

# Distribution and sources of sedimentary organic matter in different aquaculture areas of northeastern Zhanjiang Bay using stable carbon and nitrogen isotopes

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## Abstract

Zhanjiang Bay is a major aquaculture area in China with many types of mariculture products (such as oysters, fish, and shrimp). The culture area and shrimp output in Zhanjiang Bay are ranked first in China. We investigated the total organic carbon (TOC), total nitrogen (TN), TOC/TN ratio, and stable isotopes ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) of the fish and shrimp feed, fish and shrimp feces, and sedimentary organic matter (SOM) in and around different aquaculture areas of northeastern Zhanjiang Bay to study the impact of aquaculture activities on SOM. The average TOC contents of fish and shrimp feed were  $39.20\% \pm 0.91\%$  and  $39.29\% \pm 0.21\%$ , respectively. The average TOC content in the surface sediments of the oyster culture area, the mixed (fish and shrimp) culture area, and the cage fish farm area were  $0.66\%$ ,  $0.88\% \pm 0.10\%$ , and  $0.58\% \pm 0.19\%$ , respectively, which may indicate that mixed culture had a greater impact on SOM. The relatively high TOC and TN contents and relatively low TOC/TN ratios, and  $\delta^{15}\text{N}$  values in the upper layer of the core sediment in the mixed culture area could also support the significant influence of mixed culture. The average  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of fish and shrimp feed were  $-20.6\% \pm 2.2\%$  and  $1.8\% \pm 1.2\%$ , respectively, which were different from the isotopic values of SOM in the study area.  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for SOM in different aquaculture areas were different from those of nearby reference stations, probably reflecting the influence of aquaculture. The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in the oyster culture area ( $-25.9\%$  and  $6.0\%$ , respectively) seemed to have reduced  $\delta^{13}\text{C}$  and enriched  $\delta^{15}\text{N}$  relative to those of the reference station ( $-24.6\%$  and  $5.8\%$ , respectively). This may reflect the influence of organic matter on oyster culture. The  $\delta^{15}\text{N}$  value of the station in the mixed culture area ( $7.1\% \pm 0.4\%$ ) seemed to be relatively enriched in  $\delta^{15}\text{N}$  relative to that of the reference station ( $6.6\%$ ). Sedimentation and the subsequent degradation of organic matter from mixed cultures may have contributed to this phenomenon. The surface sediment at the cage fish farm area seemed to be affected by fish feces and primary production based on the indication of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values. The sediment core at the mixed culture region (NS6) had lower TOC/TN ratios and more positive  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values than the sediment core at the oyster culture area, suggesting a higher proportionate contribution of marine organic matter in the mixed culture area. In summary, oyster culture, mixed culture, and cage fish culture in northeastern Zhanjiang Bay had a certain degree of impact on SOM, and mixed culture had more significant influences on SOM based on the high TOC contents and the significant vertical variations of TOC/TN ratio and  $\delta^{15}\text{N}$  value in the sediment of this area. This study provides new insights into the impact of aquaculture activities on SOM content.

**Key words:** sedimentary organic matter, aquaculture, stable isotopes, sources, Zhanjiang Bay

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## 1 Introduction

China has large fisheries and aquaculture industries, most of which are located in coastal areas. Large volumes of uneaten feed and excrement are thrown into aquaculture water due to the businesses' rapid growth in the fisheries and aquaculture sectors (Liu et al., 2018). This results in the accumulation of organic mat-

ter, anoxic conditions, or the production of ammonium and sulfides in sediments (Zhuang et al., 2023), which may have adverse effects on aquaculture and affect its sustainable development (Yokoyama et al., 2006; Jiang et al., 2012; Srithongouthai and Tada, 2017; Liu, 2019; Rubio-Portillo et al., 2019; Wang et al., 2022).

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Considerable researchers have used stable isotopes of carbon and nitrogen ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) and the total organic carbon/total nitrogen ratio (TOC/TN ratio) to trace the sources of sedimentary organic matter (SOM) in marine and coastal regions (Yamada et al., 2003; Sarkar et al., 2016; Zhou et al., 2018; Liao et al., 2018; Lu et al., 2020). This is because the stable carbon and nitrogen isotope compositions and TOC/TN ratios from different sources differ (Brandes and Devol, 2002; Lamb et al., 2006; Wada and Hattori, 1991; Bouillon et al., 2008). Many studies have indicated that  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and TOC/TN are valuable indicators of aquaculture environments (Yamada et al., 2003; Pan et al., 2019; Wang et al., 2022). For example, Yamada et al. (2003) indicated that the sedimentary  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in fish farms reflect the changes in feed. The aquaculture-discharged organic matter had different isotopic compositions from those of autochthonous organic matter. Therefore,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  can be used to evaluate the impact of aquaculture (Wang et al., 2022).

Many studies have investigated the influence of aquaculture on the environment based on  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and TOC/TN ratio. Yamada et al. (2003) investigated the vertical profile of TOC/TN in sediments at the Hazamaura fish farm. They found that relatively low TOC/TN ratios in the upper layers may reflect the effects of feeding. Yokoyama et al. (2006) studied  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in the surface sediments of a coastal fish farm in Japan. They suggested that the accumulation of aquaculture-derived organic matter reduced  $\delta^{13}\text{C}$  and enriched  $\delta^{15}\text{N}$ . Jiang et al. (2009) used the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of sediments to trace the sources of SOM in the cage culture area of Nansha Port and found that fish feed and feces were the primary sources of SOM in this area. Some studies have indicated that shellfish biological sediments have a significant impact on the ecological environment in aquaculture areas (Bouchet and Sauriau, 2008; Ren et al., 2015). The effect of aquaculture on Zhanjiang Bay has, however, been the subject of very few studies. As one of the major aquaculture areas in China, Zhanjiang Bay has many types of mariculture, including shellfish (e.g., oysters), fish, shrimp, and crabs. The culture area, seeding, output, and export of shrimp in Zhanjiang Bay rank first in China

(Wu and Yang, 2011). To understand the impact of aquaculture on Zhanjiang Bay and its sustainable development, it is necessary to study the sources and changes in SOM in this region.

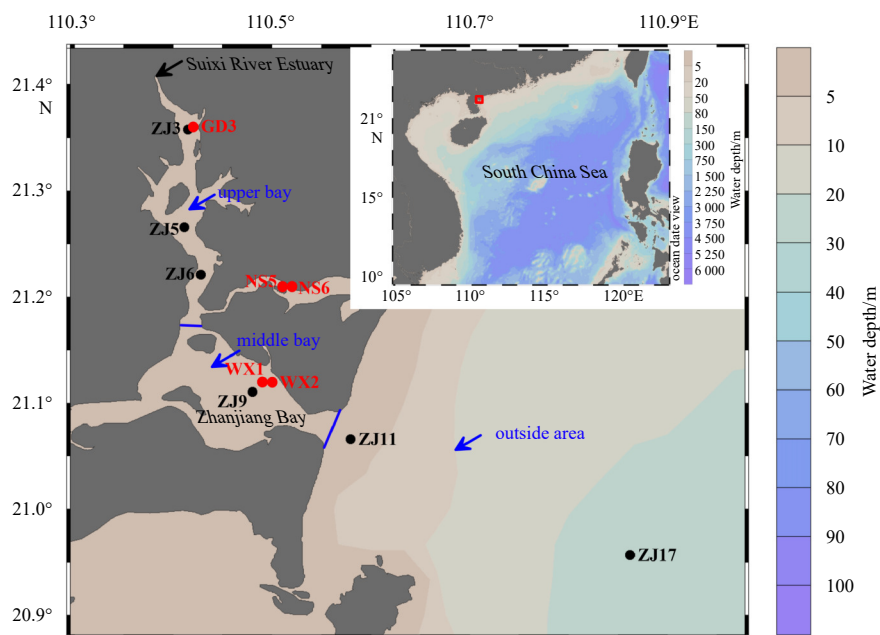
In this study, we investigated the horizontal and vertical distribution of TOC, TN, TOC/TN ratio and stable isotopes ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) of sediments in northeastern and southeastern Zhanjiang Bay, which has many types of aquaculture. The TOC, TN, TOC/TN ratio,  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  of fish and shrimp feed as well as fish and shrimp feces in Zhanjiang Bay were also measured. The purpose of this study was to determine the sources, changes, and effects of aquaculture on SOM in Zhanjiang Bay.

## 2 Materials and methods

### 2.1 Study area

Zhanjiang Bay is located in southern China adjacent to the northwestern South China Sea. It has a subtropical oceanic monsoon climate with suitable seawater temperature and sufficient sunshine time, providing superior natural conditions for the development of marine aquaculture. At the beginning of the 1990s, Zhanjiang aquaculture entered the sea from land, and traditional cages with wooden frame structures were mostly found in the bay. The aquaculture industry in Zhanjiang has developed rapidly since 2000. With the continuous increase in the aquaculture area, the total output of aquaculture in Zhanjiang has also increased. Zhanjiang City is a large aquatic producing city in China. Zhanjiang Bay, the primary aquaculture region of Zhanjiang City, is home to a variety of aquaculture practices, including mixed (fish and shrimp) cultures, oyster, and cage fish. Generally, oyster cultures don't need to be fed. Cage and mixed cultures generally require large amounts of feed to support the rapid growth of fish and shrimp.

Nine surface sediment samples (ZJ3, ZJ5, ZJ6, ZJ9, NS5, WX1, WX2, ZJ11 and ZJ17) and two sediment cores (GD3 and NS6) were collected from northeastern and southeastern Zhanjiang Bay in March 2021 (Fig. 1). The study area can be divided into three regions: the upper bay (downstream of the Suixi River Estu-



**Fig. 1.** Sampling stations in and near Zhanjiang Bay. Stations in red color indicates the stations in aquaculture areas of Zhanjiang Bay. GD3 is located in the oyster culture area; NS5 and NS6 are located in the mixed (fishes and shrimps) culture area; and WX1 and WX2 are located in the cage fish culture area.

ary), the middle bay (the major broad waters in Zhanjiang Bay), and the area outside Zhanjiang Bay (outside area) (Fig. 1). The upper and middle bays are mainly influenced by terrestrial inputs, intensive marine culture, agricultural runoff, and dredging (Li et al., 2020; Lu et al., 2020; Lao et al., 2022a, 2023c). In the upper and middle bays, there are three main types of aquaculture areas: oyster, cage, and mixed (fish and shrimp). Station GD3 was located near the oyster culture area (approximately 100 m), stations NS5 and NS6 were located near the mixed culture area (approximately 100 m), and stations WX1 and WX2 were near the cage fish farm area (approximately 100 m). According to previous studies, aquaculture waste can be dispersed up to 400 m from the aquaculture area (Jiang et al., 2012; Ren et al., 2015; Wang et al., 2022). Therefore, stations GD3, NS5, NS6, WX1, and WX2 were considered to be located in aquaculture areas. Stations ZJ3, ZJ6, and ZJ9 were located close to stations GD3, NS5, and WX1, respectively. In the field of aquaculture, these stations were thought of as references. Zhanjiang Bay was not home to stations ZJ11 and ZJ17. The West Guangdong Coastal Current flows westward along the coast of western Guangdong (Lao et al., 2022b, 2023b), which strongly influences station ZJ11 (Lao et al., 2022a, 2023c).

## 2.2 Sampling and analysis

Surface sediment samples were collected using a Van Veen grab sampler, and sediment core samples were collected using a gravity corer. The sediment core samples were sliced at 2 cm intervals. Then, samples were packed in pre-cleaned polyethylene bags and stored at  $-20^{\circ}\text{C}$  until further treatment. From the aquaculture area in Zhanjiang Bay, a number of fish and shrimp were harvested. Fish and shrimp excrement was removed from their intestines and kept in storage at  $-20^{\circ}\text{C}$ . Local shrimp and fish farmers provided the feed for the fish and shrimp employed in Zhanjiang Bay's aquaculture sector.

The sediment samples were dried at  $50^{\circ}\text{C}$  and grounded to pass through a 100-mesh sieve for analysis. The fish, shrimp feed, and fish and shrimp feces (aquacultural wastes) were also dried at  $50^{\circ}\text{C}$  and grounded. For the analysis of total organic carbon concentration (TOC) and  $\delta^{13}\text{C}$ , sediment samples were pre-treated with 1 mol/L HCl to remove carbonates. Then, the samples were rinsed with ultrapure water, dried at  $50^{\circ}\text{C}$  and homogenized with a pestle and mortar. Total nitrogen concentration (TN) and  $\delta^{15}\text{N}$  were analyzed without pretreatment with HCl. The fish and shrimp feed and fish and shrimp feces (aquacultural wastes) were rinsed with ultrapure water, dried at  $50^{\circ}\text{C}$  and grounded for TOC, TN,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  analysis. The concentrations of TOC and TN, as well as the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values were measured using an elemental analyzer integrated with an isotope ratio mass spectrometer (Flash EA 1112 HT-Delta V Advantages, Thermo). The precisions of duplicate analyses of samples were  $\pm 2\%$  for TOC and  $\pm 3\%$  for TN.  $\delta^{13}\text{C}$  was given as ‰-deviation from the isotope composition of the Vienna PeeDee Belemnite standard (V-PDB).  $\delta^{15}\text{N}$  was given as ‰-deviation from the isotope composition of atmospheric nitrogen. The analytical precision was  $\pm 0.2\%$  for  $\delta^{13}\text{C}$  and  $\pm 0.25\%$  for  $\delta^{15}\text{N}$ .

## 3 Results and discussion

### 3.1 The TOC and TN contents and isotope compositions of fish and shrimp feed and feces

The TOC content for the five fish feed samples used in the aquaculture areas of Zhanjiang Bay ranged from 38.05% to 40.22%, with an average of  $39.20\% \pm 0.91\%$  (Table 1). The TN content of these fish feed samples ranged from 1.64% to 2.57%, with an average of  $2.13\% \pm 0.36\%$ . The TOC/TN ratio for these fish feed samples ranged from 17.3 to 28.6, with an average of  $22.0 \pm 4.3$  (Table 1). In the fish feces of the aquaculture areas in Zhanjiang Bay, the TOC/TN ratio, TOC, and TN contents were 9.2, 53.10%, and 6.71%, respectively (Table 1). The TOC content for the two different dry shrimp feed samples used in the aquaculture areas of Zhanjiang Bay ranged from 39.14% to 39.44%, with an average of  $39.29\% \pm 0.21\%$ . The TN content of these dry shrimp feed samples ranged from 2.39% to 3.27%, with an average of  $2.83\% \pm 0.62\%$ . The TOC/TN ratio for these dry shrimp feed samples ranged from 14.0 to 19.2, with an average of  $16.6 \pm 3.8$  (Table 1). In shrimp feces from the aquaculture areas in Zhanjiang Bay, the TOC/TN ratio and TOC and TN contents were 4.2, 41.76%, and 11.50%, respectively. Fish and shrimp feces were significantly enriched in TOC and TN compared to the fish and shrimp feed (Table 1). The TOC/TN ratio of fish and shrimp feces was significantly lower than that of fish and shrimp feed (Table 1). Fish and shrimp have the ability to drastically alter the composition of organic matter through digestion and absorption, as evidenced by changes in the levels of TOC and TN as well as the TOC/TN ratio.

The  $\delta^{13}\text{C}$  value for the five different fish feed samples used in the aquaculture areas of Zhanjiang Bay ranged from  $-23.2\%$  to  $-17.1\%$ . The  $\delta^{15}\text{N}$  value of these five different fish feed samples ranged from 0.2‰ to 3.2‰. The average  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for fish feed were  $-20.6\% \pm 2.2\%$  and  $1.8\% \pm 1.2\%$ , respectively (Table 1). The  $\delta^{13}\text{C}$  value for the two shrimp feed samples used in the aquacultures of Zhanjiang Bay ranged from  $-20.5\%$  to  $-16.8\%$ . The  $\delta^{15}\text{N}$  value of these two different shrimp feed samples ranged from 1.1‰ to 1.6‰. The average  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for the shrimp feed were  $-18.6\% \pm 2.6\%$  and  $1.4\% \pm 0.4\%$ , respectively (Table 1).

The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for the fish feces in the aquaculture areas of Zhanjiang Bay were  $-25.0\%$  and 6.2‰, respectively. Compared with the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of fish feed obtained in this study, the fish feces were 4.4‰ reduced in  $\delta^{13}\text{C}$  and 4.5‰ increased in  $\delta^{15}\text{N}$ , respectively. The decrease of  $\delta^{13}\text{C}$  and increase of  $\delta^{15}\text{N}$  in fish feces were also reported by other studies (Yokoyama et al., 2006; Franco-Nava et al., 2004; Wang et al., 2022). The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for the shrimp feces in the aquaculture areas of Zhanjiang Bay were  $-23.3\%$  and 4.4‰, respectively. Compared with the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of shrimp feed obtained in this study, the shrimp feces were 4.7‰ reduced in  $\delta^{13}\text{C}$  and 3.0‰ increased in  $\delta^{15}\text{N}$ , respectively. The reduction of  $\delta^{13}\text{C}$  and increase in  $\delta^{15}\text{N}$  can be explained by the effective digestion of feed that has relatively enriched  $\delta^{13}\text{C}$  and decreased  $\delta^{15}\text{N}$ , then leading to the excretion of undigested materials that have reduced  $\delta^{13}\text{C}$  and increased  $\delta^{15}\text{N}$  value.

**Table 1.** The carbon and nitrogen contents,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of fish and shrimp feed and feces in the study area

	$\delta^{13}\text{C}/\text{‰}$	$\delta^{15}\text{N}/\text{‰}$	TOC/%	TN/%	TOC/TN ratio
Fish feed	$-20.6 \pm 2.2$	$1.8 \pm 1.2$	$39.20 \pm 0.91$	$2.13 \pm 0.36$	$22.0 \pm 4.3$
Fish feces <sup>a</sup>	-25.0	6.2	53.10	6.71	9.2
Shrimp feed	$-18.6 \pm 2.6$	$1.4 \pm 0.4$	$39.29 \pm 0.21$	$2.83 \pm 0.62$	$16.6 \pm 3.8$
Shrimp feces <sup>a</sup>	-23.3	4.4	41.76	11.50	4.2

Note: <sup>a</sup> Due to the small amount of fish and shrimp feces, only one sample of fish feces and one sample of shrimp feces were analyzed.

### 3.2 Distributions and implications of TOC and TN in sediments of northeastern and southeastern Zhanjiang Bay

In this study, the TOC contents of surface sediments and sediment core samples collected from northeastern and southeastern Zhanjiang Bay were in the range of 0.44%–1.32% (Figs 2 and 3a). In the surface sediments, TOC content ranged from 0.44% to 1.09%, with the lowest value at station WX1 and the highest value at ZJ6. The average TOC content of surface sediment in the oyster culture area (GD3), mixed culture area (NS5 and NS6) and cage fish farm area (WX1 and WX2) were 0.66%, 0.88%  $\pm$  0.10%, and 0.58%  $\pm$  0.19%, respectively. The area with mixed culture (fish and shrimp) exhibited the highest TOC content in relation to the other two sections, potentially suggesting that aquaculture had a significant impact on this area. Compared with the corresponding reference stations (ZJ3, ZJ6, and ZJ9), the TOC contents in the aquaculture areas were not high (Fig. 1, Table 2). This may be because these reference stations are affected by terrestrial inputs, wastewater discharge, dredging, etc. (Lu et al., 2020; Lao et al., 2022a, 2023c). Zhanjiang Bay is also frequently affected by typhoons (Chen et al., 2021a). Typhoons can cause sediment disturbance and accelerate the decomposition of organic matter (Zhou et al., 2021; Lu et al., 2022; Lao et al., 2023a), which may also be an important reason why the TOC content in the aquaculture areas of Zhanjiang Bay was not very high compared to that at the reference stations.

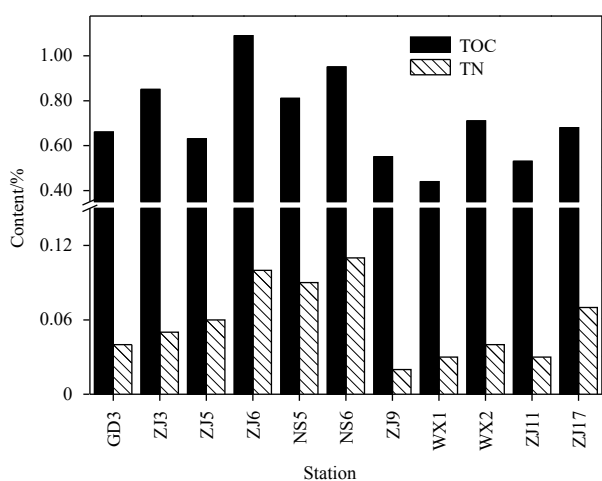


Fig. 2. The distributions of TOC and TN in surface sediments of northeastern and southeastern Zhanjiang Bay.

Vertical profiles of TOC in the sediment cores showed that TOC varied between 0.54% and 0.85% at station GD3, and between 0.67% and 1.32% at station NS6 (Fig. 3a). The average TOC content at core NS6 (0.95%  $\pm$  0.15%) was significantly higher than that of GD3 (0.68%  $\pm$  0.10%). In core NS6, the TOC content of the upper layers was generally higher than that of the deeper layers (Fig. 3a). A similar vertical distribution pattern of TOC was observed in the fish farming area of Gokasho Bay (Yamada et al., 2003).

The TN content of the surface and core sediment samples ranged from 0.02% to 0.12% (Figs 2 and 3b). In the surface sediments, the TN content ranged from 0.02% to 0.11%, with the lowest value at station ZJ9 and the highest at station NS6 (Fig. 2, Table 2). In the sediment cores, TN varied between 0.03% and 0.05% at station GD3 and varied between 0.07% and 0.12% at station NS6 (Fig. 3b). Similar to the vertical distribution of TOC, the TN content in the upper layers of core NS6 was higher than that in the deeper layers (Fig. 3b). In this sediment core, TN ranged from 0.08% at 24–26 cm to 0.11% at 0–2 cm. The relatively high values of TOC and TN at core NS6 in the upper layers indicated the accumulation of high concentrations of organic material, which is likely related to the rapid development of aquaculture in Zhanjiang Bay in recent years. The high TOC and TN contents of the aquaculture waste (fish and shrimp feed, and feces) support this conclusion (Table 1). Station NS6 was located in the mixed (fish and shrimp) culture area of Zhanjiang Bay. Large amounts of fish and shrimp feed were added to support the rapid growth of fish and shrimp. Therefore, much uneaten feed and fish and shrimp feces can accumulate in the sediment, leading to high TOC and TN contents in the upper layers of core NS6. Previous studies have also shown that aquaculture can increase TOC content in sediments (Yamada et al., 2003; Liu et al., 2014). The relatively low  $\delta^{15}\text{N}$  value and low TOC/TN ratio in the upper layers of core NS6 could also reflect the influence of aquaculture (see the discussion in Sections 3.3 and 3.4). In both the surface and core samples taken from northeastern and southeastern Zhanjiang Bay, a substantial positive linear association was discovered between TOC and TN ( $R^2 = 0.77$ ,  $P < 0.001$ ; Fig. 4). This indicates that the sources of TN were similar to those of TOC in the study area. The TOC and TN sources in the study area may have been affected by a combination of terrestrial inputs, aquaculture, and marine autogenesis. The later section will discuss the specific sources for TOC and TN in different areas based on other parameters such as TOC/TN and  $\delta^{15}\text{N}$ .

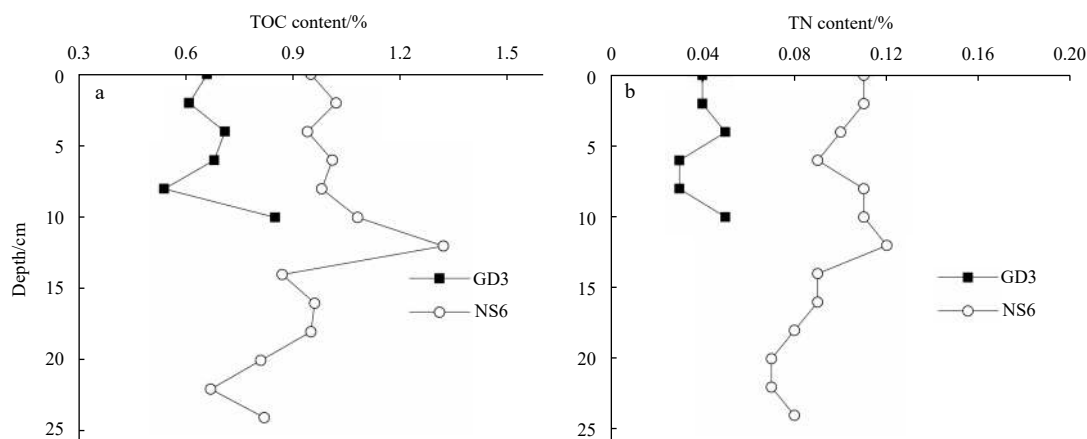
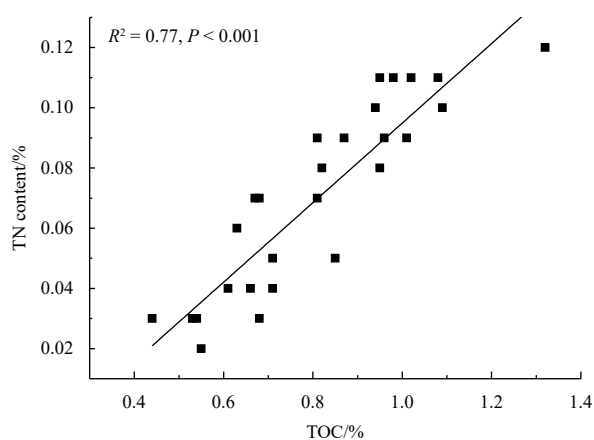


Fig. 3. Vertical distribution of TOC (a) and TN (b) in core sediments of GD3 and NS6.

**Table 2.** The TOC and TN contents, TOC/TN ratio,  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  in surface sediments at each station

Station	TOC/%	TN/%	TOC/TN ratio	$\delta^{13}\text{C}/\text{‰}$	$\delta^{15}\text{N}/\text{‰}$
GD3	0.66	0.04	19.4	-25.9	6.0
NS5	0.81	0.09	10.6	-23.5	7.3
NS6	0.95	0.11	10.1	-23.5	6.8
WX1	0.44	0.03	19.1	-25.3	7.9
WX2	0.71	0.04	19.1	-24.4	6.8
ZJ3	0.85	0.05	19.3	-24.6	5.8
ZJ5	0.63	0.06	11.3	-24.3	7.5
ZJ6	1.09	0.10	12.7	-24.0	6.6
ZJ9	0.55	0.02	31.3	-25.3	7.8
ZJ11	0.53	0.03	20.2	-27.3	4.2
ZJ17	0.68	0.07	11.6	-23.2	6.2

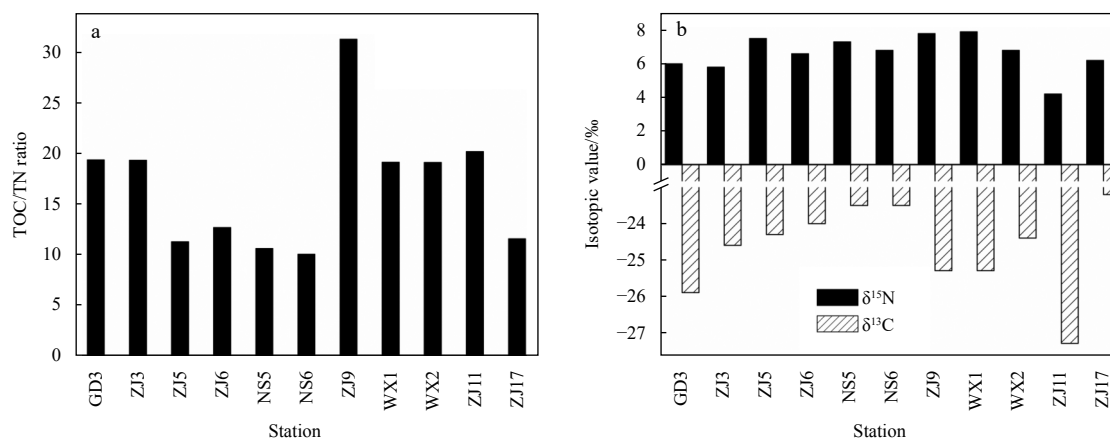
**Fig. 4.** The relationship between TOC and TN in surface and core sediments of northeastern and southeastern Zhanjiang Bay.

### 3.3 Distributions and implications of TOC/TN ratios in sediments of northeastern and southeastern Zhanjiang Bay

The TOC/TN ratio is defined as the molar ratio of TOC to TN. The TOC/TN ratio is usually used to identify the sources of organic matter in coastal and estuarine systems because TOC/TN ratios from different sources are usually different (Krishnamurthy et al., 1986; Wang et al., 2014). Generally, the TOC/TN ratios of terrestrial organic matter are higher than 15, whereas those of marine organic matter range from 5 to 8 (Redfield et al., 1963; LaZerte, 1983). The widespread vascular plants around

Zhanjiang Bay, such as eucalyptus, have TOC/TN ratios of approximately 36 (Kaiser et al., 2014). The mean TOC/TN ratios of fish feed and feces in the aquaculture areas of Zhanjiang Bay obtained in this study were 22.0 and 9.2, respectively (Table 1). The mean TOC/TN ratios of shrimp feed and feces in the aquaculture areas of Zhanjiang Bay were 16.6 and 4.2, respectively (Table 1). Furthermore, post-depositional activities (such as organic matter degradation and inorganic nitrogen adsorption on clay minerals) have the potential to modify the TOC/TN ratio, which could restrict its applicability as a source indicator (Meyers, 1997; Schubert and Calvert, 2001). In this study, the TOC/TN ratios of surface sediments and sediment core samples collected from the northeastern and southeastern coastal Zhanjiang Bay were in the range of 9.9–31.3 (Figs 5a and 6a), which were mainly between the values of marine phytoplankton and terrestrial plants. This ratio indicates that the SOM in the study area was affected by both terrestrial and marine sources. The average TOC/TN ratio in this study was 15.2, which may indicate that terrestrial input was the primary source of organic matter in the sediments of northeastern and southeastern coastal Zhanjiang Bay. However, this result requires further confirmation because organic matter degradation may affect the TOC/TN ratio (Meyers, 1997; Schubert and Calvert, 2001).

In the surface sediments, the TOC/TN ratios ranged from 10.1 to 31.3, with the lowest value at station NS6 and the highest at station ZJ9. The highest TOC/TN ratio at station ZJ9 may be related to dredging and organic matter decomposition (Lu et al., 2020). In core NS6, the TOC/TN ratio varied from 10.1 to 13.9 (Fig. 6a). The TOC/TN ratios at this core sediment were slightly higher at the deeper layer (12 cm to 26 cm), from 11.6 to 13.9 (average: 12.5), and slightly lower at the upper layer (0 cm to 12 cm), from 10.1 to 13.3 (average: 11.2). In core GD3, the TOC/TN ratio ranged from 16.1 to 22.8, which was higher than that in core NS6 (Fig. 6a). This indicates that core GD3 was greatly affected by terrestrial sources. Similar results were obtained for  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  (Sections 3.4 and 3.5). These results confirmed that the TOC/TN ratio was reliable. The TOC/TN ratios at GD3 were higher in the deeper layers (6–12 cm) and lower in the upper layers (0–6 cm), which is comparable to the vertical distribution of core NS6 (Fig. 6a). The upper layer at NS6 and GD3 was loaded with low-TOC/TN material, which may reflect the impact of aquaculture (Yamada et al., 2003). The relatively low TOC/TN ratios in fish and shrimp feces obtained in this study support this conclusion (Table 1). In addition, the increased TOC/TN ratios in the deeper

**Fig. 5.** The distributions of TOC/TN ratio (a),  $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$  (b) in surface sediments of northeastern and southeastern Zhanjiang Bay.

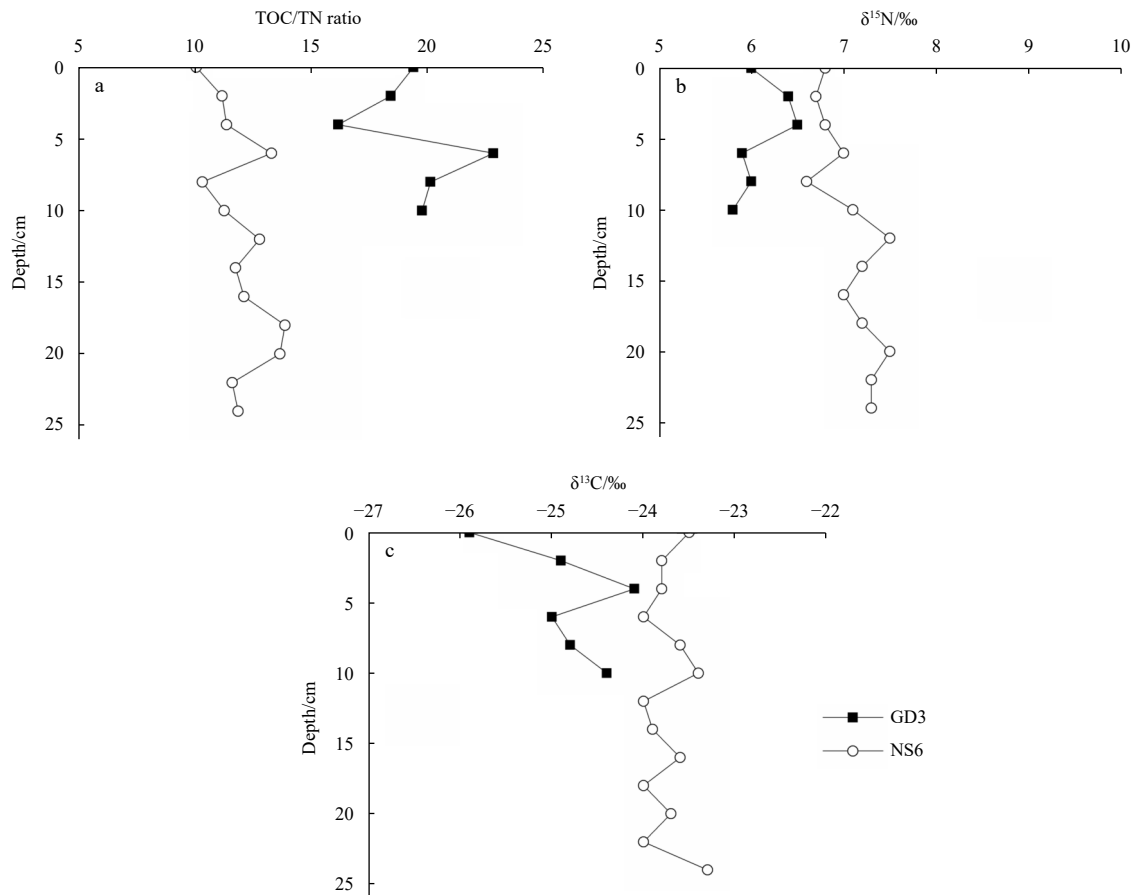


Fig. 6. Vertical distribution of TOC/TN ratio (a),  $\delta^{15}\text{N}$  (b) and  $\delta^{13}\text{C}$  (c) in core sediments of GD3 and NS6.

sections may be related to the influence of diagenesis (preferential loss of N) (Zhang et al., 2009).

### 3.4 Distributions and implications of $\delta^{15}\text{N}$ values in sediments of northeastern and southeastern Zhanjiang Bay

Generally, different organic matter sources have distinct ranges of  $\delta^{15}\text{N}$  values. The  $\delta^{15}\text{N}$  values of organic matter derived from marine phytoplankton range from 5.0‰ to 7.0‰ (Brandes and Devol, 2002; Lamb et al., 2006). The  $\delta^{15}\text{N}$  values of terrestrial vascular plants range from -5‰ to +18‰, with an average value of -3‰ (Wada and Hattori, 1991). Nitrogen from urban sewage and agriculture has high  $\delta^{15}\text{N}$  values (10‰–22‰) (Cole et al., 2004). The average  $\delta^{15}\text{N}$  values of fish and shrimp feed used in the aquaculture areas in Zhanjiang are 1.8‰ and 1.4‰, respectively (Table 1). The average  $\delta^{15}\text{N}$  values of fish and shrimp feces in the aquaculture areas of Zhanjiang are 6.2‰ and 4.4‰, respectively (Table 1).

The  $\delta^{15}\text{N}$  values of surface and sediment core samples collected from northeastern and southeastern Zhanjiang Bay ranged from 4.2‰ to 7.8‰ (Figs 5b and 6b), which were within the ranges of  $\delta^{15}\text{N}$  values of marine phytoplankton, aquaculture wastes, sewage, and terrestrial plants, indicating the impact of these sources. In the surface sediments of the study area,  $\delta^{15}\text{N}$  values were highly variable, with the lowest value (4.2‰) at the bay mouth (ZJ11) and the highest value (7.9‰) at the area of middle Zhanjiang Bay (WX1). Station ZJ9 also had a high  $\delta^{15}\text{N}$  value (7.8‰). Nitrogen from urban sewage and agriculture is enriched in  $\delta^{15}\text{N}$ , with  $\delta^{15}\text{N}$  values ranging from 10‰ to 25‰ (Kendall, 1998; Cole et al., 2004). The high  $\delta^{15}\text{N}$  values at ZJ9 and

WX1 may suggest pollution from sewage and agricultural runoff (Liao et al., 2018). According to Miyake and Wada (1971), isotopic fractionation by bacteria during organic decomposition can increase the  $\delta^{15}\text{N}$  value of remaining organic matter. Because stations ZJ9 and WX1 were located in the central and coastal parts of middle Zhanjiang Bay, respectively, they were less likely to be affected by wastewater than the other stations in Zhanjiang Bay. Therefore, we concluded that the high  $\delta^{15}\text{N}$  values at ZJ9 and WX1 were probably related to the degradation of organic matter. The West-Guangdong Coastal Current may be the primary physical factor for the anomaly of a low  $\delta^{15}\text{N}$  value at station ZJ11. Because the coastal current is made up of diluted Zhujiang River water, it is heavily contaminated on land (Lao et al., 2022b, 2023b). The  $\delta^{15}\text{N}$  value of terrestrial vascular plant has an average value of -3‰ (Wada and Hattori, 1991). The anomaly of a low  $\delta^{15}\text{N}$  value at station ZJ11 may be related to the terrestrial organic matter carried by the West-Guangdong Coastal Current. This result could also be supported by the distribution of  $\delta^{13}\text{C}$ , which will be discussed in Section 3.5. The  $\delta^{15}\text{N}$  values at the surface sediments of ZJ3 and GD3 were generally low compared with the other stations (except station ZJ11) (Fig. 5b, Table 2), suggesting the influence of terrestrial organic matter from the Suixi River. The relatively low  $\delta^{13}\text{C}$  could also reflect the influence of terrestrial input (Fig. 5b, Section 3.5).

The  $\delta^{15}\text{N}$  values ranged from 5.8‰ to 6.5‰ in the core sediment of GD3, and from 6.6‰ to 7.5‰ in the core sediment of NS6 (Fig. 6b). The relatively higher  $\delta^{15}\text{N}$  values of NS6 (average: 7.1‰) than those of GD3 (average: 6.1‰) provide evidence for the different sources of SOM in these areas. The stronger influ-

ence of marine organic matter at station NS6 may contribute to its relatively higher  $\delta^{15}\text{N}$  values, which can also be supported by the result of  $\delta^{13}\text{C}$  (Fig. 5b; Section 3.5). The  $\delta^{15}\text{N}$  value of WX2 is 6.8‰, significantly lower than that of WX1 (7.9‰). Because stations WX1 and WX2 were in the cage culture area, they were also influenced by fish feed and feces. The influence of different degrees of organic decomposition may cause the different  $\delta^{15}\text{N}$  values of these two stations. Central Zhanjiang Bay, where the navigation channel is located, is affected by dredging (Zhang et al., 2012; Lu et al., 2020). Dredging can partially expose surface sediments to solar heating, which can cause the severe degradation of organic matter (Talbot and Livingstone, 1989; Das et al., 2008). The WX1 station could be significantly impacted by dredging due to its close proximity to Zhanjiang Bay's center. Consequently, organic matter decomposition had a significant impact on station WX1, as evidenced by its high  $\delta^{15}\text{N}$  value (Miyake and Wada, 1971; Lu et al., 2020).

The  $\delta^{15}\text{N}$  values of core NS6 exhibited a gradual upward decrease, from 7.5‰ at 22 cm to 6.7‰ at 2 cm (Fig. 6b). Station NS6 is located in a mixed aquaculture area. The  $\delta^{15}\text{N}$  value for fish feces obtained in this study was 6.2‰ (Table 1). The increasing aquaculture activities in recent years can explain the upward decrease in  $\delta^{15}\text{N}$  value at station NS6 (McGhie et al., 2000; Franco-Nava et al., 2004). A study by Anderson et al. (2017) discovered a similar phenomenon. The TOC/TN ratio also reflects the influence of aquaculture at station NS6 (Section 3.3). Besides, the increase of  $\delta^{15}\text{N}$  value at the deeper depth of NS6 may also be related to organic matter degradation (Miyake and Wada, 1971), similar to the result of the TOC/TN ratio (Section 3.3). The  $\delta^{15}\text{N}$  value of core GD3 varied little, probably because this sediment core is short (12 cm long).

### 3.5 Distributions and implications of $\delta^{13}\text{C}$ values in sediments of northeastern and southeastern Zhanjiang Bay

Organic matter from land has an average  $\delta^{13}\text{C}$  value of  $-27\text{‰}$  and  $-14\text{‰}$  for  $\text{C}_3$  and  $\text{C}_4$  plants, respectively. Organic matter from marine phytoplankton has  $\delta^{13}\text{C}$  values between  $-23\text{‰}$  and  $-17\text{‰}$  (Bouillon et al., 2008). The different values of  $\delta^{13}\text{C}$  from different sources make  $\delta^{13}\text{C}$  a ruler for distinguishing between terrestrial and marine origins. The natural ecosystem surrounding Zhanjiang Bay is a subtropical forest, and the dominant cultivated plant is rice ( $\text{C}_3$  plant). Therefore, the contribution of  $\text{C}_4$  plants to SOM was neglected in this study.  $\delta^{13}\text{C}$  in the surface sediments and sediment core samples collected from the northeastern and southeastern Zhanjiang Bay ranged from  $-27.3\text{‰}$  to  $-23.1\text{‰}$  (Figs 5b and 6c). A simple two-end member mixing model can be used to quantify the sources of terrestrial and marine organic matter. The equation used is as follows:

$$\delta^{13}\text{C}_{\text{sample}} = \delta^{13}\text{C}_{\text{ter}} \times f_{\text{ter}} + \delta^{13}\text{C}_{\text{mar}} \times f_{\text{mar}}$$

where  $\delta^{13}\text{C}_{\text{ter}}$  and  $\delta^{13}\text{C}_{\text{mar}}$  are the terrestrial and marine organic matter end-members, respectively;  $f_{\text{ter}}$  and  $f_{\text{mar}}$  are the proportion of organic matter from terrestrial and marine organic matter, respectively; and  $\delta^{13}\text{C}_{\text{sample}}$  is the  $\delta^{13}\text{C}$  value of a certain station. The average  $\delta^{13}\text{C}$  value of the  $\text{C}_3$  plant ( $-27\text{‰}$ ; Meyers, 1997; Pancost and Boot, 2004) and the  $\delta^{13}\text{C}$  value of phytoplankton ( $-20.8\text{‰}$ ), which was collected from the northern South China Sea (He et al., 2010), was chosen as the terrestrial and marine organic matter end-members, respectively. The calculated results showed that approximately 60% of SOM in the study area originated from terrestrial organic matter. This result is consistent with

the TOC/TN ratio, in which terrestrial input was the primary source of organic matter in the sediments of the northeastern and southeastern coast of Zhanjiang Bay.

Relatively negative  $\delta^{13}\text{C}$  values occurred close to the Suixi River Estuary (Fig. 5b), reflecting the influence of terrestrial input. The highest  $\delta^{13}\text{C}$  value was observed at station ZJ17, located in the northwestern South China Sea and relatively far away from Zhanjiang Bay. This indicates that marine organic matter had a significant influence on this station. The lowest  $\delta^{13}\text{C}$  value occurred at ZJ11, outside the Zhanjiang Bay mouth. Organic matter from coastal areas and the West-Guangdong Coastal Current may contribute to the lowest  $\delta^{13}\text{C}$  value at this station. Section 3.4 discusses the impact of the West Guangdong Coastal Current on station ZJ11. The anomaly of low  $\delta^{15}\text{N}$  value at this station could also reflect the impact of the West-Guangdong Coastal Current.

In core NS6, the  $\delta^{13}\text{C}$  values range from  $-24.0\text{‰}$  to  $-23.3\text{‰}$ . In core GD3, the  $\delta^{13}\text{C}$  values ranged from  $-25.9\text{‰}$  to  $-24.1\text{‰}$ . The relatively low  $\delta^{13}\text{C}$  values at GD3 indicate the strong terrestrial influence at this station. There was no apparent vertical distribution pattern for the  $\delta^{13}\text{C}$  values in cores NS6 and GD3.

### 3.6 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values in aquaculture areas and their implications

More information is needed regarding the stable isotope composition of waste feed and feces-derived organic matter in aquaculture. The related data are presented in Table 3. The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  of fish feed and feces from different aquaculture areas generally have different values (Ye et al., 1991; Yokoyama et al., 2006; Jiang et al., 2012; Wang et al., 2022; Table 3). The fish feed used in Zhanjiang Bay's aquaculture sites showed generally lower  $\delta^{15}\text{N}$  values when compared to the findings of these earlier research (Table 3). Little information is available on the stable isotope composition of organic matter in oysters and shrimp. Ren et al. (2015) reported that the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of organic matter from oysters were  $-18.49\text{‰}$  and  $6.53\text{‰}$ , respectively. Our study provides the isotopic values of shrimp feed and feces in Zhanjiang Bay, China. The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of shrimp feed from the aquaculture areas of Zhanjiang Bay were  $-18.6\text{‰}$  and  $1.4\text{‰}$ , respectively. The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of shrimp feces from the aquaculture areas of Zhanjiang Bay were  $-23.3\text{‰}$  and  $4.4\text{‰}$ , respectively.

There were some differences in the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for SOM between stations in the aquaculture areas and those nearby reference stations (Table 2). Station GD3 is located in the oyster culture area of Zhanjiang Bay. Relative to the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of the reference station ZJ3 ( $-24.6\text{‰}$  and  $5.8\text{‰}$ ), the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of station GD3 ( $-25.9\text{‰}$  and  $6.0\text{‰}$ , respectively) seemed to have decreased  $\delta^{13}\text{C}$  and enriched  $\delta^{15}\text{N}$  to some extent. The  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of suspended particulate matter collected at GD3 were  $-27.3\text{‰}$  and  $7.6\text{‰}$ , respectively (unpublished data). The differences in  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values between GD3 and ZJ3 seem to reflect the deposition of organic matter from oysters. However, further research is required to confirm these findings.

The average  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values in the surface sediment of the mixed (fish and shrimp) culture area were  $-23.5\text{‰} \pm 0.0\text{‰}$  and  $7.1\text{‰} \pm 0.4\text{‰}$ , respectively (Table 3). The surface sediment at this area seemed to have enriched  $\delta^{13}\text{C}$  ( $\Delta\delta^{13}\text{C} = 0.5\text{‰}$ ) and enriched  $\delta^{15}\text{N}$  ( $\Delta\delta^{15}\text{N} = 0.5\text{‰}$ ) values relative to the nearby reference station ZJ6 ( $-24.0\text{‰}$  and  $6.6\text{‰}$ , respectively) (Table 2). Compared with the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of fish feed, fish feces, shrimp feed and shrimp feces, the  $\delta^{15}\text{N}$  values of the surface sediment in the mixed culture area were also relatively high. This is

**Table 3.**  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values of samples from different aquaculture areas

Study area	Sample type	$\delta^{13}\text{C}/\text{‰}$	$\delta^{15}\text{N}/\text{‰}$	References
Cage fish area in Poyang Lake, China	Fish feed	-25.1	10.1	Wang et al. (2022)
	Fish feces	-26.3 ± 0.0	14.4 ± 1.6	Wang et al. (2022)
Fish farm in Gokasho Bay, Japan	Fish feed	-20.2	9.7	Yokoyama et al. (2006)
	Fish feces	-24.3	6.3	Yokoyama et al. (2006)
Fish farm in Gokasho Bay, Japan	Fish feed	-21.2	10.4	Yamada et al. (2003)
Cage fish area in Nansha Port, China	Fish feed	-20.38 ± 0.24	6.20 ± 0.13	Jiang et al. (2012)
	Fish feces	-17.57 ± 0.18	7.59 ± 0.34	Jiang et al. (2012)
Oyster culture area in Sanggou Bay, China	Sediment from oyster	-18.49	6.53	Ren et al. (2015)
Aquaculture area in Zhanjiang Bay, China	Fish feed	-20.6 ± 2.2	1.8 ± 1.2	This study
	Fish feces	-25.0	6.2	This study
	Shrimp feed	-18.6 ± 2.6	1.4 ± 0.4	This study
	Shrimp feces	-23.3	4.4	This study
Oyster culture area in Zhanjian Bay, China	Surface sediment	-25.9	6.0	This study
Mixed culture area in Zhanjian Bay, China	Surface Sediment	-23.5 ± 0.0	7.1 ± 0.4	This study
Cage fish farm area in Zhanjian Bay, China	Surface sediment	-24.4	6.8	This study

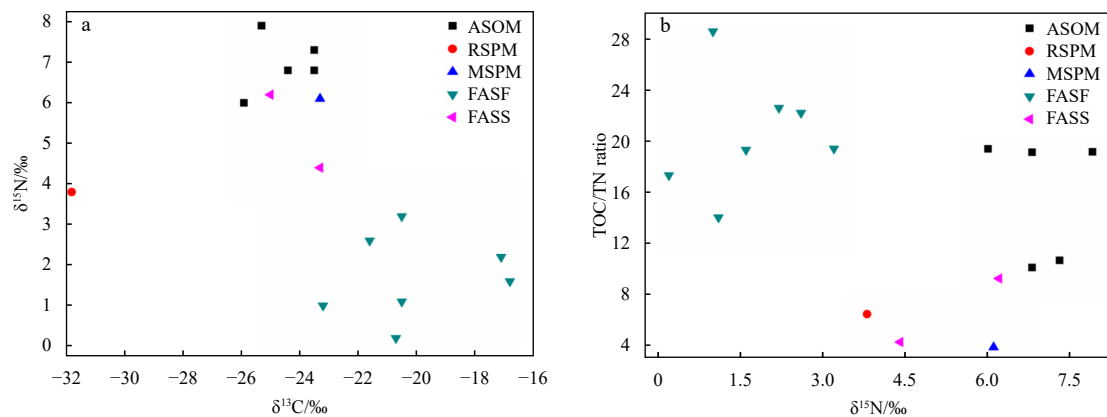
most likely because there was enough degradation of the surface sediment in the mixed (fish and shrimp) culture area, which is formed from the settling of particulate organic matter (fish feed, fish feces, shrimp feed, and shrimp feces). Organic decomposition can increase the  $\delta^{15}\text{N}$  value of remaining organic matter (Miyake and Wada, 1971). At station WX2, the surface sediments had enriched  $\delta^{13}\text{C}$  ( $\Delta\delta^{13}\text{C} = 0.9\text{‰}$ ) and reduced  $\delta^{15}\text{N}$  ( $\Delta\delta^{15}\text{N} = 1.0\text{‰}$ ) values relative to the nearby reference station ZJ9. The relatively low  $\delta^{15}\text{N}$  values of fish feces measured in this study may probably contribute to the reduced  $\delta^{15}\text{N}$  value in the cage fish farm area. The enriched  $\delta^{13}\text{C}$  in this area may be related to the high primary production (Ke et al., 2020; Chen et al., 2021b). Cage fish farming requires large amounts of fish feed, which can provide abundant nutrients and lead to high primary production (La Rosa et al., 2002; Liu, 2019).

### 3.7 SOM sources in aquaculture areas

SOM in different aquaculture areas of Zhanjiang Bay (ASOM) is likely from many sources, such as suspended particulate matter from river input (RSPM) (unpublished data), suspended particulate matter from marine autogenic sources (MSPM) (unpublished data), fish and shrimp feed (FASF), and fish and shrimp feces (FASS). The  $\delta^{13}\text{C}$ ,  $\delta^{15}\text{N}$  and TOC/TN values of SOM in different aquaculture areas of Zhanjiang Bay and the organic matter from different sources were drawn with scatter plots (Fig. 7). In

Fig. 7a, we can see that the  $\delta^{13}\text{C}$  values of ASOM were within the  $\delta^{13}\text{C}$  values of RSPM and MSPM and were close to the  $\delta^{13}\text{C}$  values of FASS and MSPM, indicating that marine organic matter and fish and shrimp feces had a significant contribution to the SOM in aquaculture areas. However, the  $\delta^{15}\text{N}$  values of ASOM were not within the range of RSPM and MSPM and were generally higher than the  $\delta^{15}\text{N}$  values of fish and shrimp feces. The most probable reason for this is that suspended particulate matter and aquaculture waste (including feed and feces) undergo extensive degradation during and after settlement. Isotopic fractionation by bacteria during organic degradation can increase the  $\delta^{15}\text{N}$  value of remaining organic matter (Miyake and Wada, 1971; Cifuentes et al., 1988). In Fig. 7b, the ASOM points were not within the area formed by potential sources, probably due to the influence of organic matter degradation, aquatic feeding, and other processes. The combination of stable isotopes and the end-member mixing model was unable to quantify the impacts of aquaculture since the ASOM locations were outside the region formed by potential sources.

The aquaculture area in Zhanjiang Bay is a complex system affected by multiple processes, such as the settlement of fish and shrimp feces, suspended particulate matter from marine autogenic and river inputs, and organic matter degradation. Quantifying the sources of organic matter in aquaculture using such complex systems is difficult. A previous study also noted that it is diffi-



**Fig. 7.** The plot of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values and  $\delta^{15}\text{N}$  and TOC/TN values for sedimentary organic matter in aquaculture areas (ASOM) and the potential organic matter sources (RSPM: suspended particulate matter from river input; MSPM: suspended particulate matter from marine autogenic; FASF: fish and shrimp feed; FASS: fish and shrimp feces).



cult to identify the sources of organic matter in complex systems (Cifuentes et al., 1988). As was covered in the preceding section, we were able to get qualitative data about the influence of aquaculture even though our study did not yield quantitative results on the sources of SOM in aquaculture areas. Some studies have quantitatively analyzed the contribution of aquaculture to SOM, but these research areas were mainly located in temperate regions such as Ailian Bay, China (Pan et al., 2019) and Gokasho Bay, Japan (Yokoyama et al., 2006). Regions located at relatively high latitudes may have insufficient degradation of organic matter and relatively small isotopic fractionation owing to the relatively low temperatures in these areas.

#### 4 Conclusions

This study investigated the horizontal and vertical distribution of TOC, TN, TOC/TN ratio, and stable isotopes ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) of sedimentary organic matter in different aquaculture areas of northeastern Zhanjiang Bay. It measured the  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values and TOC and TN contents of fish and shrimp feed and feces from the aquaculture areas of Zhanjiang Bay. Compared to the oyster culture and cage fish farm areas, the mixed (fish and shrimp) culture area had a relatively higher TOC content, which may indicate that mixed culture had a relatively strong impact on sedimentary organic matter (SOM). The relatively high TOC and TN contents and relatively low TOC/TN ratios, and  $\delta^{15}\text{N}$  values in the upper layer of the core sediment in the mixed culture area could also support the significant influence of mixed culture.

$\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values for sedimentary organic matter in aquaculture areas differed from those of nearby reference stations. The surface sediment at the oyster culture area seemed to have reduced  $\delta^{13}\text{C}$  and enriched  $\delta^{15}\text{N}$  values, which may reflect organic carbon deposition from oysters. The  $\delta^{15}\text{N}$  ( $\Delta\delta^{15}\text{N} = 0.5\text{‰}$ ) values of the surface sediment at the mixed culture region were comparatively enriched, suggesting the impact of both organic matter decomposition and mixed culture. The surface sediment at the cage fish farm area was affected by fish feces and high primary production based on the indication of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$  values.

In summary, oyster culture, mixed culture, and cage fish culture in northeastern Zhanjiang Bay all had a certain degree of impact on SOM, and mixed culture had more significant influences on SOM based on the high TOC contents and the significant vertical variations of the TOC/TN ratio and  $\delta^{15}\text{N}$  value in the sediment of this area.

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