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A persistent increase in primary productivity east off Hainan Island (northwestern South China Sea) over the last decades as inferred from sediment records

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ABSTRACT

Sediment cores were analyzed from the continental shelf of the northwestern South China Sea aiming to understand the change history of primary productivity and provide insights into key changes of environmental conditions in this region over the past ~100 years. Multiple proxies including stable carbon isotopic composition ($\delta^{13}C$) of sedimentary organic matter, diatom abundance and biogenic silica burial flux were applied along with ^{210}Pb chronology. Notably, these independent evidences consistently point to a steady increase of primary production in this region only after ~1960s. We propose that increasing atmospheric deposition due to dramatically enhanced human activities especially from China supplies essential nitrogen nutrients to the N-poor region and probably acts a major reason for the observed enhancement of marine primary production. Our study provides insights into better understanding how human perturbation may have profoundly impacted biogeochemical cycling in marginal seas in the last decades.

1. Introduction

Coastal and shelf sea regions are the most valuable and vulnerable (Jickells, 1998). Such regimes support substantial economic activity, such as a significant fraction of global fisheries (Watson and Pauly, 2001). They play a disproportionately important role in marine biogeochemistry and sequestration of atmospheric CO_2 (Zúñiga et al., 2019). For instance, > 10% of global new production takes place there while these regions account for only ~1% of the global ocean surface area (Walker and McCarthy, 2012). Primary production is one of the most fundamental processes driving carbon sequestration in marine environments. The ability to understand past changes of marine productivity on decadal- to centennial-scale is crucial to future projection (Chavez et al., 2011; Holt et al., 2012). However, unlike the sea surface temperature (SST), biological and chemical time-series are not available for the past decades or even ~100 years. On the other hand, environmental perturbation may be more complex and dynamic in the coastal and shelf waters than the open ocean. How marine productivity had changed in the context of increasing human impact on time scales of decades remains unclear and needs to be examined case by case.

The South China Sea (SCS), spanning from 1.5 to 23°N, is the largest (3.35 million km^2) marginal sea of the western Pacific Ocean. The SCS provides an ideal site to examine the change history of upper ocean production under the human impact on a decadal-centurial scale for several reasons. First, the SCS has extensive continental shelves (~41% as the shelf, bottom depth < 100 m). The biogeochemistry of continental shelf waters is dynamic and also potentially sensitive to global change (Liu et al., 2010). Second, as there are dramatically growing human activities (also at a high level when evaluated globally) on the highly populated and rapidly developing East Asia continent, it is reasonable to speculate that there is potentially imprints of human perturbation especially the northwestern SCS. Indeed, atmospheric deposition of anthropogenic nitrogen had left a surprisingly visible signal in the coral-bound nitrogen isotopic composition in a remote (several hundred kilometers from mainland China) northern SCS coral reef atoll (Ren et al., 2017). Although field measurements of primary production in the northern SCS are becoming more abundant (Chen et al., 1998; Chen and Chen, 2006; Ning et al., 2009; Song et al., 2012; Zhang et al., 2015), there lacks a long-term dataset that record possible changes in primary production over the past ~100 years in the shelf waters of the SCS. Therefore, biogeochemical indicators of marine productivity preserved in sediments of the SCS shelf should be invaluable, and likely

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provide important insights into understanding past changes in environmental conditions.

In the present study, we analyzed sediment records from the northwestern SCS spanning from the inner continental shelf to the slope based on Polonium-210 (^{210}Po) chronology. Independent biogeochemical proxies, including the stable carbon isotopic composition of sedimentary organic matter ($\delta^{13}C_{org}$), sedimentary diatom abundance (SDA), and biogenic silica burial flux (BF), were applied to try to infer past changes of primary productivity in the northwestern SCS. The isotopic composition $\delta^{13}C_{org}$ has been widely used as an indicator of the relative contribution of marine versus terrestrial organic matter (Guo et al., 2004; Meyers, 1997; Ogrinc et al., 2005). Diatom dominates the phytoplankton community in the coastal and shelf waters of the northwestern SCS (Luo et al., 2015), thus acts as a dominant contributor to carbon export as observed in similar coastal regimes (Abrantes et al., 2016). In coastal waters, a relatively large fraction of production is generally exported (Muller-Karger et al., 2005). Therefore, examining the records of diatom-related properties (i.e., SDA and BF) in such regimes will potentially reflect the changes in the upper ocean carbon production and provide implications for understanding possible shifts of environmental conditions concerning primary production.

In this study, we aim to: 1) reconstruct the change history of biological production in the northwestern SCS over the past ~100 years; and 2) explain the regulating factors for this change, especially for the past decades when both natural forcing and human impacts had been increasing. This study will aid the understanding of the biogeochemistry of coastal and shelf sea areas in the context of global change and human perturbation.

2. Materials and methods

2.1. Regional setting

The South China Sea is subject to East Asia monsoon, with southwesterly prevailing in summer and northeasterly prevailing in winter, respectively. In the northwestern South China Sea off the Hainan Island, a seasonal coastal upwelling (Qiongdong upwelling) driven by the Ekman offshore transport occurs during the southwestern monsoon in summer (Wang et al., 2016). This region receives a limited amount of freshwater discharge from rivers in the eastern Hainan Island. As the largest river in this region, the Wanquan River has a runoff $\sim 4.95 \times 10^9$ m^3 discharging into the coastal waters (Xu et al., 2014).

2.2. Sample collection

Five sediment cores were sampled using a box corer along a transect in the northwestern SCS during August 2013, spanning a region (18°10′-20°10′N, 108°37′-111°3′E) from the inner shelf to the continental slope (water depth 47–983 m; Fig. 1). The upper ~40 cm sliced at an interval of 1 cm. The samples were stored frozen (−20 °C) while onboard and dried at 60 °C to constant weight and grinded to 100-mesh size in the land laboratory. The sediment mass accumulation rates (i.e. MAR) at the study stations had been determined via ^{210}Pb technique using the constant rate of supply model (i.e. CRS) (Zhang, 2016), which can indicate the variability in sedimentation environments (Sanchez-Cabeza and Ruiz-Fernández, 2012; Yang et al., 2016).

2.3. Mass-accumulation rate and sediment chronology

The mass-accumulation rate (MAR) and sediment chronology were determined using the ^{210}Pb approach (Yang et al., 2016; Zhang, 2016). In the past half century, anthropogenic activities and natural processes have largely disturbed the sedimentation environments of Chinese coasts while the atmospheric deposition of ^{210}Pb seemed to show little variation (Yang et al., 2016; Yang et al., 2018). These specific scenarios appear to meet the precondition of the constant rate of ^{210}Pb supply model (CRS) than other ^{210}Pb-chronology model (Sanchez-Cabeza and Ruiz-Fernández, 2012). Thus, the CRS model was adopted to constrain the MAR and chronology to reveal the decadal variation of MAR in this study. ^{210}Po-derived ^{210}Pb activities and the detailed MAR and chronology determination can be found in Zhang (2016).

2.4. Burial flux of biogenic silica (BF)

Burial flux of biogenic silica (BF) was calculated from the product of MAR and biogenic silica (BSi) content. Acid silicomolybdenum-blue spectrophotometry was used for BSi analysis (Mortlock and Froelich, 1989). Briefly, ~60 mg crushed sample powder was transferred to a polypropylene centrifugal tube. The organic phase was removed by H_2O_2 (10%) and the carbonate was removed by HCl (1 N), respectively. Then the tube was added with Milli-Q water and was centrifuged at 3000 r min^{-1}. The precipitation was dried (60 °C) and the BSi was then extracted using Na_2CO_3 solution (2 mol L^{-1}). BSi was determined via spectrophotometry. BF (μmol Si m^{-2} $year^{-1}$) was calculated as:

$$BF = BSi_i \times MAR$$

2.5. Sedimentary diatom abundance (SDA)

Dried subsamples for sedimentary diatom abundance (SDA) analysis were soaked in pure water (24 h) and washed onto a 10 μm sieve. The particles retained were transferred to a centrifuge tube, and successively added with the heavy solution (2.2 g mL^{-1} ZnI_2) and acetic acid (1%) for gravity separation. Thereafter the sample was moved into a graduated centrifugal tube to constant volume 1 mL (Li et al., 2004). An aliquot of 30 μL was collected and counted for SDA (ind g^{-1}, i.e., individuals of diatom shells per gram dry sediment) on a light microscope (OLYMPUS C011).

2.6. Stable isotopic composition of sedimentary organic matter ($\delta^{13}C_{org}$)

Inorganic carbon was eliminated using 1 N HCl before analysis of stable isotopic composition of sedimentary organic carbon ($\delta^{13}C_{org}$). After rinsing with Milli-Q water and re-drying, the sample was weighed (~10 mg) and transferred into a clean tin capsule. $\delta^{13}C_{org}$ was measured on an elemental analyzer (FLASH 1112) coupled to isotope ratio mass spectrometer (Thermo Finnigan DELTA V ADVANTAGE). Two isotope standards, IAEA-C8 ($\delta^{13}C = -18.3‰$ vs VPDB) and USGS-40 ($\delta^{13}C = -26.4‰$ vs VPDB) were inserted with samples. The isotopic composition is expressed by δ notation ($\delta^{13}C = [(R_{sample} / R_{standard}) - 1] \times 1000$, where R is the $^{13}C/^{12}C$ atom ratio of sample or standard). The precision of $\delta^{13}C$ measurements was within 0.2‰. $\delta^{13}C_{org}$ values were corrected for the Suess effect (the admixture of ^{13}C depleted CO_2 from fossil-fuel combustion) after Schelske and Hodell (1995) on ^{210}Pb chronology.

3. Results

3.1. Burial flux (BF) of biogenic silica

Burial flux (BF) of biogenic silica ranged between 12 and 256 μmol Si cm^{-2} $year^{-1}$ (Fig. 2). The stations (S0401, S0403) closer to the coast had higher BF (31–256 μmol Si cm^{-2} $year^{-1}$). The rest stations showed much lower BF, ranging from 12 to 63 μmol Si cm^{-2} $year^{-1}$. BF generally showed a drastic increasing trend since ~1960, while before that BF remained relatively stable or varied slightly for all the five cores.

3.2. Sedimentary diatom abundance (SDA)

Sediment diatom abundance (SDA) fell in a wide range of 279–11,206 ind g^{-1} at the three stations sampled (Fig. 2). Station S0401 closest to the coast had SDA ranging from 1225 to 11,206 ind

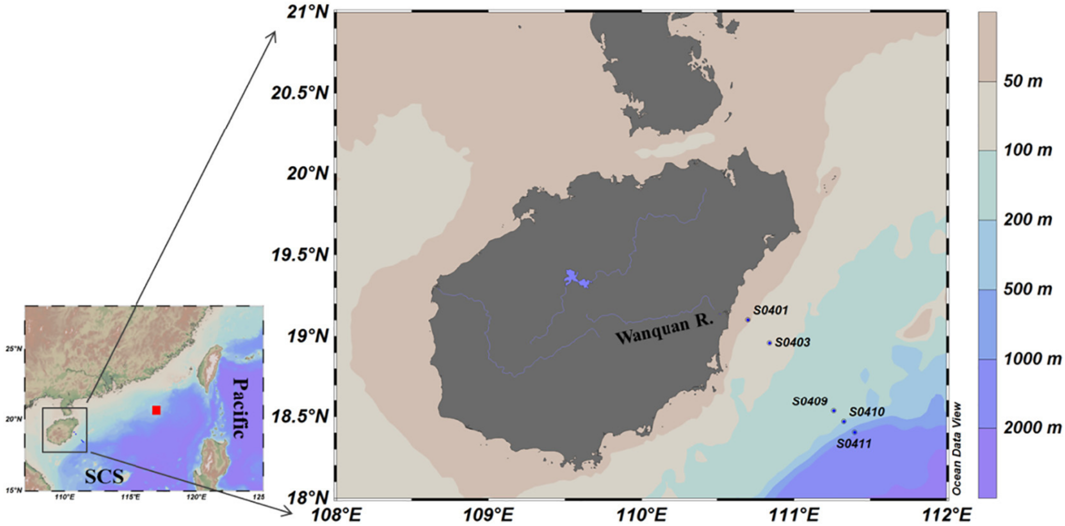

Fig. 1. Sampling locations (n = 5) of sediment cores off eastern Hainan Island in the northwestern South China Sea. Wanquan River and Dongsha Atoll (red rectangle) are also denoted. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

g^{-1}. The stations (S0409 and S0410) away from the coast had generally lower SDA ranging from 279 to 5233 ind g^{-1}. An increasing trend in the last decades for SDA is evident.

3.3. Stable carbon isotopic composition ($\delta^{13}C_{org}$)

Sedimentary $\delta^{13}C_{org}$ ranged from −21.6 to −18.7‰ over the past ~100 years (Fig. 3). Before ~1960, $\delta^{13}C_{org}$ showed relatively small variability in general. Then there appeared a drastic increase in $\delta^{13}C_{org}$ over the last decades, regardless the sediment cores were collected closer to the coast (Stn S0401) or on the far side (Stn S0411, water depth ~ 1000 m).

4. Discussion

4.1. Enhanced primary productivity since the 1960s

A novel finding from sedimentary records in this study is that primary production in the northwestern SCS may have increased steadily over the last decades, especially since ~1960s. Three lines of independent evidence supported this argument. First, SDA increased dramatically over the past decades in this study, implying an increase in primary production (Fig. 4b). Like in similar coastal upwelling regions where SDA had been proved a useful proxy of surface productivity (Abrantes et al., 2016), diatom dominates phytoplankton composition in the northwestern SCS continental shelf (Luo et al., 2015). Second, biogenic silica flux to sediments, an index of diatom productivity (Colman and Bratton, 2003), remained little varied before 1950–1960 and started to increase apparently thereafter (Fig. 4a), also supporting productivity increase over the past decades. Third, $\delta^{13}C$ of sedimentary organic matter ($\delta^{13}C_{org}$) have generally been used to reflect the relative contribution of terrestrial relative to marine organic matter in continental shelf sediments due to the distinct endmember characteristics of these two organic matter sources (lower terrestrial $\delta^{13}C$ vs higher marine $\delta^{13}C$) (Meyers, 1994; Li et al., 2013; Xiao et al., 2020; Ye et al., 2018; Zhang et al., 2013). In this study, sedimentary $\delta^{13}C_{org}$ fell in a range of −21.6 and − 18.7‰ (Fig. 3), implying a major contribution of marine production (Meyers, 1994; Li et al., 2013; Liu et al., 2007;

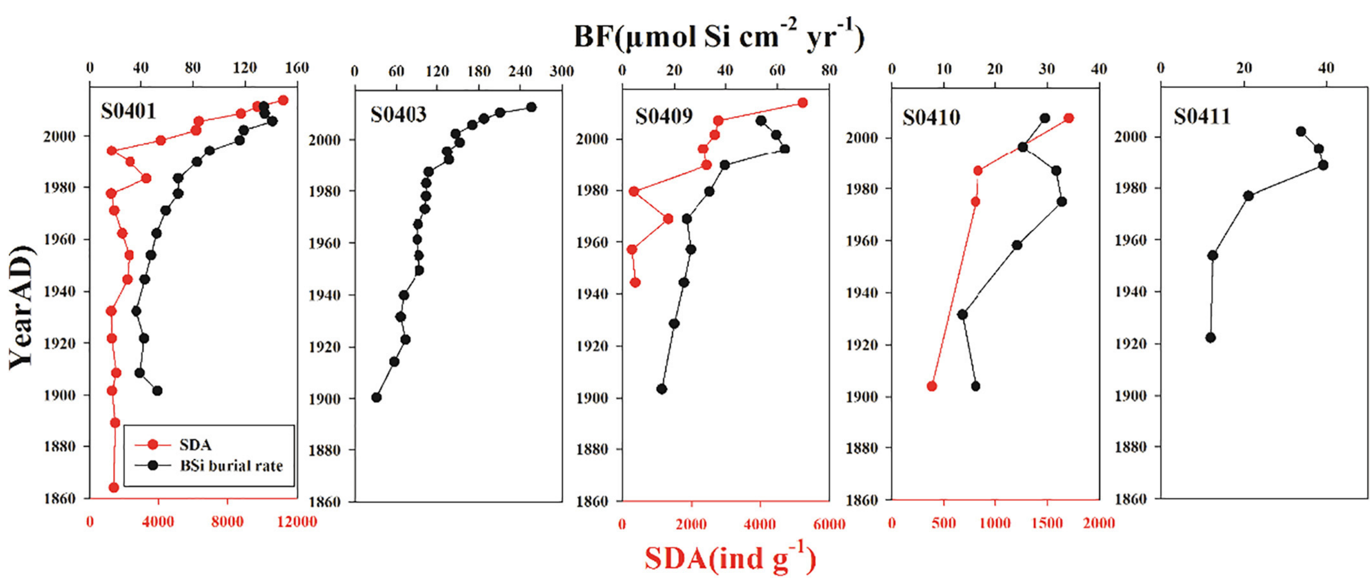

Fig. 2. Down-core variations of BSi burial flux (in black) and sedimentary diatom abundance (in red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

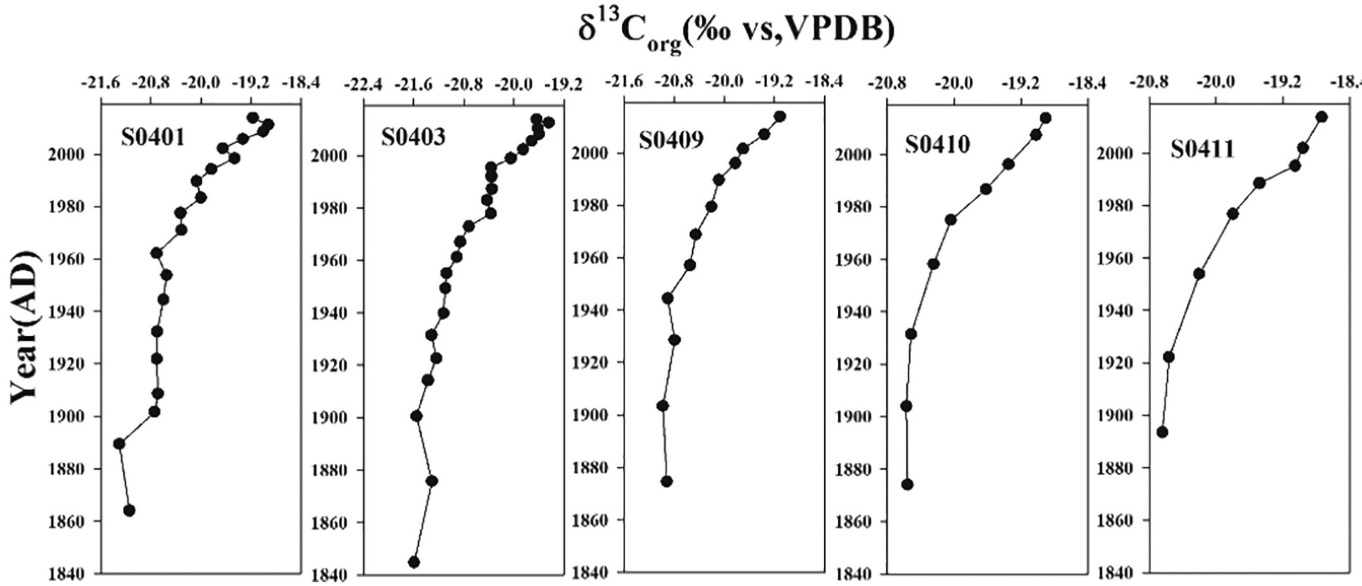

Fig. 3. Down-core variations of $\delta^{13}C_{org}$.

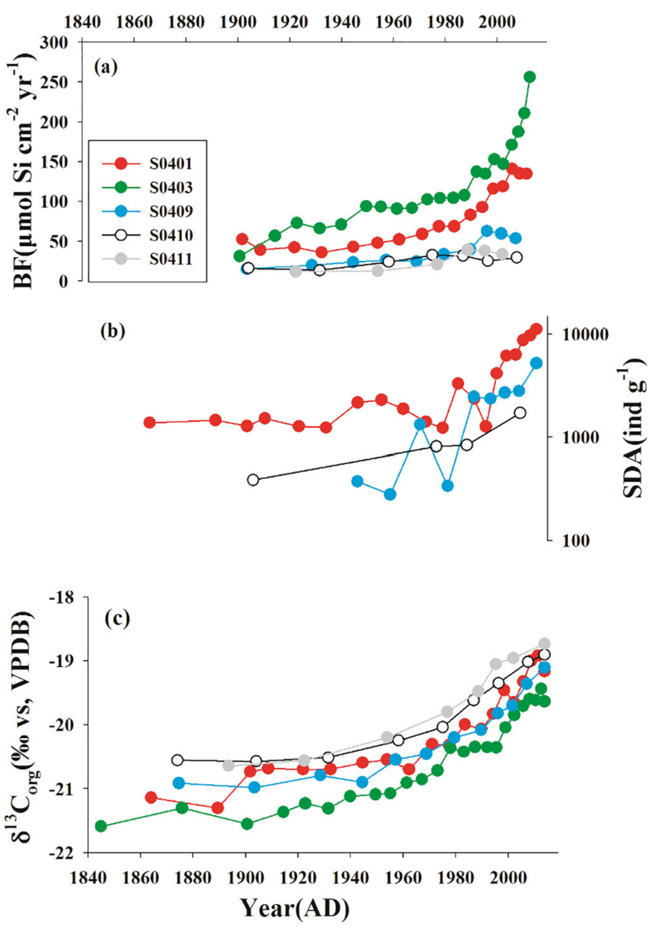

Fig. 4. Reconstructed change history of (a) BSi burial flux, (b) sediment diatom abundance, and (c) sedimentary organic $\delta^{13}C$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Zhang et al., 2013). Liu et al. (2007) reported a similar range for surface sedimentary δ^{13}Corg (−21.5% to −18.8%) in three stations of the northern South China Sea. We did observe a steady increase in sedimentary $\delta^{13}C_{org}$ since the 1960s, while before 1960s $\delta^{13}C_{org}$ showed little variations and no clear trend exists. The increased $\delta^{13}C_{org}$ can be explained by a) an increase in the fraction of marine organic matter, and/or b) a decrease in the fraction of terrestrial organic matter (Wu et al., 2017; Xiao et al., 2020). Likewise, phytoplankton $\delta^{13}C$ had been suggested to be an indicator for eutrophication, as they may be sensitive to changes in primary production that result from anthropogenic nutrient inputs (Oczkowski et al., 2014). Taking the evidence of sedimentary diatom abundance and biogenic silica burial flux into account, we suggest that such pattern of $\delta^{13}C_{org}$ is reasonably explained by an increased contribution from primary production, likely being induced by changes of environmental conditions (such as inorganic nutrient availability) in this region over the past decades.

Our results showed that the increase in primary production was not limited to coastal waters, at least for the SCS, which suggests a large spatial scale variation in the SCS over the past decades. Interestingly, our finding of increased primary production since ~1960s is consistent with prior observations in the subtropical Pearl River estuary (SCS) (Hu et al., 2008; Jia et al., 2013) and temperate Changjiang estuary (~31°N, East China Sea) (Feng et al., 2008; Xing et al., 2016), likely implying that an increased production over the past decades is common in China marginal seas.

4.2. Interpretation of enhanced primary production

4.2.1. Increased anthropogenic atmospheric deposition as a major cause

We propose that the increasing input of new nitrogen and other nutrients from atmospheric deposition should be a major cause for the increasing productivity in the northwestern SCS. It has been well accepted that nitrogen (N) a key limiting nutrient for primary production in the SCS (Chen and Chen, 2006; Liu et al., 2010). Riverine input of N is trapped within the shallow shelf and is minimal for the open water stations in the SCS (Cai et al., 2004). The major land river (Wanquan River) emptying into the study area has a very limited freshwater discharge (Xu et al., 2014), and its contribution as a nutrient input to the study area should also be spatially minimal. In contrast, anthropogenic N input via atmospheric deposition can impact over much broader spatial range in the SCS on annual basis (Kim et al., 2014; Ren et al., 2017). The combination of high levels of atmospheric N input and relatively low existing ocean productivity makes the SCS more vulnerable to anthropogenic N deposition (Kim et al., 2014). Atmospheric anthropogenic nitrogen deposition has become an increasingly important source of nitrogen input to the surface ocean because of the increase of

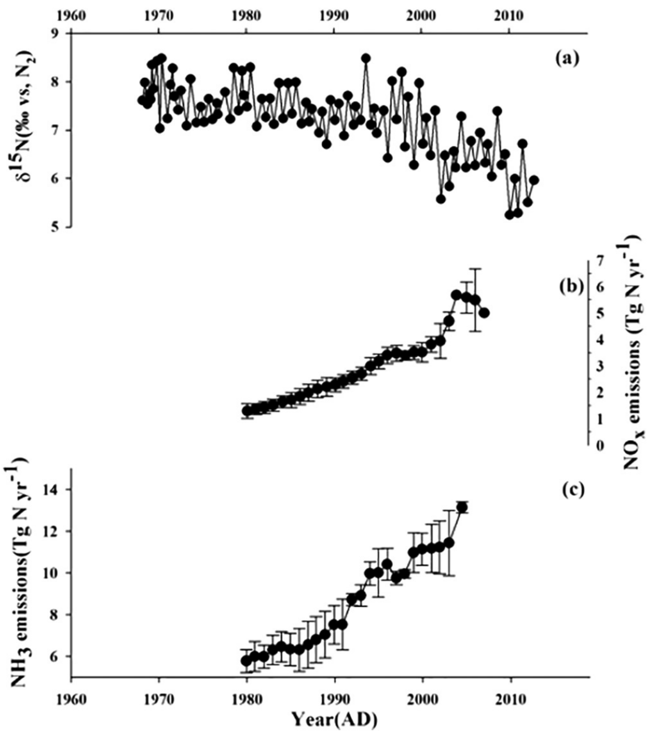

Fig. 5. (a) Coral skeleton δ^{15}N record in Dongsha Atoll (20°40′N, 116°50′E) of the northern SCS. (b) Total anthropogenic emissions (Tg N year^{-1}) of (b) NO_x and (c) NH_3 in China during 1980 and 2007. Data of δ^{15}N and anthropogenic N emissions are from Ren et al. (2017) and Liu et al. (2011), respectively.

the combustion of fossil fuels and emissions from fertilizer usage (Kim et al., 2014; Ren et al., 2017, references therein). Ren et al. (2017) found that the coral-bound nitrogen δ^{15}N in the remote Dongsha Atoll (340 km from land, also see Fig. 1) in the northern SCS showed a decreasing trend over the past decades, implying an increased imprint of atmospheric deposition of reactive nitrogen sourced from the rapid urbanization and industrialization of mainland China (Fig. 5). Ren et al. (2017) estimated a rate of atmospheric anthropogenic N deposition of ~50 mmol m^{-2} year^{-1}, which is more than twice the N_2 fixation rate (~20 mmol N m^{-2} year^{-1}) and is equal to ~25% of the vertical nitrate supply rate (~204 mmol N m^{-2} year^{-1}). Based on monitoring network and extrapolation (Kim et al., 2014) also reported similar atmospheric N deposition rate (55 mmol N m^{-2} year^{-1}) in the SCS. This contribution is much higher than the majority of the rest of world ocean (Duce et al., 2008; Jickells et al., 2017), thus should have played a significant role in shaping marine biogeochemistry as primary production in the SCS (even for the continental waters) is N-limited (Liu et al., 2010). Interestingly, the onset of the elevated $\delta^{13}C_{org}$ observed in this study is also consistence with the finding of increased coal combustion and vehicle emission based on sedimentary record of polycyclic aromatic hydrocarbons (PAHs) in this sea region in the past decades (Cai et al., 2017), further corroborating an increasing anthropogenic impact on the northern SCS.

4.2.2. Natural forcing

Upwelling may regulate the upward flux of nutrients into the euphotic zone and play a role in controlling primary productivity in coastal waters. However, we suggest that the variability of upwelling intensity (if any) over the past decades may not be a major controlling factor for the observed increase in primary productivity. There are at least two reasons. First, from a temporal perspective, the impact of coastal upwelling in the northwestern SCS does not exist all the year-round. It is a seasonal phenomenon, occurring in warm season (~June–September) only when the local wind (SW monsoon) is favorable (Jing et al., 2011). Second, the coastal upwelling in the northwestern SCS is topographically induced (parallel coastline to SW monsoon), and is active within a narrow band (restricted to < 50 m isobath) during its occurrence from a spatial perspective (Jing et al., 2011; Wang et al., 2014). Nevertheless, more investigations with higher temporal resolution are called for better understanding of whether there exists a decadal change of upwelling intensity, and how this seasonal phenomenon may impact primary production on annual basis in the northwestern SCS.

Globally, oceanic primary production has been suggested to be linked to SST and sea level, implying a tight physical and biological coupling, though the exact mechanism remains unclear (Chavez et al., 2011; Ito et al., 2010). SST reconstructed from corals in the SCS showed an increasing trend over the past decades (Yan et al., 2019). Besides, there is a persistent acceleration in global sea-level rise since the 1960s (Dangendorf et al., 2019). While in the South China Sea, sea level rise rate had been found to be significantly higher than the global mean rate for the same periods (Feng et al., 2012). It appears that the increasing patterns of SST and sea level are consistent with primary production in the northwestern South China Sea, and further studies are called for to better understand possible controls on primary production and ecosystem-level shift in marginal seas under increasing human impacts.

5. Conclusions

Based on ~100 year long sediment records ($\delta^{13}C_{org}$, biogenic silica burial flux, and sedimentary diatom abundance), we find a steady increase of primary production in the northwestern South China Sea since ~1960s. Enhanced atmospheric deposition of nitrogen sourced from intensifying human activities may have played a key role in stimulating phytoplankton production over the N-poor sea area in the last decades. Our findings thus provide insights into understanding of the biogeochemistry of coastal and shelf sea areas in the context of strengthening human perturbation on decadal-centennial scale and would help test climate model predictions.

CRediT authorship contribution statement

Mingyang Liu: Investigation, Formal analysis, Writing - original draft. **Chao Li:** Methodology, Formal analysis. **Fang Zhang:** Investigation, Formal analysis, Writing - original draft. **Run Zhang:** Conceptualization, Writing - original draft, Writing - review & editing. **Weifeng Yang:** Methodology, Formal analysis. **Min Chen:** Methodology, Formal analysis. **Minfang Zheng:** Formal analysis. **Yusheng Qiu:** Formal analysis.

Declaration of competing interest

The authors declare no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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