Temporal distribution, accumulation, speciation and ecological risk of heavy metals in the sediment of an urban Lagoon catchment at Xiamen in China

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ABSTRACT
The present study investigated the temporal distribution, accumulation, speciation and potential ecological risk of heavy metals in the sediment of an urban Yundang Lagoon catchment (YLC) at Xiamen in China. Total heavy metals concentration of Cr, Mn, Ni, Cu, Zn, As, Cd and Pb showed a significant seasonal variation in the sediment. Therefore, the average total metals concentration of Cr, Mn, Ni, Cu, Zn, As, Cd and Pb was 61.95, 529.95, 28.03, 64.95, 351.22, 10.29, 1.38 and 56.30 mg/kg for spring; 66.78, 501.22, 30.55, 67.72, 326.42, 10.91, 0.89 and 58.12 mg/kg for summer; 64.87, 472.80, 24.09, 66.26, 323.40, 5.51, 0.64 and 58.68 mg/kg for autumn; 82.36, 1589.05, 27.27, 82.64, 369.31, 11.79, 0.60 and 562.24 mg/kg for winter, respectively. Temporal metals speciation indicates a considerable seasonal variation of metal fraction concentrations. Igeo values have indicated considerable seasonal variation of pollution sources in the sediment of YLC. The temporal pollution load index indicates that winter sediment has the highest pollution load than other seasons.

1. Introduction
Heavy metals pollution in coastal environments such as lagoons, bays and estuaries has become the main concern over the last two decades [1–3]. Due to rapid industrialisation and urbanisation, heavy metals are continuously carried to coastal lagoon sediments from the surrounding areas and upstream tributaries [4,5]. The main inputs of heavy metals in coastal sediments are from natural sources such as geogenic inputs and anthropogenic sources such as domestic sewage, agricultural activities, atmospheric deposition and urban runoff [6–11]. Higher levels of heavy metals from the anthropogenic sources can accumulate in sediments and reduce the sediment quality [12,13]. The contaminated sediment could affect the water quality and enhance the bioaccumulation of heavy metals in aquatic organisms [14]. The polluted water and aquatic organisms could have potential long-term effects on human health and ecosystems [4]. To quantify the toxicity of heavy
metals, total metal content in sediments and several geo-chemical indices have been used widely by several researchers [15–17]. The most common geo-chemical indices are the geo-accumulation index, pollution load index and ecological risk index [18,19]. However, the quantification of total metal content can evaluate the potential elemental load in the aquatic ecosystem sediments [20]. But chemical fractionation study of heavy metals can evaluate the presence of possible species of heavy metals in sediments, which is also widely used to evaluate their potential mobility [21–22]. Therefore, fractionation studies can depict the origin, bioavailability, bioactivity, transport and risk of heavy metals in sediments [19,20,23]. In the aquatic ecosystems, the accumulation and distribution of heavy metals are mostly related to the local climatic factors such as rainy or dry seasons and sediment properties such as sediment grain size, the content of organic carbon, sediment pH, etc. [24–26]. These factors have substantial effects on the mobility of heavy metals in sediments [26]. So, the quantification of heavy metals concentration in sediments is an essential concern for the improvement of ecosystem health. In most of the heavy metal studies in aquatic ecosystems such as coastal areas, lagoons, and lakes, the spatial scale of metals distribution is mostly evaluated than the temporal scale [11,17,27,28]. But monitoring of sediment quality, as well as spatial and temporal processes have a large impact on the heavy metals accumulation and speciation in the aquatic ecosystems [1,14,24].

Several studies indicated the temporal variation of heavy metals concentration due to changes of geochemical processes in various dynamic aquatic environments. For example, temporal changes of Fe/Mn hydroxide formation in the estuarine environment [29], changes of oxidation and reduction conditions, and variation of sulfides in lakes and sedimentary basins [24,30–31]. Moreover, in a coastal wetland, temporal variability of total metals concentration reported in the sediment was related to historical deposition [32]. Lau [33] reported that temporal variability of total metals concentration in a coastal wetland was due to seasonal changes. Therefore, 20–30% variation of total metals concentration was due to small-scale spatial and temporal variability in a fluvial dominant aquatic environment [34]. Yundang Lagoon is an urban lagoon under continuous remediation and management scheme due to heavy metals pollution. Several studies indicated that heavy metal concentrations are higher than Chinese Marine Sediment Quality values [17]. But most of the studies were related to the spatial distribution of heavy metals concentration [14,17,21]. To our knowledge, no study is available on a temporal scale which is very important for the indentation of pollution sources, improvement of lagoon ecological health and changes of future management strategy.

The present study aims to investigate the temporal distribution and speciation of heavy metals in the sediment of the Yundang Lagoon catchment (YLC). Therefore, this study investigates the temporal variation of pollution sources and pollution load in the YLC sediment. Moreover, the relationships between heavy metals and sediment properties were assessed to identify the important factors that have an impact on heavy metals accumulation in the YLC sediments.

2. Materials and methods

2.1. Study area

Yundang Lagoon is an urban lagoon that is located in the western part of Xiamen Island at Xiamen in China (Figure 1). This lagoon was a natural harbour about 40 years ago and
connected to western Xiamen Bay. The YLC is composed of Tiandi Lake, Sonbai Lake and Yundang Lagoon and the total area of this catchment is about 37 km² [17,35]. A large land reclamation project had been conducted in the Yundang Lagoon in the early 1970s. As a result, the lagoon lost its water exchange capacity to the Bay and was considered as a dead lagoon due to the high amount of pollutants accumulated in the sediments [14,21]. This lagoon pollution received attention globally and Xiamen is the first city in China to adopt integrated coastal zone management under the supervision and support of the United Nations Environment Program [35]. After that several derigging and remediation engineering had been conducted in Yundang Lagoon, such as building an inlet channel, water pumping station and water control gate to improve the sediment and water quality [14,35]. During a natural tidal cycle, 1.3 × 10⁶ m³ seawater is input into this lagoon and is retained for three days [14]. Sea water is also pumped from the Yundang Lagoon to upstream lakes such as Tiandi and Songbai. Xiamen is a densely populated city with a sub-tropical monsoon climate and is one of the top tourist attractions in China.

2.2. Sediment sampling and preparation

Seasonal (spring, summer, autumn, and winter) sediment samples (54 samples from each season) were collected from the inlet channel, inner lagoon, outer lagoon and upstream lakes (Tiandi and Songbai Lake) of the YLC (Figure 1). Bottom surface sediments were collected from 18 sampling sites using a Peterson grabber. After that, 100 g of surface sediments were collected through a plastic spoon and put in a Ziploc plastic bag. Then, the
sediment samples were stored in an ice box container at 4°C during transport to the laboratory. Samples were stored at −18°C in the laboratory until further analysis. After that samples were oven dried at 80°C for more than 72 h until all the water content was removed from the samples. Then, sediments were ground with mortar and pestle, all the debris was removed and sieved through a 160 nylon mesh fibre [14,17]. Sediments were stored in a dry place until further chemical analysis.

2.3. Analysis of sequential extraction and digestion of residual fraction and total metals

The modified European Community Bureau of Reference (BCR) sequential extraction method was used to extract the heavy metal fractions from the sediment samples [14,36–37]. The extraction methods are described in Table S1. After the extraction of the oxidesable fraction, the residue sediment sample was oven-dried and stored in a dry place for further analysis. For the digestion of residual and total metals, 0.25 g of dried sediment samples were placed into a Teflon vessel and added with 6 mL of nitric acid, 2 mL of hydrochloric acid and 1 mL of hydrogen peroxide sequentially [17,37]. Then, the Teflon vessel was put into a microwave system for digestion (Coolpex, Preekem, China). The digested solution was put into an acid-washed centrifuge tube and stored at 4°C for quantification [37].

2.4. Assessment of sediment quality index

In the study of sediment quality assessment, various geochemical indexes have been used widely to evaluate the potential sources, pollution load, and risk of heavy metals in lagoon sediments.

2.4.1. Geo-accumulation index of heavy metals in sediment

The geo-accumulation index (Igeo) is a widely used method to quantify the pollution status of the studied locations. Igeo was first proposed by Muller [38] and this method can be used to evaluate the temporal variation of pollution levels in sediments that can be compared with the global average shale or crust concentration [39]. The following equation is used to calculate Igeo:

\[
I_{geo} = \log_2 \left( \frac{C_n}{1.5 \times B_n} \right)
\]

Here, \(C_n\) is the measured heavy metals concentration in the sediment of YLC and \(B_n\) is the global average shale concentration. The average shale value of Cr, Mn, Ni, Cu, Zn, As, Cd and Pb is 90, 850, 68, 45, 95, 13, 0.30 and 20 mg/kg, respectively [17,40]. The constant value of 1.5 is related to the correction factor that is used for the background values due to the lithogenic effect. According to Muller [39], Igeo values can be categorised into seven classes: \(I_{geo} < 0\), ‘Uncontaminated’ (UC); \(0 \leq I_{geo} < 1\), ‘Uncontaminated to moderately contaminated’ (UMC); \(1 \leq I_{geo} < 2\), ‘Moderately contaminated’ (MC); \(2 \leq I_{geo} < 3\), ‘Moderately to heavily contaminated’ (MHC); \(3 \leq I_{geo} < 4\), ‘Heavily
contaminated’ (HC); 4 ≤ I_{geo} < 5, ‘Heavily to extremely contaminated’ (HEC) and I_{geo} ≥ 5, ‘Extremely contaminated’ (EC) [11].

2.4.2. Pollution load index (PLI) of heavy metals in sediment

The pollution load (PLI) index is used to estimate the load of metal pollution and the degree of pollution in the sediment of aquatic ecosystems. The PLI was originally described by Tomlinson et al. [19] to determine the sediment quality of proposed sites, zones, and whole catchment in a certain period. The PLI was calculated using the following formula:

\[ \text{PLI} = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \ldots \times CF_n} \] (2)

Here, CF indicates the contamination factor of each metal, and n denotes the number of studied metals in the sediment sample. For the calculation of CF, the following equation was used:

\[ CF = \frac{[\text{metal}]_{\text{sediment}}}{[\text{metal}]_{\text{Ba}}} \] (3)

where \([\text{metal}]_{\text{sediment}}\) indicates the average concentration of metal in YLC sediment and \([\text{metal}]_{\text{Ba}}\) denotes the average global background value of the same metal. The global average crustal values described by Turekian and Wedepohl [40] were used in this equation. PLI values can be classified as follows: PLI < 1, ‘Perfect site quality or No pollution’; PLI = 1, ‘Baseline level of pollution’ and PLI > 1, ‘Deterioration in site quality’ [19].

2.4.3. Potential ecological risk of heavy metals

To evaluate the risk of each heavy metal to aquatic organisms, potential ecological risk \(E_i^r\) is the widely used method, which was proposed by the Swedish scientist Hakanson [18]. The \(E_i^r\) was calculated using the following equation:

\[ E_i^r = T_i^r \times CF \] (4)

Here, \(T_i^r\) indicates the toxic response factor for the studied heavy metals such as Ni = Cu = Pb = 5, Mn = Zn = 1, Cd = 30, Cr = 2 and As = 10, and CF is the contamination factor which is calculated from Equation 3. The \(E_i^r\) can be divided into the following categories: \(E_i^r < 40\), Low risk; \(40 \leq E_i^r < 80\), Moderate risk; \(80 \leq E_i^r < 160\), Considerable risk; \(160 \leq E_i^r < 320\), High risk and \(E_i^r \geq 320\), Very high risk [18]. The risk index (RI) indicates the combined ecological risk \(E_i^r\) of each heavy metal in the aquatic ecosystems. The RI values can be divided into the following categories: RI < 150, Low risk; 150 ≤ RI < 300, Moderate risk; 300 ≤ RI < 600, Considerable risk and RI ≥ 600, Very high risk [18].

2.5. Analysis of total organic carbon (TOC), pH, grain size and carbonate content

For the quantification of TOC percentage in sediments, 10 g of dried sediment sample was put into a 50 mL beaker, and soaked with 5% hydrochloric acid and stirred 24 h repeatedly at an interval of 6 h [14,17]. Then, the samples were washed with Milli-Q water and the acidity was removed. Samples were oven-dried at 80°C for more than 48 h. The dried sediment was ground with an agate mortar and sieved through a 100 mesh
nylon fibre [17]. The sediment TOC percentage was then analysed using an Elemental Analyser (Elemental tar Vario, EL III, Germany). The sediment pH was measured at a solid-to-liquid ratio of 1:2.5. The pH was determined using a Mettler Toledo pH metre (Columbus OH, USA). Carbonate percentage was measured by using the loss of ignition method [41].

2.6. Heavy metals fraction, total concentration quantification and quality control

The total and fraction concentrations of heavy metals (Cr, Mn, Ni, Cu, Zn, As, Cd and Pb) were quantified by using an inductively coupled plasma mass spectrometer (ICP-MS, Agilent, 7500ce, USA). All the samples were diluted by 10 times and triplicate absorbance was measured. Reagent blanks, triplicate measurement and standard reference materials were used to justify the analytical accuracy of the used method in this metals analysis. The detection limits of Cr, Mn, Ni, Cu, Zn, As, Cd and Pb was 0.45, 0.032, 0.76, 0.018, 0.14, 0.041, 0.045, and 0.003 ppb, respectively. The relative standard deviation of each measurement was less than 5% for ICP-MS. Therefore, the coastal marine standard reference sediment (GBW07314, National Institute of Standard and Technology, China) was used. The average recovery rate of heavy metals from the standard reference materials was 90% to 106%.

2.7. Data analysis

One-way ANOVA was used to determine the significant difference between variables of sampling sites [42]. Therefore, Turkey’s test was conducted to determine significant differences within variables. The relationships between total metals and sediment properties were determined using the Spearman correlation coefficient. Statistical analyses were conducted using SPSS 19.0 (SPSS for Windows, SPSS Inc.).

3. Results and discussion

3.1. Temporal distribution of sediment properties in YLC

Seasonal variations of TOC, sediment pH, and carbonate content are presented in Figure 2. TOC is an important component of sediments for the biogeochemical cycles of heavy metals. There is no significant difference \( (P > 0.05) \) between the seasonal variation of TOC concentration in sediments of the YLC (Figure 2). The average seasonal variation of TOC concentration in sediments decreased in the order of spring > autumn > summer > winter, respectively (Figure 2). Upstream lakes had a higher TOC content in sediments than the inlet channel, inner lagoon and outer lagoon during all the seasons, except spring in the outer lagoon (Table S2). TOC in the upstream lakes was 3.17 (spring), 3.56 (summer), 2.05 (autumn) and 3.43 (winter) times higher than the inlet channel sediment. Moreover, outer lagoon TOC concentration was 3.75 (spring), 3.15 (summer), 1.75 (autumn) and 3.11 (winter) times higher than the inlet channel TOC in sediments. Inlet channel sediment had the lowest TOC concentration than other sampling sites at all the seasons in YLC. Therefore, outer lagoon sediment had the highest TOC accumulation in the spring season than all other seasons and sampling
sites. Previous studies have suggested a lower amount of TOC concentration in the Yundang Lagoon sediments [21,27]. Moreover, a higher amount of TOC concentration was observed in the YLC sediments than in the surrounding vicinity of Xiamen Bay [27]. This higher TOC in YLC sediment is likely due to anthropogenic influences such as the input of urban runoff [27] and the release of domestic drainage materials into the lagoon [21]. The lagoon-borne TOC sources are mainly from phytoplankton, benthic microalgae and mangrove leaves [43]. This higher amount of TOC in the YLC sediment suggests anthropogenic influence. The seasonal variability of TOC in the YLC is most likely related to the wet and dry seasons in the study areas. Sediment pH has a considerable influence on the heavy metals accumulation and mobility in aquatic ecosystems. There is significant ($P < 0.05$) difference between the seasonal sediment pH concentration in the YLC sediments (Figure 2). Winter and autumn sediment pH concentrations were significantly ($P < 0.05$) lower than other studied seasons in the YLC (Figure 2). Moreover, winter had the lowest sediment pH concentration than other seasons. Summer sediment had the highest basic condition than other seasons sediment in the YLC. In the spring and summer seasons, there was higher alkaline sediment than autumn and winter sediment. Moreover, upstream lakes, inlet channel, and inner lagoon had higher alkaline content than outer lagoon sediment during the season of spring and summer (Table S2). In the autumn and winter seasons, there was a similar distribution pattern of sediment pH at all sampling sites in
the YLC (Table S2). Compared with the previous study, a higher alkaline condition is indicated in the present study [21]. A relatively lower alkaline content in autumn and winter (dry season) is likely related to the lower runoff from the surrounding areas into the lagoon [44]. Therefore, the variability of pH values is also related to the decay process of organic materials in the YLC [44].

Carbonate content in the sediment has a considerable influence on metal accumulation and speciation in aquatic ecosystems. There is a significant ($P < 0.05$) difference between the seasonal variation of carbonate content of the sediment in the YLC (Figure 2).

The highest and lowest average carbonate content was observed in the upstream lakes in the winter and summer season sediment (Table S2). The average carbonate concentration in the sediment was in the order of $3.70\%$ (winter) > $3.20\%$ (spring) > $3.27\%$ (autumn) > $2.75\%$ (summer), respectively (Figure 2).

### 3.2. Temporal distribution of total heavy metals concentration in the sediment of YLC

Climatic variability is an important factor for heavy metals accumulation in the sediment of aquatic ecosystems [1]. Cr concentration was significantly ($P < 0.05$) higher in the winter sediment than all other seasons in the YLC (Figure 3).

The average concentration of Cr in the sediment of the YLC was in the following decreasing order: winter > summer > autumn > spring. In winter, Cr concentration was higher in the upstream lakes and outer lagoon than in all other seasons (Table S3). In upstream lakes sediment, winter Cr concentration was 1.77, 1.55 and 1.64 times higher than spring, summer and autumn sediment concentrations, respectively (Table S3). An almost similar distribution pattern of Cr concentration was found in the upstream lakes, inlet channel, inner lagoon, and outer lagoon in spring, summer and autumn sediments (Table S3). There is significant ($P < 0.05$) seasonal variability of Mn concentration in the sediment of the YLC (Figure 3). Winter sediment has significantly ($P < 0.05$) higher Mn accumulation than all other seasons (Figure 3).

Average Mn concentration was in the following order: winter > spring > summer > autumn, respectively. In upstream lakes sediment, the average winter Mn concentration was 4.30, 5.08, and 4.70 times higher than spring, summer and autumn sediment concentrations (Table S3). Average winter sediment Mn concentration in the inner lagoon of the YLC was 2.18, 3.18, and 3.42 times higher than all other seasons sediment concentration (Table S3). In the outer lagoon, the average winter sediment Mn concentration was 2.41, 2.10 and 2.69 times higher than spring, summer and autumn concentrations (Table S3). An almost similar distribution pattern of Mn concentration was observed in spring, summer and autumn sediments in the YLC. There is a significant ($P < 0.05$) seasonal variation of Ni concentration in the sediment of the YLC (Figure 3).

Average Ni concentration was higher in the summer sediment and lowest in the autumn sediment than in all other seasons sediment (Figure 3). Ni concentration decreased in the order of summer > spring > winter > autumn. In comparison between the sampling zones, upstream lakes sediment (winter) was higher in Ni accumulation than inlet channel, inner lagoon and outer lagoon sediments (Table S3). Inlet channel sediment has the lowest Ni concentration than other sampling zones sediment in the
study area (Table S3). There is no significant ($P > 0.05$) seasonal variability of Cu accumulation in the sediment of the YLC (Figure 3).

But higher accumulation was observed in winter sediment than all other seasons sediment concentration (Figure 3). Seasonal Cu concentration decreased in the order of winter $>$ summer $>$ autumn $>$ spring, respectively (Figure 3). The highest and lowest Cu concentrations were found in the outer lagoon and inlet channel sediment (Table S3). In comparison between the sampling zones, outer lagoon Cu concentration was 2.08 (spring), 1.70 (summer), 2.22 (autumn), and 4.64 (winter) times higher than the inlet channel sediment (Table S3). In upstream lakes, a similar distribution pattern of Cu concentrations was observed in all the seasons in the YLC sediment (Table S3).

No significant ($P > 0.05$) difference between the seasonal Zn concentration in the sediment of the YLC was found (Figure 4). Higher Zn accumulation was found in the winter sediment than other seasons sediment concentration (Figure 4). Zn concentration decreased in the order of winter $>$ spring $>$ summer $>$ autumn (Figure 4). In comparison between the sampling zones, it is indicated that upstream lakes and outer lagoon have considerably higher Zn accumulation than inlet channel and inner lagoon sediment (Table S3). Inlet channel sediment has the lowest Zn concentration in all the studied seasons (Table S3).

As concentration in the YLC significantly ($P < 0.05$) varied in different seasons sediment accumulation (Figure 4).
Winter and autumn sediments had the highest and lowest As concentrations than other seasons (Figure 4). As concentration decreased in the order of winter > summer > spring > autumn. As concentration in the autumn sediment was lowest in upstream lakes, and inner and outer lagoons than in all other seasons sediment (Table S3). Summer inner lagoons sediment has the highest As concentration than all the other sites and seasons sediment in the YLC (Table S3).

There is significant ($P < 0.05$) seasonal variability of Cd concentration in the sediment of the YLC (Figure 4).

Average Cd concentration was higher in spring sampling sediment than in all other seasons sediment. Cd concentration decreased in the order of spring > summer > autumn > winter. In the spring, upstream lakes and outer lagoon were higher in Cd accumulation than all other seasons sediments in the YLC (Table S3). Inner lagoon was the lowest in Cd accumulation than upstream lakes and outer lagoon in all sampling seasons (Table S3). There is no significant ($P > 0.05$) seasonal variability of Pb concentration in the sediment of the YLC (Figure 4). In all the seasons, inner lagoon Pb concentration was lower than upstream lakes and outer lagoon sediments (Table S2).

Overall, in the YLC, the total concentration of Cr, Mn, Cu, Zn and As was higher in winter season sediment than in other studied seasons sediment indicating that there are seasonal influences on heavy metals accumulation in the sediment of the study areas [1]. This result also suggests that the winter season is considered as a hotspot of heavy metals accumulation in the YLC sediment. This may be due to the lower precipitation resulting in a
lower discharge of contaminated sediment from the lagoon to Xiamen Bay [45]. In comparison with the world average shale concentration of heavy metals, it showed that average Cu, Zn, Cd, and Pb concentrations were higher in all studied seasons sediment than average shale concentration [40]. Moreover, Cr and Mn concentrations in winter sediment were higher than the average shale concentration. Ni and As concentration was lower than the average shale concentration in all the studied seasons in YLC [40].

In comparison with China Marine Sediment Quality Criteria, it showed that the average concentration of Cu and Zn was higher than Class I pollution level of sediment quality in all the studied seasons [46]. And, average Zn concentration in spring and summer sediment was higher than Class II pollution level of sediment quality criteria [46]. Moreover, the average Cr concentration in winter sediment was greater than Class I pollution level of the sediment quality. This comparison indicates a considerable pollution level of heavy metals in the sediment of YLC [14]. The average concentration of Cu, Pb, As, and Cd found in this study was lower than in the previous study in Yundang Lagoon [21]. But Zn and Cr concentrations were higher in the present study than previously measured in the Yundang Lagoon sediment [21].

3.3. Temporal distribution of heavy metals speciation in the sediment of the YLC

Speciation is a criterion to assess the toxic potential of heavy metals which is a major concern for the ecological health of aquatic ecosystems [47]. In the aquatic environment, several factors and processes such as resuspension, redox condition, and seasonal variations are driving forces to act on the speciation of heavy metals [47].

3.3.1. Cr speciation

The temporal changes of Cr speciation in the sediment of YLC are depicted in Figure 5. This figure shows the minimal variation of fractions percentage in terms of seasonal changes in the whole catchment. Cr was dominant (range 68.01%–69.81%) in the residual fraction in all seasons. But the highest and lowest residual fractions were observed in winter and autumn sediments, respectively. The higher percentage of residual fraction in the sediments of YLC indicates that Cr is a stable metal in the sediment despite its seasonal variation. Similar results were reported by several researchers [14,48–49] which indicated that Cr is dominant in the residual fraction and stable in the coastal sediment. Oxidisable fractions were higher in spring sediment than other seasons and decreased in the order of spring > summer > autumn > winter, respectively. The seasonal percentage of oxidisable fraction was in the range of 21.18%–24.64% suggesting that under an oxidising condition, this percentage of fraction could be released in the Yundang Lagoon. The seasonal variation of the reducible fraction decreased in the order of autumn > summer > winter and > spring, respectively. Overall, in all seasons, the reducible fraction percentage was less than 11% in the sediments of YLC indicating that this fraction may not be mobilised under changing sediment pH condition. The acid-soluble fraction was less than 2% in all the seasons indicating that Cr is not bioavailable and is potentially toxic to the aquatic organisms in the YLC [21].

3.3.2. Mn speciation

The temporal changes of Mn speciation are presented in Figure 5, which shows the considerable variation of fraction percentages in seasons suggesting the seasonal variability
of Mn speciation in the sediments of YLC. In the sediments of spring, summer, and autumn, the residual fraction of Mn was dominant, while the acid-soluble fraction was dominant in winter sediment (Figure 5). The acid-soluble fraction was in the range of 20.89% to 54.30% indicating that this fraction could be released into the environment through the ion exchange process and is potentially toxic to aquatic organisms [49,50]. The highest bioavailable form of Mn was observed in the winter sediment than in other seasons in the YLC. A similar result was reported previously where a higher percentage of Mn was in the acid-soluble fraction in the sediment of YLC [14].

The seasonal variation of reducible fraction percentage decreased in the following order: spring > summer > winter > autumn, respectively. The variation of reducible fraction percentage was in the range of 15.26% to 28.08% suggesting that under the changes of redox condition and increasing acidity, this fraction could be released from the sediment of the study areas [37]. The oxidisable fraction was the lowest fraction percentage in the sediments of YLC. The seasonal variation of the oxidisable fraction was in the following decreasing order: winter > spring > summer > autumn, respectively. The range of oxidisable fraction percentage was 8.68% to 13.41% indicating that this fraction may be released under the increasing, reducing condition in the YLC sediment.

3.3.3. Ni speciation

Figure 5 shows the temporal changes of Ni speciation in the YLC sediment. As shown in Figure 5, Ni was dominant in the residual fraction in all seasons in the YLC sediment. This higher residual fraction indicates that Ni was less mobile in the sediment of YLC. The
highest residual fraction was observed in the winter sediment and the lowest was in the autumn sediment which indicates that Ni was more stable in winter sediment than in autumn sediment. Several researchers suggested that more than 50% of Ni occurs mostly in the residual fraction of the sediment [49,51]. The seasonal change of oxidisable fraction percentage in the YLC sediment decreased in the order of spring > autumn > winter > summer, respectively. The oxidisable fraction percentage was in the range of 17.08% to 14.34% suggesting that this fraction could be released from the organic matter to the aquatic environment if the reducing condition of sediment in the study area is increased [21]. The highest and lowest percentages of the reducible fraction were found in spring and winter sediments suggesting that Ni was more bioavailable in spring season than the other seasons. The seasonal variation of acid-soluble fraction decreased in the order of autumn > winter > summer > spring. This acid soluble fraction percentage was in the range of 6.18% to 13.58% indicating that this percentage could be released if it changes the pH condition of the sediment [14].

3.3.4. Cu speciation
The temporal changes of Cu speciation in the YLC sediment are shown in Figure 5. Cu was dominant in the oxidisable fraction in the sediment of YLC. The higher percentage of Cu in the oxidisable fraction suggests that organic matter and sulphides have important roles in Cu absorption in the sediment of the study area [52]. Several researchers have reported a high percentage of Cu in sediment in the oxidisable form [51,52]. In the previous study, a similar percentage of oxidisable fraction was also reported in the YLC sediment [14]. The seasonal variation of oxidisable fraction percentage decreased in the order of spring > autumn > winter > summer. The seasonal variation of reducible fraction percentage was in the range of 18.67% to 30.03% indicating that iron and manganese have significant roles in the absorption of Cu in the sediment. Therefore, the highest and lowest reducible fractions were found in summer and spring sediments. Acid-soluble fraction percentage was the lowest among the speciation fraction of Cu in the sediment of the YLC. This fraction percentage was in the range of 3.61% to 9.52% suggesting that Cu was less mobile in the sediment and less toxic to aquatic organisms.

3.3.5. Zn speciation
The temporal changes of Zn speciation are depicted in Figure 6.
Zn has higher seasonal variability of speciation fraction percentage in the sediment of the study area. The reducible fraction percentage concentration decreased in the order of spring > summer > autumn > winter. The seasonal fraction concentration was in the range of 19.50% to 37.65% indicating that a considerable percentage of Zn was bound in iron and manganese complexes in the sediment. Therefore, this fraction could be released into the surrounding environment owing to the increasing, reducing condition of the sediment. Gasparatous et al (2015) reported that about 50% of Zn was bound in a reducible fraction in the coastal sediment. The seasonal variation of acid-soluble fraction of Zn decreased in the order of autumn > winter > summer > spring, respectively. The concentration of acid-soluble fraction percentage was in the range of 18.64% to 41.20% suggesting that considerable percentage of Zn was bound with carbonates and this fraction may be potentially toxic to the aquatic organisms in the YLC sediment [21]. A similar acid-soluble fraction was reported previously in the YLC sediment [14].
3.3.6. As speciation

In Figure 6, the seasonal variation of As speciation in the sediment of YLC is shown. As was dominant in the residual fraction in all seasons. Residual fraction percentage was in the range of 50.16% to 74.09% indicating that As is a stable metal in the sediment of YLC and the highest stability was observed in the spring sediment than in other seasons (Figure 6). Similar results were reported by several researchers where more than 80% of As was bound in the residual form [22,53]. A similar result was also reported earlier [14]. The percentage of residual fraction decreased in the order of spring > summer > winter > autumn, respectively. The seasonal variation of oxidisable fraction percentage decreased in the order of autumn > spring > summer > winter. Autumn oxidisable fraction percentage was more than two times higher than in winter concentration.

3.3.7. Cd speciation

Figure 6 depicts the temporal changes of Cd speciation in the sediment. Cd speciation shows high seasonal variability of fraction percentage suggesting unstable metals in the sediment of YLC. The range of acid-soluble fraction was 36.76% to 73.34%. Higher percentage of acid soluble fraction indicates that Cd may be a potential toxic metal to aquatic organisms in the YLC [54]. A similar result was reported earlier in the Yundang Lagoon sediment [14]. There is a considerable seasonal variation of reducible fraction concentration in the sediment of the study area (Figure 6). The reducible fraction percentage
was in the range of 13.65% to 48.98% indicating that this fraction could be released in the environment under increasing, reducing condition and bioavailable to the aquatic organisms [49]. And this fraction concentration decreased in the order of spring > summer > autumn > winter, respectively.

3.3.8. Pb speciation
The seasonal variations of Pb speciation in the sediment of YLC are presented in Figure 6.

Among the speciation fraction, the highest percentage of Pb was bound in iron and manganese oxide complexes of the sediments. The seasonal variation of reducible fraction percentage was in the range of 47.52% to 64.60% indicating that with increasing, reducing condition, this fraction could be released into the surrounding environment and is potentially toxic to aquatic organisms [49]. The acid-soluble fraction was the lowest concentration percentage in all seasons suggesting less bioavailability to the aquatic organisms. The seasonal variation of residual fraction decreased in the order of spring > summer > winter > autumn, respectively. The residual fraction percentage was in the range of 22.35% to 32.25% indicating that Pb is a mobile metal in the sediment of the study area and potentially bioavailable to the aquatic organisms [14,21].

3.4. Temporal changes of heavy metals contamination in the sediment of YLC
The geo-accumulation index ($I_{geo}$) can quantify the degree of contamination of each metal to compare the geo-chemical background metal concentration of the sediment. This index is widely used to evaluate the ratio or amount of heavy metals in the sediment compared with the background level concentration [15,22]. The seasonal variation $I_{geo}$ values of the studied heavy metals of the YLC are presented in Supplementary Figures S1 to S8. The seasonal variation of Cr $I_{geo}$ values of the sediment is presented in Figure S1. Figure S1 shows the negative $I_{geo}$ values ($I_{geo} < 0$) of all the sampling locations except S1 (0 $\leq I_{geo} < 1$) in winter sampling. The $I_{geo}$ values of Cr were in the range of 1.44 to $-0.77$ in spring, $-1.44$ to $-0.59$ in summer, $-1.39$ to $-0.72$ in autumn and $-1.25$–$0.89$ in winter sediment (Figure S1). This result indicates that the sediments of YLC are not polluted with Cr and anthropogenic inputs are mostly from geogenic sources. A similar result was reported earlier [21]. Several researchers also reported natural input of Cr in the coastal sediment [15,22].

Figure S1 shows the negative $I_{geo}$ values ($I_{geo} < 0$) of all the sampling locations except S1 (0 $\leq I_{geo} < 1$) in winter sampling. The $I_{geo}$ values of Mn in the sediment of YLC are presented in Figure S2. Therefore, negative $I_{geo}$ values were found in all seasons of sediment except winter, indicating uncontaminated sediments in spring, summer and autumn. The $I_{geo}$ values were in the range of $-2.61$ to $-0.71$, $-1.93$ to $-0.03$, $-1.99$ to $-0.91$, and $-0.71$–$2.18$ for spring, summer, autumn and winter, respectively (Figure S2). This result suggests natural sources or input of Mn in the sediment in all seasons, except winter. Hasan et al. [15] reported natural sources of Mn in the sediment of a coastal area. Negative $I_{geo}$ ($I_{geo} < 0$) values of Ni in all sampling sites were observed (Figure S3). This suggests the geogenic input of Ni in the sediment of YLC. Guan et al. [22] reported a similar result in the river sediment where Ni was mostly from natural sources.

The seasonal $I_{geo}$ values of Cu are depicted in Figure S4. The $I_{geo}$ values of Cu were in the range of $-1.18$–$0.69$, $-1.20$–$0.96$, $-0.91$–$1.09$ and $-1.60$–$1.98$ in spring, summer,
autumn and winter sediments, respectively (Figure S4). This result indicates that sediments of the YLC are uncontaminated to moderately contaminated and may have anthropogenic input [39]. This result also suggests that winter sediment was moderately polluted with Cu than other seasons.

The changes of $I_{\text{geo}}$ values of Zn show considerable seasonal variation in the degree of contamination of heavy metals in the sediment of the YLC (Figure S5). The $I_{\text{geo}}$ values were in the range of $-0.05$–$2.16$ for spring, $-0.05$–$2.32$ for summer, $-0.07$–$2.48$ for autumn and $-1.13$–$3.11$ for winter, respectively (Figure S5). The maximum seasonal $I_{\text{geo}}$ values decreased in the following order: winter > autumn > summer > spring. The $I_{\text{geo}}$ values of Zn indicate that sediments were moderate to heavily contaminated in spring, summer and autumn seasons. But in winter, sediment was heavily contaminated with Zn. This suggests anthropogenic input of Zn in the sediment. A previous study in Yundang Lagoon showed that Zn was unpolluted to moderately polluted [21]. In urbanised areas, Zn is mostly used as a fungicide and painting agent and those are important sources of Zn in the YLC sediment [55].

The $I_{\text{geo}}$ values of As were negative at all sampling sites in all seasons (Figure S6). This indicates the geogenic or natural origin of As in the sediment (Muller [39]). But a previous study showed heavy contamination of As in the sediment of Yundang Lagoon [21]. This contrasting result indicates a reduction of As input from the surrounding areas of the Lagoon.

Considerable temporal variation of $I_{\text{geo}}$ values of Cd contamination was observed in different sampling sites (Figure S7) suggesting the variation of Cd sources or input in the YLC sediment. The seasonal $I_{\text{geo}}$ values were in the range of 0.52–2.32 for spring, $-0.48$–$2.12$ for summer, $-1.30$–$2.21$ for autumn and $-2.13$–$1.75$ for winter sediment (Figure S7). The maximum $I_{\text{geo}}$ values decreased in the following seasonal order: spring > autumn > summer > winter. In terms of contamination level, the spring, summer, and autumn sediments were moderate to heavily contaminated while winter sediments were moderately contaminated. This indicates the anthropogenic input of Cd [39]. The major anthropogenic sources are pesticides, discarded domestic electronics and discarded batteries from the surrounding urbanised areas [56,57].

Figure S8 shows the seasonal $I_{\text{geo}}$ values of Pb. The ranges of $I_{\text{geo}}$ values were 0.51–1.16 for spring, 0.51–1.34 for summer, 0.45–1.43 for autumn and 0.47–1.34 for winter sediment (Figure S8). This result indicates that sediments were uncontaminated to moderately contaminated in all the seasons. This suggests anthropogenic input of Pb in the YLC sediment. Industrial emission, combustion of fossil fuel and runoff from the surrounding roads could be the potential source of Pb in the sediment [14,21,56].

### 3.5. Temporal changes of pollution load in the sediment of YLC

The pollution load index (PLI) is used to indicate a load of metal pollution in terms of multi-elemental pollution levels in the sediment and trends of pollution in a certain period [19]. This index also shows the sediment pollution level with regard to sites, zones and catchment areas. It can estimate the overall quality of the whole catchment and quantify the baseline level of pollution in the sediment [19]. The seasonal PLI for the whole catchment is presented in Figure S9.
Figure S9 shows that winter has the highest pollution load while autumn has the lowest pollution load in the YLC sediment. This variability may be due to the climatic condition of Xiamen as wet seasons from May to August and October to April as dry seasons [45]. Higher precipitation can increase water circulation and discharge from the Yundang Lagoon resulting in lower accumulation after flooding events such as in autumn. The temporal changes of PLI values in the inlet channel, inner lagoon, outer lagoon and upstream lakes of YLC are presented in Table 1. As shown in Table 1, there is spatio-temporal variability of the pollution load of heavy metals in the sediment of the YLC areas. It was observed that in all the seasons, outer lagoon sediment has a higher pollution load of heavy metals than other areas. This may be due to higher depth and the outer lagoon has the water discharge channel of the YLC [14]. In the winter season, the PLI value was higher than in other seasons indicating greater accumulation or input of heavy metals from the surrounding areas of the YLC and lower discharge to Xiamen Bay during winter. A lower pollution load was observed in the inlet channel than in other areas. Autumn and winter have the lowest pollution load in the inlet channel than other areas suggesting that during these seasons, there is less heavy metals accumulation in the inlet channel sediment. The PLI value for upstream lakes was considerably higher than inlet channel and inner lagoon in all seasons indicating a higher anthropogenic input of pollutants from the surrounding upstream lakes areas. This may be due to lower water circulation in the upstream lakes and higher domestic sewage input from the surrounding household areas [14].

3.6. Temporal correlation between total heavy metals and sediment properties

Sediment properties such as TOC, pH and carbonate have considerable influence on heavy metals accumulation and bioavailability in the sediment of the aquatic ecosystem [25]. The temporal correlation between the total heavy metals concentration and sediment properties is presented in Tables S4 to S7. In the spring sediment, TOC has a significant positive correlation with Cu, Zn, Cd and Pb in the YLC sediment (Table S4). Moreover, heavy metal has a significantly higher positive correlation with each other such as Ni with Cr, Zn with Cu, As with Mn, Cd with Cu and Zn, and Pb with Cr, Cu, Zn and Cd (Table S4).

In the summer sediment, there is a significant positive correlation of Cu, Zn, Cd and Pb with TOC in the sediment of the YLC (Table S5). Heavy metal has a significantly higher positive correlation with each other such as Ni with Cr, Zn with Cu, As with Mn, Cd with Cu and Zn, and Pb with Zn (Table S5). In the autumn season sediment, Cu, Zn, and Cd have shown a significant positive correlation with TOC in the sediment of the YLC (Table S6). Heavy metal was also found to have a significant positive correlation with each other such as Ni with Cr, Zn with Cu, As with Mn, Cd with Cu and Zn, and Pb

Table 1. Seasonal variation of pollution load index values in sampling zones sediment in the YLC.

| Pollution load zone | Seasonal pollution load index value |
|---------------------|-----------------------------------|
|                     | Spring   | Summer  | Autumn | Winter |
| Upstream lakes      | 1.36     | 1.32    | 1.05   | 1.43   |
| Inlet channel       | 1.04     | 1.05    | 0.83   | 0.93   |
| Inner lagoon        | 1.28     | 1.17    | 0.91   | 1.32   |
| Outer lagoon        | 1.41     | 1.37    | 1.29   | 1.58   |
with Cu (Table S6). Moreover, carbonate showed a positive correlation with As in the sediment of the YLC (Table S6). In the winter season, TOC has shown a significant positive correlation with Cu, Zn, and Cd in the YLC sediment (Table S7).

Therefore, heavy metal has found a significant positive correlation with each other such as Ni with Cr, Zn with Cu, As with Mn, Cd with Cu and Zn, Pb with Cr, Cu, Zn, and Cd in the YLC sediment (Table S7). Moreover, pH has shown a significant positive correlation with Mn (Table S7).

Overall in all seasons, TOC has a significant positive correlation with Cu, Zn, Cd and Pb which indicates the formation of organic-carbon complexes with these metals by ligand flocculation [58]. Besides, the results of correlation also indicate that TOC is the main influencing factor of seasonal heavy metals accumulation in the sediment of YLC. This may be due to the higher surface area of TOC and greater absorption capacity of heavy metals in the sediment [22]. A similar result was reported previously in Yundang Lagoon where Cu, Zn, Cd and Pb were in a significant positive correlation with TOC [21]. Lake Nanhu sediment, Cd, Cr, Cu, Mn, Pb and Zn also have a significant positive correlation with TOC [59]. In all seasons, most of the heavy metals showed a strong positive correlation with each other indicating that these metals had similar behaviour, sources and transportation in the sediment of the YLC [58,59].

3.7. Temporal changes of ecological risk to heavy metals in the sediment of YLC

The ecological risk factor and risk index is a quantitative tool to evaluate the potential ecological risk of heavy metals in aquatic ecosystems for the purpose of water pollution control [18]. Therefore, risk factor and risk index can be used to identify the pollutants that need to be given more attention in aquatic ecosystems [18]. The seasonal variation of the risk factor of the studied metals in the YLC are presented in Tables S8 to S15. Spatio-temporal variation of the risk factor of the studied metals in the sediment of the YLC indicates a considerably different contamination factor or pollution level. For all seasons, Cr, Mn, Ni, Cu, Zn, As and Pb risk factor values were lower than 40, suggesting the ‘low potential ecological risk’ of these metals to the aquatic organisms. Among the studied metals, Cd showed the highest possible ecological risk to biota. The seasonal variation of risk factor values of Cd was in the range of 64–230 for spring, 32–196 for summer, 18–209 for autumn and 10–151 for winter sediments, respectively. The highest possible ecological risk factor was observed in spring and the lowest was in the winter sediment. Considering all seasons, the risk factor values of Cd were in the range of 10–230 suggesting the ‘low to higher potential ecological risk’ of Cd to aquatic organisms. So, Cd should be given more attention than other metals in the YLC [18]. In terms of multi-elemental potential ecological risk, the seasonal risk index values of the studied sites are presented in Table 2.

The risk index values were in the range of 92–276 for spring, 64–238 for summer, 46–261 for autumn and 39–224 for winter sediments, respectively. The risk index values were lower than 300 for all seasons indicating the ‘moderate ecological risk’ of heavy metals to the aquatic biota in the YLC. Therefore, upstream lakes and downstream areas of the outer lagoon sediments were at higher risk than other areas regarding the risk index values, suggesting a higher anthropogenic heavy metal input/ accumulation in these areas of the YLC.
4. Conclusions

This study investigated the temporal distribution, accumulation, speciation, and ecological risk of heavy metals in the sediment of the YLC. Results indicated that sediment pH and carbonate content showed significant seasonal variability whereas, TOC concentration showed a similar seasonal distribution pattern in the YLC. The total heavy metals of Cr, Mn, Ni, As, Cd and Pb showed a significant seasonal variation in concentration. Temporal heavy metals speciation indicates a considerable seasonal variation of metal fraction concentrations. Cr, Mn, Ni and As were mostly from natural sources whereas, Cu, Zn, Cd and Pb were mostly from the anthropogenic input in the sediment of YLC. TOC has a significant positive correlation with Cu, Zn, Cd and Pb indicating that it is the main influencing factor of heavy metals accumulation. The potential ecological risk suggests that Cr, Mn, Ni, Cu, Zn, As and Pb were ‘low potential ecological risk’ to the aquatic organisms in the YLC. The PLI indicates that the winter sediment has the highest pollution load than other seasons. As winter has the highest pollution load, this study recommends that higher water circulation and mixing in winter seasons could be possible strategies for lowering metals accumulation in sediments. Therefore, these seasonal metals accumulation in sediments, pollution load, and risks have a substantial influence on decisions, strategies and remediation of the polluted lake. As seasonal pollution data can identify the hotspot of pollution, which can trigger the management authorities to adopt strategies on a zonal and temporal basis.

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