ELSEVIER

Contents lists available at ScienceDirect

## Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul



# Assessment of heavy metals in water, sediment and shellfish organisms in typical areas of the Yangtze River Estuary, China



Haimei Fan, Sisi Chen\*, Zhien Li, Pengxia Liu, Caiyan Xu, Xingxing Yang

East China Sea Environmental Monitoring Center, State Oceanic Administration, Shanghai 201206, China

ARTICLE INFO

Keywords:
Yangtze River Estuary (YRE)
Frequency analysis method
Geographical detector
Heavy metals
Spatiotemporal stratified heterogeneity

#### ABSTRACT

Identifying the transformations of heavy metal in different media is a scientific issue, and geographical detector is applied to evaluate the spatiotemporal stratified heterogeneity mechanisms for heavy metals in the Yangtze River Estuary. Heavy metal concentrations in water and sediment were consistent with lognormal distributions. Their concentrations were organized into four classes. Class 1 included concentrations that were less than or equal to 25%, Class 2 included those between 25%–50%, Class 3 concentrations were between 50%–75% and Class 4 were > 75%, which were based on their lognormal distributions. In water and sediment, the mean heavy metal concentrations yearly decreased from 2012 to 2016. The Chongming area was significantly lower than those found in the other areas, which is the least affected area by anthropogenic activities. The explanatory power of sediment to spatiotemporal stratified heterogeneity of heavy metals in shellfish organisms was much greater than that of water.

### 1. Introduction

Heavy metal pollution has been an important focus in the ecological environments of estuaries (Dong et al., 2009; Buruaem et al., 2013; Bi et al., 2017; Barletta et al., 2019), bays (Gomes et al., 2009; Ahumada et al., 2011; Yu et al., 2017) and coastal waters (Vallius, 2015) around the world because of its numerous sources, non-degradability, easy accumulation and biological toxicity. The heavy metals commonly include mercury (Hg), copper (Cu), lead (Pb), cadmium (Cd), chromium (Cr), zinc (Zn) and arsenic (As). Estuaries are located at the junction of land and sea, and they are affected by both, which often makes their environmental conditions complex and highly variable. The Yangtze River Estuary (YRE) is the largest estuary in China and has been subjected to decades of rapid increases in human activities and fast socioeconomic development in the Yangtze River delta, which largely accounts for the elevated levels of heavy metals in its water and sediment (Guo and Yang, 2016). The hydrodynamic conditions are complex, and thus, heavy metal pollutants can originate from a wide range of sources. Moreover, the sources include natural weather erosion of rock soil in coastal basins, atmospheric particulate sedimentation and precipitation (Wang et al., 2014; Han et al., 2017; Du, 2018), discharge of industrial and agricultural wastewater and domestic sewage that enters the estuary (Wang, 2017; Zhang, 2013), with a significant portion being derived from runoff inputs in the upper and middle reaches

of the Yangtze River. According to reported statistics, from 2013 to 2017, the heavy metals, including Cu, Pb, Zn, Cd and Hg, were transported into the East China Sea by the Yangtze River at a staggering rate of about 10,000 tons per year (State Oceanic Administration, China, 2013-2017), which undoubtedly poses a multitude of threats to the water environment in the YRE and the nearshore waters.

Heavy metals not only affect the water environment. Due to the interactions between runoff and seawater, a large number of pollutants become bound to the sediment through certain processes, such as adsorption and flocculation (Fang et al., 2013; Zhang et al., 2007), which can ultimately affect the highly nutritious aquatic life and human health through bioaccumulation and amplification in the food chain (Cui et al., 2000; Qiu et al., 2005). The level of heavy metals in the living environment of aquatic organisms is the primary factor affecting the accumulation of heavy metals in organisms and biomass (Huang et al., 2011; Jahan and Strezov, 2018). Therefore, the study of heavy metal pollution in the YRE should not be limited to one medium. Instead, it should also focus on their spatial and temporal distribution and interconnection in different media. Previous research has been conducted on the content, morphology and distribution characteristics of heavy metals in the water and sediments of the YRE (Sun et al., 2011; Teng et al., 2012; Wang et al., 2015). Most such studies have focused on a single medium and reported that heavy metals in the YRE primarily come from land-based sources (Guo and Yang, 2016; Guo et al., 2019).

E-mail address: fhm@ecs.mnr.gov.cn (S. Chen).

<sup>\*</sup> Corresponding author.

Marine Pollution Bulletin 151 (2020) 110864

However, there are only a few studies dedicated to the spatial and temporal distribution and interconnection of heavy metals in water bodies, sediments and organisms in the same area. Therefore, based on YRE survey data from 2012 to 2016, this paper explored and discussed the spatial and temporal distribution characteristics and correlations of heavy metals in the estuary's water, sediments and organisms.

H. Fan, et al.

The geographical detector method was used to explore the statistical correlation and spatial distribution of heavy metals in different media (i.e., water, sediments and organisms) in the YRE and its adjacent seas. Geodetector is a new statistical method used to detect spatial differentiation and reveal the driving factors behind observed patterns (Wang and Xu. 2017). Its greatest advantages include the fact that the hypotheses are not linear, and the results have clear physical meaning. which can effectively overcome the limitations of the traditional statistical analysis methods that require hypotheses and data limits (Li et al., 2017). The scales of the research areas are effectively unlimited, and the possible causal relationship between the two variables can also be ascertained by examining the consistency of the spatial distribution of any two variables (Han, 2019). Due to these advantages, the method has been used in a wide range of areas, including natural and urban landscapes, such as public health, regional economy, meteorology and environmental pollution (Wang et al., 2010; Wang et al., 2016; Wang and Xu, 2017). The relationship between variables established by the geographical detector is generally more reliable than classic regression. It is necessary to discretize independent numerical variables. The variable strata could include areas, years, and concentrations, among others. Frequency analysis is often used to determine the reference state of observation elements, and it is widely used (Hu et al., 2011; Kang, 2012; Zheng et al., 2013). The frequency analysis is used to discretize the heavy metal data and determine their classes, which makes it possible to intuitively show the most natural heavy metal distribution in the study area.

In this paper, the frequency analysis method was used to determine the concentration classes of heavy metals in water and sediment. Based on geographical detectors, the spatiotemporal stratified heterogeneity and driving factors of heavy metals in water, sediment and organisms in the YRE were investigated. The results of this study may provide valuable guidance regarding the transformation mechanisms of heavy metals in different media of the YRE.

### 2. Materials and methods

### 2.1. Study region and data

The study region extended from 30°30′N to 32°00′N latitude and from 121°00′E to 122°20′E longitude. The data were collected from 67 water quality monitoring stations and 30 sediment monitoring stations, which were located in the typical areas, including the Jinshan area, Baoshan area, Fengxian area, Pudong area, Chongming area and Nanhui area. There were three areas where shellfish organism samples were collected, including Jinshan area, Chongming area and Nanhui area (Fig. 1).

From 2012 to 2016, heavy metal (Hg, Cu, Pb, Cd, Cr, Zn and As) concentration data in water and sediment were collected in the six typical areas, while organism sampling were only obtained from the three areas mentioned above. All of the data were collected from the August monitoring events over the 5-year period, and the data were provided by the East China Sea Marine Environmental Monitoring Center.

Both surface water (approximately 0.5 m deep) and the bottom water (depth  $\geq$  10 m) samples were collected and kept in well-sealed sampling bottles for cryopreservation. Surface sediment samples were collected using a grab sampler. The macrobenthos samples were collected using an intertidal sampling device. Water quality, sediment and shellfish sample pre-processing and analysis were conducted according to the specifications described in the National Standard of the People's Republic of China for Marine Monitoring (State Technical Supervise

Bureau, 2008). Quality control was based on the specification of the National Environmental Protection Standard of the People's Republic of China for offshore environmental monitoring (Ministry of Environmental Protection, 2009).

### 2.2. Frequency analysis method

In our study, the data were grouped into equal segments for the logarithmic transformation of the original data, the number of individuals in each group was counted, and all categories and their corresponding frequencies were employed to obtain the frequency distribution. Frequency distributions have both centralized trends and discrete trends. Most observations were clustered near the average value, which represented the centralized trend. The frequency gradually decreased from the central position on both sides, which depicted the discrete trend (Duan, 2012). The frequency distribution method was used to determine marine nutrient benchmarks and the reference threshold value of ecological zones. After selecting the reference point (i.e., the water body with the least impact from human activities) and the pollutant data of the target area, the upper 25% of the frequency distribution of the reference point were compared to the lower 25% of all the data, and the best ecological baseline value was selected (Office of Water, Office of Science and Technology, 2011; Hu et al., 2011; Kang, 2012; Yang, 2015; Zheng et al., 2013).

The frequency distribution method was employed to analyse the heavy metal concentrations in water and sediment. First, a small amount of non-detected data was eliminated (Zhang et al., 2017). The normal distribution test was applied to the original data and failed. Thus, the logarithmic transformation of the original data was conducted with 10 as the base, and the lognormal distribution curve was fitted with the frequency distribution, which included mean values and standard deviations. The concentration data corresponding to 25%, 50% and 75% probability intervals of the lognormal distribution were selected as the thresholds for the classes of each element. As a result, the original concentration data in water and sediment were organized into four classes. Hence, independent variables (i.e., concentrations) in geographical detectors became attribute data sets bases on their classes, which is presented in greater detail in Section 3.1.

### 2.3. Geographical detectors

Geographical detectors are based on spatial variation analysis of the strata of variables to assess the environmental features and development mechanisms. The factor detector identifies which factors are responsible for the environmental development and the relative importance of the factors; the interaction detector reveals whether the factors interact or lead independently to the environment development; and the risk detectors indicates where the transformation areas are.

Geographical detectors are a set of statistical methods used to detect spatiotemporal stratified heterogeneity and reveal the driving factors (Wang and Xu, 2017). If an independent variable X has an important influence on the dependent variable Y, their spatiotemporal feature should be similar. Geographical detectors are not only used to detect numerical data, but also qualitative data of feature attributes, which can be used to detect multi-factor interactions of independent variables. In this study, the strata were defined by attribute data of the independent variable X, including the medium attribute, element attribute, area attribute, year attribute, water attribute and sediment attribute. The details are provided in Section 3.2. The dependent variable Y was the heavy metal concentration data in YRE water, sediment or organisms.

### (1) Factor detector and interaction detector

To detect spatiotemporal stratified heterogeneity for the dependent variable Y, and to what extent values of X explain Y, the following

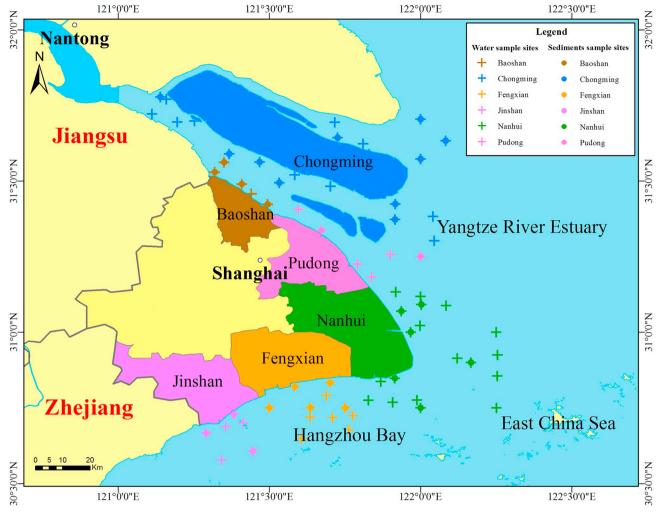


Fig. 1. Study region and location of primary sampling sites.

**Table 1** Interaction relationships between two factors  $X_1$  and  $X_2$  on variable Y.

Criteria	Interaction	Remarks
$\begin{array}{l} q(X_1 \cap X_2) &< \textit{Min} \ (q(X_1), q(X_2)) \\ \textit{Min} (q(X_1), q(X_2)) &< q(X_1 \cap X_2) &< \textit{Max} \ (q(X_1), q(X_2)) \\ q(X_1 \cap X_2) &> \textit{Max} \ (q(X_1), q(X_2)) \\ q(X_1 \cap X_2) &= q(X_1) + q(X_2) \\ q(X_1 \cap X_2) &> q(X_1) + q(X_2) \end{array}$	Nonlinearity weakens Nonlinearity weakens for single factor Enhancement of two factors Independence Enhancement of nonlinearity	Min and Max mean Minimum and maximum, respectively

### equation was as follows.

$$q = 1 - SSW/SST$$

$$SSW = \sum_{h=1}^{L} N_h \sigma_h^2, SST = N\sigma^2$$
(1)

where h=1,...,L, which is the strata of Y or X.  $N_h$  and N are separately the units of stratum h and the whole region, respectively;  $\sigma_h^2$  and  $\sigma^2$  are separately the variances of Y value in the stratum h and the whole region, respectively; and SSW and SST are the sum of variances in the stratum and the whole region, respectively. The value of q represents that X can explain 100 \* q% of Y. The range of q is [0,1], and the larger the q value, the more significant the spatiotemporal stratified heterogeneity of Y is. If the stratum is generated by independent variable X, the larger the q value is, the stronger the explanatory power of X to Y is.

A q value of 1 indicates that factor X completely controls Y, and a q value of 0 indicates that factor X has nothing to do with Y.

The interaction between different factors  $X_s$  is to ascertain whether the explanatory power to Y increases or decreases when the factors  $X_1$  and  $X_2$  work together, or whether the effects of these factors on Y are independent of each other. Overlaying stratum  $X_1$  and  $X_2$  obtains a new stratum  $X_1 \cap X_2$ , and its interaction  $q(X_1 \cap X_2)$ . Interaction type is determined by comparing  $q(X_1)$ ,  $q(X_2)$  and  $q(X_1 \cap X_2)$ . Interaction relationships between two factors  $X_1$  and  $X_2$  on variable Y are presented in Table 1.

### (2) Risk detector

T-statistics were used to test whether the mean values of Y between two strata were significantly different, and can be expressed as Eq. (2).

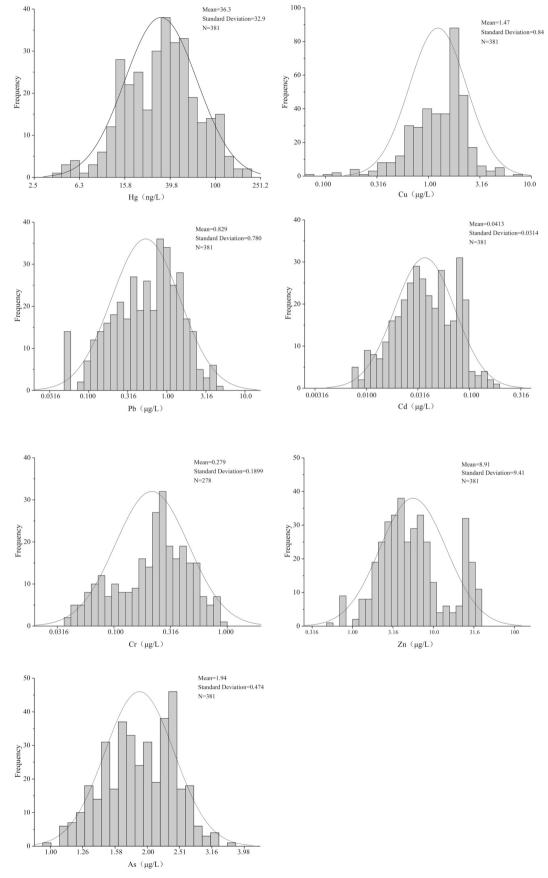


Fig. 2. Frequency distributions of heavy metals in water.

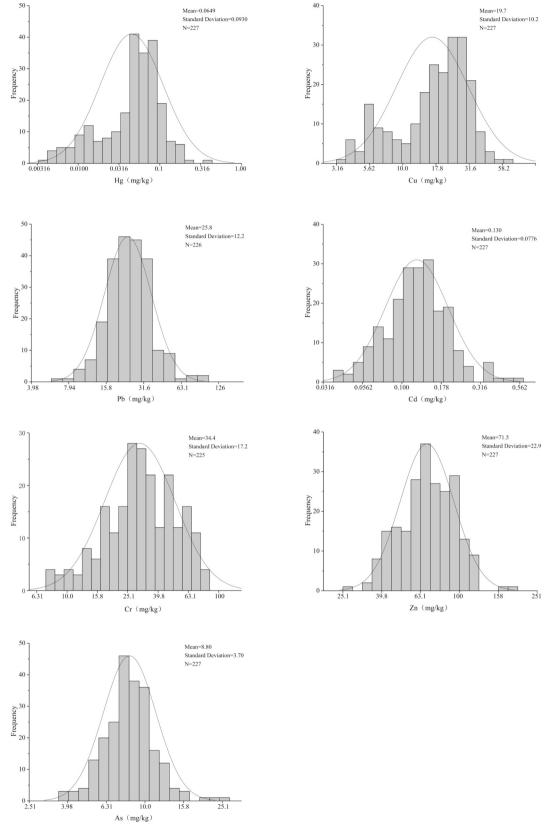


Fig. 3. Frequency distributions of heavy metals in sediment.

$$t = \frac{\overline{Y}_{h=1} - \overline{Y}_{h=2}}{\left[\frac{Var(\overline{Y}_{h=1})}{n_{h=1}} - \frac{Var(\overline{Y}_{h=2})}{n_{h=2}}\right]^{1/2}}$$
(2)

where  $\overline{Y}_h$  is the mean values of the stratum h, while  $n_h$  and Var are the

sample size and variance, respectively, with the null hypothesis  $H_0$ :  $\overline{Y}_{h=1} = \overline{Y}_{h=2}$ . There is a significant difference between the mean values of the two strata if  $H_0$  is rejected at a confidence level  $\alpha$ .

Table 2
Classes and mean values of heavy metal concentrations in water.

Water	Class 1(≤25%)	Class 2(25%-50%)	Class 3(50%-75%)	Class 4(> 75%)	Mean value
Hg(ng/L)	≤13.9	$13.9 < x \le 30.8$	$30.8 < x \le 50.0$	> 50.0	36.3
Cu(µg/L)	≤0.860	$0.860 < x \le 1.40$	$1.40 < x \le 1.94$	> 1.94	1.47
Pb(μg/L)	≤0.256	$0.256 < x \le 0.591$	$0.591 < x \le 1.16$	> 1.16	0.829
Cd(µg/L)	≤0.0185	$0.0185 < x \le 0.0329$	$0.0329 < x \le 0.0574$	> 0.0574	0.0413
Cr(µg/L)	≤0.0844	$0.0844 < x \le 0.223$	$0.223 < x \le 0.352$	> 0.352	0.279
Zn(µg/L)	≤2.92	$2.92 < x \le 4.69$	$4.69 < x \le 8.96$	> 8.96	8.91
As(μg/L)	≤1.57	$1.57 < x \le 1.92$	$1.92 < x \le 2.32$	> 2.32	1.94

 Table 3

 Classes and mean values of heavy metal concentrations in sediment dry weight.

Sediment	Class 1(≤25%)	Class 2(25%-50%)	Class 3(50%-75%)	Class 4(> 75%)	Mean value
Hg(mg/kg)	≤0.0279	$0.0279 < x \le 0.0547$	$0.0547 < x \le 0.0792$	> 0.0792	0.0649
Cu(mg/kg)	≤11.8	$11.8 < x \le 19.2$	$19.2 < x \le 26.3$	> 26.3	19.7
Pb(mg/kg)	≤17.9	$17.9 < x \le 23.7$	$23.7 < x \le 30.3$	> 30.3	25.8
Cd(mg/kg)	≤0.0853	$0.0853 < x \le 0.120$	$0.120 < x \le 0.161$	> 0.161	0.130
Cr(mg/kg)	≤21.9	$21.9 < x \le 30.7$	$30.7 < x \le 45.5$	> 45.5	34.4
Zn(mg/kg)	≤55.5	$55.5 < x \le 68.1$	$68.1 < x \le 86.9$	> 86.9	71.5
As(mg/kg)	≤7.05	$7.05 < x \le 8.21$	$8.21 < x \le 9.69$	> 9.69	8.80

**Table 4**Attribute data of independent variable X.

Medium attribute		Element a	ttribute	Area attribute		Year attrib	ute	Water attribute	Sediment attribute
Water	A1	Hg	M1	Jinshan	s1	2012	Y2	W111	S211
Sediment	A2	Cu	M2	Baoshan	s2	2013	Y3	W112	S212
Shellfish organism	A3	Pb	М3	Fengxian	s3	2014	Y4	W113	S213
-		Cd	M4	Pudong	s4	2015	Y5	W114	S214
		Cr	M5	Chongming	s5	2016	Y6	W121	S221
		Zn	M6	Nanhui	s6			W121	S222
		As	M7					W123	S223

Table 5
Driving factors and interaction relationship for heavy metals in water.

			•	
q	Year attribute	Area attribute	Water attribute	Element attribute
Year attribute Area attribute Water attribute Element attribute	0.11 0.11 0.97 0.92	0.00 0.89 0.40	0.88 0.88	0.40

Table 6
Results of risk detector about year attribute in water.

Significant level $\alpha = 0.05$	2012	2013	2014	2015	2016
Mean values (μg/L)/year 2013	5.49 <b>Yes</b>	1.59	1.41	1.34	1.01
2014	Yes	No			
2015	Yes	No	No		
2016	Yes	Yes	Yes	Yes	

<sup>&</sup>quot;Yes" means that there is significant difference between the row year and the column year; "No" means contrarily.

### 3. Results

### 3.1. Classification of heavy metals

Based on the Hg, Cu, Pb, Cd, Cr, Zn, As concentrations in water and sediments of the study areas from 2012 to 2016, the logarithmic frequency distributions and corresponding lognormal distributions were calculated using the frequency analysis method (Figs. 2 and 3). In each figure, the histogram shows logarithmic frequency distribution, and the

Table 7

Driving factors and their interaction relationship for heavy metal in sediment.

q	Year	Area	Water	Element	Sediment
	attribute	attribute	attribute	attribute	attribute
Year attribute Area attribute Water attribute Element attribute Sediment attribute	0.01 0.03 0.82 0.81	0.01 0.83 0.79	0.80 0.80 0.96	0.78 0.96	0.96

Table 8
Results of risk detector about year attribute in sediment.

Significant level $\alpha = 0.05$	2012	2013	2014	2015	2016
Mean values (mg/kg)/Year 2013	29.8 <b>Yes</b>	21.7	21.1	20.3	21.3
2014	Yes	No			
2015	Yes	No	No		
2016	Yes	No	No	No	

<sup>&</sup>quot;Yes" means that there is significant difference between the row year and the column year; "No" means contrarily.

curve shows corresponding lognormal distribution. The X-axis is the concentrations corresponding to the logarithmic scale, and the Y-axis is the frequency. The normal curves with single peaks had relatively small standard deviations, which indicated that the distribution curve of heavy metals in water and sediment was relatively steep, and concentrations were centrally distributed on both sides of the mean value.

Referring to the principle of quartile in statistics, the concentrations

Table 9
Risk detector in sediment.

Significant level $\alpha = 0.05$	Jinshan	Baoshan	Fengxian	Pudong	Chongming	Nanhui
Mean values (mg/kg)/ Area	27.4	21.1	25.7	24.4	20.7	20.7
Baoshan	No					
Fengxian	No	No				
Pudong	No	No	No			
Chongming	Yes	No	Yes	Yes		
Nanhui	No	No	No	No	No	

<sup>&</sup>quot;Yes" means that there is significant difference between the row area and the column area; "No" means contrarily.

corresponding to the 25%, 50% and 75% frequency distribution were considered as the reference states (class thresholds) separating the four classes of heavy metals in water and sediment (Tables 2 and 3). The mean values of the seven heavy metals were slightly larger than the median values, indicating that their concentration distributions in water and sediments were slightly right-biased, or there was a maximum in the original data, which inevitably pulled the mean value closer to the maximum. The concentration data for organisms were only used as the dependent variable Y when driving factors of biological quality were detected. Thus, the classes for heavy metal concentrations in organisms were not needed.

### 3.2. Spatiotemporal stratified heterogeneity of heavy metals

Based on the principle of geographical detectors, the statistical correlation and spatiotemporal stratified heterogeneity of heavy metals in different areas, years and type of media were examined. The explanatory powers of heavy metals in water and sediment were detected to those in organisms. Because heavy metal concentrations in organisms could only be collected from the Jinshan area, Chongming area and Nanhui area, driving factors of biological quality could only be obtained for these three areas.

Dependent variable Y was the concentration data of heavy metal in water, sediment and organisms. Attribute data of independent variable X included element attribute, area attribute, year attribute, water attribute, and sediment attribute (Table 4). Based on classes for heavy metal data in water or sediment (Tables 2 and 3), water or sediment attributes included three numbers, where the first number was the last number of the corresponding medium attribute, and the second number was the last number of the corresponding element attribute, and the third number was the corresponding class number of the concentrations (Table 4).

### 3.2.1. Spatiotemporal features of heavy metal in water

Dependent variable Y was the heavy metal concentrations in water. Attribute data of independent variable X included the year attribute, area attribute, water attribute and element attribute.

The water attribute (q=88%), element attribute (q=40%) and year attribute (q=11%) were three main driving factors for the spatiotemporal features of heavy metal elements in water (Table 5). The enhancement of two factors included the interaction of the element

attribute and water attribute, and the interaction of the year attribute and water attribute. The enhancement of nonlinearity included the interaction of the element attribute and year attribute, and the interaction of the area attribute and water attribute. Independence included the interaction of the element attribute and area attribute, and the interaction of the year attribute and area attribute (Table 5).

The mean values of 2012 and 2016 showed significant differences with those of the other years, and the differences between any two years from 2013 to 2015 were not significant. The yearly decreasing trend was obvious from 2012 to 2016 (Table 6), and the element differences of heavy metals in water were significant, though the spatial differences were not significant.

#### 3.2.2. Spatiotemporal features of heavy metal in sediment

The dependent variable Y was the heavy metal concentrations in sediment. The attribute data of independent variable X included the year attribute, area attribute, water attribute, element attribute and sediment attribute.

The explanatory power of the sediment attribute (q=96%), water attribute (q=80%) and element attribute (q=78%) gradually decreased with respect to the spatiotemporal features of heavy metal concentrations in sediment. The spatiotemporal features of heavy metal concentrations in water and sediment were quite similar. Except for the interactions of the sediment attribute with the others, the other interactions were almost the enhancement of nonlinearity of two factors due to fitting of the  $q(X_1 \cap X_2) > q(X_1) + q(X_2)$  relationship (Table 7).

The mean value of 2012 was significantly higher than that of the other years. Except for 2012, the difference between the other years was not significant. The yearly trend was not obvious (Table 8).

The differences between Chongming and Jinshan, Fengxian, Pudong were all significant. The risk detector showed that the concentrations in sediments of Chongming and Nanhui were much lower than those of other areas, and those of Jinshan, Fengxian and Pudong were much higher. The increasing order of heavy metal concentrations by area was Chongming < Nanhui < Baoshan < Pudong < Fengxian < Jinshan, and there were significant differences among the seven heavy metals (Table 9).

# 3.2.3. Driving factors of biological quality

The dependent variable Y was the heavy metal concentrations in organisms. Attribute data of independent variable X included the water attribute, sediment attribute, element attribute, year attribute and area attribute.

The sediment attribute (q = 59%), water attribute (q = 27%) and element attribute (q = 21%) were the three main driving factors to control the spatiotemporal features of heavy metals in organisms. The effect of sediment was evidently more significant than that of water on the level of heavy metals in the organisms (Table 10).

The enhancement of two factors included the interaction of the sediment attribute and water attribute, and the interaction of the element attribute and water attribute and the interaction of the element attribute and sediment attribute. The other interactions were all the enhancement of nonlinearity of two factors due to fitting of the q  $(X_1 \cap X_2) > q(X_1) + q(X_2)$  relationship (Table 10). In the case of small amounts of biological quality data, the spatiotemporal variation should be further detected.

Table 10

Driving factors and their interaction relationship for heavy metals in organisms.

q	Water attribute	Sediment attribute	Element attribute	Year attribute	Area attribute
Water attribute	0.27				
Sediment attribute	0.74	0.59			
Element attribute	0.28	0.60	0.21		
Year attribute	0.62	0.70	0.35	0.04	
Area attribute	0.63	0.86	0.40	0.21	0.05

#### 4. Discussion

The marine environment needs adequate protection, and heavy metals are among the most serious pollutants in the YRE. Identifying and tracking the transformations and changes of heavy metal in different media is a scientific issue, which requires constant monitoring programmes and new assessment methods. In this paper, the geographical detector was first introduced to research spatiotemporal characteristics and interactions of heavy metals in different media. Using the frequency analysis method and the geographical detector, the raw observations of heavy metal concentrations in water, sediment and organisms showed their patterns, trends and relevance in different media. Heavy metal concentrations in water and sediment exhibited lognormal distribution. The means of heavy metal concentrations decreased yearly in water and sediment. The effect of sediment on biological quality was much greater than that of water. The spatiotemporal stratified heterogeneity and relevance of heavy metals in different environmental media are expected to accurately guide environmental management. Our study offers guidance for interdisciplinary methods to explore mechanisms of the heavy metal transformation from one media to another in the YRE.

From 1984 to 2000, the median concentrations of Hg, Cu, Pb and Cd in water were 35.0 ng/L, 2.75  $\mu$ g/L, 2.22  $\mu$ g/L and 0.078  $\mu$ g/L in the YRE and its adjacent sea, respectively (Wang et al., 2003). The median values of Hg, Cu, Pb and Cd in water were 30.8 ng/L, 1.40  $\mu$ g/L, 0.591  $\mu$ g/L and 0.0329  $\mu$ g/L in this study. The concentration of Hg only slightly changed, while those of Cu, Pb, and Cd obviously decreased. The spatiotemporal feature of heavy metal concentration in water showed that the spatial variation was not significant, though it decreased with time (Yang and Xu, 2015). The mean concentrations of Hg, Cu, Pb, Cd, Cr, Zn and As in sediments from 2012 to 2016 were 0.0649 mg/kg, 19.7 mg/kg, 25.8 mg/kg, 0.130 mg/kg, 34.4 mg/kg, 71.5 mg/kg and 8.80 mg/kg, respectively. Under the guidance of the national action plan for pollution control, it is inevitable that pollutants will fluctuate and decline in the YRE (Fan et al., 2019).

The spatiotemporal feature of heavy metal concentration in sediment showed that Chongming was significantly lower than the other areas and decreased with time. This should benefit from Chongming Island's adherence to the path of ecological priority and green development, and vigorously building a world-class ecological island (Fan et al., 2019). The spatiotemporal stratified heterogeneity of heavy metals in sediments could be explained about 80% by that of water, while driving factors of heavy metals in shellfish organisms were mainly associated with concentrations in sediment and water, and q was 59% and 27%, respectively. The effect of sediment on biological quality was much greater than that of water.

The classes of heavy metal concentrations were based on equal probability intervals of 25%, 50% and 75% for the lognormal distribution. The water attribute had a high explanatory power of 88% for the spatiotemporal characteristics of water quality elements, and the sediment attribute had much higher explanatory power of 96% for the spatiotemporal characteristics of sediments. It showed that the classes of heavy metal elements in water and sediments were reasonable, and the classes of heavy metal concentrations in sediment were more in agreement with their variations than those of heavy metal concentrations in water. The classes of heavy metal concentrations in organisms could not be obtained due to the relatively small amount of biological data. Thus, the spatiotemporal features of heavy metals in organisms were not derived by the geographical detector. When enough data of heavy metals in organisms will be collected, their classifications and the spatiotemporal stratified heterogeneity mechanisms should be further researched. In order to obtain reasonable results with the method of geographical detector, classification is one of the difficulty points, and the classification complemented the spatiotemporal distribution of heavy metal elements (Wang et al., 2010).

The previous study revealed that the changes in sediment quality

were closely related with the anthropogenic activities and economic development (Guo et al., 2019), and the accumulation of heavy metals (Cu, Cr, Pb and Zn) in sediment had increased over the past decades (Guo and Yang, 2016). Although serious environmental pollution was not observed in our study area, the strong enrichment and the relative stability of heavy metals still exhibited slow toxic and side effects on benthic organisms (Chen et al., 2017), and they affected human health through the food chain. Incidental pollution could lead to the unprecedented increase of heavy metal concentrations in marine water or sediment, resulting in loss of ecological habitat and ecosystem disasters (Barletta et al., 2019). In our study area, the runoff of the Yangtze River was a very important source of many pollutants (Chen et al., 2012; Guo et al., 2019), and the other sources of pollution included the accidents of hazardous chemical leakage, sewage discharges from agriculture and aquaculture, drainage from the coastal enterprises and the collision of maritime shipping vessels (Fan et al., 2019). Further research on pollution sources is needed, and pollutant transformations will be significant issues in coastal areas around the world (Barletta et al., 2019).

### Authors' contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

The first author: study concept and design, writing and revised the MS, processing data by the method of Geodetector;

The second author: processing data by the method of classification and distribution, study concept, writing introduction of the MS;

The third author: collecting the heavy metal data in water; The fourth author: collecting the heavy metal data in sediment; The fifth author: collecting the heavy metal data in organisms; The sixth author: supplying some literature information.

## Declaration of competing interest

We declared that we have no conflicts of interest. We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

### Acknowledgments

This work was supported by the National Key Research and Development Program of China (grant numbers 2017YFC1404500, 2016YFC1400806 and 2017YFC1405002).

### References

- Ahumada, I., Rudolph, A., González, E., Fones, G., Saldías, G., Ahumada-Rudolph, R., 2011. Dissolved trace metals in the water column of Reloncaví Fjord, Chile. Lat. Am. J. Aquat. Res. 39, 567–574.
- Barletta, M., Lima, A.R.A., Costa, M.F., 2019. Distribution, sources and consequences of nutrients, persistent organic pollutants, metals and microplastics in South American estuaries. Sci. Total Environ. 651, 1199–1218.
- Bi, S.P., Yang, Y., Xu, C.F., Zhang, Y., Zhang, X.B., Zhang, X.R., 2017. Distribution of heavy metals and environmental assessment of surface sediment of typical estuaries in eastern China. Mar. Pollut. Bull. 121, 357–366.
- Buruaem, L.M., Araujo, G.S., Rosa, P.A., Nicodemo, S.C., Porto, V.F., Fonseca, J.R., Cruz, J.V., Medeiros, G.F., Abessa, D.M.S., 2013. Assessment of sediment toxicity from the Areia Branca off-shore harbor and the Potengi river estuary(RN), northeastern Brazil. Panam. J. Aquat. Sci. 8, 312–326.
- Chen, Y.X., Liu, R.M., Sun, C.C., Zhang, P.P., Feng, C.H., Shen, Z.Y., 2012. Spatial and temporal variations in nitrogen and phosphorous nutrients in the Yangtze River Estuary. Mar. Pollut. Bull. 64, 2083–2089.
- Chen, B., Liu, J., Qiu, J.D., Zhang, X.L., Wang, S., Liu, J.Q., 2017. Spatio-temporal distribution and environmental risk of sedimentary heavy metals in the Yangtze River Estuary and its adjacent areas. Mar. Pollut. Bull. 116, 469–478.
- Cui, Y., Song, Y.L., Chen, B.J., 2000. The assessment on the contents of the heavy metals in organisms along coast of Huanghua. Mar. Fish. Res. 21, 55–60.
- Dong, A.G., Zhai, S.K., Matthias, Z., Yu, Z.H., 2009. The distribution of heavy metal contents in surface sediments of the Changjiang Estuary in China and surrounding coastal areas. Acta Oceanol. Sin. 31, 54–68.

- Du, W., 2018. In: East China Normal University (Ed.), Distribution Characteristics and Environment Risk of Trace Elements in Surface Sediments of the Yangtze Estuary and Adiacent Waters.
- Duan, M., 2012. In: Nankai University (Ed.), Deriving Ecological Criteria Based on the Variation of Plankton Communities: Methodology and Case Study.
- Fan, H.M., Jiang, X.S., Ji, H.H., Liu, P. X., Hu, M.G., Qin, Y.T., 2019. Integrated evaluation of the marine ecological environment in the Yangtze River Estuary and its adjacent area. Acta Ecologica Sinica/Shengtai Xuebao 39, 4660–4675.
- Fang, M., Wu, Y.J., Liu, H., Jia, Y., Zhang, Y., Zhang, X.T., Wu, M.H., Zhang, C.L., 2013. Distribution, sources and ecological risk assessment of heavy metals in sediments of the Yangtze River Estuary. Acta Scien. Circum. 33, 563–569.
- Gomes, F., Godoy, J.M., Godoy, M.L., Lara de Carvalho, Z., Tadeu Lopes, R., Sanchez-Cabeza, J.A., Drude de Lacerda, L., Cesar Wasserman, J., 2009. Metal concentrations, fluxes, inventories and chronologies in sediments from Sepetiba and Ribeira Bays: a comparative study. Mar. Pollut. Bull. 59, 123–133.
- Guo, Y., Yang, S., 2016. Heavy metal enrichments in the Changjiang (Yangtze River) catchment and on the inner shelf of the East China Sea over the last 150 years. Sci. Total Environ. 543, 105–115.
- Guo, X.Y., Feng, C.H., Gu, E., Tian, C.H., Shen, Z.Y., 2019. Spatial distribution, source apportionment and risk assessment of antibiotics in the surface water and sediments of the Yangtze Estuary. Sci. Total Environ. 671, 548–557.
- Han, J.Y., 2019. Study on evolution and driving factors of ecological environment in China's coastal waters. Resou. Develop. & Mar. 35, 1133–1137.
- Han, D.M., Cheng, J.P., Hu, X.F., Jiang, Z.Y., Mo, L., Xu, H., Ma, Y.N., Chen, X.J., Wang, H.L., 2017. Spatial distribution, risk assessment and source identification of heavy metals in sediments of the Yangtze River Estuary, China. Mar. Pollut. Bull. 115, 141–148
- Hu, Y.Y., Wang, J.Y., Zhang, Z.F., Zong, H.M., Chen, S.M., 2011. Approach of nutrient criteria establishment and application in coastal area of Liaohe Estuary. China Environ. Sci. 31, 996–1000.
- Huang, H.J., Ping, X.Y., Li, L., Liao, Y., Shen, X.Q., 2011. Concentrations and assessment of heavy metals in seawater, sediments and aquatic organisms at Yangtze River Estuary in spring and summer. Ecol. Environ. Sci. 20, 898–903.
- Jahan, S., Strezov, V., 2018. Comparison of pollution indices for the assessment of heavy metals in the sediments of seaports of NSW, Australia. Mar. Pollut. Bull. 128, 295–306.
- Kang, L.J., 2012. Research on nutrient criteria establishment in Dianshan lake by the frequency distribution method. Resour. Environ. Yangtze Basin 21, 627–633.
- Li, Y., Feng, Y., Peng, F., Chen, S.D., 2017. Pattern evolvement of ecological land in Tianjin based on geodetector. Econ. Geogr. 37, 180–189.
- Ministry of Environmental Protection, 2009. People's Republic of China. HJ442–2008 the Specification for Offshore Environmental Monitoring. Environmental Science Press of China. Beijing.
- Office of Water, Office of Science and Technology, 2011, Nutrient Criteria Technical Guidance Manual Estuarine and Coastal Marine Waters (EPA-822-B-01-003). Retrieved from <a href="http://www.epa.gov/nutrient-policy-data/criteria-development-guidance-estuarine-and-coastal-waters">http://www.epa.gov/nutrient-policy-data/criteria-development-guidance-estuarine-and-coastal-waters</a>.
- Qiu, Y.W., Yan, W., Wang, Z.D., Zhang, G., 2005. Distributions of heavy metals in seawater, sediments and organisms at Daya bay and their ecological harm. J. Trop. Oceanogr./Redai Haiyang Xuebao 24, 69–76.

- State Oceanic Administration, China, 2013-2017. East China Sea marine environmental bulletin. Retrieved from. http://ecs.mnr.gov.cn/.
- State Technical Supervise Bureau, People's Republic of China, 2008. GB17378–2007 the Specification for Marine Monitoring. Standards Press of China, Beijing.
- Sun, Y., Mu, Q.L., She, Y.Y., Wang, Y.H., Wang, J., 2011. Distribution patterns of heavy metals in sediments of the Yangtze estuary and its adjacent areas and ecological risk assessment. J. Zhanjiang Ocean Univ. 30, 107–112 Nat. Sci.).
- Teng, D.Q., Lv, S.H., Guo, F.X., Jiang, T., 2012. Distribution pattern of heavy matals in surface sediments of the Yangtze River Estuary and adjacent areas and its ecological risk. Mar. Geol. & Quatern. Geol. 32.
- Vallius, H., 2015. Quality of the surface sediments of the northern coast of the Gulf of Finland, Baltic Sea. Mar. Pollut. Bull. 99, 250–255.
- Wang, H.B., 2017. The Study on the Distribution Characteristics and Controlling Factors of Particulate Metal Elements in the Changjiang Estuary. East China Normal University
- Wang, J.F., Xu, C.D., 2017. Geodetector: principle and prospective. Acta Geogr. Sin./Dili Xuebao 72, 116–134.
- Wang, B.S., Liu, A.C., Chen, Z.Y., 2003. Distribution changes of dissolved heavy metal concentration in Changjiang estuary and its adjacent Waters, 1984–2000. Mar. Sci. Bull. 22, 32–38.
- Wang, J.F., Li, X.H., Christakos, G., Liao, Y.L., Zhang, T., Gu, X., Zheng, X.Y., 2010.
  Geographical detectors-based health risk assessment and its application in the neural tube defects study of the Heshun region, China. Int. J. Geogr. Inf. Sci. 24, 107–127.
- Wang, J.W., Liu, R.M., Zhang, P.P., Yu, W.W., Shen, Z.Y., Feng, C.H., 2014. Spatial variation, environmental assessment and source identification of heavy metals in sediments of the Yangtze River Estuary. Mar. Pollut. Bull. 87, 364–373.
- Wang, H.T., Wang, J.W., Liu, R.M., Yu, W.W., Shen, Z.Y., 2015. Spatial variation, environmental risk and biological hazard assessment of heavy metals in surface sediments of the Yangtze River Estuary. Mar. Pollut. Bull. 93, 250–258.
- Wang, J.F., Zhang, T.L., Fu, B.J., 2016. A measure of spatial stratified heterogeneity. Ecol.
- Yang, F.X., 2015. Approaches of Nutrient Criteria Establishment and its Influencing Factors in the Daliaohe Estuary. Ocean University of China.
- Yang, Y., Xu, R., 2015. The environment variation trend in the Changjiang River Estuary in the past 30a. Mar. Sci. 39, 101–107.
- Yu, S., Hong, B., Ma, J., Chen, Y.S., Xi, X.P., Gao, J.B., Hu, X.Q., Xu, X.R., Sun, Y.X., 2017. Surface sediment quality relative to port activities: a contaminant-spectrum assessment. Sci. Total Environ. 596–597, 342–350.
- Zhang, X.L., 2013. In: Ocean University of China (Ed.), Distribution Characteristics and Controlling Factors of Heavy Metals in the Yangtze River Estuary, the Yellow River Estuary and Adjacent Sea.
- Zhang, L.X., Jiang, X.S., Zhao, M., Li, Z.E., 2007. Pollution of surface sediments and its assessment of potential ecological risk in the Yangtze Estuary. Ecol. Environ. Sci. 16, 389–393.
- Zhang, B., Wang, F., Cheng, L.J., Wang, X., Zhang, D.S., Zhao, Y., Li, X.G., Chen, S., Wang, Q.L., 2017. Observation data processing method and product development of key parameters of marine environmental change. China Basic Sci 5, 6–11.
- Zheng, B.H., Zhu, Y.Z., Liu, L.S., Zhou, J., Lin, K.X., 2013. The reference condition for Eutrophication Indictor in the Yangtze River Estuary and adjacent waters response variables. Acta Ecol. Sin./Shengtai Xuebao 33, 2768–2779.