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# Reconstructing Environmental Changes of a Coastal Lagoon with Coral Reefs in Southeastern Hainan Island

ZHOU Liang[1, 2], GAO Shu[1, 2, 3], GAO Jianhua[1, 2], ZHAO Yangyang[2], HAN Zhuochen[2], YANG Yang[2], JIA Peihong[1, 2]

(1. *Collaborative Innovation Center of South China Sea Studies*, *Nanjing University*, *Nanjing* 210023, *China*; 2. *Ministry of Education Key Laboratory for Coast and Island Development*, *Nanjing University*, *Nanjing* 210023, *China*; 3. *State Key Laboratory of Estuarine and Coastal Research*, *East China Normal University*, *Shanghai* 200046, *China*)

**Abstract:** Coastal lagoons with small catchment basins are highly sensitive to natural processes and anthropogenic activities. To figure out the environmental changes of a coastal lagoon and its contribution to carbon burial, two sediment cores were collected in Xincun Lagoon, southeastern Hainan Island and $^{210}$Pb activities, grain size parameters, total organic carbon (TOC), total nitrogen (TN), total inorganic carbon (TIC) and stable carbon isotopes ($\delta^{13}$C) were measured. The results show that in 1770–1815, the decreasing water exchange capacity with outer open water, probably caused by the shifting and narrowing of the tidal inlet, not only diminished the currents and fined the sediments in the lagoon, but also reduced the organic matter of marine sources. From 1815 to 1950, the sedimentary environment of Xincun Lagoon was frequently influenced by storm events. These extreme events resulted in the high fluctuation of sediment grain size and sorting, as well as the great variation in contributions of terrestrial (higher plants, soils) and marine sources (phytoplankton, algae, seagrass). The extremely high content of TIC, compared to TOC before 1950 could be attributed to the large-scale coverage of coral reefs. However, with the boost of seawater aquaculture activities after 1970, the health growth of coral species was severely threatened, and corresponding production and inorganic carbon burial flux reduced. The apparent enhanced inorganic carbon burial rate after 1990 might result from the concomitant carbonate debris produced by seawater aquaculture. This result is important for local government long-term coastal management and environmental planning.

**Keywords:** environmental change; carbon burial flux; organic matter; coral reef; human activity; coastal lagoon; southeastern Hainan Island

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## 1 Introduction

Coastal lagoons represent a considerable sink for autochthonous matters produced by aquatic organisms (e.g., plankton, corals, seagrass) and allochthonous materials derived from terrestrial river inputs and exchanged with outer open water (Koop *et al*., 1987; Anthony and Blivi, 1999; Devlin and Brodie, 2005; Ryan *et al*., 2008). In some tropical regions, coastal lagoons or/and sheltered embayments are natural habitats to coral species (Alongi *et al*., 2006; 2007; Ryan *et al*., 2008; Fabricius *et al*., 2013). The relatively weak hydrodynamic conditions and high productivities result in large quantities of fine-grained organic-rich and inorganic materials accumulated in these lagoons (Brevik and Homburg, 2004; Alongi *et al*., 2006, 2007; Briand *et al*., 2015). As for small tropical catchment basins, coastal lagoons tend to be highly sensitive to external driving forces, such as typhoons, floodings, sea-level rising and human activities, which might cause remark-

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Corresponding author: GAO Shu. E-mail: shugao@nju.edu.cn

able changes in flow and sediment characteristics, geomorphological evolution, ecosystem structures and even biogeochemical cycles (Jackson *et al*., 2001; Bruno *et al*., 2003; Carpenter *et al*., 2008; Houser *et al*., 2008; Storms *et al*., 2008; Herbeck *et al*., 2011; Liu *et al*., 2011). Influenced by artificial reduction of river inflow and land reclamation, the flushing efficiency of the lagoon has deteriorated greatly and the spatial distribution pattern of the residual currents has changed significantly over past decades (Gong *et al*., 2008a; Yang *et al*., 2016). An intense storm might enhance resuspension of sediments and alter their spatial distributions, as well as lead to overwash or sands transported into the lagoon by channel cut (Bray and Carter, 1992; Sabatier *et al*., 2008; Zhou *et al*., 2015). These processes have been demonstrated to be well preserved in the sedimentary records of coastal lagoons and sheltered embayments (Sfriso *et al*., 1992; Li *et al*., 2008; Sabatier *et al*., 2008; Maanan *et al*., 2014), which in turn have also been recognized as ideal archives for environmental evolutions under the influences of natural changes and human activities (Ge *et al*., 2003; Gao and Jia, 2004; Leroy *et al*., 2006; Jia *et al*., 2012). Compared to large river deltas and estuaries, however, the processes of sediment accumulation and the variation of carbon burial in tropical coastal lagoons (Gao and Jia, 2004; Hu *et al*., 2006; Li *et al*., 2008; Jia *et al*., 2012), especially in a coral reef setting, remain unclear.

Coastal lagoons on Hainan Island, the second largest island of China, cover an area of 19 541 ha in total, accounting for nearly a tenth of the entire area of the coastal wetlands on this island (Song, 1984). On the southeast Hainan Island, numerous shallow embayments are host to a variety species of coral reefs, density seagrass beds and mangrove vegetation (Zhao *et al*., 1999; Wu and Zhang, 2012; Jiang *et al*., 2015). Unfortunately, the rapid economic developments of the coastal zone along Hainan Island, as the emergence of shrimp pools, fish ponds and constructing site (Li *et al*., 2014), has exerted immense pressures on the biological communities over the past decades (Fiege *et al*., 1994; Wu and Zhang, 2012). Over 80% of coral reefs have suffered devastation to various degrees and some even been faced with the risk of extinction (Wu and Zhang, 2012). Furthermore, typhoons and tropical cyclones from the western Pacific Ocean and the South China Sea frequently hit the southeastern coast of Hainan Island during July to October (Wang *et al*., 2001). Large storm surges and waves, with accompanied flood during typhoon periods, often facilitate strong sediment resuspension and transport and accelerate degradation of coastal ecosystems, which eventually result in changes of organic matter deposition and carbon burial budget in coastal lagoons (Lambert *et al*., 2008; Herbeck *et al*., 2011; Li *et al*., 2014). For this point, sediment composition (total organic carbon (TOC) content, total inorganic carbon (TIC) content) also could provide information of environmental changes due to natural and artificial forcing.

Xincun Lagoon, a representative small coastal catchment basin system located at southeastern coast of Hainan Island, has been extensively affected by natural processes (typhoons and storms) and human activities (agriculture, aquaculture and tourism) (Zou *et al*., 1975; Yang and Yang, 2009; Li *et al*., 2010). Short sediment cores were collected in this lagoon to identify the carbon sources and determine the carbon burial flux by geochemical indicators, including TOC, TIC, total nitrogen (TN), stable carbon isotopes ($\delta^{13}C$) and TOC/TN, the last two of which, combined with geochronology studies, have been widely used to elucidate the source and fate of organic matter (OM) in lagoons and estuaries (Shultz and Calder, 1976; Yamamuro, 2000; Gonneea *et al*., 2004; Li *et al*., 2008). In addition, sediment characteristics, together with TOC, TIC and TN, were analyzed to examine the environmental evolution patterns, responding to the impacts of both natural processes and human activities.

## 2 Study Area and Methods

### 2.1 Study area

Xincun Lagoon, located in Lingshui County, southeast Hainan Island, China, has an area of approximately 22.5 $km^2$ with an average depth of 4.2 m (relative to the mean sea level) (Gong *et al*., 2008b) (Fig. 1). It is characterized by a network of channels, marine-built terrace, tidal marshes, coastal dune and islets. Though a single tidal channel, the lagoon is connected to the South China Sea for the exchange of water and sediments mainly by tidal currents (Fig. 1). Mixed semidiurnal micro-tides dominate the lagoon with the mean tidal range below 1 m and the maximum tidal range of 1.55 m (Gong *et al*., 2008b). More than 56 coral species were found during the ecological investigation in the 1960s (Zou *et al*., 1975) (Fig. 1), which made it well-known for the rich biodiversity of coral reefs. Large areas of seagrass beds were distributed within this lagoon, which won it the only Tropical Seagrass Reserve in China (Jiang *et al*., 2015). Corals and seagrass are vulnerable to the tropical storms and typhoons, which hit this area frequently during July to November every year, bringing about large amounts of rainfall (Yang and Yang, 2009; Yang *et al*., 2015).

Xincun Lagoon was one of the most important fishing ports for foreign trade during the Ming and Qing dynasties. The port relocated from Tongqi to Zaozai and to Xincun over the past 200 years (Linshui County Chronicles Compilation Committee, 2007) (Fig. 1). Prior to the 1960s, Xincun Lagoon was characterized by a rich biodiversity of coral reefs, seagrass and mangroves (Zou *et al*., 1975; Zhao *et al*., 1999). During the last few decades, these ecosystems have yet been interfered extensively by the intensive human activities (*e.g.*, aquaculture and tourism) (Zou *et al*., 1975; Yang and Yang, 2009). The coral species in Xincun Lagoon decreased to below 30 in the 1980s (Zhao *et al*., 1999; Chinese Compilation Committee of Embayment, 1999), and to only few species left in 2013 based on our field investigation. The beaches of the sand spit in the south of the lagoon have developed for tourism. Satellite images displayed that the majority of the catchment basin has been cultivated for crops since the 1980s, and the tidal basin with its adjacent areas have been lost through reclamation for aquaculture ponds (mainly for shrimps and fish) since the 1970s. Red tides often occurred in the Xincun Bay due to eutrophication by excess anthropogenic nutrient inputs (Li *et al*., 2010).

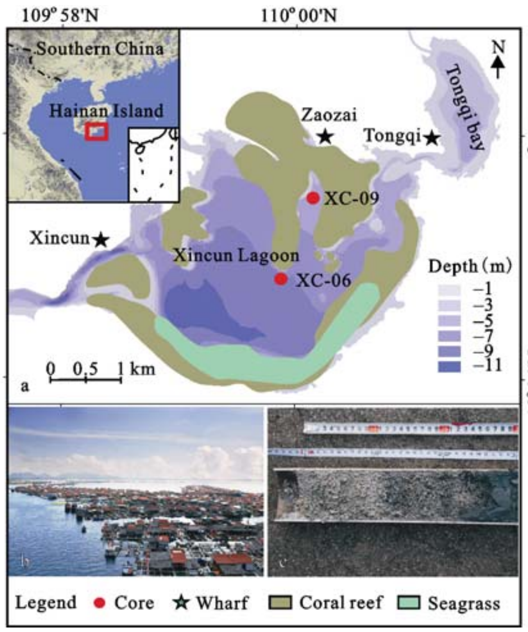


**Fig. 1** Maps of study area and sampling sites. a, Map of Xincun Lagoon, southeastern Hainan Island, with coral reef distribution in the 1960s (modified after Zou *et al.*, 1975; Zhao *et al*., 1999) and seagrass distribution patterns; b, crowded cage culture area in Xincun Lagoon; c, carbonate debris in the upper sediment layer produced by marine aquaculture

## 2.2 Methods

Two sediment cores (XC-06 and XC-09) were collected using a piston corer in August 2013 (Fig. 1a), with the lengths of 120 cm and 142 cm, respectively. Each core was sectioned at 2 cm intervals and samples were then packed into clean plastic bags and immediately stored under 40°C in laboratory. The wet and dry masses of sediment samples were weighed before and after freeze-drying, respectively, and dry bulk density and water content were calculated. After freeze-dry, a part of each sample was measured by a Mastersizer 2000 laser analyzer (with a relative error below 3% for replicated measurements) to obtain the grain size distribution, while the other part of each sample was grounded with an agate mortar pestle for following measurements. Grain size parameters were calculated by the moment method (McManus, 1988).

TC and TN contents of bulk samples were measured with a Flash 1112EA Series CNS elemental analyzer (linked to a microbalance system) and calculated by comparison with standard samples, with a precision of ±0.1% for TC and ±0.01% for TN (1SD), respectively. TIC was determined by a $CO_2$ coulometry with coulometrics 5030 and 5014 coulometers, based on the standard of $CaCO_3$ (Merck KGAA, Darmstadt, Germany). The accuracy of the measurements was verified by a standard reference material (USGS-MAG-1). Therefore, TOC was calculated as the difference between TC and TIC, and TOC/TN as the ratio of TOC and TN. Thirty-one samples throughout core XC-06 were selected to measure stable carbon isotope of total organic carbon on a DELTA$^{plus}$XL isotope ratio mass spectrometer, and the results were reported in the conventional δ-notation ($\delta^{13}C$) with respect to Peedee Belemnite (PDB) standard.

Dating of sediments by the $^{210}Pb$ method was performed in the Ministry of Education Key Laboratory for Coast and Island Development, Nanjing University. Total amount of $^{210}$Pb was determined by its granddaughter $^{210}$Po with an alpha spectrometry, assuming the secular equilibrium has been achieved. $^{210}$Po and $^{209}$Po (yield tracer) were spontaneously deposited onto a silver disc (Flynn, 1968) and counted on a standard silicon surface barrier detector. In this study, the constant initial concentration (CIC) model (Appleby and Oldfield, 1978; 1992) was employed to calculate the ages and deposition rates for sediment cores, with an assumption of a constant mass accumulation rate.

## 3 Results

### 3.1 $^{210}$Pb dating

The results of $^{210}$Pb dating for cores XC-06 and XC-09 are displayed in Fig. 2. The thickness of the mixing layers was identified to be 3 cm in core XC-06 and 17 cm in core XC-09, respectively, as for the uniform $^{210}$Pb activities. Thus, the excess $^{210}$Pb activities in mixing layers were not applied to the calculation of the sedimentation rates. It was evident to identify the constant value of $^{210}$Pb activity below the depth of 61 cm in core XC-06 (Fig. 2a), even though the $^{210}$Pb activity decreased gradually downward. In core XC-09, there was a sharp gradient of $^{210}$Pb activity at the depth of 58 cm, below which it kept stable downward (Fig. 2b). These constant values below the depths of 61 cm (core XC-06) and 58 cm (core XC-09) were averaged to be the background values, respectively. Except for the mixing layers, the logarithms of excess $^{210}$Pb activities are plotted against depth (Fig. 2) and show a significant linear declines with depth in core XC-09, indicating a constant sedimentation rate of 0.57 cm/yr (Fig. 2b). In core XC-06, the sedimentation rate experienced a noticeable change from 0.36 cm/yr to 0.60 cm/yr in the 1950s. On the basis of sedimentation rates, ages of sediments at different depths could be deduced supposed that the sedimentation rate was relatively stable throughout each period (Gonneea *et al*., 2004; Leroy *et al*., 2006; Li *et al*., 2008; Jia *et al*., 2012; Zhou *et al*., 2016). Based on the sedimentation rates and the lengths of these two cores, the bottom layers of core XC-06 and core XC-09 were dated to around 1770 AD and 1725 AD, respectively, enabling us to reconstruct the environmental changes in the southeastern Hainan Island over the last 240 years.

### 3.2 Grain size characteristics and TOC, TN and TIC contents

Both core sediments were mainly consisted of olive-grey clay silts, with the mean grain size ranging from 9.35 μm to 59.94 μm in core XC-06 and from 9.47 μm to 37.01 μm in core XC-09 (Fig. 3) and an average of 17.53 μm (core XC-06) and 15.51 μm (core XC-09), respectively. Their sorting was 1.87–3.40 (core XC-06) and 1.53–2.82 (core XC-09), and skewness varied from –0.35 to –0.04 (core XC-06) and from –0.33 to 0.05 (core XC-09), respectively. Their TOC content ranged from 0.30% to 1.42% in core XC-06 and 0.09% to 2.20% in core XC-09 (Fig. 3), and TN contents varied from 0.05% to 0.13% (core XC-06) and from 0.05% to 0.25% (core XC-09), respectively. The TIC values were in the range of 3.52%–5.82% in core XC-06 and 4.14%–7.16% in core XC-09. Based on the vertical distribution patterns of sediment grain size and TOC, TN and TIC contents, the two cores could be roughly

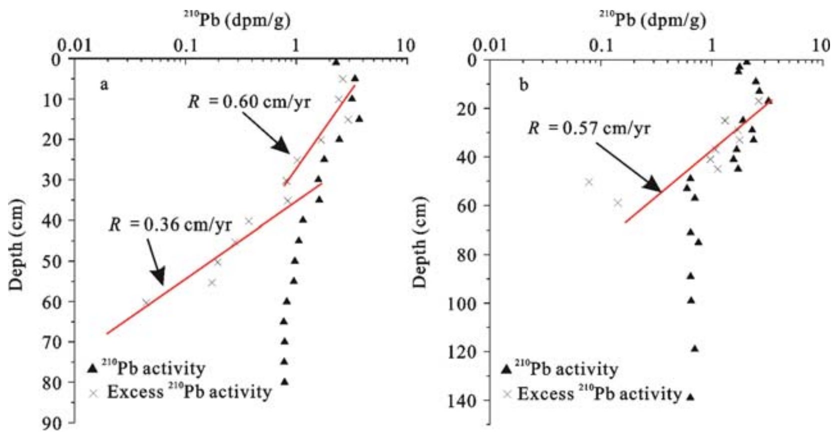


**Fig. 2** Profiles of $^{210}$Pb activity and excess $^{210}$Pb activity of sediment core XC-06 (a) and core XC-09 (b). *R* is the sedimentation rate

divided into three sections from top to bottom, i.e., section I (1950–2012), section II (1815–1950) and section III (1770–1815) (Fig. 3). Their average values of each parameter are listed in Table 1.

In core XC-06, mean grain size and sorting showed a slightly upward decrease from section III to section II and to section I, with relatively higher variations in section III, while grain size was more positively skewed in section II than both section III and section I (Fig. 3a). However, the vertical distribution of TOC and TN presented an inverse trend, increasing upward slightly with the highest values of TOC (1.42%) and TN (0.13%) at the top layer (Fig. 3a). The distribution of TIC was rather complicated, with a drop (upward) at the depth of ~90 cm in section III, keeping stable (about 4.27%) throughout section II, and a rapid rise (upward) in top 10 cm in section I (Fig. 3).

In core XC-09, all the grain size parameters (mean grain size, sorting and skewness) exhibited relatively higher variations in section II, while TOC in section II was lowest of these three sections with the mean value of only about 0.38% (Fig. 3b; Table 1). TN kept quite stable around 0.07% in section III and II, but increase significantly to 0.16% in the top 34 cm in section I (Fig. 3b). TIC showed a remarkable upward increase in section III, and fluctuated around 6.52% in section II. Across section II and section I, TIC decreased rapidly from 7.12% at the depth 46 cm to 5.86% at the depth of 26 cm, followed by a significant drop to 4.14% in depth 24–26 cm (Fig. 3b). Up to the top 10 cm, TIC rose rapidly to a relative high value around 5.50%.

### 3.3 Variations of TOC/TN and $\delta^{13}C$

In the core XC-06, $\delta^{13}C$ varied from –19.28‰ to –16.73‰ (Fig. 4) with different distribution patterns in three sections. In the section III, $\delta^{13}C$ increased from

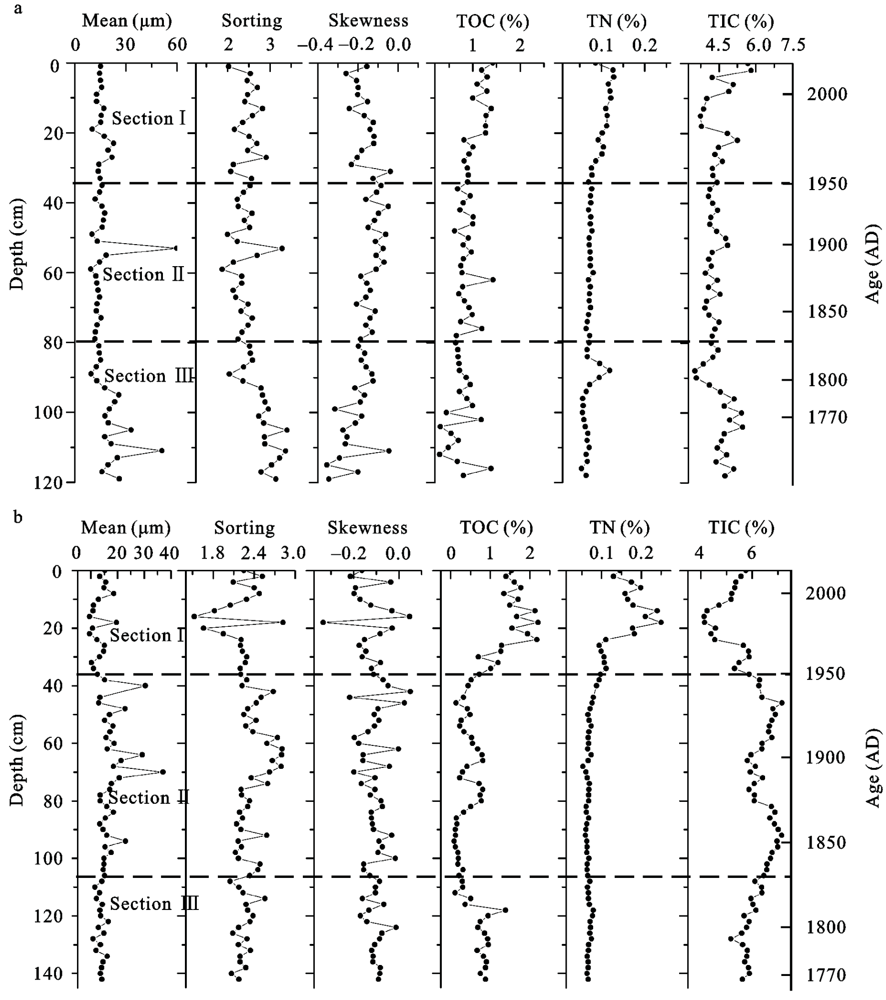


**Fig. 3** Vertical distribution pattern of grain size, TOC, TN and TIC in sediment core XC-06 (a) and core XC-09 (b)

**Table 1** Sediment grain size characteristics, TOC, TN and TIC in three sections (Mean±SD)

| | | Mean (μm) | Sorting | Skewness | TOC (%) | TN (%) | TIC (%) |
|---|---|---|---|---|---|---|---|
| Core XC-06 | Section I (1950–2012) | 15.84±3.09 | 2.47±0.25 | –0.17±0.06 | 1.08±0.23 | 0.10±0.02 | 4.54±0.61 |
| | Section II (1815–1950) | 15.88±9.64 | 2.38±0.27 | –0.13±0.06 | 0.85±0.19 | 0.07±0.01 | 4.27±0.24 |
| | Section III (1770–1815) | 16.89±5.33 | 2.74±0.24 | –0.20±0.04 | 0.77±0.17 | 0.08±0.02 | 5.12±0.64 |
| Core XC-09 | Section I (1950–2012) | 13.24±2.95 | 2.18±0.31 | –0.13±0.09 | 1.55±0.40 | 0.16±0.05 | 5.08±0.61 |
| | Section II (1815–1950) | 17.42±5.24 | 2.37±0.26 | –0.11±0.05 | 0.38±0.23 | 0.07±0.01 | 6.50±0.38 |
| | Section III (1770–1815) | 13.76±1.51 | 2.91±0.25 | –0.23±0.05 | 0.82±0.23 | 0.07±0.01 | 5.77±0.23 |

Note: SD is standard deviation

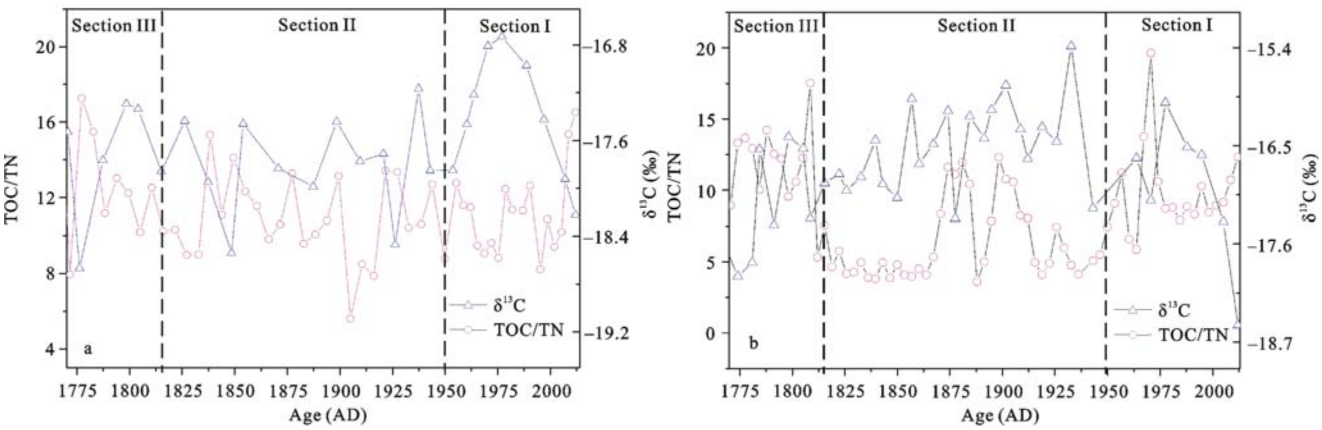


**Fig. 4** Variations of TOC/TN and $\delta^{13}C$ with depth in core XC-06 (a) and XC-09 (b)

–19.28‰ to –17.29‰ over 1770–1815, which was kept in section II but with a relatively large fluctuation. Staring from 1950 in section I, $\delta^{13}C$ increased to –16.73‰ around 1975 and then decreased to nearly –18.2‰ at present. TOC/TN ratio displayed a different distribution pattern. It increased from 7.95 to 17.26 in 1770–1777 and decreased rapidly to about 10 in 1815. In section II and I, it showed an overall general decrease with time except for an abrupt rise after about 2005. In the core XC-09, the value of $\delta^{13}C$ increased from –17.96‰ to –16.40‰ with a relatively higher variation in section III, and kept increasing in section II but with a relatively high variation (Fig. 4b). In section I, $\delta^{13}C$ increased considerably from −17.20‰ to −16.02‰ during 1950–1977, and decreased to −18.51‰ at the present. After a slightly increase from 1770 to 1778, the TOC/TN ratio decreased from 17.52 to 3.60 from section III to section II. In section II and I, the ratio of TOC/TN varied substantially in the period of 1865–1933 and then increased to the present (especially after about 2005) with high peaks around 1965–1970.

## 4 Discussion

### 4.1 Temporal changes of sediment grain size, TOC and TN

Grain size characteristics of lagoon sediment records are important proxies of hydrodynamic conditions and environment changes (Ge *et al*., 2003; Jia *et al*., 2003, 2012; Liu and Ge, 2012; Zhou *et al*., 2015). In 1770–1815, mean grain size decreased significantly in core XC-06 and showed a slightly declining trend in core XC-09, but the sorting of bulk sediments became better (smaller) in core XC-06 while relatively worse (larger) in core XC-09. These characteristics might indicate a dramatic adjustment of hydrodynamic conditions in the lagoon, probably resulting from the shifting and narrowing of the tidal inlets. The relatively smaller variation of sediment grain size in core XC-09 than that in core XC-06 could be attributed to the smaller water depth and the reduced flow buffered by aquatic communities (Fig. 1), such as corals and seagrass. In 1815–1950, similar peak mean grain sizes and sorting in both cores around 1890–1900 were well corresponded to several large storm events attacking this area (Zhou *et al.*, 2015). For this point, core XC-09 recorded more storm events than core XC-06 (Fig. 3), indicating that the shallow-water region was more accessible to be disturbed by enhanced currents and waves under the extreme hydrological conditions and to deposit coarse-grained materials transported from nearby beaches (Zhou *et al.*, 2015). The variation of sorting was higher than the mean grain size at some depth in XC-09 (Fig. 3). The possible reason is that the storm hydrodynamic conditions were relative weak in those periods and the storm wave effects on seabed were dominated by sediment resuspension. Therefore, the frequent perturbations by storms were responsible for the high variations in the grain size parameters of core XC-09. The fining-upward trend at both cores since 1950 may interpreted as the decline of hydrodynamic energy. The fining-upward trend in both cores since 1950 could be interpreted as the decline of hydrodynamic energy, as the average tidal range in this lagoon has decreased from 0.68 m in 1978 to 0.65 m in 1987, and to 0.50 m in 2013 (Zhang, 1987; Yang *et al.*, 2016). This may be likely to be caused by increasing aquaculture activities and coastal engineering over the past 40 years (Gong *et al.*, 2008a; Yang *et al.*, 2016), resulting in the squeezed tidal prism and reduced flow velocity.

TOC and TN of lagoon sediments reflect the net amount of marine organic matter produced by benthic and pelagic organisms and terrestrial organic matter discharged from the corresponding watershed (Ge *et al.*, 2003; Jia *et al.*, 2012). In 1770–1815, the decrease of TOC or its variation could be attributed to the decline of marine production exchanged with outer open water, since the terrestrial nutrient input and marine production within the lagoon were probably not diminished as TN kept stable or a rise after 1800 (Fig. 3). From 1815 to 1950, the low contents of TOC were coincided with the peak mean grain size (e.g., at the depth of 5–55 cm in core XC-06 and 65–70 cm in core XC-09), resulting from the greater input of coarse grain materials but lack of organic matter by storm attacks (Zhou *et al.*, 2015). After 1950, the rapid increase of TOC and TN contents, especially in core XC-09, reflected the dramatic increase of material inputs into the basin, in particular, the large amount of nutrient input (Fig. 3). This phenomenon has been found in a large amount of tropical lagoons, such as Xiaohai Lagoon (Gong *et al.*, 2008a; Liu and Ge, 2012) and Shamei Lagoon (Jia *et al.*, 2012) in the eastern Hainan Island, which were greatly impacted by human activities in past decades. With the continuous growth of population (Li *et al.*, 2014) and extensive increase of aquaculture areas in Lingshui County, the aquatic production increased from $6.19\times10^3$ t in 1957 to $9.45\times10^4$ t in 2012 (Fig. 5), nearly 15 times over 55 years. The accompanying input of aquaculture wastewater and unconsumed fish feed would undoubtedly elevate the nutrient level (Gong *et al.*, 2008a; Li *et al.*, 2010; Liu and Ge, 2012; Li *et al.*, 2014), which enhanced the autochthonous production.

### 4.2 Factors controlling evolution of $\delta^{13}C$ and TOC/TN in Xincun Lagoon sediments

The $\delta^{13}C$ and TOC/TN ratio are widely used to trace the biological origins of TOC in lagoon sediments (Ge *et al.*, 2003; Jia *et al.*, 2012). Organic matter produced by terrestrial vegetation has an average $\delta^{13}C$ value of approximately –27‰ for C3 plants and –14‰ for C4 plants, while organic matter produced by marine algae from aquatic bicarbonate has an average $\delta^{13}C$ value between –20‰ and –22‰ (Meyers, 1997). Besides, the $\delta^{13}C$ of organic matter produced from seagrass has also been measured to be ca. –14‰ (Moncreiff and Sullivan, 2001; Gonneea *et al.*, 2004; Miyajima *et al.*, 2014). As $\delta^{13}C$ in both cores varied between –19.28‰ and –15.38‰, terrestrial C4 plants and seagrass might contribute a high proportion of organic matter in this lagoon. However, C4 plants (e.g., sugarcane and corn) were not widely distributed in the catchment and seagrass ecosystem dominated Xincun Lagoon, implying a

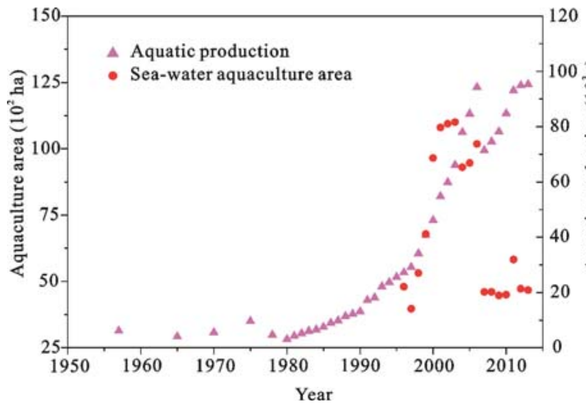


**Fig. 5** Changes of aquatic production and sea-water aquaculture area in Lingshui County from 1955 to 2013 (Hainan Provincial Bureau of Statistics, 1986–2013)

relatively larger contribution of seagrass to sedimentary organic matter. The TOC/TN ratio is usually >20 in higher plants, about 7 in phytoplankton and approximately 3–4 in bacteria (Hedges *et al*., 1997). The TOC/TN ratio in both cores rangd from 3.60 to ~20.00, mainly between 4 and 15, which well corresponds to the signal information of $\delta^{13}C$ for a mixed source of aquatic organic matter (phytoplankton) and terrigenous organic matter (higher plants) (Fig. 6). In addition, a proportion of organic matter might be derived from terrestrial soils, since the average TOC/TN ratio of the soils in the eastern Hainan Island is 13.4±1.7 (Unger *et al*., 2013).

The large fluctuation of $\delta^{13}C$ and TOC/TN ratio in 1770–1815 indicates the variable amounts of organic matter controlled by the water exchange capacity of the lagoon with outer open water. The remarkable increase of $\delta^{13}C$ and TOC/TN ratio might refer to the increased contribution of terrestrial inputs when compared to the diminished amount of autochthonous production, as the water exchange capacity might become weaker as a result of the gradual closure of the tidal inlet. To be noted that the decreasing area of this tidal basin also played a significant role in the variations of these chemical-biological indicators. Historical documents recorded the geomorphological evolution of Xincun Lagoon during this period; that is, the Tongqi Port was gradually disused because of the severe siltation in the Tongqi Bay (Fig. 1) during the Jiaqing-Daoguang period (1795–1850) of the late Qing Dynasty (Lingshui County Chronicles Compilation Committee, 2007). Thus, the declined tidal prism would further lower the water exchange capacity of Xincun Lagoon and result in the reduced contribution of organic matter from the oceanic source.

During the period of 1815–1950, the $\delta^{13}C$ and TOC/TN ratio were greatly influenced by the frequent storm events, as revealed by sediment characteristics. The accompanied strong rainfall enhanced the scouring and erosion of the catchment basin, leading to the sharp increase of terrigenous materials, $\delta^{13}C$ therefore decreased during the storm events. As terrestrial soils contained relatively high percentage of nitrogen- depleted lignin and cellulose (Gonneea *et al.*, 2004), compared with marine organic matters, the TOC/TN ratio became larger when typhoons hit this area. From this perspective, the high fluctuations of $\delta^{13}C$ and TOC/TN (Fig. 4) may indicate the frequent attacks of typhoons and tropical cyclones from 1815 to 1950.

After 1950, $\delta^{13}C$ experienced a rapid rise until ~1980 and then a rapid decrease to the present, revealing the expanding and shrinking history of the seagrass bed area as $\delta^{13}C$ of seagrass-derived organic matter (around –14‰) (Moncreiff and Sullivan, 2001; Gonneea *et al*., 2004; Miyajima *et al*., 2015) was much lighter than that of C3 plant-produced organic matter (–27‰) (Meyers, 1997) and aquaculture-derived organic matter (from –24.0‰ to –26.0‰) (Herbeck *et al*., 2011; Bao *et al*., 2013). With the boost of seagrass beds in the lagoon before 1980, seagrass provided an increasing contribution to the buried organic matter in sediments, which

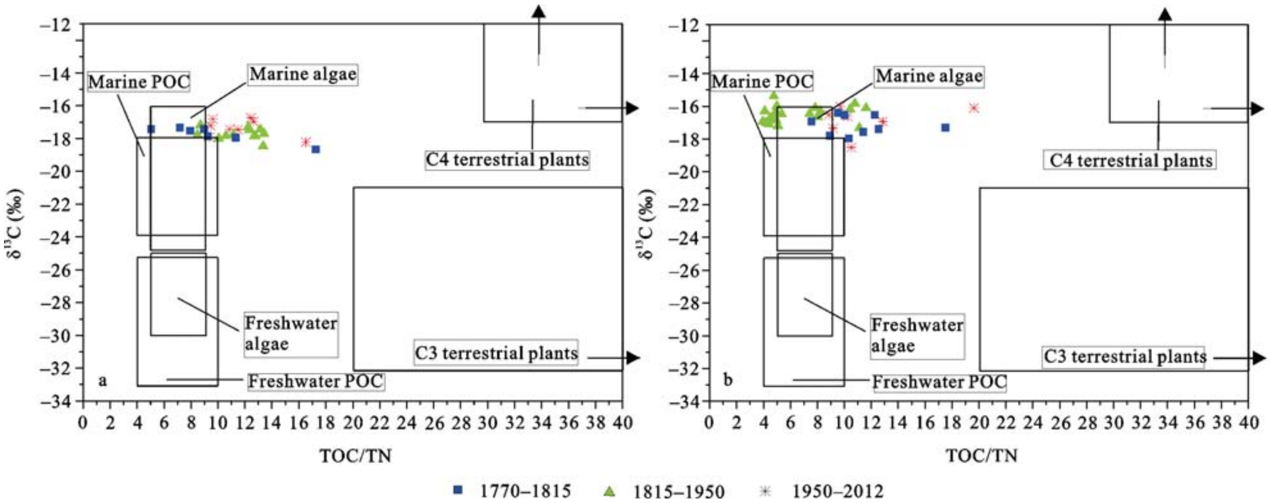


**Fig. 6** Identification of sources of organic matter with TOC/TN ratio and $\delta^{13}C$ from core XC-06 (a) and XC-09 (b). POC denotes particulate organic carbon

decreased the TOC/TN ratio at the same time (Fig. 4). Notably, the sharply decreased $\delta^{13}C$ and significantly increased TOC/TN ratio around 1965–1970 in the XC-09 (near the river mouth) indicate the rapidly increased terrigenous organic inputs, which may suggest the occurrence of storm events around 1970 (Zhou *et al*., 2016). Since ~1980, the seawater aquaculture area and aquatic production in Lingshui County have increased almost 5.1 and 15.4 times, respectively. The high concentrations of suspended organic solids, carbon, nitrogen and phosphorus in aquaculture effluents greatly enhanced the primary production of phytoplankton, macroalgae and epiphytic algae, but they also shaded seagrass and corals and deteriorated their health (Hughes, 1994; Hauxwell *et al*., 2001; Herbeck *et al*., 2013). Yang and Yang (2009) and Chen *et al*. (2015) reported a sharp declining trend in the seagrass bed area recently in Xincun Lagoon, where seagrass coverage has decreased from 66.0% to 36.2% in 2004–2012. As a result, the $\delta^{13}C$ value decreases with the declining supply of seagrass-derived organic matter. Meanwhile, the increased organic matter supply from phytoplankton and algae elevated the TOC/TN ratio. The sharp rise of the TOC/TN ratio after ~2005 in both cores was coincided with the decrease of seawater aquaculture area, which largely elevated the proportion of terrigenous organic matter preserved in lagoon sediments. Therefore, the environmental evolution of the lagoon since 1950 has been balanced by the changes of seagrass coverage and aquaculture activities.

### 4.3 Sources and burial flux of inorganic carbon

To understand the causes of variations of inorganic carbon burial, it is necessary to figure out the origin and source of inorganic matter in the lagoon. Compared to a relative low TOC content in both cores (0.09%–2.20%) (Fig. 3, Table 1), the TIC content was extremely higher, up to 3.52%–7.16%. Because of the lack of calcareous rocks and sandy coastal terrace surrounding Xincun Lagoon (Chinese Compilation Committee of China's Coast Embayments, 1999), the terrestrial inputs of inorganic carbon were limited. Zhang *et al*. (2015) reported that within the northern part of the South China Sea, the carbonate content of surficial sediments is less than 1% in general. The tropical lagoons without the growth of corals in Hainan Island have an ignorable low content of inorganic carbon (Shamei Lagoon) (Ge, 2006). In semi-enclosed coral reef lagoons, nearly 50%–90% of total $CaCO_3$ production of coral reefs could be deposited mud zones (Alongi *et al*., 2006). Thus, large numbers of corals living within this lagoon could be taken for the principal contributor to the carbonate materials deposited on the lagoon floor (Koop *et al*., 1987; Hansen *et al*., 1992; Alongi *et al*., 2006), as they produce a large amount of carbonate materials due to their high biological productivity (Alongi *et al*., 2006).

From 1950 to 2012, TIC in both cores experienced the similar changes (Fig. 3). To better quantify the potential associated with predicted factors (here, human activities), the burial flux of inorganic matter is calculated for two cores as follows:

$$M_0 = \rho_{dry} \times S \times C_s \tag{1}$$

where $M_0$ is the inorganic carbon burial rate (g/(m$^2$·yr)); $\rho_{dry}$ is the drying density of sediment (kg/m$^3$); $S$ is the sedimentation rate (m/yr); and $C_s$ is the inorganic carbon content in sediment in terms of mass (g/kg). The estimation of $\rho_{dry}$ is based upon the method adopted by Gao and Jia (2004):

$$\rho_{dry} = (\rho_s - \rho_w)/(w \times \rho_s + \rho_w) \tag{2}$$

where $\rho_s$ is the grain density of sediment (2650 kg/m$^3$); $\rho_w$ is the sea water density (taken as 1025 kg/m$^3$); and $w$ is the water content of buck sediments.

The calculated inorganic carbon burial rates in the lagoon are shown in Fig. 7. The overall inorganic carbon burial rates at XC-09 (375 g/(m$^2$·yr)) were much greater than those at XC-06 (230 g/(m$^2$·yr)), except for 1960–1990. This could be ascribed to the closer distance of the XC-09 site to center coral reefs distribution area. Since 1950, the burial rate decreased to below 200 g/(m$^2$·yr) until 1980, much lower than that in 1770–1815 (370 g/(m$^2$·yr)) and 1815–1950 (423 g/(m$^2$·yr)) at XC-09. The inorganic carbon burial rate also declined at XC-06 after 1950. The sharp decrease of inorganic carbon accretion rates was temporally coincided with the decline of coral species and coral reef coverage area in this lagoon. Based on the earliest recorded reports on coral reefs (Zou *et al*., 1975) and the interview with original chief investigators, the coral coverage area was estimated to be around 40%–50% in the 1960s (Fig. 1). The ecological investigation in 1970–1990 (Zhao *et al*., 1999; Chinese Compilation Committee of Embayment, 1999) showed that the coral coverage area in Xincun

Lagoon decreased to only 10%–20% in the 1980s. Recent surveys in 2013 indicated that few living corals existed in Xincun Lagoon (Fig. 7) and large areas of coral reef platforms have been covered by muddy sediments or replaced by aquaculture ponds. Therefore, the rapid decline of coral reefs could be responsible for the rapid decreases of inorganic carbon burial flux in lagoon sediments. It was also interesting to find that the inorganic carbon burial rate showed a gradual increase after ~1990 (Fig. 7), which was likely to be associated with the increasing seawater aquaculture activities (Fig. 1b, Fig. 5). The accelerated and disordered development of seawater aquaculture produced a quantity of carbonate debris to be deposit on the lagoon beds and mixed with sediments (Fig. 1c), which significantly altered the carbonate sedimentation flux and rate.

## 5 Conclusions

Combining total organic carbon and nitrogen, total inorganic carbon, stable carbon isotopes and sediment characteristics has allowed us to reconstruct the environmental evolution of Xincun Lagoon, a growing reef lagoon, southeastern Hainan Island. Based on these parameters and indicators, the environmental evolution can be divided into three stages. In 1770–1815, the decreasing water exchange capacity with outer open water, probably caused by the shifting and narrowing of the tidal inlet, not only diminished the currents and fined the sediments in the lagoon, but also reduced the organic matter of marine sources. From 1815 to 1950, the sedimentary environment of Xincun Lagoon was frequently influenced by storm events. These extreme events resulted in the high fluctuation of sediment grain size and sorting, as well as the great variation in contributions of terrestrial (higher plants, soils) and marine sources (phytoplankton, algae, seagrass). After 1950, the evolution of Xincun Lagoon was predominantly influenced by the increasing human activities. The source of the organic matter was greatly affected by the narrower distribution of seagrass and the increasing input of terrigenous organic matter, especially the boost of seawater aquaculture activities.

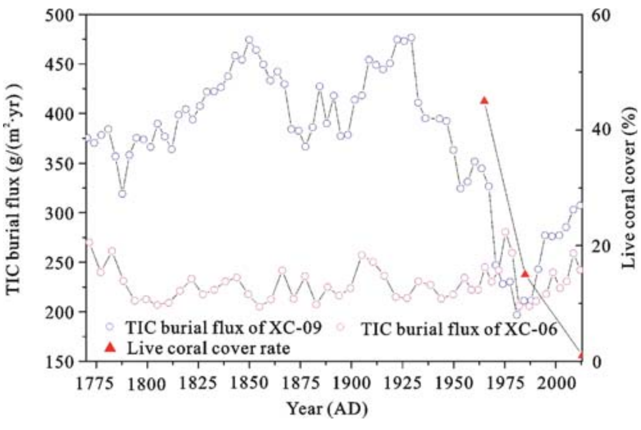


**Fig. 7** Variations of TIC burial flux and living coral coverage with time at core sites in Xincun Lagoon

In addition, the extremely high content of TIC (3.52%–7.16%) compared to TOC (0.09%–1.38%) in both cores before 1950 could be attributed to the wide distribution of coral reefs in this lagoon, which contributed a high inorganic carbon burial flux (375 g/(m$^2$·yr) in core XC-09 and 230 g/(m$^2$·yr) in core XC-06) to the lagoon beds. However, the rapid development of seawater aquaculture posted an extensive threat on the health growth of coral species and reduced the coral- derived inorganic carbon burial flux in the last 40 years, especially in the period of 1970–1990, as the effluents from aquaculture ponds deteriorated the water quality and shaded the coral reefs. Meanwhile, the concomitant carbonate debris produced by seawater aquaculture apparently enhanced the inorganic carbon burial rate in lagoon sediments since 1990.

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