Cenozoic coastal carbonate deposits of Qatar: Evidence for dolomite preservation bias in highly-arid systems

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ABSTRACT

In the ancient rock record, early replacement of metastable marine calcium carbonate deposits by dolomite has long been associated with evidence of arid depositional environments. Such associations led to the development of the seepage reflux dolomitization model, whereby magnesium-rich marine waters concentrated by evaporation descend into underlying sediments, replacing primary aragonite and calcite deposits with dolomite through rock–water interaction. In the modern arid coastal systems of Qatar, where marine waters concentrate to halite saturation, both aqueous geochemical measurements and mineralogical investigations show that replacement of calcium carbonate deposits by dolomite is not occurring to any consequential degree. At best dolomite formation is minor and localized. Instead, modern and mid-Holocene coastal deposits retain their original mineralogy, showing little evidence of carbonate precipitation reactions with the notable exception of beachrock formation. Pleistocene coastal deposits are mostly absent in comparison with both Qatar Holocene coastal deposits, and Pleistocene coastal deposits from more humid settings. The lack of onshore Pleistocene-aged carbonate outcrops in Qatar, as well as regionally, is interpreted to reflect coastal sediment denudation during emergence due to: (i) the absence of coastal marine lithification; and (ii) the absence of meteoric cementation and root stabilization in the highly-arid realm. In contrast, marine Palaeogene and Miocene carbonate deposits are preserved in outcrop in Qatar, having suffered marine lithification by dolomite, thus promoting retention of carbonate strata through subsequent lowstands. These older deposits formed during times of ocean acidification, which, based on natural system and laboratory investigations, is interpreted to promote metastable carbonate dissolution and dolomite formation. The highly-arid Holocene and Pleistocene coastal systems of Qatar represent limestone factories, but these deposits are not being retained to the rock record. Based on these observations, the association between the occurrence of dolomite and arid depositional settings may be to some degree a preservation bias.

Keywords Arid, dolomite, gypsum, saturation, seepage reflux.
INTRODUCTION

Dolomite is an important mineral of the sedimentary rock record, commonly inferred to have replaced marine calcium carbonate minerals, aragonite and calcite (Land, 1985; Morrow, 1998; Warren, 2000; Machel, 2004; Petrash et al., 2017). The general absence of dolomite in recent sedimentary deposits, as well as its variable presence in ancient sedimentary carbonates, and the difficulties of its laboratory synthesis at earth-surface temperatures, has caused consternation among earth scientists for over 100 years, and led to the coining of the term ‘dolomite problem’ (Van Tuyl, 1916). Explanations for the dichotomic nature of dolomite presence through time have not to this point moved towards consensus (Gregg et al., 2015; Petrash et al., 2017; Kaczmarek et al., 2017). The questions of whether large (1000s km²) platforms of marine carbonates suffer replacement by dolomite in near surface settings (e.g. Zentmyer et al., 2011) or require burial and extended periods of geological time (Ryb & Eiler, 2018) loom large in the literature. If dolomitization predominates in the near-surface, can platform-scale dolomitization occur quickly relative to modern sea-level fluctuations (Manche & Kaczmarek, 2019), or are flooded continents and long marine residence times required (Sibley, 1991; Nordeng & Sibley, 2003)? What is the role of ocean chemistry variability over geological time, specifically the importance of oxygen levels (Burns et al., 2000), Mg/Ca ratios (Folk & Land, 1975; Kaczmarek & Sibley, 2011) and carbonate saturation states (Given & Wilkinson, 1987)? Other factors have also been considered, including the role of microbes (Vasconcelos et al., 1995; Petrash et al., 2017) and the presence of particular depositional environments (Sun, 1994).

The long-observed association between arid depositional settings and early replacement of marine carbonates by dolomite led to it being termed an ‘evaporite mineral’ (Friedman, 1980). Such observations also brought about the development a central model for dolomite formation. Seepage reflux (Adams & Rhodes, 1960; Deffeyes et al., 1964), the downward movement of evaporatively-concentrated seawater into underlying sediments and rocks, has been cited in publications hundreds of times as a model for early-marine dolomitization of shallow-platform carbonate deposits. The model centres on the reaction between marine waters with properties thought to promote dolomitization including elevated temperatures, salinities and Mg/Ca ratios (Machel & Mountjoy, 1986; Morrow, 1990), with underlying skeletal and non-skeletal calcitic deposits. The reaction between concentrated seawater and calcium carbonates leading to dolomite formation in modern evaporative coastal settings, however, has not been commonly observed.

The peninsula of Qatar sits in a highly-arid desert setting, and its coastal waters form part of a ‘classic’ shallow-water photozoan–carbonate-producing realm westward of the ‘Great Pearl Bank’ (Wagner & Togt, 1973; Purser & Evans, 1973) (Fig. 1). Due to the lack of land plants and root formation, onshore sediments above the capillary fringe of the water table (i.e. above the Stokes surface – Stokes, 1968) have been mostly denuded by the ‘Shamal wind’ and transported to southern Qatar in dune complexes. As a result, most of central and northern Qatar is mantled by exposed Eocene and Miocene age carbonate rocks (Figs 1 and 2). Such aridity also causes waters in some coastal lagoons and tidal ponds to be concentrated beyond gypsum and even halite saturation (Purser, 1973; Rivers et al., 2019a). Studies of ongoing dolomitization in the coastal regions of Qatar have focused on sabkha environments (e.g. Illing et al., 1965; de Groot, 1973; Illing & Taylor, 1993) and associated microbial deposits (Paulo & Dittrich, 2013; Brauchli et al., 2016; Diloreto et al., 2019). However, no systematic investigation as to the speed and extent of dolomite formation in the Holocene coastal deposits of Qatar has been undertaken. In addition to the modern evaporative coastal marine system, Qatar hosts older Cenozoic carbonate deposits that formed in similar settings, including sediments and rocks of mid-Holocene (Shinn, 1973a; Billeaud et al., 2014; Strohmenger & Jameson, 2015; Purkis et al., 2017; Rivers et al., 2019b), Pleistocene (Holail, 1999; Williams & Walkden, 2002), Miocene (Dill et al., 2005) and Eocene (Cavelier et al., 1970; Abu-Zeid & Boukhary 1984; Abu-Zeid 1991; Boukhary & Alsharhan, 1998; Dill et al., 2003; Al-Saad, 2005; Rivers et al., 2019c; Ryan et al., 2019, 2020) age (Fig. 2). Whereas these units have been individually investigated, no comparative assessment as to the degree and nature of dolomite formation in these older deposits is available. Because these deposits represent both icehouse (Miocene to recent) and greenhouse (Eocene) periods, they can offer insight as to how the nature of dolomite formation compares across these system types.
This study relates a perspective of dolomite formation in Qatar based on observations that have been previously published. It reviews both evidence of dolomite precipitation in the modern coastal system (centred on water chemistry and sediment mineralogy), and its occurrence and interpreted mode of formation in equivalent deposits of the ancient rock record. The goal of this study is to understand whether the nature of modern shallow-marine dolomite formation is consistent with interpretations regarding dolomite formation in equivalent coastal deposits of the older rock record. An additional intention of this work is to explain the association between ancient evaporative depositional systems, and the formation of replacive dolomite, by constraining the timing and environment of this reaction.

CENOZOIC DOLOMITE FORMATION IN QATAR

Modern systems and mid-Holocene (Northgrippian) deposits

It has long been reported that protected coastal sabkha systems of Qatar are sites of dolomite formation (Illing et al., 1965; Illing & Taylor, 1993; Paulo & Dittrich, 2013; Brauchli et al.,...
Significant quantities of dolomite (>50% carbonate sediments) have only been found in muddy sediments of highly-evaporative upper intertidal zones in Sabkha Faishakh (Illing et al., 1965; Illing & Taylor, 1993; Fig. 1) and appear to be associated with dissolution of skeletal carbonate grains, replacement of aragonite mud and precipitation of gypsum. Scanning electron microscopy (SEM)-based observations show minor amounts of dolomite and are also found in the microbial mats of the lower intertidal zone of that sabkha (Brauchli et al., 2016). In the restricted Inner Lagoon of Khor Al Adaid (the Inland Sea, southern Qatar; Fig. 1), subtidal coastal marine waters range to 90 psu (Rivers et al., 2019a), overlie a carpet of aragonite mud (Rivers et al., 2020), where only scant dolomite is observed, and which is interpreted to be detrital in origin based on petrographic analysis (likely wind-blown Eocene-age material). Microbial mats ubiquitously form in the intertidal zone ringing the Inner Lagoon, and minor amounts of dolomite have been discovered forming as a primary precipitate (Diloreto et al., 2019). Isolated tidal ponds near Khor Al Adaid and in other protected coastal areas are likewise colonized by microbial mats and reach halite saturation during the summer (>350 psu). In spite of the minor dolomite formation described above, geochemical data show no significant deflection of Mg or Mg/Ca ratio in water samples taken from across the Khor Al Adaid lagoon, either from marine subtidal or intertidal pond settings (Rivers et al., 2019a). The only evidence of significant Mg loss was observed in an isolated coastal pond where meteoric waters filled a stranded marine salt flat during a rare flooding event (Rivers et al., 2019a), and where poorly-ordered dolomite was identified in aragonitic mud underlying the pond.

Coastal areas along the less-restricted windward margin of Qatar at Al Ruwais, as well as along the eastern coastline of Qatar, including Al Thakhira and Mesaieed, have been the subject of several sedimentological investigations (de Groot, 1973; Shinn, 1973a; Billeaud et al., 2014; Strohmenger & Jameson, 2015; Purkis et al., 2017; Rivers et al., 2019b; Fig. 1). Recent sedimentation in these areas commenced in the mid-Holocene (ca 6000 BP) (Northgrippian) when sea level rose during the ‘Flandrian transgression’ to ca 1.5 m higher than current level, before receding approximately 1500 years BP (Lokier et al., 2015). Along the eastern coast, Shinn (1973a) observed that dolomite in intertidal and nearshore carbonate deposits was eroded from Tertiary outcrops. Based on data from the Mesaieed sabkha south of Doha, de Groot (1973) concluded that dolomite is found only in ‘small amounts’ (0.2%), but that there was some evidence of Mg loss in the pore waters. Similarly, Strohmenger & Jameson (2015) concluded that there was an extensive overprint

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of gypsum cementation, but made no mention of modern dolomite formation in the Mesaieed sabkha. At Al Thakhira, dolomite observed in nearshore settings is interpreted to be derived from the weathering of Eocene rocks, and intertidal areas in particular are observed to have ‘very little’ dolomite (Billeaud et al., 2014). At Al Ruwais modern sedimentation associated with carbonate barrier islands was shown to include minor amounts of dolomite (2%) that were ‘solely associated’ with Eocene bedrock erosion (Purkis et al., 2017). Most recently, Rivers et al. (2019b) showed that mid-Holocene deposits are mostly formed of aragonite and calcite (Fig. 3A), and what little dolomite is observed is derived from underlying Eocene rocks. Notably, stromatolitic deposits there, interpreted to have formed in the highstand intertidal zone, were lithified and cemented by micritic high-Mg calcite cement, with no evidence of active dolomitization.

In Qatar the vast majority of nearshore modern and Holocene sediments are uncemented. Minor exceptions include thin (centimetre-thick) intervals of cemented/lithified sediments at the base of tidal channels (Shinn, 1973b), or intermittently associated with back barrier mid-Holocene deposits such as the stromatolites referenced above at Al Ruwais (Rivers et al., 2019b). The only major exception is beachrock cementation (Fig. 3A), which is common along beaches and spits that formed in the mid-Holocene and today (Shinn, 1973b; Billeaud et al., 2014; Rivers et al., 2019b). This observation stands in contrast to those of deeper subtidal areas in the Arabian Gulf, where centimetre to decimetre-thick layers lithified by marine cementation are common (Shinn, 1969; Purkis et al., 2005).

### Pleistocene rocks

Rare Pleistocene outcrops (Fuwayrit Formation) are observed in Qatar (making up less than 1% of the surface area below 25 m elevation), with most lying intermittently along coastal areas (Cavelier et al., 1970; Williams & Walkden, 2002; Fig. 1). These rocks are in large part interpreted to be aeolian deposits (Holail, 1999; Williams & Walkden, 2002), characterized as undolomitized ooid grainstones with metre-thick cross-beds, and cemented by low-Mg crystalline calcite. However, undolomitized Pleistocene marine deposits (Fig. 3B) are observed in Qatar, at elevations consistent with the marine isotope

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**Fig. 3.** Representative thin-section images of Qatar rock samples taken from: (A) a mid-Holocene (Northgrippian) outcrop; (B) the Pleistocene Fuwayrit Formation; (C) the Miocene Dam Formation; and (D) the Eocene Rus Formation. See references in text for detailed descriptions of these deposits. X-ray diffraction (XRD)-based mineralogy for each sample is shown above the images (‘Q’ quartz; ‘A’ aragonite; ‘C’ calcite; ‘D’ dolomite; ‘Cl’ clay). In sample (B), ‘D’ indicates detrital dolomite, and the red arrows point to micritic meniscus cements that are also commonly observed associated with modern beachrock deposits. Sample (D) includes ostracods (OST), as well as small miliolid (ML) and rotalid (RT) foraminifera, and is interpreted to represent a restricted lagoonal assemblage. Equivalent Holocene age lagoonal deposits are uncemented, and Pleistocene-age lagoonal deposits are missing and inferred to have been denuded during lowstand by wind energy.

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stage 5 (MIS 5) highstand (Williams & Walkden, 2002). In southern Qatar, many Pleistocene outcrops (Fig. 4) are capped by planar and landward-dipping cross-bedded mollusc and ooid-rich grainstones, that overlie bioturbated packstones. These outcrops, which are typically a few tens of metres wide, less than 5 m high, but kilometre-scale in length (Fig. 5), are interpreted here to represent transgressive barrier islands or land-attached spits, with landward-dipping cross-beds formed by barrier washover processes. They appear exactly analogous to barrier island deposits found to have formed over the past 6000 years at Al Ruwais, along the windward margin of Qatar, which are lithified by beachrock-type cementation (Rivers et al., 2019b). Stained thin-sections from several of the Pleistocene marine outcrops confirm that these rocks are not dolomitized to any observable degree, and demonstrate that they bear micritic meniscus-type calcite cements (Fig. 3B), identical to Holocene beachrock cements.

Both Chafetz et al. (1988) and Williams (1999) reported dolomitized carbonate strata in cores taken from offshore areas of the southern Arabian Gulf. The dolomite-rich intervals are typically greater than 10 m thick, and occur at depths of generally >10 m below the seafloor. Chafetz et al. (1988) interpreted these deposits to be of Pleistocene age, based on a single bulk rock $^{14}$C measurement that yielded an age of approximately 37 kyr before present, but also noted that some of these rocks might be older than Pleistocene. Based on bathymetric data compared with established sea-level curves, Williams (1999) likewise inferred a Pleistocene age for similar offshore dolomitized strata. More recently, however, Buchem et al. (2014) showed that seismically-thick sections of offshore but near-surface strata that onlap an Eocene-age unconformity north of Qatar, are of the Miocene Dam Formation. The offshore dolomitized deposits described by Chafetz et al. (1988) and Williams (1999) are similar to those described by Dill et al. (2005) for the Miocene Dam Formation onshore, bearing gypsum layers and having similar bulk stable isotope values (Chafetz & Rush, 1994; Dill et al., 2005). The accuracy of $^{14}$C measurements yielding dates of greater than 25 000 years is dubious (Schellmann & Radtke, 1997), and minor contamination by more recent material can cause samples older than 50 000 years to yield younger age dates (Burr et al., 1992; Rosenberg et al., 2013). It is therefore considered more likely that dolomitized intervals described by Chafetz et al. (1988) and Williams (1999) are of Miocene age.

### Miocene and Eocene rocks

Extensive shallow marine clastics, carbonates and, more locally, evaporites were deposited within the Zagros foreland basin during the Cenozoic (Ziegler, 2001). In Qatar these deposits include thick (>300 m) sections of Palaeocene–Eocene strata (Umm Er Radhuma, Rus and Dam-mam formations) and more localized and

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**Fig. 4.** Pleistocene outcrop in southern Qatar interpreted to represent barrier island or coastal spit deposits. Note the cross-bedding, interpreted to indicate landward migration of a barrier beach over a lagoon by washover processes. Person for scale is ca 1.8 m tall. See Fig. 1 for location.

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thinner (<70 m) Miocene deposits (Dam Formation). Investigations of the Umm Er Radhuma, Rus and Dam formations show that they are pervasively dolomitized (Figs 2, 3C and 4D), whereas the middle Eocene Dammam Formation is partially dolomitized (Dill et al., 2005; Rivers et al., 2019c; Ryan et al., 2019, 2020). The prior studies of the three pervasively-dolomitized formations indicated that initial dolomitization likely occurred very early as a syn-depositional diagenetic phase, with dolomite replacing marine calcium carbonate deposits prior to exposure-related diagenesis. Meteoric or mixed meteoric-marine cements are without exception interpreted to have formed after dolomite formation (e.g. Dill et al., 2005; Ryan et al., 2020). Metre-thick gypsum deposits are interbedded with the dolomitized rocks of the Rus Formation in southern Qatar (Al-Saad, 2003) and decimetre-thick gypsum beds are locally present in the Dam Formation (Dill et al., 2005). The presence of dolomite in these rocks, however, is ubiquitous, and so does not correlate well with the presence of evaporites, or even with evidence of shallow-restricted environments, where concentrated seawater might be expected (Rivers et al., 2019c; Ryan et al., 2020). In contrast, bulk isotope and cross-cutting relationships in the partially-dolomitized Eocene Dammam Formation point to dolomite replacement after stabilization to low Mg-calcite in meteoric waters. Furthermore, relative to the underlying dolomitized Eocene deposits, the Dammam appears to have suffered karstification between deposition of member units, perhaps suggesting stabilization during a more humid period (Rivers et al., 2019c). Notably, the Eocene and Miocene dolomitization occurred on a basin-wide scale (Mukhopadhyay et al., 1996; Fox & Brown, 1968; Ziegler, 2001; Saller et al., 2014). Furthermore, these rocks are above 400 m depth in Qatar (Rivers et al., 2019c) and are not interpreted to have suffered significant burial, with Eocene rocks being exposed during the Oligocene (van Bucham et al., 2014), and later capped only locally by up to ca 90 m of Miocene rocks (Dill et al., 2005) which have since undergone uplift (Rivers & Larson, 2018).

**INTERPRETATION**

Sediment-buffered waters (cf. Higgins et al., 2018) of modern marine systems in which ongoing dolomitization is observed, can show associated reduction in Mg and Mg/Ca ratios (e.g. Compton, 1992; Eberli et al., 1997; Rivers et al., 2012). In Qatar, Rivers et al. (2019a) interpreted ongoing dolomite formation in an unusual coastal pond. In this example, a marine salt flat was flooded by meteoric waters, and low Mg levels and Mg/Ca ratios accompanied the presence of poorly-ordered dolomite in the sediment. Based on the above studies, and consistent with laboratory results (Kaczmarek &
Sibley, 2011), small amounts (ca 5% replacement) of ongoing dolomite formation in near-surface sediment is expected to cause deflection of Mg and Mg/Ca ratio values in the immediately overlying seawater, at least in isolated and restricted settings such as the Inner Lagoon of Khor Al Adaïd in southern Qatar, and surrounding tidal ponds. Measurements of Qatar coastal marine Mg levels and Mg/Ca ratios, however, show no evidence of ongoing dolomitization to even this degree. Indeed, evidence indicates that calcium carbonate precipitation and dissolution is fairly minor in waters concentrating from near-normal marine to halite saturation (Rivers et al., 2019a). This geochemical interpretation is consistent with the lack of sedimentary evidence for widespread dolomitization or even significant carbonate precipitation (i.e. cementation) in coastal Qatar deposits, with the exception of beachrock formation.

The paucity of sediment lithification in evaporative Holocene coastal carbonates has important implications. For instance, un cemented deposits could easily be denuded by wind as the Stokes surface falls with sea level during initiation of glacial periods. This inference is supported by the overall lack of Pleistocene deposits in the coastal areas of Qatar relative to modern equivalents. Yet sparse deposits exist at approximately the correct elevation for the MIS 5 highstand (Fig. 6A), indicating that the Pleistocene ocean did reach the modern Qatar coastal zone at least once, and likely many times. Many of the Pleistocene-age coastal marine rocks that are retained are capped by beach deposits, which appear to have suffered the same beachrock cementation as Holocene equivalents (Fig. 3B). Such early lithification was likely a key step in their eventual retention in the face of wind-related denudation processes during subsequent sea-level fall and exposure. The associated lagoonal and seaward near-shore ramp deposits, the facies exhibiting little evidence of cementation in the modern, have been entirely removed (Fig. 5).

Contrasting the observations from the Holocene and Pleistocene systems, are those of the Qatar Miocene and Eocene deposits. Rocks comprising these strata are interpreted to record prolonged highstand sedimentation in near-shore lagoonal and tidal-flat environments (Dill et al., 2005; Al-Saad, 2003). Some of these rocks include bedded evaporites, and so doubtless were formed in evaporative settings of similar latitude as today (Marlow et al., 2014). Why these ancient deposits were retained into the rock record, given the lack of retention observed for similar deposits associated with such settings of the recent past, is an important question. Two possibilities are envisioned. First, the deposition of bedded crystalline evaporites may serve to protect underlying carbonate grains from aeolian transport during lowstand. In the Qatar rock record, however, bedded evaporite deposits are only locally observed. In the Eocene deposits, for example, bedded gypsum is present only in an isolated basin that included the area of southern Qatar (Al-Saad, 2003). Equivalent strata in northern Qatar bear no bedded evaporites, although their proximity to the southern basin as well as their palaeo-latitude dictates that they too formed in an arid setting. Second, and the more favoured interpretation, is that the process of synsedimentary dolomitization that these Eocene and Miocene strata appear to have undergone prior to exposure (Dill et al., 2005; Ryan et al., 2020) caused their lithification, through the formation of interlocking diagenetic dolomite crystals (Rivers & Kaczmarek, 2020). This interpretation is supported by observations of deposits from restricted near-surface modern systems where consequential dolomitization is occurring. For instance, Deffeyes et al. (1964) refers to the supratidal dolomites (up to 95% pure) of Bonaire as ‘crusts’ lithified enough to form ‘clasts’ and host vugs. In Qatar, Illing et al. (1965) refers to the dolomitized muds of the Faishakh Sabkha of Qatar as being ‘stiff’ and capable of hosting moulds. Dravis & Wanless (2018) show that dolomite replacement of lime sediment below a coastal salina on West Caicos Island is undoubtedly a lithifying process.

If the preferred interpretation is correct, a consequence of significant synsedimentary dolomitization is early lithification of highstand deposits, and a consequence of such lithification is preservation during subsequent lowstand, and ultimately rock record retention, when exposure-related denudation would otherwise occur. Thus, the association between highly-arid depositional environments (i.e. lacking land plants) and dolomite replacement in the rock record may not be primarily causal, but instead reflect preservation bias. If the present is the key to the past, it would appear that there are geological times, most notably the Holocene and Pleistocene, but also likely others, when evaporated seawater in Qatar’s nearshore carbonate systems did not react with sediment and cause significant dolomitization. These un lithified lime
deposits, however, were largely lost to the rock record during subsequent exposure.

**DISCUSSION**

**Speed, degree and depositional environment of ancient syn-sedimentary dolomitization in Qatar**

Constraints can be surmised as to how quickly dolomite formed in the ancient highly-arid coastal systems of Qatar, since dolomite formation sufficient for lithification must occur prior to exposure. The amount of dolomite replacement required for lithification may depend on the texture of the rock if, for instance, interlocking dolomite crystals are concentrated at grain boundaries in carbonate sands. Fine-grained slope sediments from southern Australia are mostly between 5% and 10% dolomite, and remain unlithified (Rivers et al., 2012). Similarly the common presence of between 4% and 12% dolomite in the modern sabkha of Abu Dhabi does not cause lithification (Evans et al., 1969; McKenzie et al., 1980). Alternatively, the dolomitic ‘crusts’ of Bonaire (Deffeyes et al., 1964) are between 80% and 95% dolomite, forming lithified pellet-bearing carbonate sands and muds. Carballo & Land (1987) reported that the thickness of dolomite crusts forming in Holocene sediments of Sugarloaf Key, Florida, increased with percentage of dolomite present (up to 80% dolomite in some locations), and

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Fig. 6. Comparison of the extent of Pleistocene deposits in: (A) the highly-arid setting of Qatar (Cavelier et al., 1970; Seltrust Engineering Ltd., 1980); versus the more humid settings of (B) San Salvador Island (after Robinson & Davis, 1999) and (C) Okinawa Island Japan (after Sagae et al., 2012). Location of map (A) is shown on Fig. 1. The coastal area of Qatar and almost the entirety of San Salvador Island lie at elevations of less than 25 m above sea level. Recent uplift in Okinawa has resulted in Pleistocene deposits there (Ryukyu Group) being predominantly greater than 25 m above sea level (Kizaki, 1986).
that lithification was absent in undolomitized sediments. Observations from high temperature experiments where dolomite is synthesized from calcite and aragonite powder (e.g., Kaczmarek & Sibley, 2007, 2014) show that lithification initiates at ca 30% conversion to dolomite, and is complete at ca 70% conversion. All of these observations would seem to point to a significant degree of dolomite replacement (>50%) being required for effective lithification, although this topic clearly requires further investigation.

The denudation ‘problem’ points to early lithification due to dolomite formation in highly-arid settings occurring on a cycle-by-cycle basis, as opposed to multiple cycles being dolomitized downward from a master surface (Manche & Kaczmarek, 2019). That is, to prevent denudation in such settings, dolomitization of one depositional cycle must occur before subsequent exposure. For most coastal deposits, exposure would presumably be caused by sea-level falls associated with Milinkovic cycles (Hays et al., 1976; Hinnov, 2013). Cyclostratigraphic analysis reveals that the Oligo-Miocene includes 41 kyr cycles associated with obliquity and 100 kyr cycles associated with eccentricity (Zachos et al., 2001; Holbourn et al., 2007). The rapid dolomite formation required for lithification prior to exposure caused by such cyclic sea-level change is consistent with the relatively short dolomite induction periods (<2000 years at 30°C) inferred based on extrapolation calculations from recent laboratory growth studies (Kaczmarek & Thornton, 2017).

The presence or absence of dolomite in the rock record of Qatar does not correlate well with any particular set of depositional environments. Synsedimentary dolomite replacement is interpreted to have occurred to sediments in ancient environments ranging from intertidal to open ramp (Ryan et al., 2020), and within both evaporite-bearing and evaporite-free basins (Rivers et al., 2019c). Conversely, since at least the Pleistocene, dolomite replacement of Qatar coastal sediment in environments ranging from highly-restricted lagoons to open-marine settings has been largely absent. This general lack of association between depositional environments and the presence or absence of dolomite runs counter to expectations assuming a seepage reflux model, where dolomite is expected to form preferentially in the presence of marine waters concentrated by evaporation, ostensibly in near-shore lagoonal environments, perhaps colonized by microbial mat communities tolerant of extremely saline conditions. Additionally, many Cenozoic dolomites are interpreted to have formed in unvegetated seawater (Budd, 1997).

In summary, the presence of dolomite in the ancient Cenozoic deposits of Qatar and, conversely, its absence in more recent deposits cannot be well explained either by significant differences in the amount of time available for the reaction to occur, or by any obvious differences in depositional environments. It is therefore deduced that the present ocean does not have the capacity to dolomitize nearshore sediments with the rapidity or to the degree it did in some ages of the geological past, a conclusion reached by many other investigators based on other lines of evidence (Daly, 1909; Schmoker et al., 1985; Given & Wilkinson, 1987; Sibley, 1991; Stanley & Hardie, 1999; Burns et al., 2000; Holland & Zimmerman, 2000).

Global applicability of Qatar-based observations

As in Qatar, Pleistocene-age highstand coastal carbonate deposits are generally absent along the southern margin of the Arabian Gulf. Williams & Walkden (2002) reported that onshore Pleistocene outcrops of the southern Arabian Gulf are ‘scattered’ and ‘rare’ but have a wide distribution. Pleistocene deposits along the arid Red Sea coastline (Plaziat et al., 1998) and in the Gulf of Aqaba (Bar et al., 2018) consist mainly of reefs and units of beachrock able to resist subaerial erosion. Erosion of unlithified beach and reef-flat sediment has been noted (Plaziat et al., 1998), and it is possible that fluval processes during glacial periods contributed to removal of back reef deposits (Gvirtzman et al., 1977). In southernmost Tunisia, the lack of Pleistocene deposits (Ben Hadj Ali, 1985) correlates with the presence of arid settings (Gouta et al., 2019). In the arid climate of south-western Australia, Feary & James (1998) demonstrated that Pleistocene-age inner shelf deposits of the Great Australian Bight are generally absent, perhaps removed by ravinement. By contrast, in more humid settings such as San Salvador Island in the Bahamas (Fig. 6B) (Robinson & Davis, 1999), the island of Bermuda (Vacher et al., 1989), the Yucatan Peninsula (Szabo et al., 1978) and the Ryukyu Islands of Japan (Fig. 6C) (Sagae et al., 2012), deposits of carbonate rocks from multiple Pleistocene marine isotope stages veneer the surface kilometres inland.
from the coast. Doubtless there are exceptions and uncertainties with regard to palaeoclimate, but it may be a general hallmark of highly-arid settings that near-shore highstand carbonate deposits are not retained, but largely denuded by aeolian or possibly ravinement processes as a result of sea-level change.

Although the timing of initial dolomite replacement in the rock record is commonly equivocal, many ancient dolomites associated with evaporite deposits are interpreted to be of very early diagenetic origin (Warren, 2000; Machel, 2004). Examples of very early limestone replacement by dolomite in evaporite-forming coastal settings include the Permian rocks of West Texas, USA (Saller & Henderson, 1998) and Iran (Rahimpour-Bonab et al., 2010), the Jurassic rocks of the Arab D reservoir (Cantrell et al., 2014) in Saudi Arabia, and the Cretaceous carbonates of the Upper Glen Rose Formation in Central Texas (Manche & Kaczmarek, 2019). Therefore, the interpretation of synsedimentary timing for dolomite formation in the evaporative Eocene and Miocene rocks of Qatar (Dill et al., 2005; Ryan et al., 2020) is by no means unusual, and the evidence pointing to a retention bias for dolomitized rocks relative to undolomitized equivalents in highly arid coastal depositional systems, past and present, is generally consistent with Qatar-based observations.

**Inferring the drive for dolomite formation in ancient oceans of Qatar**

It is notable that in spite of significant differences between the Eocene and Miocene oceans in absolute sea level (Kominz et al., 1998), Mg concentration (Holland & Zimmerman, 2000; Gothmann et al., 2017) and sea water temperature (Hansen et al., 2013), regional platform-scale dolomitization was able to proceed to similar degrees in the Zagros foreland basin. In a review of global dolomite volume assessments, Holland & Zimmerman (2000) postulated that increased dolomite formation in greenhouse systems, which likely caused associated low Mg/Ca ratios, resulted from the widespread occurrence of calcium carbonate reacting with concentrated seawater on flooded continents. Whereas refluxing waters of various concentration may contribute to early dolomitization, the presence of concentrated marine waters, which doubtlessly refluxed into the Holocene and Pleistocene coastal sediments immediately below, does not in itself cause dolomitization. Based on the modelled atmospheric oxygen levels of Lasaga (1989), Burns et al. (2000) argued that extensive dolomitization in the geological past was promoted by the greater presence of anaerobic microbes during times of low atmospheric O₂. Whereas this possibility in a general sense cannot be discounted, oxygen levels during most of the Cenozoic were higher than today (Lasaga, 1989; Berner, 2009), making this a less likely explanation for Qatar Eocene and Miocene platform dolomites. Qatar platform dolomites also indicate that burial and long geological time frames (Ryb & Eiler, 2018) are not required for platform-scale dolomitization. The common preservation of delicate depositional structures, such as fenestral pores in early dolomites, lends supporting evidence to significant dolomitization prior to virtually any burial (Morrow, 1990; Manche & Kaczmarek, 2019).

One important difference between the recent and ancient Cenozoic oceans is saturation state. The dolomitized Umm Er Radhuma and Rus formations of Qatar formed in the late Palaeocene and Early Eocene (Rivers et al., 2019c), which corresponds with the timing the Palaeocene–Eocene Thermal Maximum (Kennett & Stott, 1991; Koch et al., 1992), an event associated with ocean acidification (Penman et al., 2014) and warming of surface waters. Similarly, the Miocene Dam Formation formed in the Burdigalian, a stage that included the Middle Miocene Climate Optimum (MMCO). Ocean acidification during the MMCO resulted in the lowest aragonite saturation state for marine surface waters in the last 22 Myr (Sosdian et al., 2018). Given & Wilkinson (1987), Wilkinson & Algeo (1989) and Koch et al. (1992) concluded that low carbonate saturation states promote dolomitization during greenhouse periods. The association between low carbonate saturation states, ocean acidification and dolomite formation, was also noted by Mackenzie & Andersson (2013). In contrast, Burns et al. (2000), argued against the importance of low carbonate saturation states in explaining dolomite formation during greenhouse times, contending that: “pCO₂ level has no role in determining calcite saturation”. Studies show that rising atmospheric CO₂ levels, however, do cause lowering of marine carbonate saturation states, and result in carbonate dissolution (Feely et al., 2004). Similarly, relative to warm water settings, surficial marine waters of the cool-water carbonate realm have lower carbonate saturation states (James et al., 2005). This, in turn, leads to
relatively lower marine pore-water saturation states, which through dissolution, result in wholesale conversion of a mostly aragonitic depositional assemblage, to a dominantly calcitic one within the Milinkovic-cycle time frame (Rivers et al., 2008).

The potential influence of low carbonate saturation states on dolomite formation is apparent when the reaction of limestone replacement by dolomite is considered. Because low-temperature solid-state diffusion can be discounted as a mode for rapid dolomite replacement of limestone (Fisler & Cygan, 1999), the reaction should be envisioned as separate calcium carbonate dissolution and dolomite precipitation reactions, with the processes, even if they occur across a thin envelope of water (e.g. Wardlaw et al., 1978), written in two steps:

\[
\text{CaCO}_3 \rightarrow \text{Ca}^{2+} + \text{CO}_3^{2-}
\]

(1)

\[
\text{Ca}^{2+} + \text{Mg}^{2+} + 2\text{CO}_3^{-} \rightarrow \text{CaMg(CO}_3\text{)}_2
\]

(2)

(note that if the carbonate ion is sourced solely from Eq. 2, an excess of Ca\( ^{2+} \) will remain). Undersaturation with respect to metastable calcium carbonate minerals undoubtedly can drive dissolution in step 1. Although the force of crystallization (Maliva & Siever, 1988) cannot be discounted as an additional driver for calcium carbonate dissolution, sustained undersaturation with respect to \( \text{CaCO}_3 \) may be critical for conversion of limestone to dolostone in geologically-short time frames. Theoretically such undersaturation can be self-generated initially by dolomite precipitation as a cement or as an overgrowth on detrital dolomite, lowering the carbonate ion concentration of the fluid. This might lead to dissolution of metastable carbonate that can source dolomite crystallization, continuing the cycle. Precipitation of dolomite in the modern arid coastal system of Qatar is not broadly observed, however, likely being inhibited, at least in part, because the waters are highly saturated with respect to calcite (12–43×) and aragonite (4–20×) (Rivers et al., 2019a). These metastable minerals can form more quickly than dolomite when conditions promoting carbonate mineral formation and growth arise (Morse & Mackenzie, 1990). It would seem more likely that initial undersaturation with respect to metastable carbonate minerals is typical for dolomitization (Murray, 1960). Such undersaturation, observed to result from a lowering of pH in seawater when concentrated to gypsum saturation (Lazar et al., 1983), was envisioned by Sun (1992) as a driver for seepage reflux dolomitization. Additionally, maintaining undersaturation with respect to the \( \text{CaCO}_3 \) precursors during influx of Mg-bearing marine waters, may require externally-forced low carbonate saturation states. For instance, small quantities of dolomite are formed in association with an acid-producing (\( \text{H}_2\text{S}-\text{oxidizing} \)) reaction zone in southern Australian slope sediments (Rivers et al., 2012). With increasing depth below this zone, low-Mg calcite replaces dolomite as a diagenetic product (see fig. 3B, Rivers et al., 2012), which likely reflects increasing carbonate saturation states away from the \( \text{H}_2\text{S} \) oxidation zone, leading to a higher degree of saturation with respect to calcite. In spite of dolomite nucleation having been achieved, growth of dolomite does not continue, and Mg levels begin to recover, perhaps pointing to the importance of acidified seawater for both nucleation and growth of dolomite. A very similar system of limited dolomite formation that is not sustained with depth is observed in Ocean Drilling Program (ODP) Hole 1005, on the western margin of the Great Bahama Bank (Eberli et al., 1997). It is also notable that, in the southern Australian system, lime sediment above the acidified reaction zone has been bathed in near-normal marine pore waters for about 130 kyr based on time-averaged sedimentation rates (Feary et al., 2000), and no dolomite formation, replacive or otherwise, has occurred.

Whatever the cause, evidence of undersaturation with respect to metastable carbonate during replacive dolomite formation is apparent in many modern evaporative systems (Deffeyes et al., 1964; Illing et al., 1965; Dravis & Wanless, 2018). Deffeyes et al. (1964) states of the Bonaire dolomite crusts: “The fact that leached pellets and shells are found only in dolomite crust suggests that dissolution is part of the dolomitization process”. Illing et al. (1965) reached the same conclusion after observing ‘leaching’ of calcite and aragonite allochems in the Qatar Sabkha. Dravis & Wanless (2018) showed that rocks formed due to seepage reflux on Caicos Island included moulds of dissolved bivalve shells. Manche & Kaczmarek (2019) also reported mouldic pores in the early formed dolomites of the Upper Glen Rose Formation in central Texas. Mouldic pores cannot be formed by dissolution through force of crystallization. Dolomitized rocks forming the ancient near-surface rock record of Qatar are interpreted to have formed early and in
evaporated seawater (Dill et al., 2005), and show identical dissolution of aragonitic allochems and mouldic porosity (Rivers et al., 2019c). Additionally, abiotic dolomite has been formed in laboratory conditions at earth surface temperatures in several experiments involving calcite dissolution by pulsed CO₂ flood (Liebermann, 1967; Deelman, 1999; Hobbs & Xu, 2020). Sibley (1990) also showed faster dolomitization rates in high-temperature experiments when CO₂ was bubbled through fluids prior to reaction when compared to N₂ control.

Finally, whereas surface seawaters during geological times of elevated CO₂ remained saturated with respect to metastable carbonate minerals, rising carbonate compensation depths (e.g. Rea & Lyle, 2005) indicate that less geochemical work was likely required for the fluids to become undersaturated with respect to aragonite and calcite. Therefore, acidification resulting from evaporative seawater concentration (Lazar et al., 1983), or from aerobic or anaerobic organic matter oxidation in underlying pore waters (Sanders, 2003), may have caused more sustained undersaturation with respect to metastable carbonate minerals relative to the modern ocean–sediment system. Such externally-forced dissolution and associated dolomite precipitation may not be an equilibrium process. For instance, at equilibrium dolomite should be isotopically enriched with respect to ¹³C relative to co-occurring calcite, and have δ¹³C values positively offset by 2.2‰ (Sheppard & Schwarcz, 1970). In the southern Australian slope system, below the acidified reaction zone, there is no observed ¹³C fractionation between bulk carbonate (mostly low-Mg calcite) and dolomite separates (Rivers et al., 2012), indicating that dolomite crystal growth is isotopically-indiscriminant, and not in equilibrium. Ultimately, some platform-scale synsedimentary dolomitization events may be the result of marine pore water disequilibrium forced in part by atmospheric CO₂-related acidification.

CONCLUSIONS

With minor exceptions, the mid to late-Holocene coastal skeletal deposits of Qatar remain mostly unlithified. Similar lack of marine cementation, as well as a general absence of meteoric cementation and root formation in this highly arid setting, likely permitted wind-related denudation of highstand Pleistocene deposits, explaining their general absence relative to more humid settings. In Qatar, rare outcrops of marine Pleistocene deposits bear no evidence of dolomitization, and appear to have been initially cemented by beachrock-type lithification, allowing their preservation during exposure. In contrast, the Eocene and Miocene rocks of Qatar are dolomitized, but otherwise similar in terms of depositional features and settings to uncedmented examples observed in modern coastal areas. The retention rather than denudation of these older deposits is interpreted to reflect the synsedimentary lithification on a cycle-by-cycle basis due to the dolomitizing process. Because, in highly-arid settings, undolomitized and thus un lithified carbonate sediments are removed from the rock record by aeolian processes during meteoric exposure, the long-observed correlation between arid deposition settings and the presence of synsedimentary dolomite is best explained as a preservation bias. The age of the older Cenozoic rocks in Qatar suggests that they formed during times of sustained ocean acidification. Their early-marine dolomitization may have resulted from concomitant lower carbonate saturation states and metastable carbonate dissolution, which have been shown to promote dolomite formation in both natural and laboratory settings.

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