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Distribution of black carbon in sediments from mangrove wetlands in China

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Black carbon (BC) is a carbonaceous residue generated by the incomplete combustion of fossil fuels and biomass. It forms an important component of the mangrove carbon pool. Although coastal and marine sediments have long been recognized as important sinks for BC, there are few reports on its distribution in mangrove sediments. This study investigated BC distribution and the associated environmental implications in sediments of various size fractions. Mangrove sediments were collected from 44 sites in the Guangdong, Guangxi, Hainan, and Fujian Provinces of China. Sediments from the Hainan Province were found to have much higher organic carbon (OC) and BC content than those from other provinces. Sediment OC and the BC content showed a significant positive relationship. Sediments from the Guangdong, Guangxi, and Hainan provinces showed significant BC accumulation with an enrichment factor $>$ 1, especially in the size fractions 0.053–0.25 mm and 0.25–2 mm. Sediments from different sampling sites did not show significantly different BC distribution between the various size fractions. A higher mass loading of BC was observed among fine particles in samples from the Fujian Province, and among the larger particles in samples from the Guangdong, Guangxi, and Hainan provinces. Among all the samples, the BC/OC ratio ranged from 0.21 to 0.29, indicating that BC originates from a combination of biomass and mineral combustion. The average $\delta^{13}C$ values in the sediments varied between the sampling plots. The highest $\delta^{13}C$ values were recorded in the Fujian province, which indicates the proportion of BC from biomass combustion in mangroves in Fujian Province is higher than that in other Provinces.

KEYWORDS

mangroves, sediments, black carbon, distribution, organic carbon

Introduction

Mangrove wetlands are distributed in the tropical and subtropical intertidal coastal zones. They have unique morphological and structural characteristics, physiological and ecological processes, and strong adaptability to intertidal tidal flat environments, and accounting for 30–40% of tropical forest wetlands worldwide (Perez et al., 2018; Hu et al., 2019). Mangroves have many functions, including attenuating the effects of wind and waves, embankment protection, silt promotion, and water purification. They also serve as important habitats for many sea birds, fish, shrimp, and shellfish (Bouillon et al., 2008; Duarte et al., 2013; Taillardat et al., 2018).

Mangroves, saltmarshes, and seagrasses are recognized as blue carbon ecosystems for their disproportionately large organic carbon (OC) storage. When they are mature, vegetal biomass is deposited and conserved within sediments over millennia (Perez et al., 2018; Taillardat et al., 2018). Mangroves have among the highest carbon sequestration capacity of any ecosystem worldwide. The ecosystem is in a long-term flooded state, with a slow rate of OC decomposition and a long carbon cycle, making it one of the most important coastal zone carbon sinks (Alongi, 2014; Sanders et al., 2014). Mangroves play a vital role in terrestrial and oceanic carbon cycling (Alongi, 2012; Donato et al., 2012; Liu et al., 2014). Although they make up 0.7% of global coastal zones, they contribute 10% of the total net primary production and 25% of the carbon burial in the coastal zone worldwide (Kathiresan and Bingham, 2001; Kathiresan et al., 2013). As a coastal "blue carbon" wetland, mangroves play a vital role in mitigating global climate change and reducing greenhouse gas emissions.

In addition to organic carbon, the black carbon (BC) in the mangrove carbon pool has attracted considerable attention. BC is a carbonaceous residue generated from the incomplete combustion of fossil fuels and biomass (Kuhlbusch, 1998; Masiello and Druffel, 1998; Wang et al., 2022). It predominantly comprises highly polymerized polycyclic aromatic hydrocarbons that are extremely difficult to degrade at ambient temperatures. Owing to this unique property, BC has a considerable influence on the carbon budget and the refractory carbon pool worldwide. It is considered to be an inert carbon pool and an important component of the global carbon sink (Guo et al., 2018). BC is mainly produced on land through natural and anthropogenic activities and is transported to the sea in the form of aerosols and/or through soil erosion and river discharge. It may also accumulate in coastal and marine sediments (Luz et al., 2010). Above formation and storage mechanisms may account for a substantial proportion of the total marine sedimentary organic carbon (Zhang et al., 2022). Mangrove is one of the most carbon-rich coastal wetlands that sequester and store carbon to help mitigate global climate change. BC is very likely to constitute an important component of coastal carbon pool owing to its allochthonous and refractory, and it might have a significant implication on the carbon sequestration in mangrove sediments and then affect the distribution of mangrove carbon stocks and the global blue carbon budget (Perez et al., 2018; Taillardat et al., 2018; Zhang et al., 2022). Therefore, it is necessary to clarify the content and characteristics of BC in mangrove forests. This will also important for us to assess and predict accurately the role of mangroves in mitigating climate change (Chew and Gallagher, 2018). However, relatively few studies have been undertaken on BC in mangrove sediments. Therefore, it is necessary to explore the content and distribution of BC in mangrove sediment. The specific aims of this study were to: (1) determine the BC content of sediments from mangrove wetlands in China, (2) determine the distribution characteristics of BC in sediments of different particle sizes, and (3) identify BC sources in sediments and the associated environmental implications.

Materials and methods

Sediment sampling

Surface sediments were collected in 2018 and 2019 from mangrove wetlands in Shenzhen, Guangzhou, and Zhuhai (Guangdong Province, $n = 13$); Fangchenggang, Shankou, Qinzhou, and Beicanghe (Guangxi Province, $n = 16$); Danchang and Huachang (Hainan Province, $n = 5$); and Quanzhou, Fuding, and Longhai (Fujian Province, $n = 10$) (**Figure 1**), In these wetland areas, *Sonneratia apetala*, *Aegiceras corniculatum*, *Rhizophora stylosa*, and *Kandelia candel* are the dominant mangrove species, respectively. Sediment samples (0–5 cm depth) were collected with a precleaned stainless steel scoop, transported to the laboratory, air-dried, and passed through a 2 mm sieve within 1 week. Roots, rocks, and visible residues were manually removed. If we need to accurately calculate the carbon pool content of sediments in mangrove, the sampling depth should be at least 50 cm, or even 100 cm. However, there is no common protocol for the sampling depth to study the distribution characteristics of BC (Eid and Shaltout, 2016; Chew and Gallagher, 2018; Guo et al., 2018). Our study mainly focuses on the distribution characteristics of BC in mangrove surface sediments in China, therefore, the sampling depth of sediments are at depth of 0–5 cm.

Laboratory analysis

The OC content of the sediments was measured using the H_2SO_4-$K_2Cr_2O_7$ oxidation method (Yeomans and Bremner, 1988). A total of 0.5 g sediment that had been air dried through a 0.149 mm sieve was acidified using a dichromate (H_2SO_4-$K_2Cr_2O_7$) solution for 30 min in a Pyrex digestion tube placed within a 40-tube block digester that had been preheated to

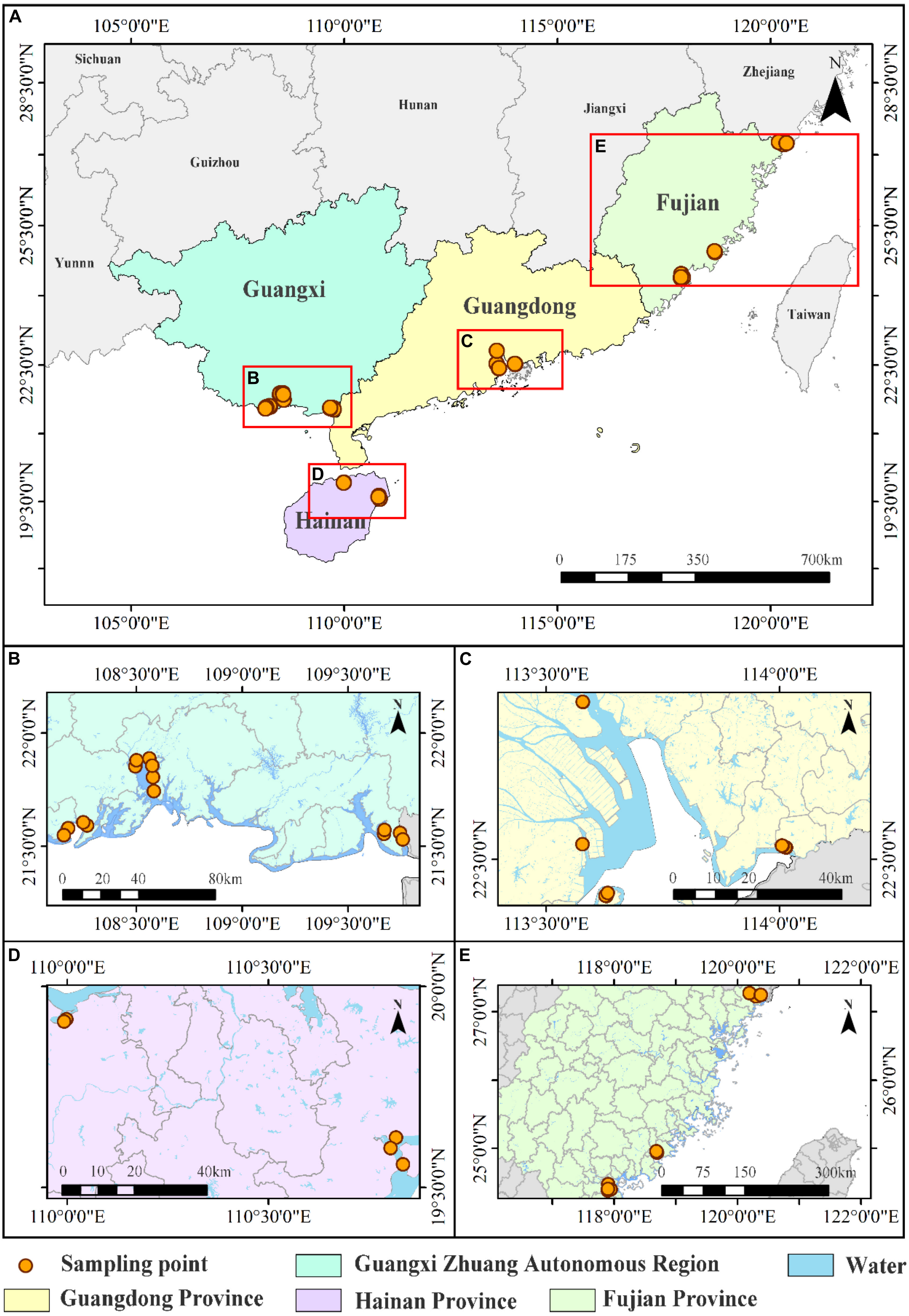

FIGURE 1
Map of mangrove sampling locations in China used for the study. **(A)** All sampling locations, **(B)** sampling locations in Guangxi Province, **(C)** sampling locations in Guangdong Province, **(D)** sampling locations in Hainan Province, and **(E)** sampling locations in Fujian Province.

170°C. The unreacted dichromate was then estimated using titration of the cooled digest with an acidified solution of ferrous ammonium sulfate using N-phenylanthranilic acid as an indicator. The BC content of the sediments was determined using the wet chemical oxidation method proposed by Lim and Cachier (1996). Briefly, 3 g of air-dried sediment (through a 0.149 mm sieve) was added to a centrifugal tube and then treated with 15 mL of 3 mol L^{-1} HCl for 24 h to remove the inorganic carbon. After centrifugation, the supernatant was decanted and 15 mL of 10 mol L^{-1} HCl was added for 24 h to remove the CaF_2. The residue was then oxidized with 15 mL of an acid potassium dichromate solution (0.1 mol L^{-1} $K_2Cr_2O_7$ and 2 mol L^{-1} H_2SO_4, 1:1, v:v) at 55 ± 1°C for 60 h. During the reaction, $K_2Cr_2O_7$ was added until the color changed to maintain the oxidant in excess, and deionized water was added several times to maintain the solution volume. The carbon residue obtained after this treatment was regarded as BC. The BC content and $\delta^{13}C$ values of the residues were determined using an elemental analyzer (FLASH EA-DELTA V; Thermo Fisher Scientific, Waltham, MA, United States).

The sediments were fractionated using modified wet sieving (Smith et al., 2014; Márquez et al., 2019); 100 g of sediment that had been air dried through a 2 mm sieve was placed in a 0.25 mm sieve and then immersed in deionized water for 5 min. It was then agitated vertically at 3 cm in amplitude 50 times within 2 min. The sediments in the 0.25 mm sieve were transferred into aluminum cans and then oven-dried at 60°C to constant weight. Using the same procedure, the remainder of the sediment and the water mixture was passed through a 0.053 mm sieve to remove aggregates of 0.053–0.25 mm. The remaining sediment and water mixture that had previously been passed through the 0.053 mm sieve was then centrifuged and dried to obtain aggregates <0.053 mm. The dried aggregates with differing particle size fractions were used to measure the BC content using this method.

Calculations and statistical analysis

The ratio of BC to OC reflects the sources of BC, calculated as:

$$BC/OC = \text{BC content in sediments}/\text{OC content in sediments}.$$

The enrichment factor (EF) reflects the addition of BC to the sediments as derived from anthropogenic activity and is calculated as:

$$EF = C_{sample}/C_{ref}$$

where C_{sample} and C_{ref} are the BC content in the sediment sample and the background value for BC, respectively. In the present study, the average BC concentration for undisturbed samples from the bottom of the soil profiles was ascertained, to represent the background value for soil BC, which was calculated as 0.47 g kg^{-1}.

The distribution factor (DF), defined as the ratio of BC concentration between the individual size fraction and bulk sediment sample, is used to estimate the size fraction with which the BC is preferentially enriched, which is calculated as follows (Zong et al., 2016):

$$DF = C_{fraction}/C_{bulk}$$

where $C_{fraction}$ and C_{bulk} represent the BC concentration in a given size fraction and the bulk sediment samples, respectively. A DF value of > 1 indicates that BC has preferentially accumulated in this particular sediment size fraction.

The mass loading of BC is an important index for assessing BC accumulation in grain-size fractions. The mass loading for the grain size fraction ($GSF_{loading}$) was calculated by combining the BC concentration and the mass percentage in a given size fraction, as follows:

$$GSF_{loading} = \frac{CM_i \times GS_i}{\sum_{i=1}^{3} (CM_i \times GS_i) \times 100}$$

where CM_i and GS_i are the BC concentration in the individual size fraction (i) and the mass percentage of the individual fraction, respectively. The summation of $GSF_{loading}$ values for the size fractions of an individual sample equaled 100%.

The statistical analyses were performed using SPSS Statistics v.21 (IBM, Armonk, NY, United States). One-way ANOVA was used to explore the differences of BC among different sampling sites. Tukey's multiple-range test was performed to determine whether significant differences occurred among different sampling sites based on the means at a significance level of 0.05. The figures were plotted using OriginPro 2016 (OriginLab Corporation, Northampton, MA, United States).

Results

Contents and relationship of organic carbon and black carbon in mangrove sediments

The OC content for different mangrove sediments ranged from 1.32 to 5.31 g kg^{-1}. The amount of OC in the sediments varied according to the sampling location. Sediments from the Hainan Province contained considerably more OC than those from the other provinces ($F = 3.214$, $P = 0.033$, **Figure 2A**). The BC content for sediments in our study area ranged from 0.30 to 1.83 g kg^{-1}, and varied according to the sampling province. Sediments from the Hainan Province had substantially higher BC than those from the other provinces ($F = 3.239$, $P = 0.033$,

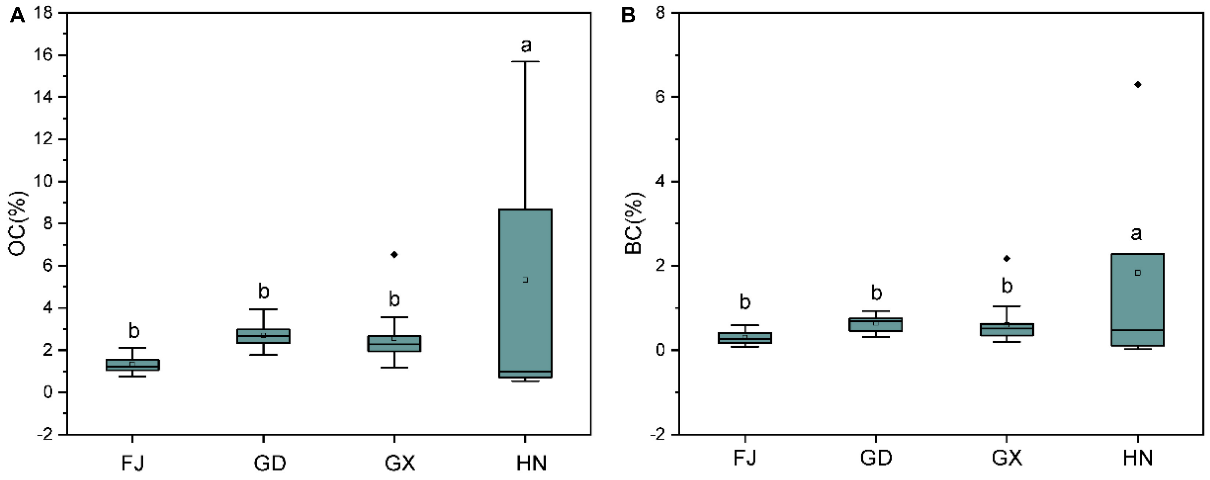

FIGURE 2
Boxplots showing the organic carbon (OC; **A**) and black carbon (BC; **B**) content of sediments from different mangrove wetlands in China. FJ, Fujian; GD, Guangdong; GX, Guangxi; HN, Hainan. Different lowercase letters indicate significant differences (α = 0.05).

Figure 2B). The OC and BC content of our sediments were positively correlated ($R^2 = 0.951$, $P < 0.05$, Figure 3), indicating that the distribution characteristics of the BC content depend on those of the OC.

Black carbon enrichment factors in different sediment size fractions

Sediment fractions from the Guangdong, Guangxi, and Hainan provinces showed significant accumulation of BC (EF > 1), especially in the 0.25–2 mm and 0.053–0.25 mm size fractions. The highest concentration was found in the 0.25–2 mm fraction, while the lowest was in the <0.053 mm fraction. Sediments from Fujian ($F = 3.338$, $P = 0.048$) and Guangdong ($F = 3.530$, $P = 0.039$) showed significantly different BC enrichment factors according to the particle size fraction. The BC concentrations were the highest in the 0.053–0.25 mm fraction from the Fujian Province, and in the 0.25–2 mm fraction from the Guangdong Province. In contrast, the sediments from Guangxi ($F = 1.515$, $P = 0.236$) and Hainan ($F = 0.406$, $P = 0.752$) showed no significant differences in BC EF among the different sediment size fractions (Figure 4).

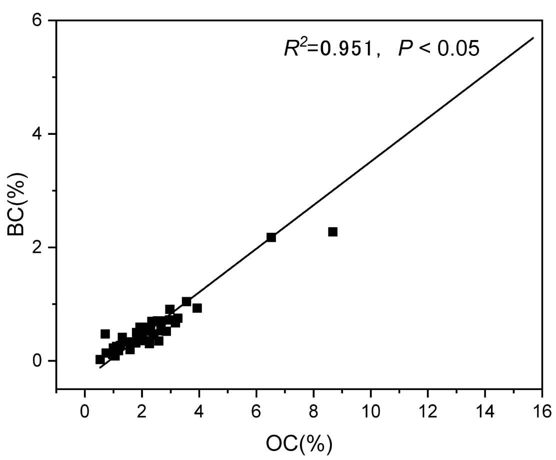

FIGURE 3
Correlation between organic carbon (OC) and black carbon (BC) content of sediments from different mangrove wetlands in China. R^2 and P indicate the fitness and significance of the linear regression models, respectively.

Distribution characteristics of black carbon in sediments with different particle sizes

The DF of BC was calculated to estimate the size fractions of BC that were preferentially enriched (see Figure 5A). The DFs of BC in sediments from different sampling sites did not significantly differ among the different size fractions. The DFs for BC were within the ranges of 1.38–2.02 for the 0.25–2 mm fraction, 1.18–2.22 for the 0.053–0.25 mm fraction, and 0.54–1.95 for the <0.053 mm fraction. With the exception of the <0.053 mm fraction from the Hainan Province, the larger sediment fractions (0.053–0.25 mm and 0.25–2 mm) had a higher DF than those with small fractions (<0.053 mm).

To estimate the contribution from each size fraction to the total BC content of the bulk sediments, mass loading was calculated as shown in Figure 5B. In the sediment from the Fujian Province, the largest BC mass loading with an average of 57% was found in the <0.053

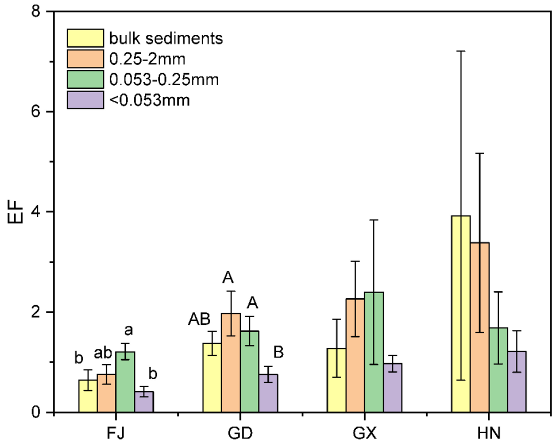

FIGURE 4
Enrichment factors (EF) for black carbon in sediment fractions from different mangrove wetlands in China. Different lowercase and uppercase letters indicate significant differences (α = 0.05) in the samples from Fujian (FJ) and Guangdong (GD), respectively. GX, Guangxi; HN, Hainan.

mm fraction, followed by the 0.053–0.25 mm fraction (28%). The three size fractions between 0.25 and 2 mm accounted for 15% of the BC storage in the sediments. In contrast, among the sediments from the Guangdong, Guangxi, and Hainan provinces, the BC mass loading was higher among the larger particles and the lowest among the fine particles.

Correlation between different black carbon indexes and its sources

The BC/OC ratio in our mangrove sediments ranged from 0.21 to 0.29, and there was no difference among the sampling plots (**Figure 6A**). The BC/OC ratio is linked with the sources of BC accumulation (Wang et al., 2022). A BC/OC value of approximately 0.1 indicates that the BC is mainly derived from biomass combustion, whereas a value of approximately 0.5 indicates that the BC is mainly derived from mineral combustion (Ruellan and Cachier, 2001; Bucheli et al., 2004). The BC/OC ratio in our is higher than 0.1, but lower than 0.5, which indicated that the BC originated from a mixture of biomass and mineral combustion. The average $\delta^{13}C$ value across all the sampled sites ranged from –28.07 to –26.26‰. The average value of $\delta^{13}C$ in the sediments varied according to the sampling plot, and the sediments from the Fujian Province had considerably higher $\delta^{13}C$ values than those from the other provinces (F = 7.36, P = 0.031, **Figure 6B**), which indicated the proportion of BC from biomass combustion in mangroves in Fujian Province is higher than that in other Provinces.

Discussion

Compared with the other regions, the OC content in our study area was lower than those reported for mangrove forests in Australia (Alongi et al., 2000, 2003), Thailand (Alongi et al., 2001, 2002), Brazil (Sanders et al., 2010), and Micronesia (Kauffman et al., 2011). However, they were similar to those reported for Japan (Khan et al., 2007), India (Ray et al., 2011), and Vietnam (Tue et al., 2012). This indicated that there are significant differences in OC concentration among different regions. Previous studies showed that OC content is predominantly influenced by tidal gradient, mangrove forest age, biomass and productivity, species composition, and sedimentation of suspended matter (Gleason and Ewel, 2002; Khan et al., 2007; Cerón-Bretón et al., 2011). In our results, OC and BC contents in Hainan Province where *Rhizophora stylosa* is a dominant species was significantly higher than other Provinces. Since BC content is affected by mangrove tree species (Guo et al., 2018), this might be the driving factor of a higher accumulation of BC in Hainan province. In addition, mangrove cover, climate factors and anthropogenic disturbances also had impact on BC content in sediments (Charles et al., 2020; Meng et al., 2022). Therefore, these factors mentioned above should be considered in the future researches.

In addition, BC accumulates in all sampling sites because of its high resistance to oxidation (Luz et al., 2010; Guo et al., 2018). Significantly high BC content recorded from the larger size fractions indicates that pronounced accumulation of BC occurred in these fractions. Larger size fractions could enrich more BC than the fine fractions due to their larger pores and easier adsorption and BC wrapping (Zong et al., 2016). The contribution from large particles to the BC content was generally greater than that of the small particles. However, fine particles are easily suspended and can be a source of atmospheric aerosols in the environment, and then might have impact on the air quality of mangrove ecosystems. Therefore, the tendency for BC to be adsorbed into finer fractions should be taken into consideration during environmental risk assessments and atmospheric pollution control (Liu et al., 2019). If we could immobilize airborne BC in mangrove sediments, it would mitigate the effects of BC on climate and human health. However, a higher mass loading of BC was observed among fine particles in samples from the Fujian Province, and among the larger particles in samples from the Guangdong, Guangxi, and Hainan provinces. Previous studies showed that BC produced by biomass would form a well-developed pore structure and specific surface area, while BC produced by fossil fuel would inhibit the formation of its pore structure (King et al., 2008; Edmondson et al., 2015; Gao et al., 2016; Zong et al., 2016). BC with well-developed pore structure and specific surface area is easier to get into fine particles. Our results indicated that the proportion of BC from biomass in mangroves in Fujian Province is higher than that in other Provinces, this might be the

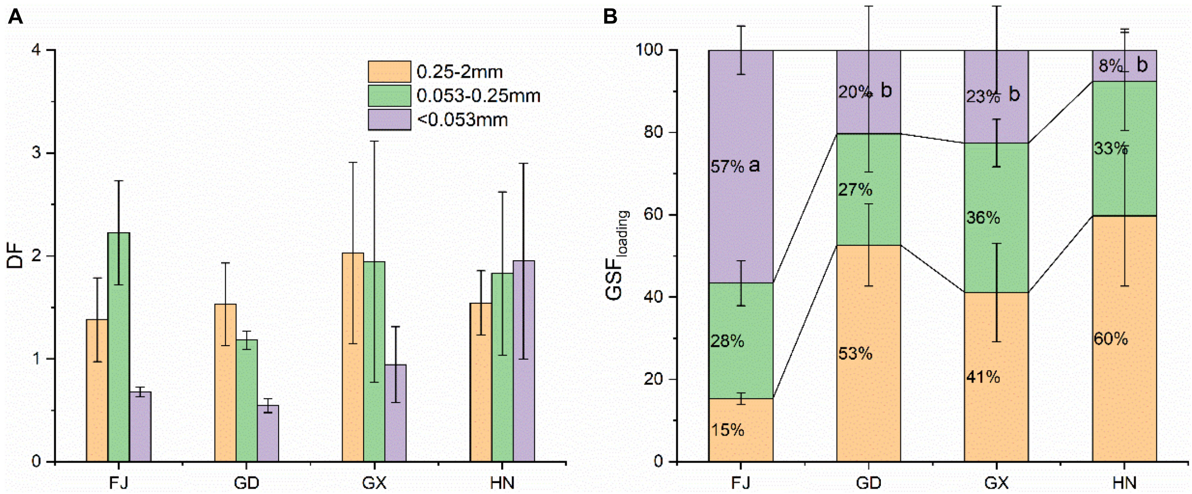

FIGURE 5
Distribution factors (DF, **A**) and mass loading ($GSF_{loading}$, **B**) for different size sediment fractions from mangrove wetlands in China. Lowercase letters indicate significant differences (α = 0.05). FJ, Fujian; GD, Guangdong; GX, Guangxi; HN, Hainan.

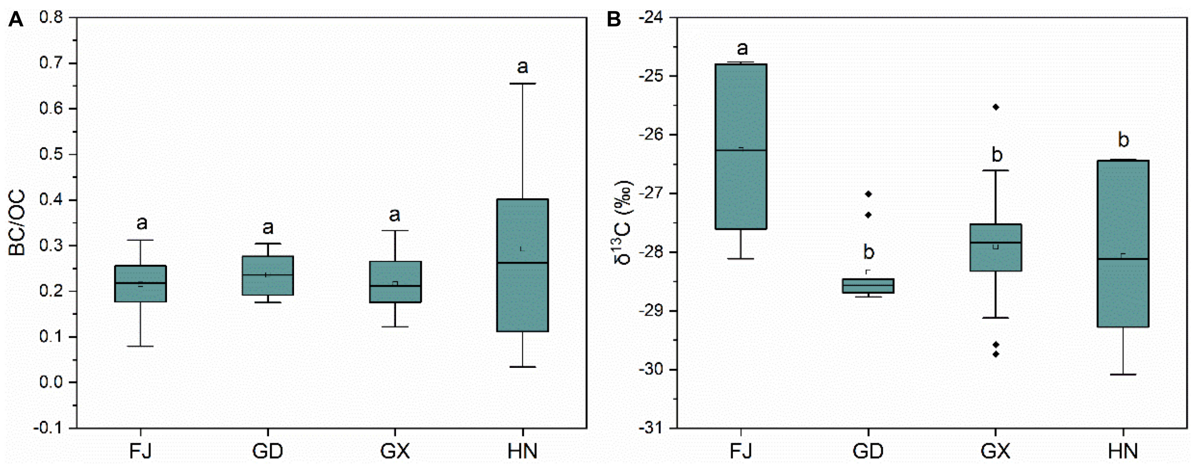

FIGURE 6
Boxplots showing the ratio of black carbon (BC) to the organic carbon (OC) **(A)** and $\delta^{13}C$ content **(B)** in the sediments from different mangrove wetlands in China. Different lowercase letters indicate significant differences (α = 0.05). FJ, Fujian; GD, Guangdong; GX, Guangxi; HN, Hainan.

reason of a higher accumulation of BC in fine particles Fujian province.

At present, many methods have been applied to identify the source of BC: ^{14}C analysis, ^{13}C analysis, BC/OC ratio, polycyclic aromatic hydrocarbons isomer ratio, particle size and morphological characteristics (Wang et al., 2012). Among them, the BC/OC ratio is often used to distinguish the source of BC due to its high reliability and low cost. However, we cannot determine the main source of BC when the source of BC is a combination of biomass and mineral combustion. Therefore, ^{13}C analysis is often combined with the BC/OC ratio to identify the sources of BC (Wang et al., 2013). Generally, the $\delta^{13}C$ value generated by fossil fuel combustion is much lower than that of plant carbon isotopes. For example, the average $\delta^{13}C$ value of petroleum is about −28‰, the average $\delta^{13}C$ value of coal is about −24.5‰, and the $\delta^{13}C$ value of BC particles in motor vehicle exhaust is between −25.9 and −27.6‰ (Guo et al., 2016). Therefore, if a large amount of BC produced by fossil fuel enters the atmosphere or soil, the $\delta^{13}C$ value of BC in atmospheric or soil will decrease. The $\delta^{13}C$ value of BC in Guangdong, Guangxi and Hainan was lower than that in Fujian, which indicated that the proportion of BC originating from fossil fuels in these three provinces is higher than that in Fujian Province. In addition, the contribution of fossil carbon to the BC pool was spatially heterogeneous, which could be related to differences in the distance to landmass, land cover and socioeconomic development. In conclusion, BC has accumulation in some degree in mangrove area in China. Although BC accumulation is beneficial to mangrove carbon storage, BC will enter the atmosphere firstly, which may aggravate the greenhouse effect and endanger human health. Therefore, we should pay more attention to the BC content in the atmosphere in this area, try to reduce the BC content in the atmosphere, and increase the BC content in the soil and sediments. In addition, BC content generally increases with depth (Guo et al., 2018). However, our sampling depth was only 5 cm, therefore, our results only showed the distribution characteristics of the surface sediments, this is also a limitation of this study.

Conclusion

In this study, the content and distribution of BC was analyzed in mangrove sediments from China. Sediments from the Hainan Province had much higher OC and BC content than those from the other provinces. The sediment OC content showed a significant positive relationship with the BC content. The BC showed a significant accumulation in sediments of differing particle sizes, especially in the size fractions of 0.25–2 mm and 0.053–0.25 mm. The significantly high BC content found in the larger size fractions indicates that pronounced accumulation of BC occurred in these fractions. The contribution of large particles to the BC content was generally greater than that of the small particles. The BC/OC ratio in sediments from different mangroves ranged from 0.21 to 0.29, which indicates that BC originated from a mixture of biomass and mineral combustion. Average $\delta^{13}C$ values in the sediments varied between the sampling plots, and sediments from the Fujian Province showed considerably higher $\delta^{13}C$ values than those from the other provinces. This indicates the proportion of BC from biomass combustion in mangroves in Fujian Province is higher than other Provinces. Our findings indicate that BC content and its $\delta^{13}C$ values in sediments vary between provinces, which provides theoretical support and guidance for future carbon storage estimation and carbon sink increases in mangrove forests. However, we did not estimate the contribution of mangrove BC storage to the sediment carbon pool, which should form the focus of further study.

Data availability statement

The original contributions presented in this study are included in the article/supplementary material, further inquiries can be directed to the corresponding author/s.

Author contributions

MW: conceptualization, methodology, software, and writing-original draft preparation. YS: data curation. HZ and WW: visualization and investigation. LD: software and validation. PT: supervision, writing-reviewing and editing. All authors contributed to the article and approved the submitted version.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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