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The potential of contribution of mangrove-derived organic matter in intertidal sediments as a proxy of mangrove development in the northern Beibu Gulf

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Abstract

Located between terrestrial and marine ecosystems, mangrove forests are sensitive to changes in climate. The responses of mangrove ecosystems to climate change in the future can be understood by reconstructing past mangrove dynamics using proxies preserved in the intertidal sediments. Considering the complexity of the proxies commonly used, it is necessary to develop a relatively simple, inexpensive proxy. In this study, available chemical tracers ($\delta^{13}C_{org}$ and C:N) of the four cores (YLW02, YLW03, O18, and Q37) from the intertidal zone of the northern Beibu Gulf (NBG) and a three-end-member (mangrove, sea grass, and suspended particulate matter) model was utilized to determine the contribution of mangrove-derived organic matter (CMOM) in carbonate-free sediments. Compared with the summed concentration of mangrove pollen (SCMP), a significant positive correlation between CMOM and SCMP is displayed. The calculated CMOM for an additional ^{210}Pb-dated sediment core from the Yingluo Bay, NBG (YLW01) clearly indicates a mangrove development going through degradation, flourishing, relative degradation, and relative flourishing, which are separately in correspondence with the lowest, highest, lower, and higher air temperature and rainfall in the time intervals of 1890–1918 AD, 1919–1956 AD, 1957–1990 AD, and 1991–2010 AD. This suggests that CMOM preserved in intertidal sediments has a potential to reconstruct historical mangrove development in high resolution, at the very least, along the coasts of the NBG.

Key words: mangrove-derived organic matter (CMOM), carbonate-free sediments, mangrove development, mangrove pollen, potential proxy

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1 Introduction

Located between terrestrial and marine ecosystems, mangrove forests are particularly sensitive to changes in climate, especially fluctuations in sea level (Ellison, 1993; Parkinson et al., 1994; Yulianto et al., 2004; Gillman et al., 2007). The response of mangrove ecosystems to future climate change can be understood by reconstructing past mangrove dynamics using signals preserved in the intertidal sediments from the tropical and subtropical coasts (Ellison, 2008). So far, the summed concentration of mangrove pollen (SCMP) has proven to be the most straightforward and effective proxy (Li et al., 2008) and is widely used to trace the historical development of mangrove forests during the Holocene (Wooller et al., 2007; Monacci et al., 2009; Li et al., 2012). However, this proxy requires more technical expertise and cost during sample fore-treatment and identification (Versteegh et al., 2004; Wooller et al., 2007; Li et al., 2008). Therefore, its use is limited in high-resolution studies of mangrove development, especially when the pollen concentration is very low. Taraxerol, a biomarker, was suggested as another useful proxy to trace past mangrove development decadal years ago (Versteegh et al., 2004). It has not been used worldwide for mangrove ecosystems due to its presence in a wide variety of higher-stand land plants (Vilegas et al., 1997; Cordeiro et al., 1999) rather than merely in the organisms of most mangrove species.

The contribution of mangrove-derived organic matter (CMOM) in carbonate-free sediments from sites along the tropical and subtropical coasts, where mangrove forests widely distribute, depends strongly on the growth status (flourishing/degradation) under relatively stable sea-level stands (Gonneea et al., 2004), and therefore it presumably has potential to be used as a proxy of mangrove development. In this study, this assumption was examined using available data of organic carbon stable isotope ($\delta^{13}C_{org}$), concentrations of total organic carbon (TOC) and nitrogen (TN), and mangrove pollen of the four cores (YLW02, YLW03, O18, and Q37) from the intertidal zone of the northern Beibu Gulf (NBG) (Xia et al., 2015b, 2017; Meng et al., 2016,

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2017). As a case of using this proxy, a new ^{210}Pb-dated sediment core (YLW01) from the Yingluo Bay, NBG was selected to reconstruct mangrove development in the bay over the past 120 years.

2 Regional settings

The coastal zones of the NBG (21°24′–22°01′N, 107°56′–109°47′E) are in the junction of the southwestern portion of Chinese mainland and the northern region of the gulf (Fig. 1a). These zones experience a tropical monsoon climate, with a rainy season between April and September and a dry season between October and March of next year. The average air temperature ranges from 22°C to 23°C, and the average annual precipitation is 2 000 mm. The tides on the coast are diurnal, with an average tidal height of 2.24 m (Jia et al., 2015). The average tide water salinity of four seasons is around 25, lower than that of the adjacent sea water (around 33) owing to the fresh water input from the coastal six rivers (Fan et al., 2011).

Six rivers, the Nanliu River, the Maoling River, the Qinjiang River, the Dafeng River, the Fangcheng River, and the Beilun River (Fig. 1a), discharge riverine loads into the NBG with average annual fluxes of 1.5×10^6 t, 5.5×10^5 t, 4.7×10^5 t, 3.6×10^5 t, 1.4×10^5 t, and 2.2×10^5 t, respectively (Fan et al., 2011), forming a series of intertidal shoals along the northern edge of the gulf.

Mangroves and seagrasses are the most typical ecosystems along the coast. Mangrove forests are broadly distributed in the intertidal zone, particularly concentrated in the Yingluo Bay, Tieshan Harbor, Lianzhou Bay, Qinzhou Bay, Zhenzhu Bay, and Beilun River Estuary (Fig. 1a). The mangrove forests are primarily composed of five pioneer species, *Aegiceras corniculatum*, *Kandelia obovata*, *Rhizophora stylosa*, *Bruguiera gymmorrhiza*, and *Avicennia marina*. Seagrass sporadically distributes in the Tieshan Harbor, Qinzhou Bay, Zhenzhu Bay, and Fangcheng Bay, which is primarily composed of *Halophila ovalis*, *Halophila beccarii*, *Zostera japonica*, *Halodule uninervis*, *Halodule pinifolia*, and *Halophila minor* (Fan et al., 2011).

3 Materials and methods

3.1 *Study sites and sampling*

In May 2010, five short sediment cores were collected from the intertidal zone of the NBG using PVC tubes with length of 120 cm and diameter of 10 cm. Q37 is located at bare flat in the northern Qinzhou Bay (Fig. 1b), while others (YLW01, YLW02, YLW03, and O18) are located at the core part of the Shankou Mangrove National Nature Reserve (Yingluo Bay) (Fig. 1c). Cores YLW02, YLW03, O18, and Q37 have been described in detail and their data (TOC, TN, $\delta^{13}C_{org}$and mangrove pollen) are available from Xia et al. (2015b, 2017) and Meng et al. (2016, 2017), so we select them as examined cores to verify the potential of CMOM as a proxy of mangrove development. Core YLW01 is selected as a case core to exhibit the effectiveness of using this proxy.

Core YLW01, with a length of 65 cm, was collected from the interior of the mangrove forest (21°29′49″N, 109°45′36″E), which is composed of *Kandelia obovata* and *Rhizophora stylosa.* Similar to other four cores, the core was cut into subsamples at intervals of 2 cm, and each of the subsamples was divided into four aliquots to permit measurement of $\delta^{13}C_{org}$, TOC, and TN, as well as moisture content in sediments and activities of ^{210}Pb. The numbers of samples used for measurements of moisture content and ^{210}Pb are 33 and 12, respectively. While the numbers of samples used for analyses of $\delta^{13}C_{org}$, TOC, and TN are 25, which was determined depending on the effective depth (49 cm) of excess ^{210}Pb ($^{210}Pb_{ex}$) (see following text).

3.2 *Analytical methods*

3.2.1 *Measurement of moisture content and DBD*

Around 20 g wet subsamples were put into small weighted boxes, and then totally weighted to determine their weights by subtracting the weights of boxes from total weights. Subsequently, the weighted wet subsamples were dried under temperature of 105–110°C for 10 h, and then were cooled completely to weight dried subsamples. Following the two steps above, the weight percents of moisture contents in each wet subsamples were determined by subtracting the weights of dried subsamples from those of wet subsamples. Under an assumption that the pore in sediment is full of seawater (Xia et al., 2015a), for compact (non-pore) intertidal sediments from the NBG, the dried bulk density (DBD, g/cm^3) of each subsample was calculated by

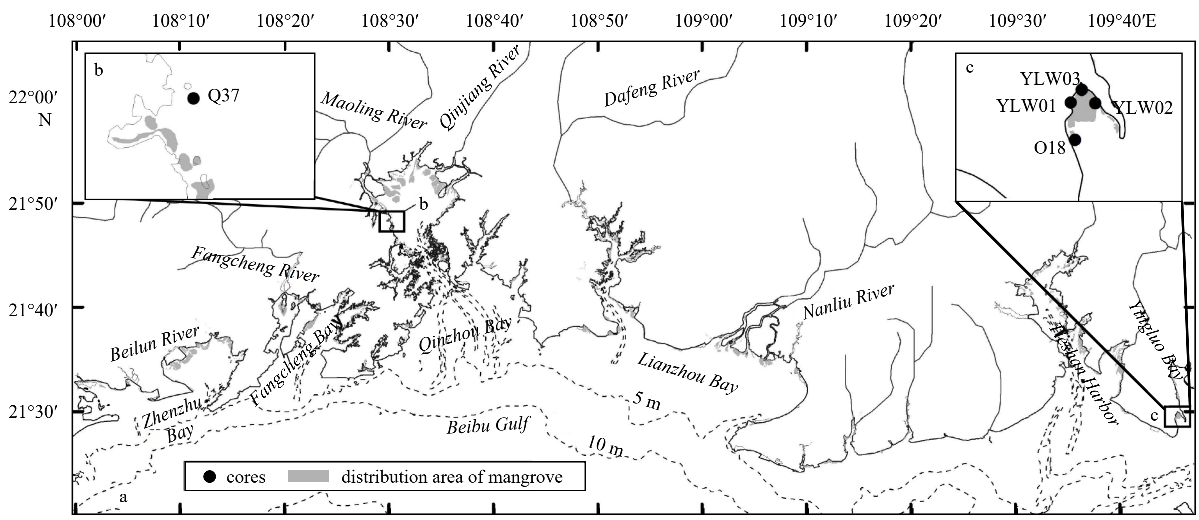

Fig. 1. A map showing the distribution of mangrove forests along the coast of the northern Beibu Gulf (a), and the sites of the sediment cores, of which YLW02, YLW03, O18(b), and Q37 (c) were used for examination of the effectiveness of the contribution of mangrove-derived organic matter (CMOM) as a proxy of mangrove development, YLW01 was used as a case of using CMOM as a proxy to reconstruct mangrove development in the Yingluo Bay.

the following empirical formula:

$$\mathrm{DBD} = (D_s \times C_w \times D_w) / [(D_w \times C_w + D_s \times (1 - C_w)], \quad (1)$$

where C_w represents moisture contents. D_s and D_w denote constant densities of non-pore shale and seawater, and are suggested to be 2.60 g/cm^3 and 1.05 g/cm^3, respectively (Xia et al., 2015a).

3.2.2 *Measurement of ^{210}Pb activity*

^{210}Pb activity was determined by analyzing the radioactivity of the decay product ^{210}Po at the Qingdao Institute of Marine Geology, assuming that both were at equilibrium (Moore, 1984). The Po was extracted, purified and self-plated onto Ag disks. ^{209}Po was used as a yield monitor and tracer in quantification. Samples were counted using computerized multi-channel α spectrometry with Au-Si surface barrier detectors. In order to obtain the excess ^{210}Pb ($^{210}Pb_{ex}$), measured ^{210}Pb activity was then corrected for ^{226}Ra-supported ^{210}Pb activity by subtracting the stable, low level activity at the bottom of the cores.

3.2.3 *Analysis of $\delta^{13}C_{org}$, TOC, and TN*

Aliquots of the subsamples were air-dried and finely grounded after shells and living biomass had been removed. These aliquots were then treated overnight with 1 mol/L HCl at room temperature to remove carbonates, followed by triple-washing with distilled water and oven-drying at 40°C (Wang et al., 2008). The stable organic carbon (OC) isotope compositions were determined in the Key Laboratory of Plant-Soil Interaction, Ministry of Education in China Agricultural University (Beijing), using a Delta Plus XP mass spectrometer in the continuous flow mode. The ^{13}C to ^{12}C ratios are reported in δ-notation as relative differences in parts per thousand ($\delta^{13}C_{org}$) from the V-PDB standard. The precision of the $\delta^{13}C_{org}$ measurements was better than 0.20‰. The TOC and TN concentrations were determined in the Key Laboratory of Marine Geology and Metallogeny, Ministry of Natural Resources, using a Vario EL-III Elemental Analyzer. Replicate weight analyses of one standard sample (GSD-9) provided a precision value of ±0.02% for TOC and ±0.005% for TN.

3.3 *Quantitative partition of the preserved organic matter according to their sources*

3.3.1 *Potential sources of organic matter in intertidal sediments from the NBG and their end-member values of $\delta^{13}C_{org}$ and atomic C:N*

In general, the organic matter (OM) stored in intertidal sediments of mangrove ecosystems originates from mangrove litters, fine roots, microbes (bacteria), seagrass debris, and suspended particulate matter (SPM) (Bouillon et al., 2004; Gonneea et al., 2004; Robertson and Alongi, 2016), which is usually a mixture of terrestrial debris and marine phytoplankton in some proportion (Bouillon et al., 2003), especially for mangrove systems with catchments. However, the contribution of OC from bacteria only accounts for less than 1% of the TOC in mangrove sediments, and hence should be ignored for the purpose of OM source discrimination (Bouillon et al., 2004). Therefore, the end-members of OM source in the bulk sediments of the Yingluo Bay and the Qinzhou Bay were reasonably regarded as mangrove-derived input, seagrass, and SPM.

Since fine roots have $\delta^{13}C_{org}$ values within the range of the $\delta^{13}C_{org}$ values of litters from different mangrove species (Badeck et al., 2005; Wei et al., 2008), we only selected senescent leaves from the two bays to represent the mangrove-derived OM end-member, for which the averaged $\delta^{13}C_{org}$ values and atomic C:N ratios have been regarded as –28.4‰ and 33.6, respectively, by Xia et al. (2015b) and Meng et al. (2016); the averaged values of $\delta^{13}C_{org}$ and atomic C:N ratio of the 14 SPM samples collected from adjacent sea water in the NBG and of 19 seagrass (leaves and roots) samples collected from the coast of the NBG are –21.3‰ and 5.52 (He et al., 2014), and –13.5‰ and 27.2, respectively (Fan et al., 2011). These average values of the $\delta^{13}C_{org}$ and the atomic C:N ratios were cited as end-member values of the mangrove, SPM, and seagrass OM sources, respectively.

3.3.2 *Quantitative partition of the OM contribution from different sources using a three-end-member model*

A three-end-member model (Dittmar et al., 2001; Gonneea et al., 2004), characterized by a ternary mixing diagram depicting $\delta^{13}C_{org}$ and C:N of three OM sources, can be used to discriminate the true provenances of the OM in sediment cores collected from the two bays. In this model, each of mangrove, SPM and seagrass OM sources occupies a corner of a triangle (ΔMSG) (a strict triangle), and an expanded triangle (ΔM′S′G′) that is similar to the strict triangle with a given tolerance interval (τ), which was determined as a averaged value (30%) of atomic C:N variation coefficients of the five sediments cores, was jointly employed to determine OM sources for these sediment cores.

When the samples of the sediment Cores YLW02, YLW03, O18, and Q37 are plotted on the ternary plot, it can be seen that 146 of the total samples (n=153) of the four sediment cores fall within ΔM′S′G′ (Fig. 2a), indicating that 95% of the total samples can be explained in terms of three-end-member model. According to the rules for discrimination of true OM provenances (Dittmar et al., 2001; Gonneea et al., 2004), the falling of 30 and 19 samples separately from Cores O18 and Q37 within the ΔMSG indicates 71% and 49% samples of the two cores can be explained in terms of mangrove, SPM, seagrass sources; the falling of 25, 22, 16, 4 samples separately from Cores YLW02, YLW03, Q37, and O18 within the expanded area limited by Lines SM and S′M′ indicates 100%, 88%, 39%, and 10% samples of these cores can be explained in terms of mangrove and SPM sources; merely 5 and 3 samples separately from Cores O18 and Q37 fall within the expanded area limited by Lines MG and M′G′, indicating 12% and 7% samples of the two cores can be explained in terms of mangrove and seagrass sources. Other samples that fall completely outside of the expanded triangle area indicate the presence of additional OM sources or diagenetic alterations of the original signal and cannot be characterized by the three-end-member model. For Core YLW01, all samples (n=25) fall within the expanded area limited by Lines SM and S′M′ of ΔM′S′G′ (Fig. 2b), indicating that 100% samples of this core can be explained in terms of mangrove and SPM sources.

The OM contributions of the samples that can be explained by two sources (mangrove and SPM, or mangrove and seagrass) can be calculated using the isotopic mass-balance equation (Calder and Parker, 1968; Schultz and Calder, 1976). For the samples that can be explained by three sources (mangrove, SPM, and seagrass), their OM contributions can be calculated using the isotopic mass-balance equation combined with an mass-conservation equation of C:N under limit that the total OM contribution from three sources is equal to 100% (Goñi et al., 1997; Hu et al., 2006; Ramaswamy et al., 2008).

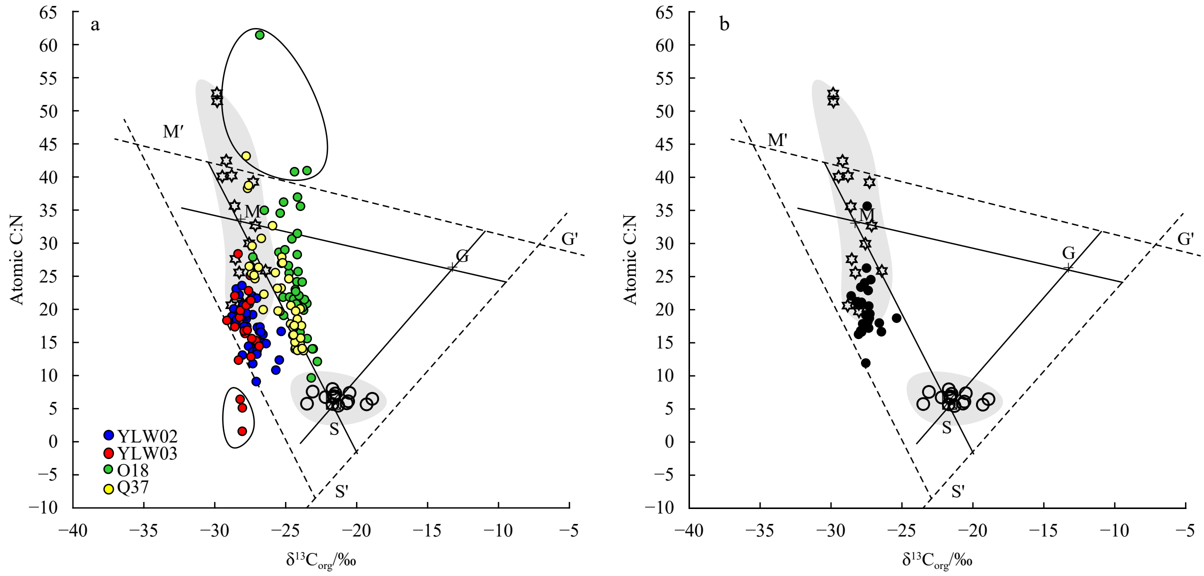

Fig. 2. Plots of the atomic C:N ratios versus $\delta^{13}C_{org}$ used to differentiate the sources of the preserved organic matter in the four examined sediment cores (a) and in the case Core YLW01 (b). The capital letters, M, S, and G, represent the mangrove, SPM, and seagrass end-members, respectively; ΔMSG is the strict triangle determined by the end-member values of the three potential sources; and ΔM′S′G′ is the extended triangle, similar to ΔMSG, with tolerance intervals of 30%.

4 Results

4.1 *Results for examination of CMOM as a proxy of mangrove development*

4.1.1 *Variations in contributions of three OM sources in the examined cores*

The calculated results for the four sediment cores indicate that the CMOM in the sediments ranges from 57% to 100% with an average of 88% for YLW02 (Fig. 3a), from 78% to 100% with an average of 92% for YLW03 (Fig. 3b), from 21% to 87% with an average of 55% for O18 (Fig. 3c), and from 35% to 92% with an average of 61% for Q37 (Fig. 3d). The contributions of SPM (CSPOM) to the sediments ranges from 0% to 43% with an average of 12% for YLW02 (Fig. 3a), from 0% to 22% with an average of 8% for YLW03 (Fig. 3b), from 0% to 79% with an average of 35% for O18 (Fig. 3c), and from 0% to 65% with an average of 35% for Q37 (Fig. 3d). The contributions of OM from seagrass (CSGOM) range from 0% to 30% with an average of 10% for O18 (Fig. 3c) and from 0% to 18% with an average of 4% for Q37 (Fig. 3d).

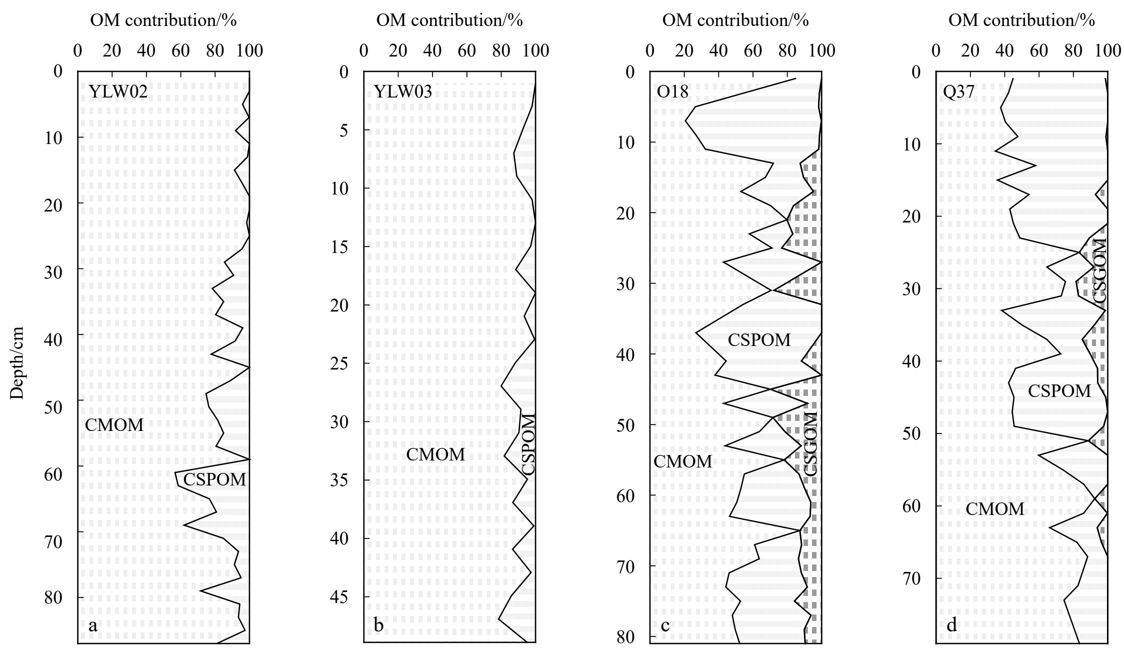

Fig. 3. Vertical distributions of the contributions OM from mangrove litters (CMOM), suspended particulate matter (CSPOM), and seagrass (CSGOM) sources for the four examined sediment cores.

4.1.2 *Relationships between CMOM and SCMP for the examined cores*

Because the SCMP has been widely accepted as a most effective proxy of mangrove development, the relationships between CMOM and SCMP for Cores YLW02, YLW03, O18, and Q37 were examined through their parallelisms of vertical distributions, in combined with their statistical correlations.

For the four sediment cores, the vertical distributions of CMOM are approximately parallel to those of the SCMP. For YLW02, CMOM exhibits a three-step variation from the bottom of the core to 70 cm, from 70 cm to 44 cm, and from 44 cm to the surface, with averages of 89%, 79%, and 94%, respectively, separately corresponding to the SCMP average values of 23%, 13%, and 46% (Fig. 4a). Very similar to YLW02, the CMOM in YLW03 also shows a three-step variation from the bottom of the core to 36 cm, from 36 cm to 24 cm, and from 24 cm to the surface, with averages of 91%, 86%, and 95%, respectively (Fig. 4b), separately corresponding to the SCMP average values of 23%, 17%, and 25% (Fig. 4b). Because Core O18 is located just at the margin of a mangrove forest in the Yingluo Bay, both CMOM and SCMP in the sediment of this core were much lower than those of Cores YLW02 and YLW03, and a five-step co-variation is seen from the bottom to 66 cm, from 66 cm to 34 cm, from 34 cm to 14 cm, from 14 cm to 4 cm, and from 4 cm to the surface (Fig. 4c), with average values of 56%, 51%, 65%, 27%, and 70%, respectively, for CMOM, separately corresponding to the average values of 13%, 11%, 13%, 6%, and 8% for the SCMP. For Q37, both CMOM and SCMP exhibit three-step decreases from the bottom to 50 cm, from 50 cm to 26 cm, and from 26 cm to the surface, with averages of 80%, 57%, and 44%, respectively, separately corresponding to the SCMP average values of 14%, 10%, and 8% for the total pollen (Fig. 4d).

The parallelism between CMOM and SCMP for each sediment core is also reflected by their individual correlation coefficients, which are 0.73 (P=0.01, n=44), 0.42 (P=0.05, n=25), 0.67 (P=0.01, n=14), and 0.87 (P=0.01, n=10) for YLW02, YLW03, O18, and Q37, respectively (Figs 4a–d). A better logarithmic fit (R=0.69, P=0.01, n=93) for the array of all samples of the four cores in a scatter plot of CMOM vs. SCMP further indicates there is a significant positive relationship between CMOM and SCMP (Fig. 5).

4.2 *Results for effectiveness of using CMOM as a proxy of mangrove development in the Yingluo Bay*

4.2.1 *^{210}Pb chronology of Core YLW01*

The measured total ^{210}Pb activity ($^{210}Pb_{tot}$) clearly exhibits a decrease from the surface (7 cm) to the depth of 49 cm and stabilizes between 55 cm and 61 cm, with an average of 0.54 dpm/g (Fig. 6a). This indicates 49 cm is the effective depth for ^{210}Pb-dating and $^{210}Pb_{ex}$ for the section from the surface to 49 cm can be obtained by subtracting 0.54 dpm/g from the $^{210}Pb_{tot}$. The DBD slightly and abruptly increases from the 49 cm to the bottom of the core, in contrast to the variation in $^{210}Pb_{tot}$. This indicates $^{210}Pb_{tot}$ is influenced by sedimentary compaction and the depths corresponding to $^{210}Pb_{ex}$ layers need to be corrected using the technique suggested by Lynch et al. (1989). According to the profile

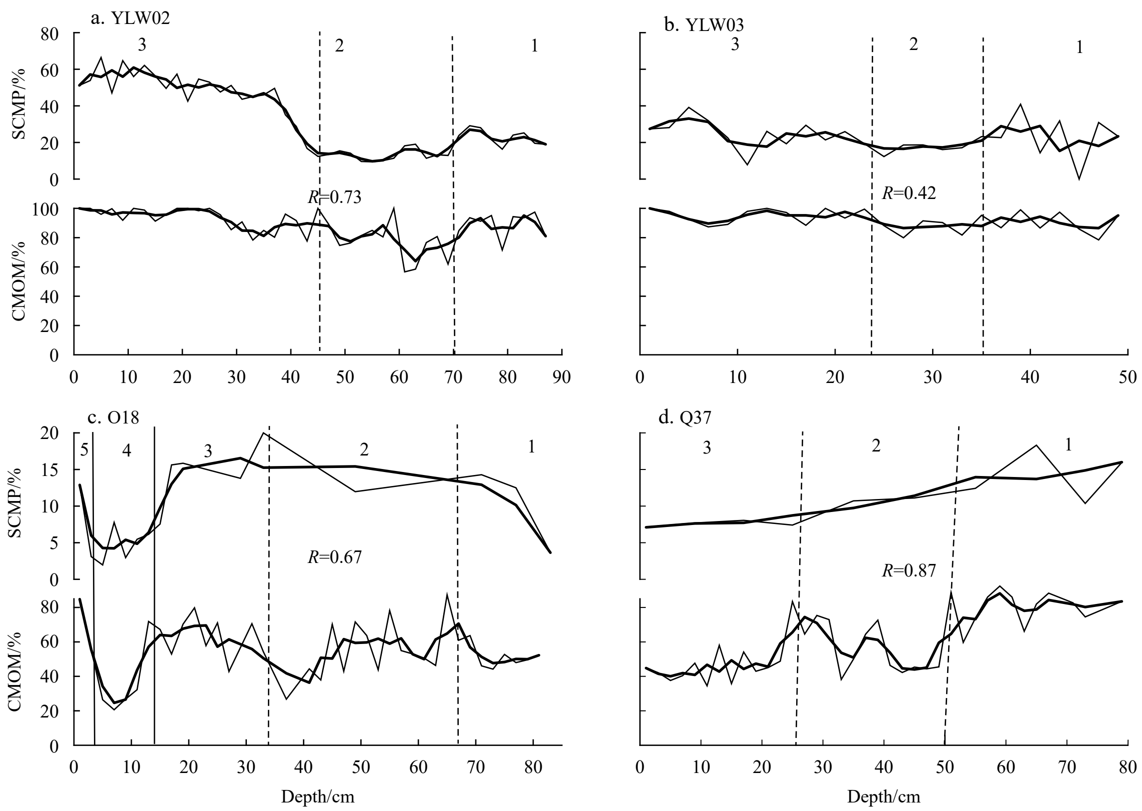

Fig. 4. Separate comparison between the vertical distribution of the contribution of mangrove-derived organic matter (CMOM) and the summed concentration of mangrove pollen (SCMP) for the four examined sediment cores (YLW02, YLW03, O18, and Q37), showing an obvious parallelism of CMOM to SCMP with significant positive correlation. The bold lines denote the three-point smoothed CMOM and SCMP for each sediment core. The upper number denotes the stages of co-variation between CMOM and SCMP.

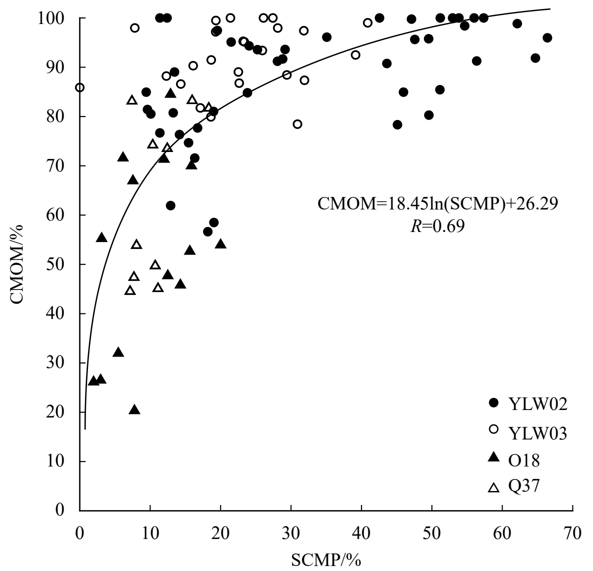

Fig. 5. Plot of CMOM vs. SCMP for the total samples of four examined sediment cores, showing a more significant positive correlation.

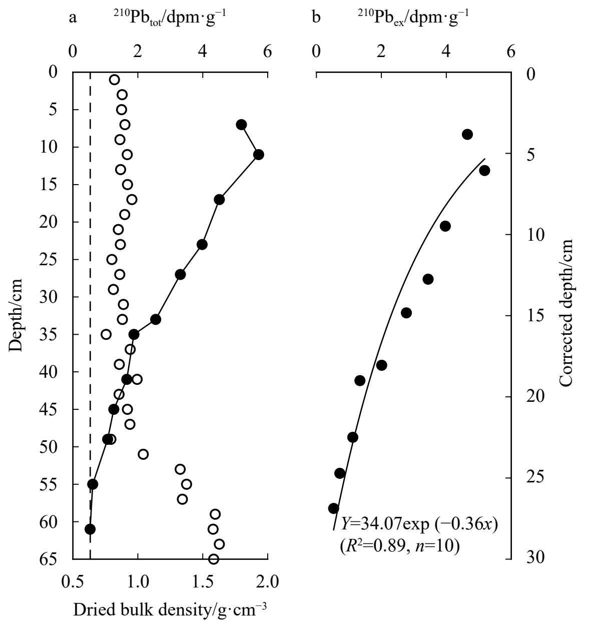

Fig. 6. Down-core variations in total ^{210}Pb ($^{210}Pb_{tot}$) and dried bulk density of the case core (YLW01) (a), in which, the solid cycles and blank cycles represent the activities of total ^{210}Pb ($^{210}Pb_{tot}$) and dried bulk densities, respectively; and excess ^{210}Pb ($^{210}Pb_{ex}$) against corrected depth (b).

of $^{210}Pb_{ex}$ activity versus compaction-corrected depth (Fig. 6b), the averaged sedimentation rate (SR) at the site of Core YLW01 was determined as 0.36 cm/a. Based on the average SR of the coring site and time (2010 AD), the effective age node for Core YLW01 was determined as around 1890 AD, meaning that the core archives the mangrove development over the last 120 years in the Yingluo Bay.

4.2.2 *Variation in CMOM of Core YLW01 and its indication for mangrove development in the Yingluo Bay*

Using the analyzed $\delta^{13}C_{org}$, the CMOM in the sediment Core YLW01 was calculated following isotopic mass-balance equation and mass-conservation equation of C:N. It can be seen that the CMOM exhibits four-stage variation from 1890 AD to 2010 AD: from 1890 to 1918 AD, from 1919 to 1956 AD, from 1957 to 1990 AD, and from 1991 to 2010 AD, with the lowest, highest, lower, higher averaged values of 43%, 76%, 64%, and 73%, respectively (Fig. 7a), very similar to that in SCMP calculated from regressed equation for the four examined cores (Fig. 7b). The variation in CMOM, in combined with that in SCMP, indicates mangrove in the Yingluo Bay has gone through degradation, flourishing, relative degradation, and relative flourishing from 1890 to 1918 AD, from 1919 to 1956 AD, from 1957 to 1990 AD, and from 1991 to 2010 AD, respectively (Fig. 7c). This development is exactly in correspondence with the lowest, highest, lower, and higher air temperature (Fig. 7d) and rainfall (Fig. 7e) for each of four time intervals.

5 Discussion

As illustrated above, the CMOM is a derivative proxy calculated from indicators of OM sources. To spread its use in reconstruction of mangrove development, there are a few notes and arguments still needed to discuss.

5.1 *The universality of three-end-member model used for discrimination of OM sources in mangrove sediments*

For most mangrove systems worldwide, the OM in sediments can usually be considered to be a mixture of two or three sources, and the contribution from each source can be calculated using a three-end-member model. However, for some cases, where the number of potential OM sources is greater than three and hence the three-end-member model is not applicable, the number of OM sources should be reasonably simplified or integrated to be two or three, such as in this study (where the terrestrial input and marine phytoplankton were integrated into SPM), making the three-end-member model universally applicable.

5.2 *The universality and limitation of $\delta^{13}C_{org}$ as an indicator of OM sources in mangrove sediments*

The critical precondition for differentiation of OM sources in the mangrove sediment is that the end-member values of the used indicators of the potential OM sources must be significantly distinguishable from each other. For mangrove systems without catchments, the potential sources of preserved OM in sediments usually involve mangrove litters, seagrass, and marine phytoplankton (micro- and macro-algae) (Gonneea et al., 2004; Bouillon et al., 2008), each of which have relatively stable but distinctive $\delta^{13}C_{org}$ values of around 28.1‰, around 20‰, and around 12.1‰, respectively, even though they are likely to vary slightly with local environments (Bouillon et al., 2008) and be affected by early diagenesis (Wooller et al., 2003). Therefore, $\delta^{13}C_{org}$ is considered to be a valid OM indicator. For mangrove systems with catchments, the terrestrial (fluvial) input is an important source of preserved OM in sediments, which makes the discrimination of the OM source more complicated due to the dependence of terrestrial OM on the proportion of plant types (C_3:C_4 ratio) in the catchments. Only in situations where the catchments are dominated by C_4 plants and the C_3:C_4 ratios do not strongly change with the time, can the $\delta^{13}C_{org}$ value be used to indicate the OM source of the mangrove sediments? Fortunately, previous studies have illustrated that C_4 plants are widely distributed on the land in the tropics and subtropics, where mangrove forests are distributed in the intertidal zones due to the hot climate (Huang et al., 2001; Rao et al., 2010). If C_3 plants are predominate in the catchments, such as for the Caeté River in northern Brazil where the $\delta^{13}C_{org}$ value range of the terrestrial plants (–27.4±0.8)‰ overlaps that of

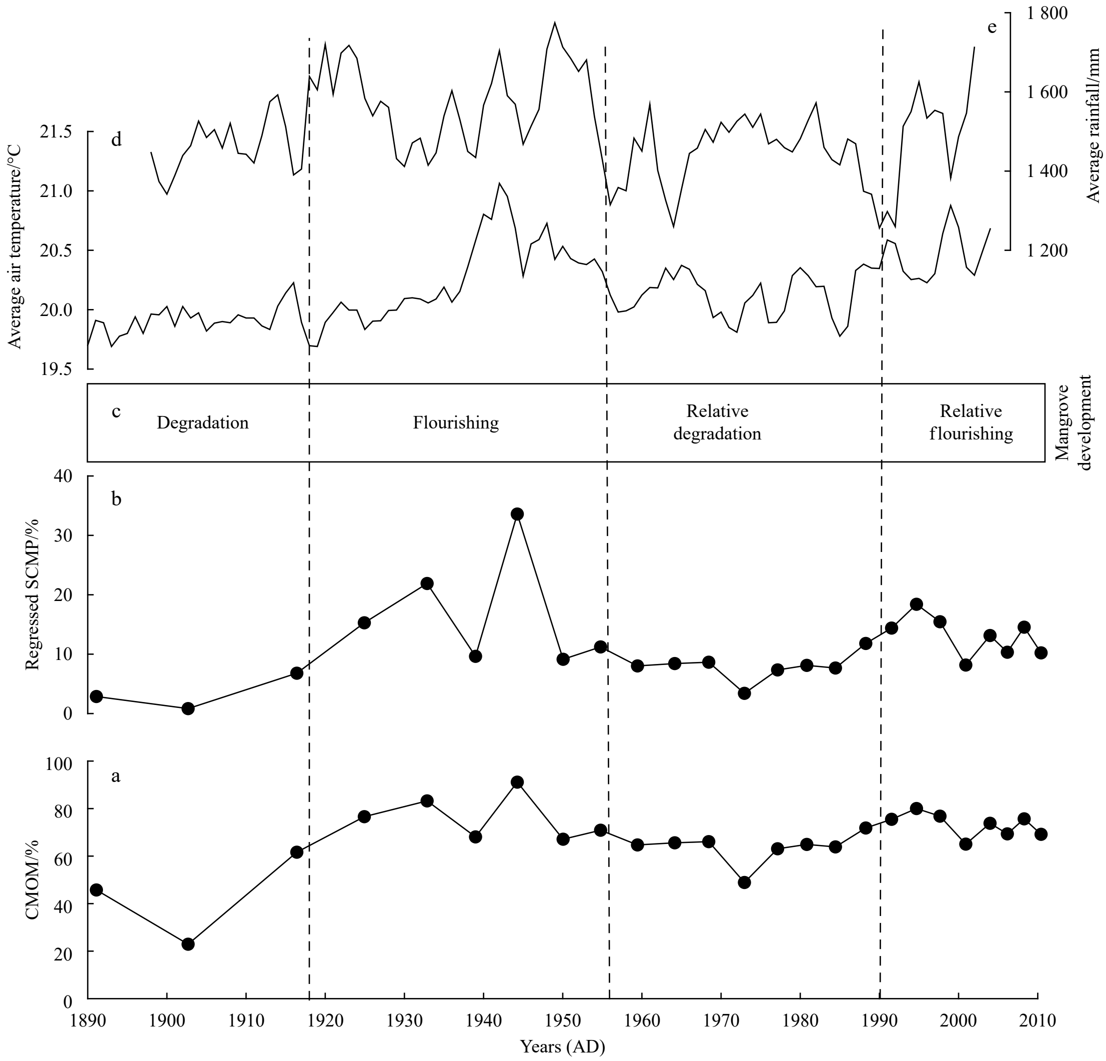

Fig. 7. Four-stage variations in CMOM (a) and regressed SCMP (b), and their indication of mangrove development since 1890 AD (c), in comparison with the changes of averaged air temperature (d) and rainfall (e).

the mangrove forests (–28.1±1.5)‰, then $\delta^{13}C_{org}$ can only be used as an assistant indicator and other indicators, such as lignin, need to be used as the key indicator of OM source of the mangrove sediments (Dittmar et al., 2001).

5.3 *The argument against the use of C:N ratio as one of OM sources indicators in mangrove sediments*

$\delta^{13}C_{org}$ alone is not sufficient to qualitatively discriminate OM sources and quantitatively estimate their contributions in sediments for most mangrove ecosystems with OM inputs from three sources, and other assistant tracers should be combined with $\delta^{13}C_{org}$ in these cases. One widely used assistant tracer is atomic C:N ratio (Andrews et al., 1998; Lamb et al., 2006; Bouillon et al., 2008). However, there have been arguments against the atomic C:N ratio used as an indicator for discriminating OM sources in marine sediments due to its post-deposition alteration (Andrews et al., 1998; Yamamuro, 2000; Graham et al., 2001). In general, the variation in the C:N ratios of sedimentary OM over time or with depth in a core exhibits three patterns: increases, small changes, and decreases. Increases have been interpreted as indicative of the preferential loss of N via processes including ammonification, nitrification, and denitrification (Andrews et al., 1998), while small changes suggest that C and N are mineralized or preserved at the same rate. Decreases in C:N ratios have been explained as the result of absorption of organic or inorganic N onto silicate clay surfaces (Macko et al., 1993) or the incorporation of N by bacteria into decaying OM (Cifuentes et al., 1996). However, if small changes in the C:N ratio are observed between the sources and the decaying OM pools, as in most mangrove systems where the decomposition rate is diminished and N can be preserved due to anoxic or tannin-rich depositional environments (Gonzalez-Farias and Mee, 1988), this element ratio can be used as a tracer of OM sources (Gonneea et al., 2004). In fact, the atomic C:N ratios of sediment cores from most mangrove forests co-vary with $\delta^{13}C_{org}$ in depth with significant negative correlations rather than a monotonous increase or decrease, such as those studied here (Fig. 2), mangrove sediments from the Ba Lat Estuary in Vietnam (Tue et al., 2011), and sediments from Ilha do Cardoso in southeastern Brazil (Pessenda et al., 2012). This indicates that the C:N ratio is a valid assistant indicator of OM sources for most mangrove sediments.

5.4 *The argument against the use of CMOM as a proxy of mangrove development*

The calculated CMOM depends not only on the status (flourishing or degrading) and change in the habitat of mangrove forests but also on the contributions from other organic sources. The existences of parallelism of the vertical distribution of CMOM in each separate sediment core to SCMP with a significant linear correlation (Fig. 4) and of a better positive correlation between CMOM and SCMP for the total samples (Fig. 5). This suggests that CMOM depends primarily on the status and habitat of the mangrove forest and can be used as a proxy for mangrove development if the core sediments are dated. The calculated CMOM in Core YLW01 (Figs 7a, c) indicates a mangrove development going through degradation, flourishing, relative degradation, and relative flourishing, which are separately in correspondence with the lowest, highest, lower, and higher air temperature and rainfall in the time intervals of 1890–1918 AD, 1919–1956 AD, 1957–1990 AD, and 1991–2010 AD. The exact correspondence truly reflects the fact that mangrove plants prefer to grow under a warmer and wetter condition. This further validates that CMOM in the intertidal sediment core can be used as a proxy of mangrove development at least along the coasts of the NBG.

5.5 *The strengths and weaknesses of CMOM used as a proxy of mangrove development*

Any proxy used to trace mangrove development has its strengths and weaknesses. Compared to SCMP that has been widely used to retrospectively study mangrove forests (Monacci et al., 2009; Ellison, 2008; Li et al., 2008, 2012), the strength of CMOM includes relatively simple chemical pre-treatment, inexpensive instrumental analyses of $\delta^{13}C_{org}$ and concentrations of TOC and TN for a large amount of fine-cut sub-samples. In particular, the CMOM is calculated for bulk carbonate-free sediments rather than only for coarser fraction of sediments like SCMP (greater than 10 μm) (Li et al., 2008, 2012), and hence the CMOM is usually much higher than SCMP, which derives another strength of CMOM for reconstruction of mangrove development in the case of less mangrove pollen kept in sediments. These strengths provide a possibility for reconstruction of mangrove development in higher resolution. The weakness of CMOM is that it cannot trace the community structure of mangrove forests as pollen assemblages of mangrove species do. Therefore, the best strategy to more perfectly reconstruct mangrove development is the use of multiple proxies.

As a potential proxy for tracing mangrove development, the CMOM indeed needs to further prove its reliability via testing for mangrove forests worldwide. At the very least, however, this proxy is reliable for most mangrove forests located in sites similar to the NBG.

6 Conclusions

To validate the use of CMOM in bulk sediments as a proxy for mangrove development, the correlation between CMOM and SCMP was examined using available data of $\delta^{13}C_{org}$, TOC, TN, and mangrove pollen of four sediment cores from the intertidal zone of the NBG, where mangrove forests are broadly distributed. The results show that the vertical distribution of CMOM parallels that of SCMP with a significant positive correlation for each sediment core. There is an obvious logarithmic relationship between CMOM and SCMP for the integrated samples of the four sediment cores, with a much more significant positive correlation coefficient than for the individual cores. Using CMOM combined with ^{210}Pb-age model of a sediment core (YLW01), mangrove development going through degradation, flourishing, relative degradation, and relative flourishing in the Yingluo Bay over the past 120 years was successfully reconstructed. This indicates that CMOM in bulk sediments from the intertidal sites is similar to the NBG, which can be used as a proxy of mangrove development if an age model is established. This proxy allows mangrove development to be studied in high resolution owing to its simple chemical treatment, inexpensive instrumental analyses of $\delta^{13}C_{org}$ and concentrations of TOC and TN for a large amount of fine-cut sub-samples. At the very least, CMOM can be used as one of multiple proxies to trace mangrove development along the coasts of the NBG.

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