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Article

Sedimentological Analysis of Regional Differentiation and Sediment Provenance in the Lu'erhuan River Sea Area of Qinzhou Bay, Guangxi Province

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Abstract: Globally, coastal regions are vital areas of human activity and, as such, are centers of population growth and urban and economic development. Long-term human development has had a major impact on the ecological environment of coastal zones. Therefore, exploring the distribution and provenance of marine sediment types in coastal areas heavily influenced by human activities can provide scientific evidence and references for the current and future ecological management of these sensitive environments. For this reason, we conducted an analysis of the sediment grain size, endmembers, and organic matter content and geochemical elements in the Lu'erhuan River-Malan Island-Sandun Island area in the eastern part of Qinzhou Bay, a region heavily influenced by human activities. The sediment grain size clearly differs throughout the study site and the material provenances and hydrodynamic conditions also vary, likely due to the local environmental conditions and the significant impact that human activities have had on the area. The finest-grained sediment is imported from either inland or coastal areas via rivers and weak tidal currents, the next finest component comes from coastal areas through weak tidal currents, and the moderately coarse component mainly originates from nearby beaches. The two coarsest-grained sediment components are influenced by the combination of human activities, tidal currents and waves and enter the water via erosion. The organic matter provenance resembles that of the sediment components, exhibiting varied characteristics. Due to the combination of natural and human activities in the bay, the organic matter in the upper reaches of the Lu'erhuan River originates from the river and coastal paddy fields, with obvious terrigenous characteristics; the organic matter in northern Malan Island mainly comes from external sources related to oyster farming, while the organic matter in eastern Sandun Island is mainly produced endogenously by marine plankton. Al, Ti, Fe, Mg, K, Ga and other elements indicate that terrestrial sediments are significantly disturbed by human activities. However, Mn reflects the marine distribution of terrestrial sediments from the Lu'erhuan River to Jishuimen. Ca and Sr, which are indicators of marine sediments, are distributed in the eastern offshore area of Sandun Island, which is connected to open waters. Due to the influence of human activities, As and Cd are highly enriched in the study area, while Cu is less affected by human activities.

Keywords: Qinzhou Bay; sediment provenance; human activities; endmember analysis

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1. Introduction

Throughout the world, coastal regions are areas with the largest concentrations of human, urban and economic activities. Over years, high-intensity human activities have had a major impact on the ecological environment of coastal zones, making them among the world's most vulnerable ecosystems. A series of increasingly prominent ecological and environmental challenges, such as severely reduced coastal wetlands, reduced biodiversity, destroyed mangroves, environmental pollution, seawater eutrophication and coastal erosion, have all become topics of great concern. Under the guidance of the strategic decision of ecological civilization construction [1], coastal ecological restoration projects have been gradually implemented.

Among coastal zones, gulf regions most frequently bear a burden imposed by human activities; thus, they are prime areas for ecological restoration. Qinzhou Bay, located in the northern part of the South China Sea, is a major area for oyster breeding in China due to its superior geographical location and abundant mangrove natural resources. It is known in China as the "hometown of oysters" [2] and promotes the social and economic development of the region. Over the years, the construction of ports, wharves, highways and bridges has had a major impact on shoals and beaches. This development has caused damage to the ecological environment and has had serious impacts on aquaculture [3]. In the eastern part of Qinzhou Bay, the abundance of human activities, such as the construction of coastal highways, the Sandun Marine Highway and Sandun Port, has divided the marine area around Lu'erhuan River-Malan Island-Sandun Island from Qinzhou Bay. This environment change has had a significant impact on the coastal hydrological characteristics and surface sediments, which in turn pose serious problems for ecological safety and aquaculture. Therefore, this study, which considers the larger regional context of intense human activities, and analyzes the distribution characteristics and provenance of sediments in this bay, can provide scientific evidence and a reference for current and future ecological construction projects in coastal areas.

Grain size analysis, endmember analysis and geochemical element analysis of bay sediments are effective ways to explore sediment distribution characteristics and material provenance [3–17]. In addition, elemental analysis under the local impact of human activities, grainsize and end-element analysis can effectively reflect the sedimentary dynamics, material source and sedimentary environment. Sediment properties such as organic matter content, heavy metal content and magnetism can also reflect the source of sediment [18]. Organic matter in the Gulf of Tonkin's sediments is transported mainly by rivers to the bays, and the distribution of terrestrial organic carbon is limited by coastal and adjacent areas. The seasonal variation in phosphorus in Zhanjiang Bay coastal waters is related to the seasonal emission from terrestrial sources. Analysis of heavy metals in the sediments of the Gulf of Tonkin Bay shows that the concentrations of Cd, Pb and Cu in the sediments of Qinzhou Bay are significantly higher than their background values [19]. The proportion of man-made sources is close to 70%, and the contribution of terrestrial sources to heavy metals is significant [20,21]. Moreover, the sediments in Qinzhou Bay contain high levels of micro-plastics, especially near shellfish farms. The environmental magnetic properties of the sediments in the Gulf of Tonkin suggest that the sediments in the eastern part of China were imported from the Qinzhou Bay, the Honghe River and the rivers of Hainan Island. At present, there has been little systematic discussion on the characteristics of coastal sediments and terrestrial source analysis, so it is of practical significance to study the characteristics of the sediments in the Qinzhou Bay area.

This study aims to provide reliable suggestion sand references for coastal development and ecological environment management by exploring the following: (1) sediment types and their distribution characteristics; (2) endmember analysis and material provenance of sediments impacted by intense human activities; (3) distribution characteristics and the provenance of organic matter impacted by intense human activities. This paper achieves the goal on the basis of these analytical methods and in combination with the characteristics of high-intensity human activities in the marine area of Lu'erhuan River-Malan Island-Sandun Island.

2. The Study Area

Qinzhou Bay (21°33′20′′–21°54′30′′ N, 108°28′20′′–108°45′30′′ E) is located on the northern coast of the Beibu Gulf in the South China Sea (Figure 1). The area of the bay in 2020 was 380 km^2, with beach composing approximately 200 km^2 of the total. Qinzhou Bay is the largest bay in the coastal zone of Guangxi province. However, the bay area has shrunk to 320 km^2, and the areal extent of the tidal flats has correspondingly decreased in recent years. The Lu'erhuan River-Malan Island-Sandun Island marine area along the east side of Qinzhou Bay (21°35′18″–21°44′40″ N, 108°40′53″–108°44′28″ E) is narrow and runs from north to south. The Lu'erhuan River and Jinwo Reservoir are connected on the north side, and the river flows southward from Malan Island towards Sandun Island passing through Jishui Mountain. In recent years, the coastal environment in this area has been strongly disturbed. In addition, the hydrodynamic conditions, sediment transport and redistribution characteristics of the bay have undergone substantial changes, which have led to difficulties in the comprehensive regulation and ecological restoration of the Lu'erhuan River-Malan Island marine area.

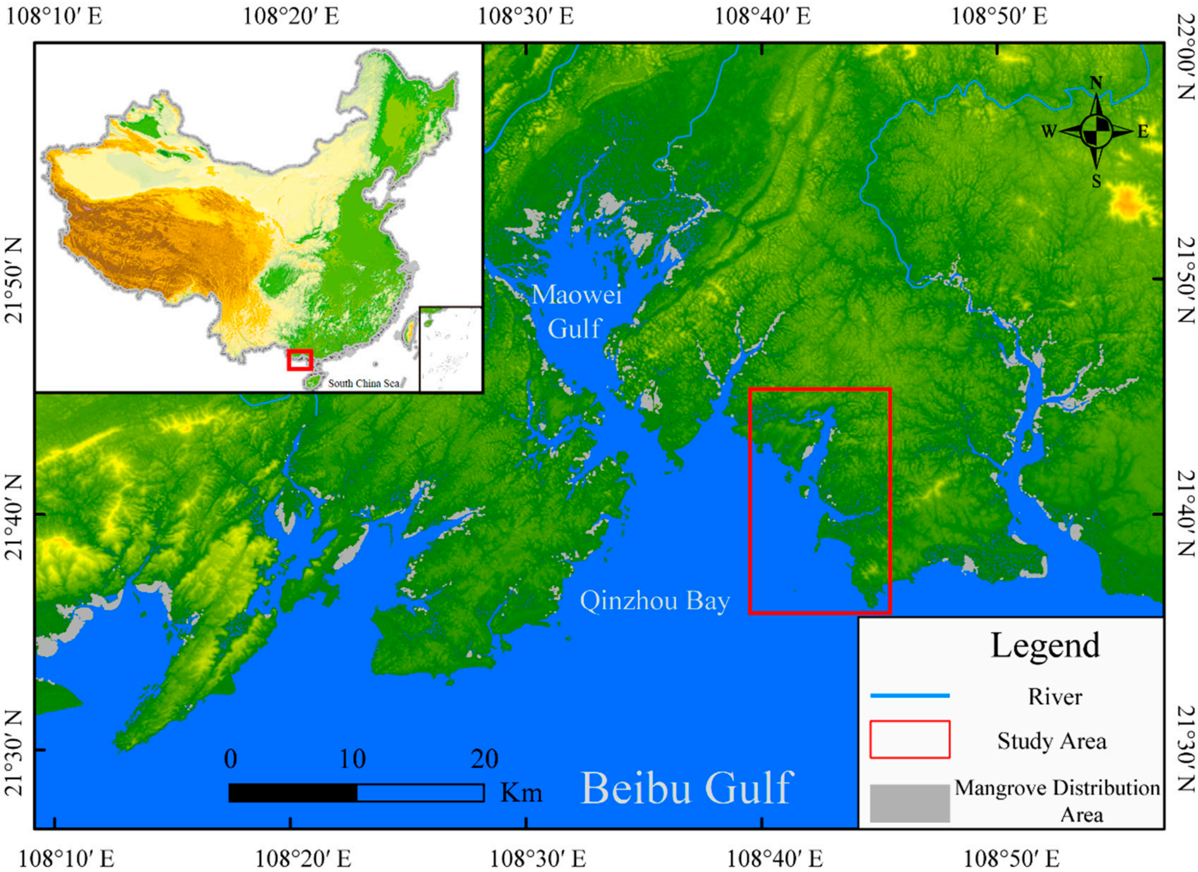

Figure 1. Geographical location and mangrove distribution area in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province. The red rectangle is the study area and the grey shaded areas are mangrove growth areas.

3. Material and Methods

3.1. Sampling of Surface Sediments

Considering the natural environment and characteristics of human activity in the area around Lu'erhuan River-Malan Island-Sandun Island, the study area was divided into five partitions from north to south, called areas A, B, C, D and E (Figure 2, Table 1). Area A, which mostly consists of the Lu'erhuan River, is more clearly affected by the Jinwo Reservoir located upstream and terrestrial runoff from both banks, and the sediments here clearly have a terrigenous provenance. The north and east sides of area B are significantly affected by the Lu'erhuan and Dazao Rivers, while the south side is connected to area D and more significantly affected by ocean tides. Area B is surrounded by the other four areas, and separated from the other four areas by the east-west coastal highway, the north-south coastal highway, the islands and reefs along Jishui Mountain and the Sandun Marine Highway. Due to the impact of coastal roads, bridges, the Sandun Marine Highway and Sandun Island reclamation projects, the hydrological and sedimentary environment of each area differs. Area C is mainly affected by the Dazao River flowing westward, which is similar to area A. The sediments indicate terrigenous origins. Areas D and E are connected with Qinzhou Bay and are mainly affected by ocean tides.

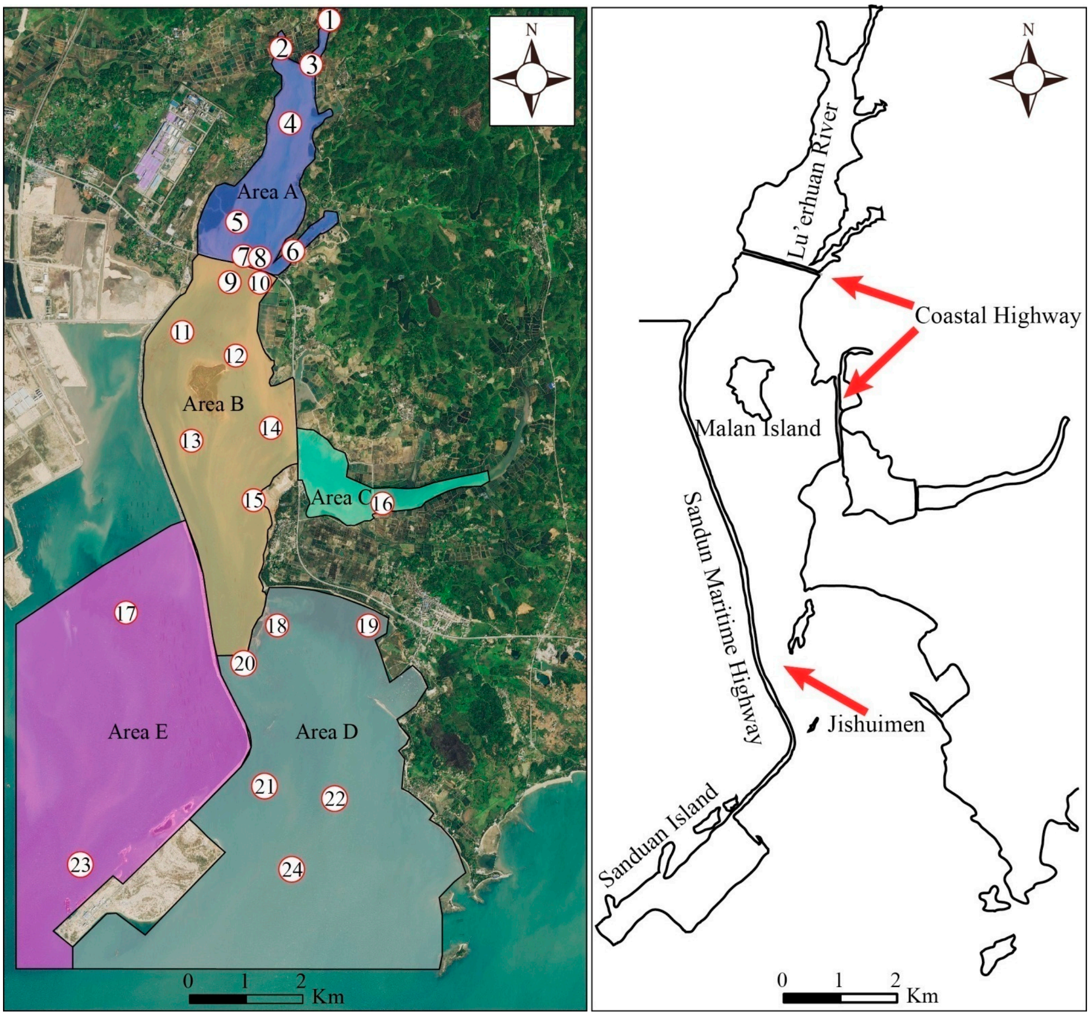

Figure 2. Map of sampling sites in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province. The remote sensing image (**the left**) and diagram (**the right**) of the study area. The colored shaded areas in the left panel are the five study areas A, B, C, D, E, and the circles in red and marked with serial numbers represent the sampling points. The right panel shows a sketch of the outline of study area.

In August 2020, a total of 24 surface sediment samples were obtained by a clam grab sampler, with eight, seven, one, six and two samples obtained from areas A, B, C, D and E, respectively. Area C is dominated by the influence of the Dazao River, which is a very small river and is connected to Area B only by a small bridge, so the depositional environment here is more stable, and one sample can represent its sediment characteristics. Area E, which is connected to Qinzhou Bay in the same way as Area D, was not more intensively sampled because the depositional environments of both these areas are subject to significant anthropogenic disturbance in addition to tidal influence, and their roles are similar in this paper. The samples were sealed in plastic self-sealing bags and labeled according to their geographical location, number and other information and then returned to the laboratory for subsequent analysis.

Table 1. Information on sampling location and average grain size of surface sediments in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

Sample Number	Latitude (N)	Longitude (E)	Average Grain Size (μm)	Sample Number	Latitude (N)	Longitude (E)	Average Grain Size (μm)
1	21°44′26″	108°43′20″	7.58	13	21°40′26″	108°42′43″	5.78
2	21°44′2″	108°42′51″	7.56	14	21°39′38″	108°42′33″	0.57
3	21°43′20″	108°42′57″	7.36	15	21°39′38″	108°43′53″	6.84
4	21°42′22″	108°42′23″	7.20	16	21°39′33″	108°41′1″	6.84
5	21°42′4″	108°42′56″	7.49	17	21°38′31″	108°42′48″	5.33
6	21°42′0″	108°42′25″	6.11	18	21°38′30″	108°43′42″	3.63
7	21°42′1″	108°42′38″	1.73	19	21°38′8″	108°42′23″	5.37
8	21°41′48″	108°42′13″	6.67	20	21°36′54″	108°42′37″	0.15
9	21°41′46″	108°42′36″	7.10	21	21°36′50″	108°43′23″	7.10
10	21°41′14″	108°41′47″	7.13	22	21°36′6″	108°40′42″	7.35
11	21°41′4″	108°42′21″	6.46	23	21°36′1″	108°42′56″	6.90
12	21°40′12″	108°41′51″	0.56	24	21°36′1″	108°42′58″	6.84

3.2. Sedimentological and Geochemical Analyses

Grain size, as one of the main physical characteristics of sediment, is controlled by factors such as the transport energy and transport mode and can offer insight into the nature of the sedimentary environment [22]. In this study, according to the characteristics of the regional sedimentary environment, grain size analysis was conducted with the following procedure: First, an appropriate amount of each sediment sample was selected and placed into a centrifuge tube, and the samples were numbered appropriately. Next, an appropriate amount of 10% HCl was added to the centrifuge tubes to remove the calcium carbonate sediment in the sample. Distilled water was added to wash and neutralize the samples after the reactions had completed. Organic matter was removed from the samples with an appropriate amount of 30% H_2O_2 solution. After washing again with distilled water to neutralize the samples, 10 mL of 0.05 mol/L $(NaPO_3)_6$ solution was added as a deflocculating agent, and the mixture was sonicated in an ultrasonic cleaner for 15 min. Finally, a laser particle size analyzer (Microtrac S3500;Microtrac Inc., in Montgomeryville and York, PA, USA) was used to analyze the grain size of the processed samples. In order to reduce the error caused by uneven sampling, each sample was measured three times. When no substantial differences were recorded, results were averaged.

On the basis of grain size analysis, endmember analysis was also carried out to better identify the sediment provenance. The analysis was performed using AnalySize1.2.1 software [23]. Organic matter refers to a variety of organic compound forms within sediments, and its content is an important indicator in environmental research [24]. Loss-on-ignition 550 °C ($LOI_{550\ °C}$) is an effective method for measuring the organic content of sediment [25]. $LOI_{550\ °C}$ provides the percentage of a sample lost (organic matter) during high temperature combustion at 550 °C. The specific experimental procedure is as follows: Sediment samples of approximately 10–20 g were ground into powder, and then dried in a drying oven at 105 °C. The dried samples were transferred into the dried ceramic crucibles with a known weight and dried again at 105 °C. Then, the dried crucibles and samples were weighed to obtain the initial sample weights. Next, the ceramic crucibles were placed in a muffle furnace and burned at 550 °C for 4–6 h. Finally, when the temperature in the furnace dropped below 200 °C, the ceramic crucibles were removed for final weighing, and the organic matter content was calculated (the drying oven was used to store the samples throughout the experiment).

Elemental geochemistry is an important component of marine sediment research. Changes in the sedimentary composition and distribution can reflect the provenance of sediment [26,27]. In this study, some samples were subjected to elemental analysis. First, selected sediment samples were dried at 105 °C, and then the dried samples were ground into a powder. Then, 0.1 g of the powdered sample was weighed and placed in a sealed clean thromboethylene digestion tank. Next, 5 mL of HNO_3 and 1 mL of HF were added, and the solution was placed into a microwave digester. After the digestion was completed and the sample had cooled, it was moved to the acid evaporator to remove the excess acid, and 1 mL of perchloric acid was added. Then, the processed sample was held at a constant volume and placed in a 50 mL volumetric flask for testing. Finally, an appropriate amount of the liquid to be measured was added to the inductively coupled plasma optical emission spectrometer (IPC-OS: 5900 ICP-OES Instrument; Agilent Technologies, CA, USA) and the carbon and nitrogen analyzers to test the contents of metal elements (K, Ca, Mg, Al, Fe, Co, Ni, Cu, Zn, Ti, Mn, Cr, Ga, Cd, Sr, Sn, Ba, Pb) and nonmetal elements (P, As, N).

4. Results

4.1. Surface Sediment Types

Because the sea area around Lu'erhuan River-Malan Island-Sandun Island is affected by intense human activities, the grain sizes of the sediments differ greatly. The grain size analysis results of the 24 surface sediment samples are presented here. According to the Folk sediment classification method [28] (Figure 3), the surface sediments in the study area were divided into four types. The grain size categories are, from fine to coarse, mud (I), silt (II), sandy silt (III) and sand (IV). The finest mud was mainly from five samples from the northernmost Lu'erhuan River (area A). In addition to area A, silt was also present in the other four areas. Sandy silt was mainly present in areas B and D along the route from Malan Island to Sandun Island. One sample of sandy silt was obtained from area A located near the water outlet of the bridge in the southernmost part of the area. This location is extremely vulnerable to the influence of area B during high tide. The sediment with the coarsest grain size was mainly distributed in areas B and D, with two samples each. One sample in area A and one in area E were identified as sand, and were obtained near the coastal highway and Sandun artificial island, respectively.

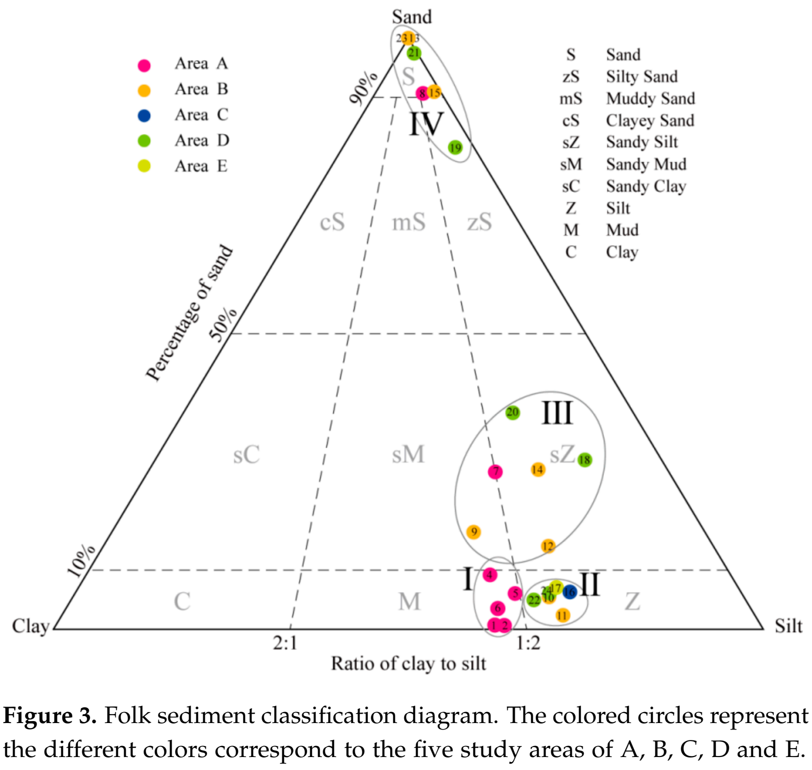

Figure 3. Folk sediment classification diagram. The colored circles represent different samples, and the different colors correspond to the five study areas of A, B, C, D and E. I, II, III and IV are the sediment types of the samples, representing mud, silt, sandy silt and sand, respectively.

The grain size frequency curves and average grain sizes of the four types of sediments in the study area, mud (I), silt (II), sandy silt (III) and sand (IV), clearly differ (Figure 4). Type I and II show typical unimodal morphology with average grain sizes of 5.99 μm and 7.89 μm, respectively. Moreover, these sediments are not widely distributed, indicating that the dynamic sediment environment is local. The sediment grain size frequency curve of type III is bimodal, and the frequency of the two peaks is close to 5%, indicating that the sediment dynamics of type III are distinctly different from those of types I and II and more complex. The average grain size is between 9.94 and 22.74 μm, with a mean value of 16.40 μm, which is relatively broad (Figure 3). The sediment grain size frequency curve of type IV has two or more peaks, all of which exceed 100 μm or even 1000 μm. The proportion is close to 40%, and the average grain size of sediments is scattered, ranging from 86.46 to 1044.34 μm, with an average of 628.34 μm.

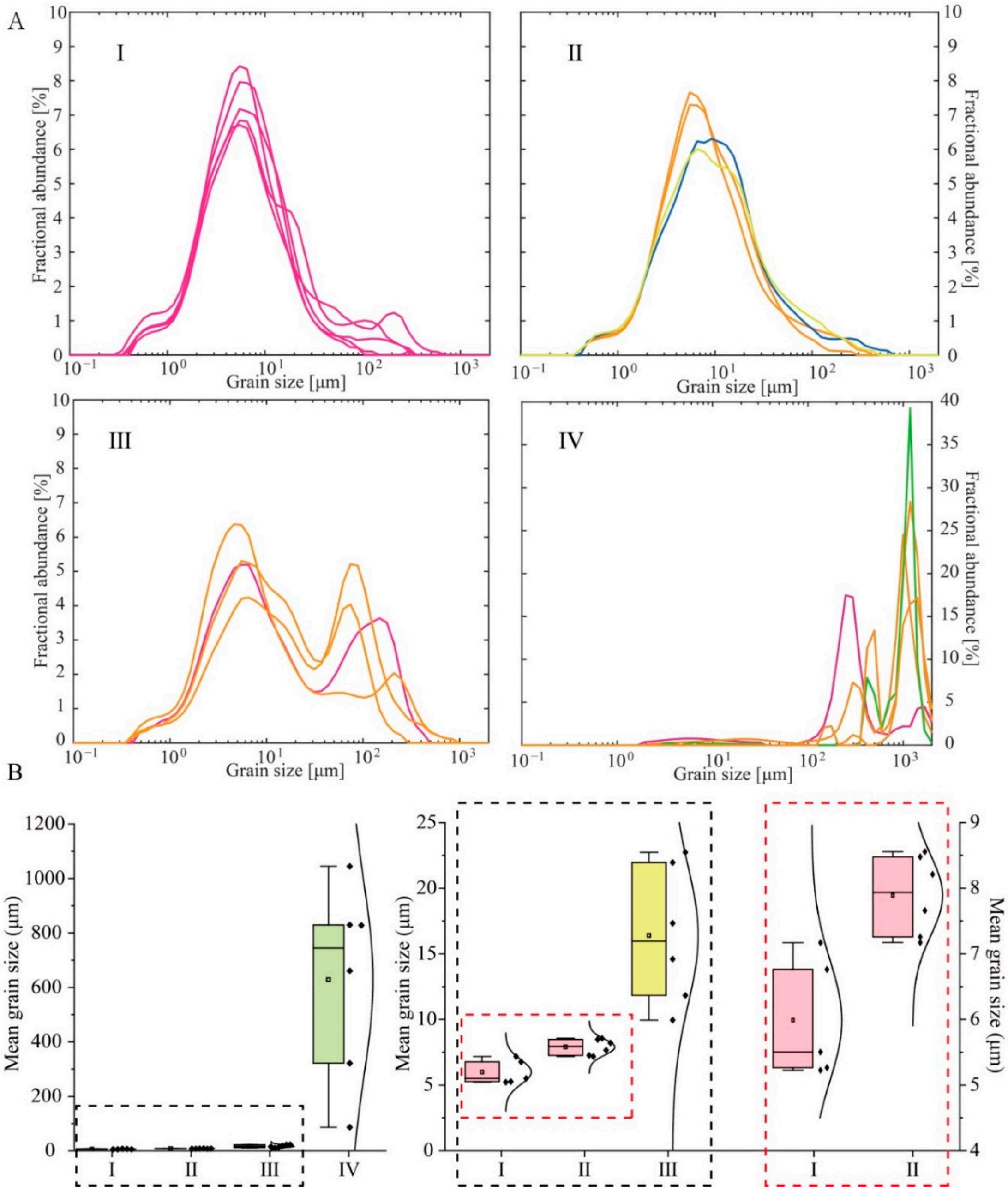

Figure 4. Sediment grain size frequency curve (**A**) and average grain size distribution (**B**). I, II, III and IV in the figure correspond to mud, silt, sandy silt and sand sediment types in the Folk sediment classification diagram, respectively. The curves in the four rectangles in zone A are grain size distribution curves of the samples, and the color of the curves corresponds to the color of the partitions in the Folk sediment classification diagram. The black and red dashed rectangles in area B are the enlarged portions of the box-plot.

4.2. Surface Sediment Grain Size Distribution

The spatial distributions of the sand (63–2000 μm), silt (4–63 μm) and clay (<4 μm) contents in the sediment samples in the study area were analyzed (Figure 5). The average grain size of the sample exhibits positive correlation with the percentage content of sand and a negative correlation with the contents of silt and clay, showing either an exponential increase or decrease. The average grain size of the entire study area is fine, with several peaks on the north side of the coastal highway at the junction of areas A and B, the south side of Malan Island in area B, and the east and west sides of Sandun Island. Due to the significant positive correlation with the average grain size, the percent sand distribution is very similar to the average grain size distribution. However, an area of high percent sand values appears near the eastern coast of the study area, and the six high value areas are also consistent with sampling point IV shown in Figure 3. In contrast, the distributions of fine sand and clay are relatively similar, and are mainly distributed along the route from the Lu'erhuan River to Malan Island in the north of the study area, and along the east and west offshore areas located far from Sandun Island and the SandunMarine Highway.

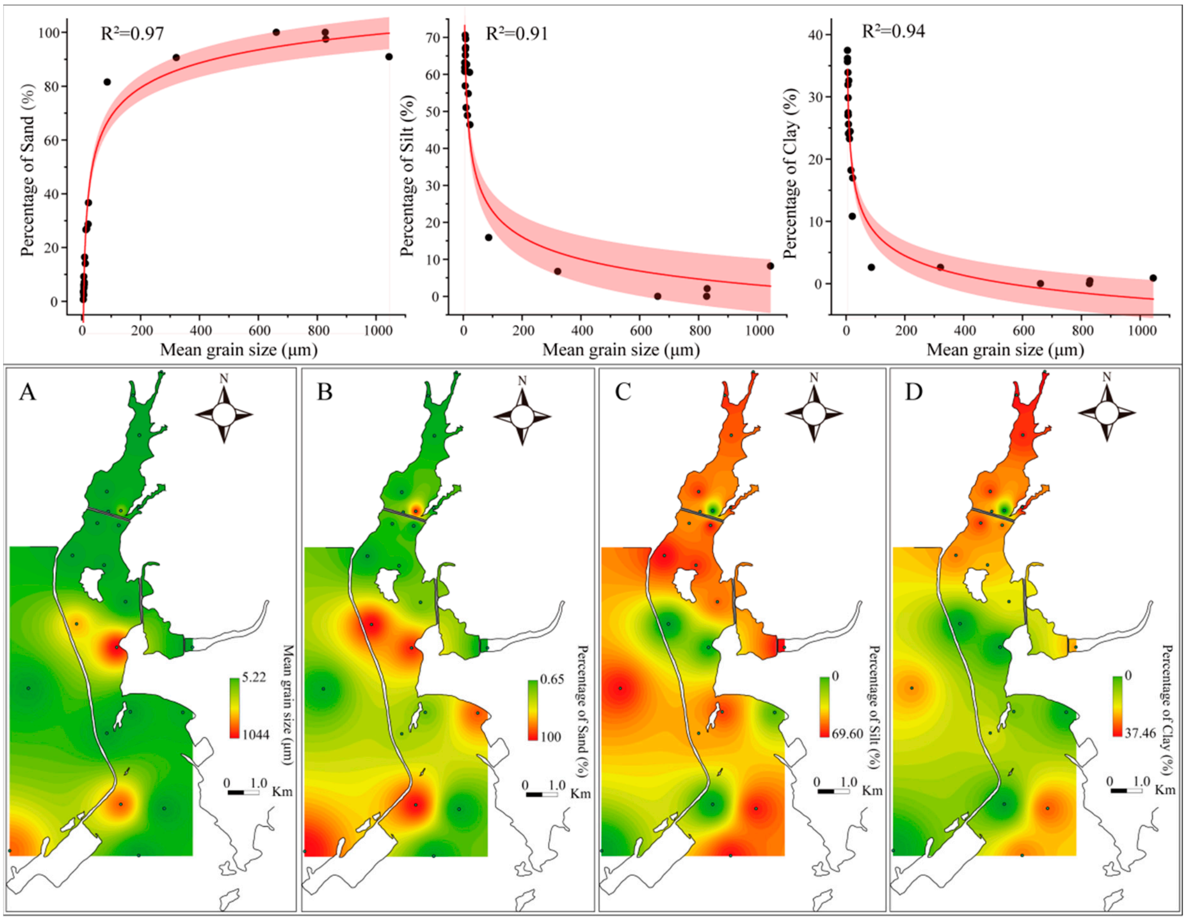

Figure 5. The correlation between mean grain sizes, sand and spatial distribution of silt and sand in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province. The (**A**–**D**) in the Figure 5 respectively shows the content distribution of mean grain sizes and percentage of sand, silt and clay.

The grain size analysis and sediment type distribution of surface sediments in the study area suggest that the fine-grained sediments are mainly mud (I) and silt (II), with average grain sizes of only 5.99 and 7.89 μm, respectively. They are mainly distributed along the route from the Lu'erhuan River to Malan Island (north of areas A, C and B). In addition, the barrier between the Sandun Marine Highway and the coastal highway makes this a relatively enclosed area. For these reasons, the area is less affected by tidal actions, and contains many fine-grained materials. Moreover, the grain size distribution of sediments is unimodal, indicating stable hydrodynamic conditions and a weak sedimentary environment. The surface sediments in this area are assumed to originate mainly from terrestrial runoff, being transported by rivers or surface water, suspended in water bodies in estuaries and accumulated in a relatively stable hydrodynamic environment. Fine-grained clay and silt accumulation in the coastal area are common throughout the Beibu Gulf [29–32], which differs significantly from coastal areas subjected to wave-induced scouring. In addition, east of Sandun Island (area D), the fine-grained sediment component content is also high, yet the sediment grain size distribution shows multiple peaks, indicating the presence of sediment from multiple sources and varied dynamic conditions. One possible reason for this is that this location is connected to the open sea, and is thus affected by multiple tidal currents and waves [33]. The coarse-grained sediments are mainly sand, with an average grain size of 628.34 μm. They are mainly distributed on the south side of Malan Island and near SandunIsland. This sampling point is connected to the open sea. Affected by tidal currents and breaking waves, coastal erosion is severe at this location. In addition, the large volume of debris produced by intense human activities, such as land reclamation, marine highway construction and oyster breeding, has caused the sediment distribution here to take on a chaotic multimodal shape, as shown in Figure 4.

Overall, the offshore area around Lu'erhuanRiver-Malan Island-Sandun Island is subject to intense human activities, and the grain sizes of the sediments in this location are obviously varied. Taking the vicinity of Malan Island as the boundary, the sediments on the north side have a singular provenance and are subjected to stable hydrodynamic conditions, and the sediment grain size is relatively small. Finally, the sediment on the south side is complex, and the average grain size is relatively large due to the combined impacts of human activities, tidal currents and waves.

4.3. The Characteristics of Endmember Analysis of Sediments

To more accurately explore the sedimentary environment and sediment provenances in the Lu'erhuanRiver-Malan Island-Sandun Island area, endmember analysis of sediment grain size data was also conducted. Regarding the selection of the endmember analysis model, after comparing the analysis results of nonparametric and parametric models, we ultimately selected the fitting scheme of the general Weibull distribution parameter for endmember analysis [34]. The grain size endmember fitting results (Figure 6) show that the endmember numbers 2, 3, 4, 5 and 6 correspond to R^2 values (linear correlation: >0.8 indicates that the fitting requirements are basically met) of 0.794, 0.904, 0.923, 0.957 and 0.957, respectively, and angular deviation values 24.0, 14.3, 13.0, 9.3 and 9.2, respectively. The R^2 and angular deviation values tended to be stable as the number of endmembers increased. Through comparison, the best fitting results were obtained in the case of five endmembers (Table 2). Each endmember presents a single peak, and the correlation is 0.013, indicating that the separation effect between endmembers is excellent. The composition data of each endmember (Table 3) show that EM1 (1.69–13.37 μm) and EM2 (5.99–74 μm) are primarily composed of clay and silt, while EM3 (55.9–132.24 μm), EM4 (144.66–336.53 μm) and EM5 (705.20–1335.68 μm) are mainly composed of sand (subdivided into very fine sand (63–125 μm), fine sand (125–250 μm), medium sand (250–500 μm), coarse sand (500–1000 μm) and very coarse sand (1000–2000 μm)).

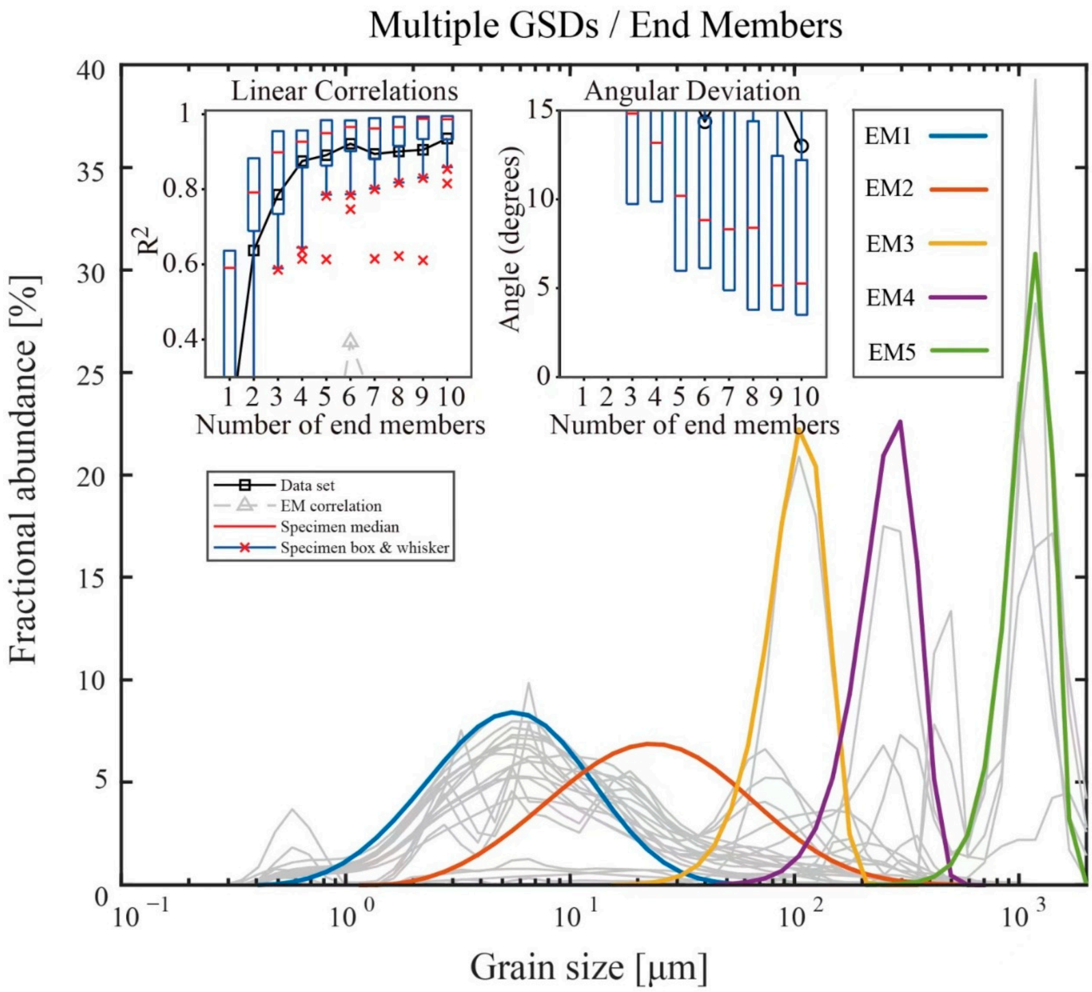

Figure 6. Endmember analysis decomposition diagram. The gray curve shows the grain size distribution of the 24 samples, and the five colored curves show the five endmember components, the R^2 values and angular deviations corresponding to different number of endmember are presented in the upper two rectangles.

Table 2. Endmember parameter characteristics of EM 1 to 4 of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

Endmember	Mean (μm)	Separability (φ)	Skewness (φ)	Kurtosis (φ)
EM 1	6.43	2.85	0.04	1.06
EM 2	75.11	2.03	−0.47	1.90
EM 3	315.74	3.42	0.01	3.39
EM 4	949.59	1.47	−0.35	1.66
EM 5	625.24	1.67	−0.25	1.56

Table 3. The five endmember components of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

Endmember	P10 (μm)	P25 (μm)	P50 (μm)	P75 (μm)	P90 (μm)
EM 1	1.69	2.78	4.89	8.43	13.37
EM 2	5.99	10.74	21.11	41.40	74.00
EM 3	55.94	72.99	92.72	113.42	132.24
EM 4	144.66	187.01	237.54	288.48	336.53
EM 5	705.20	856.34	1025.70	1178.83	1335.68

5. Discussion

5.1. Endmember Analysis of Sediments' Provenances

Based on the comparison between the component distribution of each endmember and the spatial distribution of the average grain size (Figure 7), a substantial inverse relationship exists between the distribution of fine-grained EM1 and EM2 components and the distribution of the average grain size. An area of high values is formed in the region ranging from the Lu'erhuan River to Malan Island, while the east and west sides of the Sandun Marine Highway, from Malan Island to Sandun Island, exhibit the inverse trend. The distribution of EM5 components with the coarsest grain size is consistent with the average grain size distribution, and five relatively significant high value centers are formed in the study area. The distribution of other coarse-grained EM3 and EM4 components bears no obvious relationship with the average grain size distribution, and two high value centers are formed in the easternmost coastal area of area D and the northern part of the southernmost Marine Highway in area A.

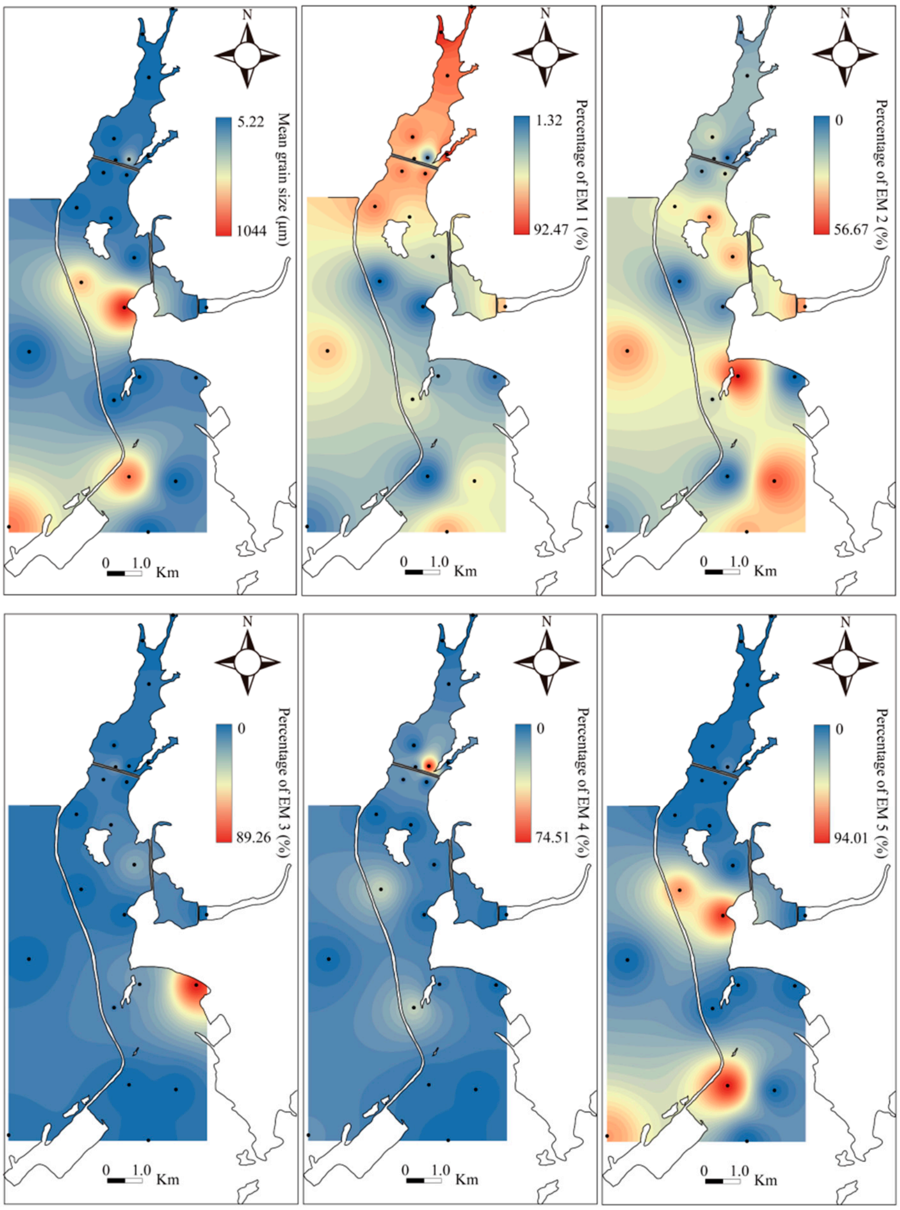

Figure 7. Distribution of average grain size and endmember components of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

The sediments in the area around the Lu'erhuan River-Malan Island-Sandun Island area have been deeply affected by long-term human activities. The materials' provenances and hydrodynamic conditions are varied. Among the five endmember components fitted by the endmember analysis, the spatial distribution of the finest EM1 component is essentially consistent with the clay distribution in the grain size analysis. These finer sediments are mainly distributed in the area from the Lu'erhuan River to Malan Island in the northern part of the study area, while the EM1 component content to the south of Malan Island is generally low. In general, such fine-grained sediments are typically suspended in the water column and enter the system via the river and weak tidal currents and are gradually deposited under relatively stable hydrodynamic conditions. The area from the Lu'erhuan River to Malan Island is enclosed due to the construction of the Sandun Marine Highway and the coastal highway, which simply creates a relatively stable sedimentary environment leading to the deposition of fine-grained materials in this area. In addition, the mangroves distributed on the east and west sides of the Lu'erhuan River dampen the disturbance of the tidal current [35] while also providing muddy sediments for the shoals from the Lu'erhuan River to Malan Island. In short, the materials of EM1 mainly originate from inland or coastal areas and are transported by rivers and weak currents. The spatial distribution of the finer EM2 component is basically consistent with the distribution of silt from the grain size analysis, but its content is relatively low; thus, it is not the main sediment type in the study area. Silt is mainly distributed near Malan Island and in areas D and E, far from Sandun Island and the SandunMarine Highway. The sediment with this grain size is widely distributed in the coastal area of the Beibu Gulf [33] and is deposited under the stable hydrodynamic conditions. This sediment is imported from the near shore through a weak tidal current. The rather coarse EM3 component is concentrated in only one location in the study area, near the beach on the northeast side of area D and connected with the vast inland sea of Qinzhou Bay. The sediment of the EM3 component mainly originates from the nearby beach and is input through the erosion of the coast by weak waves and tidal currents. Coarse-grained EM4 and EM5 are unevenly distributed in the study area (mainly distributed in areas with intense human activities, such as Sandun Island, the Sandun Marine Highway and coastal highway). We speculate that these components are affected by the combination of human activities, tidal currents and waves and deposited by ocean water after coastal erosion.

5.2. The Influence of Intense Human Activitieson the Organic Matter's Provenance

Overall, the organic matter content was low (Figure 8), with an average of only 4.26%, ranging between 0.72% and 9.49%. In terms of spatial distribution, the organic matter is mainly distributed in the offshore areas from the Lu'erhuan River to Malan Island and the eastern marine area of Sandun Island. Two high value centers are formed in the upper reaches of the Lu'erhuan River and the north-western side of Malan Island. Near Sandun Island, the Sandun Marine Highway and the coastal highway, which have undergone intense human activities, the organic matter content is relatively lower than other areas.

The content of organic matter in the sediment is closely related to its provenance [36,37]. The correlations between the organic matter content, grain size and endmember components show that the organic matter content of the sediments in the study area has a relatively significant negative correlation with the average grain size of the sediment, the percentage content of sand and the percentage content of coarse-grained EM5 (R2values of 0.32, 0.46 and 0.30, respectively) in Figure 9. However, the relationship between these variables seems to be nonlinear. Linear regression may not capture it correctly because there appears to be obvious clustering in the data. Meanwhile, a relatively significant positive correlation was found with the percentage content of silt, clay and fine-grained EM1sediments (R^2 values of 0.42, 0.445 and 0.42, respectively), but no significant correlation was observed with the components of EM2, EM3 and EM4 (R^2 values of 0.11, 0.05 and 0.06, respectively).

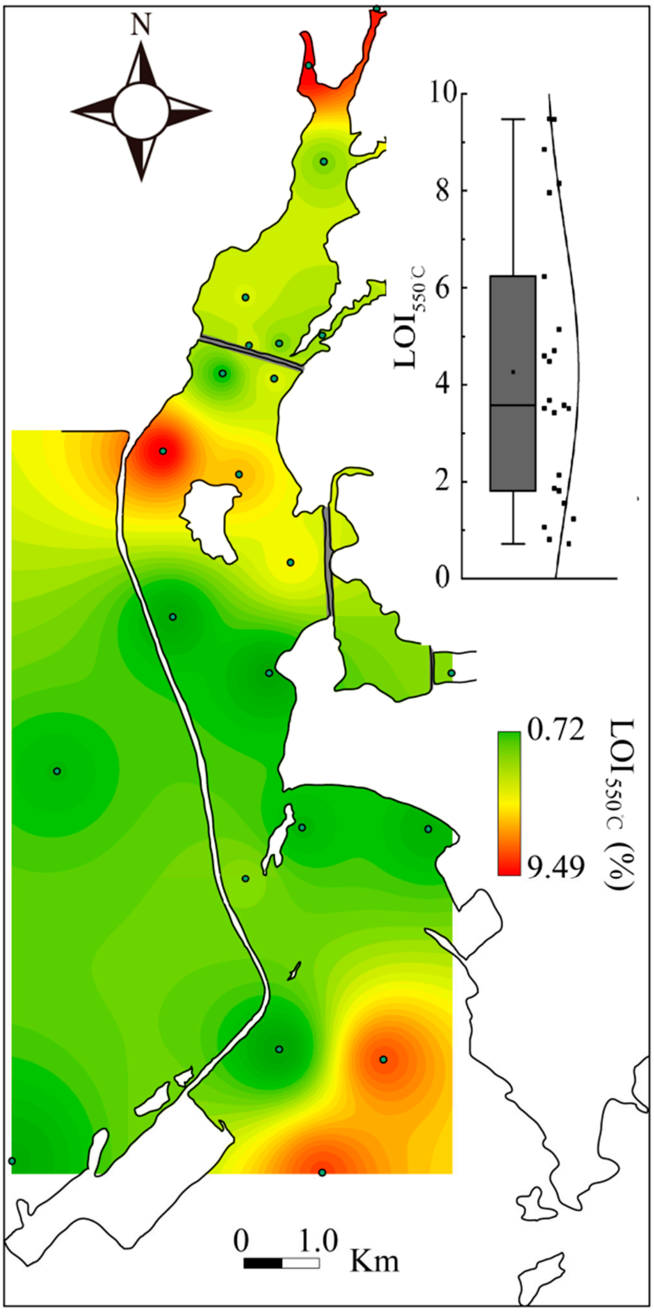

Figure 8. Organic matter content and its spatial distribution of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

The organic matter contained within shallow sea sediments can be primarily divided into endogenous and exogenous material. The former is the accumulation of organic matter produced by organisms within the water body (microorganisms, animals, plants, etc.), while the latter is mainly the particulate or dissolved organic matter imported by rivers and terrestrial runoff. Research on the spatial distribution of the organic matter content and the correlation between the organic matter and grain size endmembers in the offshore area around Lu'erhuan River-Malan Island-Sandun Island has shown that the organic matter content has a significant positive correlation with the fine-grained sediments and a negative correlation with the coarse-grained sediments. The organic matter content in the study area may be affected by the provenance of the sediments. The organic matter content in the relatively enclosed area from the Lu'erhuan River to Malan Island is relatively high. In particular, the upper reaches of the Lu'erhuan River (the northernmost side of area A) are replenished by inland rivers and the upstream Jinwo Reservoir. Large areas of paddy fields, which can input a large amount of granular or dissolved organic matter into the river, are distributed along the coast. This is the primary factor causing the high organic matter content in the surface sediments of this area, thus indicating the enrichment of organic matter as the combined result of natural and human activities. In addition, the content of organic matter on the north side of Malan Island is high. This area is less affected by the Lu'erhuan River and tidal currents. The relatively weak hydrodynamic environment is conducive to the sedimentation of fine-grained sediments and promotes the accumulation of organic matter. In addition, silt and clay have small pores and poor water permeability, which are conducive to the accumulation of organic matter compared to sand. Moreover, the large-scale oyster culture in this area inevitably leads to the input of exogenous organic matter and plankton enrichment [38,39]. This is the main reason for the increase in organic matter in the sediments of the offshore area. In the area connected with the open sea to the east of Sandun Island, the sediment grain size is relatively fine due to the action of tidal currents and waves. In addition, the organic matter content in this area is obviously affected by marine plankton, which is basically consistent with previous research results in the Beibu Gulf.

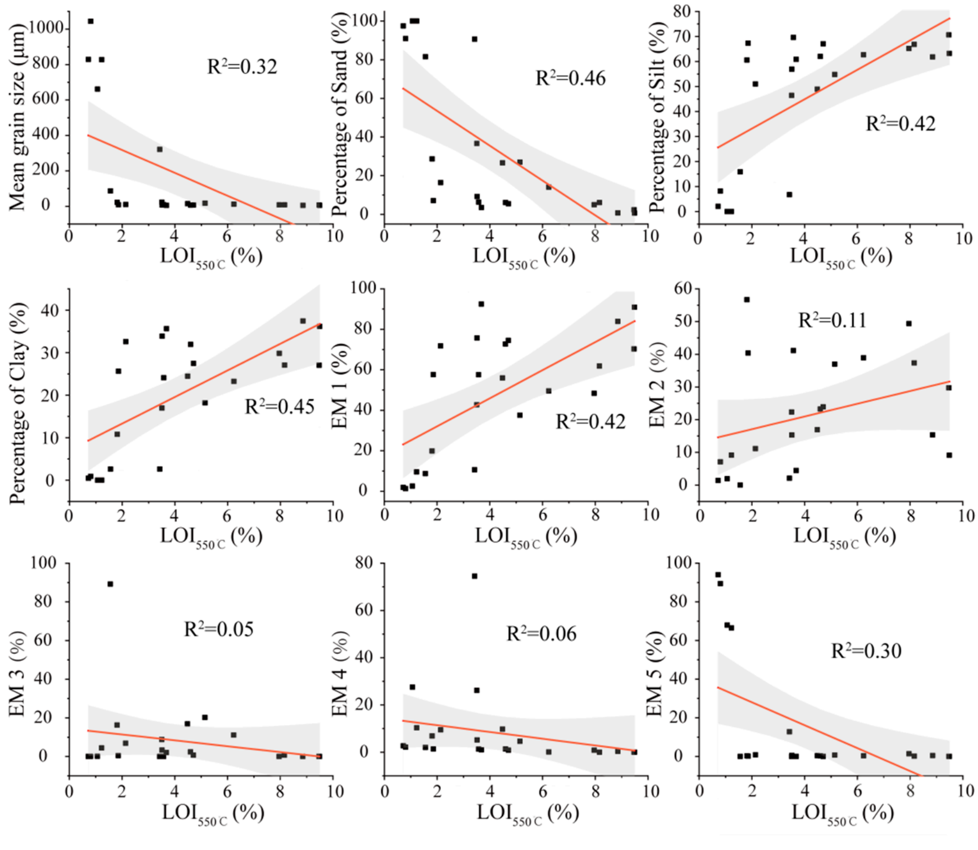

Figure 9. Correlation between organic matter content and grain size along with endmember components of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

In summary, the provenance of organic matter in the study area resembles that of the sediment endmember components, exhibiting diversified characteristics resulting from the combined impact of natural and human activities. The organic matter in the upper reaches of the Lu'erhuan River originates from the river and the paddy fields along the river and has terrigenous characteristics. The organic matter to the north of Malan Island mainly originates from the external inputs produced by oyster culture, while the organic matter to the east of Sandun Island is mainly produced internally by marine plankton.

5.3. Elemental Analysis and Discussion about Provenances

In shallow marine sediments, the species, abundance and regional distribution of elements are significantly affected by their provenance [40]. Through analyzing 15 heavy metal elements, four common metal elements and two nonmetal elements, we found that the distributions of Pb and Cr are similar and mainly located in the offshore areas from the Lu'erhuan River to Malan Island (Table 4, Figure 10). The distributions of Fe, Ti, Ga, Co, Sn, Zn, Ni, P, K, Al and Mg are similar and mainly located near Malan Island and the offshore area to the east of Sandun Island (Figure 11). The distributions of As and Cd are similar, and they are concentrated near the Lu'erhuan River (area A) and Jishuimen (a narrow channel at the junction of areas B and D). Both Cu and Ba have similar distribution characteristics and are concentrated in the offshore area from the Lu'erhuan River to Malan Island and the offshore area to the east of Sandun Island (area D). Two biophilic elements, Sr and Ca, which are markers of nonterrigenous sedimentation, show the same distribution characteristics and are mainly distributed in the offshore region to the east of Sandun Island, which is connected to the open sea. The abundance of Mn may be affected by rivers, and is mainly distributed in areas A, B and C from the Lu'erhuan River to Jishuimen. In addition, the change in N is not obvious throughout the study area, but Nis relatively concentrated along the east coast of Sandun Island.

Table 4. The content of each metal of sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

Sample	P (cps)	Mg (cps)	Mn (cps)	K (cps)	Ca (cps)
4	6378	9,778,145	1,985,847	21,025,771	1,382,918
11	11,645	19,100,219	4,115,123	42,273,623	2,087,401
14	7463	18,099,022	3,066,164	26,212,772	4,548,484
20	4140	11,772,609	2,960,292	9,667,056	12,193,893
22	11,848	24,181,639	6,383,927	43,192,880	6,195,244
24	10,706	22,109,094	5,067,886	37,562,231	6,469,540
	N (cps)	TOC%	C/N%	Zn/Pb%	Ca/Mg
4	0.63	0.04	0.06	101.8	0.1414
11	0.53	0.10	0.18	−21.8	0.1093
14	0.53	0.05	0.10	55.7	0.2513
20	0.46	0.04	0.08	43.4	1.0358
22	0.87	0.08	0.09	65.9	0.2562
24	0.52	0.08	0.16	73.6	0.2926

Compared with heavy metals, and other metals, N and P, Al mainly appears in the crystal lattice of clay mineral sand and is the characteristic element of clay minerals. As an indicator of terrigenous components in offshore sediments, it is relatively stable during the transportation process from the mainland to the sea. Inert elements including Ti, Fe, Mg and K elements with a positive correlation to Al can also indicate terrigenous input. In addition, Ga is very sensitive to weathering processes and can be transported and deposited with particles. Classified as lithophilic elements, almost all Ga is derived from classic materials and can indicate terrigenous classic deposits. In this study, the above elements are mainly distributed near Malan Island and the eastern offshore area of Sandun Island. However, no obvious increase in the elements in the Lu'erhuan River (area A) that could be attributed to reverie input was observed. The north side of Malan Island is an area of dense oyster breeding. A popular breeding method in this area is to place rough cement columns or cement plates under oyster rows to create a living environment for oysters. Due to being submerged in corrosive seawater, Al, Fe and other elements in rough cement columns will gradually leach out and lead to local enrichment. This process is believed to be the main reason for the high content of these elements near Malan Island. In the offshore area on the eastern side of Sandun Island, human activities, such as high-intensity land reclamation, have introduced many terrestrial types of sediment. Combined with the erosion forces of waves and rainstorms, the sediments enter the bay mouth with the tide and lead to the enrichment of Al, Fe and Ga. As human activities have intensified in the region, a clear deviation in the terrestrial indicator elements within the sediments in the study area can be observed. However, it is worth noting that the heavy metal element Mn, which mainly originates from reverie inputs and oxidation precipitation in shallow sea sediments, can partially indicate terrigenous sediments. The Mn in the study area is mainly distributed along the route from the Lu'erhuan River to Jishuimen (areas A, B and C). This area is subject to the confluence of river discharge and terrestrial runoff, and portions of the terrestrial materials are deposited here.

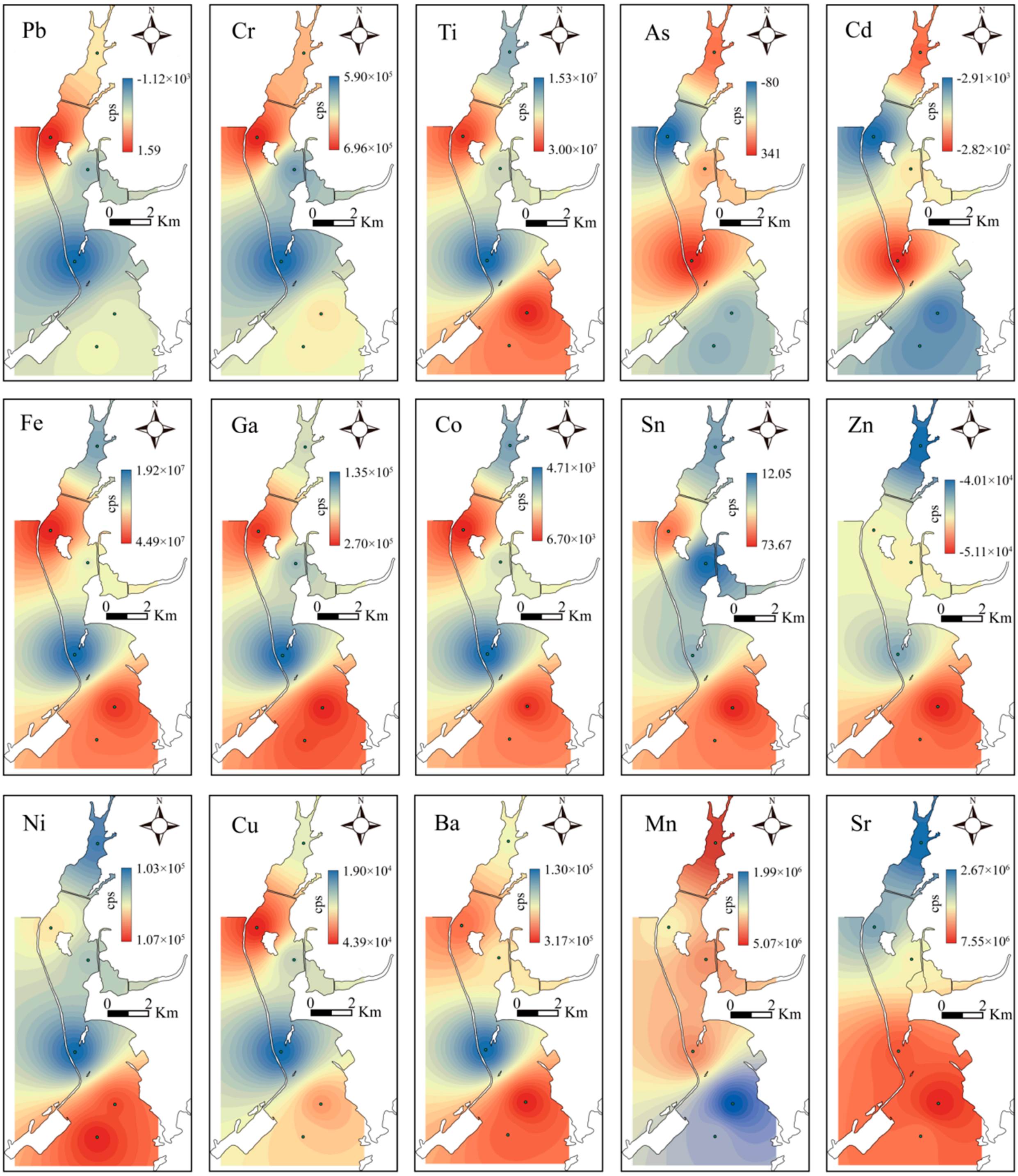

Figure 10. Spatial distribution of heavy metal elements of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

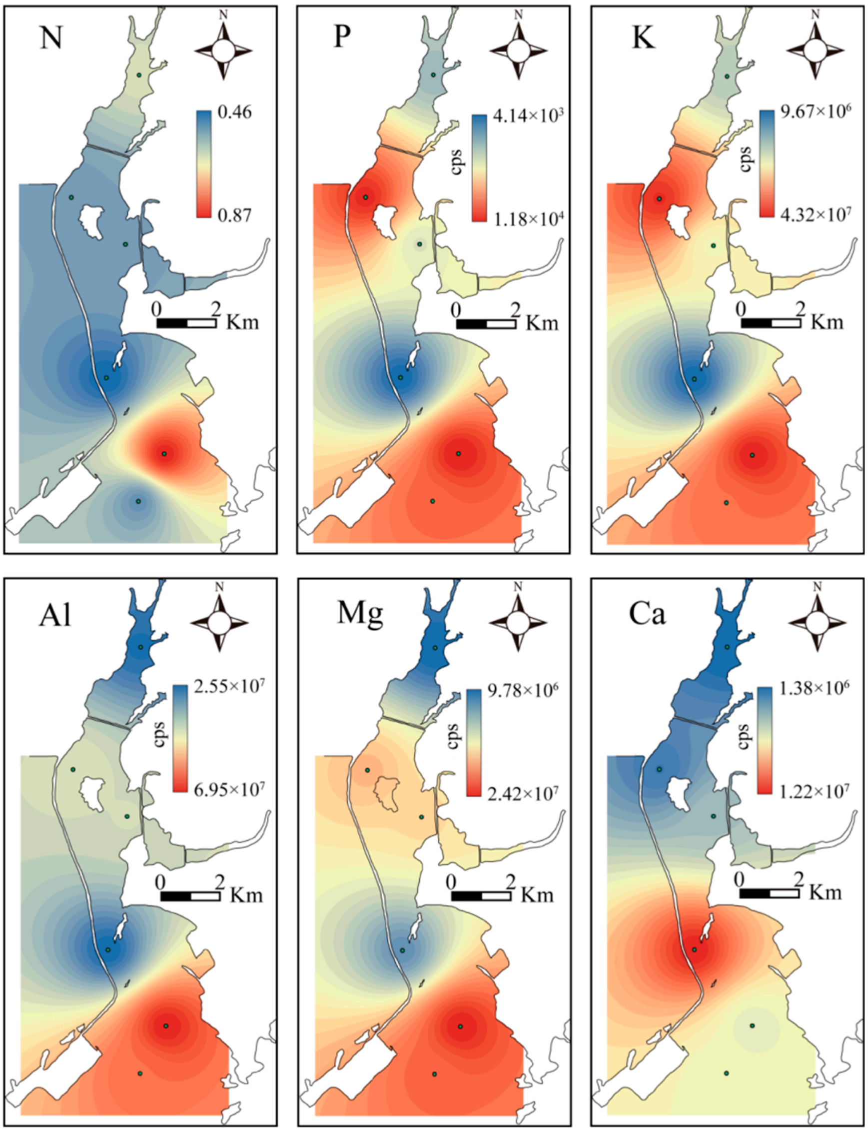

Figure 11. Spatial distribution of N, P and other metal elements of 24 sediment samples in the Lu'erhuan River Sea area of Qinzhou Bay, Guangxi Province.

Unlike Al, Ti terrestrial indicator elements, Ca and Sr, are characteristic elements and biophilic elements of marine sediments, respectively [41], that represent the provenance of marine sediments. In this study, the spatial distributions of Ca and Sr bear some similarities. They are mainly distributed in the offshore area to the east of Sandun Island, which connects with the open sea. Under the action of tides and waves, marine sediments are easily carried to this location. Additionally, this location, especially near Jishuimen (the narrow waterway at the junction of areas B and D), is also affected by human activities, and Ca is significantly enriched here. The cement used in oyster breeding and engineering construction is assumed to decompose and be deposited here by tides. In general, although the Ca and Sr in the study area are affected by human activities, they can still indicate the provenance of marine sediments in the area.

The form and enrichment degree of transition elements (Cr, Mn, Fe, Co, Ni, Cu, Zn and Cd) in the study area represent, to a certain extent, the input flux of organic matter in the corresponding depositional period [42]. As and Cd are clearly affected by human activities, while Cu is less affected by human activities. The enrichment degree of As and Cd is quite apparent, especially in the vicinity of the Lu'erhuan River (area A) and Jishuimen.

The reason for this may be related to industrial and agricultural wastewater and other human activities [43].

In general, although the sediments in the study area are strongly affected by human activities, Ca and Sr, which can indicate the provenance of marine sediments, may still be distributed in the eastern sea area of Sandun Island, which is connected to the open sea. Meanwhile, Al, Ti, Fe, Mg, K, Ga and other elements are significantly impacted by human activities when used as terrigenous indicators. However, the distribution of Mn in this area compensates for this defect and can better indicate the distribution of terrigenous sediments. Mn is mainly distributed in the offshore area from the Lu'erhuan River to Jishuimen. The influence of human activities on the enrichment of As and Cd in the study area is very obvious, while Cu is less affected by human activities.

6. Conclusions

In this study, through the grain size analysis, endmember analysis and geochemical element analysis of surface sediments in the Lu'erhuan River-Malan Island-Sandun Island area, we successfully explored the regional differentiation and sediment provenance under intense human activities. This study showed:

(1) The offshore area around Lu'erhuan River-Malan Island-Sandun Island is affected by intense human activities; thus, the sediment grain sizes differed significantly. Taking the vicinity of Malan Island as the boundary, the sediment on the north side has a single provenance, the hydrodynamic conditions vary slightly and the sediment grain size is relatively fine (argillaceous sediment). However, the sediment on the south side is complex, and the average grain size is relatively large due to the multiple impacts of human activities, tidal currents and waves.

(2) The endmember analysis showed that, due to long-term intense human activities, the material provenances and hydrodynamic conditions in the offshore area around Lu'erhuan River-Malan Island-Sandun Island are varied. The EM1 component is mainly transported by rivers and weak tidal currents from inland or coastal areas; the EM2 component is mainly input from near shore sources through the weak tidal current; and the rather coarse EM3 component mainly comes from the nearby beach and is input through coastal erosion by weak waves and tidal currents. However, the coarse-grained EM4 and EM5 are unevenly distributed in the study area. They are affected by human activities, tidal currents and waves, and are deposited through the marine transport after coastal erosion.

(3) The provenance of organic matter in the waters around Lu'erhuan River-Malan Island-Sandun Island resembles that of the sediment endmember components, exhibiting diversified characteristics as a result of the superposed impact of natural and human activities. The organic matter in the upper reaches of the Lu'erhuan River originates from the river and the paddy fields along it and exhibits obvious terrigenous characteristics. Meanwhile, the organic matter to the north of Malan Island mainly comes from the external input produced by oyster breeding, while organic matter to the east of Sandun Island mainly comes from the internal input produced by marine plankton.

(4) In the elemental analysis, when used as indicators of terrigenous sediments, the elements (Al, Ti, Fe, Mg, K, Ga, et al.) are significantly disturbed by human activities. However, the element Mn can still sufficiently indicate the distribution characteristics of terrigenous sediments, which are mainly distributed in the offshore area from the Lu'erhuan River to Jishuimen. In addition, Ca and Sr, which indicate the provenance of marine sediments, are located in the eastern offshore area of Sandun Island, which connects with the open sea. Finally, affected by human activities, the enrichment of As and Cd in the study area is clearly a result of human activities, while Cu is less affected by human activities.

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