Marine Pollution Bulletin 174 (2022) 113278

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

# Distribution, pollution, and human health risks of persistent and potentially toxic elements in the sediments around Hainan Island, China

Zhiwei Che [a,1], Waqas Ahmed [b,c,1], Jiechang Weng [d], Liu Wenjie [e], Mohsin Mahmood [b,c], Juha M. Alatalo [f], Ou Wenjie [b,c], Mir Muhammad Nizamani [g], Wang Lu [b,c], Fu Xiu Xian [b,c], Yang Jie [b,c], Wang Yunting [b,c], Weidong Li [b,c,*], Sajid Mehmood [b,c,*]

[a] *Haikou Marine Environmental Monitoring Center, State Oceanic Administration, Haikou 570000, Hainan, China*
[b] *Key Laboratory of Agro-Forestry Environmental Processes and Ecological Regulation of Hainan Province (Hainan University), Haikou 570228, China*
[c] *College of Ecology and Environment, Hainan University, Haikou City, 570100, P.R China*
[d] *Hainan Provincial Ecological and Environmental Monitoring Center, 571126, China*
[e] *College of Ecology and Environment, Hainan University, Haikou 570228, China*
[f] *Environmental Science Center, Qatar University, Doha, Qatar*
[g] *Key Laboratory of Tropical Biological Resources of Ministry of Education, College of Tropical Crops, Hainan University, Haikou, China*

## ARTICLE INFO

*Keywords:*
Pollution indices
Potentially toxic elements
Source identification
Sediments
Human health risk assessment

## ABSTRACT

Human activities have changed the global concentration of potentially toxic elements (PTEs) and significantly altered the marine ecosystem. Little is known about the concentrations of these PTEs around Hainan Island in China, or their distribution and human health risks. Understanding the variability of PTEs in marine sediments and how they accumulate is important not only for biodiversity and ecological conservation, but also for management of aquatic natural resources and human health risk assessments. This study showed that the concentrations of six PTEs (Cd, Cu, Zn, As, Pb, and Hg), sampled in nine different cities, were linked to human activities. In order to understand the ecological risks associated with PTE pollution, we calculated the contamination factor (CF), enrichment factor (EF), pollution load index (PLI), and geo-accumulation index (Igeo) of each element in each city. These indicators suggest that the pollution of Cd and Zn in the sediments of these cities is higher than that of the other PTEs. We also carried out a human health risk assessment which demonstrated the carcinogenic effects of Zn on children and adults in ChengMai, while Pb showed non-carcinogenic effects at all the studied sites, suggesting that Zn pollution in the sediments of ChengMai may pose human health risks. We would therefore advise that follow-up studies endeavor to monitor the levels of PTEs in the flora and fauna of these cities.

## 1. Introduction

Advances in agricultural technology and industry development near coastal areas have brought about a significant increase in potential risks to human health, on account of increased levels of potentially toxic elements (PTEs) (Shaheen et al., 2020; Xu et al., 2017). Industrial development is normally associated with the release of PTEs into aquatic environments (Amin et al., 2020; Arisekar et al., 2020), and the risks posed by PTEs to these environments are inherently related to their interactions with water and sediments. In recent years, shipping and industrialization have come to be considered the main sources of Cd, Co, Cr, Cu, Ti and Zn in surface sediments (Zhuang et al., 2019), while Cd is understood to be primarily transferred through agricultural production (Khusnidinov et al., 2020), industrial effluents and smelting slags, owing to rough smelting processes and inefficient recycling methods (Wang et al., 2019). The adsorption and desorption reactions of water and sediments cause PTEs to bind on sediments surfaces, interactions which are related to the mineral content, surface area and cation-exchange capacity of the sediments. Surface sediments are rich in organic (living or dead flora and fauna) and inorganic (soil particles from bedrock) materials (Hasimuna et al., 2021). They are also home to many benthic organisms and serve as a food source for some species of fish

---

* Corresponding authors.
*E-mail addresses:* weidongli@hainanu.edu.cn (W. Li), drsajid@hainanu.edu.cn (S. Mehmood).

[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.marpolbul.2021.113278
Received 13 October 2021; Received in revised form 11 December 2021; Accepted 16 December 2021

(catfish, common carp) (Hasimuna et al., 2021). Due to their continuous biological toxicity, PTEs in sediments have a direct effect on benthic organisms and a negative impact on human health (Li et al., 2021), with diseases such as cancer, anemia, diabetes and osteomalacia being attributed to human exposure to PTEs (Mohammadi et al., 2018). The main routes by which humans can be exposed to PTEs are water intake, seafood, and skin contact while swimming (Isa et al., 2015). It is recommended to reduce the intake of tilapia fish because of the pollution of PTEs such as copper, nickel, and chromium in riverside wastewater sources (Tayebi and Sobhanardakani, 2020).

Aquaculture sedimentation is essential for outlining the spatial distribution of PTEs (Li et al., 2019a, 2019b), as it can bring about the reverse reaction of the release of toxic elements into aquatic organisms and ultimately affect human health (H. Wang et al., 2015). The Chinese seafood market is developing rapidly due to the country's vast territory and abundant natural resources, and the lifestyles and diets of residents vary significantly between different regions (Zhang et al., 2021). The report also found that fish consumption in China has increased by 50% in the last decade and is expected to see further increases, from 44 kg per capita to 50 kg. According to Liu et al. (2019a, 2019b) consumption of aquatic organisms which have been contaminated by PTEs can adversely affect human health, meaning that spatial distribution of PTEs has a vital role to play in establishing the degree and source of pollution (Cai et al., 2021; H. Zhang et al., 2019). PTEs like zinc (Zn), lead (Pb), copper (Cu), cadmium (Cd), mercury (Hg), and arsenic (As) in marine sediments are generally considered to be indicators of potential environmental hazards (Khan et al., 2017).

Hainan Island, located in the northwestern part of the South China Sea (Cai et al., 2021), is a major tourist hub (Zhao et al., 2020). Fish tend to prefer strong upwelling regions (Cai et al., 2021), and Hainan accounts for 0.1% of all ocean upwelling regions worldwide (Ryther, 1969). The recent surge in coastal urbanization on Hainan Island has seen an increase in wastewater discharge, agricultural development, and human activities, which have polluted Hainan Island to a serious degree (Li et al., 2014; Xu et al., 2018; Zhang et al., 2017). These increases are expected to worsen the contamination of marine sediments, which will have a direct negative impact on aquatic organisms. Minimal literature is available on the distribution and source of PTEs on Hainan Island (Hu et al., 2013; Zhao et al., 2020), but given its population increase and surge in agricultural development in recent years, investigation into PTE pollution around the Island is going to be necessary if the environmental health of the aquaculture is to be maintained (Zhao et al., 2020). Indeed, sediment pollution poses serious risks due to its toxicity, lack of biodegradability, and food chain accumulation characteristics (Mennillo et al., 2020; Vitali et al., 2019). Despite impressive gains, the aquaculture sector still faces serious challenges that, in some cases, undermine its ability to achieve sustainable outcomes (Naylor et al., 2021).

While there is research available regarding the offshore of Hainan Island (Cai et al., 2021), there are limited published studies on PTE pollution in the marine sediments of the Island's cities. Previous studies on the spatial distribution of PTEs only reflected a narrow area and a limited number of contaminants and did not provide a comprehensive evaluation of PTEs around the Island (Ge et al., 2018; Lu et al., 2021; D. Wang et al., 2015; Zang et al., 2021). Our study covered nine different Island locations (Lingao, Haikou, Qionghai, Sanya, Wenchang, Lingshui, Dongfang, ChengMai, and Ledong), and aimed to assess the distribution of six (6) PTEs in the marine sediments of nine (9) different cities, thereby establishing the risk these PTEs may pose to the health of the residents. The areas studied are significant in terms of agriculture and fish production, so the present study assumes that any pollution taking place there, be it minor or extreme, will have an overall bearing on the Island.

## 2. Materials and methods

### 2.1. Study areas

Hainan Island (19.5664°N, 109.9497°E) is located at the southernmost tip of China (Fig. 1). It is China's second largest island, with an offshore area of approximately 2 million km$^2$, accounting for 42.3% of the country's sea area. The total coastline length of Hainan Province is 1928 km, of which the coastline of Hainan Island is 1618 km. The province's annual average temperature (T) is 25.1 °C, and the annual precipitation is 2176.7 mm (Zhang et al., 2019a, 2019b), although it may vary from area to area. Hainan Island has a tropical humid monsoon climate, with mountains in the center and hills and alluvial plains along the coast (Zhang et al., 2019a, 2019b).

### 2.2. Site selection and surface sediment collection

The area under consideration covers a total distance of 555 km around Hainan Island and a total of nine marine sediment sample survey stations. A total of 285 samples were collected from all survey sites. The specific locations of these areas, as well as sampling points, are shown in Fig. 1, and the locations of the survey samples are shown in Table S1. A manual auger was used to collect surface sediment samples from designated sampling points at different locations in triplicate (300 g each). After each collection, the auger was cleaned thoroughly to avoid impurities. These samples were collected from a depth of 20 cm (Zhu et al., 2018).

Based on their topography and physical characteristics (such as non-rock and sand), three locations were selected for sampling in each of the nine cities. Further criteria for choosing the cities and sampling locations included the relative waste generated and injected into the ocean, posing a danger to aquatic life and, ultimately, to humans. Lingao is the administrative area of Hainan Island's tourist attractions and considered China's largest cage breeding base. Haikou is the capital of the Island, a famous port city located on the northern coast of Hainan Province. Qionghai is located in the center of China, at the southern tip of Hainan Island, and is perhaps the most sparsely populated province in the country. Sanya, which is famous for its tropical climate, has become a popular tourist destination. Wenchang is located in the northeast of the Island, and is dominated by mountains and hills, combined with forest and agricultural areas. Lingshui, in the southeast of the Island, is famous for its tourism and marine and mineral resources. Dongfeng, in the southwest, has a vast land area, while ChengMai, also in the southwest, has a pleasant climate and has become attractive to tourists. Ledong, another city in the southwest, is known for its dense tropical rainforests and beautiful natural scenery. Due to variability in morphology and physical characteristics, the distance between stations was not equal.

### 2.3. Sediment preparation

The wet sediment samples were air-dried at room temperature in the laboratory of Hainan University. After that, the sample was ground using a mortar and pestle and sieved through a 2 mm sieve. Dried samples were then tightly packed in a polyethylene bag for storage, until the potentially toxic elements (PTEs) could be determined.

### 2.4. Chemical analysis of collected sediments

The analysis method was implemented in accordance with the "Marine Monitoring Regulations" (GB 17378.5-2007), and the quality assurance in accordance with the "Marine Monitoring Regulations" promulgated by the state (Zhao et al., 2019). PTEs such as Cd, Cu, Zn, As, Pb, and Hg were determined from the marine sediment samples collected (Table 1). The monitoring and analysis methods used in the investigation are shown in Table S2. The method for analyzing these PTEs from the sediment proceeded as follows: 1 g of a finely ground

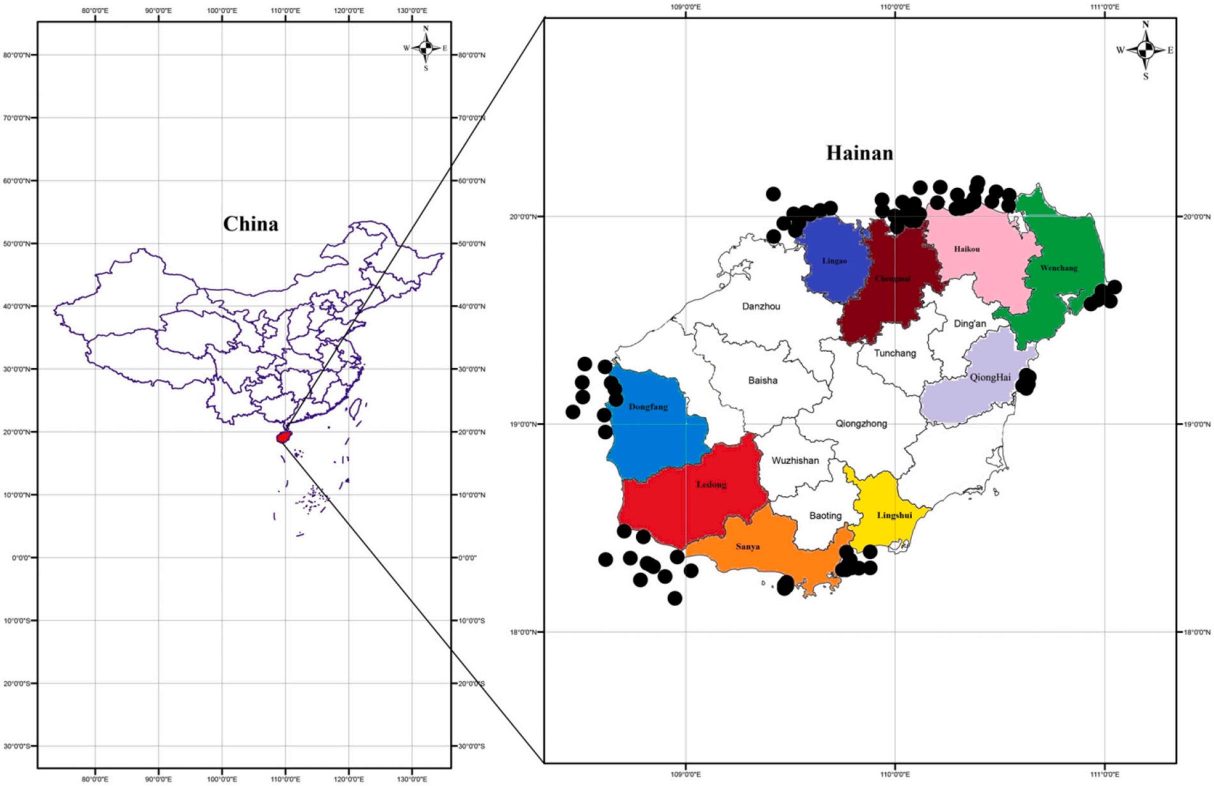

**Fig. 1.** Geographic location and geological map of Hainan Island, with locations of sampling sites. Black dots on the map represent sampling locations. Note that some of the sampling points overlap.

**Table 1**
Statistics and mean concentration of heavy metal (mg/kg) in nine (9) different cities around Hainan, Island.

| PTE | ChengMai | DongFang | Haikou | Ledong | Lingao | LingShui | QiongHai | Sanya | Wenchang |
|---|---|---|---|---|---|---|---|---|---|
| Zn | 43.9462a ± 20.679 | 0.232b ± 0.124 | 0.5363b ± 0.49913 | 0.325b ± 0.082 | 0.403b ± 0.148 | 0.352b ± 0.149 | 0.121b ± 0.096 | 0.311b ± 0.199 | 0.058b ± 0.013 |
| Pb | 19.2677a ± 4.434 | 0.309b ± 0.211 | 0.2995b ± 0.15686 | 0.271b ± 0.054 | 0.533b ± 0.219 | 0.281b ± 0.065 | 0.075b ± 0.041 | 0.142b ± 0.073 | 0.520 ± 0.077 |
| Cu | 12.9562a ± 5.489 | 0.284b ± 0.214 | 0.3232b ± 0.26729 | 0.205b ± 0.053 | 0.334b ± 0.141 | 0.311b ± 0.155 | 0.137b ± 0.112 | 0.208b ± 0.058 | 0.075b ± 0.025 |
| Cd | 0.2444cd ± 0.134 | 0.097c ± 0.022 | 0.3747b ± 0.29802 | 0.125c ± 0.098 | 0.743a ± 0.274 | 0.403b ± 0.123 | 0.117c ± 0.050 | 0.237 cd ± 0.099 | 0.190c ± 0.056 |
| Hg | 0.0344c ± 0.010 | 0.152bc ± 0.050 | 0.3311a ± 0.40770 | 0.049c ± 0.022 | 0.237ab ± 0.057 | 0.115bc ± 0.055 | 0.101bc ± 0.033 | 0.058bc ± 0.032 | 0.056bc ± 0.005 |
| As | 15.0338a ± 7.760 | 0.649b ± 0.213 | 0.2895b ± 0.12136 | 0.378b ± 0.151 | 0.606b ± 0.147 | 0.416b ± 0.237 | 0.586b ± 0.127 | 0.273b ± 0.118 | 0.616b ± 0.161 |

sediment sample was placed in a Teflon crucible, and a few drops of deionized water added to wet the sample. After that, 1 ml of $HNO_3$ (65% *W/V*) and HF (40% W/V) was added to the wet sediment sample, before it was heated at 150 °C. After heating, a further 1 ml $HNO_3$ was added, and the sample subjected to heat once more, until dry. 3 ml $HNO_3$ (30%) was then added to the dry sample, which was heated at 120 °C. The digested sediment samples were washed with a 0.1 M $HNO_3$ solution and made to 100 ml volume using deionized water (Duncan et al., 2018). After carrying out this procedure for all the sediment samples, we ran Atomic Absorption Spectrophotometer (AAS: model An Analyst 500, PerkinElmer Inc., Shelton, USA) to determine the concentration of six potentially toxic elements (PTEs) (Cd, Cu, Zn, As, Pb, and Hg). Before running AAS for PTE analysis, it was calibrated and standardized according to appropriate standards (Manoj and Padhy, 2014). The results given for each sediment sample represent the average of three readings. A calibration curve of absorbance and concentration was drawn for each element, and the concentration of each of the six PTEs determined from the calibration map by interpolation. Standard samples were run after every 10 samples. Throughout the course of the research process, all reagents used were of analytical grade.

## *2.5. Pollution indices used in the assessment of surface sediments*

Various indicators have been used to measure the pollution level of aquatic ecosystems. These include contamination factor (CF), enrichment factor (EF), geo-accumulation index (Igeo) and pollution load index (PLI) (Enuneku et al., 2018; Hasimuna et al., 2021). Each of these indices has advantages and disadvantages, and the specific index used tends to depend on the user and the amount of information available to them. Some indices, for example, might require certain information, such as background data pertaining to elements before any major human activities began to occur. In the absence of such data, then, the user would have to select a different pollution index for which the data is not required. When estimating sediment pollution, the CF EF, Igeo and PLI of each of the PTEs was calculated, which provided general information about specific elements and sediment pollution at a given research site

(Manoj and Padhy, 2014).

### 2.5.1. Geo-accumulation index (Igeo)

The geo-accumulation index (Igeo), proposed by Müller (1969), is used to calculate the quantitative measure of the degree of PTE pollution in the collected sediment samples, as shown in Table 2. The geo-accumulation of PTEs was calculated using Eq. (1).

$$Igeo = log2\left(\frac{Cn}{1.5\ Bn}\right) \tag{1}$$

where Cn is the total concentration of the PTE measured in the collected sediment samples, Bn is the average background concentration of the PTE, and 1.5 is the factor (correction factor) that compensates for the background data. Igeo of less than 0 signifies practically no pollution, 0 < Igeo < 1 signifies non-pollution to moderate pollution, 2 < Igeo < 3 signifies moderate to severe pollution, 3 < Igeo < 4 signifies severe pollution, 4 < Igeo < 5 signifies strong to extreme pollution, and Igeo > 5 signifies extreme pollution (Loska et al., 2003).

### 2.5.2. Contamination factor (CF)

Contamination factor (CF) is a measure of the concentration of heavy metals relative to the metals' background value.

$$CF = \frac{PTEs\ in\ the\ collected\ sediments}{Background\ values\ of\ the\ PTEs} \tag{2}$$

Hakanson (1980) proposed that CF < 1 suggests low pollution, 1 < CF < 3 suggests moderate pollution, 3 < CF < 6 suggests a considerable degree of pollution, and CF > 6 suggests a very high degree of pollution.

### 2.5.3. Enrichment factor (EF)

Enrichment factor (EF) assesses contamination in environmental media by following Eq. (3).

$$EF = \frac{\left(\frac{C_i}{C_s}\right)sediments}{\left(\frac{C_i}{C_s}\right)background} \tag{3}$$

where (Ci/Cs) sediments are the ratio of elements in the sediment sample to the reference element, and (Ci/Cs) background is the ratio of the corresponding element in the reference background. An element is regarded as a reference element if it is of low occurrence variability and is present in the element in trace amounts. It is also possible to apply an element of a geochemical nature which occurs in substantial quantities in the environment but has no characteristic effects i.e., synergism or antagonism towards an examined element. The categories of contamination are determined according to the enrichment factor:

EF < 2 indicates a lack of enrichment or only minimum enrichment, EF = 2–5 indicates moderate enrichment, EF = 5–20 indicates severe enrichment, EF = 20–40 indicates very high enrichment, and EF > 40 indicates extremely high enrichment (Merciai et al., 2014). On account of its general formula, the enrichment factor is a relatively simple and easy tool for evaluating the degree of enrichment and comparing different instances of environmental pollution.

**Table 2**
Geo-accumulation index (Igeo) of heavy metals in nine (9) different cities around Hainan Island.

| City | Zn | Pb | Cu | Cd | Hg | As |
|---|---|---|---|---|---|---|
| | Igeo | Igeo | Igeo | Igeo | Igeo | Igeo |
| Ledong | 0.005 ± 0.001 | 0.007 ± 0.001 | 0.022 ± 0.006 | 0.208 ± 0.165 | 0.193 ± 0.089 | 0.188 ± 0.075 |
| ChengMai | 0.659 ± 0.310 | 0.527 ± 0.121 | 1.416 ± 0.600 | 0.282 ± 0.268 | 0.135 ± 0.042 | 7.480 ± 3.861 |
| DongFang | 0.003 ± 0.002 | 0.008 ± 0.006 | 0.028 ± 0.024 | 0.130 ± 0.076 | 0.596 ± 0.200 | 0.323 ± 0.106 |
| LingShui | 0.005 ± 0.002 | 0.008 ± 0.002 | 0.034 ± 0.017 | 0.672 ± 0.206 | 0.451 ± 0.216 | 0.207 ± 0.118 |
| Wenchang | 0.001 ± 0.000 | 0.014 ± 0.002 | 0.008 ± 0.003 | 0.317 ± 0.094 | 0.222 ± 0.020 | 0.307 ± 0.081 |
| Sanya | 0.004 ± 0.003 | 0.004 ± 0.002 | 0.023 ± 0.006 | 0.396 ± 0.167 | 0.230 ± 0.003 | 0.136 ± 0.003 |
| QingHai | 0.002 ± 0.001 | 0.002 ± 0.001 | 0.015 ± 0.012 | 0.196 ± 0.084 | 0.400 ± 0.132 | 0.292 ± 0.063 |
| Haikou | 0.008 ± 0.007 | 0.008 ± 0.004 | 0.008 ± 0.029 | 0.008 ± 0.497 | 0.008 ± 1.599 | 0.008 ± 0.060 |
| Lingao | 0.006 ± 0.002 | 0.015 ± 0.006 | 0.037 ± 0.015 | 1.238 ± 0.457 | 0.929 ± 0.226 | 0.301 ± 0.074 |

### 2.5.4. Pollution load index (PLI)

Pollution load index (PLI) represents the number of times the metal content in the sediment exceeds the background concentration, and offers evidence for the quality of the aquatic environment and valuable information about its pollution status (Guan et al., 2018). In addition, PLI provides comprehensive information about the toxicity of metals in specific samples (Enuneku et al., 2018). The pollution load index (PLI) is defined as the nth root of the product of concentration (Enuneku et al., 2018). If the PLI value is greater than 1 (>1), it means that the sediment is contaminated; if the PLI value is less than 1 (<1), it means that there is little to no pollution (Barakat et al., 2012). The present study used the following formula, proposed by Tomlinson et al. (1980), to evaluate PLI.

$$PLI = (CF1 \times CF2 \times CF3 \times - - - \times CFn)^{1/n} \tag{4}$$

where n is the number of metals (six in the present study), and CF is the contamination factor.

## 2.6. Human health risk assessment

The potential health risk posed by PTEs to human beings was calculated by assessing both the concentrations of the PTEs in sediments and the routes through which humans might be exposed to them.

### 2.6.1. Carcinogenic and non-carcinogenic risks

The cumulative carcinogenic and non-carcinogenic risks of all PTEs were calculated using total carcinogenic risk (TCR) and total hazard index (THI), as shown in algorithms (5) and (6). All other related parameters, such as lifetime cancer incidence risk index (CR), average daily dose (ADD), and hazard quotient (HQ), are detailed in supplementary information, and calculated using Eqs. S1, S2 and S3, respectively.

$$\mathrm{TCR} = \sum_{\mathrm{i=1}}^{6} \mathrm{CRi} - - - - \tag{5}$$

$$\mathrm{THI} = \sum_{\mathrm{i=1}}^{6} \mathrm{HQi} - - - - \tag{6}$$

Cumulative risk refers to the sum of the estimated risks, based on the pollution levels of the six PTEs under consideration. If the HQi (see supplementary information) and HI values are less than or equal to 1, there are no adverse non-carcinogenic effects on human health. If the HQi and THIi values are greater than 1, the PTE may have a negative impact on human health (Alipour et al., 2015). If the values of TCR are less than or equal to $10^{-6}$, then the risk of carcinogenesis is negligible; if the value is greater than $10^{-4}$, it is unacceptable; and if the value is between $10^{-6}$ and $10^{-4}$, it is acceptable (Rinklebe et al., 2019).

## 2.7. Statistical analysis

Statistical analyses and data visualization were performed using Microsoft Excel 2019 software, ArcMap 10.5, and OriginPro 9.1. The means for the variables were tested using one-way ANOVA with using LSD test at a significance level of $p < 0.05$. OriginPro 9.1 b215

(OriginLab Corporation, Northampton, USA) was used to create the figures. Figs. 1–5 were drawn on ArcMap 10.5, and Fig. 6 was drawn on R software using package corrplot.

## 3. Results

The study found that all six of the selected PTEs were present in every sample that was collected, although concentrations varied for different areas. The highest recorded concentrations of Cu, Pb, Zn and As were in ChengMai, which demonstrated a statistically significant difference compared with the eight other cities. The other cities, meanwhile, demonstrated a statistically non-significant difference when compared with each other. Wenchang recorded the lowest levels of Zn, but the second highest of As. Hg had the lowest concentration at all the sites, with the highest being 0.33 mg/kg, in Haikou. In general, contamination levels followed this order: Zn > Pb > As > Cu. In terms of Cd, Sanya and ChengMai had the lowest concentrations, while Lingao had the highest. Lingshui, QiongHai, Wenchang, and Dongfang, on the other hand, had lower concentrations of all the elements, which could be attributed to fewer anthropogenic activities on account of their lower population densities (Fig. 2).

### 3.1. Analytical technique and accuracy check

All treatments and experiments were performed in triplicates. Quality control of the extraction efficiency of the metal concentrations was performed using certified reference samples. The average recovery percentages were 94.60% for Cd, 95.85% for Cu, 108.33% for Zn, 99.60% for As, 96.60% for Pb and 103.7% for Hg. Also, blank and triplicate measurements were employed for analyses. Standard solutions (Merck) were routinely used to guarantee high-quality results. The maximum relative standard deviation (RSD) between replicates was set to 10%. An atomic absorption spectrophotometer (AAS: Analyst 500, PerkinElmer Inc., Shelton, USA) was used for the determination of potential toxic elements (PTE). All reagents used are of analytical grade, and ultra-pure deionized water is used for all analyses. All glassware is cleaned by immersing them in a warm 5% (*V*/V) aqueous solution of nitric acid for 6–7 h, and then rinsed with ultra-pure deionized water.

### 3.2. Evaluation of potential ecological risks

In this study we used contamination factor (CF), enrichment factor (EF), and pollution load index (PLI) to ascertain the health of the flora and fauna and, ultimately, the health of human beings (refer to Figs. 3–5). Fig. 3 shows that the surface sediments in ChengMai had a high degree of As contamination (CF > 6). The results for Zn, meanwhile, indicated a low degree of contamination (CF < 1) for all the cities. The CF for Cd at Lingao showed a very high degree of contamination (CF > 1), and the results for Hg implied a low degree of contamination (CF < 1) for all cities except Haikou and Lingao, which exhibited high degrees of contamination (CF > 1). Meanwhile, the results for Cu indicated a low degree of contamination (CF < 1) for all cities except ChengMai, which had a high degree of contamination (1 < CF < 3). Overall, the CF of all the metals showed a low degree of contamination (CF < 1) at all the sites in each city, except for Cd in Lingao, Cu and As in ChengMai, and Hg in Haikou and Lingao.

The EF of Cd was more than 40 in Lingao, which showed extremely high enrichment of Cd in this area, and 20–40 in Lingshui and Haikou, also suggesting high enrichment. All the other cities saw only moderate enrichment of Cd. Apart from ChengMai, all cities had deficient or only minimum enrichment of Cu, As, Zn and Pb, while ChengMai saw extremely high, high, severe, and minimal enrichment of As, Cu, Pb and Zn, respectively. Enrichment of Hg, meanwhile, was relatively high at all the sampling sites, with the lowest value calculated in ChengMai (8.09), and the highest in Haikou (77.89), suggesting that all the sampled cities around Hainan Island are contaminated by Hg metal.

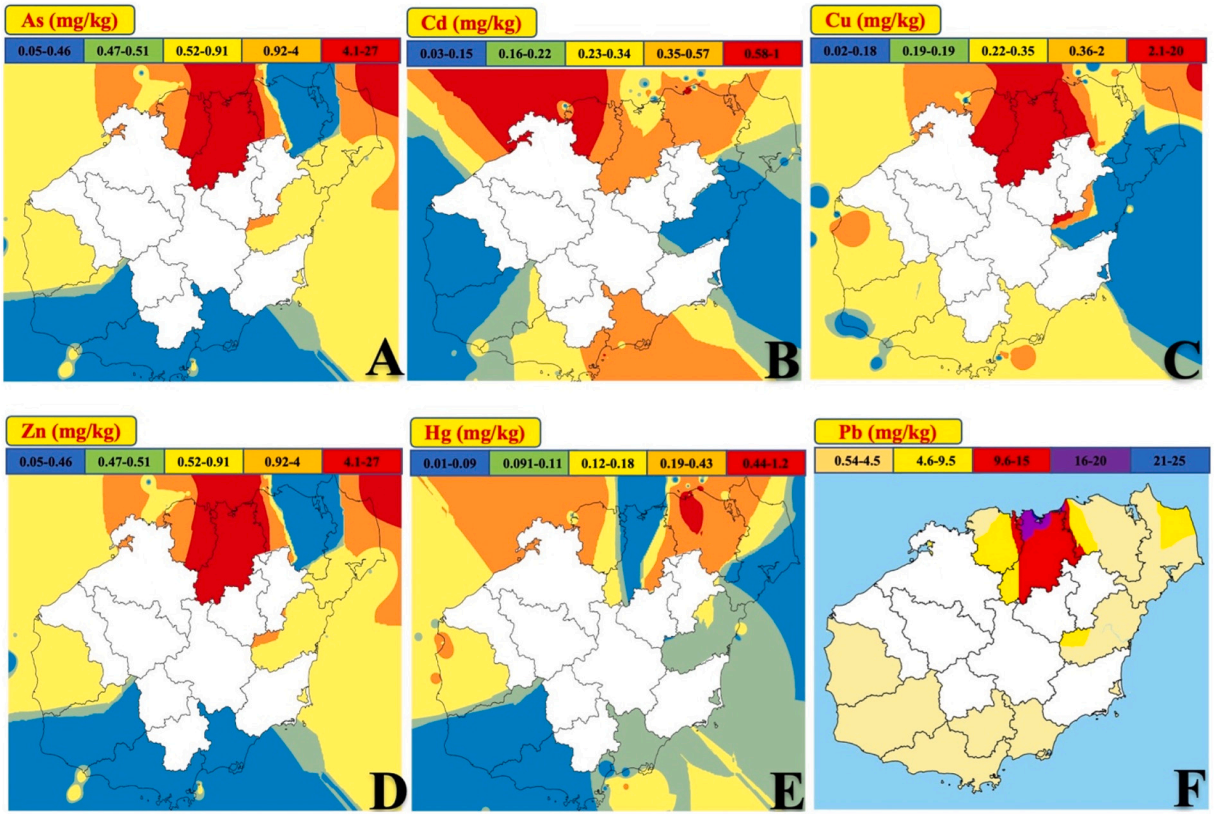

**Fig. 2.** Spatial distribution of potentially toxic elements (PTEs) (mg/kg) in the sediments collected from 9 different cities around Hainan Island.

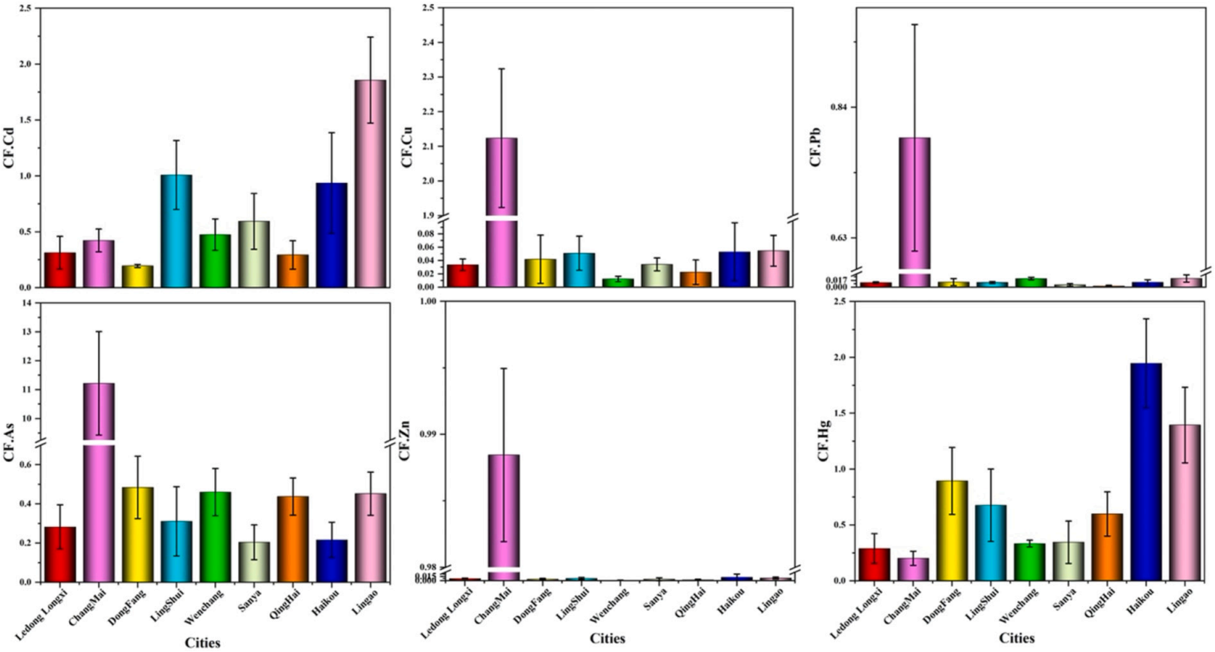

**Fig. 3.** Contamination factor (CF) of potentially toxic elements (PTEs) (mg/kg) in the sediments collected from 9 different cities around Hainan Island.

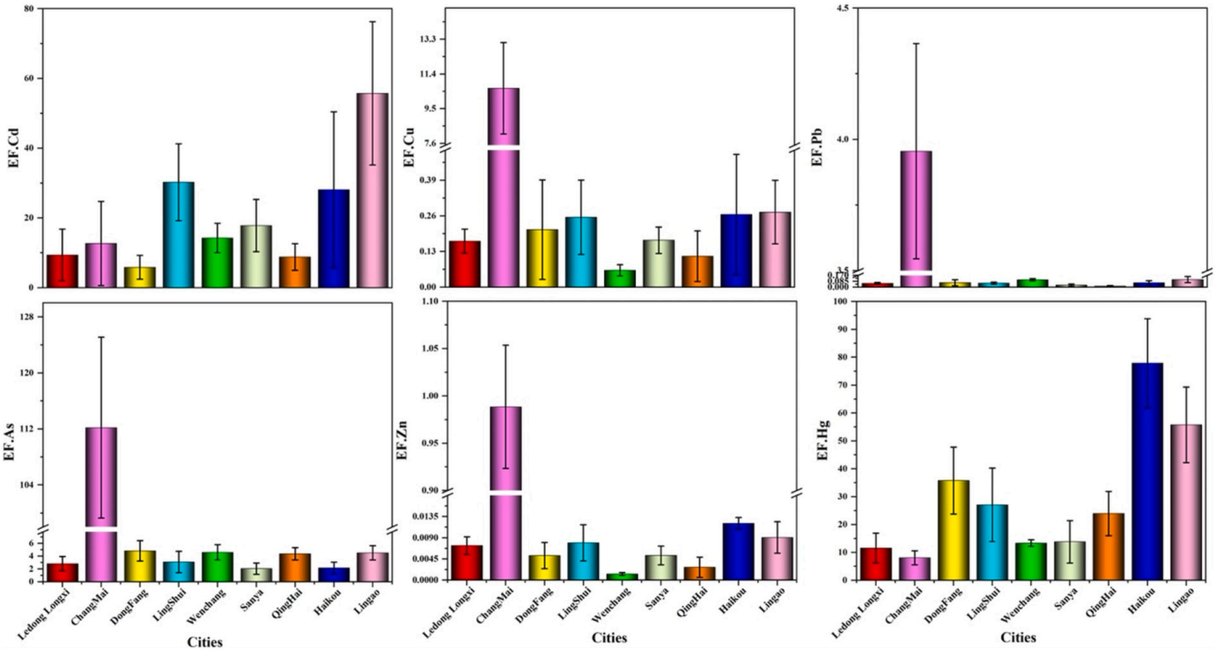

**Fig. 4.** Enrichment factor (EF) of potentially toxic elements (PTEs) (mg/kg) in the sediments collected from 9 different cities around Hainan Island.

The pollution load index (PLI) for Haikou, Lingao and ChengMai indicated heavy metal pollution in these areas. PLI for Ledong, Dongfang, Lingshui, Sanya, Wechang, and QiongHai, on the other hand, indicated moderate degrees of contamination or none at all (Fig. 5), suggesting they were less polluted than others in the province.

In addition to being possible secondary sources of contaminants, sediments usually act as sinks to water columns or organisms in aquatic environments (Lee et al., 2021), so their pollution has the potential to seriously degrade aquatic ecosystems. Consequently, it is of vital importance to understand the sources of these pollutants, as well as how they behave and to what extent, in order to better establish pollution control measures. The correlations between the heavy metals analyzed in the sediment are shown in Fig. 6, where dark blue dots show a strong positive correlation between the parameters while the red dots indicate a strong negative correlation. The lighter reds and blues show less positive and negative correlations. For example, Pb shows strong positive correlations with As, Zn, and Cu, and less positive correlations with all the other research parameters.

### 3.3. *Human health risk assessment*

In the present study, non-carcinogenicity of PTEs to humans was determined by Total Hazard Index (THI) (Table 3), while the potential carcinogenicity of PTEs to humans was determined by Total Cancer Risk

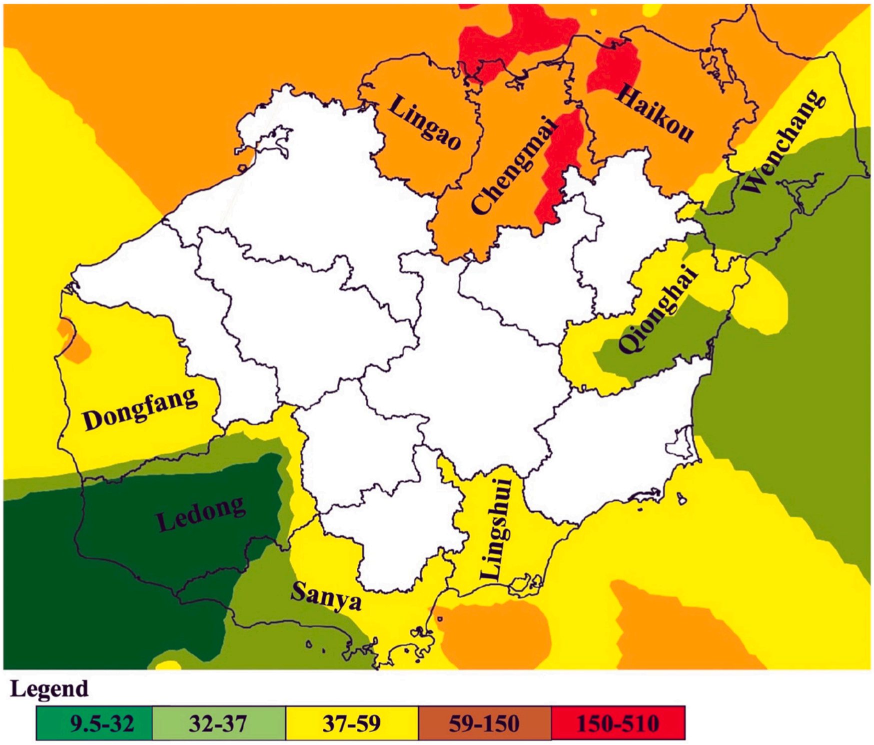

**Fig. 5.** Pollution load index (PLI) of potentially toxic elements (PTEs) (mg/kg) in the sediments collected from 9 different cities around Hainan Island.

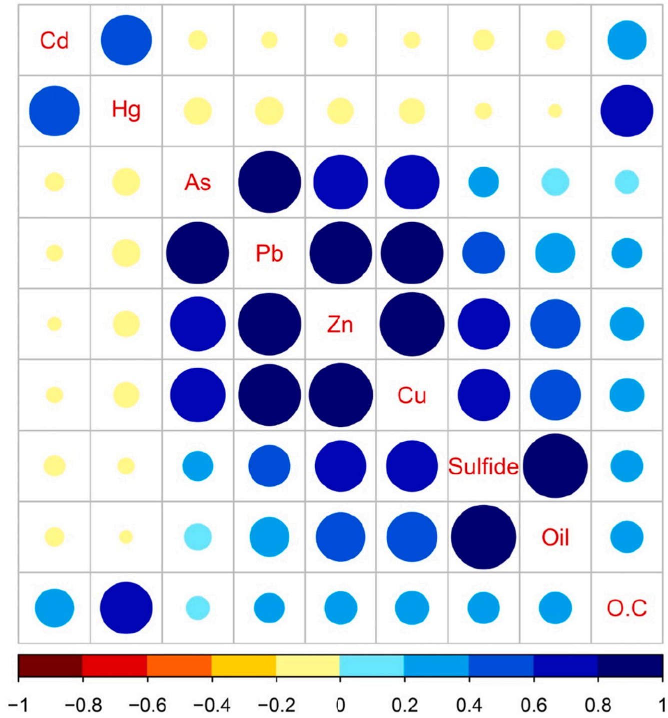

**Fig. 6.** Pearson's correlation coefficients for relationships between potential toxic element (PTE) concentrations in the sediments collected from 9 different cities around Hainan Island.

(TCR) (Table 3). The results showed that at all sampling points, THI values were higher for children than adults. Children were found to have higher THI in ChengMai, where the Zn and As contents were particularly high. Lead, on the other hand, was found at all sampling points to have a non-carcinogenic risk for both children and adults. In general, THI for children followed this order: Zn > As> Cd > Hg > Cu > Pb, while adult THI followed this order: Zn > As> Cu > Cd > Hg > Pb. Overall, our results tell us about that the children in the ChengMai area are most susceptible to the health risks posed by PTEs in collected sediments, and that they are more likely to be affected by these PTEs than adults (Table 4).

## 4. Discussion

We investigated the concentrations of six potentially toxic elements (PTEs) in the sediments of nine (9) different cities around Hainan Island. Samples of marine sediments were collected from each city at three different sites for Cd, Cu, Pb, As, Zn and Hg analysis, in order to determine their concentrations (Table 1). In September 2019, 285 sediment samples were collected from the nine cities around the Island. The locations of sample collection sites are detailed in Fig. 1. The study found that all six of the selected PTEs were present in every sample that was collected, although concentrations varied for different areas. The increased levels of these metals in the sediment samples collected from ChengMai could be attributed to anthropogenic activities such as agricultural, fishing, and tourism activities, which are natural pathways to heavy metal uptake in plants and therefore a major source of heavy metal pollution (El-Naggar et al., 2021; Hasimuna et al., 2021). While, Cai et al. (2021) found that the concentration of heavy metals gradually increased in the areas near the sea possibly caused by a higher degree of industrialization on the Hainan Island. One particular study found that, due to anthropogenic activities in the area, the concentrations of heavy metals in collected vegetable samples were far higher than FAO/WHO permissible values (Mensah et al., 2009). The increased levels of heavy metals in Lingao could be a consequence of industrial discharge along the coastline, leakage and petrol emissions due to busy marine transport routes, or atmospheric depositions (Sany et al., 2013). These results reflect previous findings reporting that urbanization and industrial development have resulted in increased levels of heavy metal pollution in the People's Republic of China (Zhang et al., 2020). Other anthropogenic activities, such as agriculture and production of household

**Table 3**

Results of non-carcinogenic risks of heavy metals for children and adults in the study regions around Hainan Island.

| | Cd | | Pb | | Zn | | Cu | | Hg | | As | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Children | Adults | Children | Adults | Children | Adults | Children | Adults | Children | Adults | Children | Adults |
| City | | | | | | | | | | | | |
| Ledong | 0.0459 | 0.0114 | 0.0015 | 0.0003 | 0.1196 | 0.0298 | 0.0011 | 0.0002 | 0.0181 | 0.0045 | 0.0021 | 0.0003 |
| ChengMai | 0.0621 | 0.0155 | 0.1066 | 0.0196 | 16.1352 | 4.0169 | 0.0717 | 0.0132 | 0.0126 | 0.0031 | 0.0832 | 0.0111 |
| DongFang | 0.0286 | 0.0071 | 0.0017 | 0.0003 | 0.0852 | 0.0212 | 0.0014 | 0.0003 | 0.0558 | 0.0139 | 0.0036 | 0.0005 |
| LingShui | 0.1481 | 0.0369 | 0.0016 | 0.0003 | 0.1293 | 0.0322 | 0.0017 | 0.0003 | 0.0422 | 0.0105 | 0.0023 | 0.0003 |
| Wenchang | 0.0698 | 0.0174 | 0.0029 | 0.0005 | 0.0214 | 0.0053 | 0.0004 | 0.0001 | 0.0208 | 0.0052 | 0.0034 | 0.0005 |
| Sanya | 0.0872 | 0.0217 | 0.0008 | 0.0001 | 0.0858 | 0.0214 | 0.0012 | 0.0002 | 0.0216 | 0.0054 | 0.0015 | 0.0002 |
| QiongHai | 0.0431 | 0.0107 | 0.0004 | 0.0001 | 0.0445 | 0.0111 | 0.0008 | 0.0001 | 0.0374 | 0.0093 | 0.0032 | 0.0004 |
| Haikou | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 | 0.0080 |
| Lingao | 0.2728 | 0.0679 | 0.0029 | 0.0005 | 0.1480 | 0.0368 | 0.0018 | 0.0003 | 0.0870 | 0.0217 | 0.0034 | 0.0004 |

**Table 4**

Results of carcinogenic risks of heavy metals for children and adults in the study regions around Hainan Island.

| City | Cd | | Pb | | As | | Cu | | Zn | | Hg | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Children | Adult | Children | Adult | Children | Adult | Children | Adult | Children | Adult | Children | Adult |
| Ledong | 3.231E−06 | 4.785E−07 | 7.148E−06 | 7.831E−07 | 1.757E−06 | 2.040E−08 | ND | ND | ND | ND | ND | ND |
| ChengMai | 4.374E−06 | 6.479E−07 | 5.069E−04 | 5.554E−05 | 6.980E−05 | 2.924E−07 | ND | ND | ND | ND | ND | ND |
| DongFang | 2.016E−06 | 2.986E−07 | 8.130E−06 | 8.907E−07 | 3.013E−06 | 1.262E−08 | ND | ND | ND | ND | ND | ND |
| LingShui | 1.043E−05 | 1.544E−06 | 7.396E−06 | 8.103E−07 | 1.935E−06 | 8.103E−09 | ND | ND | ND | ND | ND | ND |
| Wenchang | 4.911E−06 | 7.274E−07 | 1.368E−05 | 1.499E−06 | 2.863E−06 | 1.199E−08 | ND | ND | ND | ND | ND | ND |
| Sanya | 6.139E−06 | 9.092E−07 | 3.749E−06 | 4.108E−07 | 1.271E−06 | 5.324E−09 | ND | ND | ND | ND | ND | ND |
| QiongHai | 3.037E−06 | 4.498E−07 | 1.973E−06 | 2.162E−07 | 2.722E−06 | 1.140E−08 | ND | ND | ND | ND | ND | ND |
| Haikou | 4.906E−02 | 4.906E−02 | 6.836E−02 | 6.836E−02 | 1.206E−02 | 1.206E−02 | ND | ND | ND | ND | ND | ND |
| Lingao | 1.921E−05 | 2.844E−06 | 1.402E−05 | 1.536E−06 | 2.814E−06 | 1.179E−08 | ND | ND | ND | ND | ND | ND |

ND = not determined.

wastewater, could also be contributing to elevated levels of PTEs. This is because the sites are close to farming areas, which can also serve as a source of the trace elements found in sediments (Hasimuna et al., 2021). On a general basis, these trace elements are essential for day-to-day life, for both humans and domesticated animals. They are harmful, however, when they exceed certain thresholds. Indeed, it has been observed that heavy metals are toxic when their concentrations exceed specific thresholds, which is when they can begin to have substantial adverse impacts on human health and the environment (Cui et al., 2020). Table 5 shows the comparison of the mean heavy metal concentrations in sediments of different cities around Hainan Island with those in other regions, it clearly shows less pollution of heavy metals compared to other regions.

PTE pollution of sediments can seriously degrade aquatic ecosystems, and their concentrations are thus considered to be reliable indicators of ecosystem health (X. Li et al., 2019). The highest degree of As contamination in ChengMai could be due to the proximity of the site to industrial waste and the abundance of anthropogenic activities in the area, such as agriculture and residential depositions. Indeed, these high levels of Cd, Cu and Hg in the sediments may be attributed to crustal and anthropogenic sources (Hasimuna et al., 2021). The enrichment factor (EF) of the concentrations of all six (6) PTEs (Cd, Cu, As, Zn, Hg and Cr) in sediment samples collected from the nine (9) cities around Hainan Island are presented in Fig. 4. The increased values of the EF suggesting that these heavy metals were mainly derived from continental clastic materials controlled by natural weathering (Cai et al., 2021). The Enrichment Factor (EF) values between 0.5 and 1.5 indicates that the metals are entirely from the coastal materials whereas EF values greater than 1.5 indicates that the sources are most likely to be anthropogenic activities (Chang et al., 2014). The higher levels of Hg in the sediment

**Table 5**

Comparison of the mean heavy metal concentrations in sediments of different cities around Hainan Island with those in other regions.

| Location | Unit | Cd | Cu | Zn | As | Pb | Hg | Reference |
|---|---|---|---|---|---|---|---|---|
| Lingao | mg/kg | 0.743 | 0.334 | 0.403 | 0.606 | 0.533 | 0.237 | This study |
| Haikou | mg/kg | 0.374 | 0.323 | 0.536 | 0.289 | 0.299 | 0.331 | This study |
| Qionghai | mg/kg | 0.117 | 0.137 | 0.121 | 0.586 | 0.075 | 0.101 | This study |
| Sanya | mg/kg | 0.237 | 0.208 | 0.311 | 0.273 | 0.142 | 0.058 | This study |
| Wenchang | mg/kg | 0.190 | 0.075 | 0.058 | 0.616 | 0.520 | 0.056 | This study |
| Lingshui | mg/kg | 0.403 | 0.311 | 0.352 | 0.416 | 0.281 | 0.115 | This study |
| Dongfang | mg/kg | 0.097 | 0.284 | 0.232 | 0.649 | 0.309 | 0.152 | This study |
| ChengMai | mg/kg | 0.244 | 12.95 | 43.94 | 15.03 | 19.26 | 0.034 | This study |
| Ledong | mg/kg | 0.125 | 0.205 | 0.325 | 0.378 | 0.271 | 0.049 | This study |
| Continental shelf of Hainan | μg/kg | ND | 14.97 | 70.50 | ND | 26.82 | ND | (Cai et al., 2021) |
| Western Taiwan Strait | mg/kg | ND | 10.2 | 51.7 | 7.5 | 18.3 | ND | (Zhai et al., 2020) |
| North Yellow Sea | μg/kg | 0.09 | 15.9 | 57.3 | ND | 24.1 | ND | (Huang et al., 2014) |
| South Yellow Sea | μg/kg | ND | 17.40 | 70.30 | ND | 20.70 | ND | (Lu et al., 2017) |
| East China Sea | μg/kg | 0.13 | 16.3 | 72.7 | ND | 33.1 | ND | (M. Liu et al., 2019) |
| Manimuthar | mg/kg | 0.294 | 0.448 | 18.020 | 1.095 | 1.053 | ND | (Arisekar et al., 2020) |
| Tirunelveli | mg/kg | 0.643 | 9.510 | 29.958 | 0.940 | 1.343 | ND | (Arisekar et al., 2020) |
| Srivaikuntam | mg/kg | 1.620 | 20.97 | 70.414 | 0.987 | 1.338 | ND | (Arisekar et al., 2020) |
| Authoor | mg/kg | 3.123 | 35.23 | 93.278 | 2.061 | 2.964 | ND | (Arisekar et al., 2020) |

ND = no data.

samples could be attributed to anthropogenic activities (Nemati et al., 2011), since Haikou is a city of ports with a considerably high volume of metal works, and a city which frequently sees gasoline burnt and engine oil spilt from generators. The pollution load index (PLI) indicated pollution in Haikou, Lingao and ChengMai might be due to anthropogenic activities, including increases in population density and more frequent dumping of household or industrial waste in these areas. This is in keeping with the findings reporting that sediments in the coastal areas of an industrial basin in Northeast China were extremely contaminated following many years of random dumping of hazardous waste and free discharge of effluents by industries smelting Zn, companies dealing in chemical engineering, and other factories (Marrugo-Negrete et al., 2021). Their research also suggested that urbanization and economic development in coastal areas was leading to environmental problems on a global scale. Furthermore, it was noted that economic activities, including mining, metal fabrication, construction of housing, streets and highways, increased automobile traffic, use of septic tanks, municipal/industrial waste effluents, and illegal dumping of waste products from homes and businesses, are contributing to the emergence in varied ecosystems of contaminants such as esters, phthalates, derivatives of petroleum-based fuels, and heavy metals (Ortiz Colon, 2016). Twenty years ago, Naylor et al. (2000) reviewed the description of aquaculture as a possible solution and contributor to the reduction of global fishery resources. Unless the industry reduces the use of wild fish in feed as well as its environmental impact (Naylor et al., 2021).

The carcinogenic risks are usually estimated by the lifetime cancer risk index, the lifetime cancer risk, and the cumulative cancer risk (Antoniadis et al., 2019); whereas the hazard quotient, the target hazard quotient, and the total hazard index are used to assess non-carcinogenic risks (Alves et al., 2014). Industrialization and advancements in agriculture are increasingly the cause of environmental concerns and need to be addressed (Arisekar et al., 2020). The potential health risks posed by sediments to humans were established by evaluating the concentrations of potentially toxic elements (PTEs) in the sediments and the ways in which humans might be exposed to them. Metals with $HI > 1$ indicate a higher carcinogenic risk (USEPA, 2015), while metals with $HI < 1$ indicate no carcinogenic risk. The high levels of Zn at the ChengMai site may be attributed to human activities, as high concentrations of PTEs were found in the industrial area (Hoang et al., 2021), which would then enter the food chain and ultimately pose a risk to human health (Hoang et al., 2021). Zn is considered harmless than other HMs in the soil and sediments but its exposure to high doses has toxic effects, making acute zinc intoxication a rare event (Plum et al., 2010). In addition to acute intoxication, long-term, high-dose zinc supplementation interferes with the uptake of copper. Hence, many of its toxic effects are in fact due to copper deficiency. There are three major routes of entry for zinc into the human body: by inhalation, through the skin, or by ingestion (Atlanta, USA, 2002). All the other studies HMs in the sediments in all the cities were found within the permissible range, so it can be concluded that except Zn in ChengMai all the other cities have good and healthy living environment.

## 5. Conclusions and recommendations

The present study investigated the distribution of potentially toxic elements (PTEs) in the surface sediments of different cities on Hainan Island, China, with the basic goal of measuring the content of PTEs in sediments and assessing the potential health risks these metals may pose to humans. The study concludes that surface sediments around Hainan Island are significantly polluted by PTEs, especially by Cd and Zn. Analysis of PLI further confirmed the belief that PTE pollution may threaten the ecosystem of the relevant area. In the case of Cd, the element was only moderately enriched in some areas, and may have come mainly from pesticides and fertilizers. Furthermore, our human health risk assessment revealed that it is Zn in ChengMai which poses the most significant risk, for both children and adults, followed by As. Although all the PTEs (except Cd in the city of Haikou) exhibit low ecological risks in general, future research would be well-advised to account for the province's rapid population and industry growth, and to conduct experiments which can better our understanding of the bioaccumulation of these PTEs in animals and plants. Such research will contribute to an understanding of the abundance and distribution of PTEs, an important endeavor given that insufficient distribution of aquatic species is usually attributed to the large number of pollutants discharged directly or indirectly into the water by industrial processes. It will also emphasize the dangers of consuming aquatic products in these areas and reveal the possible biological amplification of some species of fish, or even plants. We also recommend the study of microplastics, and PTEs in fish from aquaculture farms and cages, as a research direction that would be worthy of pursuit.

## CRediT authorship contribution statement

**Zhiwei Che**: Performing the experiment, data collection and treatment, creating figures, writing the draft of the manuscript

**Waqas Ahmed**: Performing the experiment, investigation, analysis, and writing

**Juha Alatalo**: review, editing and proof reading

**Mohsin Mahmood**: Data analyses, writing, and corrections

**Jiechang Weng**: review, correction, editing and proof reading

**Liu Wenjie**: accurateness of data analysis, correction, and editing

**Ou Wenjie**: review, correction, editing and proof reading

**Mir Muhammad Nizamani**: review, correction, editing and proof reading

**Wang Lu**: review, correction, editing and proof reading

**Fu Xiu Xian**: review, correction, editing and proof reading

**Wang Yunting**: review, correction, editing and proof reading

**Sajid Mehmood**: research idea, concept, writing, review, correction, editing and proof reading, and corresponding

**Weidong Li**: supervision, writing, review, correction, editing and proof reading, and corresponding

## Declaration of competing interest

The authors declare no conflict of interest. Moreover, the manuscript fits well with the aim and scope of Marine Pollution Bulletin. The manuscript is an original work that has not been published previously and it is not under consideration for publication elsewhere. The publication has been approved by all co-authors.

### *Acknowledgement*

The National Natural Science Foundation of China (NSFC-31860728) and Key R & D Project of Hainan Province (No. ZDYF2021XDNY185) financially supported this research.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2021.113278.

## References

Alipour, H., Pourkhabbaz, A., Hassanpour, M., 2015. Estimation of potential health risks for some metallic elements by consumption of fish. Water Qual Expo Health. https://doi.org/10.1007/s12403-014-0137-3.

Alves, R.I.S., Sampaio, C.F., Nadal, M., Schuhmacher, M., Domingo, J.L., Segura-Muñoz, S.I., 2014. Metal concentrations in surface water and sediments from Pardo River, Brazil: human health risks. Environ. Res. https://doi.org/10.1016/j.envres.2014.05.012.

Amin, H.A., Saleh, H.N., Omar, M.Y., Mostafa, A.R., Ebraham, Y.E., 2020. Spatial distribution and assessment of heavy metals pollution in sediments of Tobruk Bay (Libya). In: Advances in Intelligent Systems and Computing. https://doi.org/10.1007/978-3-030-36671-1_55.

Antoniadis, V., Shaheen, S.M., Levizou, E., Shahid, M., Niazi, N.K., Vithanage, M., Ok, Y.S., Bolan, N., Rinklebe, J., 2019. A critical prospective analysis of the potential toxicity of trace element regulation limits in soils worldwide: are they protective concerning health risk assessment? - A review. Environ. Int. https://doi.org/10.1016/j.envint.2019.03.039.

Arisekar, U., Shakila, R.J., Shalini, R., Jeyasekaran, G., 2020. Human health risk assessment of heavy metals in aquatic sediments and freshwater fish caught from Thamirabarani River, the Western Ghats of South Tamil Nadu. Mar. Pollut. Bull. https://doi.org/10.1016/j.marpolbul.2020.111496.

Barakat, A., El Baghdadi, M., Rais, J., Nadem, S., 2012. Assessment of heavy metal in surface sediments of Day River at Beni-Mellal region, Morocco. Res. J. Environ. Earth Sci. 4, 797–806.

Cai, P., Cai, G., Chen, X., Li, S., Zhao, L., 2021. The concentration distribution and biohazard assessment of heavy metal elements in surface sediments from the continental shelf of Hainan Island. Mar. Pollut. Bull. 166 https://doi.org/10.1016/j.marpolbul.2021.112254.

Chang, C.Y., Yu, H.Y., Chen, J.J., Li, F.B., Zhang, H.H., Liu, C.P., 2014. Accumulation of heavy metals in leaf vegetables from agricultural soils and associated potential health risks in the Pearl River Delta, South China. Environ. Monit. Assess. https://doi.org/10.1007/s10661-013-3472-0.

Cui, X., Wang, X., Liu, B., 2020. The characteristics of heavy metal pollution in surface dust in Tangshan, a heavily industrialized city in North China, and an assessment of associated health risks. J. Geochem. Explor. https://doi.org/10.1016/j.gexplo.2019.106432.

Duncan, A.E., de Vries, N., Nyarko, K.B., 2018. Assessment of heavy metal pollution in the sediments of the River Pra and Its Tributaries. Water Air Soil Pollut. https://doi.org/10.1007/s11270-018-3899-6.

El-Naggar, A., Shaheen, S.M., Chang, S.X., Hou, D., Ok, Y.S., Rinklebe, J., 2021. Biochar surface functionality plays a vital role in (Im)Mobilization and phytoavailability of soil vanadium. ACS Sustain. Chem. Eng. 9, 6864–6874. https://doi.org/10.1021/acssuschemeng.1c01656.

Enuneku, A., Omoruyi, O., Tongo, I., Ogbomida, E., Ogbeide, O., Ezemonye, L., 2018. Evaluating the potential health risks of heavy metal pollution in sediment and selected benthic fauna of Benin River, Southern Nigeria. Appl. Water Sci. https://doi.org/10.1007/s13201-018-0873-9.

Ge, Q., Xue, Z.G., Chu, F., 2018. Spatial distribution and contamination assessment of surface heavy metals off the western Guangdong Province and northeastern Hainan Island. Int. J. Environ. Res. Public Health. https://doi.org/10.3390/ijerph15091897.

Guan, J., Wang, J., Pan, H., Yang, C., Qu, J., Lu, N., Yuan, X., 2018. Heavy metals in Yinma River sediment in a major Phaeozems zone, Northeast China: distribution, chemical fraction, contamination assessment and source apportionment. Sci. Rep. https://doi.org/10.1038/s41598-018-30197-z.

Hakanson, L., 1980. An ecological risk index for aquatic pollution control. A sedimentological approach. Water Res. 14 (8), 975–1001.

Hasimuna, O.J., Chibesa, M., Ellender, B.R., Maulu, S., 2021. Variability of selected heavy metals in surface sediments and ecological risks in the Solwezi and Kifubwa Rivers, Northwestern province, Zambia. Sci. Afr. 12. https://doi.org/10.1016/j.sciaf.2021.e00822.

Hoang, H.G., Chiang, C.F., Lin, C., Wu, C.Y., Lee, C.W., Cheruiyot, N.K., Tran, H.T., Bui, X.T., 2021. Human health risk simulation and assessment of heavy metal contamination in a river affected by industrial activities. Environ. Pollut. https://doi.org/10.1016/j.envpol.2021.117414.

Hu, B., Li, J., Zhao, J., Yang, J., Bai, F., Dou, Y., 2013. Heavy metal in surface sediments of the Liaodong Bay, Bohai Sea: distribution, contamination, and sources. Environ. Monit. Assess. 185, 5071–5083. https://doi.org/10.1007/s10661-012-2926-0.

Huang, P., Li, T.Gang, Li, A.Chun, Yu, X.Ke, Hu, N.J., 2014. Distribution, enrichment and sources of heavy metals in surface sediments of the North Yellow Sea. Cont. Shelf Res. https://doi.org/10.1016/j.csr.2013.11.014.

Isa, B.K., Amina, S.B., Aminu, U., Sabo, Y., 2015. Health risk assessment of heavy metals in water, air, soil and fish. Afr. J. Pure Appl. Chem. 9, 204–210. https://doi.org/10.5897/ajpac2015.0654.

Khan, M.Z.H., Hasan, M.R., Khan, M., Aktar, S., Fatema, K., 2017. Distribution of heavy metals in surface sediments of the bay of Bengal coast. J. Toxicol. 2017 https://doi.org/10.1155/2017/9235764.

Khusnidinov, S.K., Zamaschikov, R.V., Dmitriev, N.N., Butyrin, V., Sosnitskaya, T.N., 2020. Assessment of crop production quality in case of technogenic soil contamination. In: IOP Conference Series: Earth and Environmental Science. https://doi.org/10.1088/1755-1315/548/6/062092.

Lee, S., Chu, M.L., Guzman, J.A., 2021. Sediment fate and transport: influence of sediment source and rainfall. J. Hydrol. https://doi.org/10.1016/j.jhydrol.2021.125980.

Li, R.H., Liu, S.M., Li, Y.W., Zhang, G.L., Ren, J.L., Zhang, J., 2014. Nutrient dynamics in tropical rivers, lagoons, and coastal ecosystems of eastern Hainan Island, South China Sea. Biogeosciences 11, 481–506. https://doi.org/10.5194/bg-11-481-2014.

Li, R., Tang, C., Li, X., Jiang, T., Shi, Y., Cao, Y., 2019. Reconstructing the historical pollution levels and ecological risks over the past sixty years in sediments of the Beijiang River, South China. Sci. Total Environ. 649, 448–460. https://doi.org/10.1016/j.scitotenv.2018.08.283.

Li, X., Shen, H., Zhao, Y., Cao, W., Hu, C., Sun, C., 2019. Distribution and potential ecological risk of heavy metals in water, sediments, and aquatic macrophytes: A case study of the junction of four rivers in Linyi City, China. Int. J. Environ. Res. Public Health. https://doi.org/10.3390/ijerph16162861.

Li, X., Yang, Y., Yang, J., Fan, Y., Qian, X., Li, H., 2021. Rapid diagnosis of heavy metal pollution in lake sediments based on environmental magnetism and machine learning. J. Hazard. Mater. 416 https://doi.org/10.1016/j.jhazmat.2021.126163.

Liu, H., Liu, G., Yuan, Z., Ge, M., Wang, S., Liu, Y., Da, C., 2019. Occurrence, potential health risk of heavy metals in aquatic organisms from Laizhou Bay, China. Mar. Pollut. Bull. 140, 388–394. https://doi.org/10.1016/j.marpolbul.2019.01.067.

Liu, M., Chen, J., Sun, X., Hu, Z., Fan, D., 2019. Accumulation and transformation of heavy metals in surface sediments from the Yangtze River estuary to the East China Sea shelf. Environ. Pollut. https://doi.org/10.1016/j.envpol.2018.10.128.

Loska, K., Wiechula, D., Barska, B., Cebula, E., Chojnecka, A., 2003. Assessment of arsenic enrichment of cultivated soils in Southern Poland. Pol. J. Environ. Stud. 12 (2), 187–192.

Lu, J., Li, A., Huang, P., 2017. Distribution, sources and contamination assessment of heavy metals in surface sediments of the South Yellow Sea and northern part of the East China Sea. Mar. Pollut. Bull. https://doi.org/10.1016/j.marpolbul.2017.07.007.

Lu, X., Huang, C., Chen, F., Zhang, S., Lao, Q., Chen, C., Wu, J., Jin, G., Zhu, Q., 2021. Carbon and nitrogen isotopic compositions of particulate organic matter in the upwelling zone off the east coast of Hainan Island, China. Mar. Pollut. Bull. https://doi.org/10.1016/j.marpolbul.2021.112349.

Manoj, K., Padhy, P.K., 2014. Distribution, enrichment and ecological risk assessment of six elements in bed sediments of a Tropical River, chottanagpur plateau: a spatial and temporal appraisal. J. Environ. Prot. (Irvine, Calif). https://doi.org/10.4236/jep.2014.514136.

Marrugo-Negrete, J., Pinedo-Hernández, J., Marrugo-Madrid, S., Navarro-Frómeta, E., Díez, S., 2021. Sea cucumber as bioindicator of trace metal pollution in coastal sediments. Biol. Trace Elem. Res. https://doi.org/10.1007/s12011-020-02308-3.

Mennillo, E., Adeogun, A.O., Arukwe, A., 2020. Quality screening of the Lagos lagoon sediment by assessing the cytotoxicity and toxicological responses of rat hepatoma H4IIE and fish PLHC-1 cell-lines using different extraction approaches. Environ. Res. 182 https://doi.org/10.1016/j.envres.2019.108986.

Mensah, E., Kyei-Baffour, N., Ofori, E., Obeng, G., 2009. Influence of human activities and land use on heavy metal concentrations in irrigated vegetables in Ghana and their health implications. In: Appropriate Technologies for Environmental Protection in the Developing World - Selected Papers from ERTEP 2007, pp. 9–14. https://doi.org/10.1007/978-1-4020-9139-1_2.

Merciai, R., Guasch, H., Kumar, A., Sabater, S., García-Berthou, E., 2014. Trace metal concentration and fish size: variation among fish species in a Mediterranean river. Ecotoxicol. Environ. Saf. https://doi.org/10.1016/j.ecoenv.2014.05.006.

Mohammadi, M.J., Yari, A.R., Saghazadeh, M., Sobhanardakani, S., Geravandi, S., Afkar, A., Salehi, S.Z., Valipour, A., Biglari, H., Hosseini, S.A., Rastegarimehr, B., Vosoughi, M., Omidi Khaniabadi, Y., 2018. A health risk assessment of heavy metals in people consuming Sohan in Qom, Iran. Toxin Rev. 37, 278–286. https://doi.org/10.1080/15569543.2017.1362655.

Müller, G., 1969. Index of geoaccumulation in sediments of the Rhine River. Geol. J. 2, 108–118.

Naylor, R.L., Goldburg, R.J., Primavera, J.H., Kautsky, N., Beveridge, M.C.M., Clay, J., Folke, C., Lubchenco, J., Mooney, H., Troell, M., 2000. Effect of aquaculture on world fish supplies. Nature. https://doi.org/10.1038/35016500.

Naylor, R.L., Hardy, R.W., Buschmann, A.H., Bush, S.R., Cao, L., Klinger, D.H., Little, D.C., Lubchenco, J., Shumway, S.E., Troell, M., 2021. A 20-year retrospective review of global aquaculture. Nature. https://doi.org/10.1038/s41586-021-03308-6.

Nemati, K., Bakar, N.K.A., Abas, M.R., Sobhanzadeh, E., 2011. Speciation of heavy metals by modified BCR sequential extraction procedure in different depths of sediments from Sungai Buloh, Selangor, Malaysia. J. Hazard. Mater. https://doi.org/10.1016/j.jhazmat.2011.05.039.

Ortiz Colon, A.I., 2016. Assessment of concentrations of heavy metals and phthalates in two urban Rivers of the northeast of Puerto Rico. J. Environ. Anal. Toxicol. https://doi.org/10.4172/2161-0525.1000353.

Plum, L.M., Rink, L., Hajo, H., 2010. The essential toxin: impact of zinc on human health. Int. J. Environ. Res. Public Health. https://doi.org/10.3390/ijerph7041342.

Rinklebe, J., Antoniadis, V., Shaheen, S.M., Rosche, O., Altermann, M., 2019. Health risk assessment of potentially toxic elements in soils along the Central Elbe River, Germany. Environ. Int. https://doi.org/10.1016/j.envint.2019.02.011.

Ryther, J.H., 1969. Photosynthesis and fish production in the sea. Science (80-.). https://doi.org/10.1126/science.166.3901.72.

Sany, S.B.T., Salleh, A., Sulaiman, A.H., Sasekumar, A., Rezayi, M., Tehrani, G.M., 2013. Heavy metal contamination in water and sediment of the Port Klang coastal area, Selangor, Malaysia. Environ. Earth Sci. 69, 2013–2025. https://doi.org/10.1007/s12665-012-2038-8.

Shaheen, S.M., El-Naggar, A., Antoniadis, V., Moghanm, F.S., Zhang, Z., Tsang, D.C.W., Ok, Y.S., Rinklebe, J., 2020. Release of toxic elements in fishpond sediments under dynamic redox conditions: assessing the potential environmental risk for a safe management of fisheries systems and degraded waterlogged sediments. J. Environ. Manag. 255 https://doi.org/10.1016/j.jenvman.2019.109778.

Tayebi, L., Sobhanardakani, S., 2020. Analysis of heavy metal contents and non-carcinogenic health risk assessment through consumption of tilapia fish (Oreochromis niloticus). Pollution 6, 59–67. https://doi.org/10.22059/poll.2019.284500.639.

Tomlinson, D.L., Wilson, J.G., Harris, C.R., Jeffrey, D.W., 1980. Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. Helgoländer Meeresun. https://doi.org/10.1007/BF02414780.

USEPA, 2015. Regional Screening Level (RSL) Summary Table (TR=1E-06, HQ=1) November 2015. Usepa.

Vitali, F., Mandalakis, M., Chatzinikolaou, E., Dailianis, T., Senatore, G., Casalone, E., Mastromei, G., Sergi, S., Lussu, R., Arvanitidis, C., Tamburini, E., 2019. Benthic prokaryotic community response to polycyclic aromatic hydrocarbon chronic exposure: importance of emission sources in mediterranean ports. Front. Mar. Sci. 6 https://doi.org/10.3389/fmars.2019.00590.

Wang, D., Dang, Z., Feng, H., Wang, R., 2015. Distribution of anthropogenic cadmium and arsenic in arable land soils of Hainan, China. Toxicol. Environ. Chem. https://doi.org/10.1080/02772248.2015.1050193.

Wang, H., Wang, J., Liu, R., Yu, W., Shen, Z., 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. https://doi.org/10.1016/j.marpolbul.2015.01.026.

Wang, X.X., Yu, S.J., Wang, X.K., 2019. Removal of radionuclides by metal-organic framework-based materials. Wuji Cailiao Xuebao J. Inorg. Mater. https://doi.org/10.15541/jim20180211.

Xu, Fangjian, Liu, Z., Cao, Y., Qiu, L., Feng, J., Xu, Feng, Tian, X., 2017. Assessment of heavy metal contamination in urban river sediments in the Jiaozhou Bay catchment, Qingdao, China. Catena 150, 9–16. https://doi.org/10.1016/j.catena.2016.11.004.

Xu, F., Hu, B., Dou, Y., Song, Z., Liu, X., Yuan, S., Sun, Z., Li, A., Yin, X., 2018. Prehistoric heavy metal pollution on the continental shelf off Hainan Island, South China Sea: from natural to anthropogenic impacts around 4.0 kyr BP. The Holocene. https://doi.org/10.1177/0959683617729445.

Zang, Z., Li, Y., Liu, S., Li, H., Hao, Z., Xu, Y., 2021. Assessment of the heavy metal pollution and health risks of rice cultivated in Hainan Island, China. Environ. Forensic. https://doi.org/10.1080/15275922.2020.1836081.

Zhai, B., Liu, Z., Wang, X., Bai, F., Wang, L., Chen, Z., Zhang, X., 2020. Assessment of heavy metal contamination in surface sediments in the western Taiwan Strait. Mar. Pollut. Bull. https://doi.org/10.1016/j.marpolbul.2020.111492.

Zhang, K., Yang, F., Zhang, H., Su, D., Li, Q.Q., 2017. Morphological characterization of coral reefs by combining lidar and MBES data: a case study from Yuanzhi Island, South China Sea. J. Geophys. Res. Oceans. https://doi.org/10.1002/2016JC012507.

Zhang, H., Walker, T.R., Davis, E., Ma, G., 2019. Spatiotemporal characterization of metals in small craft harbour sediments in Nova Scotia, Canada. Mar. Pollut. Bull. 140, 493–502. https://doi.org/10.1016/j.marpolbul.2019.02.004.

Zhang, J.B., Zhang, P., Dai, P.D., Lai, J.Y., Chen, Y., 2019. Spatiotemporal distributions of DIP and the eutrophication in Hainan Island adjacent coastal water. Zhongguo Huanjing Kexue/China Environ. Sci. 39 (6), 2541–2548.

Zhang, M., Sun, X., Xu, J., 2020. Heavy metal pollution in the East China Sea: a review. Mar. Pollut. Bull. https://doi.org/10.1016/j.marpolbul.2020.111473.

Zhang, H., Sun, C., Wang, Z., Che, B., 2021. Seafood consumption patterns and affecting factors in urban China: a field survey from six cities. Aquac. Rep. https://doi.org/10.1016/j.aqrep.2021.100608.

Zhao, W., Chen, H., Wang, J., Zhang, M., Chen, K., Guo, Y., Ke, H., Huang, W., Liu, L., Yang, S., Cai, M., 2019. Current status, challenges, and policy recommendations of china's marine monitoring systems for coastal persistent organic pollution based on experts' questionnaire analysis. Int. J. Environ. Res. Public Health. https://doi.org/10.3390/ijerph16173083.

Zhao, L., Liu, J., Cai, G., Huang, L., Luo, W., 2020. Distribution, source, and pollution assessment of heavy metals in Sanya offshore area, South Hainan Island of China. Mar. Pollut. Bull. 160 https://doi.org/10.1016/j.marpolbul.2020.111561.

Zhu, D., Wu, S., Han, J., Wang, L., Qi, M., 2018. Evaluation of nutrients and heavy metals in the sediments of the Heer River, Shenzhen, China. Environ. Monit. Assess. https://doi.org/10.1007/s10661-018-6740-1.

Zhuang, W., Liu, Y., Tang, L., Yue, W., Liu, J., Ren, Y., Wang, X., Xu, S., Tai, S., Zhang, J., Zheng, Y., Guo, F., Wang, Q., Song, J., Duan, L., Chen, Q., 2019. Thallium concentrations, sources and ecological risk in the surface sediments of the Yangtze estuary and its adjacent East China marginal sea: a baseline study. Mar. Pollut. Bull. 138, 206–212. https://doi.org/10.1016/j.marpolbul.2018.11.049.