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Risk Assessment of Heavy Metal Distribution in Soil of a Typical Tourist Island in South China

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Abstract: *The concentrations of seven heavy metals (Hg, Cd, Pb, Cu, As, Cr and Zn) were measured in 20 topsoil samples collected from Sanjiaozhou Island to (1) reveal the distribution of heavy metals in soils after long-term human activities on an uninhabited island, and (2) analyze the ecological risks associated with soil heavy metals. According to the measured heavy metal concentrations, Zn (88.07 mg/kg) had the highest concentration, which was followed by Pb, Cr, Cu, As, Hg and Cd. Additionally, the average concentrations of Pb (73.86 mg/kg) and Hg (0.17 mg/kg) exceeded the background values 1.23 and 1.31 times, respectively. The analyzed contamination degrees, based on the contamination factor index (P_i) and geoaccumulation index (I_{geo}) values, were mostly low, but were high for Hg and Pb. The mean value of the potential ecological index (RI) of heavy metals in soils was 80.95, indicating a relatively low potential ecological risk. In terms of the individual metal ecological index (E_i) value, Hg was the highest followed by Cd, Pb, Cu, As, Cr and Zn. Hg resulted in a higher ecological risk, as its average E_i was 52.31, suggesting a moderate risk to the environment. Based on the distribution of the metals, domestic sewage discharge and agricultural inputs are most likely to be the primary sources of metal pollution on the island.*

Keywords: *island soil, heavy metal, pollution index, risk assessment*

1. Introduction

Islands are not only a vital part of the marine ecosystem, but also are key nodes for human protection and utilization of the ocean. Additionally, they have enormous ecological, economic and social value. With their unique natural landscapes creating the potential for wonderful experiences, islands make for indispensable tourist destinations [1]. As a result, tourism and tourism-induced activities, such as tourist visits, catering and accommodation, and tourism infrastructure, will inevitability pose an increasing threat to the environments of islands [2]. Due to the inherent vulnerability caused by the spatial location, isolated space and limited area, island ecosystems respond sensitively to multiple natural and anthropogenic disturbances and present variable responses of ecological resilience [3,4]. It is difficult to restore these ecosystems once they have been damaged. However, a growing number of human activities have resulted in the decline in biodiversity and ecosystem damage on islands. As human understanding and the development of islands increase, the ecological security and sustainable development of islands are becoming increasingly important. Despite this, the health of island ecosystems has been given little attention.

In previous studies, vegetation, soil and landscape have been identified as fundamental parts of an island ecosystem [5].Additionally, the soil is a link between the atmosphere, lithosphere, hydrosphere and biosphere [6], and is an essential element of ecosystem functioning and services [7]. There are many severe problems due to soil contamination in China, particularly related to heavy metals [8]. Because of the toxicity, bioaccumulation and persistence of heavy metals, they pose a significant threat to the natural environment [9]. Understanding the concentration and spatial distribution of soil heavy metals and assessing their ecological risks are critical for the accurate and effective implementation of risk management and control of soil heavy metal pollution [10]. In terms of soil heavy metal distribution characteristics and ecological risk assessments, most current research has focused on mainland ecosystems, with little attention paid to islands [9,11-13]. In this study, Sanjiaozhou Island, an important and typical tourist island in Daya Bay, southern China, was selected to investigate the heavy metal pollution in the surface soils and to identify the influence of human activities. The objectives were: (1)

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to reveal the distribution of soil heavy metals after long-term human activities on an uninhabited island, and (2) to assess the potential ecological risks of soil heavy metals. The results of this study will be useful for the planning, risk assessing, and decision making carried out by environmental managers in this region.

2. Materials and Methods

2.1. Study area and sampling

Surface soil samples were collected from Sanjiaozhou Island, Huidong County, located between E 114°43′37″~114°43′56″ and N 22°37′25″~22°37′46″ in Daya Bay, southern China. Sanjiaozhou Island is an uninhabited island that has been developed for tourism. It is approximately 1.10 km from the mainland, with many surrounding ports. The study area is a rocky island consisting of two parts: the east island and west island, together covering approximately 0.10 km^2. The island has a warm and humid climate with an average temperature of 22.5 ℃, an annual precipitation of 1434.9 mm and a relative humidity of 75%. The soil type on the island is mainly lateritic red soil originating from granite. There are large areas of artificial orchards on the island. The soil of the island forest is relatively thin, and the soil particle size is coarse.

About 20 composite soil samples were collected during December 2021 from the study area (Figure 1). Portable global positioning systems (GPS) were used for locating sampling sites. Soil samples were collected in the form of sub-samples at each sampling site (up to a 20 cm depth). A composite sample was created by thoroughly mixing five sub-samples from each site. Following the removal of stones, coarse plant roots and residues from the samples, the samples weighed approximately 1000 g. A zip-lock bag with labels was used to store the samples, which were then returned to the laboratory. The air-drying soil samples under natural conditions was carried out to avoid external interference, and they were ground and sieved through a 0.15 mm sieve to ensure even mixing for the later analysis [14].

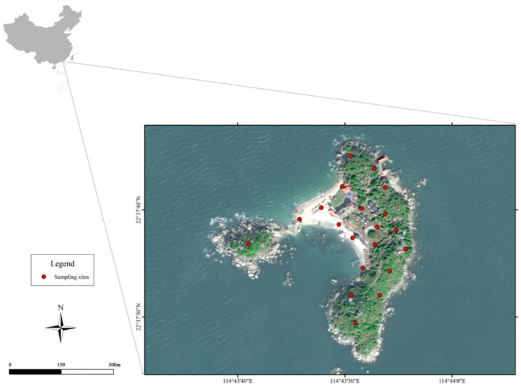

Figure 1: The map of locations of sampling sites in the study area.

2.2. Sample analysis

Weighing and placing soil samples into Teflon digestion vessels for trace element analysis involved approximately 0.300 g of soil samples. First, a mixture of 6 ml of 12 M HCl and 2 ml of 17 M HNO_3 was used to treat the soil samples. After pretreatment, a microwave digestion system (SpeedWave MWS-4, Germany) with automatic pressure and temperature regulation was employed to digest the samples. After that, the digested solution was filtered through a 0.45-micron filter into a centrifuge tube for further analysis.

A flame atomic absorption spectrophotometer (Agilent AA240FS, USA) was used to determine the concentrations of Pb, Cr, Cu and Zn in the solution, whereas a graphite furnace atomic absorption spectrophotometer (Agilent AA240Z, USA) was used to determine the concentrations of Cd [15]. Additionally, the concentrations of Hg and as were determined by atomic fluorescence spectrometry (Aurora Lumina 3400, Canada) [16]. All chemicals were analytical-grade reagents. In order to ensure quality control, analytical-grade reagents were used in all chemical analyses, and duplicate analyses were performed for each sample.

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2.3. Calculation of soil pollution indices

The contamination factor (P_i) and geoaccumulation index (I_{geo}) were used to measure the pollution extent, while the ecological risk index (E_i) was used to assess the potential degree of impact of metals in soils on the environment.

2.3.1. Contamination factor (P_i) and degree of contamination (C_{deg})

The P_i was quantified as the ratio of the measured concentration to the baseline or background value of a single heavy metal. Its expression is shown in equation (1).

$$P_i = \frac{C_i}{S_i} \tag{1}$$

where C_i and S_i represent the measured concentrations and the background concentrations of each heavy metal, respectively. In this paper, the background values are reference values of metals (mg/kg) obtained from the standards for soil environmental quality in the Pearl River Delta, Guangdong Province (DB 44/ T1415-2014).

The P_i values can reflect soil enrichment over time with a given metal [11]. According to the P_i values, the intensity of contamination can be classified from 1 to 6: a low degree when $P_i < 1$, moderate degree when $1 \le P_i < 3$, considerable degree when $3 \le P_i < 6$ and very high degree when $P_i \ge 6$ [17].

The degree of contamination (C_{deg}) was calculated by adding together the derived contamination factors (P_i) as indicated in equation (2).

$$C_{deg} = \sum_{i=1}^{n} Pi \tag{2}$$

The grades of the degree of contamination (C_{deg}) are: low degree ($C_{deg} < 8$), moderate degree ($8 \le C_{deg} < 16$), considerable degree ($16 \le C_{deg} < 32$) and very high degree ($C_{deg} \ge 32$) [18].

2.3.2. Geoaccumulation index (I_{geo})

Initially proposed by Muller in 1969 [19], the geoaccumulation index (I_{geo}) has been widely used in heavy metal studies ever since. With the I_{geo}, soil contamination can be assessed by comparing current concentrations with preindustrial ones and assessing the accumulation of heavy metals in soil [20]. The I_{geo} is computed using equation (3)

$$I_{geo} = \log_2[C_i / (k * B_i)] \tag{3}$$

In this equation, the measured concentration of heavy metal i in soil is C_i, and B_i is its geochemical background concentration or pristine value. In the present study, the B_i of an individual metal is a reference value of metals (mg/kg) obtained from the standards for soil environmental quality in the Pearl River Delta, Guangdong Province (DB 44/ T1415-2014). The term k is 1.5, which is the constant factor introduced to analyze natural variations in the contents of a given metal based on its surroundings and very small anthropogenic influences. The I_{geo} was distinguished into seven classes by Muller [21], as shown in Table 1.

Table 1: Pollution grades of the geoaccumulation index of the metals.

I_{geo} value	I_{geo} class	Grades of quality
≤ 0	0	Unpolluted
0 ~ 1	1	Unpolluted to moderately polluted
1 ~ 2	2	Moderately polluted
2 ~ 3	3	Moderately to heavily polluted
3 ~ 4	4	Heavily polluted
4 ~ 5	5	Heavily to extremely polluted
> 5	6	Extremely polluted

2.3.3. Potential ecological risk index (RI)

The ecological risk factor (E_i) quantitatively expresses the potential threat to the ecological environment associated with a given single contaminant. Additionally, the potential ecological risk index (*RI*) evaluates the adverse effects of the human-caused contaminants on the environment, which is the sum of *Ei*. This index was proposed by Hakanson in 1980 [22] in order to more accurately assess

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the toxicity and ecological sensitivity of contaminants. Equation (4) shows the calculation of the *RI*.

$$RI = \sum E_i = \sum T_i(C_s^i / C_n^i) \tag{4}$$

where C^i_s and C^i_n are the measured concentrations and the background concentrations of the heavy metal *i*, respectively. T_i is the response coefficient for the toxicity of the single heavy metal *i*. The toxic response factors for Zn, Cr, Pb, Cu, As, Cd and Hg were 1, 2, 5, 5, 10, 30 and 40, respectively [23,24]. The *RI* is the comprehensive potential ecological index, which is the sum of *Ei*. The grading standards for the potential ecological risk of heavy metals are shown in Table 2 [22].

Table 2: Indices and grades of ecological risk of heavy metal contamination.

Scope of E_i	Risk grade of single metal	Scope of *RI*	Risk grade of multiple metals
< 40	Low	< 150	Low
40 ~ 80	Moderate	150 ~ 300	Moderate
80 ~ 160	Considerable	300 ~ 600	Considerable
160 ~ 320	High	≥ 600	Serious
≥ 320	Serious		

2.4. Statistical analysis

Statistical analysis was conducted with SPSS 22.0. The means and standard deviations of the trace element concentrations in soils were calculated. Different heavy metals were correlated using Pearson's correlation analysis. The spatial analysis of metal concentrations and the potential ecological risk indices were mapped by inverse distance weight interpolation in ArcGIS [25].

3. Results and discussion

3.1. Heavy metal concentrations

Table 3: Heavy metal levels in soils on Sanjiaozhou Island: descriptive statistics (n = 20, mg/kg), [a] background values and threshold values [b]: according to the standards for soil environmental quality in the Pearl River Delta, Guangdong Province (DB 44/ T1415-2014).

Metals	Min value	Max value	Mean (±SD)	CV(%)	Background values [a]	Threshold values [b]
Zn	12.08	277.61	88.07 ± 62.68	71.17	97.00	320.00
Cr	8.96	80.62	50.59 ± 22.66	44.78	77.00	260.00
Pb	24.70	126.33	73.86 ± 30.55	41.37	60.000	100.00
Cu	6.23	37.38	15.33 ± 6.92	45.13	32.00	145.00
As	1.06	18.00	3.75 ± 3.68	98.20	25.00	40.00
Cd	0.01	0.16	0.06 ± 0.04	79.46	0.11	0.80
Hg	0.05	0.32	0.17 ± 0.07	40.18	0.13	1.00

Concentrations of Zn, Cr, Pb, Cu, As, Cd and Hg, as well as the background values and threshold values for these pollutants in soil, are listed in Table 3. According to the measured heavy metal concentrations, Zn had the highest concentration, and was followed by Pb, Cr, Cu, As, Hg and Cd. The maximum concentration of Zn was 277.61 mg/kg and its average concentration was 88.07 mg/kg, which was probably influenced by external factors including human activities. The concentration values of the seven heavy metals were lower in the study area than the threshold values from the soil environmental quality standards of the Pearl River Delta, Guangdong Province. However, compared with the background values of heavy metals in the Pearl River Delta, the average concentrations of Pb and Hg exceeded the standards 1.23 and 1.31 times, respectively. The results indicate that soils were contaminated with Pb and Hg at the majority of study locations. In addition, many sampling sites had higher heavy metal concentrations than their corresponding background values by several orders of magnitude. Additionally, it was observed that from Table 3, most of the measured concentrations of heavy metals showed high coefficients of variation (C.V.). Moreover, the concentrations of heavy metals were found to be within a wide range as well, such that there was a more than five-fold difference in values between the maximum and minimum measured of each heavy metal. Such a deviation might be traced to a lack of uniformity in the elemental distribution across the island, indicating the characteristics of the point source of pollution in the study area. A significant portion of these heavy metals of soils in this research area are likely to be imported from anthropogenic sources [26].Based on the above analysis, there is a greater threat from Hg and Pb pollution, and thus, they deserve special attention. Although the average concentration of Hg was low, being only 0.17 mg/kg, it is toxic even at low concentrations.

3.2. Heavy metal correlation analysis and spatial distribution

Generally, there are two sources of heavy metals found in soil: a natural origin and anthropogenic inputs. The soil's parent material may be a major factor in controlling heavy metals in the soil on an uninhabited island. However, with the development of island tourism, human activities introduce more forms of heavy metal pollution. Seven heavy metals were correlated using Pearson's correlation analysis, and the results are shown in Table 4. Significant correlated coefficients between metal elements in soils could indicate a common source [27]. For instance, a high correlation was observed between Zn, Cd and Cu in this study, which was in agreement with the results of other investigations published in the past [8,28]. Additionally, researchers have shown that the main source of Zn, Cu and Cd is anthropogenic inputs from agricultural activities [29].

The spatial distribution characteristics of the seven heavy metal contents in the soils of Sanjiaozhou Island are presented in Figure 2. Overall, high levels of all metals are located at sites located in the north and middle of the east island, which correspond with areas of crop farming and where various tourism and entertainment facilities exist. Additionally, these areas are low-lying and prone to heavy metal enrichment. According to Figure 2 and Table 4, Pb and Cd have similar spatial distributions, and the correlation between Pb and Cd was found to be significant, suggesting that there is a possibility that Pb and Cd had the same source. In fact, it has been established that Cd often exists with Pb in nature [30]. As seen in Figure 2, an area where soils were polluted with Cr and Zn was located in the center of the east island and was surrounded with orchard. Agricultural inputs were probably the main cause of these high concentrations. For example, the use of phosphate fertilizers contributed significantly to the introduction of heavy metals into agricultural soils, particularly for the metals Zn, Cr, Cd and Cu [31]. In addition, it is also important to note that atmospheric deposition and the stacking of various wastes contribute a great deal to soil pollution. In Ogundele's studies [12], it was determined that the heavy metal ingredients of solid wastes are mainly absorbed in soil. Much previous research has demonstrated that one of the main source of heavy metals in soil is atmospheric deposition [31-33]. Additionally, compared with other measurements around the world, the deposition flux in the Pearl River Delta region was found to be relatively higher [32].

Soil heavy metal pollution is a long-term and dynamic process that is easily influenced by regional environmental changes or human activities. For areas with high heavy metal contents, corresponding preventive measures should be taken, such as strengthening the prevention and control of domestic sewage discharge. For areas with low heavy metal contents, monitoring and forecasting should be strengthened.

Table 4: Result of Pearson's correlation analysis for seven heavy metals in soils. is significant at the 0.01 level.*

Metals	Zn	Cr	Pb	Cu	As	Cd	Hg
Zn	1						
Cr	0.565*	1					
Pb	0.240	0.252	1				
Cu	0.454*	0.534*	0.437	1			
As	-0.093	0.250	-0.114	0.206	1		
Cd	0.491*	0.281	0.512*	0.735*	0.031	1	
Hg	0.180	0.611*	0.279	0.425	0.665*	0.144	1

3.3. Contamination levels of soil heavy metals

According to Table 5, in terms of Zn, Cr, Cu, As and Cd, the P_i values were lower than 1.0, meaning there was low pollution. In terms of Pb and Hg, the P_i values were in the range of 1.0 and 3.0, indicating moderate contamination. A low degree of contamination is implied by the overall degree of contamination (C_{deg}) of 5.29, which is less than the lower limit of 8. We analyzed all 20 samples and showed that only the P_i of As was less than 1 in all samples, indicating that arsenic has a low contamination status in all soils on the island. In other words, the contamination levels of the other heavy metals are unevenly distributed and wide-ranging. The highest P_i values obtained for Zn ranged from 0.12 to 2.86. Additionally, the P_i values of Hg and Pb were greater than 1.0 in 75% and 65% of the samples, respectively.

As shown in Table 6, the contamination levels of heavy metals in the study area were in the decreasing order of Hg > Pb > Zn > Cr > Cd > Cu > As based on the I_{geo} value. The average values of the I_{geo} of heavy metals ranged from -3.32 to -0.20, since these values were less than 0, the soils were indicated as being uncontaminated. However, in a proportion of the samples the levels of Zn, Pb and Hg indicated uncontaminated to moderately contaminated soils. In summary, heavy metal pollution was mainly caused by Hg in the study areas, which is similar to previous results from heavy metal pollution assessments of sediments in Daya Bay [34]. The average concentration of Hg in island soils in Daya Bay was 0.107 mg/kg in 1990 [35], which is much lower than the soil environmental background value in the Pearl River Delta. In this survey, the average Hg content in the soils of Sanjiaozhou Island increased by 59%, exceeding the soil environmental background value in the Pearl River Delta.

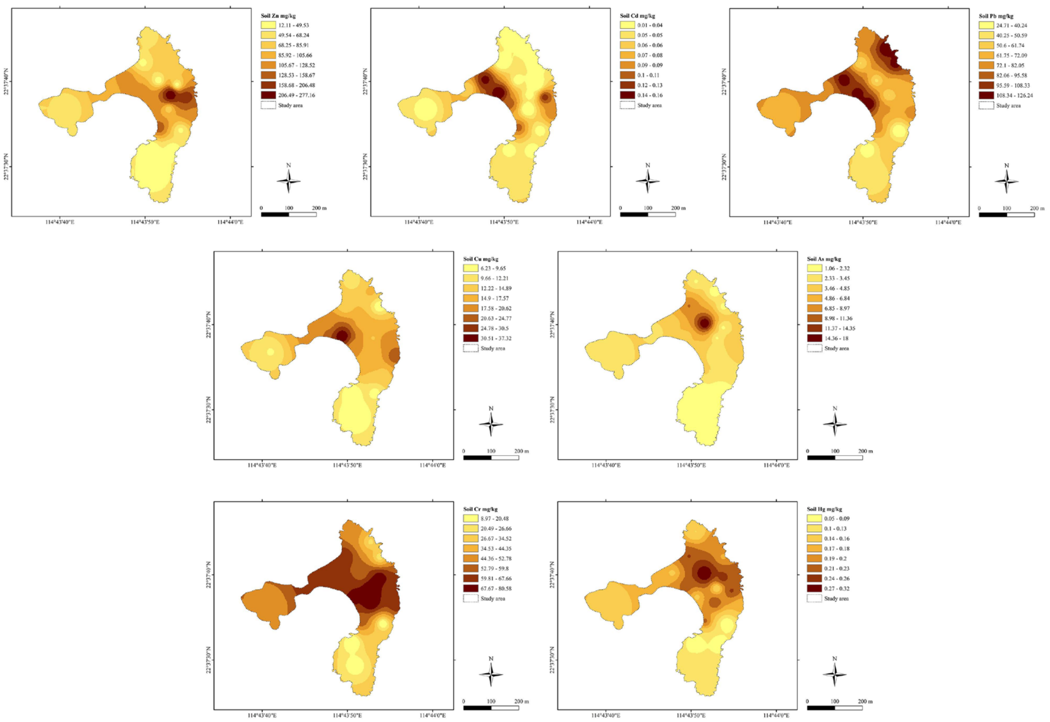

Figure 2: Spatial distribution of seven heavy metals in Sanjiaozhou Island soils

Table 5: Contamination factors of heavy metals in soils from Sanjiaozhou Island.

Metals	Range of P_i	Mean of P_i	Proportion of $P_i < 1$	Proportion of $1.0 \leq P_i < 3.0$
Zn	0.12 ~ 2.86	0.91	65%	35%
Cr	0.12 ~ 1.05	0.66	90%	10%
Pb	0.41 ~ 2.11	1.23	35%	65%
Cu	0.19 ~ 1.17	0.48	95%	5%
As	0.04 ~ 0.72	0.15	100%	0
Cd	0.09 ~ 1.45	0.55	85%	15%
Hg	0.38 ~ 2.46	1.31	25%	75%

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Table 6: Geoaccumulation index of heavy metals in soil samples.

Metals	I_{geo}			*Proportion of sample and grades of quality*	
	Min	*Max*	*Mean*	*Uncontaminated*	*Uncontaminated to moderately contaminated*
Zn	*-3.59*	*0.93*	*-0.72*	*85%*	*15%*
Cr	*-3.69*	*-0.52*	*-1.19*	*100%*	*0*
Pb	*-1.87*	*0.49*	*-0.29*	*75%*	*25%*
Cu	*-2.95*	*-0.36*	*-1.65*	*100%*	*0*
As	*-5.14*	*-1.06*	*-3.32*	*100%*	*0*
Cd	*-4.04*	*-0.04*	*-1.46*	*100%*	*0*
Hg	*-2.05*	*0.73*	*-0.20*	*65%*	*35%*

3.4. Potential ecological risk index results

The ecological risk (E_i) of soil was calculated using the P_i values in Table 4, and the results are shown in Table 7. There were less than 40 values of E_i at all sites for Zn, Cr, Cu, As and Cd, thus suggesting little ecological risk from these five metals on Sanjiaozhou Island. Hg and Cd represent the main potential ecological risk, especially as Hg's average value of E_i was 52.31, suggesting a moderate potential ecological risk. Out of all the sampling sites, 75% had a moderate to considerable ecological risk due to Hg, and 5% had a moderate risk due to Cd. There was a trend in the E_i values of the seven heavy metals as follows: Hg > Cd > Pb > Cu > As > Cr > Zn. This trend shows distinct differences between the E_i and I_{geo} ranking of the seven heavy metals. The geoaccumulation index method mainly reflects the degree of heavy metal enrichment [24,36], while ignoring the toxic response factors of the different heavy metals. Relatively high Zn and Pb contents lead to correspondingly higher geoaccumulation index values. The ecological risk index mainly considers the toxicity of the different heavy metals [9]. Additionally, the toxic response factors of Cd and Hg are much higher than those of other heavy metals, which leads to larger E_i values for Cd and Hg. Moreover, certain heavy metals such as Cr and Zn have a granule affinity. They are prone to migrate with other particulate matter and enter the soil for mineralization and burial, causing their biological toxicity and ecological risk to be reduced.

There is a low potential ecological risk in the study area, with an *RI* value between 28.69 and 128.05, and a mean value of 80.95. Hg was the highest contributor to the *RI*, accounting for 64.62%, which was followed closely by Cd, accounting for 20.21%. The potential ecological risk indices for all the studied soil samples were distributed based on their site-specific characteristics (Figure 3.). Overall, the areas with the most significant ecological risk are located in the northern part of Sanjiaozhou Island's east island. This characteristic is in agreement with the distribution of human activities. This may indicate that these heavy metals in the studied regional soil possibly result mainly from human activities, such as domestic sewage discharge and agricultural inputs. The other areas were covered by extensive virginal vegetation and had relatively lower ecological risk index values. Vegetation such as ferns can reduce the contents of heavy metals in the surrounding soils as their roots uptake them from the soils [13,37].

Table 7: The ecological risk factor (Ei) of soil heavy metals.

Metals	E_i			Proportion of sample and risk degree		
	Min	Max	Mean	Low	Moderate	Considerable
Zn	0.12	2.86	0.91	100%	0	0
Cr	0.23	2.09	1.31	100%	0	0
Pb	2.06	10.53	6.16	100%	0	0
Cu	0.97	5.84	2.40	100%	0	0
As	0.42	7.2	1.50	100%	0	0
Cd	2.73	43.64	16.36	95%	5%	0
Hg	14.46	99.69	52.31	25%	70%	5%

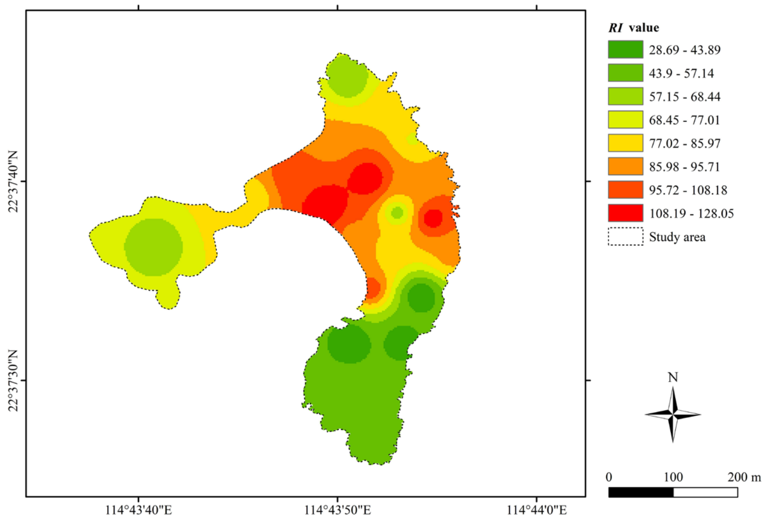

Figure 3: Distribution of potential ecological risk of heavy metals in Sanjiaozhou Island soil.

4. Conclusions

In this study, a series of metal (Zn, Cr, Pb, Cu, As, Cd and Hg) concentrations were determined in soils of Sanjiaozhou Island. Furthermore, the pollution status and potential ecological risk of the heavy metals were evaluated. Zn had a maximum average concentration of 88.07 mg/kg, and the average concentrations of Pb (73.86 mg/kg) and Hg (0.17 mg/kg) were 1.23 and 1.31 times their background values, respectively. According to the P_i and I_{geo} values, the Hg contamination level was highest in the study area, followed by Pb, Zn, Cr, Cd, Cu and As in decreasing order.

An average value of 80.95 was found for the *RI* in the study area, with values ranging from 28.68 to 128.14. Hg was the highest contributor to the *RI*, accounting for 64.62%, and its average E_i was 52.31, suggesting a moderate potential ecological risk. The order of the mean *Ei* values is Hg > Cd > Pb > Cu > As > Cr > Zn. The distribution of these metals in the soils of the island indicate that domestic sewage discharge and agricultural inputs are possibly mainly responsible for metal pollution. By combining P_i, I_{geo}, E_i and *RI* with their grade classifications, it was found that the overall contamination level and potential ecological risks of heavy metals on Sanjiaozhou Island were relatively low. For individual metals, Hg displayed a higher ecological risk, as it was found to be a moderate to considerable ecological risk for most of the sites. The long-term monitoring of heavy metal concentrations in the soil is recommended in order to protect the health of tourists and the soil environment.

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