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# Impacts of Fishing Vessels on the Heavy Metal Contamination in Sediments: A Case Study of Qianzhen Fishing Port in Southern Taiwan

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**Abstract:** Routine maintenance of fishing vessels and wastewater discharges are primary sources of heavy metals in fishing ports. Sediment pollution assessment is necessary in fishing port management, including sediment dredging and disposal, sewage treatment facility construction, and pollution source control. In this study, sediment heavy metal contents in Qianzhen Fishing Port, the largest pelagic fishery port in Taiwan, were investigated to assess the contamination levels and related potential ecological risks using multiple sediment pollution indices. Normalization methods were applied to identify the potential sources of heavy metals in fishing port sediments. Results showed that Cu, Zn, Pb, and Cr contents in the sediments of the inner fishing port (averages of 276, 742, 113, and 221 mg/kg, respectively) were 3–5 times greater compared to those along the port entrance and outside, indicating the strong impacts of anthropogenic pollution ($EF_{Cu}$: 5.6–12.5; $EF_{Zn}$: 2.8–4.3; $EF_{Pb}$: 2.4–5.4; $EF_{Cr}$: 1.1–3.2). Copper pollution was more severe, with high maxima contamination factor ($CF_{Cu}$: 15.1–24.8), probably contributed by copper-based antifouling paints used in fishing vessels. The sediments in the inner fishing port are categorized as having considerable ecological risk and toxicity ($mERMq$: 0.61–0.91; $\Sigma TU$: 7.5–11.7) that can potentially cause adverse effects on benthic organisms. Qianzhen Fishing Port sediments can be characterized as high Cu/Fe and Pb/Fe, moderate Zn/Fe, and high total grease content, indicating that the potential sources of heavy metals are primarily antifouling paints and oil spills from the fishing vessels. This study provides valuable data for pollution control, remediation, and environmental management of fishing ports.

**Keywords:** heavy metal; sediment; fishing port; fishing vessel; pollution index; antifouling paint

## 1. Introduction

Sediments act as a significant sink of heavy metals in the aquatic environment, especially in estuarine, coastal, and port regions where anthropogenic heavy metals are frequently accumulated [1,2]. Most heavy metals tend to be adsorbed on suspended solids and organic matter in seawater, then finally deposited in sediments [3]. Thus, heavy metal

contents in the sediments can better correspond to the surrounding aquatic environment quality, contrasting to the generally low concentration in the water column [4]. Even though some heavy metals such as Cu, Zn, and Ni are essential for aquatic life, they could be toxic at a certain level, posing risks to aquatic biological diversity [5,6]. As a result, heavy metals accumulated in sediments potentially cause adverse effects on aquatic organisms, resulting in long-term impacts on the aquatic ecosystem, even threatening human health through the contaminated food chain [7,8]. Therefore, sediments have been used as an important environmental indicator in assessing the impacts of anthropogenic heavy metal pollution [9,10].

Seaports are important areas for global economic and trade activities, and hence have become a land–sea coordinated area strongly influenced by human activities [11]. Sea transportation, fishery, ship maintenance, and commercial activities surrounding seaports release pollutants into the adjacent waters and sediments [12]. Ports are generally a semi-enclosed water area with dike constructions, resulting in limited water circulation and slow renewal after being polluted. Therefore, port areas are prone to accumulating considerable pollutants, particularly in the sediments which are considered as hot spots of anthropogenic pollution [13,14].

Fishing port management is an essential government role in environmental and economic aspects as it supports the fishery, being the primary food and economic resource, especially for coastal cities [15]. Determining the pollution status of fishing ports will help the planning of facilities, construction activities, formulation of management regulations, and control of pollution sources [16,17]. Fishing port sediments worldwide have been significantly contaminated with heavy metals [17–20]. Routine maintenance of fishing vessels and wastewater discharges may potentially be the primary sources of heavy metals in fishing port sediments.

In Kaohsiung City, Taiwan's third largest city, there are 16 fishing ports along the coastal zone, accommodating more than 3400 fishing boats with a total tonnage of approximately 350,000 tons; accounting for two-thirds of the total fishing vessel tonnage in Taiwan [21]. Qianzhen Fishing Port, constructed in the late 1960s, is currently the largest fishing port of pelagic fishery in Taiwan. Its annual landed catch is up to 320,000 tons, contributing 30% of Taiwan's fishery production [22]. This intensive fishery has caused the fishing port to face unavoidable impacts of heavy metal pollution. Sediment pollution assessment is necessary for fishing port management, particularly for sediment dredging and disposal, construction of sewage treatment facilities, and pollution source control. However, the lack of studies focusing on the heavy metal contamination in Qianzhen Fishing Port sediments and its associated ecological impacts hinder the development of appropriate pollution management strategies.

Therefore, this study examined the spatial variation, contamination levels, and the potential ecological risks of heavy metals (Cu, Zn, Pb, Cr, Ni, Hg, As, and Fe) in Qianzhen Fishing Port sediments using multiple sediment pollution indices. These indices include: pollution load index (*PLI*), Nemerow pollution index (*NPI*), enrichment factor (*EF*), geo-accumulation index ($I_{geo}$), mean *ERM* quotient (*mERMq*), and sum of toxic units ($\Sigma TU$). Moreover, the interrelationships between properties of the sediments and its heavy metal contents, and their potential sources along the fishing port area were evaluated.

## 2. Materials and Methods

### 2.1. Study Area

Qianzhen Fishing Port (22°34′11″ N, 120°18′49″ E) is a Class I fishing port situated in Qianzhen District of Kaohsiung City, southern Taiwan. The fishing port is located inside and connecting to the main waterway of Kaohsiung Port (the largest international container harbor in Taiwan). The wharf area is adjacent to the container terminals of Kaohsiung Port, fishery administration buildings, schools, residential areas, fueling stations, fish markets, and fishery-related factories. Qianzhen Fishing Port currently ranks as the largest oceanic fishing port in Taiwan, with a berth of ~0.27 km$^2$ and a wharf of ~3200 m. It can

accommodate approximately 300 large fishing vessels of over 500 tons, mainly including pelagic tuna and squid fishing vessels, purse seines, and trawls. The tonnage and fish catch of fishing vessels landing in the fishing port is the highest in Taiwan [21].

*2.2. Sample Collection and Analysis*

Surface sediments (approximately 0–10 cm below seafloor) were collected from 7 sampling sites (Figure 1) onboard the vessel using a Van-Veen grab, in August 2021. Site Q1 is located at the waterway outside Qianzhen Fishing Port. Site Q2 is located at the port entrance, while sites Q3–Q7 are located inside the fishing port. The digestion and measurement procedures of sediment heavy metals were conducted according to the standard method of the Environmental Protection Administration, Taiwan. Sediment samples were air-dried and homogenized using an agate mortar and pestle. A 0.5 g of sediment sample was digested using a microwave digestion system (CEM MARS-6, Matthews, NC, USA) with Teflon-lined digestion vessels. The digestion solution is a mixture of concentrated acid: 9 mL 65% $HNO_3$, 3 mL 37% HCl, 3 mL 40% HF, and 2 mL 35% $H_2O_2$ [23]. Typically, the reverse aqua regia ($HNO_3$/HCl = 3/1) has been documented as the best digestion solution for sediment sample [24]. The HF has been commonly applied for the digestion of aluminosilicate minerals matrix in sediments [25]. The $H_2O_2$ was used to enhance the decomposition of organic matter in sediment sample. Samples were digested at 180°C for 10 min, with a heating rate of 10 °C/min [23]. After microwave digestion process, the final solution was diluted to 25 mL with double distilled water (>18.2 MΩcm). Heavy metal concentrations (Cu, Zn, Pb, Cr, Ni, Hg, As, and Fe) were determined using a polarized Zeeman flame atomic absorption spectrophotometer (AAS, Hitachi ZA-3300, Tokyo, Japan). A hydride formation system (Hitachi HFS-3, Tokyo, Japan) was applied to determine Hg and As, of which Hg was determined using the cold vapor technique. To verify the validity of the results, a NIST SRM 1646a standard reference material (Gaithersburg, MD, USA) was analyzed with the same process. A certified reference material, CRM-029 (Sigma-Aldrich, Burlington, MD, USA) was also used for routine QAQC works of the heavy metal analysis. In addition to the AAS method, the alternative method, inductively coupled plasma optical emission spectrometry (ICP-OES), can also be used to determine heavy metal concentrations in the sediments [26].
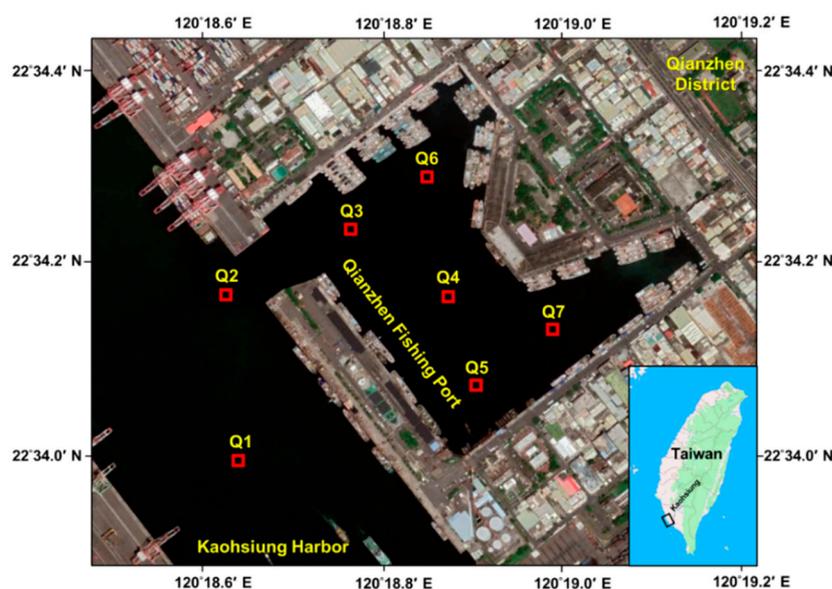


**Figure 1.** Sampling sites in Qianzhen Fishing Port, Kaohsiung City, southern Taiwan.

In addition, the sediment grain size composition was determined using a laser diffraction particle size analyzer (Beckman Coulter LS-230, Brea, CA, USA). Total organic carbon (TOC) was determined based on the Walkley–Black method [27]. The recovery of the

dextrose ($C_6H_{12}O_{16}$) standard (Sigma-Aldrich, Burlington, MD, USA) was $93 \pm 4\%$ ($n = 3$) and the reproducibility standard deviation of the TOC in sediment sample was 0.3% ($n = 5$). Total nitrogen (TN), total phosphorus (TP), and total grease (TG) were determined using USEPA Method 351.2, Method 365.1, and the APHA Method 5520E, respectively [28–30].

### 2.3. Sediment Pollution Indices

The sediment contamination levels were assessed using pollution load index (*PLI*), Nemerow pollution index (*NPI*), enrichment factor (*EF*), geo-accumulation index ($I_{geo}$), mean *ERM* quotient (*mERMq*), and sum of toxic units ($\Sigma TU$). Among them, the *EF* has been considered as a better index to compensate for the grain size effect and provide better assessment of anthropogenic pollution. Other indices, such as contamination factor (*CF*) and geo-accumulation index ($I_{geo}$), could be easily affected by the sediment particle size and mineral composition [31]. Nevertheless, those sediment pollution indices are still widely used in regional sediment heavy metal pollution assessment [32,33].

The classification of sediment contamination levels based on sediment pollution indices are detailed in Table 1. The formulas are listed as follows:

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \ldots \ldots \times CF_n}; \; CF = \frac{M_{sample}}{M_{background}} \tag{1}$$

where *PLI* is the pollution load index; $CF_i$ pertains to the contamination factor of each heavy metal; n is the number of heavy metals studied; $M_{sample}$ and $M_{background}$ are concentrations of heavy metals in fishing port sediments and the background, respectively.

**Table 1.** Classification of sediment contamination levels based on sediment pollution indices.

| Class | Contamination Level | Class | Contamination Level |
|---|---|---|---|
| | Pollution load index (*PLI*) [32] | | Nemerow pollution index (*NPI*) [34] |
| <1 | Unpolluted | <1 | Non-pollution |
| 1–2 | Slightly polluted | 1–2.5 | Low degree of pollution |
| 2–3 | Moderately polluted | 2.5–7 | A moderate degree of pollution |
| >3 | Strongly polluted | >7 | A high degree of pollution |
| | Enrichment factor (*EF*) [35] | | Geo-accumulation index ($I_{geo}$) [36] |
| <1 | No enrichment | <0 | Unpolluted |
| 1–3 | Minor enrichment | 0–1 | Unpolluted to moderately polluted |
| 3–5 | Moderate enrichment | 1–2 | Moderately polluted |
| 5–10 | Significant enrichment | 2–3 | Moderately to strongly polluted |
| 10–25 | Severe enrichment | 3–4 | Strongly polluted |
| | | 4–5 | Strongly to extremely polluted |
| | Mean *ERM* quotient (*mERMq*) [37] | | Sum of toxic units ($\Sigma TU$) [38] |
| <0.1 | Slight toxic (9% probability of being toxic) | <4 | Slight acute toxicity |
| 0.1–0.5 | Moderate toxic (21% probability of being toxic) | 4–6 | Moderate acute toxicity |
| 0.5–1.5 | Considerable toxic (49% probability of being toxic) | >6 | Significant acute toxicity |

The average contents of heavy metals in the outer Kaohsiung Harbor sediments were adopted as the background values [39].

$$NPI = \sqrt{\frac{CF_{max}^2 + CF_{mean}^2}{2}} \tag{2}$$

where *NPI* is the Nemerow pollution index; $CF_{max}$ and $CF_{mean}$ are the maxima and average contamination factor of heavy metal studied.

$$EF = \frac{(M_i/Fe)_{sample}}{(M_i/Fe)_{background}} \tag{3}$$

where *EF* is the enrichment factor; *(M_i/Fe)_{sample}* and *(M_i/Fe)_{background}* are the ratio of each heavy metal to iron content of the fishing port sediment sample and the background, respectively. This study adopted iron content for the normalization because it is abundant in the earth and not easily disturbed by anthropogenic pollution. Natural inputs mostly dominate iron sources [40]. Some studies reported that the iron content could be not appreciated for the normalization such as semi-closed waters due to early diagenesis of the redox-sensitive iron element. However, in a regional investigation, there may be no significant differences in the redox condition of sediments. The *EF* values could be more affected by the adopted background values than the selection of element used for the normalization [41]. Thus, so far, the iron content is still commonly used for the normalization of heavy metals in sediments [17,42].

$$I_{geo} = \log_2 \left( \frac{M_{sample}}{1.5 M_{background}} \right) \qquad (4)$$

where $I_{geo}$ is the geo-accumulation index; $M_{sample}$ and $M_{background}$ are concentrations of heavy metals in fishing port sediments and the background, respectively.

$$mERMq = \frac{\sum\limits_{i=1}^{n} \left( M_{sample} / M_{ERM} \right)_i}{n} \qquad (5)$$

where *mERMq* is the mean *ERM* quotient; $(M_{sample}/M_{ERM})_i$ is the ratio of each heavy metal content in the sediment sample to its effects range median (*ERM*) criteria value; *n* pertains to the number of studied heavy metals.

$$\Sigma TU = \sum\limits_{i=1}^{n} \left( \frac{M_{sample}}{M_{PEL}} \right)_i \qquad (6)$$

where $\Sigma TU$ is the sum of toxic units; $(M_{sample}/M_{PEL})_i$ is the ratio of each heavy metal content in the sediment sample to its probable effects level (*PEL*) criteria value.

## 3. Results and Discussion

### 3.1. Sediment Basic Properties

Grain size composition and organic matter contents are the two crucial properties affecting the degree of pollution in sediments. As shown in Table 2, the sediments inside Qianzhen Fishing Port (Q3–Q7) were predominately composed of silt (62.3–84.7%, 76.1% in average), followed by sand (5.2–29.0%, avg. 14.7%) and clay (6.0–10.9%, avg. 9.3%). In contrast to fine-grained sediments observed inside the fishing port, the sediments from outside the waterway (Q1) and at the port entrance (Q2) have high sand content (avg. 79.4%), but low contents of silt (avg. 18.3%) and clay (avg. 2.3%). The port is a relatively closed water area, with a 100-meter-wide entrance for water exchange, only through tides. Thus, unlike the waterways outside the fishing port where sediment dredging is frequently conducted, it is easier to deposit fine-grained sediments associated with pollutants inside the fishing port.

Similarly, sediment total organic carbon (TOC) was higher inside the fishing port (1.7–7.1%, avg. 4.3%) compared to the port entrance and outside the port (avg. 0.9%) (Table 2). In particular, the TOC content at sites Q6 and Q7 located at the innermost port area were 2–3 times greater compared to the rest of the stations inside the port (Q3–Q5). The amounts of total nitrogen (TN) in the sediments (Q3–Q7) inside the fishing port ranged between 1480–4250, while the total phosphorus (TP) was between 444–620 mg/kg. These concentrations were 2–3 times higher than sites Q1 and Q2 (Table 2). The sediment TOC, TN, and TP contents consistently showed an increasing pattern from the port entrance to the innermost port area. The total grease (TG) content also increased from the port entrance inward (Table 2), of which TG inside the port (Q4–Q7, avg. 4970 mg/kg) reached up to 17 times greater compared to outside (Q1–Q2, avg. 284 mg/kg). The high organic matter

accumulation and grease levels in the sediments may reflect the poor water exchange in the fishing port area; hence, it is classified as a pollution hot spot.

**Table 2.** Grain size (clay, silt, sand), total organic carbon (TOC), total nitrogen (TN), total phosphorus (TP), and total grease (TG) contents in the sediments of Qianzhen Fishing Port.

| Site | Clay | Silt | Sand | TOC | TN | TP | TG |
|------|------|------|------|-----|------|-----|------|
| Q1 | 2.0 | 17.4 | 80.6 | 0.8 | 1150 | 122 | 140 |
| Q2 | 2.6 | 19.1 | 78.2 | 1.1 | 1220 | 231 | 428 |
| Q3 | 10.7 | 80.1 | 9.2 | 1.7 | 1480 | 444 | 1770 |
| Q4 | 10.9 | 81.4 | 7.7 | 4.0 | 2150 | 518 | 3180 |
| Q5 | 8.7 | 62.3 | 29.0 | 2.8 | 1880 | 585 | 3110 |
| Q6 | 10.1 | 84.7 | 5.2 | 7.1 | 4250 | 538 | 7290 |
| Q7 | 6.0 | 71.8 | 22.2 | 6.1 | 3250 | 620 | 6280 |

Unit: Clay (%), Silt (%), Sand (%), TOC (%), TN (mg/kg), TP (mg/kg), TG (mg/kg).

### 3.2. Spatial Variations in Heavy Metals

Obvious spatial variation in heavy metal contents in sediments were detected in the fishing port (Table 3). Cu, Zn, and Pb exhibited an increasing pattern from the port entrance to the inner fishing port area (Figure 2). Cu, Zn, and Pb in the inner fishing port area (Q4–Q7) ranged between 226–372, 655–805, and 79–150 mg/kg, respectively. The average concentrations (276, 742, and 113 mg/kg for Cu, Zn, and Pb, respectively) were approximately 4.6, 3.2, and 3.5 times higher than those of the port entrance and outside (Q1–Q2, avg. 60, 233, and 32 mg/kg for Cu, Zn, and Pb). Comparing with the sediment quality guidelines (Table 3, Figure 2), the Cu content of site Q6 was found higher than the *ERM*, while that of sites Q4, Q5, and Q7 were relatively close. This indicates that the sediments in the innermost fishing port area have severe Cu pollution. Similarly, Zn contents in sites Q4–Q7 were much higher than the *ERM*, indicating severe Zn pollution. The contamination status of Pb was lower than Cu and Zn, displaying the content between *ERL* and *ERM* at sites Q4–Q7. Overall, it can be observed that site Q6 presented the highest heavy metal contents (e.g., Cu, Pb, Cr, and Ni) and TOC. In addition, Cr and Ni contents in site Q6 were also higher than the *ERM*, indicating serious pollution of heavy metal.

**Table 3.** Heavy metal contents (mg/kg and except for Fe in %) in the sediments of Qianzhen Fishing Port.

| Site | Cu | Zn | Pb | Cr | Ni | Hg | As | Fe |
|------|------|------|------|------|------|------|------|------|
| Q1 | 56 | 204 | 31 | 70 | 29 | 0.16 | 2.2 | 3.3 |
| Q2 | 64 | 262 | 33 | 81 | 32 | 0.12 | 3.0 | 3.7 |
| Q3 | 93 | 335 | 44 | 79 | 32 | 0.19 | 2.5 | 4.1 |
| Q4 | 226 | 655 | 79 | 177 | 38 | 0.15 | 2.5 | 3.7 |
| Q5 | 249 | 727 | 121 | 138 | 32 | 0.13 | 2.9 | 4.2 |
| Q6 | 372 | 781 | 150 | 377 | 59 | 0.16 | 2.7 | 4.5 |
| Q7 | 258 | 805 | 101 | 191 | 49 | 0.13 | 2.8 | 6.9 |
| *ERL* [a] | 34 | 150 | 46.7 | 81 | 20.9 | 0.15 | 8.2 | – [d] |
| *ERM* [a] | 270 | 410 | 218 | 370 | 51.6 | 0.71 | 70 | – |
| TEL [a] | 18.7 | 124 | 30.2 | 52.3 | 15.9 | 0.13 | 7.2 | – |
| PEL [a] | 108 | 271 | 112 | 160 | 42.8 | 0.70 | 41.6 | – |
| UCC [b] | 28 | 67 | 17 | 92 | 47 | 0.05 | 4.8 | 3.9 |
| BG [c] | 15 | 93 | 14 | 59 | 31 | 0.07 | 3.0 | 2.3 |

[a] Sediment quality guideline criteria values for effect range low (*ERL*), effect range median (*ERM*), threshold effect level (TEL), and probable effect level (PEL) for heavy metal contents in marine environments [43,44]. [b] Upper continental crust average metal contents [45]. [c] Background values of the sediments from outer Kaohsiung Port [39]. [d] –: unavailable.
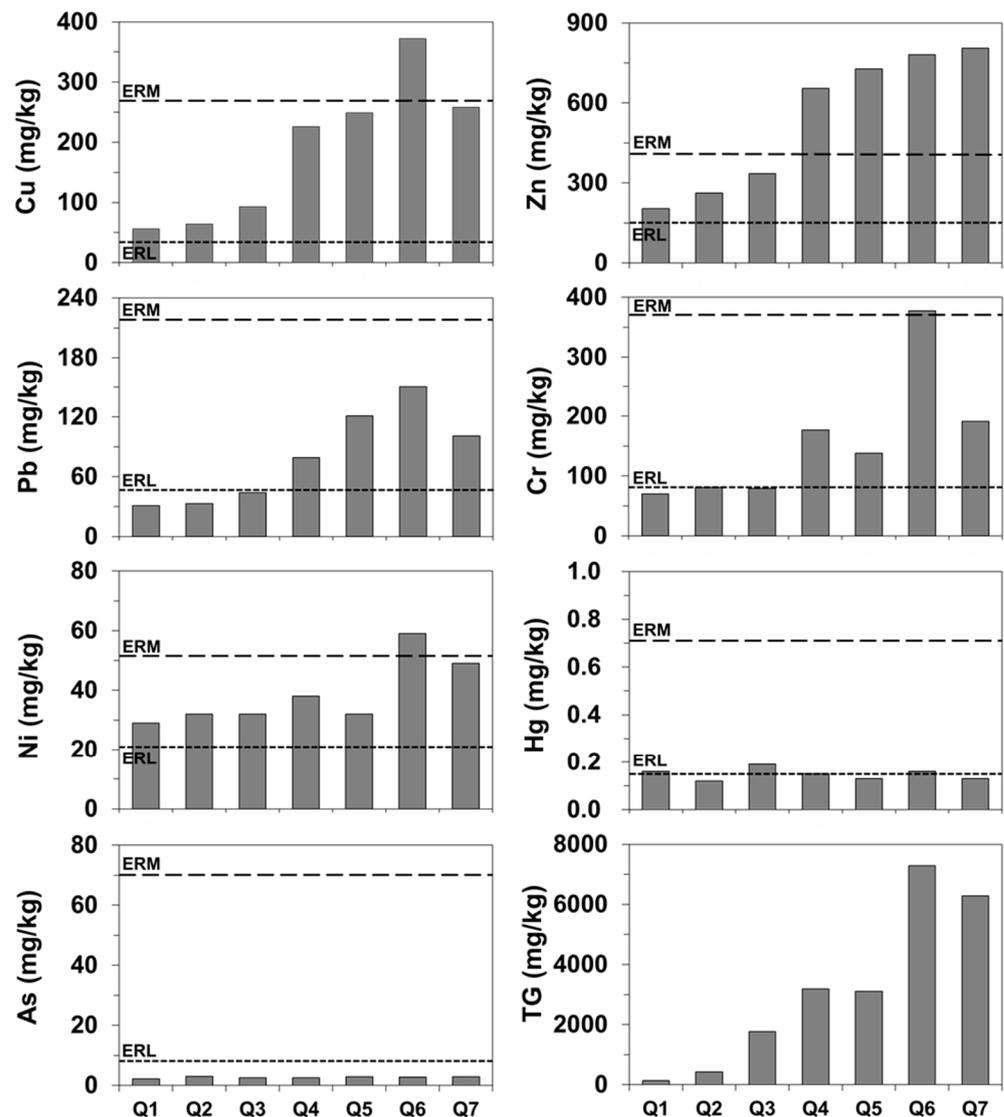
**Figure 2.** Variations in heavy metals (Cu, Zn, Pb, Cr, Pb, Ni, Hg, As) and total grease (TG) in Qianzhen Fishing Port sediments. Horizontal dotted and dashed lines indicate the values of *ERL* and *ERM*, respectively.

As indicated in Figure 1, the vicinity of site Q6 is the main berthing area of fishing boats between 200–1000 tons (categorized as CT6 and CT7). Thus, several ship hardware and repair factories are located near the quayside, increasing pollution in the area. Antifouling paints used in fishing boats typically contain heavy metals such as Cu and Zn, which can prevent the growth of fouling organisms [46]. Likewise, the paints for the decking and cabin generally contain Pb additives for corrosion resistance [47]. Heavy metals may also be enriched in the spilled oils, especially Pb [48]. Heavy metals such as Cr and Ni are commonly used in welding operations [49,50]. Cr and Ni also exist in antifouling paint residues [46]. Zhou et al. [10] reported that heavy metal contents in the sediments near the port area are primarily sourced from ship repainting and repairs. Regular maintenance of fishing boats, such as deck cleaning, ship hull painting, welding, mechanical disassembly, and repair, may flush heavy metals into the port waters [51]. Discharges of bilge water and greywater (drainage from dishwater, laundry, baths, and showers) from fishing boats also serve as potential heavy metal sources [52]. As a result, Cu, Zn, Pb, Cr, and Ni were accumulated in the sediments of the fishing port.

In contrast, As and Hg contents were relatively low in the sediments of the innermost sites (Q4–Q7), which ranged between 2.5–2.9 and 0.13–0.15 mg/kg, respectively. Concentrations were comparable between the inner fishing port area and the waterways outside the fishing port. Compared with the guidelines on sediment quality (Table 3, Figure 2), the Hg contents of sites Q1, Q3, and Q6 were slightly higher than the *ERM*, while the As contents were all lower than the *ERL*. This indicates less contamination of As and Hg in Qianzhen Fishing Port sediments in contrast with Cu, Zn, and Pb.

*3.3. Sediment Contamination Level Assessment*

Heavy metal pollution assessment in sediments is currently carried out through sediment pollution indices, normalization, and comparison with sediment quality guidelines and regulations [32,53]. In particular, pollution characteristics of heavy metal in sediments and their ecological risks can be evaluated through multiple sediment pollution indices [39,54]. Those sediment pollution indices provide comparative means for ranking the overall contamination degree of heavy metals in each site, and have been extensively utilized to obtain an integrated judgment of sediment contamination levels [32,33].

As shown in Figure 3, the sediment pollution indices exhibited an increasing trend from the port entrance to the inner fishing port area ranging between 1.7–4.8, 3.0–18.4, 0.27–0.91, and 2.9–11.7 for *PLI*, *NPI*, *mERMq*, and $\Sigma TU$, respectively. The *PLI* was proposed by Tomlinson et al. [55], which denotes how many times the overall heavy metal content in the sediment exceeds the background value. It can provide a general approach to assessing and ranking the toxicity of sediments. The highest *PLI* value was observed at site Q6 (4.8), followed by site Q7 (3.7) (Figure 3a), indicating that the inner fishing port area is strongly polluted. It is consistent with sites Q6 and Q7 being the main berthing areas of fishing boats (Figure 1). In contrast, the port entrance (Q2) and the waterway outside the fishing port (Q1) showed relatively low *PLI* values (<2), categorized as slightly polluted areas.

Like *PLI*, the *NPI* can also provide a comprehensive assessment of heavy metals in sediments. *NPI* emphasizes the most polluted metal rather than the average contamination factor (*CF*) due to the variable heavy metal impacts, even in identical studied sites [56]. The highest *NPI* value was observed at site Q6 (18.4), with a maxima contamination factor for Cu (*CF* = 24.8). Sites Q4, Q5, and Q7 showed *NPI* values of 11.2, 12.4, and 12.8, respectively (Figure 3b). These three sites also had high maxima contamination factor for Cu (*CF* = 15.1–17.2), consistently indicating that the inner fishing port area presented high degrees of metal pollution, especially for Cu where pollution is more severe. According to the *EF* assessment (Figure 3c), Cu showed significant to high enrichment (*EF* = 5.6–12.5) in the inner fishing port (Q4–Q7) sediments. Similarly, the $I_{geo}$ of Cu ranged from 3.3 to 4.0 at sites Q4–Q7 (Figure 3d), classifying these sites as strongly polluted. Copper-based antifouling paint has been used for biofouling protection of ship hulls for at least 100 years, applied in an estimated 90% of vessels worldwide [57,58]. Cu released from antifouling paints into seawater may adsorb onto suspended solids and deposit in the sediments.

The results for Zn and Pb also showed moderately to strongly polluted and moderate enrichment in the inner port sediments (Q4–Q7), with *EF* and $I_{geo}$ values ranging between 2.4–5.4 and 1.9–2.8, respectively (Figure 3c,d). In contrast, the port entrance (Q2) and the outside waterway (Q1) showed moderate enrichment for Cu (*EF* = 2.5–2.6; $I_{geo}$ = 1.3–1.5), and minor enrichment for Zn (*EF* = 2.5–2.6; $I_{geo}$ = 1.3–1.5) and Pb (*EF* = 2.5–2.6; $I_{geo}$ = 1.3–1.5). *EF* and $I_{geo}$ are useful indices to differentiate heavy metal sources between anthropogenic contamination and natural lithogenic origin [32,39]. High *EF* and $I_{geo}$ values for Cu, Zn, and Pb in inner fishing port sediments indicate that Qianzhen Fishing Port is strongly impacted by anthropogenic pollution.
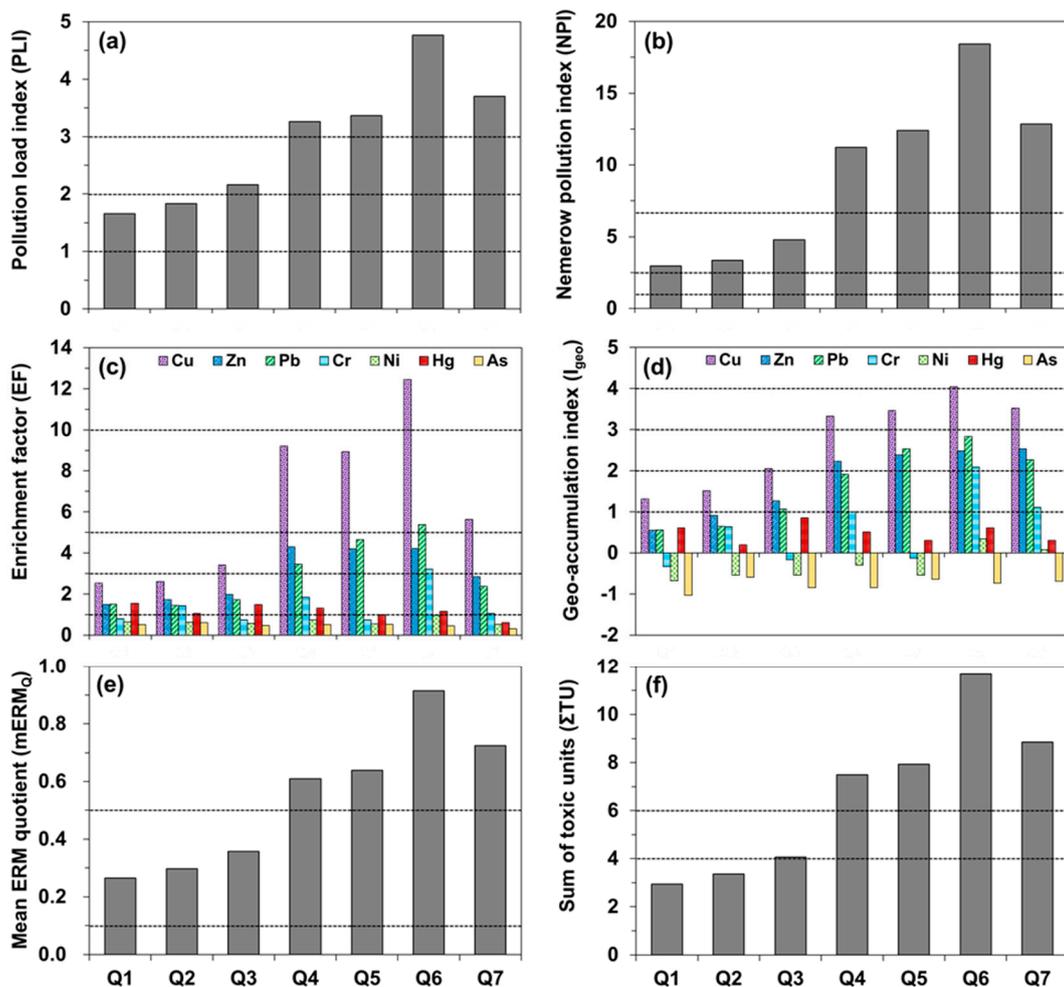
**Figure 3.** (**a**) Pollution load index (*PLI*), (**b**) Nemerow pollution index (*NPI*), (**c**) enrichment factor (*EF*), (**d**) geo-accumulation index (*I_geo*), (**e**) mean *ERM* quotient (*mERMq*), and (**f**) sum of toxic units (Σ*TU*) for heavy metals in the sediments of Qianzhen Fishing Port. The *PLI* is classified into unpolluted (<1), slightly (1–2), moderately (2–3), and strongly polluted (>3). The *NPI* is classified into non-pollution (<1), low (1–2.5), moderate (2.5–7), and high degree of pollution (>7). The *EF* is classified into nil (<1), minor (1–3), moderate (3–5), significant (5–10), and severe enrichment (10–25). The *I_geo* is divided as follows: (<0) unpolluted, (0–1) unpolluted to moderately, (1–2) moderately, (2–3) moderately to strongly, (3–4) strongly polluted, and (4–5) strongly to extremely polluted. The *mERMq* is classified into slight (<0.1), moderate (0.1–0.5), and considerable toxic (0.5–1.5). The Σ*TU* is classified into nil to slight (<4), moderate (4–6), and significant acute toxicity (>6).

### 3.4. Potential Ecological Toxicity Assessment

Several pollution indices have been proposed for sediment potential ecological toxicity assessment. Those indices are obtained by contrasting the sediment heavy metal contents with sediment quality guidelines such as the *ERL*, *ERM*, TEL, and PEL [59,60]. It is a simple way to obtain a reference for classifying sediment pollution degree and associated potential ecological risks [61]. Thus, these are commonly used methods for assessing sediment potential ecological risks, even though the actual ecological impacts of contaminated sediments need to be confirmed by on-site or laboratory toxicity experiments [9,62]. The *mERMq* and Σ*TU* were adopted for the potential ecological risk assessment in this study.

The *mERMq* was proposed by Long et al. [63], which estimates sediment toxicity for a combination of heavy metals based on the *ERM* guideline. Due to the *ERM* guideline being formulated based on numerous data that related biological effects of metals, the results of the *mERMq* assessment are generally considered reliable [60,64]. As shown in Figure 3e,

the highest *mERMq* value was observed at site Q6 (0.92), followed by site Q7 (0.72). Sites Q4 and Q5 also had high *mERMq* of 0.60–0.64. These results categorize the inner fishing port sediments as considerably toxic, presenting a 49% probability of being toxic on benthic organisms. These agree with the *PLI* and *NPI* assessment (Figure 3a,b), indicating the inner fishing port area has been severely polluted and may have adversely affected the adjacent benthic organisms.

Similarly, $\Sigma TU$ estimates the acute toxicity potential of heavy metals in the sediment studied, based on the PEL guideline. PEL is the threshold of pollutant contents where higher levels would likely cause adverse effects [65]. According to Pedersen et al. [38], $\Sigma TU$ greater than four indicates moderate toxicity on aquatic organisms, while significant mortality occurring for $\Sigma TU$ greater than six indicates significant acute toxicity. As shown in Figure 3f, the $\Sigma TU$ values of inner fishing port sediments (Q4–Q7) ranged between 7.5–11.7, with the highest in site Q6, indicating significant heavy metal toxicity in sediments. In particular, the average toxic units of heavy metals were ranked as Zn > Cu > Cr > Ni > Pb > Hg > As, indicating higher contributions of Zn, Cu, and Cr to the $\Sigma TU$. In contrast, sediments from the port entrance (Q2) and the waterway outside the port (Q1) showed relatively low $\Sigma TU$ values (2.9–3.7), indicating no potential adverse effects on aquatic organisms.

### 3.5. Potential Sources of Heavy Metals

The normalization of heavy metals is primarily used to compensate the grain-size effect [66]. Some studies also used the normalized ratio of heavy metals (e.g., metal-to-Fe ratio, metal-to-Al ratio, and metal-to-TOC ratio) to identify the characteristics and sources of metals in sediments [67,68]. In the same geological setting, similar sources and biogeochemistry of heavy metals are generally expected to have similar normalized ratios, whereas anthropogenic pollution may cause the normalized ratios to deviate from the original relationship [66].

For the geological setting of our study area, the Kaohsiung area is located on the southwestern edge of the Western Foothills Geological Zone. The whole plain ground of Kaohsiung City is mostly composed of a modern alluvial layer that is a combination of river deposits, coastal aeolian sand, and lagoon silt [69]. Hence, it is expected that the mineral compositions of the sediment around Kaohsiung area are similar. The relationships between heavy metals, total grease, and TOC contents in the sediments of Qianzhen Fishing Port are shown in Figure 4. The contents of Cu, Zn, Pb, Cr, and Ni in the sediments showed good positive linear relationships with the TOC contents ($R^2 > 0.90$), except for sites Q5 and Q6. Site Q5 displayed an outlier in Cu, Zn, and Pb from the 95% confidence interval of the regression. Similarly, site Q6 displayed an outlier in Cu, Zn, Pb, and Cr. These indicate that sites Q5 and Q6 had additional heavy metal pollution sources (e.g., antifouling paints). A strong relationship between the total grease (TG) and TOC contents was also observed in the fishing port sediments. The high TG/TOC ratio, up to 1092, suggests contamination from fuel oils.

In addition, the average normalized ratios of heavy metals (metal to Fe) in the inner fishing port sediments were 60, 160, 25, 45, 9.5, 0.03, and 0.59 for Cu/Fe, Zn/Fe, Pb/Fe, Cr/Fe, Ni/Fe, Hg/Fe, and As/Fe, respectively (Table 4). Compared to the characteristic feature of the adjacent Jen-Gen River mouth sediments, relatively higher Cu/Fe, Zn/Fe, and Pb/Fe but lower Cr/Fe were observed in the inner fishing port sediments. This suggests that heavy metal sources are not from fluvial discharge. Moreover, Cu/Fe and Pb/Fe in the inner fishing port sediments were also higher than the estuarine sediments of Love River, among the major rivers traversing through the heart of metropolitan Kaohsiung City. Hence, Qianzhen Fishing Port sediments can be characterized as high Cu/Fe and Pb/Fe, and moderate Zn/Fe, which are notably different from the characteristics of heavy metals in estuarine sediments (Table 4).
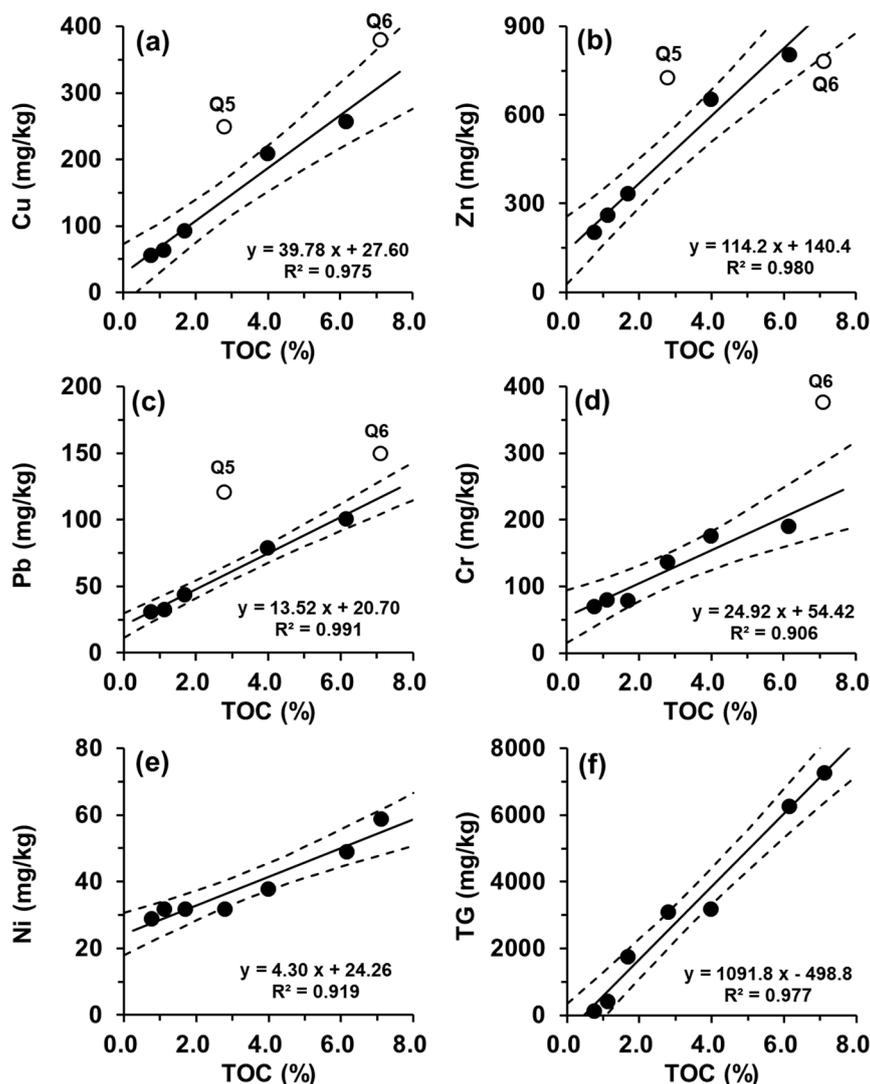
**Figure 4.** Relationships of (**a**) Cu, (**b**) Zn, (**c**) Pb, (**d**) Cr, (**e**) Ni, and (**f**) total grease (TG) with total organic carbon (TOC) contents in the sediments of Qianzhen Fishing Port. The dashed lines represent confidence interval at the 95% confidence level. The hollow circles denote the outliers.

**Table 4.** Comparison among the average contents (mg/kg) and normalized ratios of heavy metals in the sediments of Qianzhen Fishing Port and other regions worldwide.

| Station | Cu | Zn | Pb | Cr | Ni | Hg | As |
|---|---|---|---|---|---|---|---|
| Qianzhen Fishing Port, Taiwan (This study) | | | | | | | |
| Inner (Q4–Q7) | 274 (59) [a] | 742 (160) | 113 (25) | 221 (48) | 45 (9.5) | 0.14 (0.03) | 2.7 (0.59) |
| Outside (Q1–Q2) | 60 (17) | 233 (66) | 32 (9.1) | 76 (21) | 31 (8.7) | 0.14 (0.04) | 2.6 (0.74) |
| Jen-Gen River mouth, Kaohsiung, Taiwan [39] | (26) | (77) | (8.0) | (50) | (8.6) | (0.09) | – [b] |
| Love River mouth, Kaohsiung, Taiwan [39] | (42) | (220) | (14) | (52) | (14) | (0.31) | – |
| Veraval Harbor, India [17] | | | | | | | |
| Inner | 44 (15) | 765 (252) | 321 (106) | 255 (84) | 83 (27) | – | – |
| Outer | 20 (6.7) | 451 (149) | 129 (43) | 98 (32) | 40 (13) | – | – |
| Port of Ceuta, Spain [70] | | | | | | | |
| Inside | 129 (49) | 176 (89) | 110 (55) | 72 (35) | 87 (32) | – | 14 (8.1) |
| Outside | 12 (7.2) | 61 (37) | 21 (13) | 41 (25) | 38 (23) | – | 13 (7.7) |
| Ramin Port, Iran [19] | 100 (39) | 204 (79) | 99 (38) | 347 (134) | 52 (20) | – | 7.8 (3.0) |
| Inebolu Port, Turkey [51] | 452 (127) | – | – | 53 (15) | 45 (13) | – | 11 (3.0) |
| Port Kembla, Australia [71] | 839 (97) | 1824 (210) | 374 (43) | 218 (25) | – | 0.92 (0.11) | 36 (4.2) |

[a] Number in the parentheses is the normalized ratio, which was calculated by dividing the heavy metal content (mg/kg) by the iron content (%). [b] –: unavailable.

Compared to the port areas in other countries (Table 4), the sediments inside the ports similarly showed higher normalized heavy metal ratios compared to those outside the port.

The sediment characteristic featuring high Cu/Fe, Pb/Fe, and Zn/Fe probably represents a specific source of heavy metals in fishing ports—antifouling paints and oil spills. After the use of tributyltin coatings was banned in 2001, the antifouling paints composed of biocides with metal compounds, especially Cu, Pb, and Zn, have been commonly used as alternative anti-biofouling and anti-corrosive compounds [72]. The Cu, Pb, and Zn compounds released from the antifouling paints tend to adsorb onto suspended particulate matter and settle in the sediments. Paint particles had also been frequently detected in surface seawater and sediments [73]. Metal compounds such as Cu, Pb, and Zn associated with the paint particles can contribute to metal contamination in the sediments. This is why heavy metal contamination in fishing port sediments is strongly linked to ship hull cleaning operations [74]. In addition, the limited water circulation due to the wharf configuration may also enhance the deposition and accumulation of fine-grained sediments with the heavy metal pollutants in the fishing port area.

## 4. Conclusions

This study clearly showed that Qianzhen Fishing Port sediments were significantly polluted by heavy metals, particularly Cu, Zn, Pb, and Cr. Based on the *PLI* and *NPI* assessment, the sediment contamination level showed an increasing trend of up to fivefold from the port entrance to the inner fishing port sites. The high *EF* and $I_{geo}$ values of Cu, Zn, and Pb exhibited strong anthropogenic effects in the inner fishing port areas ($EF_{Cu}$: 5.6–12.5; $EF_{Zn}$: 2.8–4.3; $EF_{Pb}$: 2.4–5.4; $EF_{Cr}$: 1.1–3.2; $I_{geo\text{-}Cu}$: 3.2–4.1; $I_{geo\text{-}Zn}$: 2.2–2.5; $I_{geo\text{-}Pb}$: 1.9–2.8; $I_{geo\text{-}Cr}$: 0.6–2.1). It also poses considerable ecological risk and toxicity, considering the high values of *mERMq* (0.61–0.91) and $\Sigma TU$ (7.5–11.7). Antifouling paints used in ship hulls and oil spills from boats may have primarily contributed to the heavy metal pollution in the fishing port sediments. High Cu/Fe and Pb/Fe, moderate Zn/Fe, and high total grease contents could serve as the specific characteristic features of the fishing port sediments. Overall, this study showed the current heavy metal pollution status of Qianzhen Fishing Port, providing valuable information for fishing port management and control of heavy metal accumulation in sediments. These results further indicate the need for pollution management and remediation, e.g., sediment dredging and disposal, construction of sewage treatment facilities, and policy formulation for pollution source control.

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## References

1. Li, T.; Sun, G.; Yang, C.; Liang, K.; Ma, S.; Huang, L.; Luo, W. Source apportionment and source-to-sink transport of major and trace elements in coastal sediments: Combining positive matrix factorization and sediment trend analysis. *Sci. Total Environ.* **2019**, *651*, 344–356. [CrossRef]
2. Roussiez, V.; Ludwig, W.; Monaco, A.; Probst, J.L.; Bouloubassi, I.; Buscail, R.; Saragoni, G. Sources and sinks of sediment-bound contaminants in the Gulf of Lions (NW Mediterranean Sea): A multi-tracer approach. *Cont. Shelf Res.* **2006**, *26*, 1843–1857. [CrossRef]
3. Feng, C.; Guo, X.; Yin, S.; Tian, C.; Li, Y.; Shen, Z. Heavy metal partitioning of suspended particulate matter–water and sediment–water in the Yangtze Estuary. *Chemosphere* **2017**, *185*, 717–725. [CrossRef]
4. Singovszka, E.; Balintova, M.; Demcak, S.; Pavlikova, P. Metal pollution indices of bottom sediment and surface water affected by acid mine drainage. *Metals* **2017**, *7*, 284. [CrossRef]
5. Kumar, A.; Jigyasu, D.K.; Subrahmanyam, G.; Mondal, R.; Shabnam, A.A.; Cabral-Pinto, M.M.S.; Malyan, S.K.; Chaturvedi, A.K.; Gupta, D.K.; Fagodiya, R.K.; et al. Nickel in terrestrial biota: Comprehensive review on contamination, toxicity, tolerance and its remediation approaches. *Chemosphere* **2021**, *275*, 129996. [CrossRef]
6. Kumar, A.; Pinto, M.C.; Candeias, C.; Dinis, P.A. Baseline maps of potentially toxic elements in the soils of Garhwal Himalayas, India: Assessment of their eco-environmental and human health risks. *Land Degrad. Dev.* **2021**, *32*, 3856–3869. [CrossRef]
7. Liu, J.; Cao, L.; Dou, S. Trophic transfer, biomagnification and risk assessments of four common heavy metals in the food web of Laizhou Bay, the Bohai Sea. *Sci. Total Environ.* **2019**, *670*, 508–522. [CrossRef]
8. Pandiyan, J.; Mahboob, S.; Govindarajan, M.; Al-Ghanim, K.A.; Ahmed, Z.; Al-Mulhm, N.; Jagadheesan, R.; Krishnappa, K. An assessment of level of heavy metals pollution in the water, sediment and aquatic organisms: A perspective of tackling environmental threats for food security. *Saudi J. Biol. Sci.* **2021**, *28*, 1218–1225. [CrossRef]
9. Chen, C.F.; Ju, Y.R.; Chen, C.W.; Dong, C.D. Vertical profile, contamination assessment, and source apportionment of heavy metals in sediment cores of Kaohsiung Harbor, Taiwan. *Chemosphere* **2016**, *165*, 67–79. [CrossRef]
10. Zhou, F.; Guo, H.; Hao, Z. Spatial distribution of heavy metals in Hong Kong's marine sediments and their human impacts: A GIS-based chemometric approach. *Mar. Pollut. Bull.* **2007**, *54*, 1372–1384. [CrossRef]
11. Wang, C.; Chen, J.; Li, Z.; Nasr, E.S.A.; El-Tamimi, A.M. An indicator system for evaluating the development of land-sea coordination systems: A case study of Lianyungang port. *Ecol. Indic.* **2019**, *98*, 112–120. [CrossRef]
12. Jahan, S.; Strezov, V. Water quality assessment of Australian ports using water quality evaluation indices. *PLoS ONE* **2017**, *12*, e0189284. [CrossRef]
13. Dong, C.D.; Chen, C.F.; Ju, Y.R.; Chen, C.W. Metal pollution and ecological risk assessment in the surface sediments of Anping Harbor, Taiwan. *Desalin. Water Treat.* **2016**, *57*, 29274–29285. [CrossRef]
14. Gu, Y.G.; Gao, Y.P. An unconstrained ordination-and-GIS-based approach for identifying anthropogenic sources of heavy metal pollution in marine sediments. *Mar. Pollut. Bull.* **2019**, *146*, 100–105. [CrossRef]
15. Sciortino, J.A. *Fishing Harbour Planning, Construction and Management*; Food and Agriculture Organization of the United Nations: Rome, Italy, 2010.
16. Merhaby, D.; Ouddane, B.; Net, S.; Halwani, J. Assessment of trace metals contamination in surficial sediments along Lebanese Coastal Zone. *Mar. Pollut. Bull.* **2018**, *133*, 881–890. [CrossRef]
17. Sundararajan, S.; Khadanga, M.K.; Kumar, J.P.P.J.; Raghumaran, S.; Vijaya, R.; Jena, B.K. Ecological risk assessment of trace metal accumulation in sediments of Veraval Harbor, Gujarat, Arabian Sea. *Mar. Pollut. Bull.* **2017**, *114*, 592–601. [CrossRef]
18. Farzingohar, M.; Khakpour, Z.; Ahmadizadeh Shaghooei, M.; Soory, A. Fishing port pollution due to the vessel activities along Bandar Abbas Coast, Iran. *Int. J. Coast. Offshore Eng.* **2020**, *3*, 47–53. [CrossRef]
19. Hamzeh, M.A.; Shah-Hosseini, M.; Naderi Beni, A. Effect of fishing vessels on trace metal contamination in sediments of three harbors along Iranian Oman Sea coast. *Environ. Monit. Assess.* **2013**, *185*, 1791–1807. [CrossRef]
20. Xu, W.; Lin, T.; Hu, Z.; Yan, W. Environmental exposure and ecological risk of heavy metals in fishing harbors of the Pearl River Delta, South China. *Aquat. Ecosyst. Health Manag.* **2012**, *15*, 192–199. [CrossRef]
21. Marine Bureau, Kaohsiung City Government. 2022. Available online: https://kcmb.kcg.gov.tw (accessed on 16 February 2022). (In Chinese)
22. Fisheries Agency. *Report for Medium and Long-Term Plan (2021–2024) of Qianzhen Fishing Port Construction Project*; Council of Agriculture: Kaohsiung, Taiwan, 2021. (In Chinese)
23. Taiwan EPA. *Standard Method for Determination of Elements in Marine Discarded Materials—Microwave Digestion Method (NIEA M301.00B)*; National Institute of Environmental Analysis, Environmental Protection Administration: Taoyuan, Taiwan, 2011. (In Chinese)
24. Siaka, M.; Owens, C.M.; Birch, G.F. Evaluation of some digestion methods for the determination of heavy metals in sediment samples by flame-AAS. *Anal. Lett.* **1998**, *31*, 703–718. [CrossRef]
25. Sandroni, V.; Smith, C.M. Microwave digestion of sludge, soil and sediment samples for metal analysis by inductively coupled plasma–atomic emission spectrometry. *Anal. Chim. Acta* **2002**, *468*, 335–344. [CrossRef]
26. Benipal, G.; Harris, A.; Srirajayatsayai, C.; Tate, A.; Topalidis, V.; Eswani, Z.; Qureshi, M.; Hardaway, C.J.; Galiotos, J.; Douvris, C. Examination of Al, As, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Sb, Se, V, and Zn in sediments collected around the downtown Houston, Texas area, using inductively coupled plasma-optical emission spectroscopy. *Microchem. J.* **2017**, *130*, 255–262. [CrossRef]

27. Loring, D.H.; Rantala, R.T. Manual for the geochemical analyses of marine sediments and suspended particulate matter. *Earth-Sci. Rev.* **1992**, *32*, 235–283. [CrossRef]

28. USEPA. *Method 351.2: Determination of Total Kjeldahl Nitrogen by Semi-Automated Colorimetry*; Revision 2.0; U.S. Environmental Protection Agency: Cincinnati, OH, USA, 1993.

29. USEPA. *Method 365.1: Determination of Phosphorus by Semi-Automated Colorimetry*; Revision 2.0; U.S. Environmental Protection Agency: Cincinnati, OH, USA, 1993.

30. APHA. Method 5520: Oil and Grease. In *Standard Methods for the Examination of Water and Wastewater*, 23rd ed.; Baird, R.B., Eaton, A.D., Rice, E.W., Eds.; American Public Health Association: Washington, DC, USA, 2017.

31. Bern, C.R.; Walton-Day, K.; Naftz, D.L. Improved enrichment factor calculations through principal component analysis: Examples from soils near breccia pipe uranium mines, Arizona, USA. *Environ. Pollut.* **2019**, *248*, 90–100. [CrossRef]

32. Kim, B.S.M.; Angeli, J.L.F.; Ferreira, P.A.L.; de Mahiques, M.M.; Figueira, R.C.L. A multivariate approach and sediment quality index evaluation applied to Baixada Santista, Southeastern Brazil. *Mar. Pollut. Bull.* **2019**, *143*, 72–80. [CrossRef]

33. Duodu, G.O.; Goonetilleke, A.; Ayoko, G.A. Comparison of pollution indices for the assessment of heavy metal in Brisbane River sediment. *Environ. Pollut.* **2016**, *219*, 1077–1091. [CrossRef]

34. Yan, N.; Liu, W.; Xie, H.; Gao, L.; Han, Y.; Wang, M.; Li, H. Distribution and assessment of heavy metals in the surface sediment of Yellow River, China. *J. Environ. Sci.* **2016**, *39*, 45–51. [CrossRef]

35. Hasan, A.B.; Kabir, S.; Reza, A.S.; Zaman, M.N.; Ahsan, A.; Rashid, M. Enrichment factor and geo-accumulation index of trace metals in sediments of the ship breaking area of Sitakund Upazilla (Bhatiary–Kumira), Chittagong, Bangladesh. *J. Geochem. Explor.* **2013**, *125*, 130–137. [CrossRef]

36. Müller, G. Schwermetalle in den Sedimenten des Rheins-Veränderungen seit 1971. *Umschau* **1979**, *79*, 778–783.

37. Long, E.R.; MacDonald, D.D.; Severn, C.G.; Hong, C.B. Classifying probabilities of acute toxicity in marine sediments with empirically derived sediment quality guidelines. *Environ. Toxicol. Chem.* **2000**, *19*, 2598–2601. [CrossRef]

38. Pedersen, F.; Bjørnestad, E.; Anderson, H.V.; Kjølholt, J.; Poll, C. Characterization of sediments from Copenhagen Harbour by use of biotests. *Water Sci. Technol.* **1998**, *37*, 233–240. [CrossRef]

39. Chen, C.W.; Lim, Y.C.; Chen, C.F.; Ju, Y.R.; Dong, C.D. Spatial distribution and ecological risk assessment of sediment metals in a highly industrialized coastal zone southwestern Taiwan. *Environ. Sci. Pollut. Res.* **2019**, *26*, 14717–14731. [CrossRef]

40. Raju, K.; Vijayaraghavan, K.; Seshachalam, S.; Muthumanickam, J. Impact of anthropogenic input on physicochemical parameters and trace metals in marine surface sediments of Bay of Bengal off Chennai, India. *Environ. Monit. Assess.* **2011**, *177*, 95–114. [CrossRef]

41. Sakan, S.; Dević, G.; Relić, D.; Anđelković, I.; Sakan, N.; Đorđević, D. Evaluation of sediment contamination with heavy metals: The importance of determining appropriate background content and suitable element for normalization. *Environ. Geochem. Health* **2015**, *37*, 97–113. [CrossRef]

42. Ranjan, P.; Ramanathan, A.L.; Kumar, A.; Singhal, R.K.; Datta, D.; Venkatesh, M. Trace metal distribution, assessment and enrichment in the surface sediments of Sundarban mangrove ecosystem in India and Bangladesh. *Mar. Pollut. Bull.* **2018**, *127*, 541–547. [CrossRef]

43. Long, E.R.; MacDonald, D.D.; Smith, S.L.; Calder, F.D. Incidence of adverse biological effects within ranges of chemical concentrations in marine and estuarine sediments. *Environ. Manag.* **1995**, *19*, 81–97. [CrossRef]

44. MacDonald, D.D.; Carr, R.S.; Calder, F.D.; Long, E.R.; Ingersoll, C.G. Development and evaluation of sediment quality guidelines for Florida coastal waters. *Ecotoxicology* **1996**, *5*, 253–278. [CrossRef]

45. Rudnick, R.L.; Gao, S. Composition of the continental crust. In *The Crust, Treatise on Geochemistry*; Rudnick, R.L., Ed.; Elsevier: Amsterdam, The Netherlands, 2003; Volume 3, pp. 1–64.

46. Turner, A. Marine pollution from antifouling paint particles. *Mar. Pollut. Bull.* **2010**, *60*, 159–171. [CrossRef]

47. Zedd, H.C.; Walker, Y.P.; Hernandez, J.E.; Thomas, R.J. Lead exposures during shipboard chipping and grinding paint-removal operations. *Am. Ind. Hyg. Assoc. J.* **1993**, *54*, 392–396. [CrossRef]

48. Mustafa, A.D.; Juahir, H.; Yunus, K.; Amran, M.A.; Hasnam, C.N.C.; Azaman, F.; Abidin, I.Z.; Azmee, S.H.; Sulaiman, N.H. Oil spill related heavy metal: A review. *Malays. J. Anal. Sci.* **2015**, *19*, 1348–1360.

49. Drakopoulos, S.; Salonitis, K.; Tsoukantas, G.; Chryssolouris, G. Environmental impact of ship hull repair. *Int. J. Sustain. Manuf.* **2009**, *1*, 361–374. [CrossRef]

50. Zukauskaite, A.; Mickeviciene, R.; Karnauskaite, D.; Turkina, L. Environmental and human health issue of welding in the shipyard. In Proceedings of the 17th International Conference Transport Means, Kaunas, Lithuania, 24–25 October 2013; Kaunas University of Technology: Kaunas, Lithuania, 2013.

51. Gokkus, K.; Berber, S. Heavy metal pollution in Inebolu and Bartin Ports, Black Sea, Turkey. *Indian J. Geol-Mar. Sci.* **2019**, *48*, 1600–1608.

52. Jalkanen, J.P.; Johansson, L.; Wilewska-Bien, M.; Granhag, L.; Ytreberg, E.; Eriksson, K.M.; Yngsell, D.; Hassellöv, I.M.; Magnusson, K.; Raudsepp, U.; et al. Modelling of discharges from Baltic Sea shipping. *Ocean Sci.* **2021**, *17*, 699–728. [CrossRef]

53. Chen, C.F.; Ju, Y.R.; Chen, C.W.; Dong, C.D. Metal distributions in the Kaohsiung Ocean dredged material disposal site, Taiwan. *Desalin. Water Treat.* **2017**, *63*, 366–374. [CrossRef]

54. Lin, K.N.; Lim, Y.C.; Chen, C.W.; Chen, C.F.; Kao, C.M.; Dong, C.D. Spatiotemporal Variation and Ecological Risk Assessment of Heavy Metals in Industrialized Urban River Sediments: Fengshan River in Southern Taiwan as a Case Study. *Appl. Sci.* **2022**, *12*, 1013. [CrossRef]

55. Tomlinson, D.; Wilson, J.; Harris, C.; Jeffrey, D. Problems in the assessment of heavy-metal levels in estuaries and the formation of a pollution index. *Helgol. Meeresunters* **1980**, *33*, 566–575. [CrossRef]

56. Nemerow, N.L. *Stream, Lake, Estuary, and Ocean Pollution*; Van Nostrand Reinhold: New York, NY, USA, 1991.

57. Blossom, N.; Szafranski, F.; Yacht, A.; Lotz, A. Use of copper-based antifouling paint: A US regulatory update. *JCT Coat. Tech.* **2018**, *15*, 63–68.

58. Brooks, S.; Waldock, M. The use of copper as a biocide in marine antifouling paints. In *Advances in Marine Antifouling Coatings and Technologies*; Hellio, C., Yebra, D., Eds.; Woodhead Publishing: Sawston, UK, 2009; Volume 25, pp. 492–521.

59. Benson, N.U.; Adedapo, A.E.; Fred-Ahmadu, O.H.; Williams, A.B.; Udosen, E.D.; Ayejuyo, O.O.; Olajire, A.A. New ecological risk indices for evaluating heavy metals contamination in aquatic sediment: A case study of the Gulf of Guinea. *Reg. Stud. Mar. Sci.* **2018**, *18*, 44–56. [CrossRef]

60. Long, E.R.; Ingersoll, C.G.; MacDonald, D.D. Calculation and uses of mean sediment quality guideline quotients: A critical review. *Environ. Sci. Technol.* **2006**, *40*, 1726–1736. [CrossRef]

61. Gargouri, D.; Gzam, M.; Kharroubi, A.; Jedoui, Y. Use of sediment quality indicators for heavy metals contamination and ecological risk assessment in urbanized coastal zones. *Environ. Earth Sci.* **2018**, *77*, 381. [CrossRef]

62. Larrose, A.; Coynel, A.; Schäfer, J.; Blanc, G.; Massé, L.; Maneux, E. Assessing the current state of the Gironde Estuary by mapping priority contaminant distribution and risk potential in surface sediment. *Appl. Geochem.* **2010**, *25*, 1912–1923. [CrossRef]

63. Long, E.R.; Field, L.J.; MacDonald, D.D. Predicting toxicity in marine sediments with numerical sediment quality guidelines. *Environ. Toxicol. Chem.* **1998**, *17*, 714–727. [CrossRef]

64. Birch, G.F. A review of chemical-based sediment quality assessment methodologies for the marine environment. *Mar. Pollut. Bull.* **2018**, *133*, 218–232. [CrossRef] [PubMed]

65. Zhang, G.; Bai, J.; Zhao, Q.; Lu, Q.; Jia, J.; Wen, X. Heavy metals in wetland soils along a wetland-forming chronosequence in the Yellow River Delta of China: Levels, sources and toxic risks. *Ecol. Indic.* **2016**, *69*, 331–339. [CrossRef]

66. Kersten, M.; Smedes, F. Normalization procedures for sediment contaminants in spatial and temporal trend monitoring. *J. Environ. Monit.* **2002**, *4*, 109–115. [CrossRef]

67. Lin, S.; Hsieh, I.J.; Huang, K.M.; Wang, C.H. Influence of the Yangtze River and grain size on the spatial variations of heavy metals and organic carbon in the East China Sea continental shelf sediments. *Chem. Geol.* **2002**, *182*, 377–394. [CrossRef]

68. Sun, X.; Fan, D.; Liu, M.; Tian, Y.; Pang, Y.; Liao, H. Source identification, geochemical normalization and influence factors of heavy metals in Yangtze River Estuary sediment. *Environ. Pollut.* **2018**, *241*, 938–949. [CrossRef]

69. Tainan Hydraulics Laboratory, National Cheng Kung University. Website of Information of Land Subsidence Prevention in Taiwan. 2022. Available online: http://www.lsprc.ncku.edu.tw/en/trend.php?action=view&id=72 (accessed on 16 February 2022).

70. Guerra-García, J.M.; García-Gómez, J.C. Assessing pollution levels in sediments of a harbour with two opposing entrances. Environmental implications. *J. Environ. Manag.* **2005**, *77*, 1–11. [CrossRef]

71. He, Z.; Morrison, R.J. Changes in the marine environment of Port Kembla Harbour, NSW, Australia, 1975–1995: A review. *Mar. Pollut. Bull.* **2001**, *42*, 193–201. [CrossRef]

72. Manoj, S.; Mahesh, S.; Srikanth, N. Review of biofouling paints on the marine vessel. In Proceedings of the 2018 Asian Conference on Energy, Power and Transportation Electrification (ACEPT), Singapore, 30 October–2 November 2018; pp. 1–6.

73. Turner, A. Paint particles in the marine environment: An overlooked component of microplastics. *Water Res. X* **2021**, *12*, 100110. [CrossRef]

74. Briant, N.; Bancon-Montigny, C.; Elbaz-Poulichet, F.; Freydier, R.; Delpoux, S.; Cossa, D. Trace elements in the sediments of a large Mediterranean marina (Port Camargue, France): Levels and contamination history. *Mar. Pollut. Bull.* **2013**, *73*, 78–85. [CrossRef] [PubMed]