Distribution of grain size and organic elemental composition of the surficial sediments in Lingding Bay in the Pearl River Delta, China: A record of recent human activity

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\textbf{ABSTRACT}

The Pearl River Delta (PRD) in China is severely affected by intensive human activities. In this study, we used surficial sediments collected in its Lingding Bay (LDB) in December 2016 to analyse the grain size and organic carbon composition and to identify the recent human impact recorded in the sediments. We examined the temporal variations in the spatial distribution of the grain size and organic elemental geochemistry of the surficial sediments by comparison with previous studies. Our study shows that in 2016, there were several patches of coarse-grained sediments, which were not consistent with the hydrodynamic conditions in LDB. Compared with the organic carbon composition in 2005–2006, the $\delta^{13}$C values in 2016 were more negative and the contribution of terrigenous organic matter (TOM) to the surficial sediments was significantly greater. There were also several patches of high TOM values in the outer LDB. The overall coarser sediments and lighter $\delta^{13}$C of organic carbon were in contrast to the decline in the riverine sediment supply and the increased eutrophication in recent decades. Furthermore, the anomalous patches of coarse-grained sediments and terrestrially sourced organic carbon were mainly distributed in the navigation channels and on their adjacent shoals, reflecting channel dredging, dumping, and sand mining activities. Therefore, the origins and spatial distributions of the grain size and organic carbon composition of the surficial sediments in LDB in 2016 were not controlled by hydrodynamic conditions and land–sea interactions, but overwhelmingly by human activities. We suggest that LDB of PRD exemplifies an anthropogenically controlled bay.

\section{1. Introduction}

The estuaries and deltas of the world are facing pressures from both intensive human activities and climate change (Giosan et al., 2014). Human activities in both river basins and delta regions have increased greatly since the 1950s. These activities mainly include reservoir construction, the extraction of groundwater in delta plains, the embankment of shorelines, the dredging of navigation channel, land reclamation, etc. (Milliman, 1997; Nilsson et al., 2005; Syvitski et al., 2005, 2008; Day and Giosan, 2008; Edmonds, 2012; Pethick and Orford, 2013). The role of such activities in the evolution of many deltas, such as those of the Nile, Mississippi, and Yangtze, has exceeded the roles of the natural factors of climate and sea-level change in recent decades (Stanley and Warne, 1993; Lam et al., 2018; Wang et al., 2018). Therefore, it is essential that we clearly understand the extent of human intervention to ensure the better management of these deltas (Day and Giosan, 2008).

The Pearl River Delta (PRD) in South China (Fig. 1) is one of the most important economic centres in China. In 2015, the PRD urban agglomeration became the largest in the world, with a population...
navigation channels were dumped onto nearby shoals and the adjacent shelf (Fig. 3c; Jiang et al., 2009; GBOF, 2014; Wu et al., 2016, 2018). There has also been illegal dumping, the locations of which are still to be identified. Therefore, the channels have become deeper and the shoals have become shallower in LDB (Wu et al., 2016, 2018).

The sedimentary sequence at a river mouth is thought to provide a record of the environmental changes in both the drainage and sedimentary basins. However, it is unclear whether the signatures of the decline in sediment supply and the eutrophication in the offshore area are preserved in the sedimentary records at LDB or whether the sedimentary records have been completely perturbed by coastal engineering projects. Organic geochemical proxies are useful indicators for the variations in freshwater discharge and terrestrial input (Zong et al., 2006; Yu et al., 2010, 2012; Zhan et al., 2012) and ecological environmental changes (Zong et al., 2013). The reduced freshwater discharge and sediment load of the Pearl River (Fig. 2; Dai et al., 2008; Liu et al., 2014) and in-channel sand excavation (Luo et al., 2007; Wu et al., 2016) have weakened the fluvial functions and intensified the brackish-water intrusion into the river mouth. The eutrophication can increase the accumulation of marine-sourced organic carbon (Huang et al., 2003; Cui et al., 2018). However, coastal engineering projects, including channel dredging, sand mining, and ocean dumping, in LDB (Fig. 3c) have the potential to change the particle and geochemical composition of surficial sediments, which is naturally controlled by hydrodynamic and biogeochemical processes. Therefore, it is essential to determine the extent of the human perturbation of the surficial sediments to understand the impact on the hydrological and sediment dynamic processes and the ecological environment in LDB. In this study, we analysed the grain size and organic geochemical composition of surficial sediments collected in LDB in December 2016 (Fig. 4). We also examined the redistribution of sediments in LDB caused by recent human activities by comparing our data with the surficial sediment compositions reported in previous studies (Yu et al., 2010; Xia et al., 2013).

2. Study area

The Pearl River consists of three major tributaries (the West, North, and East Rivers) and has a catchment area of 425,700 km² (Fig. 1a). The mean annual precipitation in the catchment is 1200–2200 mm, mainly occurring during the wet season from April to September (Zhang et al., 2008). The annual discharge from the whole Pearl River system was 282.5 × 10^9 m³ during the period 1957–2016 (Fig. 2a; Pearl River Sediment Bulletin). The average annual sediment load was 81.5 Mt/yr in 1957–1995, but declined rapidly to 48.3 Mt/yr in 1996–2008 and 24.1 Mt/yr in 2009–2016 (Fig. 2a; Pearl River Sediment Bulletin) as a result of the construction of more than 8600 reservoirs from the late 1990s (Wu et al., 2016). The West River is the largest tributary, supplying 216.8 × 10^9 m³/yr freshwater in 1957–2016 (Fig. 2b). The suspended sediment load was 72.7 Mt/yr in 1957–1995, but declined significantly after 1996 and was only 15.4 Mt/yr after 2009 (Fig. 2b). There was no obvious change in the freshwater discharge or sediment load from the North River, which were 41.9 × 10^9 m³/yr and 5.5 Mt/yr, respectively, in the period 1955–2016 (Fig. 2c). The freshwater discharge from the East River was 23.3 × 10^9 m³/yr and the sediment load was 2.2 Mt/yr in 1954–2016 (Fig. 2d).

The PRD is located at 21°5′–23°25′ N and 112°33′–114°10′ E, in South China. One-fifth of the delta region is scattered with bedrock outcrops and terraces, which form a network of river channels on the delta plain and eight outlets to the sea (Fig. 1b; Huang et al., 1982). The Holocene delta is mainly built in the palaeo-Pearl River mouth surrounded by outcrops (Zong et al., 2012). Among the eight present-day outlets, only the Modaomen faces the open sea. Two outlets, the Yamen and Hutiaomen, are located in the sheltered Huangmiao Sea in the western part of the PRD and four (Humen, Jiaomen, Honggili, and Hengmen) are located in LDB in the eastern part of the delta.
The water depth in LDB is usually shallower than 30 m (Fig. 3). The bay is divided into the inner and outer bays by the line from inner Lingding Island to Qi’ao Island (Fig. 1b). Its present-day subaqueous topography is characterized by four shoals and three channels (Fig. 4). The four shoals are the east, middle, west and south shoals, where the water depth is shallower than 5 m. The three channels are all navigable, including the western trough (Lingding Channel), eastern trough (Chuanbi-Longxue-Fanshi Channel; CLF Channel), and southern trough (Tonggu Channel), with water depths between 10 and 20 m. Before the 1970s, only the CLF Channel and the outer Lingding Channel were present in the bay (Fig. 3b; Wu et al., 2018). In the period 1970–1995, sand mining and dredging activities formed the inner Lingding Channel, which is connected to the outer Lingding Channel today (Fig. 3c). Intensive sand mining occurred in 1995–2015, changing most of the middle shoal to deep water (Tang et al., 2011). Dredging also occurred in the east trough during this period. This dredging and sand mining widened the CLF Channel and extended it seaward (Fig. 3c). The Tonggu Channel is an artificial waterway excavated in 2008 (Wu et al., 2016, 2018). There are three important ports, Shekou, Dachanwan, and Mawan, located on the southeast coast of the inner LDB and at the terminal of Tonggu Channel. These three ports, with water depths of 10–20 m, were also artificially excavated in 1995–2005 (Figs. 3c and 4). By contrast, the subaqueous topography on both sides of the Tonggu Channel and the outer Lingding Channel shallowed at a rate of > 10 cm/yr in 1995–2015 (Wu et al., 2018). The southwest part (close to Luhuan Island) of the outer LDB also shallowed significantly (Fig. 3c), which may have been caused by the dumping of dredged material (Wu et al., 2016, 2018).

Irregular semidiurnal tides occur in LDB, with an average tidal range of 0.86–1.57 m and a maximum range of 2.29–3.36 m (Huang et al., 1982). Rectilinear tidal currents occur in the inner LDB and rotary tidal currents in the southeast part of the bay (Mao et al., 2004). The water column is highly stratified in LDB in the wet season (Fig. 3d) and partially mixed in the dry season (Liu et al., 2016). The suspended sediment concentration is usually < 0.5 kg/m³ in the inner LDB and < 0.2 kg/m³ in the outer LDB (Fig. 3e). It is much higher at the bottom than at the surface of the water column owing to resuspension (Liu et al., 2016).

3. Material and methods

We collected a total of 128 surficial sediments from LDB in the PRD in December 2016. These surficial samples were distributed in the following 10 geomorphic units (Fig. 4): 1) CLF Channel, connected to the Humen outlet in the northeast LDB; 2) the inner Lingding Channel, connected to the Humen outlet, which is an artificial shipping channel constructed based on the original west trough in the inner LDB; 3) the outer Lingding Channel, connected to the inner Lingding Channel, which is the artificial shipping channel in the outer LDB; 4) the Qingzhou and Daxi (QD) waterway in the area west of the Lingding Channel, where the water depth is between 5 and 10 m; 5) Tonggu Channel, which is also an artificial shipping channel in the outer LDB; 6) the Qinzhong and Daxi (QD) waterway in the area west of the Lingding Channel, where the water depth is between 5 and 10 m; 7) the east shoal in the north-eastern LDB, where the water depth is shallower than 5 m; 8) the middle shoal, between the CLF and inner Lingding Channels; 9) the west shoal, in the western LDB, where the water depth is shallower than 5 m; and 10) the south shoal, the area east of the outer Lingding Channel in the outer LDB (Fig. 4). All the samples were analysed for grain size and most samples were analysed for organic elemental composition, except when the sample remaining after the grain size analysis was insufficient (Table 1).

The pre-treatment in grain size analysis was as follows. About 5 ml of 10% H₂O₂ was added to each specimen and allowed to react for 24 h at room temperature, after which the sample was heated for 1 h to
remove the organic matter. A further 5 ml of 10% HCl was added and allowed to react on a platen heater. The beaker was filled with distilled water and the reactants were allowed to settle for 24 h before the overlying liquid was removed. This procedure was repeated three times. To disperse the specimen remaining in the beaker, 5 ml of 0.05 mol/l Calgon (NaPO₃)₆ was added and the specimen was shaken in an ultrasonic bath for 15 min to prevent flocculation. The grain size was measured with a Mastersizer 2000 laser diffraction particle size analyser (Malvern, UK), with a measurement range of 0.02–2000 μm.

Samples containing gravels were measured with a Camsizer® X2 particle size and shape analyser (Retsch Technology, Germany).

The pre-treatment in the organic geochemical analysis was as follows. The samples were lyophilized with a vacuum freeze-dryer (SRK, Germany). The freeze-dried specimen was ground with an agate mortar and then sieved through 200 mesh. Half the specimen was wrapped for the measurement of total carbon (TC) and total nitrogen (TN). To the remaining half of each specimen was added 40 ml of 1 mol/l HCl in a centrifuge tube. The specimen-filled centrifuge tube was placed in a water bath for 90 min to fully remove the carbonates. The specimen was centrifuged and washed with ultrapure water four to five times until the pH of the supernatant was neutral. After the specimen was dried and ground, it was ready for measurements of δ¹³C and total organic carbon (TOC). TOC, TC, and TN were measured with a vario EL III element analyser (Germany), using national geochemical reference standard GSD-9, with an accuracy of 0.5%. The δ¹³C measurements were made with a Delta Plus XP isotope ratio mass spectrometer (Thermo Finnigan, USA), with reference standards caffeine (IAEA-600), cellulose (IAEA–CH–3), and black carbon (GBW04407 and GBW04408), with an accuracy < 0.1‰. Both the grain size and organic geochemical analyses were performed at the State Key Laboratory of Estuarine and Coastal Research, East China Normal University.

We compiled the spatial distribution of the grain size of surficial sediments collected in LDB in 1975 and 2003–2004, reported by Xia et al. (2013), and the results of organic geochemical analyses of surficial sediments collected in 2005–2006, reported by Yu et al. (2010). To compare the results of our study with the organic geochemical properties recorded in 2005–2006, we used the same method and end members used by Yu et al. (2010) to estimate the relative proportion of terrigenous organic matter (TOM) in the surficial sediments, with the equation (Calder and Parker, 1968; Schultz and Calder, 1976):

% TOM = (δ¹³Cmarine − δ¹³Corg)/(δ¹³Cmarine − δ¹³Cterrestrial) × 100%

Where δ¹³Cmarine is δ¹³C of the marine end member (−21.0‰; Yu et al., 2010), δ¹³Cterrestrial is δ¹³C of the terrestrial end member (−25.0‰; Yu et al., 2010), and δ¹³Corg is the measured δ¹³C of a given sample in this study.

4. Results

4.1. Grain size distribution

In 2016, the spatial distribution of the grain sizes in the surficial sediments in LDB was characterized by several separated patches of coarse-grained sediments (Fig. 5). It was difficult to define the
relationship between the grain size and the geomorphic units. The particle composition of the sediments in each geomorphic unit was characterized by large variations (Table 1). In general, coarse-grained and fine-grained sediments appeared alternatively from north to south in the whole LDB, and more coarse-grained sediments occurred in both the shoals and channels in the inner bay. The isolines of the mean grain sizes and the percentages of sand, silt, and clay fractions were east–west oriented, roughly perpendicular to the bathymetric contours (Fig. 5). In the delta front platform, west of the inner Lingding Channel, the coarsest sediment occurred in its eastern part, close to the inner Lingding Channel, where the sand content exceeded 90% and the gravel content was ~4% (Table 1). Individual samples from the inner Lingding Channel were also composed of gravelly sand, with a sand content of 93.6% and a gravel content of 4.4% (Table 1). In the outer LDB, the coarsest sediments occurred in the outer Lingding Channel and Mawan port (Fig. 5). The gravel content reached 15.5% in Mawan port (Table 1). The coarse-grained sediment in the outer Lingding Channel was composed of >90% sand and ~2.6% gravel at a location close to its intersection with the Tonggu Channel (Fig. 5, Table 1). The surficial sediments were also coarser on both sides of the outer Lingding Channel. In the remaining area of the outer LDB, especially the western part, fine-grained sediments prevailed, with a clay content of >30% and a sand content of <10% (Fig. 5), but there was no relationship between the grain size contours and the bathymetric contours.

### Table 1

Grain size and organic carbon characteristics of surficial sediments in LDB. Locations of the geomorphic units are indicated in Fig. 4. For some geomorphic units, some samples were of insufficient size after grain size analysis and were therefore not subjected to organic elemental analysis. In such cases, the number of samples analysed is indicated in brackets.

<table>
<thead>
<tr>
<th>Units</th>
<th>Number of samples</th>
<th>Statistics parameters</th>
<th>Mz (%)</th>
<th>Gravel (%)</th>
<th>Sand (%)</th>
<th>Silt (%)</th>
<th>Clay (%)</th>
<th>δ13C (‰)</th>
<th>TOC (%)</th>
<th>TN (%)</th>
<th>C/N</th>
</tr>
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<tbody>
<tr>
<td>1 17</td>
<td>AVE 6.39 0</td>
<td>19.0 53.1 27.9</td>
<td></td>
<td>–25.54</td>
<td>0.93</td>
<td>0.12</td>
<td>9.4</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>2 6 (2)</td>
<td>AVE 5.55 0.7</td>
<td>23.7 55.4 20.2</td>
<td></td>
<td>–26.75</td>
<td>0.76</td>
<td>0.09</td>
<td>10.2</td>
<td></td>
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<td></td>
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<tr>
<td>3 16 (15)</td>
<td>AVE 6.01 0.2</td>
<td>20.2 56.2 22.9</td>
<td></td>
<td>–23.72</td>
<td>0.65</td>
<td>0.09</td>
<td>8.6</td>
<td></td>
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</tr>
<tr>
<td>4 28</td>
<td>AVE 6.75 0</td>
<td>9.0 63.2 27.8</td>
<td></td>
<td>–23.98</td>
<td>0.75</td>
<td>0.11</td>
<td>8.4</td>
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<tr>
<td>5 3</td>
<td>AVE 6.28 0</td>
<td>19.9 52.9 27.1</td>
<td></td>
<td>–24.00</td>
<td>0.60</td>
<td>0.11</td>
<td>8.4</td>
<td></td>
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</tr>
<tr>
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<td>AVE 5.36 3.9</td>
<td>26.4 47.4 22.3</td>
<td></td>
<td>–24.02</td>
<td>0.74</td>
<td>0.10</td>
<td>8.9</td>
<td></td>
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<tr>
<td>7 10</td>
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<td>19.0 55.0 26.0</td>
<td></td>
<td>–25.32</td>
<td>1.13</td>
<td>0.15</td>
<td>9.1</td>
<td></td>
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</tr>
<tr>
<td>8 8</td>
<td>AVE 6.25 0</td>
<td>18.4 57.2 24.4</td>
<td></td>
<td>–24.76</td>
<td>0.73</td>
<td>0.10</td>
<td>8.7</td>
<td></td>
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</tr>
<tr>
<td>9 27 (25)</td>
<td>AVE 6.28 0.15</td>
<td>20.2 52.8 26.8</td>
<td></td>
<td>–24.24</td>
<td>0.85</td>
<td>0.12</td>
<td>8.4</td>
<td></td>
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<tr>
<td>10 9</td>
<td>AVE 5.78 0</td>
<td>24.4 53.7 21.9</td>
<td></td>
<td>–24.16</td>
<td>0.65</td>
<td>0.09</td>
<td>8.6</td>
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4.2. Organic geochemical properties

The spatial distributions of TOC, the carbon/nitrogen ratio (C/N), and δ13C in the surficial sediments showed some regularity (Fig. 6). The TOC value was high, mostly >0.8%, in the east and west shoals. The highest TOC content (1.68%) occurred in the northeast part of the east shoal (Fig. 6a, Table 1). The TOC content was <0.8% in most samples from the middle part of the inner LDB and in areas of water depth

**Fig. 4.** Distribution of surficial samples collected from LDB in December 2016. Numbers on the map represent geomorphic units: 1, CLF Channel; 2, inner Lingding Channel; 3, outer Lingding Channel; 4, QD waterway; 5, Tonggu Channel; 6, Mawan port; 7, east shoal; 8, middle shoal; 9, west shoal; 10, south shoal. (1)–(4) represent the islands in LDB, i.e., Qi’ao Island, inner Lingding Island, Luhuan Island, Dayu Mountain, respectively.
exceeding 5 m in the outer LDB, except in some individual samples. The lowest TOC occurred in the outer Lingding Channel (Table 1). The spatial distribution of the C/N ratio was complex, with some irregular patches of high and low values (Fig. 6b). In general, the C/N ratios close to the four outlets in the northern part of the inner LDB were high (> 9). The C/N ratios were low in the area north of inner Lingding Island (southern part of the middle shoal), but were significantly higher in the area east of the island and in Mawan port. C/N was usually < 9, but with some isolated patches of high values (9–11) in the outer LDB, and the lowest value occurred in the south-western part.

The δ13C values increased from north to south in LDB, and then decreased again towards the open sea outside LDB (Fig. 6c). It was generally less than −25‰ in the inner LDB, and low values were distributed along the inner Lingding Channel. It was also lower in the area east of the inner Lingding Channel than in the west shoal. The δ13C values were clearly increased in the outer LDB, and were usually higher than −24‰ in areas with water depth greater than 5 m, except in some isolated patches. In areas with water depth less than 5 m, the δ13C values were lower than −24‰ in the north and increased to higher than −24‰ in the south.

The proportion of TOM (Fig. 6d) generally exceeded 90% in the inner LDB, and was even 100% in some parts of the inner Lingding Channel, CLF Channel, and east shoal. Low values of TOM (40%–70%) were mainly distributed along the outer Lingding Channel. It decreased southward and increased both eastward and westward. However, high values of TOM (> 90%) appeared again near the bay mouth and on the shelf in the open sea.

Plots of C/N vs δ13C (Lamb et al., 2006) showed that the organic carbon in the surficial sediments from the CLF Channel and inner Lingding Channel mainly fell within the ranges of freshwater particulate organic carbon (POC), freshwater algae, and the transitional zone to C3 terrestrial plants (Fig. 7a), indicating a predominantly terrestrial source of the organic carbon. The surficial sediments from the east shoal all fell within the ranges of freshwater algae/POC and the transitional zone to C3 plants (Fig. 7b). Some of samples from the west shoal were derived from freshwater algae/POC and C3 plants, and others were from marine algae/POC (Fig. 7b). Most of the samples from the middle shoal fell within the ranges of C3 plants and freshwater algae/POC, but a few were in the range of marine algae/POC (Fig. 7c). The samples from the south shoal were predominantly distributed within the range of marine algae/POC, with individual exceptions attributable to freshwater algae/POC and the transitional zone to C3 plants (Fig. 7c).
The samples from Mawan port and Tonggu Channel also showed a mixture of terrestrial and marine contributions (Fig. 7c). The samples from the outer Lingding Channel and QD waterway fell mainly within the range of marine algae/POC, but a few samples were in the ranges of freshwater algae/POC and the transitional zone to C3 plants (Fig. 7d).

5. Discussion

5.1. Temporal change in grain size distribution and controlling mechanism

Previous studies (Chen, 1995; Xia et al., 2013) have shown that the distribution of surficial sediments in LDB in earlier years (1975, 2003–2004) was mainly controlled by the hydrodynamic conditions of the freshwater discharge and tidal currents. Therefore, the spatial distribution of grain size was characterized by three subareas in LDB and the offshore shelf (Fig. 8a and b). The first subarea comprised the northern and north-western LDB, where the highest sand content occurred because fluvial processes dominated. The second sub-area comprised the north-eastern, central, and southern parts of the bay, where fine-grained sediments occurred and tides dominated. This sub-area corresponded to the location of turbidity maximum, where resuspension and vertical circulation prevailed (Liu et al., 2016). The third subarea comprised the shelf outside LDB, where the relict sand from the late Pleistocene was directly exposed on the seabed (Fig. 8a and b). However, the present study demonstrates that there was no longer such a regular pattern in 2016 (Figs. 5 and 8c). We suggest that strong human perturbation is responsible for these changes, as discussed below.

Let us take the percentage of sand as an example. In 1975, the sand content was high in the delta front platform at the four outlets in the northern LDB, and low in the northeast and southern inner LDB and the outer LDB (Fig. 8a). In 2003–2004, the sand content in the delta front platform had clearly decreased (Fig. 8b). Furthermore, the area of high sand content had shrunk and high sand contents were only present in areas off the Humen and Jiaomen outlets. Previous studies have suggested that such fining of the surficial sediments in the delta front platform was mainly caused by the weakened runoff that resulted from sand mining in the river channels on the delta plain (Luo et al., 2007; Xia et al., 2013), because this would disperse less of the sand fraction to the river mouths. We also speculate that the greater sand fraction in the suspended particulates was trapped in the reservoirs in the drainage basin, leading to a fining of the suspended sediments supplied to the

Fig. 6. Spatial distribution of organic carbon characteristics in the surficial sediments in LDB. The dashed line in each inset represents the boundary of the inner and outer LDB.
river mouth. The suspended sediments continued to be trapped in the reservoirs after 2004, as inferred from the further decline in the sediment load, whereas the freshwater discharge was much the same as in previous decades (Fig. 2; Wu et al., 2016). Therefore, it is difficult to attribute the coarse-grained surficial sediment in the northwest LDB in 2016 to the strengthening of fluvial processes. In particular, the surficial sediments were coarser at locations close to the Lingding Channel than at locations close to the outlets (Fig. 8c), which is opposite the change in the fluvial processes. A simulation by Heise et al. (2010) indicated that tidal currents do not cause erosion in this area. Therefore, we suggest that the coarsening of the surficial sediments in the northwest part of the inner LDB was caused by human activities.

In the spatial distribution pattern of grain size in 2016, the coarse-grained sediments predominantly occurred in the channels and ports, and the shoals close to them (Figs. 5 and 8c). There were gravel-bearing sediments in the inner and outer Lingding Channel, Mawan port, and west shoal (Table 1). By contrast, gravelly sand was only reported in the channel close to the Humen outlet, whereas mud dominated in the inner and outer Lingding Channel in 2003–2004 (Xia et al., 2013). We suggest that the dredging of the channels exposed the coarse-grained sediments deposited during the early Holocene or late Pleistocene (Xia et al., 2013; Zhang et al., 2010) on the channel bed. The dumping of dredged material onto the adjacent shoals (Fig. 3c; Jiang et al., 2009; GBOF, 2014) redistributed the ‘old’ sediments. These ‘old’ sediments were then sorted during the resuspension induced by tidal currents, which left the coarse-grained particles on the shoals. The irregular patches of sand-dominated surficial sediments in the zone of turbidity maximum in the dry season of 2016 (Fig. 8c) indicated either less deposition of fine-grained sediments there or very intensive dredging and sand mining, which allowed no preservation of the newly deposited sediments. Additional on-site research is required to determine the sediment dynamic processes in this highly anthropogenically perturbed area.

We also suggest that the fining of the surficial sediments on the shelf in the open sea in 2016 (Fig. 8) implies that dredged sediments were dumped in the open sea. A previous study reported a very low suspended sediment concentration (< 0.08 kg/m³) at the mouth of LDB (Liu et al., 2016), which suggests that there has been little seaward
dispersal of fine-grained sediments from the Pearl River mouth. By contrast, there is a dumping area on the shelf for sediments dredged from LDB (Fig. 3c; Jiang et al., 2009; GBOF, 2014). The dumped sediments have possibly been redistributed by the action of tidal currents and other sediment dynamic processes.

5.2. Temporal change in organic geochemical properties and controlling mechanism

An organic geochemical analysis of the surficial sediments collected in 2016 confirmed the strong human perturbation of the distribution of surficial sediments in LDB and on the adjacent continental shelf. When we compared the $\delta^{13}C$ and TOM values from this study with those of 2005–2006 (Yu et al., 2010, Fig. 9), two major changes were evident. First, the $\delta^{13}C$ values decreased and the proportion of TOM increased significantly in 2016. Second, the regular distribution patterns of $\delta^{13}C$ and TOM observed in 2005–2006 no longer existed in 2016.

The overall increase in the contribution of terrestrial organic carbon to the surficial sediments in 2016 contradicts the significant reduction in the terrestrial sediment load from the Pearl River (Fig. 2) and the eutrophication trend in LDB and on the adjacent shelf over recent decades. Previous studies have reported that eutrophication has strengthened in the past 10 years with the increased input of sewage, industrial wastewater, and agricultural fertilizer (Huang et al., 2003; Cui et al., 2018). Eutrophication increased the productivity of marine algae, causing hypoxia in the outer LDB and the open sea outside the bay (Cui et al., 2018). Such ecological environmental changes would have increased the contribution of marine-sourced organic carbon, increasing the $\delta^{13}C$ values and reducing TOM (Müller and Voss, 1999; Lamb et al., 2007). We also suggest that the spatial distributions of $\delta^{13}C$ and TOM in natural deposits should be similar to the patterns in 2005–2006, reported by Yu et al. (2010; Fig. 9a and b), which were consistent with the hydro- and sediment-dynamic processes in LDB and on the adjacent shelf. In the report of Yu et al. (2010), the shoal sediments off the four outlets in the north-western inner LDB were dominated by terrestrial organic carbon, resulting from the prevalence of freshwater discharges (Huang et al., 1982), whereas the contribution of terrestrial organic carbon was lower in the offshore area of the Humen outlet, where tides dominated (Huang et al., 1982). In the outer LDB and on the shelf in the open sea, marine-sourced organic carbon prevailed, which was consistent with the prevalence of saltwater and the stratification of the water column in this area (Liu et al., 2016; Cui et al., 2018). A previous study reported landward net sediment transport from the shelf to the outer LDB, based on an investigation of the grain size of the surficial sediments in 2007, which was consistent with a simulation of residual bottom currents (Zhang et al., 2013). We suggest that such a net sediment transport pattern would also result in a predominance of marine-sourced organic carbon in the outer LDB and on the adjacent shelf, as reported by Yu et al. (2010).

However, the results of our study showed no such regular pattern of
organic carbon in the surficial sediments in 2016. We speculate that the overall increase in TOM in the surficial sediments in 2016 is the result of human activities, including sand mining, dredging, and dumping. These human activities exposed and redistributed early Holocene or late Pleistocene sediments, which were formed in a near-shore or terrestrial environment and were dominated by terrestrial organic carbon because the sea level at that time was low (Zong, 2004, 2006). In particular, much higher TOM values in the outer LDB and adjacent shelf in 2016 (Fig. 9d) indicate that there was significant dumping there, as well as on the shoals along the Tonggu and outer Lingding Channels (Fig. 3c). This inference is consistent with the rapid increase in the subaqueous topography, reported by Wu et al. (2018), and the finer surficial sediments on the shelf in the open sea, reported in the present study (Fig. 8). We also speculate that dumped sediments were redistributed by tidal currents or other hydro- and sediment-dynamic processes because the area dominated by terrestrial organic carbon is much larger than the dumping area in the outer LDB and adjacent shelf (Figs. 3c and 9d).

5.3. Implications of the highly human-perturbed surficial sediments at the Pearl River mouth

Based on the discussion above, we suggest that the surficial sediments in LDB and on the adjacent shelf have been completely altered by dredging, dumping, and sand mining, particularly in the inner LDB and the eastern part of the outer LDB. In the western part of the outer LDB, the sediment particle composition was similar to that in earlier periods, but the organic elemental composition has been completely altered, reflected in the obvious increase in TOM. We suggest that such changes in the composition of the surficial sediments will have unknown ecological consequences, which must be considered in coastal management strategies. First, the submarine groundwater discharged through the confined and unconfined aquifers in the subaqueous PRD reportedly makes an important contribution to the eutrophication and hypoxia there (Liu et al., 2018; Luo et al., 2018). Dredging and sand mining have increased the exposure of the confined aquifers on the seabed, evidence of which is the new occurrence in 2016 of gravelly sands in the shipping channels and sand mining pits (Fig. 5). The chemical flux to the LDB from those newly exposed confined aquifers will increase, exacerbating eutrophication. However, the dumping of fine-grained sediments on the shelf could cover the outcrops of aquifer on the seabed and change the groundwater cycle. Therefore, it is necessary to investigate the new balance between the submarine groundwater discharge and the ecological environment in LDB and on the adjacent shelf. The change from marine-sourced to terrestrial-dominated organic carbon in the surficial sediments of LDB and the adjacent shelf could also influence food cycles and induce chain reactions in the ecology of this area. Therefore, we suggest that more research is required into the ecological responses to the changes in the composition of surficial sediments at the Pearl River mouth, to improve coastal management. We also believe that understanding the responses of natural processes to the intensive human activities in LDB will improve the coastal management in other river mouths around the world.

6. Conclusions

In this study, we analysed the spatial distributions of the grain size and organic geochemical characteristics of surficial sediments collected in LDB, in the PRD, in December 2016. By comparing them with values and distribution patterns reported in previous studies, we found that the grain size and organic geochemical characteristics of the surficial sediments have changed significantly, as described below.

1) Irregular patches of coarse-grained sediments mainly occurred in the shipping channels and nearby shoals in both the inner and outer LDB in 2016.

2) A dramatic reduction in the δ13C values for organic carbon and an increase in the proportion of TOM were detected in LDB and on the adjacent shelf in the open sea in 2016.

These features (grain size and organic carbon composition) of the surficial sediments in 2016 were not consistent with the spatial variations in the hydrodynamic and sediment dynamic processes in LDB, and were inconsistent with the decline in the riverine sediment supply and the eutrophication in the outer LDB and adjacent shelf in past decades. Human activities have overwhelmed the natural processes at the present-day Pearl River mouth. The coarser sediments and lighter δ13C indicate the re-exposure of near-shore or terrestrial sediments formed in the early Holocene or late Pleistocene on the present seabed, which has been caused by sand mining, dredging, and dumping in LDB in recent years. Unknown ecological responses will be induced by the exposure of ‘old’ coarse-grained sediments from the confined aquifers by dredging and sand mining and by the overall increase in TOM in LDB and on the adjacent shelf.

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