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Appraising ecotoxicological risk of mercury species and their mixtures in sediments to aquatic biota using diffusive gradients in thin films (DGT)

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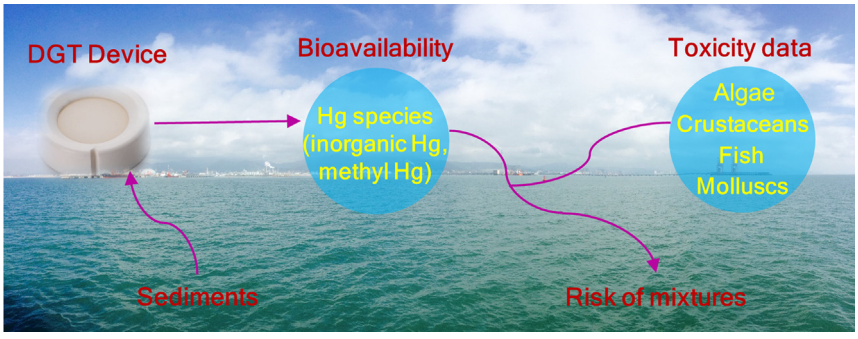
HIGHLIGHTS

- Diffusive gradients in thin films to assess bioavailabilities of Hg species in sediments
- Fist time to evaluate ecotoxicological of Hg species and their mixtures to aquatic biota based on bioavailability
- Potential toxicities for individual Hg species and mixtures have occurred.

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury (Hg) is a global, persistent and inevitable pollutant, the toxicity of which is mostly reflected in its species including inorganic Hg (InHg) and methyl mercury (MeHg). Using diffusive gradients in thin films (DGT) is deemed as a reliable technique to determine the bioavailability of pollutants. This study is the first attempt to assess the integrated toxicity of mercury species mixtures in sediments to the aquatic biota based on the DGT technique. In the course, the Daya Bay under serious anthropogenic influences was selected as the study case. The results showed that the DGT concentrations of InHg and MeHg were detected as 0.30–1.93 μg/L and 0.28–1.94 μg/L respectively in the surface sediments collected from the Daya Bay. In terms of the toxicity of single mercury species, the risk quotient (RQ) values of InHg and MeHg significantly exceeded 1, indicating that the adverse effects of InHg and MeHg should not be ignored. In terms of the integrated toxicity of mercury species mixtures, the probabilistic biological risk assessment results demonstrate that Daya Bay features low (3.32%) probability of toxic effects in its surface sediments to the aquatic biota.

1. Introduction

As a global, persistent and irreversible pollutant, (Podar et al., 2015; Azaroff et al., 2019; Wang et al., 2021), mercury (Hg) has contaminated the coastal and estuarine ecosystems owing to anthropogenic sources (Amirbahman et al., 2013; Jonsson et al., 2017). Whenever Hg species (inorganic Hg (InHg) and methyl Hg (MeHg)) are detected in the aquatic ecosystem, the sediments, in comparison with the waterbody or other aquatic

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organisms, are functioning as the dominant "sink" of mercury (Jonsson et al., 2017; Lei et al., 2019; Gerson et al., 2020; Yu et al., 2021). When the aquatic environment changes, for example, the pH value, the mercury species once contained in the sediments can possibly permeate into the waterbody, which may affect the aquatic organisms adversely (Trefry et al., 2007; Podar et al., 2015; Fleck et al., 2016; Jonsson et al., 2017; Lei et al., 2019). Nevertheless, no study has been performed up to now on the integrated toxic effects resulting from Hg species mixtures on the aquatic biota involved in the entire aquatic ecosystem, within the sediments, in particular.

The toxicity of Hg mostly depends on its species, and organic Hg is commonly known of higher toxicity than inorganic Hg. Although InHg can be uneasily absorbed by people's gastrointestinal tract, it can also be effortlessly changed into MeHg through biogeochemical processes (Clarisse et al., 2011; Azaroff et al., 2019), which will also influence the biological risks due to the bioavailability of Hg species (InHg and MeHg, for example) (Amirbahman et al., 2013; Azaroff et al., 2019). It has been proved that traditional methods (involving sediment total mercury concentrations, sequential extraction and simultaneously extracted metals (SEM)–acid-volatile sulfide (AVS) models) cannot well predict the bioavailability of Hg species in sediments (Yin et al., 2014); in this sense, more effective methods are in urgent demand.

DGT, as a passive sampling technique established for measuring the bioavailable concentrations of Hg species and the distribution of different Hg species solute in waters, sediments, and soils, is believed to be an effective technique of measuring the bioavailability of Hg species (Clarisse et al., 2011; Ndu et al., 2018; Pelcová et al., 2018; Ren et al., 2018) and has been lately undertaken to forecast the toxicity and bioavailability of Hg species causing to the surrounding benthic organisms (Amirbahman et al., 2013; Yin et al., 2014; Pelcová et al., 2018).

The Daya Bay, located at the northeast Pearl River Estuary, is a busy area that has been greatly impacted by anthropogenic activities and its surrounding area is a densely populated and economically important part of Guangdong Province (Fig. 1; Gu et al., 2016b). Anthropogenic activities related to petrochemical, plastic, printing and other industries as well as harbors are available in its surrounding area (Gu et al., 2016b). Two nuclear power stations, the Daya Bay Power Station and Ling'ao Power Station, which came into service in 1994 and 2002 respectively, are located along the Bay's western coast (Fig. 1; Gu et al., 2016b). Previous studies show that the Daya Bay's sedimental Hg has exhibited strong Hakanson's ecological risks based on the total concentration of Hg (Hakanson, 1980; Yang et al., 2020). In the meantime, the Daya Bay is deemed as an important Fishery Resource Reserve at the provincial level (Gu et al., 2016a). Considering both its characteristics and significance, this Bay was then selected as the study case.

Upon prudent literature review, no study has been previously carried out about the integrated toxicity of mixtures of Hg species (InHg and MeHg) in sediments through DGT analysis. Therefore, the present study aims to (1) assess the concentrations of Hg species contained in the surface sediments of the Daya Bay through DGT, (2) depict the distributions of Hg species, and (3) evaluate the risk of Hg species and its mixtures, in hopes that such study can provide a new and extensive approach for assessing the risk of Hg species in sediments.

2. Materials and methods

2.1. Study area and sample collection

The Daya Bay, which is situated in the northern part of the South China Sea, is a shallow semi-enclosed and subsidence mountain drowned valley bay (Fig. 1; CBCCC (China Bay Chronicles Compilation Committee), 1998), highlighting typical subtropical marine climate. It covers an area of 516 km^2 with its water depth varying from 5 m to 18 m (CBCCC, 1998), the annual average temperature, the average annual rainfall, and the annual average relative humidity of 21.7 °C, 1984.4 mm and 82%, respectively (Hao et al., 2016). As there's no major rivers flowing into the bay with only three small rivers discharging into Dapeng Cove (Fig. 1A), the majority water in the Daya Bay sources from the South China Sea.

In January 2020, the surface sediments of 3 cm thick were collected from 24 stations (Fig. 1) using a Peterson-grab sampler. The samples from each site were then split into three parts, two parts of which were put separately into two self-sealing polyethylene bags, with the remaining part for sulfide put into a brown glass reagent bottle. An amount of zinc acetate (100 g/L) was added and then mixed with the samples thoroughly. Then all samples were frozen preserved on the way to the laboratory and stored at −20 °C for further analysis.

2.2. Analytic method

The procedures regarding the total organic carbon (TOC) determined by potassium dichromate oxidation-reduction capacity method and the sulfide determined by methylene blue spectrophotometric method were conducted strictly in accordance with China National Standards (GB 17378.5-2007). The particle size was pretreated in compliance with China National Standards (GB/T12763.8-2007), and the granulometry of the sediment samples was measured using a Malvern Mastersizer 2000 laser diffractometer.

The DGT procedures were undertaken in compliance with the methods established by environmental scientists from the laboratory of EasySensor Ltd. (Nanjing, China) (Ren et al., 2018). Briefly speaking, a microcosm deployment assay was conducted with a TCH-95 DGT probe and then bathed and incubated in water at the temperature of 25 °C for 3 weeks. Afterwards, the DGT probe was inserted into the sediment sample vertically and then taken out after 24 h. In the course, a cutter featuring stacked ceramic blades (the laboratory of EasySensor Ltd., Nanjing, China) was used to slice the binding gel into smaller pieces of 4.0 mm each, which were eluted separately with the solution of 10 mL. Then the eluents were gathered to identify the concentrations of MeHg and InHg via cold vapor atomic fluorescence spectrometry (CV-AFS). The absolute difference between two independent determination results obtained under repeatability conditions was less than 10% of the arithmetic mean value. The whole operation and detection of DGT procedures were implemented in the laboratory of EasySensor Ltd. (Nanjing, China).

2.3. Equations for DGT calculation

In accordance with Fick's first law related to the diffusive flux to the gradient of the concentration, the concentration of Hg contained in the DGT extract liquor can be converted into the Hg mass (M) by Eq. (1):

$$M = \frac{C_e\left(V_e + V_g\right)}{f_e} \tag{1}$$

where C_e refers to the concentration of Hg contained in the extract liquor, V_e and V_g refer to the volumes of the extract-liquor (mL) and the gel (mL) respectively, f_e refers to the extraction rate of Hg (1.00 for MgHg and 0.536 for InHg). The concentration of DGT-labile Hg can be then computed by Eq. (2):

$$C_{DGT} = \frac{M\triangle g}{DAt} \tag{2}$$

where $\triangle g$ refers to the thickness of the diffusive-layer (0.9 mm), D refers to the diffusion coefficient of Hg through the diffusion layer (9.94×10^{-6} cm^2s^{-1} for MeHg at 25 °C and 7.65×10^{-6} cm^2s^{-1} for InHg at 25 °C), with D value for the investigated Hg based on the method established by environmental scientists from the laboratory of EasySensor Ltd. (Nanjing, China) (Ren et al., 2018), A refers to the DGT-device exposed surface area (3000 mm^2), and t refers to the deployment time, which was 24 h in this study. In this study, the value of gel volume was 1200 mm^3.

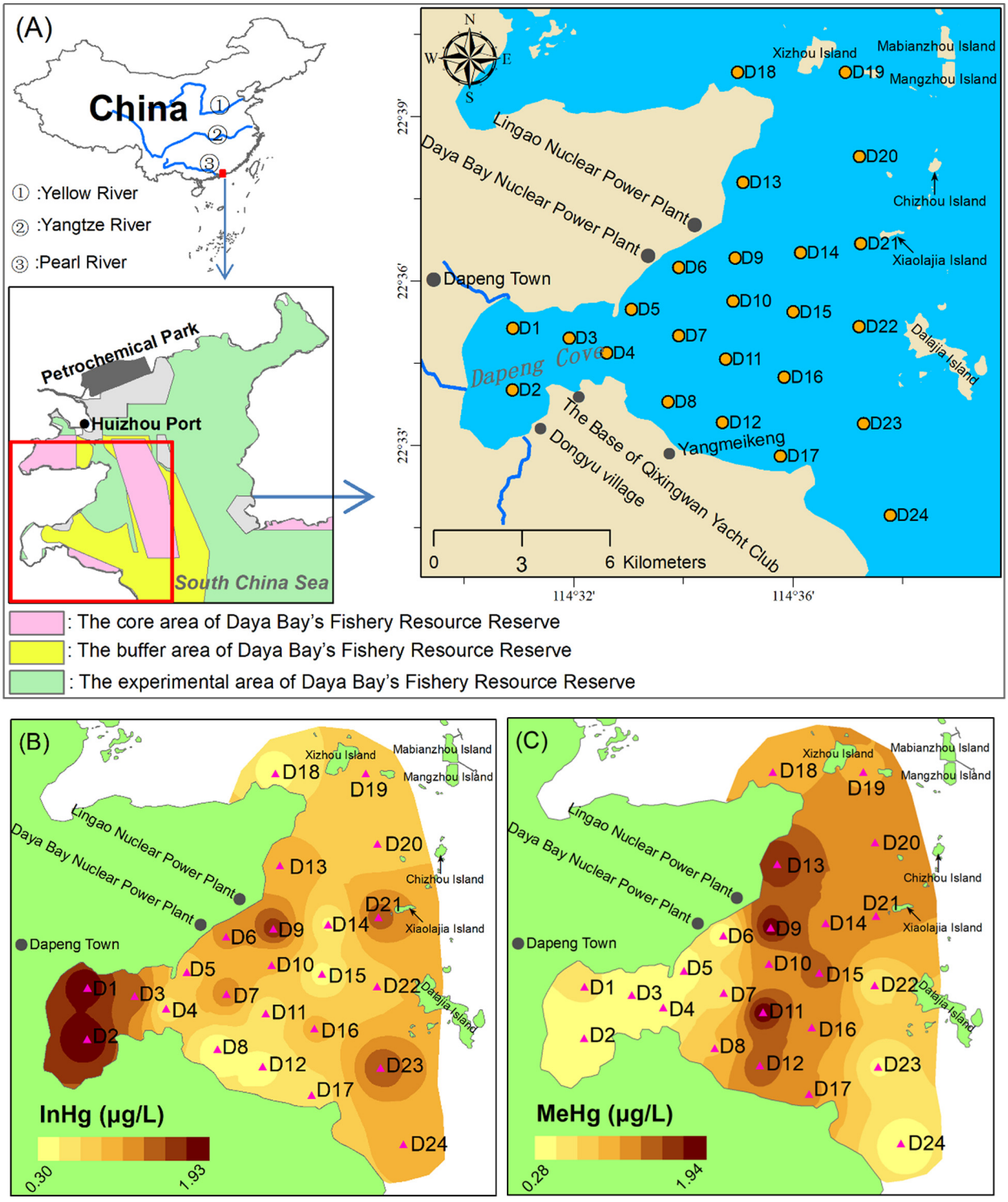

Fig. 1. Map demonstrating the surrounding scenario of the study area and the sampling stations (A) and the spatial distributions of mercury species (inorganic mercury (InHg) and methyl mercury (MeHg)) in the surface sediments (B and C) of the Daya Bay, China.

2.4. Collection of the toxicity data

Data related to the acute toxicity of MeHg and InHg were collected from the USEPA ECOTOX database (https://cfpub.epa.gov/ecotox/). Data (LC50 or EC50) related to the acute toxicity of aquatic species in saltwater, from such four different trophic-level groups as algae, fish, crustacean, and mollusc, are indicated in Table 1 respectively. Detailed information including the values of such toxicity data and specific aquatic species are listed in the supplementary material.

The species sensitivity distribution (SSD) method was built based on the standard method delineated by USEPA (U.S. Environmental Protection Agency) (2001, 2018) in accordance with the subsequent steps. First of all, select an appropriate endpoint (e.g.: the lethality, the growth, and the reproduction, etc.); then collect the toxicity data from the literature

Table 1
Data related to the acute toxicity of the mercury species and values of the predicted no-effect concentration (PNEC) (μg/L).

Mercury species	Algae		Crustaceans		Fish		Molluscs		PNEC[b]
	EC50	N[a]	EC50/LC50	N	EC50/LC50	N	EC50/LC50	N	
InHg	25.50	14	39.00	158	71.00	44	40.00	71	0.0255
MeHg	n.a.	n.a.	78.00	2	65.50	6	40.00	2	0.040

n.a.: no available.
[a] *N* refers to the numbers of toxicity.
[b] PNEC was computed in terms of the acute toxicity in algae, crustaceans, and fish respectively, with the minimum data chosen and then divided by the assessment factor (=1000) (Gu et al., 2020).

covering as many related species as possible; and thirdly, characterize the distribution of such toxicity data across species in accordance with an appropriate probability density. The detailed SSD method was elaborated by USEPA (USEPA (U.S. Environmental Protection Agency), 2001; USEPA (U.S. Environmental Protection Agency), 2018).

2.5. Ecotoxicological risk models

2.5.1. Risk assessment of single mercury species

The risk quotient (RQ) was utilized for evaluating the ecotoxicological risk caused by each DGT-labile Hg, that is, the ratio between the measured concentrations of MeHg and InHG respectively in environment (DCR) and PNEC, as shown in Eq.(3):

$$RQ = \frac{DCR}{PNEC} \tag{3}$$

In Table 1, indicating the established PNEC values of MeHg and InHg in this study, RQ is effective to assess the ecotoxicological risk of a single pollutant that is ubiquitous in water, sediments and soils (Gu, 2021; Riva et al., 2019; Vašíčková et al., 2019). The RQ value, if equaling to or being less than 1, indicates relatively lower risk; if being more than 1, indicates potential ecotoxicological risk and non-ignorable environmental risk (Gu et al., 2020; Riva et al., 2019). The higher the value is, the higher potential ecological risk will be (Gu et al., 2020).

2.5.2. Risk assessment of Hg species mixtures

Before assessing the risk of mixtures of Hg species, the risk probability of single Hg to aquatic biota was computed according to the probabilistic risk assessment (PRA) protocols (USEPA (U.S. Environmental Protection Agency), 2001). On this basis, the risk probabilities of MeHg and InHg were conducted to calculate the integrated risk of mixtures of MeHg and InHg to aquatic biota by the sum of such probabilities (Steen et al., 1999), as indicated in Eq. (4):

$$\begin{aligned}\Phi[A_1 + A_2 + \ldots + A_n] = {} & \sum_{i-1}^{n} \Phi[A_i] - \sum_{i1<i2} \Phi[A_{i1}A_{i2}] + \ldots \\ & + (-1)^{r+1} \sum_{i1<i2<\ldots<ir} \Phi[A_{i1}A_{i2}\ldots A_{ir}] + \ldots \\ & + (-1)^{n+1} \Phi[A_1 + A_2 + \ldots + A_n]\end{aligned} \tag{4}$$

where Φ refers to the probability; $\Sigma\Phi[A_{i1}A_{i2}\ldots A_{ir}]$ takes over all of the $\binom{n}{r}$ possible subsets of size r for the set $\{1, 2, n\}$.

2.6. Statistical analysis

Inverse distance weighting (IDW) method was conducted for spatial interpolation. The statistic results of IDW interpolation showed a good agreement between the measured line and the predicted line with the acceptable mean error (<0.05) and Root-Mean-Square (<0.05) (using ArcGIS 10.4). In terms of statistical analysis, SPSS 19.0 for Windows was used for the factor analysis with the principal component analysis (FA-PCA) and Kolmogorov-Smirnov (KS) test, with Windows STATISTICA 8.0 for Box-Cox data transformation, and MASS package in R4.04 software for computing the overlap area between the exposure data and the toxicity data.

3. Results and discussion

3.1. Sediment traits

In this study, the content of total organic carbon (TOC), the sulfide, and the composition of particles were measured to establish the general traits of sediments, as shown in Table 2. The content of TOC varied from 0.17% to 1.58%, 0.93% on average. The content of sulfide was measured between 6.70 mg/kg and 201.00 mg/kg in the dry sediment, 50.03 mg/kg on average. And the median grain size ranged from 3.34 to 6.97 phi (Φ), 5.83 Φ on average.

The content of TOC and the particle size are significant controlling factors that may determine the metal content in natural sediments (Gu, 2021). In addition, the sulfide involves the distribution of natural metals (Dang et al., 2015). In this study, we have found that median grain size and sulfide were significantly correlated with TOC ($p < 0.001$) (Fig. 2A and B). We have also found that sulfide was not correlated with median grain size (Fig. 2C). These results demonstrated that distributions of sulfide and particle size were mainly controlled by TOC.

Table 2
Concentrations of mercury species, total organic carbon (TOC), sulfide, and median grain size distributions in surface sediments from the Daya Bay, China.

Site	InHg (μg/L)	MeHg (μg/L)	TOC (%)	Sulfide (%)	Grain size (median, Φ)
D1	1.87	0.53	0.50	14.40	4.77
D2	1.93	0.28	0.85	23.50	5.75
D3	1.55	0.38	1.58	174.00	5.79
D4	0.55	0.34	1.26	15.00	6.52
D5	0.76	0.30	0.62	14.00	5.43
D6	1.17	0.32	0.17	6.80	3.34
D7	1.33	0.46	0.67	29.50	5.95
D8	0.30	0.89	0.94	57.30	5.62
D9	1.69	1.87	0.56	18.30	5.52
D10	0.92	1.18	0.22	6.70	4.83
D11	0.52	1.94	0.82	25.60	6.15
D12	0.41	1.46	1.19	56.80	5.94
D13	1.00	1.72	1.04	201.00	5.72
D14	0.45	1.26	1.25	92.00	5.76
D15	0.36	1.42	1.14	38.00	6.19
D16	0.99	1.21	1.10	24.20	5.90
D17	0.59	1.13	0.85	30.90	6.01
D18	0.42	0.91	1.31	50.50	6.97
D19	0.76	1.09	1.34	96.90	6.30
D20	0.78	1.15	1.26	18.90	6.23
D21	1.44	1.16	1.34	103.00	6.22
D22	0.87	0.48	0.72	23.90	6.38
D23	1.52	0.44	0.80	39.20	6.37
D24	0.95	0.33	0.81	40.40	6.28
Min.	0.30	0.28	0.17	6.70	3.34
Max.	1.93	1.94	1.58	201.00	6.97
Mean	0.96	0.93	0.93	50.03	5.83
S.D.	0.50	0.53	0.36	50.36	0.72

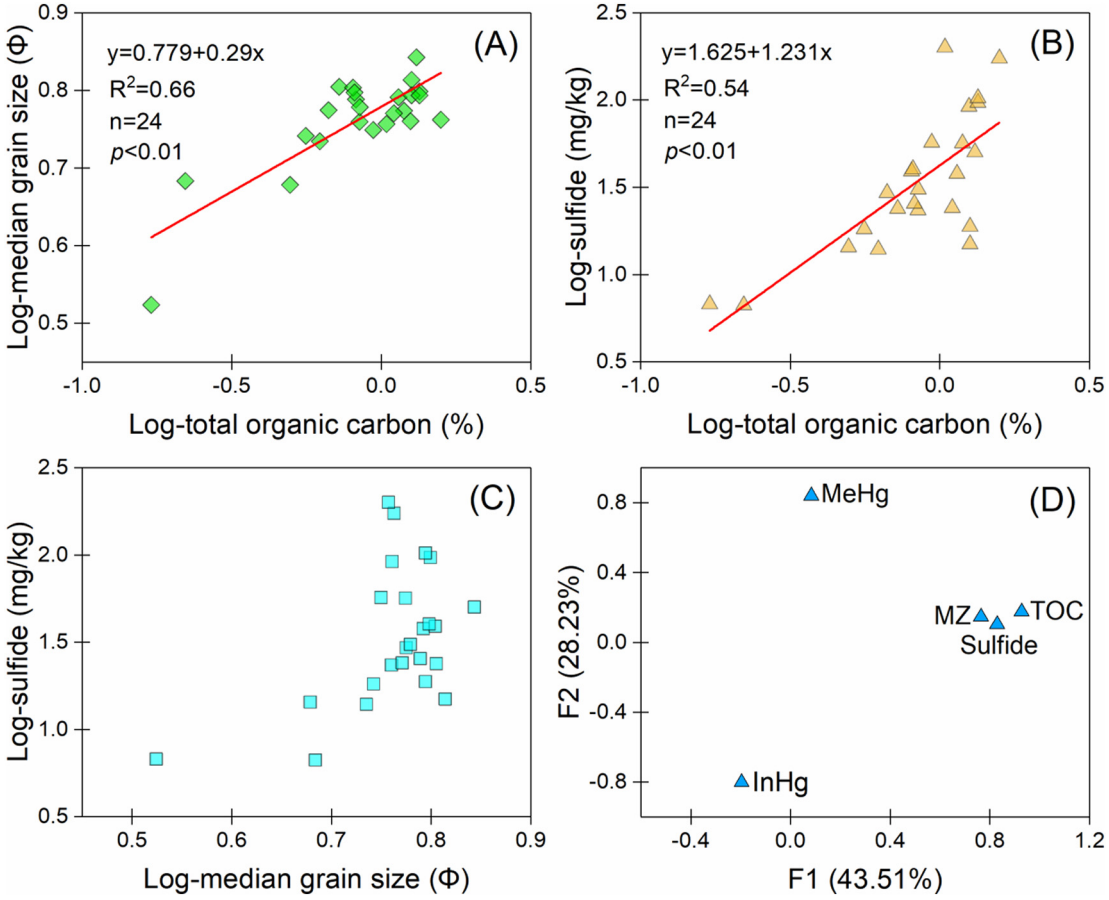

Fig. 2. Correlations between TOC and the median grain size (A) and sulfide (B), and between the median grain size and sulfide (C); loadings of variables on VARIMAX rotated factors of the variables (D). Log: the logarithm base 10; MZ: median grain size.

3.2. Concentration of DGT-labile Hg species in sediments

As illustrated in Fig. 1B, Fig. 1C and Table 2, the highest value of InHg was detected at D2, the highest value of MeHg was found at D11, the concentrations of DGT-labile Hg species (μg/L) in the surface sediments covered in this study varied widely, with InHg ranging from 0.30 to1.93 and MeHg ranging from 0.28 to 1.94.

The total Hg concentration reported in the literature was 0.012–0.045 μg/L (mixed acid extraction method) in seawater from the Daya Bay (China) (Liu et al., 2022) and 0.01–0.03 μg/L (DGT technique) in seawater from the Boat Harbor (Canada) (Chaudhary et al., 2020). MeHg concentration (DGT technique) was 0.0006–0.00082 μg/L in seawater from the Boat Harbor (Canada) (Chaudhary et al., 2020) and 0.0029–0.00354 μg/L in sea pore water from Gulf of Trieste (Northern Adriatic) (Bratkič et al., 2019).

It has been proved that FA-PCA is powerful to confirm the possible relationship between the sediment-trait parameters and DGT-labile metals (Gu, 2021). Thus, this approach was adopted in this paper for data analysis to identify the possible relationship between the former and DGT-labile mercury species. In addition, FA (VARIMAX rotation mode) was applied to discern the two factors (Fs) as extracted based on the eigenvalues (>1), representing 60.00% data. The communalities explained by the variables (sediment-trait parameters and DGT-labile mercury species), while considering the two Fs, varied from 60.5% for the median grain size (MZ) to 89.1% for TOC. All variables were well represented by the two Fs (F1 and F2), contributing 43.51% and 28.23% data respectively. And the loadings of mercury species and the sediment-trait parameters were demonstrated in Fig. 2D. Among them, F1 featured strong positive loadings for TOC, sulfide, and median grain size (MZ) (>0.5) with F2 loaded with InHg and MeHg (>0.5), indicating that the mercury species were not influenced by the sediment-trait parameters.

3.3. Ecotoxicological risk assessment

3.3.1. Risk of individual metals

Fig. 3 illustrates RQ values of mercury species (InHg and MeHg) in the surface sediments of the Daya Bay. In the figure, the RQ values of InHg varied from 11.76 to 75.69, 37.79 on average, and the RQ values of MeHg were between 7.00 and 48.50 with an average of 23.18, implying the existence of adverse ecological effects and the fact that the mixture of mercury species can also present ecotoxicological risk. Accordingly, further studies are urgently demanded based on the current efforts.

3.3.2. Risk of mercury mixtures

In order to estimate the possible biological risk (integrated toxicity) caused by mercury species, the concentration of DGT-labile Hg mixtures and the toxicity data (the species sensitivity distribution, SSD) should be measured (USEPA (U.S. Environmental Protection Agency), 2001; Gu et al., 2020). KS test was adopted to identify whether the exposure and the toxic data fitted the normal distribution. As demonstrated by the test results, the transformed data for exposure and toxicity indicated normal distributions ($p > 0.05$) with the mean, standard deviation (SD), and KS test results for mercury species demonstrated in Table 3 respectively, which were subsequently used to create the transformed-normal probability distribution curves for biological risk analysis (Fig. 4). In the figure, the overlap area between the exposure and the corresponding toxicity of single Hg species was computed (Fig. 4). Results showed that InHg and MeHg had a

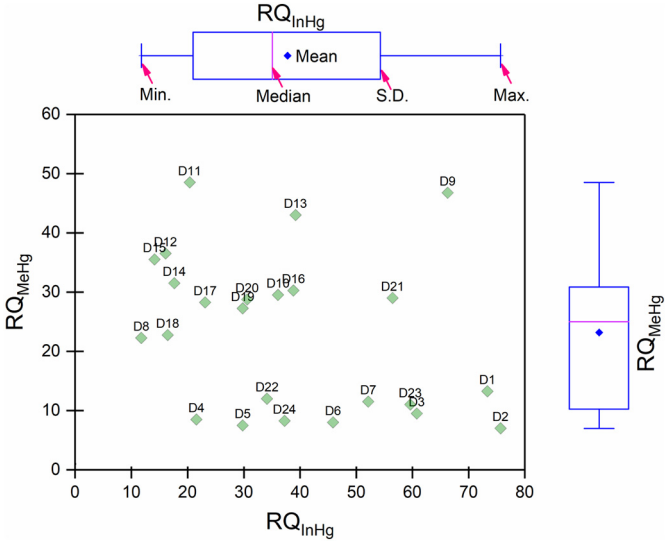

Fig. 3. Values of RQ for mercury species (InHg and MeHg) regarding each sampling point in the surface sediments of the Daya Bay of China.

Table 3
Exposure and acute toxicity data for mercury species: the transformed-normal distribution, the calculated means, the standard deviations (SD), and the KS test results, among which, the exposure and toxicity concentrations of InHg have been Box-Cox transformed with those of MeHg Log-transformed.

Element	Transformed exposure concentrations			Transformed toxicity concentrations		
	Mean	SD	K-S test	Mean	SD	K-S test
InHg	−0.14	0.53	0.99	6.94	3.03	0.68
MeHg	−0.11	0.28	0.28	1.83	0.28	0.88

3.32% and 5.32E-02% probability of ecotoxicological effects respectively. The sum of toxic-effect probabilities (Gu et al., 2020; Steen et al., 1999) of mercury species was then used to evaluate the integrated risks of mercury species mixtures. Results demonstrated that the surface sediments of the Daya Bay exhibited an integrated 3.32% probability of exerting toxic effects on the aquatic organisms.

RQ is deemed conservative in nature because it has established the exposure and effect assumptions that represent the upper limit of available toxicity data (Fig. 3; Garber et al., 2014). As this evaluation method assumes both the exposure and the toxicity endpoints conservatively, it is inherently protective and fails to identify the general risk extent in a quantitative manner. By contrast, the PRA technique, on the basis of SSD, has overcome such limitation and provided a quantitative probabilistic risk assessment (Fig. 3; Gu et al., 2020). As a methodology for determining the toxicity of mercury species mixtures, the sum of probabilities, together with the PRA method, was efficient in quantifying the potential risk to aquatic biota in this research. Therefore, this unique technique is critical for conserving and managing the ecological environment.

4. Conclusion

It is challenging to diagnose the integrated toxicity of Hg species to the aquatic biota in natural aquatic ecosystem, peculiarly in sediments, where the Hg species mostly sink and concentrate. To determine the bioavailability of pollutant substances, DGT is a profitable approach. In this study, the bioavailable (DGT-labile) concentrations of mercury species were computed with the DGT approach. The PRA approach, taking the toxicity of Hg species to aquatic groups at four different trophic levels into account, was established in terms of the SSD method and DGT-labile concentrations. Subsequently, a new approach, based on the sum of probabilities in combination with the PRA approach, was thus developed to assess the toxicity of mercury species mixtures in sediments.

The Daya Bay, as seriously influenced by people, was selected as the case study. The PRA results revealed that InHg and MeHg had a 3.32% and 5.32E-02% biological risk to the aquatic biota. This new method showed that surface sediments of PRE had a 3.32% probability of inducing toxic effects on aquatic organisms. In this sense, the biological risk of Hg species mixtures in sediments using this new method coupled with the DGT technique is useful and can be adopted as an effective approach to monitor and protect the aquatic ecosystem.

CRediT authorship contribution statement

Yang-Guang Gu: Methodology and Writing- Original draft preparation, Data curation, Supervision, Conceptualization, Visualization, Writing-Reviewing, Editing. **Hong-Hui Huang:** Investigation and Validation. **Shi-Jun Jiang:** Investigation and Validation. **Xiu-Yu Gong:** Investigation. **Xiu-Li Liao:** Investigation. **Dai Ming:** Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

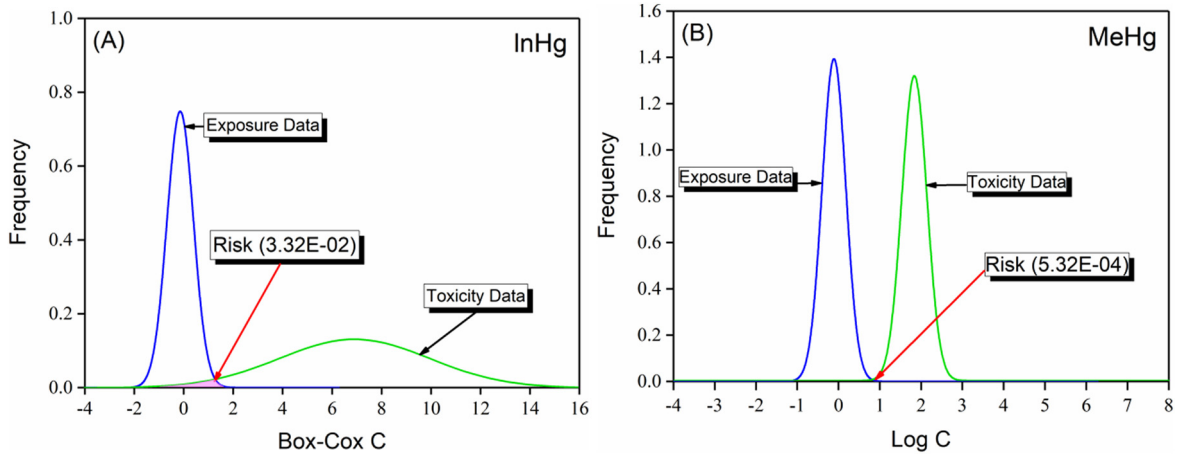

Fig. 4. Probability density distribution curves of the exposure and toxicity concentrations of mercury species in the surface sediments of the Daya Bay of China; Log C: base-10-logarithm-transformation concentrations.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.154069.

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