crystals which nucleate from solutions and either accumulate as primary sediment or precipitate into megascopic pores as cement...) (Land [24]) and 'secondary' (showing clear evidence of replacement of a pre-existing solid material) dolomites. There seems to be a consensus that primary dolomites are very rare and, in most cases, they form by replacement of a precursor carbonate (Mg-calcite) (Hsu [25], Usdowski [1], Lippmann [2], Chillingar *et al.* [26], Morrow [4]). However, the actual mechanism of dolomitization is still being debated and the difference of opinion persists about the source of Mg²⁺ ions and the process by which the dolomitizing fluid is pumped through the carbonate sediments.

The key idea is that the dolomite is a difficult mineral to form because of the precise Ca-Mg ordering required (Goldsmith [27]). Ideal dolomite consists of carbonate layers alternating with layers consisting entirely of Ca atoms or entirely of Mg atoms. Recent and very young dolomites, both from marine and playa environments, often show strongly disordered crystal structures (so-called 'proto-dolomites') characterized by non-stoichiometry (Ca_{>1}Mg_{<1}) and disorder of the distinct layers (Lippmann [2]). With increasing diagenesis, the structure becomes more and more ordered, forming layers of CaCO₃ and MgCO₃ (Lippmann [2]), and a stoichiometric ratio of Ca and Mg (1:1) is attained too. Later diagenetic changes in contact with reducing Fe²⁺ rich solutions, the dolomite structure may incorporate Fe²⁺ or Mn²⁺ ions, thus showing the formula, Ca(Mg, Fe²⁺, Mn)(CO₃)₂.

At Sambhar Lake located at the eastern margin of the Thar desert, India (Fig. 1), dolomite occurs in the superficial duricrust (Roy *et al.* [28]) and in deeper sediments as well (Sinha and Raymahashay [29, 30], Sinha *et al.* [31]). The amount and character of dolomite vary quite remarkably in the playa sediments, as does the association of detrital and evaporitic minerals. The purpose of this study is to correlate the dolomite properties with the mineralogical and the geochemical properties of the lacustrine sediments and to discuss the reasons of the mineral's striking variety. For this, the samples from a borehole of ~23 m depth from the Sambhar Lake, available from

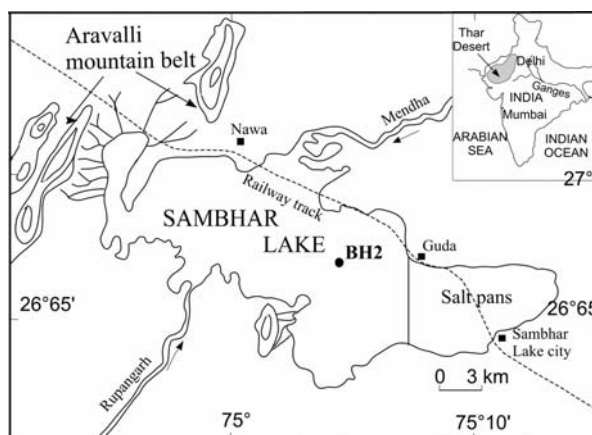


Fig. 1 Location of the Sambhar Lake playa in Thar desert, western India; BH2 is the site of deep borehole utilized for this study

earlier investigations (Sinha and Raymahashay [29, 30]), were analyzed for dolomite mineralogy and crystal chemistry.

Methods

The crystal chemical and crystal physical (=structural) properties of dolomites can well be characterized by X-ray and thermal methods. In XRD charts, the main interference at 1014 at 2.88 Å gets broader by additional ion substitution for Mg (e.g. Fe²⁺, Mn, Ni, Zn), and in case of Fe-rich variety ('ankerites'), it is shifted to higher d-values around 2.90 Å (Graf [32], Goldsmith *et al.* [33], Lippmann [2], Brindley and Brown [34]).

In differential thermal analysis (DTA), the decomposition of dolomite is mirrored by two endothermic effects representing the decomposition of MgCO₃ component around 800°C and the CaCO₃ component around 900°C (Cuthbert and Rowland [35], Haul and Heystek [36], Smykatz-Kloss [37, 40], Trdlička [38], Webb and Krüger [39], Smith *et al.* [41]). Graf [42] and Murray *et al.* [52] reported a decrease of about 30°C in these decomposition temperatures by the influence of salt solutions. Bradley *et al.* [44], Dasgupta [45], Smykatz-Kloss [40, 46, 47] and Warne and Dubrawski [48] investigated the interrelation of crystal chemical composition, mainly the substitution of Mg by Fe²⁺ and Mn²⁺ and thermal decomposition effects. Iron-bearing dolomites show a third endothermic peak before the MgCO₃ decomposition. The temperature of this third endotherm decreases with increasing Fe→Mg substitution (Smykatz-Kloss [40, 47]). Using standardized conditions of preparation and analysis, the determination of the FeCO₃ decomposition peak may be used for the determination of the incorporated iron (Smykatz-Kloss [40, 46, 47]).

In the present paper, the dolomite is characterized mainly by differential thermal analysis (DTA), using standard conditions of analysis as proposed by Smykatz-Kloss (1974), e.g. using heating rate of 10 K min⁻¹, Al₂O₃ crucibles, 100 mg sample amount, standing air for furnace atmosphere, and Pt/Pt₉₀Rh₁₀ thermocouples. The sam-

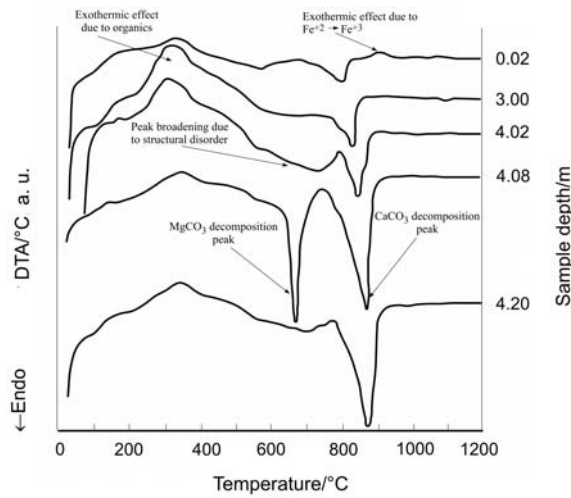


Fig. 2 Representative DTA curves for the samples rich in dolomite from upper 4.20 m of the BH2 core. Note that all samples, except at 4.08 m depth, show poorly ordered dolomites and high organic matter. The sample at 4.08 m depth shows well-ordered dolomite

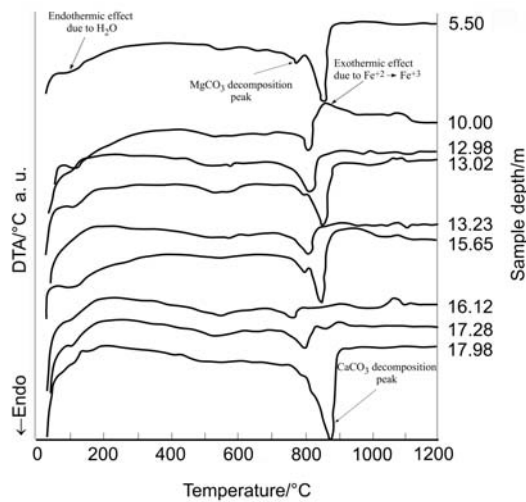


Fig. 3 Representative DTA curves for the bottom part of the BH2 core (below ~5 m). Note that most of the samples have poorly ordered, low Mg dolomite and low organic matter

ples were desalted by water dissolution and dried at 50°C for 2 days. After desalting, only silicates, carbonates and a part of gypsum remained in the samples which were analyzed for their thermal properties. The DTA curves obtained for the Sambhar Lake samples are shown in Figs 2 and 3. For comparison of DTA results, XRD charts were used (Smykatz-Kloss [40]).

Results

A total of 67 samples from the ~23 m deep borehole were analyzed for bulk mineralogy using XRD (Sinha *et al.* [31]). For the purpose of dolomite characterization, 16 samples, rich in dolomite, were selected and their bulk mineralogy is summarized in Table 1. The bulk samples exhibited over 10 different evaporite minerals amounting to about 40–50% of the sediments. Thermal properties of the desalted samples are summarized in Table 2. The major minerals identified on DTA charts of these samples are dolomite, calcite, quartz, and illitic mica.

The burning of organic matter creates a broad, and sometimes very intensive, exothermic peak (320–340°C), characterizing the samples of the uppermost 4 m of the profile (Tables 1 and 2, Fig. 2). The samples from the lower part of the core are generally devoid of organic matter. Around 500–600°C, the associated illitic mica and quartz cause a broad, generally not very intensive, endotherm.

The first endotherm of the decomposition of the studied dolomites is also quite broad (~700–800°C). In some samples (e.g. 4.02 m and 4.20 m depth, Fig. 2), this endotherm is even broader, starting at 600°C. The second endothermic effect due to CaCO₃ decomposition, i.e. between 800–870°C, is the cumulative effect of the decomposition of CaCO₃-component of dolomite and of the associated calcite. This peak is generally of higher intensity than the MgCO₃ decomposition peak in all samples. Shortly after the decomposition of (Fe-bearing) dolomite, a comparatively weak exothermic effect mirrors the oxidation of the Fe²⁺ to Fe³⁺ (e.g. samples at 0.02 m, 10.0 m and 15.65 m, Figs 2 and 3).

The amount of carbonates varies from 10 mass% at the surface to about 70% at ~4 m depth (Table 2). The dolomite structure is generally strongly disordered (except for one sample at 4.08 m) as is typical of (sub-) recent proto-dolomites. This is manifested in chemical disorder, too; some samples contain Fe²⁺ substituting Mg as may be seen in the typical exothermic peak (Figs 2 and 3). In the case of Fe-rich samples, the intensity of the MgCO₃ decomposition peak decreases.

The exceptional sample at 4.08 m shows a relatively well-ordered and Fe-free structure quite unusual for this profile. It is also striking that the first endotherm (representing the decomposition of MgCO₃ component) of this sample occurs at a temperature more than 100°C lower than that reported for 'normal' dolomites (Webb and Krüger [39], Smykatz-Kloss [40]).

Discussion

The fact that organic matter is abundant only in the uppermost 4 m of the profile indicates that the diagenetic processes destroyed the organic matter at deeper levels with increasing time, due to the effects of circulation of salty pore solutions. The character of the dolomite changed several times (as is mirrored in the DTA curves, Figs 2 and 3) from an iron-free and stoichiometric type e.g. CaMg(CO₃)₂ (samples at 4.08 m, 5.50 m and 15.65 m) to strongly disordered, Fe-bearing type e.g. Ca_{>1.0}(Mg,Fe)_{<1.0}(CO₃)₂ (samples 0.02 m, 4.02 m, 4.20 m, 10.0 m and 13.02 m). With the exception of 4.08 m sample which has well-ordered and iron-free dolomite, all other samples exhibit remarkable

Table 1 Bulk mineralogy as determined by XRD for the selected samples, Sambhar Lake playa, Thar desert, India

Depth/m	Qtz	Felds	Calcite	Dolomite	Gypsum	Bassanite	Halite	Polyhalite	Trace minerals except marked as*
0.02	++++	++	+++	+			++	+++	Glauberite, Pentahydrate, Starkeyite
3.00	++	+	++	++			++++		Anhydrite
4.02	+			++++			++++	+	Bloedite
4.08	+			++			+++		Aragonite, Bloedite
4.20	++++	+++	+	++			++		Aragonite*
5.50	++++		++	++++	+++		+++	++	Glauberite
10.00	+++	+	++	+++			+++		Stark/Mirabilite, Bloedite
12.98	++++		++	++	++		+++		
13.02	++++	+	++	++	+	+	++		
13.23	++++	+	++	+++			+++		
14.99	++++	+	+	+++	++		+++		
15.65	+++	++		++++	+++		++	++	
16.12	++++	++		++	+++	+	+++		Sylvite
17.00	++++	+	++	++			+++		
17.28	++++	+		++	+++				
17.98	++++	+	+++	+++			++		

++++ Very high; +++ Major component; ++ Minor component; + Trace

Table 2 Thermal characterization of Sambhar lake playa sediments, Thar desert, Rajasthan

Sample depth/m	Water/%	Organic matter (exo DTA effect)		Mica		Quartz		Carbonates		Exothermic effect due to $\text{Fe}^{2+} \rightarrow \text{Fe}^{3+}$	
		°C	μV	°C	$-\mu\text{V}$	Amt	Amount/%	Fe^{2+} in dolomite and crystallinity	°C	Amt	
0.02	1.8	340	3.3	550	2.4	+	10 (Cc*)		900	++	
3.00	3	335	16.5	*			17 (Cc+dol*)				
4.02	2	320	17.0	*			30 (dol)				
4.08		342	5.7	550	1.8		70 (dol)	Fe-poor, very well ordered			
4.20	1.2	330	6.5	550	1.5		70 (dol+Cc)	Fe-poor, disordered	945	+	
5.50	1.3			550	1.5		45-50 (dol+Cc)	Fe-poor, disordered			
10.00	3			550	1.7		10 (dol+Cc)	Fe-rich, disordered	870	+++	
12.98	0.8			540	2.0	++	15 (Cc+dol)	Fe-rich, disordered	900	++	

Table 2 Continued

Sample depth/m	Water/%	Organic matter (exo DTA effect)		Mica		Quartz		Carbonates		Exothermic effect due to $Fe^{2+} \rightarrow Fe^{3+}$	
		°C	μV	+°C	- μV	Amt	Amt	Amount/%	Fe^{2+} in dolomite and crystallinity		°C
13.02	3.2			550	2.7	++		50 (Cc*+dol*)	Fe-poor, disordered		
13.23	0.7			550	2.8	+++		15 (dol+Cc)	Fe-rich, disordered	900	++
14.99	0.6			550	2.8	++		20 (Cc+dol)	Fe-rich, disordered	910	+
15.65	1.3			550	1.2	+		30 (dol)	Fe-poor, disordered	903	+++
16.12	2.15			540	3.0	+		10 (dol)	Fe-rich, disordered	900	+
17.00	2.3			540	3.0	+		20 (Cc+dol)	Fe-rich, disordered	915	++
17.28	2.8			540	3.0	+		10 (dol)	Fe-rich, disordered	905	++
17.98	1.5			540	2.3	+		55 (Cc+dol)	Fe-rich, disordered	910	+

* Cc - calcite, dol - dolomite

structural disorder of the dolomite which may be caused by chemical substitution (by Fe, as mentioned above) or by physical effects (samples at 12.98 m, 13.23 m, 16.12 m, 17.28 m), as is typical of non-stoichiometric proto-dolomites. Such strongly (chemically and physically) disordered dolomites occur not only in very young, (sub-) recent inorganic marine muds as a natural precipitate and precursor of stoichiometric and well-ordered dolomite (as described by Skinner [8] and von der Borch *et al.* [9]) but can also form as an alteration product of well-ordered dolomite in continental (semi-) arid regions as well. Goebelbecker [49] and Smykatz-Kloss and Goebelbecker [50] described (secondary) proto-dolomites from Mesozoic carbonate rocks of southern Tunisia which are similar to the type observed in Thar desert (this paper). Both marine and continental types of these dolomites are characterized by:

a) the partial substitution of Mg by Fe^{2+} as manifested by the decrease of MgCO_3 component decomposition peak, and by the occurrence of a typical exothermic effect due to oxidation of Fe^{2+} to Fe^{3+} ;

b) a non-stoichiometric ratio of Ca:(Mg, Fe) e.g. $\text{Ca}_{>1.0}(\text{Mg,Fe})_{<1.0}(\text{CO}_3)_2$, as reflected in peak areas of the decomposition peaks; and

c) the broadening of all the decomposition peaks with increasing structural disorder.

Detailed geochemical data including stable isotope analysis of carbonate fraction are being published elsewhere (Sinha *et al.* [31]) but we would make use of some relevant data coupled with published literature to comment upon the origin of dolomite in the Sambhar Lake playa sediments. Firstly, it may be relevant to point out here that there are no carbonate rocks in the Aravalli source region (Heron [51], Sen and Ramalingam [52], Gupta *et al.* [53], Abu-Hamatteh [54]) of the Sambhar Lake playa. The catchment rocks also do not include any ultrabasic rocks which could yield high concentrations of Mg, and the pedogenic carbonates in alluvium and aeolian sediments in the region consist of low-Mg calcite. Therefore, dolomites in the Sambhar playa sediments must be authigenic, and the source of Mg-rich brine to favour dolomitization must be within the lake. Secondly, the present-day lake water is characterized by low Mg/Ca ratio (0.12, Yadav [55], 0.5–0.7, Bhattacharya *et al.* [56]). Similarly, the Mendha river feeding the lake from the NE and the groundwater in the region are characterized by Mg/Ca ratios of 1.56 and 1.61 which is still far too low to precipitate dolomite (Folk and Land [57]). It seems likely therefore that the brine was enriched in Mg by a secondary process such as rapid evaporation in hyperarid condition to favour dolomitization. Our conclusion is supported by the fact that all dolomite-rich samples are characterized by a high (MgO/MgO + CaO) ratio and high $\delta^{18}\text{O}$ values suggesting higher temperature during intense evaporation (Sinha *et al.* [31]). Thirdly, Table 1 shows that most of the dolomite-rich samples are poor in calcite and the samples below ~5 m are rich in gypsum. No gypsum occurs above ~5 m, and except the sample at 4.02 m, the amount of dolomite is relatively lower in upper horizons than in lower horizons. This indicates that the precipitation of gypsum at moderate salinity may be one of the mechanisms to remove Ca from the brine under arid conditions thereby increasing the Mg/Ca ratio. Fourthly, our geochemical data show that the dolomite-rich samples have higher concentrations of evaporitic components such as Sr and Br and low concentrations of clastic elements such as Ti, Rb and Zr. This again suggests a temperature influence (high salinity) and low clastic influx at the time of formation of dolomite. Finally, the dolomite rich samples also show a high mobile/immo-

bile element ratio (e.g. Na/Al, Na/Ti) for the clastic fraction. These effects are particularly enhanced at ~4 m (e.g. Sr 3332 ppm, Br 161 ppm, Zr 50 ppm at 4.02 m, Na/Al=0.68, Na/Ti=13.09), and interestingly, the samples are nearly free of calcite at this level. Instead, aragonite occurs as the dominant CaCO₃ mineral at 4.20 m and in trace in other samples.

The exceptionally well-ordered dolomite (4.08 m sample) exhibits some anomaly in its thermal decomposition, too. In spite of being well-ordered (as deduced from the relatively sharp endotherms of the decomposition), the first endotherm at ~660°C (Fig. 2), which mirrors the decomposition of the MgCO₃ component, occurs unusually early, more than 120°C lower than for 'normal' dolomites. The interpretation of the (standardized) DTA curve of this sample (4.08 m) leads to the following two questions:

i) What is the reason for the exceptionally well-ordered dolomite structure of this layer at 4.08 m among all the disordered dolomites of the profile?

ii) What is the reason for this extreme decrease in the decomposition temperature of the MgCO₃ component of the 4.08 m sample?

The first question remains largely unanswered except the presumption that the layer at 4.08 m depth came in contact with high energy (hydrothermal?) solution (circulating at the border of the layer?). To answer the second question, the study of the literature reporting DTA of dolomites may be useful. Among the numerous publications on the TA of dolomites, only three offer similar DTA curves. Smykatz-Kloss and Goebelbecker [50], early contributions of Murray *et al.* [43] and Graf [42] reported an interrelation between temperature decrease of this endotherm and a contact of the dolomite with salt-bearing solutions (mainly with NaCl). While Graf [42] and later Smykatz-Kloss and Goebelbecker [50] reported 'natural' dolomite occurrences from (semi) arid regions where dolomite is associated with halite (NaCl) as is the case for the Thar playa dolomites. Murray *et al.* [43] have conducted laboratory experiments to observe the changes in dolomite structure when brought in contact with artificial evaporitic solutions. Graf [42] found a decrease in the decomposition temperature of the MgCO₃ component by ~30°C. Murray *et al.* [43] found relations similar to our results e.g. a decrease of the MgCO₃ component decomposition temperature of more than 100°C compared with 'normal' dolomites. The decrease in decomposition temperature of MgCO₃ component of dolomites in the 4.08 m sample may thus be explained by contact with salt-bearing evaporitic or pore solutions which altered the structural stability of the MgCO₃ layers of the dolomites.

Conclusions

The characteristics of dolomite from the Sambhar Lake playa are variable in the vertical profile and are manifested very well in terms of thermal properties. The dolomites are generally disordered (proto-dolomite) except for one sample at 4.08 m which shows reasonably well-ordered structure, albeit with anomalous thermal properties. The presence of Fe affects the first endothermic peak of dolomite decomposition and is also manifested in a small exothermic peak mirroring the oxidation of Fe²⁺ to Fe³⁺. The origin of lacustrine dolomite in the Sambhar Lake playa sediments in the Thar

desert seems to be authigenic and perhaps derived from Mg-calcite. Although we can not confirm the derivation of (proto-) dolomite from Mg-calcite by means of the available data, there are indirect evidences which support our interpretation viz. absence of carbonate or high Mg rocks in the source area, low Mg/Ca ratio of lake, river and ground water, positive correlation of dolomite occurrence with $\delta^{18}\text{O}$ values and mobile/immobile element ratios. We conclude that the dolomitization of a precursor calcite has been favored by hyperarid conditions in the Thar desert, and this interpretation seems to be valid for (sub-) recent as well as older dolomites in the sequence.

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