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Source and sequestration of sediment organic carbon from different elevation zones in estuarine wetland, North China

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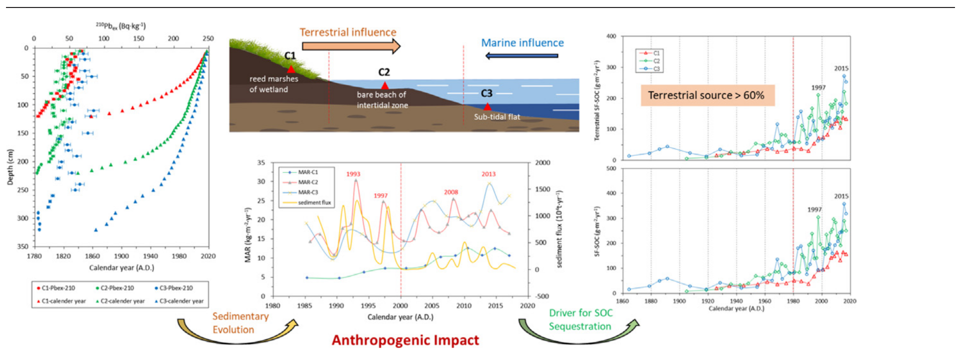
HIGHLIGHTS

- The $^{210}Pb_{ex}$ chronologies in sediments over the past century were established.
- SOC source showed that the terrestrial input was dominant.
- Increasing MAR linked to changes from riverine input and land use since the 1980s.
- SF-SOC dominantly controlled by MAR, and changed following elevation and TOC.

GRAPHICAL ABSTRACT



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ABSTRACT

Estuarine wetland plays an important role in regulating global carbon cycle due to high terrestrial carbon input and burial. However, it is unclear how the source and sequestration of sediment organic carbon (SOC) in estuarine wetlands changes under the anthropogenic impact in the past century. In this study, combining parameters of TOC/TN ratios, $\delta^{13}C$, $\delta^{15}N$ and ^{210}Pb-chronology, temporal trends of SOC source and sequestration flux in Liaohe estuarine wetland were studied. The results showed that the source of organic carbon in Liaohe estuarine wetland was dominated by terrestrial input (contribution >60 %). Due to vegetation, TOC in shallow reed marsh was significantly higher than that of bare beach and subtidal flat. Affected by elevation, the sediment mass accumulation rate (MAR, $kg{\cdot}m^{-2}{\cdot}yr^{-1}$) showed differences in reed marsh (C1), bare beach (C2) and subtidal flat (C3), which were 6.57, 13.56 and 13.25 respectively in the past century. MAR fluctuated over time, it showed an overall increasing trend, especially since the 1980s. Correspondingly, the sequestration flux of SOC (SF-SOC, $g{\cdot}m^{-2}{\cdot}yr^{-1}$) showed an overall increasing trend with average of 82.84 (reed marsh), 151.93 (bare beach) and 123.71 (subtidal flat). Comparing to TOC, the higher MAR had a more distinct effect on carbon sequestration in Liaohe estuarine wetland. The difference in sedimentation rate and carbon sequestration are linked to the changes in sediment flux of riverine input and land utilization in the catchment area due to human activities in recent decades, including the construction of reservoirs, dams and local ditch wharf.

1. Introduction

Located between terrestrial and marine ecosystem, the estuarine wetland is a vital part of the coastal habitats (Sutula et al., 2001). Coastal vegetated habitats, which are only around 0.2 % of the ocean surface in size, can contribute 50 % of carbon burial in marine sediments (Duarte et al., 2013). The burial of carbon is believed to be one of the most cost-effective

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ways to mitigate CO_2 increase (Janzen, 2004; Xu et al., 2011). This highlights the important role that estuarine wetland plays in the context of the global carbon cycle as well as supporting climate change and adaptation policies (Duarte et al., 2005; Macreadie et al., 2014). Thus, a carbon storage index has been proposed to estimate the ability of wetlands as a carbon sink which is one of the goals from the Ocean Health Index (Halpern et al., 2012). Over last decades, a large number of studies have been carried out on the fate of organic carbon in coastal habitats (Callaway et al., 2012; Kirwan and Mudd, 2012; Ye et al., 2015), especially in estuarine wetlands, including mangroves and saltmarshes (Chmura et al., 2003; Palomo and Niell, 2009). It was reported that mangroves ecosystem contains carbon stocks as average of 956 $Mg \cdot ha^{-1}$ and saltmarshes contain 593 $Mg \cdot ha^{-1}$ (Alongi, 2014), while their averaged carbon sequestration rates are about 226 $g \cdot m^{-2} \cdot yr^{-1}$ and 218 $g \cdot m^{-2} \cdot yr^{-1}$, respectively (Nelleman et al., 2009). However, these stocks and sequestration vary widely with the changes in geographical locations, vegetation type, wetland age, etc. (McLeod et al., 2011; Osland et al., 2012). Meanwhile there is also local variability with the changes in elevation and vegetation density. For example, low marshes near the coastline have greater organic carbon sequestration capacity than high marshes due to high primary productivity and sediment accretion (Callaway et al., 1996; Radabaugh et al., 2017). In addition, the changes in land utilization and other anthropogenic disturbance can also alter the natural process of estuarine wetlands, and threaten the preservation of vegetation and ecological function (Barbier et al., 2011; Dahl et al., 2022), resulting in the oxidation of organic carbon and the sink-source conversion of carbon dioxide (Nelleman et al., 2009). Thus, the recognition and quantification of organic carbon source and sequestration are of great significance for a better understanding of spatial variability in coastal carbon cycle process in estuarine wetland as a result of preservation, degradation, and construction under anthropogenic impact (Allison et al., 2007; Krishna et al., 2013; Hu et al., 2016; Radabaugh et al., 2017; Pellegrini et al., 2021).

Liaohe estuarine wetland is a typical coastal wetland affected by riverine input and human land use, it's located in the alluvial plain of the lower Liaohe River, its coastline has moved in and out frequently and the vegetation biocoenosis are complex and diverse (Zuo et al., 2013). In recent years, the sediment sources, components and structure have shifted, and the vegetation has undergone continuous degradation due to human activities such as upstream river water cutoff and seawall dam construction (Zhao et al., 2016). Consequently, the characteristics of sedimentation, as well as organic carbon input and sequestration in Liaohe estuarine wetland have been strongly influenced. There had been a few studies on the transport of suspended sediment and evolution history of Liaohe River delta (Zhu et al., 2010), as well as the sediment organic carbon stocks and carbon accumulation rate in some part of wetlands of Liaohe River delta (Ye et al., 2015; Zhao et al., 2016). However, the sources and relative composition of organic carbon buried in sediment from different elevation zones are not well understood. Moreover, the influence of river discharge on sedimentation and organic carbon sequestration is poorly investigated, especially under the pressure of human activities such as upstream river water cutoff and seawall dam construction happening since the 1980s. So, the focus of this study is on the sediment organic carbon processes from different elevation zones in Liaohe estuarine wetland, including reed marshes, bare beach of intertidal zone and subtidal flat. The objectives are to (I) calculate sedimentation rate by excess ^{210}Pb chronology, (II) investigate the variation of organic carbon sources using carbon and nitrogen elements and their stable isotopes, and (III) reveal the temporal and spatial variation of organic carbon sequestration flux under the influence of vegetation and anthropogenic disturbance. Findings from this study could be helpful in the evaluation of carbon sink capacity in estuarine wetlands, particularly in response to continuous human pressure.

2. Material and methods

2.1. Study area and sampling

Located in northeastern China, landforms of Liaohe estuarine wetland were formed by accretion of sediment from Liaohe River which is 1396 km long with a drainage area of 2.19×10^5 km^2 (Zhao et al., 2016). The reed marsh of Liaohe estuarine wetland represents one of the largest reed fields in the world (Brix et al., 2014).

In this study, three sediments were collected from the west side of Shuangtaizi (main branch of Liaohe River) estuarine wetland in July 2017 (Fig. 1). Samples were collected in the reed marshes of wetland (C1), the bare beach of intertidal zone (C2) and the subtidal flat (C3) at different elevations, and the water depth of C3 site was 0.5 m. C1 and C2 sediment cores were collected by hand-held non-destructive columnar sampler and then sectioned into subsamples of 5 cm thickness. C3 sediment core sample was collected by sediment borehole sampler and then sectioned into subsamples of 10 cm thickness. The length (Φ 5 cm) of sediment core for C1, C2 and C3 was 100 cm, 200 cm and 320 cm, respectively.

2.2. Analysis

The details of laboratory procedures for the treatment of the sediment cores and the subsequent analysis were described in a previous work (Liu et al., 2019). In short, the frozen-dried samples were used for the analyses of total organic carbon (TOC), total nitrogen (TN), stable isotopic composition of carbon (^{13}C) and nitrogen (^{15}N) utilizing Stable Isotope Ratio Mass Spectrometer (Model: Delta plus XP, Thermo Finnigan) connected with a Flash EA 1112 analyzer. The TOC, ^{13}C, and ^{15}N were measured after the removal of inorganic carbon through acid-treatment with HCl (1 M), while no acid treatment was performed on the TN samples. The isotope ratios were reported in δ (‰); ^{13}C and ^{15}N were relative to the V-PDB standard and atmospheric N, respectively. The precision of the analysis was better than 0.1 ‰ and 0.2 ‰ for carbon and nitrogen, respectively.

Samples for ^{210}Pb chronology need to be sealed at least 20 days before analysis. Excess ^{210}Pb ($^{210}Pb_{ex}$) analysis was measured by the high-purity germanium γ spectrometer (Model: GEM-MX7080P4). The counting time was 24 h. The counting efficiency was calibrated with a standard sample (CJ130621, the Chinese Academy of Metrology). Radionuclides and their peak energies used in measurement were ^{226}Ra (295.2, 351.9 and 609.3 keV), ^{210}Pb (46.5 keV), ^{137}Cs (661.66 keV). The $^{210}Pb_{ex}$ was calculated from the subtraction between ^{210}Pb and ^{226}Ra activities.

2.3. Chronology of sediment cores

The method for sediment chronology had been described in our previous work (Du et al., 2019, 2021), which we will briefly outline here as follows: the total activity of ^{210}Pb in sediment consists of two components: supported ($^{210}Pb_{sup}$) and excess ($^{210}Pb_{ex}$). $^{210}Pb_{sup}$ usually equilibrates with ^{226}Ra, while $^{210}Pb_{ex}$ is eventually buried in sediment (Alvarez-Iglesias et al., 2007). $^{210}Pb_{ex}$ constant rate of supply model (i.e. CRS, Appleby and Oldfield, 1978) is commonly used to date sediment from the last 100 years. Applying the CRS model, sediment age at a certain layer could be calculated (Sanchez-Cabeza and Ruiz-Fernández, 2012). The basis of ^{137}Cs chronology is that the first peak layer after ^{137}Cs's detection in the sediment core corresponds to the most intensive nuclear test year of 1963, and the last peak layer corresponds to the Chernobyl accident of 1986 (Madsen et al., 2005).

3. Results

3.1. Characteristics of TOC, TN, $\delta^{13}C$ and $\delta^{15}N$ in sediment cores

TOC showed significant differences at C1, C2 and C3 (Table 1). Its average value of 12.14 $mg \cdot g^{-1}$ at C1 was the highest, while C2 and C3 showed lower average values of 8.87 $mg \cdot g^{-1}$ and 11.02 $mg \cdot g^{-1}$ respectively. The variation range of TN content was small, its values were all <1.0 $mg \cdot g^{-1}$. Similarly, the TN average value ($mg \cdot g^{-1}$) showed the following order: C1 (0.69) > C2 (0.6) > C3 (0.4). The variation range of $\delta^{13}C$ (‰) was very small, the average values were relatively high at C2 (−24.20),low at C1 (−24.74)., and in the middle for C3(−24.51). Similar to that of $\delta^{13}C$, the $\delta^{15}N$ values were almost at the same level, and the average values were in this order: C2 (6.39) > C3 (6.24) > C1 (6.18).

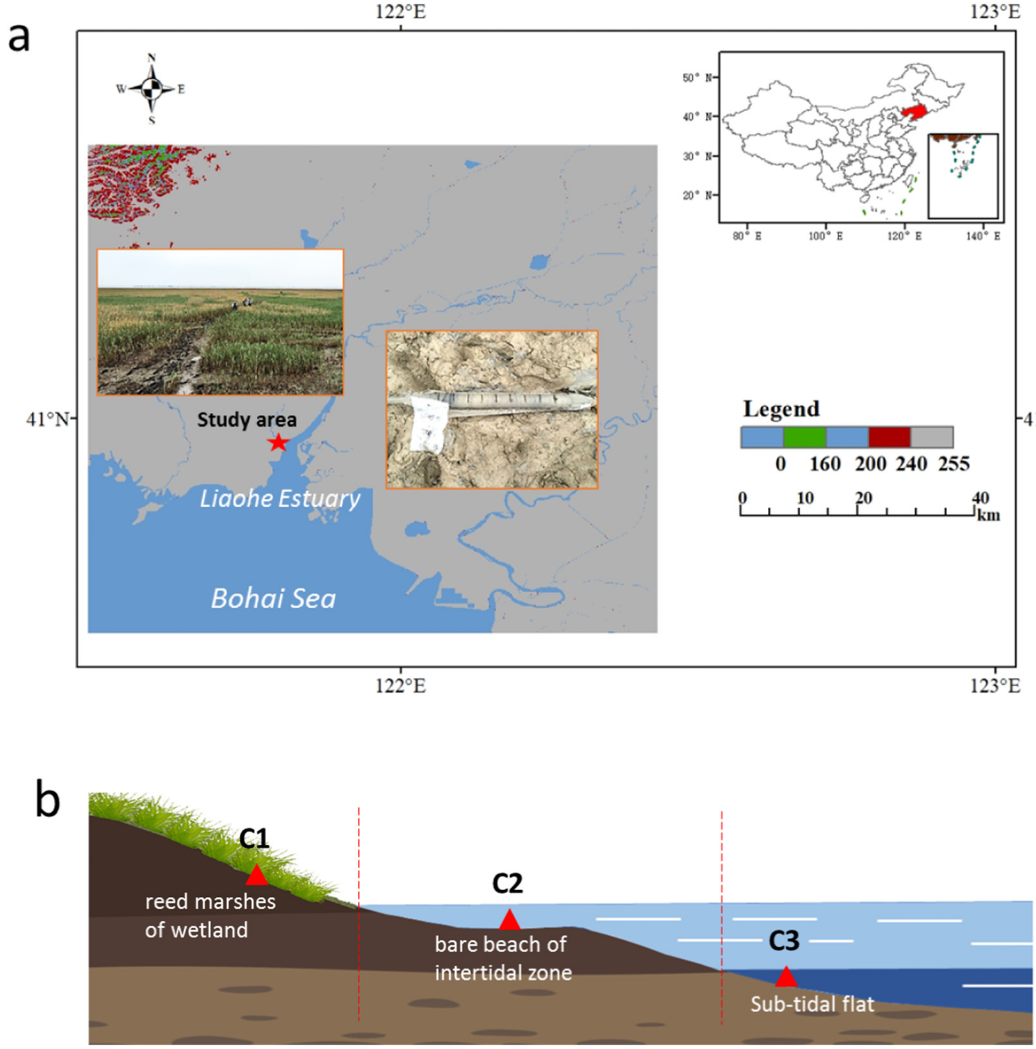

Fig. 1. Location of Liaohe estuarine wetland (a) and sampling stations (b).

The vertical distribution of TOC, TN, $\delta^{13}C$ and $\delta^{15}N$ with depths in sediments of C1, C2 and C3 are shown in Fig. 2. Similarly, results from all three cores are: both TOC and TN contents were high at the surface layer, and then fluctuated significantly in decreasing trend with the increasing depths. The $\delta^{13}C$ values of these three sediment cores were low at the surface, and then slightly increased with the increasing depths although it fluctuated in the deep layers. The variation of $\delta^{15}N$ value with depth was not obvious at C1, but those at C2 and C3 fluctuated significantly from the surface to the bottom.

3.2. Radionuclides in sediment cores and their chronologies

The vertical distribution of $^{210}Pb_{ex}$ with depths in sediments of C1, C2 and C3 are shown in Fig. 3a. As expected, $^{210}Pb_{ex}$ activity showed a significant overall decreasing trend with depth. Assuming that the input of $^{210}Pb_{ex}$ is constant, the ages of sediments at different layers were then calculated by the CRS model (Du et al., 2012; Baskaran, 2012; Sanchez-Cabeza and Ruiz-Fernández, 2012). The chronological ages of sediments were correspondingly plotted in Fig. 3. It showed that the sampling depths of 100 cm, 200 cm and 320 cm at reed marsh, bare beach and the subtidal flat at different elevations corresponded to the year of 1926, 1905 and 1864. The changes of sediment age with depth at various elevation zones were different, reflecting the changes in sedimentation rates.

The vertical distribution of ^{137}Cs with depth in sediments of C1, C2 and C3 are shown in Fig. 3b. ^{137}Cs activities showed fluctuation with depths, the first peak since its detection was very pronounced at C1, while they were less pronounced at C2 and C3 although still discernable. The first peaks of ^{137}Cs activities after detection in sediment from C1, C2 and C3 appeared at 75 cm, 160 cm and 210 cm layers, respectively, corresponding to the year 1963 (Walling and He, 1997; Madsen et al., 2005). When applying the $^{210}Pb_{ex}$ method, the same aforementioned layers corresponded to the

Table 1
Characteristics of *TOC, TN,* $\delta^{13}C$ and $\delta^{15}N$ from the sediment cores of Liaohe estuarine wetland.

Core ID	Sediment depth (cm)	Habitat	Data type	TOC ($mg·g^{-1}$)	TN ($mg·g^{-1}$)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)
C1	100	Reed marsh	Range	8.10–14.93	0.55–0.95	−25.07 ~ −24.49	5.98–6.55
			Average	12.14	0.69	−24.74	6.18
C2	200	Bare beach	Range	6.94–15.99	0.32–0.85	−24.72 ~ −23.59	5.56–6.99
			Average	11.02	0.60	−24.20	6.39
C3	320	Subtidal flat	Range	4.71–14.74	0.21–0.58	−25.21 ~ −23.91	5.59–6.96
			Average	8.87	0.40	−24.51	6.24

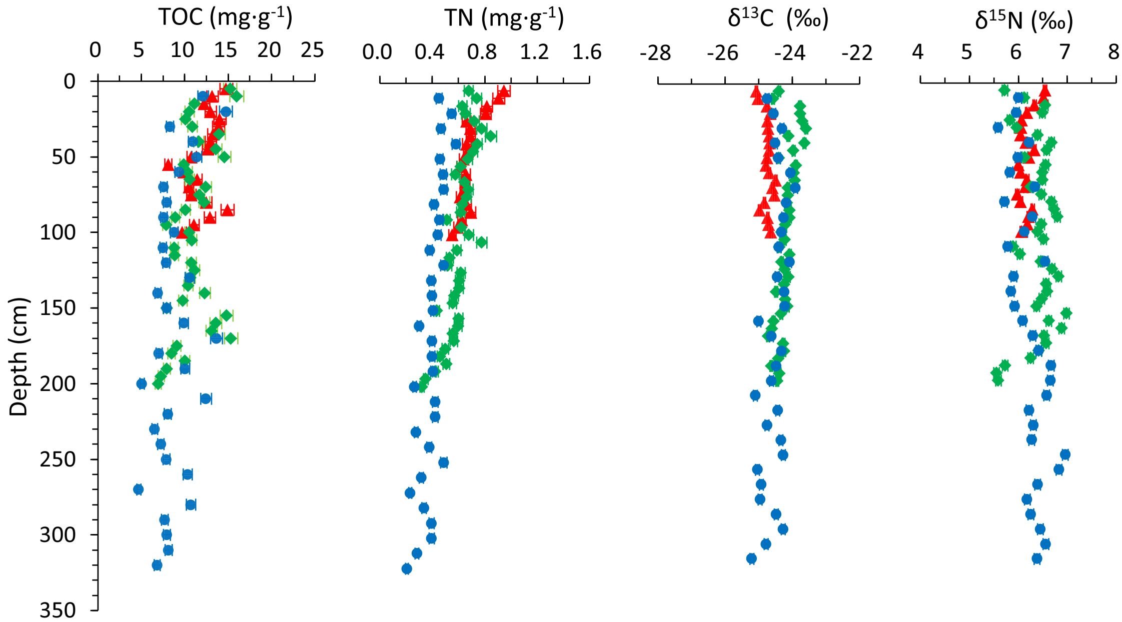

Fig. 2. Vertical distributions of TOC, TN, $\delta^{13}C$ and $\delta^{15}N$ in sediment cores at the reed marshes (C1, ▲), the bare beach of intertidal zone (C2, ◆) and the subtidal flat (C3, ●) of Liaohe estuarine wetland.

years 1968, 1963 and 1968 for C1, C2 and C3 respectively. The chronological age results from the two methods corroborated each other quite well (Fig. 3).

4. Discussion

4.1. Identification of sediment organic carbon sources in different elevation zones

Organic carbon in sediment cores is mainly influenced by particle organic matter (POM) inputs and outputs, as well as vertical accumulation and degradation (Pendleton et al., 2012). In present-day studies, C/N ratios and isotopic composition of $\delta^{13}C$ and $\delta^{15}N$ are the most common indicators to elucidate the source of sediment organic carbon (SOC), which have been widely applied in coastal area, including estuarine wetlands (Liu et al., 2006; Koziorowska et al., 2016; Sun et al., 2020; Guan, 2022). The variation of TOC/TN ratios and $\delta^{13}C$, $\delta^{15}N$ values over time in sediments at different elevation zones in Liaohe estuarine wetland were shown in Fig. 4. The ranges of TOC/TN ratio at C1, C2 and C3 were 13.12–21.44 (n = 20), 12.65–27.18 (n = 40) and 15.56–34.22 (n = 32), with an

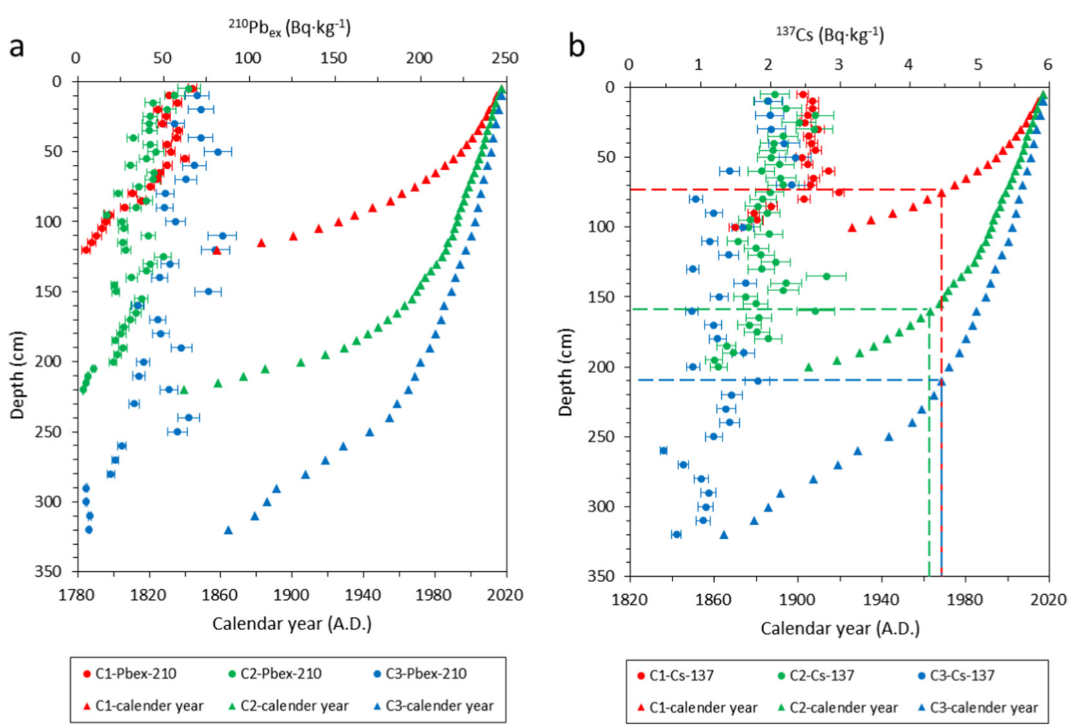

Fig. 3. (a) Vertical distributions of $^{210}Pb_{ex}$ and (b) ^{137}Cs in sediment cores, and their chronologies at the reed marshes (C1), the bare beach of intertidal zone(C2) and the subtidal flat (C3) of Liaohe estuarine wetland, the chronologies for $^{210}Pb_{ex}$ at C1 and C2 have been extended to below 100 cm and 200 cm respectively based on CRS model.

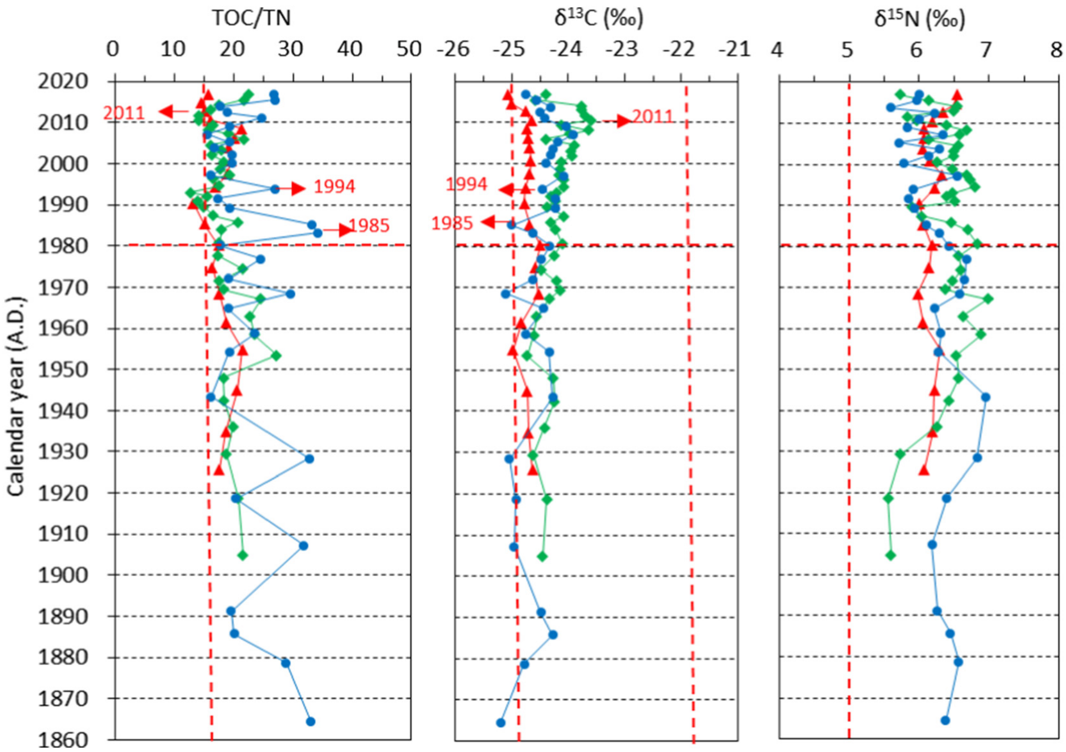

Fig. 4. Variation of TOC/TN and δ^{13}C over calendar year at the reed marshes (C1, ▲), the bare beach of intertidal zone (C2, ◆) and the subtidal flat (C3, ●) in Liaohe estuarine wetland.

average ratio of 17.68, 18.45 and 22.72, respectively. In general, the TOC/TN ratios from terrestrial sources and marine sources have much difference (Goñi et al., 2003; Sun et al., 2020). The terrestrial POM usually has a wide range of TOC/TN ratios >15, but those TOC/TN ratios of marine-derived POM are less variable, ranging from 4 to 9 (Meyers, 1994; Hedges and Oades, 1997; Hu et al., 2006). Moreover, typical isotopic compositions of δ^{13}C for terrestrial POM are from −33 ‰ to −25 ‰ (Hedges and Oades, 1997; Barth et al., 1998) and from −22 ‰ to −18 ‰ (Wada et al., 1987; Vizzini et al., 2005) for marine-derived POM. The ranges of δ^{13}C values at C1, C2 and C3 were − 25.07 ~ −24.49 ‰, −24.72 ~ −23.59 ‰ and − 25.21 ~ −23.91 ‰, respectively. Therefore, the ratios of TOC/TN (12.65–34.22) and isotopic compositions of δ^{13}C (−25.21 ~ −23.59 ‰) in our work are between the ranges for terrestrial and marine end-members, indicating clearly that the SOC at reed marshes, bare beach of intertidal zone and subtidal flat were almost consistent, from a mixture of terrestrial and marine sources. Similarly, the ranges of δ^{15}N values at C1, C2 and C3 were between 5.56 and6.99 ‰, indicating their marine-derived sources (5 ‰ to 12 ‰) (Thornton and McManus, 1994; Lamb et al., 2006), since δ^{15}N from terrestrial POM sources have ranges of 0 ‰ to 4 ‰. Actually, the determined δ^{15}N in sediment included not only POM (DON), but also DIN. Meanwhile, the isotope fractionation in the sediment has a significant impact on the organic nitrogen composition. Therefore, in terms of indicating SOC sources, the δ^{13}C value is more reliable than the δ^{15}N value (Raymond and Bauer, 2001; Sun et al., 2020).

The identification of SOC source using TOC/TN ratio and δ^{13}C value agreed with each other in the study area of Liaohe estuarine wetland. From the variation of TOC/TN ratio and δ^{13}C value with depths from the three cores over time, the change trend of these two values basically showed a mirror-image relationship, especially the maximum and minimum layers with strong influence of terrestrial or marine input at certain years (1985,1994 and 2011), the corresponding relationship is distinct. Furthermore, the correlation between the TOC/TN ratio and the δ^{13}C value was analyzed and shown in Fig. 5, which was consistent with previous results in coastal areas of China (Wang et al., 2015a, 2015b; Liu et al., 2015; Dun et al., 2019; Zhang and Wang, 2019; Sun et al., 2020). As the TOC/TN ratio increases, the δ^{13}C value showed a decreasing trend (Zhang and Wang, 2019). It showed the same pattern at C2 and C3, but the correlation between the TOC/TN ratio and the δ^{13}C value is not obvious at C1, this may be related to the influence from local land-based vegetation of reed (Volvoikar et al., 2014; Krishna et al., 2013). The Fig. 5 also shows the eigenvalue ranges of both terrestrial and marine inputs, the data of our study partially fall within the range of the terrestrial source input, a small part of data of coastal seas (Bohai Sea and Sanggou Bay) from previous studies fall within the range of the marine source input (Gao et al., 2016; Xia et al., 2014), and most of the data from estuary, salt marsh and our study fall within the range of both end-member values, indicating that terrestrial-sourced and marine-sourced inputs were mixed at these areas, attributed to the strong land-sea interactions in the estuarine region (Krishna et al., 2013).

4.2. Quantifying sediment organic carbon from different sources

Based on the TOC/TN ratio and δ^{13}C value, the sources of SOC in the study area showed the characteristics of a mixture of both terrestrial and marine inputs, with the terrestrial input being more significant (Pradhan et al., 2014). In order to further distinguish the relative contribution between terrestrial and marine inputs to SOC, and to identify the proportion of organic carbon from different sources, the δ^{13}C value terrestrial ($\delta^{13}C_t$) –marine ($\delta^{13}C_m$) two-end member model was used, the contribution proportion of terrestrial and marine organic carbon inputs can be calculated as shown in Eq. (1) (Zhang et al., 2007; Cai et al., 2014).

$$\delta^{13}C_s = \delta^{13}C_t \times f_t + \delta^{13}C_m \times f_m \quad (1)$$

$$f_t + f_m = 1 \quad (2)$$

where $\delta^{13}C_s$ is the stable carbon isotope value in the sediment (‰), f_t and f_m are the percentages of the terrestrial and marine sources in SOC (%), respectively. Generally, the end-member values of organic carbon are different, which need to be determined according to the local input sources (Gao

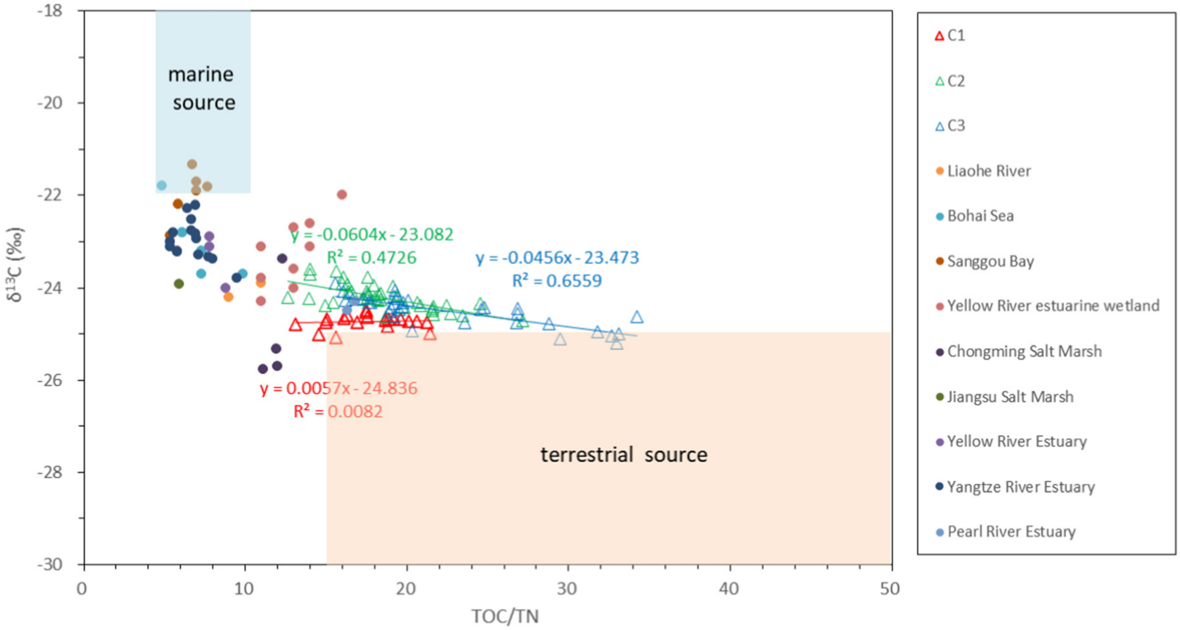

Fig. 5. Plots of the TOC/TN ratio vs the δ^{13}C value in the sediment of Liaohe estuarine wetland and other typical zones: Wang et al. (2015a, 2015b) (Liaohe); Gao et al. (2016) (Bohai); Xia et al. (2014) (Sanggou Bay); Zhang and Wang (2019) (Yellow River Estuarine Wetland); Dun et al. (2019) (Chongming Salt Marsh); Wang et al. (2013) (Jiangsu Salt Marsh); Liu et al. (2015) (Yellow River Estuary); Wang et al. (2015a, 2015b), Sun et al. (2020) (Yangtze River Estuary); Zhang et al. (2013) (Pearl River Estuary).

et al., 2016). In this study, $\delta^{13}C$ values with −26 ‰ and − 20 ‰ were selected as the end-members for terrestrial and marine sources, respectively (Cai et al., 2014). The result in Fig. 6 showed that the contribution of terrestrial organic carbon input was significantly higher than that of marine-sourced input. The f_t values at bare beach of intertidal zone and subtidal flat were at the same level (> 60 %), but they were lower than that of the reed marshes (> 70 %), indicating that the accumulation of terrestrial source SOC at the estuarine zone can be facilitated by vegetation (Zhang and Wang, 2019). Meanwhile, the f_t values in sediment cores of three zones fluctuated over time (Fig. 6), especially since the 1980s, in which the f_t values tended to decrease first and then increase again. It was reported that the soil from different areas is the main source of SOC in estuary because the change in land-use (i.e., reclamation and development of aquaculture/agriculture) can increase the export of soil-derived organic carbon stored in estuary sediments (Bianchi, 2011; Raymond and Bauer, 2001; Tao et al., 2015). With the rapid growth and spread of human population, human activities have introduced new types of disturbances into natural estuarine wetland, which can greatly influence the input of SOC (Bianchi and Allison, 2009), therefore the significant changes since the 1980s are more likely attributed to anthropogenic disturbances around Liaohe estuarine wetland. At the same time, the degree of disturbances in estuarine wetland is also affected by the elevation.

4.3. Evolution of sedimentation rate and its influencing factors

On the basis of the sediment dating by $^{210}Pb_{ex}$ method (Baskaran, 2012; Sanchez-Cabeza and Ruiz-Fernández, 2012), combined with sediment bulk density, the sedimentation rate (mass accumulation rate: MAR) in different

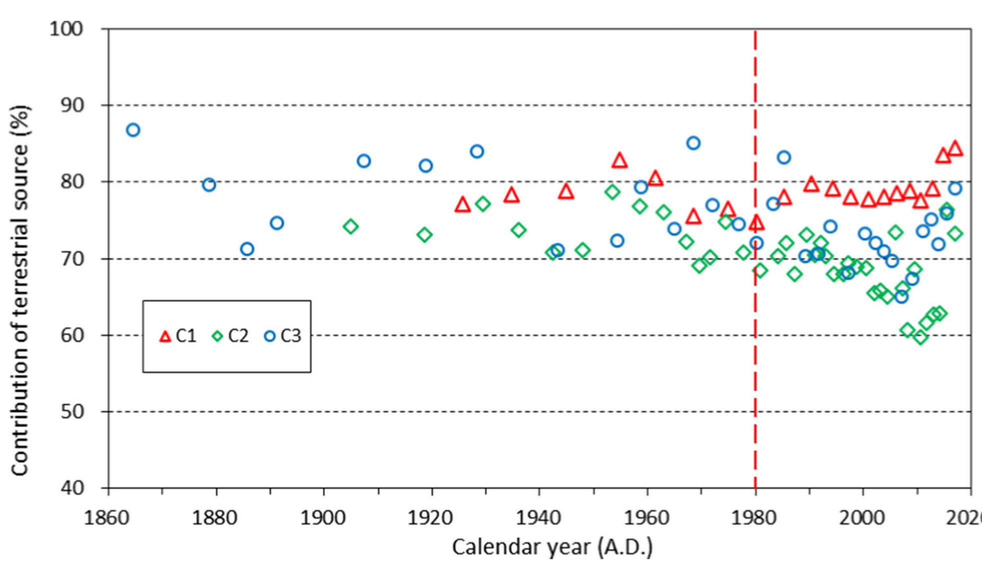

Fig. 6. Variation in terrestrial source contribution of SOC from the reed marshes of wetland(C1), the bare beach of intertidal zone(C2) and the subtidal flat (C3) of Liaohe estuarine wetland.

periods was calculated as shown in Fig. 7a. In the Liaohe estuarine wetland, MAR fluctuated over time at different elevation zones, and the overall trend was increasing. Since the 1980s, MAR fluctuation was more intense and the magnitude of the increase even more significant, suggesting human-induced impacts due to increased terrestrial material loading (Cuellar-Martinez et al., 2020; Dahl et al., 2022). Comparing the different elevations, MAR values and change characteristics at the bare beach and the subtidal flat were similar, and both of them were significantly greater than that of the reed marsh.

The variations of sediment accretion in estuarine wetland are controlled by the riverine discharge into the sea (Lagomasino et al., 2013), and additionally affected by the change of land use pattern caused by human activities (Yang et al., 2004; Wang et al., 2020). It was reported that the runoff at Liujianfang Station upstream of the Shuangtaizi estuary on the main stream of Liaohe River fluctuated significantly over time, especially since the 1980s (According to the report of the series "Statistic Bulletin on China River Sediment" published by the Ministry of Water Resources, P.R. China). The variation of annual sediment flux with runoff during 1987–2017 were plotted in Fig. 7b, the flux was quite low at the year of 2000 which is the turning point. During the period of 1987 to 2000, the annual average sediment flux was 624 × 10^4 t, with the peak values appearing around the year of 1993 and 1997. During the period of 2000 to 2017, the annual average sediment flux is 147 × 10^4 t, with the maximum value appearing around the year of 2010. After the year of 2000, the annual average sediment flux decreased due to human activities such as water consumption and the construction of hundreds of reservoirs (Zhang et al., 2014; Zhao et al., 2016).

As expected, riverine suspended sediment into the estuarine wetland can strongly affect the sedimentary environment of coastal zone, especially the sedimentation rate (Falcini et al., 2012; Li et al., 2017). Similar with those for riverine sediment discharge, the MAR of the Liaohe estuarine wetland fluctuated over time since the 1980s (Fig. 7b). The minimum value of MAR occurred around the year of 2000, and the maximum values around the year of 1993, 1997 and 2010. The variation in MAR is corresponding well to the changes in sediment flux of Liaohe River (Fig. 7b), reflecting that the riverine input was the dominant factor for sedimentation rate of Liaohe estuarine wetland (Ye et al., 2015). However, while riverine sediment flux has shown an overall decreasing trend since the 1980s, the MAR showed an overall increasing trend, suggesting the impact of land use change due to human activities in the local area of Liaohe estuary (Zhu et al., 2010). Land reclamation is one of the most prevalent causes of estuarine wetland decline (Ewers Lewis et al., 2019), estimates of coastal habitat loss due to reclamation reaches ~755,000 ha in China between 1985 and 2010 (Tian et al., 2016), and > 80 % of coastal wetlands developed in China (Wu et al., 2018). During the last few decades, Liaohe estuarine wetland has experienced rapid development (Zhao et al., 2016), the

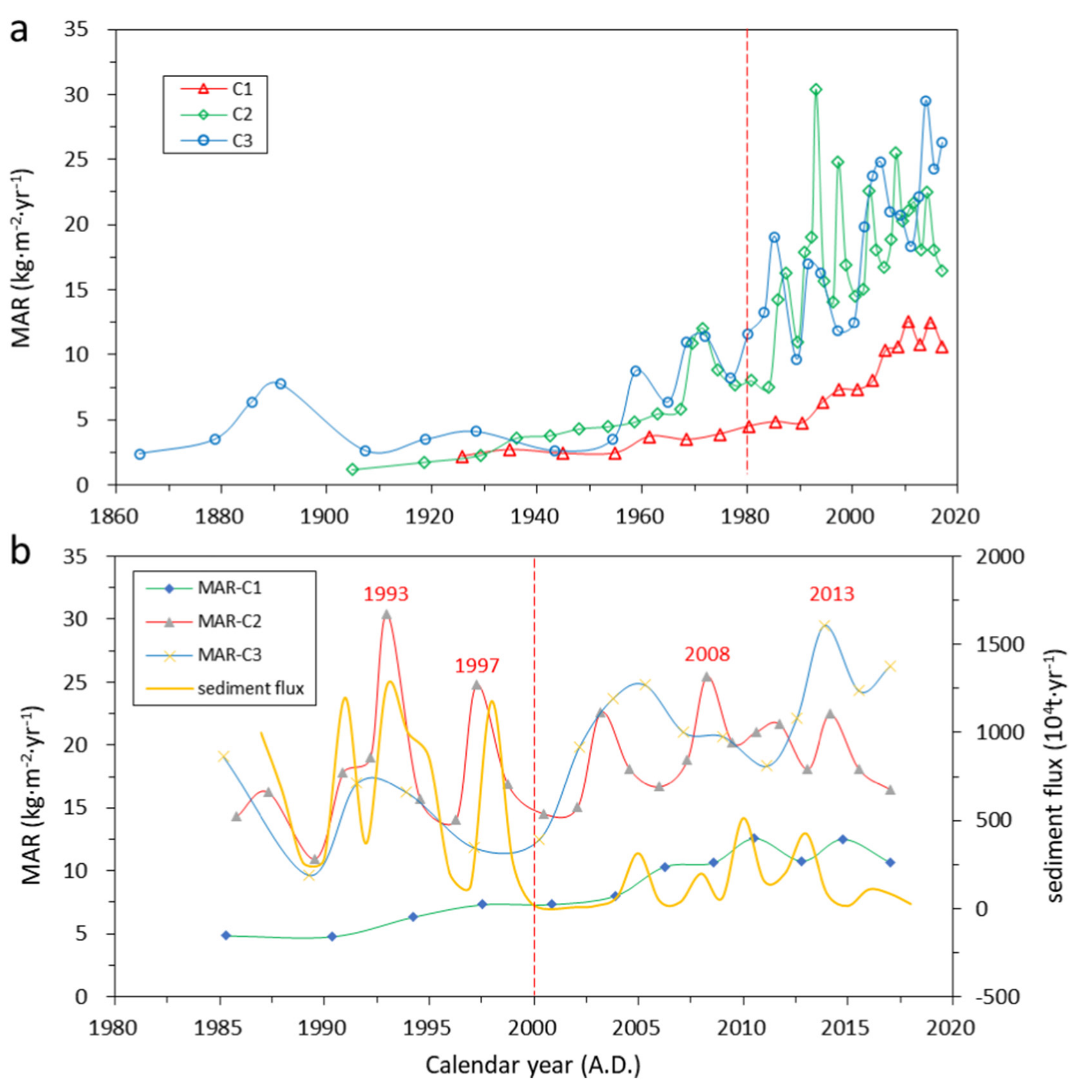

Fig. 7. (a) Temporal variation of MAR at different elevation zones in Liaohe estuarine wetland. (b) Temporal variation of sediment flux ("Statistic Bulletin on China River Sediment" published by the Ministry of Water Resources, P.R. China) in Liaohe River (Liujianfang Station) VS. MAR at different elevation zones in Liaohe estuarine wetland since the 1980s.

construction of Yuanyang ditch Wharf and Cross-sea Bridge occurred around the year of 2008 and 2013 corresponding to the peak values of sedimentation rate (especially at the bare beach and the subtidal flat) (Lu et al., 2018). Estuarine wetland is particularly vulnerable to human disturbance in that they have been targeted for settlement, livestock grazing, and fishing (Gedan et al., 2009). Different elevation zones have different sediment dynamics because of tidal and vegetation influence, in the form of death and oxidation of root materials, which add structural complexity and strength to the sediment matrix, can lead to collapse and compaction of peaty sediments (Delaune et al., 1994; Ewers Lewis et al., 2019). Therefore, the response of sedimentation rate at different elevation zones to human activities are different, where the reed marsh is relatively stable compared to the bare beach and subtidal flat.

4.4. Trends of organic carbon sequestration and its driver for change

High sedimentation rate, low organic matter degradation rate, and organic carbon from vegetation in deep burial would promote the sequestration of SOC in estuarine wetland, enhancing its carbon sink function and sink effectiveness (Chmura et al., 2003; Duarte et al., 2013). Based on MAR and SOC, the sequestration flux of SOC (SF-SOC) over time in the Liaohe estuarine wetland were calculated and results were shown in Fig. 8. It was noted that the SF-SOC in this study lied in the range of 20–500 $g \cdot m^{-2} \cdot yr^{-1}$ for coastal wetland (Reddy and Delaune, 2008; Ye et al., 2015). Similar to that of MAR, the SF-SOC fluctuated over time at different elevation zones although it showed an overall increasing trend, especially since the 1980s. This phenomenon was consistent with the conclusion that under the most common time horizon (100 year) used for carbon reporting under the United Nations Framework Convention on Climate Change (UNFCCC), SOC sequestration for decades or longer becomes increasingly relevant (Marland et al., 2001; Levasseur et al., 2012). Meanwhile, SF-SOC was significantly higher at the bare beach and the subtidal flat than that of the reed marsh. There were a few peak values ($g \cdot m^{-2} \cdot yr^{-1}$) at the bare beach and the subtidal flat with the highest SF-SOC of 304.19 (C2) and 358.41 (C3) occurring around the year of 1997 and 2015. Meanwhile, the SF-SOC contributed by terrestrial sources was also drawn in Fig. 8, it showed that the terrestrial sequestration flux of SOC is basically consistent with the characteristics of total SOC sequestration flux over time at different elevation zones. It confirmed the dominant role of terrestrial-sourced influence on organic carbon input and sequestration in estuarine wetland (Tao et al., 2015).

In order to further illustrate the main influencing factor to organic carbon sequestration at different elevation zones in Liaohe estuarine wetland, the linear correlation between SF-SOC and MAR, SF-SOC and TOC were plotted in Fig. 9. It was noted that SF-SOC and MAR showed a significant positive linear correlation, indicating the major control from sedimentation rate on the organic carbon sequestration in estuarine wetland (Chmura et al., 2003; Duarte et al., 2013). There was also an obvious positive linear relationship between TOC and SF-SOC, but the correlation coefficient was relatively low compared to that of MAR. It means that organic carbon sequestration in estuarine wetland is controlled by both sedimentation and organic carbon input, while sedimentation being the more significant control factor (Cuellar-Martinez et al., 2020). At the same time, the degree of influence from sedimentation rate and organic carbon content showed variations at different elevation zones, where their influence on the reed marsh was more significant. Carbon fixation in reed leaves generally exceeds their

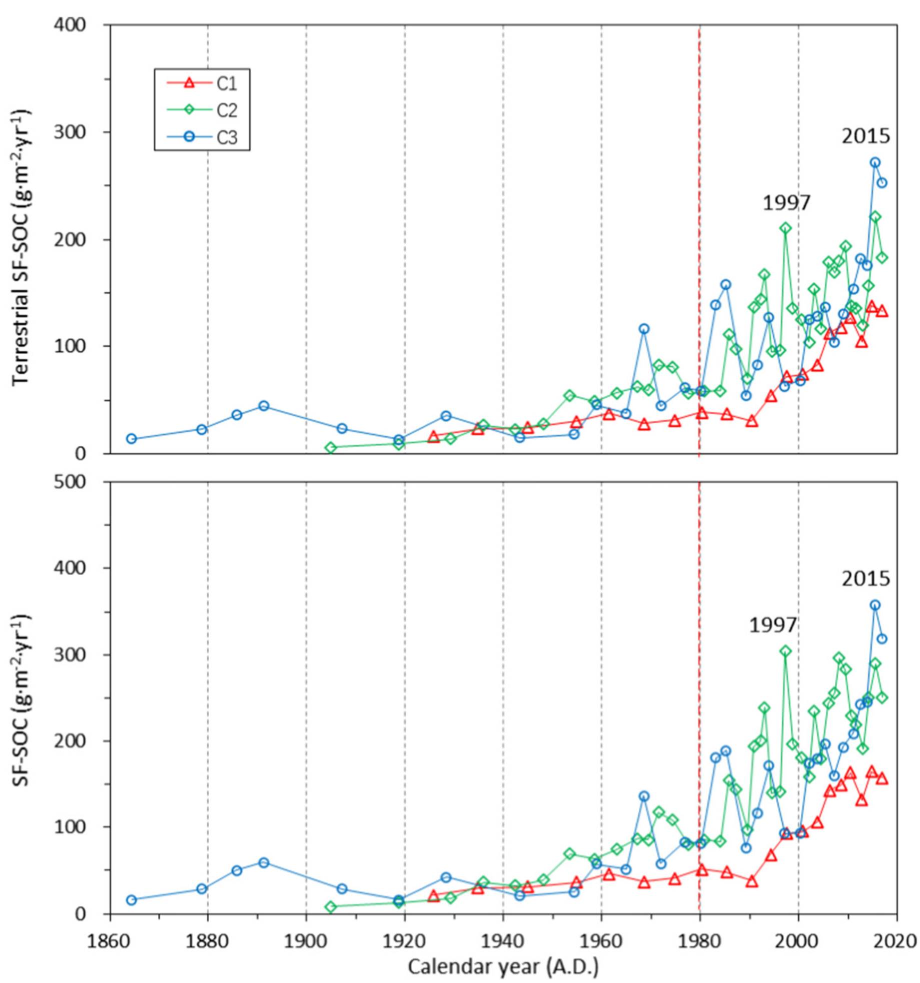

Fig. 8. Temporal variation of SF-SOC and its terrestrial proportion at different elevation zones in Liaohe estuarine wetland.

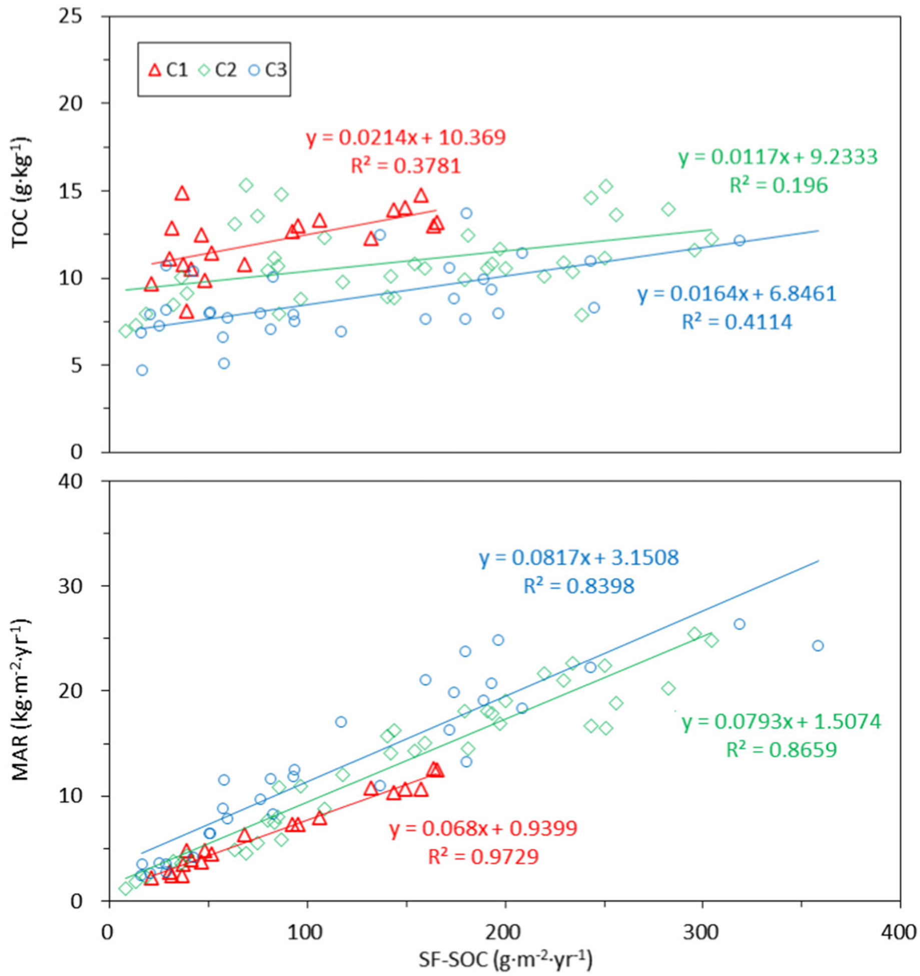

Fig. 9. Correlation between SF-SOC and MAR/TOC at different elevation zones in Liaohe estuarine wetland.

metabolic needs (Duarte et al., 2010), thus a large proportion of excess organic carbon may be transported to the roots, where it is eventually buried into the sediment (López-Mendoza et al., 2020). A large number of biotic (e.g., reed roots) and abiotic (e.g., sediment structure and water column) factors can potentially influence the degree of SOC sequestration and preservation, that can produce spatial variability in estuarine wetland at local and regional levels (Serrano et al., 2016; López-Mendoza et al., 2020).

5. Conclusions

TOC in shallow layer of reed marsh was higher than those of bare beach and subtidal flat in Liaohe estuarine wetland, the levels of TOC were equalized in deep layers. Based on the temporal and spatial variations of TOC/TN and $\delta^{13}C$, the source of organic carbon at different elevation zones of Liaohe estuarine wetland showed a mixture of both terrestrial and marine sources, with the terrestrial input being the dominant one. Since the 1980s, the variation of terrestrial contribution of SOC became more pronounced.

Based on $^{210}Pb_{ex}$ chronology method, the MAR at different elevation zones in Liaohe estuarine wetland showed a tendency of increase over the past century, and evolved more drastically since the 1980s; MAR was relatively stable in the reed marsh, compared to the bare beach and subtidal flat. The sedimentation rate of the estuarine wetland was linked to the changes of sediment flux from riverine discharge and the land use in the catchment area of estuary attributed to human activities in recent decades.

SF-SOC showed an overall increasing trend, especially since the 1980s, and it was found to be higher at the bare beach and subtidal flat than that of reed marsh. The linkage between SF-SOC and MAR was found to be stronger than that of TOC content. Compared with the organic carbon input, the effect from high sedimentation rate on organic carbon sequestration was more pronounced at estuarine wetland.

CRediT authorship contribution statement

Jinzhou Du developed the theory and performed the computations.
Jinqiu Du and Fenfen Zhang verified the analytical methods.
Jinqiu Du carried out the experiment, wrote the manuscript with support from Jinzhou Du, Fenfen Zhang and Xu Ren.
All authors provided critical feedback and helped shape the research, analysis and manuscript.

Data availability

Data will be made available on request.

Declaration of competing interest

None of the authors of this paper has a financial or personal relationship with other people or organizations that could inappropriately influence of bias the content of the paper.

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