Neogene carbonate platform development in the NW South China Sea: litho-, bio- and chemo-stratigraphic evidence

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Abstract: The variations of lithostratigraphy, foraminifera, geochemical elements, and organic biomarkers in well XK1 from the Xisha (Paracel) Islands reveal the history of carbonate platform development since the early Miocene in the South China Sea. Our results indicate that reef growth was more active in the middle Miocene than in any earlier periods, decreased in the late Miocene, revitalized in the Pliocene, and active again in the Pleistocene interglacial periods. Unlike those limestone-rich intervals, the late Miocene succession in well XK1 is characterized by dolomitized limestones with a low deposition rate (30-60 m/Ma post-compaction). These late Miocene rocks also contain relative high MgO (~20%), more Σ REE, abundant algae and some small sized foraminifera, but rare larger benthic foraminifera, very low land-sourced brGDGTs (0-0.5 ng/20g), low BIT index (0.1-0.5), and low TEX86-derived sea temperature (~20-25°C), indicating limited active reef growth in relative cooler mesophotic environments at >30 m water depths. Similar dolomitized limestones of late Miocene age also occur in other carbonate platform areas including the Nansha (Spratly) Islands in the southern South China Sea. Together, these South China Sea records provide regional evidence of a recession in global carbonate platform development starting from the late Miocene. While carbonate platform reduction in other regions may have been triggered by global cooling and changing oceanic chemistry, accelerated basin subsidence coupled with monsoon cooling are considered as the main factors causing the reef decline in the late Miocene South China Sea.

Key words: South China Sea; Xisha Islands; reef carbonate platform; geochemical elements; biomarkers

1. Introduction

The South China Sea (SCS) is a tropical-subtropical basin formed along the East Asian continental margin during the mid-Cenozoic (Fig. 1). It covers an area of about $3.5 \times 10^6 \text{ km}^2$ across over 20° of northern latitude from the equator in the western

Pacific (Wang and Li, 2009). Due to its marginal setting, a large part of the SCS is dominated by terrigenous sediments from the surrounding landmasses. On the separated continental terranes, such as the Xisha (Paracel) Islands, Zhongsha (Macclesfield Bank) Islands, and Nansha (Spratly) Islands, however, coral reefs or atolls are well developed, producing CaCO_3 at a rate of $>21 \text{ Mt/yr}$ (Yu and Zhao, 2009). Like many other low latitude reefs, most SCS reefs are built on the Miocene or younger carbonate platforms.

Reef studies in the region has been mainly driven by hydrocarbon exploration, and lately by academic research focusing on carbonate platform development as relating to the evolution of the SCS basin and global carbon cycle. Over the last 30 years, industrial drilling has revealed the existence of widespread carbonate platforms along the shelf edge and on older terranes (Erlach et al., 1993; Wang and Li, 2009 and references therein). In the southern SCS, for example, well Sampaguita-1 on the Reed Bank of the northeastern Nansha Islands recovered a reef complex of around 2100 m thick initiated from the late Oligocene (Du Bois, 1981; Taylor and Hayes, 1982; Yao et al., 2012). In Palawan, the Nido limestone of early Miocene age unconformably overlies the late Oligocene platform carbonates, which are underlain by pre-Nido limestones at some localities of NW Palawan shelf (Steuer et al., 2013), indicating carbonate buildups in the Eocene and early Oligocene. In 1974, Chinese geologists recovered a reef sequence of 1251 m thick covering the early Miocene to Pleistocene in a well named Xiyong 1 on Yongxing Island of Xisha Islands, which unconformably overlies a metamorphic complex of Precambrian to Paleozoic age (Wang et al., 1979; Yu and Zhao, 2009). During the 1980s-1990s, another 5 wells were drilled to various depths in the Xisha and Nansha areas by Chinese research organizations (Zhu et al., 1997), penetrating Miocene and younger carbonate sections (Fig. 1; Table 1). All the drilling results combined indicate that reef carbonate deposition first started in the Eocene-Oligocene in limited areas of the southern SCS, and carbonate platforms expanded to wider regions since the early Miocene following accelerated seafloor

spreading. Most Miocene reefs, however, were subsequently drowned during the later middle Miocene and late Miocene (e.g., Hutchison, 2004; 2010). Reefs on terranes largely survived through the middle-late Miocene crisis although these remaining platforms may have shrunk considerably with severe destructions occurring in places (Zhang et al., 1989; Xu et al., 1999; Wu et al., 2014).

Most of the cored materials from the above mentioned SCS wells, however, are no longer available for further detailed studies. Aiming for a better understanding of the regional reef history, a new well (XK1) was planned and drilled during 2013-2014 using the facility of China National Offshore Oil Corporation (CNOOC) Zhanjiang Branch. Preliminary results on the lithostratigraphy and geochemistry of well XK1 have been recently reported in several Chinese papers (Wang et al., 2015; Xiu et al., 2015; Zhai et al., 2015; Zhu et al., 2015). As part of a large research project on reef platform deposition and its implications for deep water buried plays, this study analyzed carbonate lithostratigraphy, foraminiferal biostratigraphy, elemental geochemistry and organic biomarkers, with a focus on the late Miocene-Pliocene history of reef development in the SCS.

2. Material and Methods

Xisha Islands are a group of atolls stubbed on an older terrane in the northwestern SCS (Fig. 1). Annual precipitation in the area ranges from 1300 mm to 2000 mm, mostly during the summer monsoon period (May to September) or typhoon season (June to November). The average SST is 26.8°C (or 22-30°C), while the salinity near surface ranges from 33.14 to 34.24‰, becoming lower at the surface layer during high rainfall (He and Zhang, 1986).

Well XK1 was drilled during 2013-2014 on the Shidao (Rocky) Island of Xisha from an altitude of +1.5 m to a bottom depth at 1268.2 m (Fig. 1; Table 1). The Cenozoic carbonate succession up to 1257.52 m thick unconformably overlies the Paleozoic or older metamorphic rocks. The well was fully cored, with core recovery

averages to ~70%, or >80% in most sections to <50% in several intervals with abundant unconsolidated bioclastics (Fig. 2).

In this study, samples spacing at ~1-5 m (201 in total for foraminifera; 1007 for elemental geochemistry including those with ~20 cm spacing from the upper 150 m) and at 5-10 m (90 samples for organic geochemistry) were taken from the upper 747.67 m of the well. Thin sections made from samples at 0.5-1 m spacing throughout the core were also examined for microtexture and facies variations as well as for foraminifera.

Samples for foraminiferal analyses were crushed and cooked for ~2 days before washing through a 63 μm sieve. Residue were then dried and stored in small containers for further analyses using a binocular microscope. Foraminiferal data from washed samples and thin sections together provide information useful for age dating and stratigraphic correlation.

Bulk sediment samples for elemental analyses were processed and measured with standard techniques using an inductively coupled plasma atomic emission spectrometer (ICP-AES) for major elements and an inductively coupled plasma emission mass spectrometer (ICP-MS) for trace elements. The procedural precision of measurements calculated from the differences of duplicate sample digestions was better than 2%rel for all analyzed elements. Major elements were based on their weight percentage, while the trace elements were expressed as parts of per million (ppm).

Samples for organic biomarker analyses were processed following the method described in Hopmans et al. (2004) and Schouten et al. (2007). Briefly, about 20 g freeze-dried samples were extracted ultrasonically five times using dichloromethane/methanol (3:1, V/V), after adding internal standards. The total lipid extracts (TLEs) were separated into two parts, with one part being used for GDGTs (glycerol dialkyl glycerol tetraethers) analysis. GDGTs were eluted isocratically with 99% hexane and 1% propanol for 5 min, followed by a linear gradient to 1.8%

propanol in 45 min with the flow rate at 0.2 ml/min. Detection was achieved using an Agilent 6460 triple-quadrupole mass spectrometer with atmospheric pressure chemical ionization (APCI) ion source. GDGTs were quantified by integration of the peak areas.

Calculation of the GDGT index BIT was based on the sum of all branched GDGTs (brGDGTs) over the total of brGDGTs and iGDGTs (Hopmans et al., 2004). Theoretically, the brGDGTs are mainly from soil bacteria and iGDGTs from archaea, thus representing land sources and marine sources, respectively. The methylation index of branched tetraethers and cyclization ratio of branched tetraethers (MBT/CBT) can also be used to infer the soil pH and mean annual air temperature based on the equations given by Weijers et al. (2007). In addition, sea water temperature based on TEX₈₆ using the equation $TEX_{86}=0.015T+0.28$ (Schouten et al., 2002) was also calculated. As this equation was primarily designed for open water samples, the results obtained and described below can only be viewed as representing a general trend rather than specific temperature values because samples from shallow water sites like XK1 could have been contaminated and therefore less reliable.

3. Results

3.1. Lithology

The lithology is dominated by reef carbonate limestones commonly in the forms of packstones and wackestones, which contain abundant coralline algae, corals (especially 21-190 m but rarely from below), benthic foraminifera, echinoids, bivalves, and skeletons of other shallow marine organisms (Fig. 2; see also Zhu et al., 2015). Dolomitized limestones and dolostones are concentrated in the following intervals: 289-303 m, 376-577 m, 623-637 m, 776.3-778.8 m, 896.27-898.48 m and 1208.92-1211.52 m. Unconsolidated or weakly consolidated sediments occur mainly in the uppermost part (<22 m) and several intervals dominated by bioclast-rich fine sand and mud with common algal remnants (Supplementary Fig. 1). In some

mudstones and siltstones with dark color spots, the concentration of organic matter is relatively high.

As also shown in Supplementary Fig. 1, typical erosional surfaces are characterized by partly dissolved hardgrounds and weathering surfaces associating with limonite concentration. Some erosional surfaces likely represent short-term hiatuses corresponding to stratal boundaries, as discussed further below.

3.2. Foraminiferal biostratigraphy

All foraminifera found in well XK1 are subtropical-tropical species, with preservation ranging from good to poor depending on diagenetic effects. Larger benthic foraminifera are common to abundant in most samples except in severely dolomitized intervals. The most common larger foraminifera include species of *Amphistegina* (*A. lessonii* and *A. madagascariensis*), *Austrotrillina* (*A. howchini*), *Calcarina* (*C. hispida*), *Cyclocypeus* (*C. indopacificus*), *Miogypsina* (*M. antillea*), *Operculina* (*O. complanata*) and *Nephrolepidina* (*N. morgana*), while other genera and species are rare. Based on the relative high abundance of the characteristic taxa, four foraminiferal assemblages were recognized : *Calcarina-Amphistegina* (0-223 m), *Amphistegina- Cyclocypeus* (223-376 m), *Miogypsina-Nephrolepidina* (577-1202 m), and *Spiroclypeus*-miliolids (1180-1257 m) (Supplementary Fig. 2). For the dolomitized late Miocene section between 375 and 577 m, no assemblage can be assigned due to heavy alternation, although some specimens of miliolids, bolivinids, and other small sized benthic and planktonic taxa may occur in less affected samples (e.g., Supplementary Fig. 2G).

Planktonic foraminifera are rare to occasionally frequent from upper 400 m and sporadic or absent below 400 m. Species frequently occurred include *Globigerinoides ruber* and *Globigerinoides sacculifer*, while species of *Orbulina*, *Dentoglobigerina* and *Globorotalia* are rare. Important datum levels include the last occurrence (LO) of common *Globigerinoides obliquus* at 215 m (>1.3 Ma), LO of *Dentoglobigerina altispira* group at 307 m (~3.5 Ma), first occurrence (FO) of *Sphaeroidinella*

dehiscens at 349 m (~4.7 Ma), and the FO of *Orbulina* spp. at 607 m (~15 Ma), with the datum ages following Wade et al. (2011) and the time scale by Gradstein et al. (2012).

The record of the FO of *Orbulina* at 607.43 m and the FO of *Globigerinoides* spp. at 643.15 m together indicates an age of ~15 Ma in the middle Miocene. This age is supported by the LO of common and continuously ranging *Miogypsina* and *Nephrolepidina* (at 577 m), both becoming extinct in the later part of the middle Miocene according to BouDagher-Fadel (2008). The sporadic occurrence of *Austrotrillina* up to 656.7 m (LO at ~13.5 Ma) also supports this section belonging to the middle Miocene. Below 1180 m, large specimens of *Spiroclypeus* were occasionally frequent, indicating the early Miocene (Table 2). Oligocene and older planktonic and larger benthic taxa (such as *Lepidocyclina* s.s.) are absent, further supporting an early Miocene age for the bottom section of the core.

The distribution of main foraminiferal taxa is summarized in Table 3, by emphasizing their variations in corresponding lithological intervals. Together with the datum levels listed in Table 2, these foraminiferal characteristics enable a fairly reliable division of the major Neogene stratigraphic units. Therefore, the sediment succession recovered in well XK1 can be subdivided into the following 4 intervals (Table 2): Pleistocene (0-216 m), Pliocene (216-376 m), late Miocene (376-577 m), middle Miocene (577-1032 m), and early Miocene with contacts falling on erosional surfaces (Fig. 2; Table 2). Comparatively, the late Miocene section features greater changes than other intervals in having a severely altered foraminiferal biofacies (Supplementary Fig. 2G-I). Finer stratigraphic divisions, however, require further studies.

3.2. Elements

As the most characteristic elements, CaO ranges from 35 to 100%, representing about 60-100% of the total CaCO₃ (Fig. 3). MgO comes second in abundance with

1-20%, showing a mirroring pattern with the abundance of CaCO_3 . Intervals with ~20% MgO fall in three intervals: (I) 623-637 m, (II) 376-577 m, and (III) 289-303 m, apparently corresponding to the dolomitized layers. Other elements contributing to the low CaCO_3 values include SiO_2 , up to 10% or more. Unlike these common components, P_2O_5 is rare, below 0.2%, with relative higher values in samples from the middle to late Pleistocene (0.08-0.15) and from the later late Miocene and Pliocene (0.05-0.1), thus showing some intermediate patterns between those of CaO and MgO. For trace elements, both B and Ba increase from ~1 to >20 ppm up-section with strong fluctuations (Fig. 3). The upward increasing trend is followed by Zr, La, Mn, Sc, Th and Ti although with much lower abundance. Li, however, shows patterns transitional between B and Ba with obvious cyclicities. Unlike all others, U is rather stable at 2-5 ppm except for peaks at 745 m, 650 m, 260 m and 225 m. The results of other elements and their ratios are presented in Supplementary Fig. 3, and only the most relevant elemental data summarized in Fig. 3 are emphasized in the following discussion.

3.3. Biomarkers

Nine brGDGTs and five iGDGTs were detected, with brGDGTs ranging from 0 to ~17 ng/20g and iGDGTs from ~0.2 to ~5 ng/20g (Fig. 4). Crenarchaeol occupies ~40% of all iGDGTs, while brGDGTs-Ia and -Ib account for ~38% and 30% of all brGDGTs, respectively. The brGDGTs are largely absent in samples from ~570 m to 215 m, while the iGDGTs are low, 0.1-1 ng/20g, in most samples from between 190 m and 20 m. The abundance of individual biomarker lipids is given in Supplementary Fig. 4.

The BIT (brGDGTs vs brGDGTs+ iGDGTs) varied between 0.1 and 1, with 0.5 or lower values mainly in the interval from 575 m to 190 m (Fig. 4). For other intervals, BIT values reach 0.5 or more with strong fluctuations, although several samples at 720 m, 620-630 m and 110 m register relative lower values.

The MBT values range from 0.3 to 0.9 and show an upward increasing trend, while the CBT values fluctuate between 0.7 and 1.0 with an upward decreasing trend. The MBT/CBT-derived pH basically follows the tendency of the MBT in having relative lower values (6-8) in the lower section and higher values (7-9) in the upper section. The TEX86-derived sea water temperature shows 4 intervals with different mean values: ~25°C at 747-577 m, ~20°C at 577-220 m, ~27°C at 220-120 m, and increasing from <20°C at 120 m to >30°C at 10 m (Fig. 4).

4. Discussion

4.1. Late Miocene carbonate deposition setting shift to mesophotic waters

All the lithological, paleobiological, elemental and biomarker data indicate a big change in the late Miocene part of the carbonate sequence recovered in well XK1 (Tables 2 and 3; Figs. 2-5). Specifically, the change is marked by a shift from carbonate limestones to algal limestones or dolomitized limestones (Fig. 2), an altered foraminiferal assemblage (Supplementary Fig. 2), a sharp increase in MgO from ~1% to ~20% (Fig. 3), and a large drop in the BIT (Fig. 4). As shown in Fig. 3, MgO values up to ~20% occur in three intervals 645-620 m, 570-376 m, and 310-290 m, respectively. Although the 570-376 m interval was truncated by some short-term decreases to 5% or lower MgO, particularly at 470 m and 420 m, it represents the entire late Miocene section with a thickness of ~200 m and abundant dolomites (Fig. 2).

In samples less affected by dolomites from the late Miocene section, algae are abundant, up to ~70%. For foraminifera, small benthic and planktonic individuals both increase (Table 3), while larger benthic forms decrease to a minimum with only a few *Amphistegina* remnants (Fig. 2). The assemblage frequented by miliolids, bolivinids, and other small sized taxa including planktonic forms may imply relatively quieter, cooler and deeper environments (Murray, 1991), compared to warmer and shallower conditions of the middle Miocene backing a vigorous reef growth and a

diverse larger benthic foraminiferal assemblage (Table 3; Fig. 2; Supplementary Fig. 2). Thus, if the middle Miocene reef carbonate buildup was occurring in 5-10 m or shallower waters, the late Miocene fine grain algal limestones might have been deposited in mesophotic water depths exceeding 30 m before being transformed into dolostones.

The late Miocene dolomites show a distinct drusy mosaic texture. Corresponding to this, most late Miocene samples are characterized by high MgO (20%) and moderate P₂O₅ (0.05-0.1 %) and Li (2-20 ppb), but minimum Sr, Ba, Ga, Rb, Ti, and Zr (Fig. 3; Supplementary Fig. 3). As a typical geochemistry problem, dolomitization involves many depositional and diagenetic processes which are largely unknown, but it seems to have been associated primarily with argillaceous carbonates as a replicative phase (Wilson, 2012). Meanwhile, a steady increase of Mg/Ca in the global ocean from 3 prior to the late Miocene to 4 afterwards (Stanley and Hardie, 1998) could have also played a role in increased dolomitization globally since the late Miocene. Although the topic will be dealt with specifically in subsequent on-going geochemical work, our preliminary elemental results reveal very low such elements as Ti or Fe, indicating a far offshore setting with very little terrestrial influence by dust or river runoff, especially in the Miocene and Pliocene. However, strong fluctuations in most elements since the early Pleistocene (Fig. 3; Supplementary Fig. 3) may suggest some continental link with intensified monsoons at glacial sea-level lows (e.g., Wei et al., 2003, 2006). Mn, however, shows a high level of variations not only in the Pleistocene but also in the late Miocene due to its paired authogenic marine and diagenetic nature. Compared to other intervals, increases in the total REE and such elements as Li in the late Miocene (Fig. 3) with abundant fine grain sediments appear to indicate also mesophotic conditions, as their enrichment often resulted from a higher rate of absorption by muddy sediments below active reef growth zone (e.g., Zhang, 1997). Together with the above-mentioned foraminiferal and elemental results, therefore, rare corals and other shelly organism remnants but maximum algae

observed in samples from the late Miocene section (Fig. 2; Supplementary Fig. 2) support the notion that the algal-rich fine-grain sediment sequence from the Xisha Islands was deposited largely in mesophotic conditions, in water depths of 30 to 150 m (e.g., Lesser et al., 2009; Weinstein et al., 2015).

The biomarker index BIT is the ratio between the bacterial-derived brGDGTs and the archaea-derived iGDGTs, thus representing the variations between the land-sourced biomarkers and marine biomarkers (Hopmans et al., 2004). The dominance of land-sourced organisms would push BIT close to 1, whereas smaller values close to 0 are often indicative of a higher level of marine influence. In well XK1, BIT fluctuates mostly below 0.4 in the late Miocene-Pliocene and part of the middle Miocene sections, but ranges between 0.5 and 0.9 in most middle Miocene and Pleistocene intervals (Fig. 3). These BIT variations may lead to a simple deduction that normal marine influence was instrumental during the late Miocene-Pliocene more than other time periods in the studied area. Schouten et al. (2007), however, has warned that some components of marine sourced brGDGTs could affect the BIT signal at shallow water settings.

The TEX86-derived temperature curve (Fig. 4) reveals strong fluctuations dividable into 4 stages: 3~0-25°C for the middle Miocene (747-577 m), ~23°C for the late Miocene to earliest Pliocene (577-220 m), ~27°C for the mid-Pliocene to early Pleistocene (220-120 m), and increasing from <20°C to >30°C since the middle Pleistocene (120-10 m). These results demonstrate a cooler depositional environment at XK1 locality during the late Miocene-Pliocene than in other intervals. The combined faunal, elemental and biomarker evidence, therefore, supports relative deeper and cooler mesophotic conditions for the deposition of the late Miocene carbonate section.

4.2. Late Miocene decline of Xisha carbonate platforms

As shown in Fig. 2 and Table 2, the ~200 m thick dolomitized late Miocene

section (375-575 m) corresponds to an average accumulation rate of ~32 m/Ma regardless of the post-depositional compaction, erosion and diagenesis. By comparison, this rate is only about half of that for the Pliocene (59 m), and about one third of that for the middle Miocene (91 m) or the Pleistocene (83 m). Although we cannot measure the age and duration of the hiatuses, some of which are believed to have concurred with the erosional surfaces marked in Fig. 2 (see also Zhu et al., 2015), these different accumulation rates may still warrant a scaling of the carbonate platform development between different time periods. At the well XK1 location in Xisha, therefore, the lithostratigraphic evidence indicates relatively active reef and platform growth during the middle Miocene, very weak during the late Miocene, and moderate to active during the Pliocene and Pleistocene.

In the studied region, the late Miocene sequence is bounded by seismic reflectors T30 (upper) and T40 (lower) (Fig. 1B). These two reflectors are prominent and closely packed in many profiles, especially in areas with apparent carbonate buildups, indicating the platforms now began to contract and backstep toward the local basement highs (Wu et al., 2014). The modern Shidao Island, where locates the well XK1, may represent one of a few atolls developed on one such basement high at the time (Fig. 1).

The late Miocene reduction in shallow water carbonate deposition was not limited to XK1 but has been recorded also in other Xisha-Nansha wells penetrating the Miocene sequence as listed in Table 1 (He and Zhang, 1986; Zhang et al., 1989; Zhu et al., 1997; Xu, 1999; Wei et al., 2008). Accompanying by a lower accumulation rate, the thickness of the late Miocene sections in most cores listed in Table 1 reduced considerably compared to the underlying older sequences. In well NY2 from the Nansha Islands, for example, strong dolomitization occurs not only in the Miocene section (>44 m) but also in the Pliocene (~100 m) and lower Pleistocene (~120 m) (Zhu et al., 1997; Xu, 1999). It thus represents a region-wide event. Obviously, the extent and intensity of dolomitization alone may not signify the healthiness of the

carbonate platforms as it could have been merely a diagenetic effect (Wilson, 2012). As pointed out by Perrin (2002) and Wilson (2008), however, late Miocene carbonate platform contraction was not a local phenomenon but has been known in many parts of SE Asia or even equator-wide around the earth. Climate cooling and strengthened winter monsoon, both probably triggered by the initiation of the Northern Hemisphere ice sheets (Zachos et al., 2001), appear to have played a significant role. We examine this issue further below.

4.3. Coupled effects of subsidence and cooling on late Miocene carbonate deposition

Carbonate platform development depends on various global and regional factors, including tectonics, CO₂, oceanography, nutrient input and precipitation patterns (Wilson, 2008, 2012). Specifically, apart from a warm-water oligophotic setting with adequate photoautotrophs and heterotrophs, other requisite conditions include reduced marine salinity and reduced nutrient upwelling, plus the absence of obvious sedimentary evaporites, coated grains or aggregates (Wilson, 2012). On a longer timescale, sea level change appears to have more important impacts because it often resulted in different basement structures and water conditions that may or may not favor reef carbonate buildup. Because the reefs on carbonate platforms are built by colonies of organisms dominated by *K*-mode specialists which are extremely sensitive to environmental changes, only minor variations in ambient water conditions or sea level change by a few to tens of meters will impose catastrophic effects on them (e.g., Montaggioni, 2005; Lukasik et al., 2008).

The decline of late Miocene platform carbonate deposition along the SCS margin has been attributed to basin-wide thermal subsidence after the end of seafloor spreading at 16-15 Ma (e.g., Hutchison, 2004, 2010; Wang and Li, 2009). In the eastern Qiongdongnan Basin, immediately north of the studied area, tectonic subsidence up to 1300 m at a rate of ~260 m/Ma during the late Miocene has been projected using seismic data (Wu et al., 2014), although the total subsidence in areas

close to the Vietnamese margin was smaller, 500-1000 m (or ~100-200 m/Ma).

Previous microfossil studies indicate a large scale rise of the late Miocene sea level in the Yinggehai and Qiongdongnan Basins driven also by basin subsidence (e.g., Hao et al., 2000), which defied the overall lowering trend in the contemporary global sea level.

In Xisha area, a subsidence rate of 200-260 m/Ma (Wu et al., 2014) would far exceed the accumulation rate (after compaction) of ~32 m/Ma recorded for the late Miocene in well XK1 (Table 2). This comparison may render support to the previous notion that late Miocene subsidence was so spectacular that the original shallow-water carbonate platforms in the region were largely drowned (Erlich et al., 1993; Wang and Li, 2009; Hutchison, 2010). The accumulation of the late Miocene carbonates at ~32 m/Ma could then be indicative of either a slow deposition in water depths over 30 m with shattered reef growth or the effects of frequent erosions, or both. The latter is confirmed by at least 6 major erosional surfaces which punctuate the late Miocene section (Fig. 2; Supplementary Fig. 2), although current stratigraphy is unable to resolve the age and duration of the corresponding hiatuses. Instead, a deeper water ramp setting (or similar environment) during the deposition of the late Miocene section as a result of rapid basement subsidence was more likely. Two questions remain, however. (1) Why platform areas of Xisha and Nansha all submerged to become mesophotic in the late Miocene? (2) What have contributed to the dolomite formation if not the action of frequent exposure and monsoon precipitation on the argillaceous carbonate? A comparable subsidence rate between localities might help to answer the first question, but all platform areas subsiding at a similar rate looked unlikely.

Our TEX86-derived temperature curve differs substantially not only from the $\delta^{18}\text{O}$ curves from ODP Sites 1148 (Miocene to Pleistocene) (Fig. 5E; Tian et al., 2008), the widely used ice volume proxies, but also from the alkenone-based surface sea temperature curve from Site 1143 (Pliocene to Pleistocene; Li et al., 2011). As

indicated by $\delta^{18}\text{O}$ records (Zachos et al., 2001; Tian et al., 2008), after the ending of the middle Miocene climate optimum (17-15 Ma), the global climate started to deteriorate in the later part of the middle Miocene and reached a large dip at ~14 Ma accompanied by major ice sheet development on Antarctica (Fig. 5E). The abrupt onset of the South Asian Monsoon at 12.9 Ma and wind-driven currents caused the accumulation of spectacular drift sediment sequences as recovered in cores from the Maldives (Betzler et al., 2016). Although a proto-warm pool appeared to build up in the early part of the late Miocene, ~10-11 Ma (Jian et al., 2006; Li et al., 2006; Nathan and Leckie, 2009), symbolized also by a relative high TEX86-derived sea temperature ~30°C at 560 m in XK1 (Fig. 4), seemed to have persisted for a time too short to support platform recovery.

In a sharp contrast to the low amplitude $\delta^{18}\text{O}$ records, however, our TEX86-derived subsurface temperature dropped by ~10°C (or 30-20°C) immediately after the demise of the proto-warm pool and remained low, ~23°C, from the late Miocene to Pliocene (Fig. 4). According to Jia et al. (2012), the TEX86-derived temperature might not represent the surface sea temperature as originally thought but better as a proxy of the annual mean subsurface sea temperature at 30-125 m water depths in the SCS. Clearly, continuing basin subsidence had provoked the main part of this long term deepening and subsurface cooling. Meanwhile, the sinking of cool waters driven by enhanced winter monsoon induced by global cooling into the low latitude region may have also played an important role on weaker reef buildup during the late Miocene at the XK1 locality.

4.4. Neogene carbonate platform development and hydroclimate perspectives

The limestone sequence recovered in well XK1 provides evidence of carbonate platform development since the early Miocene in the NW SCS. Reef growth was more active in the middle Miocene than in any earlier periods, decreased in the late Miocene, revitalized in the Pliocene, and active again in the Pleistocene interglacial

periods. The decline of late Miocene carbonate platforms can be tracked along all SCS margins, from the Qiongdongnan and Zhujiangkou Basins in the north to offshore Palawan-Borneo and Dangerous Grounds in the south (e.g., Matthews et al., 1997; Madon et al., 1999; Noad, 2001; Hutchison, 2004; 2010; Ma et al., 2011; Yao et al., 2012; Ding et al., 2013; Wilson et al., 2013). Although the onset of carbonate platform reduction in some places may have occurred slightly earlier in the later middle Miocene, the late Miocene appeared to be the most severely affected period. Only on some basement highs did reef carbonate platforms survive and continue to grow into the Pliocene and Pleistocene (Hutchison, 2010; Wu et al., 2014).

As sketched in Fig. 6, the late Miocene platforms were double hit by rapid subsidence or rise in regional sea level and by monsoon cooling, which not only accounted for the deposition of smaller volume argillaceous carbonates in a mesophotic environment but also for their subsequent transformation into dolomites seen widely in many SCS localities. Diagenetic imprints on the sections with dolomites include frequent limonite concentration (Supplementary Fig. 1) likely due to exposures and erosions. This evidence is however at odd with the previous notion that a deeper mesophotic environment was prevalent during the deposition of the late Miocene limestone. Probably, sudden sea level falls as depicted in the global sea level curve may have contributed to these short-term exposures and erosions, especially when regional subsidence was easing. At these sea level lowstands, a large “lagoon-like” bay involving all Xisha (Paracel) Islands as its rim may have developed, and subsequently maintained to become the atolls today. Evidence for such a deep bay is the deposition of organic matter-rich fine-grain sediments (Supplementary Fig. 1), indicative of a stratified water column with the poorly ventilating bottom water. Calcareous algae would flourish not only in a mesophotic environment at rising sea level but also in restricted bottom water at falling sea level, and subsequently contributed to the enrichment of Mg (derived from algal thalli, e.g., Kamenos et al., 2009).

Therefore, late Miocene dolomitization in the SCS region is accreditable to both mesophotic algal production and intensified diagenesis due to periodic subaerial exposures. Other factors, such as rigorous bottom currents, may have directly associated with the platform drowning and demise at times of strong tectonic activities (e.g., Tian et al., 2015), but their role appears to be secondary and local compared to the effect of basin-wide subsidence, climate cooling and prevalent mesophotic settings on carbonate development in the late Miocene NW SCS.

The variations of MBT/CBT-derived pH (Hopmans et al., 2004) mostly between 6 and 9 (Fig. 5B, unadjusted values) indicate a normal marine depositional environment affected only occasionally by short-term strong precipitation events implied by low pH values. The stepwise increase in pH values up-section may also imply increasing alkalinity in the SCS since the middle Miocene. The relative lower pH values from the lower part of the section may further imply stronger precipitation effects on carbonate deposition in the Miocene than in other periods if other factors are ignored. At ODP Site 1148 (18°50.17'N, 116°33.94'E, water depth 3294 m) in the northern SCS (Fig. 1), a continuous decrease in black carbon $\delta^{13}\text{C}$ from the Miocene to the Pleistocene has been attributed to long-term decreasing humidity affected by intensified winter monsoon over the SCS region (Fig. 5D; Wei et al., 2006). Thus, the geochemical data from XK1 and ODP 1148 both support a wetter climate scenario for the Miocene than today in the SCS.

In contrast, the relative higher BIT values of the Pleistocene and middle Miocene (Fig. 4) can be attributed to the enrichment of land-based biomarkers transported by dynamic currents or winds. Similarly, strong fluctuations in all elemental proxies in the Pleistocene interval (Fig. 3; Supplementary Fig. 3) appear to echo the enhanced loess deposition in China (e.g., Zheng et al., 2004) in indicating strong East Asian winter monsoon wind following the expansion of the Northern Hemisphere ice sheets about 2.5-2.7 Ma ago (e.g., Li et al., 2011). A different scenario may have existed in the middle Miocene when the SCS was expanding, and the Xisha area probably still

lied close to the margin of a new sea beginning to evolve into a deep-sea basin.

As discussed above, the late Miocene was characterized by relatively weaker reef growth and carbonate buildup. However, a different scenario of carbonate deposition existed in the deep-water SCS during the late Miocene. At ODP Site 1143 (9°21'N, 113°17'E, water depth 2772 m) in the southern SCS and Site 1146 (19°27'N, 116°16'E, water depth 2092 m) in the northern SCS (Fig. 1), for example, carbonate sediments increased from ~40% in the middle Miocene to 55% or more in the late Miocene before decreasing to lower values in the early Pliocene (Site 1143) or late Pliocene (Site 1146) (Fig. 5C; Li et al., 2006, 2007). The highest deep sea carbonate mass accumulation rate ~20 g/cm²/ka was recorded at Site 1143 in the late Miocene, followed by ~5-10 g/cm²/ka in the latest Miocene-Pliocene and <5 g/cm²/ka in the Pleistocene. This comparison demonstrates one of many different carbonate deposition features between shallow and deeper water settings since the middle Miocene in the SCS region. Future research with an integrative approach may shed light on this issue of the carbonate system and its evolution.

5. Conclusions

Lithostratigraphic, foraminiferal, elemental and organic biomarker results from well XK1 in the northwestern SCS provide evidence for a severe reduction in reef carbonate buildup during the late Miocene. The affected interval is an algal-rich carbonate section with a low deposition rate (~32 m/Ma post-compaction), and features abundant dolomites with high MgO (~20%), few benthic and planktonic foraminifera, very low land-sourced brGDGTs (0-0.5 ng/20g), low BIT index (0.1-0.5), and low TEX86-derived subsurface sea temperature (~23°C). Based on these results, we infer a dominant mesophotic environmental condition in relative cool and >30 m deep waters during the deposition of the late Miocene sequence. Together with similar records from other South China Sea localities, the carbonate section of XK1 exemplifies carbonate platform development in the low-latitude

western Pacific with a marked contraction period about 9-5 Ma ago. Although the reduced carbonate deposition in other regions was primarily driven by global cooling and changing oceanic chemistry in the late Miocene, rapid basin subsidence coupled with monsoon cooling were most likely the main factors causing the basin-wide reef decline in the South China Sea at the time. Comparatively, reef carbonate platform growth was more active in the middle Miocene and then in the Plio-Pleistocene, especially during interglacial periods. The lithostratigraphic and geochemical characteristics of carbonate succession recovered in well XK1 shed light on the history of carbonate platform development in the region.

Acknowledgements

We are grateful to Zhanjiang Branch of China National Offshore Oil Corporation Ltd for the supply of well XK1 data and samples. This research was supported by the National Scientific Foundation of China (91128207, 91228203, 41576059) and the Fundamental Research Program of the Ministry of Sciences and Technology, China (2011ZX05025-002). The manuscript benefited greatly from comments by Judy Webster, Lucien F. Montaggioni and an anonymous reviewer. Karl Stattegger and Shiguo Wu kindly reviewed an early draft. Guofu Cai, Yamin Song, Tianhui Yu, Meng Zhao and Fengfeng Zheng are thanked for assistance in sampling and/or analyses. All analyses were done using the facilities of the State Key Laboratory of Marine Geology, Tongji University.

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Figure captions

Fig. 1. (A) Locality map of well XK1 on the Rocky Island of the Yongxing Atoll (star) and other well sites mentioned in the text, including geological survey wells (yellow circles) and cores at ODP sites (red circles). Isobath = 1000 m. (B) Interpreted seismic profile along a section (location shown in A) to the west of XK1, on which the middle Miocene section is bounded by reflectors T50 and T40, late Miocene by T40 and T30 and Pliocene by T30 and T20, respectively (Wu et al., 2014 with a newly acquired profile).

Fig. 2. Lithostratigraphy of well XK1, showing core recovery, stratigraphic divisions, main facies, and typical lithologies. (A) Unconsolidated bioclastic sands, Pleistocene, 48.54 m. (B) Bioclastic limestone (packstone), Pleistocene, 54.62 m. (C) Foraminiferal bioclastic limestone, Pliocene, 231.76 m. (D) Loosely packed foraminiferal and bryozoan limestone, Pliocene, 340.92 m. (E) Dolomitized algal limestone with some unaffected calcite (red), late Miocene, 391.98 m. (F) Dolomitized algal limestone with some unaffected calcite (red), late Miocene, 456.81 m. (G) Weakly dolomitized bioclastic limestone with foraminiferal and algal skeletons, middle Miocene, 580.36 m. (H) Weakly dolomitized bioclastic limestone with algal and foraminiferal skeletons, middle Miocene, 744.48 m. Scale bars = 500 μm .

Fig. 3. Variations of geochemical components and elements in well XK1. High MgO contents ($\sim 20\%$) characterized three intervals (I to III), with the thickest interval II (376-577 m) representing the late Miocene deposition.

Fig. 4. Total abundance of iGDGTs and brGDGTs, and variations of other biomarker proxies: BIT, MBT, CBT, MBT/CBT-derived pH, and TEX86-derived temperature.

Fig. 5. Comparison between XK1 records (A, B) and deep sea records (C-E). (A) MBT/CBT-derived pH. (B) BIT. (C) CaCO₃ percentages from ODP Site 1146 (green) and Site 1143 (grey) (modified from Li et al., 2007). (D) Black carbon $\delta^{13}\text{C}$ from ODP Site 1148 (heavy line smoothed, from Wei et al., 2006). (E) $\delta^{18}\text{O}$ from ODP Site 1148 (from Tian et al., 2008).

Fig. 6. Sketches showing the generalized developmental stages of platform reefs in Xisha and Nansha Islands region. (A) Middle Miocene. (B) Late Miocene. (C) Pliocene-Pleistocene interglacials. (D) Pliocene-Pleistocene glacials. Regional subsidence and invasion of cool water by winter monsoon in the late Miocene are interpreted as playing a primary role in platform drowning and reef carbonate decline in most parts of the South China Sea.

Figure1

Figure2

Figure3

Figure4

Figure5

Figure6

(D) Pliocene-Pleistocene
low sea level (glaciation)

Frequent erosion

(C) Pliocene-Pleistocene
high sea level (interglaciation)

active reef growth

(B) late Miocene-early Pliocene
basin subsidence

platform sinking
and recovery

(A) middle Miocene

active reef growth

Table 1

Table 1. Fully cored wells drilled on Xisha and Nansha Islands by Chinese organizations

Well	Location	Lat/Long	Water	Penetration (m)	Carbonate	Driller	Year	References
	(island)		depth (m)		succession (m)			
<i>Xisha (Paracel)</i>								
Xiyong 1	Yongxing	16°50'01" N/112°20'48" E	+3	1348.68	1251	Ministry of Petroleum	1973-1974	Wang et al., 1979; Qin, 1987
Xichen 1	Chenhang	16°25'24" N/111°40'00" E	+3	802.17	802.17	Ministry of Mineral resources	1983	Zhang et al., 1989; Han and Meng, 1990
Xichen 2	Chenhang	16°25'24" N/111°40'00" E	+3	600.02	600.02	Ministry of Mineral resources	1984	Zhang et al., 1989; Han and Meng, 1990
Xishi 2	Shidao	16°50'38" N/112°20'50" E	+2	200.63	200.63	Ministry of Mineral resources	1983-1984	Zhang et al., 1989; Han and Meng, 1990
XK 1	Shidao	16°50'39" N/112°20'54 " E	+1.5	1268.2	1257.52	SNOOC	2013-2014	This study
<i>Nansha (Spratly)</i>								
Nanyong 1	Yongshu	9°32'24"N/112°52'46"E	+2.9	105.07	105.07	Chinese Academy of Science	1990	Zhu et al., 1997

Nanyong 2	Yongshu	9°32'24"N/112°52'46"E	+2	413.69	413.69	Chinese Academy of Science	1994	Zhu et al., 1997
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Table 2. Lithostratigraphy, calculated sedimentation rate (post-compaction), and key foraminiferal datums in well XK1. Stratigraphic ages of Gradstein et al. (2012) and datum ages of Wade et al. (2011) and BouDagher-Fade (2008) (correlated) are followed.

Interval (m)	Age (base Ma)	Thickness (m)	Sedi rate (m/Ma)	Main lithology
0-216	Pleistocene (2.6)	216	83	Coral packstone, wakestone
216-375	Pliocene (5.3)	160	59	Packstone, limestone
375-576.5	Late Miocene (11.6)	201	32	Dolostone, algal limestone
576.5-1032	Middle Miocene (16)	455	84	Limestone, foram-rich
1032-1257.5	Early Miocene (23)	236	34	Dolomitized limestone
1257.5-1268	Paleozoic or older			metamorphic rocks
Depth (m)	Age (Ma)	Datum		
215	>1.3	Common last occurrence of <i>Globigerinoides obliquus</i>		
307	~3.5	Last occurrence of <i>Dentoglobigerina altispira</i>		
349	~4.7	First occurrence of <i>Sphaeroidinella dehiscens</i>		
577	~11.6	Common/continuous last occurrence of <i>Miogypsina</i> and <i>Nephrolepidina</i>		
656.7	~13.5	Last occurrence of <i>Austrotrillina</i>		
1180	~17.5	Last occurrence of <i>Spiroclypeus</i>		

Table 3. Major geochemical and paleobiological characteristics of the Miocene and Plio-Pleistocene sequences in XK1 (R=rare, <5%; F=frequent, 5-10%; C=common, 10-30%; A=abundant, >30%; S=sporadic)

	Early Miocene (1257.5-1032 m)	Middle Miocene (1032-576.5 m)	Late Miocene (576.5-375 m)	Plio-Pleistocene (<375 m)
Geochemistry:				
Dolomite (MgO)	R-F	R	A	F-R
REE, Li	Low	Low	Higher	High
Small benthic forams:	R	R	R-F	R
Miliolids	R-F	R	R-F	R-F
Lagenids + Bolivinids	R	R	R-F	R-F
Large benthic forams:	R-C	C-A	R-F	C-A
<i>Calcarina</i>	R	F-C	S	F-C
<i>Amphistegina</i>	R	F	S-R	F-C
<i>Cyclocypeus</i>	R	R-F	S	F-C
<i>Austratrillina</i>	S-R	S-R		
<i>Nephrolepidina</i>	R	F	S (relict?)	R (relict?)
<i>Miogypsina</i>	R	F		
<i>Gypsina</i>	R-F	R-F	S-R	F
<i>Spiroclypeus</i>	R-F			
Planktonic forams:		S-R	R-F	R-F
<i>Orbulina</i>		S	S	R
<i>Globigerinoides</i>	?	S	S	R-F
<i>Globorotalia</i>		S	S	R
Algae	R-C	R-F	A	R-C
Interpreted environment	Reefal (<30 m)	Reefal (<30 m)	Mesophotic (>30 m)	Reefal (<30 m)