Article

Biological Risk Assessment of Heavy Metals in Sediments and Health Risk Assessment in Marine Organisms from Daya Bay, China

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Abstract: The concentrations of heavy metals in sediments and marine organisms in Daya Bay were investigated, and the Monte Carlo method was used to analyze the uncertainty of the results of geo-accumulation characteristics and ecological and health risks. The mean concentrations of metal elements in sediments were in the following order: Zn > Cr > Cu > As > Cd > Hg, while those in marine organisms were Zn > Cu > As > Cr ≈ Cd > Hg. The geo-accumulation index (I_{geo}) indicated that the primary pollutant was Hg, with 5.46% moderately polluted, and 39.52% for unpolluted to moderately polluted. Potential ecological risks (RI) were between low and high risks, and the contributions of Hg, Cd, and As to ecological risks were 50.85%, 33.92%, and 11.47%, respectively. The total hazard coefficients (THQ) were less than 1, but on the basis of total carcinogenic risks (TCR), the probability of children and adults exceeded the unacceptable risk threshold of 22.27% and 11.19%, respectively. Sensitivity analysis results showed that the concentrations of carcinogenic elements contributed to risk in the order of As > Cd > Cr. Therefore, in order to effectively control heavy metals contamination in Daya Bay, it is necessary to strengthen the management of Hg, Cd, and As emissions.

Keywords: heavy metals; ecological risk; human health risk; Monte Carlo; Daya Bay

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1. Introduction

The impact of heavy metal pollution on the sustainability of biogeochemical cycles and ecological risks has received increasing global attention [1]. Heavy metals have special eco-toxicological effects, such as toxicity, accumulation, and refractory degradation [2]. When heavy metals enter the aquatic environment, they eventually accumulate in sediments, so sediments are usually used as a main medium for evaluating heavy metal pollution [3]. However, when the environmental conditions change, these are released from the sediments, and then used as a source to return to the overlying water, threatening the aquatic environments and marine organisms [2,4,5]. Marine organisms are rich in proteins, low saturated fatty contents and Omega-3 fatty acids, and other nutrients needed by the human body [6]. However, due to the enrichment and amplification of heavy metals in marine organisms, seafood that originally provided the human body with abundant protein may become a carrier of concentrated poisons, thereby endangering human health [7–11]. Therefore, identifying the concentration distribution of heavy metals in sediments and marine organisms and assessing the ecological and health risks under heavy metal exposure are of great significance to the protection of biological habitats and human safety.

Daya Bay is a semi-enclosed bay in the northwest of the South China Sea and is a key protected area for fishery resources [5,6]. Since the 1980s, with the rapid development of industrialization and urbanization, as well as the rapid growth of population, Daya Bay has been affected by many aspects of pollution, such as aquaculture, port, petrochemical, nuclear power plant, wind power plant, etc. [12,13]. Moreover, there are reports that since the construction of the nuclear power plants, the concentrations of metal elements in sediment cores of Daya Bay had been dramatically increased [14,15]. Gu et al. [12] proposed that the “hot spot” area in the Daya Bay near the nuclear power plants seems to have contributed metallic elements through nuclear waste discharge to the sediments in study area. Although many scholars have conducted studies on the seawater, sediments, and heavy metal pollution of marine organisms in Daya Bay and reported their ecological or health risks [5,6,10,13,16,17], there has been no targeted research on the pollution of existing nuclear power plant sources, or the sampling density has been insufficient. Thus, there is an urgent need for improved high-resolution heavy metal index sampling in the sea area surrounding the nuclear power plants in the Daya Bay. In this study, we adopted the Monte Carlo method to obtain large samples of heavy metals index data through statistical sampling experiments on the computer to accommodate uncertainties associated with risk-related problems [18]. The purposes of this study were (1) to develop a combination of traditional evaluation models and Monte Carlo simulation to optimize the uncertainty of risk assessment; (2) investigate the concentration distribution and pollution of heavy metals in the sediments and marine organisms in the sea area surrounding the nuclear power plants in the Daya Bay; and (3) examine toxic effects and risk assessment under exposure of heavy metals.

2. Materials and Methods

2.1. Study Area and Samples Collection

The study area is located in the southwestern region of Daya Bay in Guangdong Province, South China. In January and March 2020, 27 surface sediment samples and 20 species of marine organism samples were collected, respectively (Figure 1). The sediment samples were collected by precleaned stainless steel grab sampler, and two sediment samples were collected at each site to prepare a mixed sample. Marine organisms were collected by horizontal trawl and intertidal surveys, and selected adult individuals as samples. The details of organism samples are shown in Table S1. All samples collected from Daya Bay were placed in polyethylene sealed bags and stored at -4°C until processing and analysis.

2.2. Chemical Analysis and Quality Assurance

The sediment samples were freeze-dried to a constant weight, ground with an agate mortar and pestle, passed through a $63\text{ }\mu\text{m}$ sieve, and stored at -20°C in brown glass bottles. Heavy metal microwave digestion was conducted in accordance with the EPA Method 3050B using an Ethos Plus Microwave Labstation (Milestone Inc., Italy).

The marine organism samples were first thawed naturally at room temperature, then rinsed with deionized water for 3–5 times. The edible tissues were removed from each organism with stainless steel scissors and a scalpel, tissues from organisms of the same species were mixed together thoroughly, crushed with a stainless steel mixer, freeze-dried to constant weight, and ground to give a homogeneous powder, which was stored in a brown glass bottle at -20°C . An aliquot of each dried and homogenized sample was digested using a microwave digestion system. The metal extraction procedure is described in detail elsewhere [1].

The quality control and assurance of sediment samples were carried out by using the standard recovery method. The recoveries of the metals were between 83% and 111%. The Chinese National Standard material (offshore marine sediment, GBW 07314) were analyzed together.

Figure 1. Location of Daya Bay and the sampling sites.

Reagent blanks were used to correct the analyses results of marine organism samples. The concentration of all seven metals in the standard muscle tissue reference material 2976 (for trace elements and methylmercury) supplied by the United States National Institute of Standards and Technology (Gaithersburg, MD, USA) were determined, as well as on samples for quality control. The recoveries of the metals were between 80% and 120%. The Cu, Zn, Cd, and Cr concentrations in samples were measured by an atomic absorption spectrophotometer (AAS-Z2000; Hitachi, Tokyo, Japan) and the As and Hg concentrations were measured by an atomic fluorescence spectrophotometer (AFS-9700; Haiguang Instruments, Beijing, China).

2.3. Evaluation Methods

2.3.1. Assessment of Sediment Contamination

The index I_{geo} has been widely used to assess heavy metal contamination in sediment and soil [19,20]. The I_{geo} values were calculated by using the following equation:

$$I_{geo} = \text{Log}_2 [C_n / (1.5B_n)] \quad (1)$$

where C_n represents the measured concentration of heavy metals and B_n represents the background reference concentration. Due to lack of data on background values for the study area, element abundances of the upper continental crust were used as the background reference [19]. For this, 1.5 was the background matrix correction factor due to lithogenic effects. The geo-accumulation index can be divided into 7 categories, as shown in Table S2.

The methodology developed by Hakanson was used to calculate the potential ecological risk (RI) of Daya Bay [21].

$$\begin{aligned} C_f^i &= C_i / B_i \\ E_r^i &= T_r^i \times C_f^i \\ RI &= \sum E_r^i \end{aligned} \quad (2)$$

where C_i and B_i represent the determined concentration and reference standard value of heavy metals, respectively. C_f^i , the pollution index of an individual element; T_r^i , the toxicity coefficient of heavy metals, for Cu, Zn, Cr, Cd, As, and Hg were 5, 1, 2, 30, 10, and 40, respectively [22]; and E_r^i is the ecological risk coefficient of an individual element.

2.3.2. Health Risk Models

Marine organisms accumulate heavy metals through respiration and ingestion, and heavy metals may endanger human health through the food chains. This study used health risk assessment index to evaluate the edible health risks of Daya Bay organisms. The calculation method was as follows:

$$\begin{aligned} EDI &= \frac{C_m \times IR \times EF \times ED}{AT \times BW} \times AR_m \\ HQ &= EDI / RfDCR = EDI \times SF \\ THQ &= HQ_1 + HQ_2 + \dots + HQ_n \\ TCR &= CR_1 + CR_2 + \dots + CR_n \end{aligned} \quad (3)$$

where C_m is the concentration of heavy metals in marine organisms (mg/kg) and IR is the ingestion rate of seafood (g/person/day). The daily consumption of seafood in Guangdong Province is 51 g for an adult, and the ingestion rate for children is estimated to be 26.5% of that of adults [23]. BW is the average body weight (adults: 70 kg; children: 15 kg) [24], EF and ED indicate the exposure frequency (365 day/year) and exposure duration (adults: 70 years; children: 6 years), respectively. AT is the total exposure time (adults: 25,550 days; children: 2190 days). AR_m is the intestinal absorption rate of heavy metals, the values of AR_m for Cu, Zn, Cd, Cr, Hg and As were 45%, 65%, 75%, 55%, 64%, and 75%, respectively [25,26]. RfD is the chronic reference dose, the chronic reference doses of Cu, Zn, Cd, Cr, Hg and As were 0.04, 0.3, 0.001, 0.003, 0.0001, and 0.0003 mg/kg/day, respectively; SF is the carcinogenic slope factor, based on values of 0.38, 0.5, and 1.5 mg/kg/day for Cd, Cr, and As, respectively [10]. Moreover, EDI indicates the established daily intake, HQ is the noncarcinogenic hazard quotient of heavy metals to human body (THQ indicates the total hazard quotient), and CR is the carcinogenic risk quotient (TCR indicates the total carcinogenic risk). In addition, the As concentration in this study was the total content, but previous studies on the health risks of As reported that only inorganic As posed threats to humans, and the inorganic As was assumed to be 10% of the total As [27].

2.4. Monte Carlo Simulation

When the uncertainty arises owing to lack of knowledge of the metal concentrations and variability of toxic response among individuals, the probability approximate solution obtained by Monte Carlo simulation can enhance the understanding of the environmental behavior of contaminants and show the uncertainties [28,29]. To capture variations of risk analysis in Guangdong costal, Monte Carlo simulation was used to handle the uncertainty of heavy metal concentrations in surface sediments and marine organism samples collected in Daya Bay. Simultaneously, sensitivity analysis was carried out to estimate the contribution of the exposure variables of the total risks to discern critical exposure factors. The main steps of Monte Carlo simulation are as following: (1) determine the random variables selected by the evaluation model; (2) define the distribution models of the random variables; and (3) sample randomly from the above distribution of the variables and output the simulation results. The concentrations of heavy metals in sediments and

organism samples were transformed by natural logarithm to satisfy normality, indicating that the concentration data obey lognormal distribution (Table S3). Although, IR and BW, exposure time parameters (e.g., EF, ED, and AT) and toxicity criteria of heavy metals (e.g., RfD and SF) could also have been changed, the current study only considered the impact of exposed heavy metal concentrations on risks, thus other exposure parameters were single-point inputs.

2.5. Statistical Analysis

SPSS (version 24.00) was used to transform the metal concentration data into natural logarithm and Q-Q plot was used to evaluate the normality of the dataset of each variable. Monte Carlo simulation was performed using Oracle Crystal Ball (version 11.1.4323.0) loaded in Microsoft Excel to run 10,000 times to obtain stable risk outputs and sensitivity index. ArcGIS 10.2 and OriginPro 2017 were used for graphics delineation.

3. Results and Discussion

3.1. Heavy Metal Concentrations in Sediments

The concentrations of Cu, Zn, Cd, Cr, Hg, and As in sediments of Daya Bay were in the ranges of 5.3–44.5, 37.1–109.0, 0.06–0.22, 13.6–72.5, 0.014–0.171, and 4.8–9.7 mg/kg, respectively (Table 2). The mean concentrations of these elements followed a decreasing order: Zn > Cr > Cu > As > Cd > Hg. From the perspective of coefficient of variation (CV), Hg and Cu reached the CV level of 49% and 47%, respectively, and the coefficients of variation of other elements were between 14% and 31%. The results of descriptive statistics indicated that the metal concentrations in sediments of Daya Bay were quite different, and there was a large uncertainty in the spatial distribution of the metal concentration levels. Compared with the limited values of Chinese Marine Sediment Quality Standard (GB18668-2002), most of the concentration levels were lower than the corresponding limits. The mean concentrations of Zn, Cd, Hg, and As were slightly higher than their background values, indicating the impact of human activities. Threshold effect level (TEL) and probable effect level (PEL) could be used to evaluate the environmental risk caused by heavy metals in sediments. When concentrations of metals in sediments are lower than TEL, adverse effects upon sediment dwelling fauna will be infrequent, while when the PEL is exceeded, adverse harmful effects on sediment-dwelling organisms are expected to occur frequently [30]. The concentrations of heavy metal in the sediments of Daya Bay were lower than TEL or between TEL and PEL.

Comparing the results of this study with the studies of the Daya Bay in different periods (Table 1), it could be found that all concentrations of metals in sediments after 2008 were higher than those in 1988 [5]. From 2008 to 2020, the metals concentrations showed a law of first decline, then increase, and then decrease [5,13,19]. The trend of regular fluctuations reflected the impact of urban development and the government's corresponding environmental management policies on heavy metal contamination in sediments of Daya Bay. However, the results of the one-way analysis of variance showed that there was no significant difference in the concentrations of heavy metals from 1988 to 2020 ($p > 0.05$), which seems to indicate that the contribution of nuclear power plants sources to the contents of heavy metals in the sediment is limited. Compared with other bays along the coast of China, the levels of heavy metal concentration in Daya Bay were slightly higher than Beibu Gulf [20], but lower than Liaodong Bay, Bohai Bay, Jiaozhou Bay, Zhoushan Bay, and Quanzhou Bay [31–35].

Table 1. Comparison of heavy metal concentrations in sediments from Daya Bay with other regions (mg/kg).

Study Area	Sampling Date	Cu	Zn	Cd	Cr	Hg	As	Reference
Daya Bay	1988	6.44	26.01	0.03	22.35	NA	2.61	[5]
Daya Bay	2008	16.46	87.81	0.07	59.03	0.04	8.16	[19]
Daya Bay	2011	10.4	59.34	0.04	30.03	NA	7.01	[13]
Daya Bay	2016	24.58	111.65	0.23	65.04	NA	12.41	[5]
Daya Bay	2020	15.1	79.8	0.13	36.1	0.08	7.16	This study
Liaodong Bay	2013	19.66	70.2	0.22	61.5	0.06	9.28	[34]
Bohai Bay	2016	32.6	95.2	0.3	75.2	0.07	12.9	[31]
Jiaozhou Bay	2015	27.31	76	0.3	86.17	NA	NA	[33]
Zhoushan Bay	2017	27.22	103.09	0.19	68.62	0.03	NA	[32]
Quanzhou Bay	2011	60.81	186.7	0.64	84.72	0.11	NA	[35]
Beibu Gulf	2017	15.07	52.37	0.06	44.42	0.06	7.82	[20]

NA: not available.

Table 2. Descriptive statistics of sediment concentrations and corresponding reference standards.

Parameters	Heavy Metal (mg/kg)					
	Cu	Zn	Cd	Cr	Hg	As
Range	5.3–44.5	37.1–109.0	0.06–0.22	13.6–72.5	0.014–0.171	4.8–9.7
Mean $\pm$ S.D.	15.1 $\pm$ 7.02	79.8 $\pm$ 18.46	0.13 $\pm$ 0.04	36.1 $\pm$ 11.36	0.079 $\pm$ 0.039	7.16 $\pm$ 0.99
Median	14.4	85.2	0.13	38.3	0.072	7.1
CV (%)	47%	23%	28%	31%	49%	14%
TEL ¹	35.7	123	0.596	37.3	0.174	5.9
PEL ¹	197	315	3.53	90	0.486	17
Background value ²	28	67	0.09	92	0.05	5
Primary standard, China ³	35	150	0.5	80	0.2	20

Mean $\pm$ S.D.: average values and standard deviation; CV: coefficients of variation. ¹ TEL: threshold effect level, PEL: probable effect level [30,36]. ² Element abundances of the upper continental crust [19]. ³ Chinese Marine Sediment Quality Standard (GB 18668-2002).

3.2. Heavy Metal Concentrations in Organisms

Table 3 lists the overall heavy metal concentrations of 20 organism species collected in Daya Bay. Heavy metal concentrations (mg/kg, dry weight) ranged from 0.7 to 102.0 for Cu, 12.5–274.0 for Zn, 0.011–1.690 for Cd, 0.10–1.99 for Cr, 0.4–4.5 for As, and 0.015–0.147 for Hg. The relative ranking order of the mean concentrations of overall marine organism samples were: Zn > Cu > As > Cr $\approx$ Cd > Hg. The concentrations of mollusks in this study were similar to the results of previous study by Yuan et al. [10]. Comparing the mean concentrations of different marine organism groups indicated that mollusks had the highest Cr and As concentrations; crustaceans had the highest concentrations of Cu, Zn, and Cd; and fish had the highest concentrations of Hg. The variance in the concentration of heavy metals in different groups of organisms is related to their feeding patterns and nutritional levels [8,26]. Additionally, this study did not determine the sexes and reproductive stages of marine organisms, and the accumulation of metals in gonad tissues as well as in eggs at different embryonic development stages will have a slight impact on the statistical heavy metal contents of marine organisms [37,38]. In terms of the mean concentrations, the heavy metals concentrations in the marine organisms from Daya Bay basically met the safety standards of the Chinese National Standard of Maximum Levels of Contaminants in Food species (GB 2762-2017) and the safety standards of the FAO guidelines [39].

Table 3. Heavy metal concentrations (mg/kg, dry weight) in 20 marine species from Daya Bay.

Group of Marine Organisms	Species	Metal Concentrations					
		Cu	Zn	Cd	Cr	As	Hg
Mollusks	<i>Perna viridis</i>	13	108	0.674	0.44	2.9	0.086
	<i>Ruditapes philippinarum</i>	6.7	82.8	0.964	0.27	4.5	0.04
	<i>Paphia undulata</i>	10	57.5	0.834	1.07	1.7	0.062
	<i>Placamen calophylla</i>	12.4	51.7	1.09	1.99	3.3	0.056
	<i>Bursa rana</i>	7.4	66.2	0.27	0.48	1.3	0.054
	<i>Glossaulax didyma</i>	18.9	86.2	0.218	0.4	1	0.04
	<i>Uroteuthis duvauceli</i>	16	44	0.15	0.14	0.6	0.04
	Mean	12.06	70.91	0.60	0.68	2.19	0.05
Crustaceans	<i>Miyakea nepa</i>	59.8	102	1.14	0.18	2.1	0.04
	<i>Harpisquilla harpax</i>	102	99	1.69	0.29	2	0.018
	<i>Portunus pelagicus</i>	72.6	130	0.108	0.48	1.6	0.015
	<i>Charybdis hongkongensis</i> Shen	70.8	169	0.11	0.76	1.1	0.066
	<i>Eucrata crenata</i>	60.2	274	0.063	0.24	1.3	0.062
	Mean	73.08	154.80	0.62	0.39	1.62	0.04
	<i>Nematalosa japonica</i>	8.1	30.7	0.093	0.2	0.4	0.017
	<i>Osteomugil ophuyseni</i>	3.2	29.1	0.051	0.1	1.6	0.04
Fish	<i>Siganus canaliculatus</i>	2.5	26.4	0.062	0.18	0.9	0.036
	<i>muraenesox cinereus</i>	1.3	17.6	0.027	0.15	0.9	0.054
	<i>Sillago sihama</i>	1.1	20.4	0.026	0.16	0.9	0.138
	<i>Pennahia anea</i>	3.3	20.6	0.015	0.14	0.9	0.054
	<i>Gerres filamentosus</i>	1.5	39	0.014	0.16	0.8	0.147
	<i>Saurida elongata</i>	0.7	12.5	0.011	0.1	0.5	0.141
	Mean	2.71	24.54	0.04	0.15	0.86	0.08
	Guidelines						
China ¹	Molluscs	NA	NA	2	2	0.5	0.5
	Crustaceans	NA	NA	0.5	2	0.5	0.5
	Fish	NA	NA	0.1	2	0.1	0.5
FAO/WHO ²	Molluscs	30	30	0.5	NA	1.5	0.5
	Crustaceans	30	30	0.5	NA	1.5	0.5
	Fish	30	30	0.5	NA	3.5	0.5

¹ The Chinese National Standard of Maximum Levels of Contaminants in Food species (GB 2762-2017). ² Food and Agriculture Organization, World Health Organization [39].

When comparing this study with the concentrations of heavy metals in marine organisms in other regions of the world, it was found that the concentrations of heavy metals in marine organisms varied widely in terms of regions and species (Table 4). Overall, the concentrations of Cd, Cr, and Hg in the marine organisms from Daya Bay were comparable to those in other regions, and the concentrations of As were much lower than those in Xiangshan Bay, Sanmen Bay, and Bangladesh coast. In addition, the concentrations of Cu and Zn in mollusks were also lower than those in other regions except the South China Sea. The geographical variation in heavy metal concentrations in marine organisms was commonly related to the local pollution status [40]. The high concentrations of metal elements in seawater and sediments contributed to the bioaccumulation of marine organisms [32].

3.3. Contamination and Ecological Risk Assessment

3.3.1. Pollution Characteristics

The I_{geo} values of heavy metals in sediments of the Daya Bay are listed in Figure 2, and ranged from -2.99 to 0.08 for Cu (average -1.60), -1.44 to 0.12 for Zn (average -0.38), -1.17 to 0.70 for Cd (average -0.14), -3.34 to -0.93 for Cr (average -2.01), -2.42 to 1.19 for Hg (average -0.14), and -0.64 to 0.37 for As (average -0.08). The average values of I_{geo} were arranged in descending order: As > Hg = Cd > Zn > Cu > Cr. The average values of I_{geo} of all elements were less than zero ($I_{geo} < 0$), suggesting limited pollution of the Daya Bay.

Table 4. Comparison of heavy metals concentrations (mg/kg, wet weight) in marine organisms from Daya Bay with other locations around the world. Data reported based on dry weights were converted into wet weight by moisture contents [41].

Sampling Area	Groups	Cu	Zn	Cd	Cr	As	Hg	References
Daya Bay	Molluscs	1.3–3.8	14.2–21.6	0.120–0.218	0.14–0.40	0.4–0.9	0.011–0.017	This study
	Crustaceans	15.0–25.5	24.8–68.5	0.016–0.423	0.05–0.19	0.3–0.5	0.004–0.017	
	Fish	0.2–2.4	3.8–11.7	0.003–0.028	0.03–0.06	0.1–0.5	0.005–0.044	
South China Sea	Molluscs	0.10–0.24	0.80–2.01	0.06–0.17	0.12–0.58	NA	NA	[26]
	Fish	0.04–0.25	1.05–2.14	0.04–0.53	0.40–0.86	NA	NA	
Xiangshan Bay	Molluscs	0.45–26.65	8.34–65.72	0.01–1.05	ND–2.89	0.42–15.9	0.003–0.037	[42]
	Crustaceans	2.28–28.13	9.58–51.22	0.001–0.32	0.02–2.14	1.60–17.89	0.001–0.25	
	Fish	0.14–0.47	2.52–5.26	0.0004–0.004	ND–0.21	0.16–8.04	0.004–0.083	
Maowei Sea	Molluscs	4.60–77.50	9.50–64.60	0.180–1.800	0.18–0.42	0.70–1.20	0.024–0.066	[43]
	Crustaceans	3.16–29.40	9.57–34.35	0.019–1.510	0.10–0.23	0.16–1.39	0.016–0.045	
	Fish	0.20–1.90	10.50–40.50	0.003–0.220	0.12–0.63	0.10–1.50	0.006–0.028	
Sanmen Bay	Molluscs	0.92–96.81	12.11–132.93	0.18–9.64	0.08–0.73	1.42–20.94	0.004–0.087	[44]
	Crustaceans	1.10–8.22	12.59–59.31	ND–0.10	0.06–0.44	2.90–10.36	0.001–0.018	
	Fish	0.12–0.61	2.38–8.34	ND–0.03	0.03–0.31	0.53–2.04	0.003–0.023	
Bangladesh coast	Crustaceans	13–400	53–1480	0.02–8.3	0.29–29	0.3–53	NA	[45]
	Fish	1.3–14	31–138	0.03–0.09	0.15–2.2	0.8–13	NA	
Saint Martin Island	Crustaceans	5.05–30.73	7.02–61.92	0.20–30.44	ND–1.41	ND–0.28	ND–0.05	[7]
	Fish	0.3–2.23	3.34–12.10	1.52–14.09	0.18–1.87	ND	0.06–0.13	

ND: not detected.

Figure 2. Geo-accumulation index (I_{geo}) of heavy metals in sediments of the Daya Bay.

For the contamination characteristics of heavy metals in the sediments of Daya Bay, the statistical results of random sampling based on the elements' concentrations distribution through Monte Carlo simulation and the class distribution of I_{geo} values are shown in Table S4 and Figure 3. The average values of I_{geo} were arranged in descending order: $Hg \approx As > Cd > Zn > Cu > Cr$, and the average values of I_{geo} of all elements were less than zero, which is similar to the evaluation results based on the geo-accumulation index method. However, the class distribution of the I_{geo} values revealed that there was relative variance in the spatial distribution of heavy metal contamination. The I_{geo} value of Hg was the highest, with a moderately polluted score of 5.46% and unpolluted to moderately polluted score of 39.52%, followed by Cd, As, and Zn, with unpolluted to moderately polluted accounting for 35.71%, 34.78%, and 12.76%, respectively. Hg has no biological function, its most important sources are fuel combustion and coal combustion [12]. Previous studies reported that Hg and As in Daya Bay were affected by the same anthropogenic input [19], and Cd originated from the discharge of domestic sewage [13]. Therefore, heavy metals contamination in Daya Bay was closely related to nearby industrial and agricultural activities.

3.3.2. Potential Ecological Risks

Figure 4 illustrated the potential ecological risks of the heavy metals in sediments of Daya Bay. In general, the RI values of sampling sites varied significantly, ranging from 58.76 to 202.48. According to the category of RI (Table S5), five sampling sites reached a moderate risk and 22 sampling sites reached a low risk. In addition, Figure 4 shows that the E_r^i values of Hg had the highest contribution to potential ecological risks, followed by Cd. The consequence of E_r^i values of the six heavy metals were ranked as $Hg (62.81) > Cd (42.47) > As (14.31) > Cu (2.69) > Zn (1.19) > Cr (0.79)$.

For the potential ecological risk characteristics, the cumulative probability of RI of heavy metals in sediments of Daya Bay based on Monte Carlo simulation were 80.94% for low risk, 18.96% for moderate risk, and 0.1% for high risk, respectively. High I_{geo} values also corresponded to high potential ecological risk indices. Hg, Cd, and As contributed 50.85%, 33.92%, and 11.47% of the potential ecological risk, respectively (Figure 5b). It has been found in many studies that heavy metals negatively affect microbial community diversity [46]. Indirect ecological risks posed by changes in microbial communities are difficult to predict [47]. However, the concentrations of heavy metals, such as Hg, measured

in this study were relatively low, and did not exceed the primary standard of the China Marine Sediment Quality standards. Even at low concentrations, the comprehensive evaluation results of I_{geo} and RI showed that Hg was the primary heavy metal pollutant in sediments, followed by Cd and As. These results were similar to the study of Ding et al. [31]. From this point of view, Hg emissions in the Daya Bay area should be effectively controlled, and it is necessary to conduct ecological risk analysis and evaluation on the uncertainty of heavy metal emissions in the Daya Bay area.

Figure 3. Class distribution of I_{geo} values of heavy metals in sediment of Daya Bay.

Figure 4. Potential ecological risk index (RI) of heavy metals in sediments of Daya Bay.

Figure 5. (a) The cumulative percentage of RI and (b) the contribution of respective heavy metal to RI in Daya Bay.

3.4. Health Risk Assessment

The EDI of the heavy metals determined as described above are shown in Table 5. For both adults and children, EDI values were below the acceptable daily intake (ADI) and HQ values were generally in decreasing order as follows: Hg > As > Cu > Cd > Zn > Cr, and the CR values for different metals generally followed as As > Cr > Cd. Monte Carlo simulation was used to evaluate the total noncarcinogenic risks (THQ) and carcinogenic risks (TCR) (Figure 6a,b). Potential risks were indicated by the probabilities corresponding to THQ values higher than 1 (THQ > 1). It was found that there was no potential noncarcinogenic risk for adults and children. But for total carcinogenic risks, the probability of adult and children exceeded the unacceptable risk value (1×10^{-4}) at 11.19% and 22.27%, respectively. Therefore, the continuous or excessive intake of these marine organisms could cause chronic carcinogenic effects. The health risk results were comparable to the results reported by Yuan et al. [10]. It should be noted that the THQ and TCR used for risk assessment were based on the concentration addition of different metals, and their calculation is a noninteractive process. Humans are exposed to more than one pollutant and suffer combined or interactive effects [48]. However, THQ and TCR cannot directly reflect the true risk because they do not define any dose–response relationship [49]. Furthermore, the loss of elements caused by cooking and human intestinal absorption rate was not considered here.

Figure 6. Cumulative percentage of (a) carcinogenic and (b) noncarcinogenic risks of eating marine organisms in Daya Bay.

Table 5. Daily intakes of heavy metals in marine organisms consumed by adults and children in Guangdong province.

Metals	Mean Concentration ¹ (mg/kg)	EDI (mg/kg/day)		ADI ² (mg/kg/day)	HQ		CR	
		Adult	Children		Adult	Children	Adult	Children
Cu	5.7	1.96×10^{-3}	2.43×10^{-3}	5.00×10^{-1}	4.91×10^{-2}	6.07×10^{-2}		
Zn	17.6	8.33×10^{-3}	1.03×10^{-2}	3.00×10^{-1}	2.78×10^{-2}	3.43×10^{-2}		
Cd	0.085	4.66×10^{-5}	5.77×10^{-5}	1.00×10^{-3}	4.66×10^{-2}	5.77×10^{-2}	1.77×10^{-5}	2.19×10^{-5}
Cr	0.09	3.61×10^{-5}	4.47×10^{-5}	3.30×10^{-3}	1.20×10^{-2}	1.49×10^{-2}	1.81×10^{-5}	2.23×10^{-5}
As	0.4	1.95×10^{-5}	2.42×10^{-5}	3.10×10^{-4}	6.52×10^{-2}	8.06×10^{-2}	2.93×10^{-5}	3.63×10^{-5}
Hg	0.016	7.32×10^{-6}	9.05×10^{-6}	2.30×10^{-4}	7.32×10^{-2}	9.05×10^{-2}		

¹ Average wet weight concentrations calculated after conversion from dry weight. ² Acceptable daily intake [6].

Figure 7 presents the sensitive analysis results of the metals concentrations of THQ and TCR estimates for humans. The highest contribution to THQ was Hg (28.45%), followed by Cd (22.75%), Cu (22.67%), and As (17.84%). Noncarcinogenic risks of Hg are also expected, because Hg exposure to human is believed to be significantly correlated with the consumption of fish among all food stuffs [8,50]. The contribution of As, Cd, and Cr to the TCR value was relatively similar, with the highest being As (37.16%), followed by Cd (32.47%) and Cr (30.37%), respectively. The carcinogenic health risk results of children and adults simulated by Monte Carlo warn people to pay more attention to the contents of carcinogenic elements in marine organisms. There were reports that the enrichment and amplification of heavy metals in organisms will harm human health through the food chain. For example, As can cause diseases of the cardiovascular system, central nervous system, and hematopoietic system [24]. Cd can lead to high blood pressure, high risk of fracture [51], and liver dysfunction [9]. Cr is involved in lipid metabolism and insulin function, and can cause damage to DNA and tissue structure [52]. Additionally, Table 1 presented that the concentrations of Cr, As, and Cd in mollusks were higher. Crustaceans have the highest concentrations of Cu, Zn, and Cd, and fish have the highest concentrations of Hg. Therefore, mollusks were the main species that caused carcinogenic risk. Since the food habits could not be easily changed, and seafood was also the main source of protein for residents in coastal areas, on the basis of the same overall consumption of seafood, the carcinogenic risk can be maintained below a safe level by reducing the intake of mollusks and supplementing the intake of crustaceans. Moreover, the intake of crustaceans also contributed to the supplement of Zn in the human body. Cu and Zn were trace elements necessary for life activities. The moderate intake of Zn from marine organisms may therefore promote health [53].

Figure 7. Sensitivity analyses of heavy metal concentration (C_m) for a total hazard quotient and cancer risk.

3.5. Limitations

For heavy metals in sediments, only certain sampling points were used to estimate the overall risk of the area, and the results were usually uncertain, especially when the study area is large and there are many pollution sources. A Monte Carlo method, based on mathematical statistics and probability theory, was used to estimate the distribution of

metal concentrations for the purpose of evaluating uncertainty in environmental risk models caused by random sampling [54]. However, the traditional Hakanson risk assessment method does not incorporate the ecosystem (species and their ecological functions) into the RI as a whole [22]. Therefore, the evaluation results cannot determine the ecological components that may be affected by heavy metals, and specify the species that need to be clearly protected. In addition, due to the spatial heterogeneity of soil/sediment, although the evaluation results can reflect the uncertainty of the contamination degree in the study area to a certain extent, it cannot combine geographic information to reveal the temporal and spatial distribution of contamination in the entire region. Moreover, the background information, such as the influence of pH, Eh, and SPM in sediments on the toxic effects of heavy metals, should also be taken into consideration.

It should be pointed out that although this study collected representative economic marine organisms in Daya Bay, the evaluation results do not represent all marine species. In addition, heavy metals can be enriched through the food chains and endanger human health. Therefore, the bioavailability and chemical speciation of heavy metals are also key factors affecting risk assessment [55]. The measurement in this study was the total concentration of marine organisms. Although the concentration and bioavailability of heavy metals have been treated with reference to a large number of previous studies, the results of the risk assessment were still uncertain. Moreover, risk models and key exposure parameters (C_m , BW, IR, RfD, and SF) are also two main sources that affect the evaluation results [28]. The RfD and SF based upon the Integrated Risk Information System and International Agency for Research on Cancer might not yield a precise picture of current toxic responses of the Chinese population [29]. On the other hand, more precise health risks should consider the biological half-life of heavy metals, as well as the differences in the interaction and metabolic processes of heavy metals in the human body.

4. Conclusions

This study combined Monte Carlo simulation with risk assessment methods, and converted the traditional single point calculations into the probability distribution of risk description, providing a more comprehensive and objective assessment result of the heavy metal contamination in Daya Bay. The contents of heavy metals in the sediments of Daya Bay were lower than that in other regions of China. However, from the evaluation results of I_{geo} and RI, Daya Bay sediment was found to be most seriously contaminated with Hg, which reached moderate pollution and accounted for 50.85% of the potential ecological risk, followed by Cd and As, which were unpolluted to moderately polluted, accounting for 33.92% and 11.47% of the potential ecological risk, respectively. Although the consumption of marine organisms in Daya Bay would not cause noncarcinogenic risks to the human body, the probability that the carcinogenic risk of adults and children exceeded the threshold (1×10^{-4}) was 11.19% and 22.27%, respectively. Sensitivity analysis pointed out that the contribution of carcinogenic elements to risk can be arranged in descending order as As > Cd > Cr. In summary, efforts to control and remediate extensive heavy metal contamination in the Daya Bay should focus on Hg, As, and Cd. Taking into account the uncertainty of the concentration information and the parameters in the evaluation models, the risk assessment of the coastal marine environment and human health should focus on the analysis of the bioavailability and toxic interaction of heavy metals in the future.

Supplementary Materials: The following are available online at <https://www.mdpi.com/2077-1312/9/1/17/s1>, Table S1: List and information of marine organisms sampled in the Daya Bay., Table S2: Seven classes of the geo-accumulation index., Table S3: Uncertain concentration (mg/kg) of heavy metals in sediments and marine organisms in Daya Bay., Table S4: Statistical results of I_{geo} and RI based on Monte Carlo simulation., Table S5: Category of potential ecological risks.

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