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Variations in methylmercury contamination levels and associated health risks in different fish species across three coastal bays in China

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The growing atmospheric mercury (Hg) emissions in China have raised ongoing concerns regarding contamination in marine fish. To better understand the pollution patterns and associated risks, we examined methylmercury (MeHq) content in demersal and pelagic fish from four commonly found families in three geographically distinct bays along the Chinese coast. We identified significant spatial variations in MeHg levels within the same fish family across regions. Specifically, fish collected from the Beibu Gulf in the South China Sea consistently exhibited significantly higher MeHg levels compared to those from the Laizhou Bay in the Northeast and/or Haizhou Bay in the East of China. In contrast, MeHg levels in fish collected from Haizhou Bay consistently remained the lowest. Within each region, we observed significantly higher MeHg concentrations in demersal species compared to pelagic species. This trend was particularly evident in fish species including bartail flathead (Platycephalus indicus), small-scale tongue sole (Cynoglossus microlepis) and greater lizardfish (Saurida tumbil) from the Beibu Gulf (0.50, 0.21, and 0.18 mg/kg dw, respectively), as well as bartail flathead and slender lizardfish (Saurida elongata) from Laizhou Bay (0.09 and 0.12 mg/kg dw, respectively). By comparison, MeHg content in silver pomfret (Pampus argenteus) from all three regions consistently remained relatively lower than in other species. Using target hazardous quotient (THQ) calculations, we estimated potential health risks in local populations associated with the consumption of the studied fish species. Our results showed a lack of apparent health risks to local residents, as all THQ values obtained from the three regions fell within the safe limits (0.02-0.94). However, it remains important to conduct additional assessments and spatiotemporal monitoring that encompass a broader range of species and regions.

KEYWORDS

heavy metals, aquatic products, neurotoxicity, potentially toxic elements, marine

1 Introduction

Mercury (Hg) is a ubiquitous and hazardous environmental contaminant, well recognized for its wide-ranging impact on ecosystems and human health (Clarkson, 1997; Boening, 2000). Various forms of Hg originating from human activities or natural geological processes can be introduced into the environment (Hammond, 1971), and undergo the methylation process and transformed into methylmercury (MeHg) (Weber, 1993; Regnell and Watras, 2019). Coastal marine sediments, rich in organic matter and microorganisms that support methylation, play a vital role in shaping methylmercury (MeHg) budgets within coastal regions and the marine food web (Hammerschmidt and Fitzgerald, 2004; Hollweg et al., 2009).

The formation and bioaccumulation of MeHg in the marine environment are influenced by a multitude of factors, such as sediment physicochemical property, organic carbon, bacterial activity, and plankton biomass, resulting in significant spatial heterogeneity (Chen et al., 2008; Zhang Y. et al., 2020). Therefore, assessing Hg levels across geographically distinct regions is essential for obtaining a comprehensive overview of distribution and accumulation patterns, and the better understand of environmental and ecological influences (Clayden et al., 2014; Barbosa et al., 2022; Médieu et al., 2022). However, inter-comparison between existing studies can be problematic due to differences in sample processing method and instrumental analysis (Chen et al., 2008).

National and international legislation efforts have been undertaken to address and mitigate Hg contamination, including the Minamata Convention (Kessler, 2013). However, MeHg remains a significant concern due to ongoing risks such as neurological damage and metabolic disruption associated with the consumption of marine fish in human populations from regions in the United States (Tollefson and Cordle, 1986; Liu et al., 2018), the European Union (Llull et al., 2017), as well as China and other regions (Cao et al., 2020; Basu et al., 2023). Furthermore, increased atmospheric Hg inputs from coal combustion in China (Wu et al., 2016) have been linked to elevated tuna Hg levels compared to other Pacific regions (Médieu et al., 2022).

In this study, we investigated MeHg concentrations in common fish species of the same family or suborder collected from three distinct coast bays in China: the Beibu Gulf (subtropical), Haizhou Bay (temperate), and Laizhou Bay (temperate). We examined regional differences in MeHg levels of the same species or close-related species within the same family, as well as across different species within each region. Given that all three studied bays were situated at urbanized regions, we hypothesized that regional differences in fish MeHg content may exist due primarily to different methylation rate across subtropical and temperate waters. Furthermore, species-specific differences in MeHg content were also expected, particularly between demersal and pelagic species, since coastal sediments are major sites for Hg methylation. We also evaluated the potential factors influencing these variations and assessed human health risks related to the consumption of these fish species for the local populations.

2 Materials and methods

2.1 Study area and sampling

Fish samples were collected during fishery surveys conducted in the offshore area along the Chinese coast to facilitate regional comparisons of MeHg content in common fish species. We selected three specific coastal bays which are geographically distinct regions with vital ecological and economic significance but also face challenges related to pollution due to human activities (Laizhou Bay, Haizhou Bay, and the Beibu Gulf; Figure 1). Fish were collected using trawls in September and October of 2021 following the Animal Research and Ethics Committee of the Ocean University of China guidelines. The sampling period coincides with the rainy season in all three regions due to their location within the East Asian Monsoon system. All samples were immediately placed on ice until returned to the laboratory for further processing. In the laboratory, biological parameters including body length and weight were first measured and recorded, we then used acid-rinsed dissection tools to collect muscle samples from the center of the fish body after removing the skin. Samples were stored in polypropylene centrifuge tubes and kept at -20°C until further analyses. This study focused on identical or closely related species found in the three bays, totalling 96 fish from four families or suborders, i.e., Cynoglossidae, Platycephalidae, Stromateoidei, and Synodontidae. The species, sample size, weight and length are listed in Table 1.

2.2 Methylmercury analysis

Fish muscle samples were freeze-dried and homogenized using a tissue-homogenizer. Approximately 0.10-0.15 g of dried tissue sample were accurately weighed using an analytical balance (Sartorius, Germany), and placed into 50 mL polypropylene centrifuge tubes. Samples were digested using 20 mL of 30% nitric acid solution at 60°C overnight. After digestion, the samples were mixed with 10 mL ultrapure water and centrifugated, and a 200 μ L of the supernatant was subsequently collected and transferred into a 50 mL pre-rinsed tube. This fractionation was adjusted to a pH of 6 using potassium hydroxide and sodium acetate solution. Finally, the mixture was added with ultrapure water and 50 μ L of a derivatization reagent [NaB(C₂H₅)₄], and left aside for 2 h at room temperature before instrumental analysis. The MeHg content was determined by using gas chromatography-cold vapor atomic fluorescence spectrometry (GC-CVAFS) (Polytech Instrumental Co., Ltd., Beijing, China). The analytical quality control was verified by the analysis of procedure blanks and a reference material, i.e., P43123B (fish powder, Guangzhou Puen Scientific Instrument Co., Ltd.). The average recovery of the reference material ($85\% \pm 10\%$; n = 13) and the blank content $(1.2 \pm 1.0 \text{ ng}; n = 13)$ were all within the acceptable range. All samples were detected at levels above the detection limit (0.06 pg). All concentrations are expressed in mg/kg dw (dry weight).



TABLE 1 Species, sample size (n), weight, and length of sampled fish species from the Beibu Gulf, Haizhou Bay, and Laizhou Bay.

Region	Species	Scientific name	Feeding habits	n	Weight (g)	Length (mm)
Beibu Gulf	Bartail flathead	Platycephalus indicus	Demersal	4	295 ± 17.8	247 ± 27.5
	Small-scale tongue sole	Cynoglossus microlepis	Demersal	11	273 ± 41.7	131 ± 64.3
	Greater lizardfish	Saurida tumbil	Demersal	6	217 ± 19.8	52.8 ± 3.04
	Brushtooth lizardfish	Saurida undosquamis	Demersal	8	206 ± 54.6	32.4 ± 8.83
	Silver pomfret	Pampus argenteus	Pelagic	3	97.1 ± 7.56	43.4 ± 5.87
	Pacific rudderfish	Psenopsis anomala	Pelagic	7	116 ± 13.1	63.5 ± 26.6
Haizhou Bay	Red tongue sole	Cynoglossus joyneri	Demersal	11	161 ± 16.9	22.1 ± 6.14
	Slender lizardfish	Saurida elongata	Demersal	4	146 ± 8.54	20.9 ± 4.40
	Silver pomfret	Pampus argenteus	Pelagic	4	154 ± 4.79	74.0 ± 6.67
Laizhou Bay	Bartail flathead	Platycephalus indicus	Demersal	10	106 ± 27.6	26.4 ± 2.37
	Chinese tongue sole	Cynoglossus semilaevis	Demersal	10	103 ± 29.8	26.7 ± 2.16
	Slender lizardfish	Saurida elongata	Demersal	8	111 ± 19.6	24.5 ± 1.17
	Silver pomfret	Pampus argenteus	Pelagic	10	64.0 ± 11.7	14.9 ± 0.98

2.3 Data analysis

In the present study, all statistical analyses and plotting of the results were performed using R 4.2.0 (R Core Team, 2022). The distribution of MeHg concentrations were checked using Shapiro-Wilk test and Levene's test, and the values were log-transformed for the analysis of variance (ANOVA; Base R). Tukey's HSD test (package "stats") was

used for *post hoc* comparisons in order to assess potential differences in MeHg concentrations of the same fish family among different regions, as well as among different species within the same region. Additionally, the relationship between fish physical parameters (weight and length) and MeHg content across different species and regions were identified using linear mixed effect regression analysis (package "lme4"; Bates et al., 2014) with species and sampling region as random effects.



Additionally, regional comparison was performed using weightadjusted MeHg content, i.e., observed MeHg of each species within a region normalized by the average weight of the same species within that region (Braaten et al., 2017).

We further assessed the human health risks associated with consumption of the studied fish species using the Target Hazard Quotient (THQ) (EPA, 1989) based on the equation below:

$$THQ = \frac{EF \times ED \times IR \times C}{RfD \times BW \times AT} \times 10^{-2}$$

where EF is exposure frequency (365 days/year); ED is total exposure duration (4 and 72 years for children and adults, respectively) (Yu et al., 2020); IR is ingestion rate of marine fish in China's coastal populations (18 and 49 g/day for children and adults, respectively) (Wang et al., 2020); C is MeHg concentration in edible portion of the fish (mg/kg on a wet weight basis employing a conversion factor of 0.8; Li et al., 2023); RfD is oral reference dose for MeHg (1×10^{-4} mg/kg/day); BW is average body weight of an adult (25 and 65 kg for children and adults, respectively); and AT is average exposure time for non-carcinogens (EF × ED). THQ < 1.0 indicates no health risks, while THQ > 1.0 suggests potential risks from fish consumption.

3 Results and discussion

3.1 Regional differences in fish MeHg content

We observed notable variations in MeHg content among the examined fish species in various regions, with fish from the Beibu

Gulf consistently exhibiting relatively high levels, while those from Haizhou Bay were consistently low (Figure 2). Specifically, significantly higher concentrations of MeHg were found in the demersal species, the small-scale tongue sole from the Beibu Gulf (0.21 mg/g) compared to the other two tongue sole species from Haizhou Bay and Laizhou Bay (both 0.04 mg/kg; P < 0.001). Similarly, significantly higher MeHg content was found between two Stromateoidei species, both pelagic (silver pomfret and Pacific rudderfish, 0.07 and 0.03 mg/kg, respectively) and two Synodontidae species, both demersal (greater lizardfish and brushtooth lizardfish, 0.18 and 0.14 mg/kg, respectively) from the Beibu Gulf compared to those from Haizhou Bay (silver pomfret, and slender lizardfish, 0.01 and 0.03 mg/kg, respectively; all $P \le 0.034$). Concentrations of MeHg in silver pomfret (0.04 mg/kg) and slender lizardfish (0.11 mg/kg) from Laizhou Bay were also significantly higher than Haizhou Bay (P < 0.001 and P = 0.046, respectively). Additionally, in bartail flathead, MeHg was significantly higher in those collected from the Beibu Gulf than Laizhou Bay (0.50 and 0.09 mg/kg, respectively; *P* < 0.001).

In general, the observed concentrations are consistent with those reported in fish collected along the coast of China. For instance, concentrations of MeHg in muscle of bartail flathead and slender lizardfish collected in Laizhou Bay were 0.07 and 0.10 mg/kg dw, respectively, in line with the present findings, whereas those reported for silver pomfret (0.10 mg/kg) was relatively higher compared to the present study (Cao et al., 2020). Such differences may be due to variations in trophic levels within the local food web in conjunction with different contamination profiles between the sediment and water bodies, and warrant in-depth investigation. Nevertheless, muscle MeHg content in silver pomfrets collected from multiple coastal cities of southeastern China was 0.05 mg/kg dw (Zhang et al.,



2020). Pacific rudderfish from the Beibu Gulf also exhibited comparable muscle MeHg levels in those previously reported $(0.04 \pm 0.01 \text{ mg/kg dw})$ (Zhu et al., 2013) and the present study $(0.03 \pm 0.01 \text{ mg/kg dw})$. Additionally, the current MeHg levels in sole and flathead from the Beibu Gulf appear to be similar to MeHg levels found in muscle of flatfish and common sole (*Solea solea*), which were approximately 0.39 and 0.44 (mg/kg dw) from the Baltic Sea and France's Atlantic Coast, respectively (Polak-Juszczak, 2017; Mille et al., 2021). However, the highest levels detected in the species of the present study were considerably lower than those reported for muscle content in common sole from the Western Mediterranean Sea, which were approximately 4.75 (mg/kg dw) (Llull et al., 2017). In fact, it has been shown that the Mediterranean waters exhibit a notable capacity for methylation and serve as a source for the nearby North Atlantic Ocean (Cossa et al., 2022).

The relatively higher MeHg concentrations in fish from the Beibu Gulf observed here compared to the other coastal bays may be associated with potentially stronger atmospheric Hg emission owing to biomass burning activities in the Indochina Peninsula (Sheu et al., 2013). Long-range atmospheric transport through the Asian Northeastern Monsoons in autumn may also contribute to elevated atmospheric Hg levels and its subsequent deposition in the northern South China Sea (Liu et al., 2016; Yuan et al., 2023). Furthermore, high precipitation rates, enhanced primary production and microbial activity could also have facilitated the speciation and bioaccumulation of MeHg (Kim et al., 2017; Zhang et al., 2020) in the subtropical Beibu Gulf compared to the other two temperate bays. Indeed, a systematic evaluation of Hg in Chinese coastal sediments has also demonstrated more elevated levels of Hg in sediments from the south coast than those from the east coast of China (Meng et al., 2019). In conjunction, MeHg production could also increase with decreasing latitude, which was primarily influenced by the elevated annual temperature (Dai et al., 2021).

It has been commonly found that MeHg contents correlate with fish size (Andersen and Depledge, 1997; Baeyens et al., 2003; Kehrig et al., 2008). The present study observed a significantly positive association between fish body weight and MeHg concentration, fish collected from the Beibu Gulf also appeared to be relatively heavier in comparison with those from the other regions (Figure 3). Nevertheless, except for bartail flathead (weight-adjusted MeHg: 0.27 and 0.14 mg/kg for Beibu Gulf and Laizhou Bay, respectively; P = 0.08), all regional differences in MeHg content detected here remained significant after adjusting body



weight (P range: 0.017 to < 0.001). These results further highlight significant regional disparities and emphasize the need for continued monitoring and assessment to investigate the sources and pathways of MeHg. Moreover, future studies should incorporate fish species of the same sizes and a larger sample size to further assess regional differences fully controlling the size-effect. In addition, within each species, there was a significant positive correlation between body weight and MeHg concentrations for bartail flathead (collected in Laizhou Bay and Beibu Gulf), and a significant negative correlation for silver pomfret (collected in all three bays), whereas no association was found for slender lizardfish (collected in Haizhou Bay and Laizhou Bay; Supplementary Figure S1). These findings suggest that the weight-MeHg concentration relationship as well as the fish size effect may be species- and region-specific, and warrants in-depth revaluation. Additionally, further investigations into the occurrence and distribution of MeHg in the sediment environment of these coastal regions are desired to better assess the baseline contamination pattern of these respective environment and provide further insight into the transfer mechanisms of Hg along the food chain.

3.2 Species-specific variation in MeHg content

In the present study, we found significant differences across species within each population. Overall, the MeHg content in demersal fish including sole, flathead, and lizardfish showed relatively high concentrations. By comparison, pelagic fish including silver pomfret and Pacific rudderfish had relatively low MeHg concentrations (Figure 4). In the Beibu Gulf, MeHg levels in Pacific rudderfish were significantly lower compared to bartail flathead, small-scale tongue sole and brushtooth lizardfish (P < 0.001, P < 0.001, and P = 0.029, respectively). In Haizhou Bay, MeHg concentrations in silver pomfret was significantly lower than red tongue sole (P < 0.001). In Laizhou Bay, MeHg content in bartail flathead and slender lizard fish was significantly higher than Chinese tongue sole and/or silver pomfret (all $P \le 0.049$).

Coastal and estuarine sediments are recognized for their ability to produce MeHg at elevated levels, primarily because of the specific biogeochemical conditions present, such as the abundance of organic matter and sulfate (Chen et al., 2008). Subsequently, MeHg enters the food web through uptake by benthic invertebrate macrofauna, and eventually becomes incorporated into fish tissues from the ingestion of contaminated prey (Mason and Lawrence, 1999; Hammerschmidt and Fitzgerald, 2006). Likewise, various previous study have found considerable species-dependent variability showing demersal species had higher MeHg than pelagic ones (Storelli et al., 2003; Anual et al., 2018; Romero-Romero et al., 2022).

3.3 Human health risks of fish MeHg contamination across different regions

Considering that the fish species examined in this study also serve as important commercial resources for human consumption, it becomes imperative to further assess the potential health risks posed to humans regarding their MeHg content, particularly among the demersal species. In this study, the THQ calculations for various species in different regions indicated no apparent health risks associated with consuming the studied species for either children or adults in the Beibu Gulf, Haizhou Bay, and Laizhou Bay populations (Table 2). Nevertheless, it's worth noting that the THQ values for both children and adults were notably elevated in the case of bartail flathead from the Beibu Gulf (close to 1.0). This

Region	Species	Scientific name	MeHg (mg/kg dw)	THQ children	THQ adult
Beibu Gulf	Bartail flathead	Platycephalus indicus	0.497 ± 0.347	0.89 ± 0.62	0.94 ± 0.65
	Small-scale tongue sole	Cynoglossus microlepis	0.213 ± 0.065	0.38 ± 0.12	0.40 ± 0.12
	Greater lizardfish	Saurida tumbil	0.185 ± 0.055	0.33 ± 0.10	0.35 ± 0.10
	Brushtooth lizardfish	Saurida undosquamis	0.136 ± 0.061	0.24 ± 0.11	0.26 ± 0.11
	Silver pomfret	Pampus argenteus	0.072 ± 0.026	0.13 ± 0.05	0.14 ± 0.05
	Pacific rudderfish	Psenopsis anomala	0.032 ± 0.016	0.06 ± 0.03	0.06 ± 0.03
Haizhou Bay	Red tongue sole	Cynoglossus joyneri	0.044 ± 0.028	0.08 ± 0.05	0.08 ± 0.05
	Slender lizardfish	Saurida elongata	0.026 ± 0.008	0.05 ± 0.01	0.05 ± 0.02
	Silver pomfret	Pampus argenteus	0.012 ± 0.006	0.02 ± 0.01	0.02 ± 0.01
Laizhou Bay	Bartail flathead	Platycephalus indicus	0.090 ± 0.077	0.16 ± 0.14	0.17 ± 0.15
	Chinese tongue sole	Cynoglossus semilaevis	0.036 ± 0.018	0.06 ± 0.03	0.07 ± 0.03
	Slender lizardfish	Saurida elongata	0.115 ± 0.062	0.21 ± 0.11	0.22 ± 0.12
	Silver pomfret	Pampus argenteus	0.045 ± 0.009	0.08 ± 0.02	0.08 ± 0.02

TABLE 2 Muscle MeHg content and target hazard quotient (THQ) in different fish species from the Beibu Gulf, Haizhou Bay, and Laizhou Bay.

aligns with the significantly higher MeHg concentrations observed in this fish from the Beibu Gulf compared to other species and other regions. Therefore, further monitoring and assessment are warranted.

Consistently, multiple prior studies have shown that Hg levels in aquatic products from the Beibu Gulf and various other regions along the Chinese coast are within safe limits, posing no apparent health risks (Gu et al., 2018; Zhao et al., 2018; Liu et al., 2019; Qin et al., 2021; Li et al., 2023). On the other hand, certain trace metals, notably arsenic (As), have been found to have elevated THQ values and potential health risks associated with the consumption of specific aquatic products from the Beibu Gulf (Wang et al., 2018; Yang et al., 2021). In the Laizhou Bay, in addition to Hg in predatory fish species (Cao et al., 2020), studies have indicated potential health risks associated with both As and cadmium (Cd) contamination in marine aquatic products (Jiao et al., 2021; Liu et al., 2022). Moreover, in the adjacent waters of the Beibu Gulf within Guangdong province, research has shown that As and Hg pose significantly higher health risk compared to other trace metals (Wang et al., 2023). Consequently, while the observed MeHg levels in fishes from the studied coastal areas are generally safe, continued monitoring of this and other contaminants in a suite of abiotic and biotic compartments is still necessary. Additionally, further studies are needed to understand the mechanisms underlying the varying methylation rates of inorganic Hg across coastal regions, as well as the uptake and accumulation in fish residing in different habitats.

Data availability statement

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

Ethics statement

Ethical approval was not required for the study involving animals in accordance with the local legislation and institutional requirements because only some fish related data was used.

Author contributions

TL: Formal Analysis, Investigation, Resources, Writing-original draft. MA: Formal Analysis, Writing-review and editing. JC: Formal Analysis, Writing-review and editing. YL: Formal Analysis, Writing-review and editing. LC: Formal Analysis, Writing-review and editing. JL: Formal Analysis, Writing-review and editing. MZ: Conceptualization, Funding acquisition, Writing-review and editing.

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Conflict of interest

Authors TL and MA were employed by China National Offshore Oil Corporation (CNOOC) Research Institute Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary Material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenvs.2024.1376882/ full#supplementary-material

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