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Fish, rice, and human hair mercury concentrations and health risks in typical Hg-contaminated areas and fish-rich areas, China

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A R T I C L E I N F O

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A B S T R A C T

Human exposure to methylmercury (MeHg) from consuming contaminated fish has been a major concern for decades. Besides, human MeHg exposure through rice consumption has been recently found to be important in some Asian countries. China is the largest country on mercury (Hg) production, consumption, and anthropogenic emission. However, the health risks of human Hg exposure are not fully understood. A total of 624 fish, 299 rice, and 994 human hair samples were collected from typical Hg-contaminated areas and major fish-rich areas to assess the health risks from human Hg exposure in China. Fish and rice samples showed relatively low Hg levels, except the rice in the Wanshan Hg mining area (WMMA). Human hair total Hg (THg) and MeHg concentrations were significantly elevated in WMMA, Zhoushan (ZS), Xiamen (XM), Qingdao (QD), and zinc smelting area (ZSA), and 85% of hair samples in WMMA, 62% in ZS, 40% in XM, 26% in QD, and 17% in ZSA had THg concentrations exceeding the limit set by the USEPA (1 μg/g). Rice consumption was the main pathway (>85%) for human MeHg exposure in the studied Hg-contaminated areas. Meanwhile, fish was the primary human MeHg exposure source (>85%) in coastal cities. Therefore, soil remediation in typical Hg-contaminated areas and scientific guidance for fish consumption in coastal provinces are urgently needed to reduce the health risks from human Hg exposure in China.

1. Introduction

Mercury (Hg) is a ubiquitously toxic element that poses potential health risks for humans and wildlife. It is considered as a global pollutant based on the fact that metal atom vapor with long lifetime dominates the Hg speciation in the atmosphere and entails long-distance transport (Driscoll et al., 2013; Yu et al., 2016). Mercury can be transformed into methylmercury (MeHg) by microorganisms after being translocated and deposited into agricultural and aquatic ecosystems. This is easily bioaccumulated along the food chain (Harris et al., 2007; Lavoie et al., 2013; Wright et al., 2018; Yu et al., 2020). Human exposure to MeHg may cause nervous system disorders, cardiovascular diseases, kidney and liver damage, and visual impairment (Beckers and Rinklebe, 2017; Clarkson and Magos, 2006; Selin, 2009). And exposure to MeHg is also considered to adversely affect a child's intelligence quota (Axelrad et al., 2007; Feng et al., 2020), resulting in a decline in individual lifetime income and even affecting the national economy (Griffiths et al., 2007; Trasande et al., 2005). Fatal MeHg poisonings documented from Japan and Iraq during the latter half of the twentieth century are well-known examples of the health hazards associated with the spread of anthropogenic organic Hg in the environment (MEGJ, 2020; Skerfving and Copplestone, 1976).

China is the largest anthropogenic Hg emitter due to its rapid economic growth in recent decades (UNEP, 2019). The total atmospheric Hg emissions from anthropogenic sources in China were estimated to be 444 t in 2017 (Liu et al., 2019a). Consequently, MeHg concentrations in certain fish and seafood from coastal and Hg mining areas exceeded the national limit (GB2762, 2017) of 1000 ng/g for carnivorous fish and

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500 ng/g for the other aquatic on a wet weight basis (Liu et al., 2014a; Qiu et al., 2009; Shao et al., 2013). Similarly, MeHg concentration in rice grains (174 ng/g) collected in Guizhou Hg mining areas also exceeded the national limit of 20 ng/g for THg (GB2762, 2017; Horvat et al., 2003; Li et al., 2015; Qiu et al., 2008). Notably, China is the largest country worldwide concerning the production and consumption of fish, seafood, and rice (FAOSTAT, 2020). Sixty-nine million tons of fish and seafood and 210 million tons of rice were harvested in China in 2016. Further, 16 million tons of fish and seafood (61% as fish) were consumed by Chinese residents in the same year (NBS, 2017). Nearly 50% of China's population live in coastal provinces, and fish and seafood are served as essential daily food needs (NBS, 2017). Furthermore, rice consumption was confirmed as the main pathway of human MeHg exposure in the Guizhou Hg mining areas (Feng et al., 2008; Li et al., 2012; Zhang et al., 2010). To evaluate the health risks of human Hg exposure, typical Hg-contaminated areas should be considered as hot spot areas. Coal combustion, Zinc and Lead smelting, and cement production were the main sectors of anthropogenic Hg emission in China. Meanwhile, Hg mining, gold mining, and chemical industries are the primary Hg pollution categories in Asia (Li et al., 2009; Wu et al., 2016).

Human hair is considered as a good biomarker for assessing human MeHg exposure and its associated health risks (Basu et al., 2018; Li et al., 2015; Li et al., 2008b; Srogi, 2007). Compared with the blood matrix, human hair samples are easier to collect, store, and transport (Clarkson, 2002; Li et al., 2015). MeHg in hair derives from that circulated in the blood during hair formation and is therefore endogenous in character (Srogi, 2007). Once absorbed into the hair, MeHg is largely preserved which can record the MeHg exposure. In general, hair contains from 250 to 300 times more total Hg than for blood, which enables a convenient analysis (Gill et al., 2002; Srogi, 2007). The international limits for hair Hg are 1.0 μg/g and 2.3 μg/g, which are adopted from the reference dose recommended by the United States Environmental Protection Agency (USEPA) and the provisional tolerable weekly intake set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA), respectively. Human hair collection to estimate Hg exposure has previously been performed in China (Cheng et al., 2009; Liu et al., 2014b; Liu et al., 2008). However, there is insufficient information for establishing a background level of human MeHg exposure, which is why continued human bio-monitoring is desirable.

This study aims to evaluate the health risks of human Hg exposure and provide effective insights for implementing the Minamata Convention in China. Rice and fish samples were collected from typical Hg-contaminated areas and major fish-rich areas across China to quantify the potential Hg pollution in paddy fields and aquatic ecosystems. Human hair samples were also systematically collected in the same areas to directly evaluate human Hg exposure's health risks. Different health endpoints on children and adults were calculated to quantify associated adverse effects based on hair THg concentrations.

2. Material and methods

2.1. Study areas

Fig. 1 shows the geographical location relative to Beijing of the four contaminated sites and the four urban centers along Chinese waterways. Following the given numbers, the first contaminated site is located near one of China's largest historic zinc smelters (ZSA), the second at Guizhou Province's largest coal-fired power plant (CFPPA), the third adjacent to a large-scale commercial gold mine in Guizhou (GMA), and finally the fourth is located in Wanshan mercury mining area (WMMA), where historically is the largest Hg mine in China.

The first place in the second category is Wuhan (WH) in Hubei Province which is the largest producer of freshwater food nationally (4.7 million tons), then follows Qingdao (QD) in Shandong Province which has the largest seafood production nationally (8.0 million tons), Xiamen (XM) in Fujian province with a high production of seafood (6.7 million tons) and finally Zhoushan (ZS) in the Yangtze River Delta that is China's largest fishing port. All statistical information in this section refers to 2016 and was retrieved from the National Bureau of Statistics (NBS, 2017). Changshun of Guizhou, where without large scale industrial activities, was used as a control site in this study (Du et al., 2018).

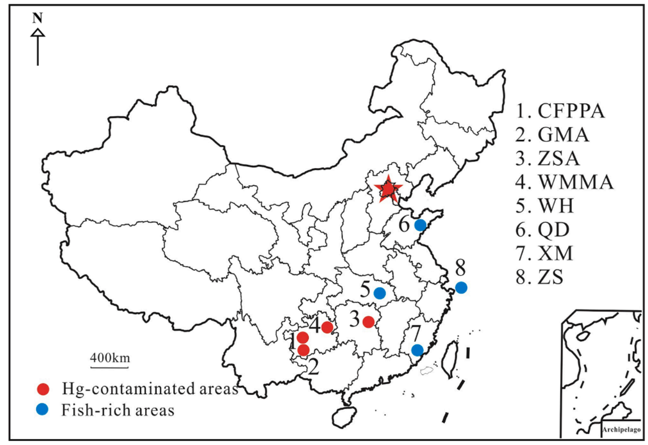

Fig.1. The spatial distribution of eight study areas Blue and red dots stand for sampling areas and the star for the location of Beijing. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.2. Sample collection

Fish were collected from typical markets in every study area, which mainly consisted of cultured fish. As listed in Table S1, the collected species covered the main fish consumed by local inhabitants in each area. The edible parts (30–80 g) of the fish were dissected by scalpel and preserved in polyethylene bags with ice before taken into the lab. In total, 624 fish samples were collected in the study areas.

Rice samples were obtained for the sites GMA, CFPPA and ZSA from the local population's housing stock and from local markets in the urban centers of WH, QD, and XM. Three sampling sites at different distances downwind of the pollution source were selected for collecting rice samples in each Hg-contaminated area. Three typical markets were selected for collecting rice samples in each fish-rich area. A total of 299 rice samples were collected in this study.

The human hair samples in the Hg-contaminated areas were collected from residents' houses, and the consumed rice samples were collected simultaneously. Meanwhile, the participants in fish-rich areas were recruited on the street. Two urban areas and one suburban area were selected for hair sample collection to ensure the hair samples' representativeness for each fish-rich city. The criterion for selecting the participants was that they had stayed in the corresponding area for more than three months, and the selected participants were no occupational exposure and covered all age and gender groups. The participants with dyed hair or dental prosthesis were excluded in this study. The recruitment strategy for this study was to include as many participants as possible. Finally, the number of human hair samples was 994 in all study areas, as shown in Table S2. Human hair samples were cut from the occipital region of every participant's scalp using stainless steel scissors, packed into a clean polyethylene bag, and transported to the laboratory for subsequent pre-treatment and analysis.

Human hair and rice Hg data in WMMA were adopted from our previous study (Li et al., 2015). Rice Hg data in ZS were adopted from Xu et al. (2019). Questionnaires were also executed in the study areas, which collected information on age, gender, body weight, height, and occupation (Table S3). This study obtained ethics approval from the Institute of Geochemistry, Chinese Academy of Sciences. All participants signed an informed consent form before engaging in the survey.

2.3. Analytical methods

After being dried in an oven (DHG-9030, Yiheng, China) at 50℃ for 48 h, rice samples were crushed by an electric mill (AQ-180E-X, Cixinaiou Electric Appliance Co., China) and sieved into 150 meshes. Fish samples were freeze-dried (EYELA, Tokyo, Japan) at −80℃ and 10 Pa for 72 h before being crushed into powder. The hair samples were washed with non-ionic detergent, ultrapure water, and acetone and dried in the oven at 45℃ overnight. The pretreated hair samples were cut into pieces with clean scissors. Finally, the rice, fish, and hair samples were stored in a dark drying box for further analysis.

The prepared samples were weighted ~ 0.05 g for hair, ~0.1 g for fish, and ~ 0.4 g for rice, respectively, and transported into quartz boats before analysis. THg concentrations in human hair, fish, and rice samples were determined by a direct mercury analyzer (DMA-80, Milestone, Italy) per the USEPA standard method 7473 (USEPA, 2007). Further, 120 rice, 361 human hair, and 287 fish samples were chosen for MeHg analysis. These samples covered high, medium, and low THg concentrations in each area. For MeHg analysis, ~0.2 g of the rice samples was digested by a KOH/solvent extraction technique, while ~ 0.1 g fish and ~ 0.05 g hair samples were digested with KOH following our previous study (Du et al., 2018). Subsequently, the digested solutions were ethylated, purged, trapped, and measured via GC-CVAFS (Brooks Rand Model III) (Liang et al., 1996). Information on quality assurance and quality control is listed in SI Section 1 and Table S4.

2.4. Calculation of probable daily intake (PDI)

PDIs of MeHg for residents in the study areas were calculated to estimate daily MeHg intake via rice and fish consumption using the following formula:

$$\mathrm{PDI} = \left(\mathrm{C} \times \mathrm{IR} \times 10^{-3}\right)/bw \tag{1}$$

where PDI is given in μg/kg/d, bw is body weight (adopted from the questionnaire), C is the MeHg concentration in the rice or fish samples (ng/g), and IR is the daily intake rate (g/d), as listed in Table S5.

2.5. Dose-response from Hg exposure

The changes in IQ and fatal heart attack risk were used as endpoints for the risk assessments of Hg exposure in children and adults, respectively (Zhang et al., 2018). Children's IQ would decrease by 0.18 points for each μg/g maternal hair Hg from fish consumption (Axelrad et al., 2007), and by 1 point for each μg/g children hair Hg via rice consumption (Feng et al., 2020).

Therefore, the change in children's IQ (ΔIQ) caused by Hg exposure via consumption of fish or rice was calculated using the following equations:

$$\Delta \mathrm{IQ} = \Delta \mathrm{IQ}_{aqu+} \Delta \mathrm{IQ}_{rice} \tag{1}$$

$$\Delta IQ_{aqu} = \alpha \times \overline{C}_{hair-aqu} \tag{2}$$

$$\Delta \mathrm{IQ}_{rice} = \gamma \times \psi \times \overline{C}_{hair-rice} \tag{3}$$

$$\overline{C}_{hair-aqu} = \xi_1 \times s \times \overline{C}_{hair} \tag{4}$$

$$\overline{C}_{hair-rice} = \xi_2 \times s \times \overline{C}_{hair} \tag{5}$$

where ΔIQ_{aqu} and ΔIQ_{rice} are the changes of children's IQ via the consumption of fish and rice, respectively; α is the coefficient of the relationship between ΔIQ_{aqu} and $\overline{C}_{hair\text{-}aqu}$; $\overline{C}_{hair\text{-}aqu}$ is the average THg hair concentration caused by fish consumption, α is the decrement in IQ point for each μg/g maternal hair Hg via consumption of fish; γ is the decrement in IQ points for each μg/g of child hair Hg via rice consumption; is the average THg concentration in maternal hair caused by consumption of rice; ψ is the ratio of $\overline{C}_{hair\text{-}rice\text{-}child}/\overline{C}_{hair\text{-}rice}$; $\overline{C}_{hair\text{-}rice\text{-}child}$ is the average hair THg concentration in child caused by consumption of rice; s refers to the ratio of THg contribution to human from rice and fish in China; ξ_1 and ξ_2 are the THg contribution ratios of rice and fish, respectively, in the study areas (Fig. S1); $\overline{C}_{hair}$ is the mean THg concentration of human hair in the study areas.

A modified risk model was used for assessing the changes in fatal heart attack risk (Rice et al., 2010; Zhang et al., 2018):

$$\Delta \mathrm{C}_f = \mathrm{C}_f \times \left(1 - \exp\left(-\varphi \times \Delta \mathrm{M}_{hair-np}\right)\right) \tag{6}$$

$$\Delta \mathrm{M}_{hair-np} = \mathrm{M}_{hair-np} - \mathrm{M}_{hair-bg} \tag{7}$$

$$\mathrm{IMB} = \Delta \mathrm{C}_f \times \mathrm{TPS} \tag{8}$$

$$\mathrm{IMT} = \Delta \mathrm{C}_f \times \mathrm{TPS} \times \theta \tag{9}$$

where ΔC_f and C_f are the change and original risk of fatal heart attack in China, respectively; φ is the response coefficient of the fatal heart attack risks; $\Delta M_{hair\text{-}np}$ and $M_{hair\text{-}np}$ are the alternation and original hair THg concentration of the non-pregnant population, respectively; $M_{hair\text{-}bg}$ is the hair THg concentration of the background population; IMB and IMT are the increase of morbidity and mortality of fatal heart attack risk, respectively; θ is the mortality of heart attacks; and TPS is the total population size at risk (see Table S6). Meanwhile, lifetime earnings would decrease with the decrement of IQ, resulting economic costs for local society (SI Section 2). Hair MeHg concentrations were positively correlated with dietary MeHg intakes, and the simulated methods were introduced in SI Section 3.

3. Results

3.1. Fish Hg

THg concentrations in fish collected from CFPPA, GMA, and ZSA were comparable, showing relatively low levels with a geomean of 5.36 ng/g (1.59–26.4 ng/g, n = 42). Meanwhile, the fish THg concentrations (geomean: 11.0 ng/g, range: 4.44–23.5 ng/g, n = 8) in WMMA were significantly elevated (Fig. 2, Table S7). Fish MeHg concentrations in CFPPA, GMA, ZSA, and WMMA were relatively low with a geomean of 2.63 ng/g (0.57–14.6 ng/g, n = 28) (Fig. 2, Table S7). These figures were much lower than the national limit (500 ng/g). Fish species collected in the Hg-contaminated areas were classified into 2 groups (omnivore and herbivore) according to the feeding habitats. In the typical Hg-contaminated areas, the geomeans of THg and MeHg concentrations in omnivore fish were 7.33 ng/g (1.35 ~ 29.9 ng/g) and 4.37 ng/g (1.40 ~ 14.6 ng/g), respectively, which were significantly ($p < 0.05$) higher than

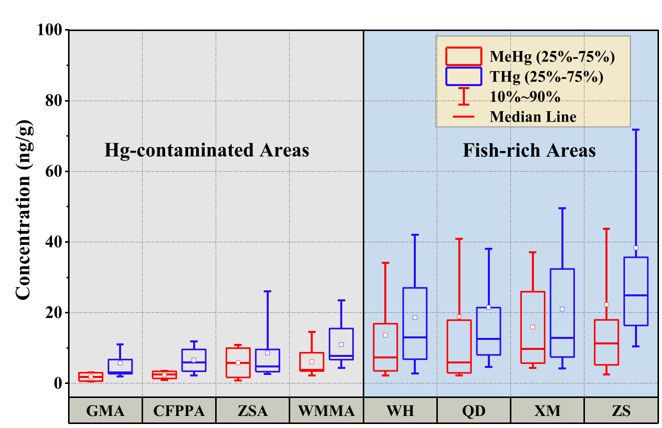

Fig.2. Box-plot of THg and MeHg concentrations in fish from the study areas. Each box represents the median quartile range (25th to 75th quantiles). The white squares indicate the mean values.

those in herbivore fish (THg: geomean: 3.19 ng/g, range: 2.01 ~ 8.20 ng/g; MeHg: geomean: 1.35 ng/g, range: 0.79 ~ 3.43 ng/g) (Fig. S2). Especially, THg concentrations in *Aristichthys nobilis* and *Pelteobagrus fulvidraco* (omnivore fish) were relatively high, which averaged at 20.5 ± 8.80 ng/g and 12.2 ± 3.88 ng/g, respectively.

The geomeans of THg concentrations in fish collected from WH, QD, and XM were 12.1 ng/g (1.39–106 ng/g, n = 118), 12.7 ng/g (0.79–221 ng/g, n = 169), and 14.4 ng/g (1.99–79.7 ng/g, n = 146), respectively (Fig. 2, Table S7). The fish THg concentrations from ZS were much higher, and the geomean was 26.9 ng/g (3.73–456 ng/g, n = 149). Meanwhile, the geomeans of MeHg concentrations in the fish from WH, QD, XM, and ZS were 8.24 ng/g (1.09–75.9 ng/g, n = 30), 7.62 ng/g (0.54–148 ng/g, n = 47), 11.3 ng/g (1.00–51.9 ng/g, n = 47), and 10.9 ng/g (0.77–341 ng/g, n = 149), respectively. None exceeded the national limit (500 ng/g) (Fig. 2, Table S7). The fish THg and MeHg concentrations in fish-rich areas were higher than those from cultured fish in the Hg-contaminated areas. Fish species collected in fish-rich areas were classified into 3 groups (carnivore, omnivore and herbivore) according to the feeding habitats. The geomeans of THg concentrations in carnivore, omnivore and herbivore fish from fish-rich areas were 24.3 ng/g (1.39 ~ 456 ng/g), 20.9 ng/g (1.99 ~ 145 ng/g), and 6.20 ng/g (2.34 ~ 22.8 ng/g), respectively (Fig. S3). Besides, the geomeans of MeHg in carnivore, omnivore and herbivore fish from fish-rich areas were 12.1 ng/g (1.09 ~ 341 ng/g), 11.5 ng/g (1.22 ~ 65.4 ng/g) and 3.76 ng/g (1.54 ~ 8.94 ng/g), respectively (Fig. S3). Particularly, the THg concentrations in *Muraenesox cinereus* and *Lateolabrax japonicus* averaged at 204 ± 168 ng/g and 136 ± 88.1 ng/g, respectively.

The average percentage of THg as MeHg (MeHg%) was around 50% in fish from the fish-rich areas and ZSA. However, it was only 30% in those from WMMA, CFPPA, and GMA.

3.2. Rice Hg

THg levels of rice collected from CFPPA and GMA averaged at 3.63 ng/g (1.05–11.4 ng/g, n = 126) and 4.46 ng/g (3.13–8.67 ng/g, n = 65), respectively. The results were comparable with the background area values (geomean: 4.73 ng/g, Fig. 3a, Table S8). Rice THg concentrations in ZSA and WMMA were significantly ($p < 0.05$) elevated, and the geomeans were 5.98 ng/g (3.02–30.7 ng/g, n = 70) and 24.0 ng/g (1.35–401 ng/g, n = 89), respectively. Approximately 1.4% of rice THg concentrations in ZSA surpassed the national limit (20 ng/g). Meanwhile, 53% in WMMA exceeded the national limit. The MeHg concentrations in rice samples collected from the four Hg-contaminated areas showed similar patterns with THg (Fig. 3, Table S8). Rice MeHg concentrations in CFPPA (2.04 ng/g, 1.01–5.80 ng/g, n = 31) and GMA (2.55 ng/g, 1.05–3.26 ng/g, n = 27) were relatively low among the Hg-contaminated areas, and the rice MeHg concentrations in ZSA (5.29 ng/g, 1.88–21.1 ng/g, n = 25) and WMMA (8.97 ng/g, 1.13–45.1 ng/g, n = 168) were significantly ($p < 0.05$) elevated.

Rice samples in WH, QD, XM, and ZS showed low THg levels (geomean: 2.46 ng/g, range: 0.61–6.52 ng/g; Fig. 3, Table S8), which were comparable with the background values and were much lower than those in ZSA and WMMA. No sample in WH, QD, XM, and ZS exceeded the national limit. The geomeans of MeHg concentrations in WH, QD, XM, and ZS were around 1 ng/g, much lower than those in the Hg-contaminated areas (Fig. 3, Table S8).

The average MeHg% in ZSA was as high as 76%±17%. In contrast, the MeHg% in rice samples in the other study areas averaged at 50%. A significant positive correlation was found between rice THg and MeHg concentration if considering all the rice samples ($r = 0.55$, $p < 0.01$).

3.3. Hair Hg

Hair THg concentrations in CFPPA and GMA averaged at 0.24 μg/g (0.08–11.93 μg/g, n = 156) and 0.33 μg/g (0.10–2.92 μg/g), respectively. These figures were comparable with background values

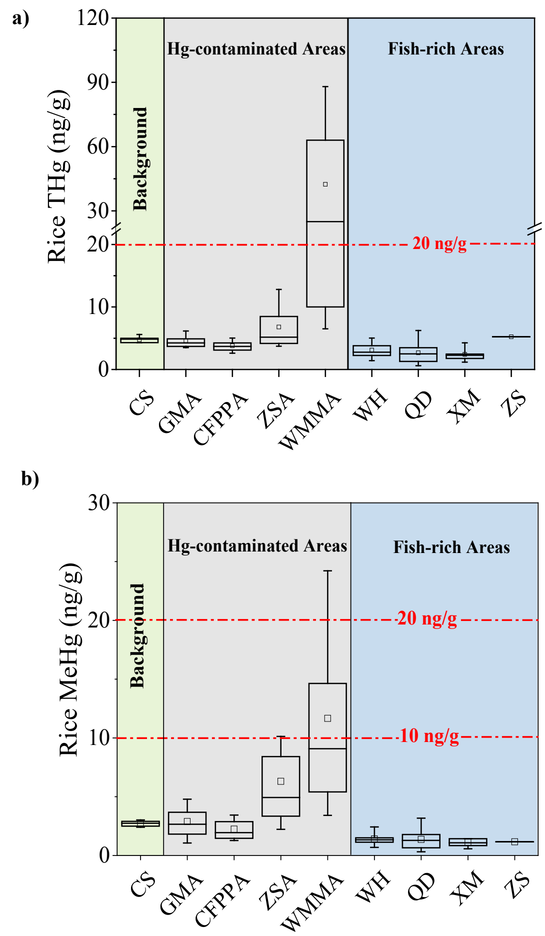

Fig.3. THg and MeHg concentrations in rice from the study areas. Rice data in the background sites are adopted from Du et al., (2018), while data in ZS and WMMA are adopted from Xu et al., (2019) and Li et al., (2015). Each box represents the median quartile range (25th to 75th quantiles). The band near the middle of the box is the 50th percentile (the median). The whisker spans from the 10th to 90th quartiles, and open squares indicate the average values.

(geomean: 0.33 μg/g) (Fig. 4, Table S2). Hair THg concentrations in ZSA and WMMA were significantly ($p < 0.05$) elevated compared with background values, and the geomeans were 0.60 μg/g (0.13–1.82 μg/g, n = 125) and 2.20 μg/g (0.41–34.1 μg/g, n = 168), respectively. About 17% of hair THg concentrations in ZSA and 85% in WMMA exceeded the limit (1 μg/g) set by the USEPA. Meanwhile, 48% of hair samples in WMMA exceeded the limit (2.3 μg/g) recommended by the WHO. In CFPPA, GMA, and ZSA, the geomean of hair MeHg concentrations was 0.17 μg/g (0.03–1.14 μg/g, n = 119), which compared with the background value (0.21 μg/g). The hair MeHg concentrations in WMMA were significantly ($p < 0.05$) elevated with a geomean of 1.52 μg/g (0.26–10.9 μg/g, n = 168).

Hair THg concentrations in WH and QD averaged at 0.41 μg/g (0.09–2.24 μg/g, n = 140) and 0.49 μg/g (0.05–5.40 μg/g, n = 150), respectively. These figures were slightly higher than the background value. However, hair THg concentrations in XM and ZS were significantly ($p < 0.05$) elevated, with the geomeans of 0.81 μg/g (0.11–9.18 μg/g, n = 160) and 1.14 μg/g (0.13–8.18 μg/g, n = 141), respectively. Hair MeHg concentrations in WH and QD were similar, and the geomeans were 0.32 μg/g (0.06–1.12 μg/g, n = 41) and 0.26 μg/g (0.05–3.31 μg/g, n = 41), respectively. Hair MeHg concentrations in XM and ZS were significantly elevated, and the geomean in XM (0.90 μg/g, 0.07–5.40 μg/g, n = 67) was much higher than that in ZS (0.50 μg/g, 0.01–5.87 μg/g, n = 139).

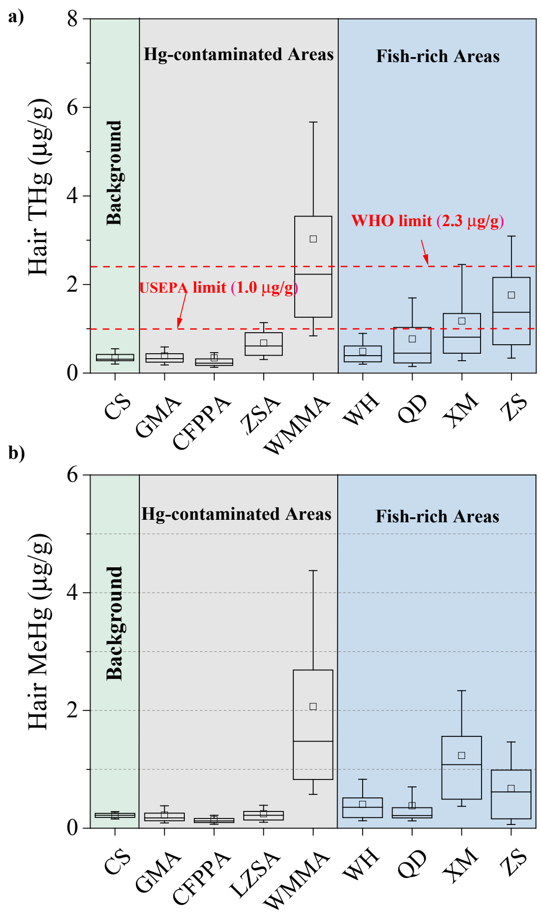

Fig.4. Human hair THg (a) and MeHg (b) concentrations in the study areas. Data from Changshun town (CS) are adopted from Du et al., (2018). Each box represents the median quartile range (25th to 75th quantiles). The band near the middle of the box is the median. The whisker spans from the 10th to 90th quartiles, and open squares indicate the mean values.

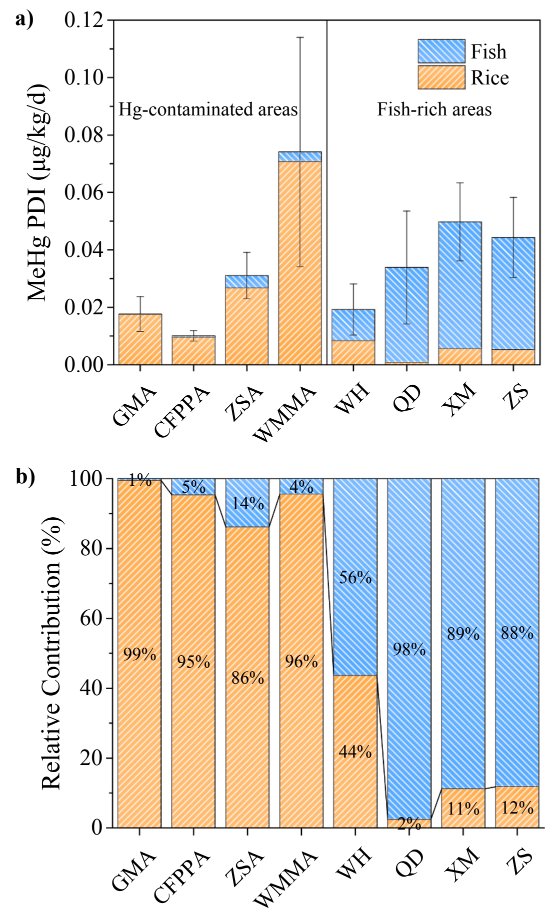

Fig. 5. The PDIs of MeHg by consuming rice and fish (a), and the relative contribution (b) in the study areas.

3.4. *Probable daily intake (PDI) of MeHg*

Based on the daily intake rate and MeHg concentrations in fish and rice, PDIs of MeHg in the study areas were calculated (Fig. 5). PDIs of MeHg in CFPPA and GMA averaged at 0.010 ± 0.002 μg/kg/d and 0.018 ± 0.006 μg/kg/d, respectively. Meanwhile, the PDIs of MeHg in ZSA (0.031 ± 0.008 μg/kg/d) and WMMA (0.075 ± 0.039 μg/kg/d) were much higher. In contrast, the PDIs of MeHg in WH and QD were 0.019 ± 0.009 μg/kg/d and 0.034 ± 0.019 μg/kg/d, respectively. The values in XM (0.050 ± 0.014 μg/kg/d) and ZS (0.044 ± 0.014 μg/kg/d) were significantly ($p < 0.05$) elevated. Based on the PDIs of MeHg in these eight study areas, hair MeHg concentrations were simulated. The simulated results showed a significant ($p < 0.05$) relationship with the measured hair MeHg concentrations (Table S2).

3.5. *Dose response and economic cost*

The individual's IQ in WMMA decreased the most due to its Hg exposure, followed by ZS and ZSA, and then GMA, CFPPA, WH, XM, and QD (Table 1). The individual IQ loss in WMMA was 0.62 points. Meanwhile, the IQ losses in GMA, CFPPA, WH, and XM showed similar levels and averaged at ~ 0.08. The total IQ loss for children in WH was largest (10540). The increased number of mortality (IMT) and morbidity (IMB) from heart attacks caused by Hg exposure for the non-pregnant population in these eight study areas are also listed in Table 1. The IMB and IMT in fish-rich areas were higher than the results in the four Hg-contaminated areas. The economic costs caused by Hg exposure in the four Hg-contaminated areas were lower than $0.05 billion. Meanwhile, these in four fish-rich areas were higher than $0.1 billion, particularly for WH, reaching up to $0.79 billion.

4. Discussion

4.1. *Food Hg pollution*

The THg and MeHg concentrations in fish from the studied Hg-contaminated areas (CFPPA, GMA, ZSA, and WMMA) were considerably low (Fig. 2, Table S7). These were much lower than those in one Hg mining area (average: 293 ng/g) (Qiu et al., 2009), an electronic waste recycling area (average: 229.8 ng/g) (Tang et al., 2015), a small-scale artisanal gold mining area (average: 460 ng/g) (Bose-O'Reilly et al., 2016), and the vicinity of a chloralkali plant in Cuba (1093 ± 319 ng/g in a catfish and 596 ± 233 ng/g in an oyster) (Feng et al., 2019). This study's fish samples were purchased from the local market. They were artificially cultured in the pond. Therefore, the food chain was very short, and the fish grew rapidly, causing low Hg concentrations. The length and body weight of fish were essential surrogates for their lifespan (Sun et al., 2020). Researchers note that the body weight and length were positively correlated with the THg and MeHg concentrations, indicating the elevated bioaccumulation of Hg with the increased lifetime (Sinkus et al., 2017; Zhu et al., 2013). However, no significant relationship was found between the Hg concentrations and the body weight and length of fish in this study (Fig. S4). The phenomenon might be influenced by that different fish species present various Hg concentrations, even for similar-sized fish consuming the same food pellets

Table 1
Health impacts and economic costs caused by mercury exposure.

	GMA	CFPPA	ZSA	WMMA	WH	QD	XM	ZS
ΔIQ	−0.10	−0.07	−0.16	−0.62	−0.08	−0.03	−0.08	−0.15
Total IQ Loss	748	349	421	1732	10,540	2489	4745	1555
ΔC_f (1/100000)	0.14	–	3.03	19.29	0.92	1.86	5.29	9.02
IMB (Persons)	0.59	–	7.67	52.07	100.07	174.65	217.36	105.57
IMT (Persons)	0.08	–	1.03	7.03	13.51	23.58	29.34	14.25
Costs (Billion$)	0.049	0.033	0.024	0.039	0.796	0.160	0.299	0.117

Note: "IMB" and "IMT" refer to the increase of morbidity and mortality of fatal heart attack risk, respectively; "-" represent no influence on heart attack risk, and "Costs" refer to the economic costs caused by mercury exposure.

(Wang and Wang, 2019). In the typical Hg-contaminated areas, the THg and MeHg concentrations in omnivore fish were 9.68 ± 7.75 ng/g and 5.48 ± 4.16 ng/g, respectively. These were significantly ($p < 0.05$) higher than those in herbivore fish (THg: 3.43 ± 1.58 ng/g; MeHg: 1.61 ± 1.11 ng/g) (Fig. S2). Meanwhile, the omnivore fish's body weight and length were comparable with those in herbivore fish ($p > 0.05$, Fig. S5).

The fish THg and MeHg concentrations in fish-rich areas (WH, QD, XM, and ZS) were higher than those in the four Hg-contaminated areas (Fig. 2, Table S7), suggesting the intensive bioaccumulation of fish Hg in fish-rich areas. The fish MeHg concentrations in fish-rich areas compared with those in the coastal area (2.3–303.6 ng/g) and Three Gorges Reservoir (0.10–199 ng/g) (Xu et al., 2018; Zhang et al., 2018). However, the results were much lower than those of wild fish in the South China Sea (mean: 105 ng/g, 6.22–358 ng/g) and the Tibetan Plateau (mean: 90.7 ng/g, 24.9–1196 ng/g) (Liu et al., 2014a; Zhang et al., 2014). It can be concluded that the cultured fish showed low THg and MeHg concentrations were comparable with the wild fish. This finding was attributed to their short lifetime and rapid growth in a cultured environment. Meanwhile, no fish from fish-rich areas exceeded the national limit (500 ng/g). The THg concentrations in fish from ZS were higher than those from XM. However, the MeHg concentrations were comparable. Water exchange in the coastal bay of XM is rather poor, resulting in an extension of the residence time of water Hg and the increase of Hg methylation (Wang and Wang, 2019). Additionally, different fish species communities may present different Hg levels. No positive relationship was found between the Hg concentrations and the body weight and length of fish in fish-rich areas (Fig. S6). This also might be attributed to various Hg concentrations between different species. Generally, the fish THg concentrations in the four fish-rich areas follow this order: carnivore > omnivore > herbivore. In contrast, MeHg concentrations follow this sequence: carnivore ~ omnivore > herbivore (Fig. S3), which were comparable with the previous result (Wang and Wang, 2019). The herbivore fish showed a higher body weight than the omnivore and carnivore fish ($p < 0.05$), while the herbivore, omnivore, and carnivore fish showed similar body lengths ($p > 0.05$, Fig. S7).

Rice samples collected from ZSA and WMMA showed elevated THg and MeHg concentrations (Fig. 3, Table S8), showing potential rice Hg pollution. CFPPA and GMA were considered typical Hg-contaminated areas in China. Still, rice THg and MeHg concentrations in these two areas were relatively low and comparable with the background values. The distributions of rice THg and MeHg concentrations in the four Hg-contaminated areas can be explained by soil and atmospheric Hg pollution. Soil and atmospheric Hg are the main sources of THg in rice grains, as revealed by stable Hg isotope ratio systematics (Yin et al., 2013). Our previous study found that the geomeans of soil THg concentrations in WMMA, ZSA, GMA, and CFPPA were 34.2 μg/g, 0.54 μg/g, 0.42 μg/g, and 0.13 μg/g, respectively (Song et al., 2021). These figures were significantly elevated compared with the national background of 0.12 μg/g (CNEMC, 1990). The ambient air TGM concentrations averaged at 34.7 ± 16.3, 10.8 ± 5.4, 4.6 ± 1.5, 5.0 ± 1.7 ng/m^3 in WMMA, ZSA, GMA, and CFPPA, respectively (Table S9) (Song et al., 2021). Meanwhile, the newly deposited Hg from the atmosphere was more readily transformed to MeHg (Branfireun et al., 2005; Meng et al., 2010). Therefore, the elevated soil THg and TGM levels in WMMA and ZSA resulted in high rice THg levels.

The rice THg and MeHg concentrations in WH, QD, XM, and ZS were far lower than those from the four Hg-contaminated areas (Fig. 3, Table S8), and none exceeded the national limit (20 ng/g). The results were comparable with the THg levels in rice samples collected on-site (Geomean: 4.74 ng/g, range: 1.06–22.7 ng/g) and purchased from markets (Geomean: 3.97 ng/g, range: 0.64–31.70 ng/g) in China (Xu et al., 2019; Zhao et al., 2019).

4.2. MeHg exposure pathway

Previous studies indicated that rice and fish consumption are the major MeHg exposure pathways in China (Cheng et al., 2009; Li et al., 2012; Liu et al., 2018; Zhang et al., 2010). In this study, rice was the main pathway (>85%) for human MeHg exposure in Hg-contaminated areas. Meanwhile, fish consumption contributed to the majority (>85%) of MeHg exposure in coastal cities. However, in WH, rice, and fish consumption contributed equally to human MeHg exposure (Fig. 5b). The relative contribution of fish consumption to human MeHg exposure decreased distinctly from coastal cities to inland areas, consistent with our previous study (Li et al., 2012). The dietary structure varies significantly in different residential regions. Therefore, the relative contributions of MeHg exposure from rice and fish consumption also vary correspondingly (Table S5). International and interprovincial food trades potentially changed the structure of rice and fish Hg concentrations in special region (Liu et al., 2018). If residents in Hg-contaminated areas consume rice from the other areas, rice consumption would contribute less MeHg exposure for human. Besides, marine fish were transported to inland areas and consumed by inland residents, which would increase MeHg exposure for the inhabitants. Previous study indicated that MeHg bioaccessibilities in rice and fish were 40.5 ± 9.4% and 61.4 ± 14.2%, respectively (Gong et al., 2018). And the study indicated that rice consumption contributed 15.1% to human MeHg exposure in Shanghai and 6.6% in Guangdong, which were comparable with the results obtained in QD (2%), XM (11%) and ZS (12%) in this study, indicating minor contributions from rice consumption in coastal provinces. Rice consumption accounted 99% of total MeHg exposure in GMA, 95% in CFPPA and 96% in WMMA, which were higher than the result in Guizhou (81.5%) evaluated by Gong et al. (2018).

Based on the PDIs of MeHg from rice and fish consumption, the hair MeHg concentrations were simulated per the model as stating in SI Section 3. The simulated and measured hair MeHg concentrations were comparable in most study areas when considering variability ($r = 0.81$, $p < 0.05$, Fig. S8). These results also confirmed that the calculation of PDIs of MeHg and analysis of human MeHg exposure sources in this study were accurate. It provided direct evidence on that consumption of rice and fish were main MeHg exposure pathways in China.

4.3. Health risk

Human hair Hg concentrations are considered good biomarkers for assessing human Hg exposure since the limit of hair THg concentrations is recommended by an international agency. As shown in Table S2, two and five hair samples in CFPPA and GMA exceeded 1 μg/g, respectively.

However, >40% of the hair samples surpassed 1 μg/g in WMMA, XM, and ZS, indicating potential human health risks from Hg exposure in these areas. Hair THg concentrations in WMMA were similar to those of occupational workers in a chlorine-alkali plant in Pakistan (mean: 2.45 μg/g) and individuals in a small-scale artisanal gold mine of the Philippines (mean: 2.72 μg/g) (Drasch et al., 1999; Elgazali et al., 2018). However, the results were much higher than those of occupational workers in a compact fluorescent lamp factory (1.36 ± 2.90 μg/g) and an electronic waste recycling factory in China (1.07 ± 0.21 μg/g) (Liang et al., 2015; Tang et al., 2015). Therefore, more attention should be paid to human Hg exposure in Hg mining areas, chlorine-alkali plants, and artisanal and small-scale gold mines. Hair THg concentrations in GMA and CFPPA were comparable with those in a cement plant area in the USA (0.44 ± 0.46 μg/g) (Dong et al., 2015). These three areas were considered as typical Hg emission areas. Still, low health risks from human Hg exposure were found in these areas. Hair THg in QD were comparable with those of Hainan islanders (geomean: 0.69 μg/g) (Liu et al., 2014b) and Shanghai citizens (geomean: 0.50 μg/g) (Liu et al., 2008). Hair THg concentrations in ZS were significantly elevated, identical to those in the Pearl River Delta (1.08 ± 0.94 μg/g) and Ningbo City (geomean: 1.04 μg/g) (Liu et al., 2008; Shao et al., 2013). The different hair Hg levels along the Chinese coastline might be due to different dietary habits of fish consumption and different fish Hg levels.

Hair MeHg concentrations in GMA, CFPPA, and ZSA were comparable with those observed in a compact fluorescent lamp manufacturing area (mean: 0.15 μg/g) and electronic waste recycling area in China (0.35 ± 0.21 μg/g). These were also similar to the background values (Fig. 4, Table S2). Hair MeHg concentrations in WH, XM, and ZS were comparable with those observed in the Pearl River Delta (Shao et al., 2013). However, they were lower than those of the ZS fishermen since fishermen consumed large amounts of fish with high MeHg levels (Cheng et al., 2009). The hair MeHg concentrations in WMMA were similar to these in a Wuchuan Hg-mining area in China (0.52–4.21 μg/g) and chlorine-alkali plant area in Pakistan (2.45 μg/g) (Elgazali et al., 2018; Li et al., 2008a), indicating high health risks from Hg exposure in these three areas.

Hair THg concentrations showed a significant relationship with age ($p < 0.05$, Fig. 6a). The hair THg concentrations increased from a median of 365 ng/g (the median) in a group of 0- to 10-year-olds to 874 ng/g in 50- to 60-year-olds before slightly decreasing. The results might be related to the capacity of human metabolism. The hair THg concentrations from males in groups of 31- to 40-, 51- to 60-, and 61-to 70-year-olds were significantly ($p < 0.05$) higher than females. Meanwhile, the THg concentrations in other age groups showed no gender differences ($p > 0.05$, Fig. 6b). The larger amount of daily diet intake may result in higher hair THg concentrations in males. Hair MeHg concentrations showed no significant trend with age ($p > 0.05$, Fig. 6c), and no gender difference of hair MeHg concentrations was observed at different age groups (Fig. 6d).

All PDIs of MeHg in GMA, CFPPA, and WH were lower than the RfD of 0.10 μg/kg/d set by the USEPA (Fig. 5a), indicating the low health risks from MeHg exposure in these areas. Approximately 24% of the PDIs of MeHg in WMMA, 11% in XM, 7% in ZS, 6% in QD, and 4% in ZSA exceeded the RfD of 0.10 μg/kg/d. This illustrates potential MeHg exposure risks in these areas. Further, the PDIs of MeHg in WMMA showed a declining trend when compared with the value calculated by Zhang et al. (2010a) since Hg pollution control has been implemented in this area. The PDIs of MeHg in the fish-rich areas (WH, QD, XM, and ZS) were comparable with those in the South China Sea (average: 0.054 μg/kg/d) (Liu et al., 2014b). However, these much lower than those in the ZS (0.88 μg/kg/d) (Cheng et al., 2009), Shunde (0.25 μg/kg/d) and Zhongshan fishermen (0.43 μg/kg/d) in the Pearl River Delta (Shao et al., 2013). The average of PDIs of MeHg in WMMA (0.074 μg/kg/d), XM (0.050 μg/kg/d) and ZS (0.044 μg/kg/d) were comparable with

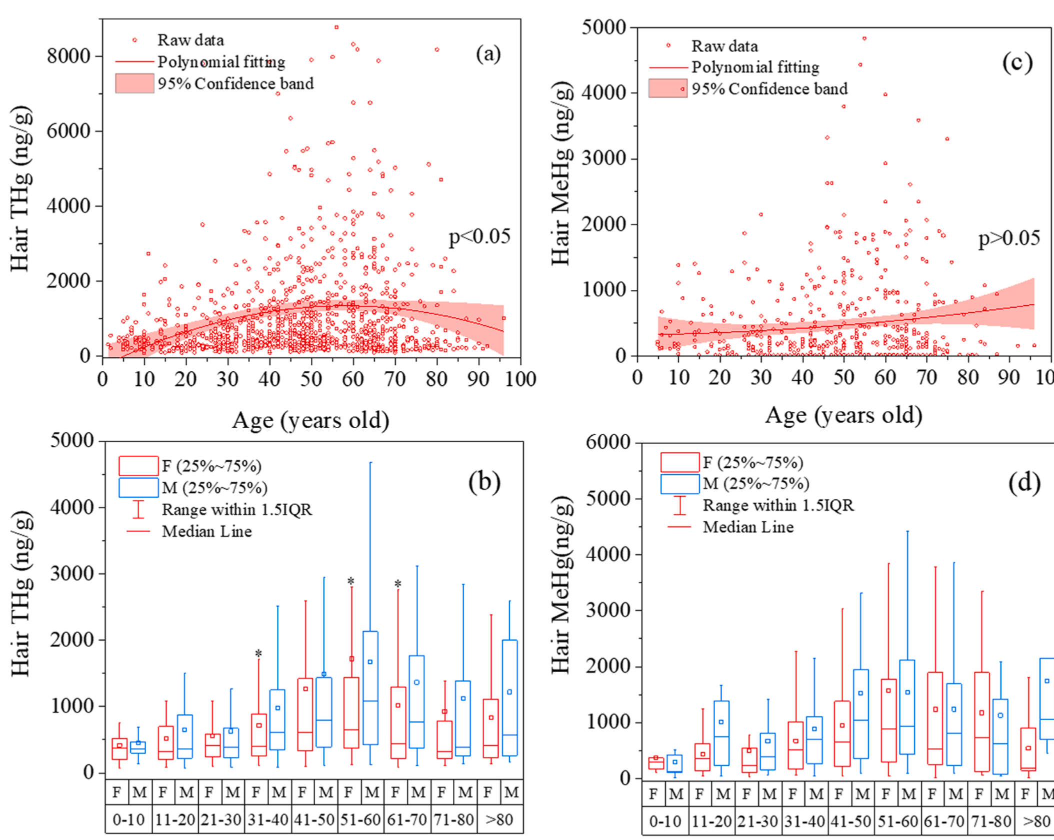

Fig. 6. The relationship between age and hair THg (a) and MeHg (c) concentrations; and the hair THg (b) and MeHg (d) concentrations in the female (F) and male (M) at different ages.

those for the Chinese population (0.057 μg/kg/d) (Liu et al., 2018), while PDIs of MeHg in CFPPA (0.010 μg/kg/d), GMA (0.018 μg/kg/d) and WH (0.019 μg/kg/d) were lower than 0.057 μg/kg/d. PDIs of MeHg from fish consumption in GMA, CFPPA, ZSA and WMMA were relatively low, and those in fish-rich areas were significantly elevated (Fig. S9). Especially, about 11% of PDIs of MeHg in XM, 7% in ZS and 6% in QD exceeded 0.10 μg/kg/d (Fig. S9), which may attribute to that specific fish species displayed extremely high MeHg concentrations (Fig. S9). Because pregnant women are sensitive to MeHg exposure, herbivore fish species were recommended for consumption to balance the benefit and risk. High-income individuals ingested more fish than the low-income population (Bowman, 2007), which potentially posed higher MeHg exposure. Therefore, socio-economic status differences may play an important role in regulating MeHg exposure level.

4.4. Health impacts

In this study, the health impacts caused by Hg exposure were directly assessed through human hair THg concentrations, ensuring low uncertainty in the calculation. The IQ points losses in GMA, CFPPA, WH, and XM were comparable with those associated with rice consumption in Bangladesh (−0.10), Philippines (−0.084) and Nepal (−0.082), and the average across China (−0.08), as contributed by various anthropogenic sources (Chen et al., 2019; Liu et al., 2019b). The total IQ loss for children in WH was 10540, which was the largest among the different study areas since this area had the most newborns (137,000) in 2016 (NBS, 2017). There is an increased heart attack risks in WMMA and ZS due to the subjects' high Hg exposure level. Generally, the IMT and IMB in the fish-rich areas were significantly higher than those in the four Hg-contaminated areas due to the large population in these big cities.

Due to decreased IQ points, economic costs were estimated, as was done by earlier studies (Rice et al., 2010; Salkever, 1995; Trasande et al., 2005). The fish-rich areas' economic costs were relatively high, and all exceeded $0.1 billion in 2016. The economic costs in GMA, CFPPA, and WMMA exceeded 1% of local total gross domestic product, suggesting the negative impacts of human Hg exposure on the local economy. Nearly half the population in China live in the coastal provinces. Therefore, high potential economic costs will be induced by human Hg exposure.

5. Conclusions and implications

Mercury pollution in rice and fish and associated human Hg exposure were investigated in typical Hg-contaminated areas and major fish-rich areas in China. Except for the rice in WMMA, THg, and MeHg concentrations in rice and fish samples were relatively low in this study. Hair THg concentrations in WMMA, ZSA, XM, and ZS were significantly elevated, suggesting potential health risks from Hg exposure for residents. Hg exposure has significant impacts on mental development and heart diseases. The largest alterations were found in WMMA, with a decreased IQ by 0.62 points and an increased risk of heart attack risk by 0.19‰ compared with cases without Hg exposure.

This study found that zinc smelting and Hg mining activities could result in severe environmental Hg pollution, Hg bioaccumulation in rice and fish, and human Hg exposure. More attention should be paid to highly Hg polluted areas (such as the Hg mining area and zinc smelting area) and the coastal provinces to implement the Minamata Convention on Mercury in China. Therefore, Hg emission control and soil remediation are urgently needed in these typical Hg-polluted areas to reduce the health risks of human Hg exposure. Balancing the risks and benefits of fish consumption has become an increasingly essential goal, and scientific advisories on fish consumption are needed to reduce Hg exposure in coastal provinces.

CRediT authorship contribution statement

Bo Wang: Investigation, Methodology, Data curation, Writing - review & editing. **Min Chen:** Investigation, Data curation. **Li Ding:** Investigation, Data curation. **Yuhang Zhao:** Investigation, Data curation. **Yi Man:** Data curation, Writing - review & editing. **Lin Feng:** Data curation, Writing - review & editing. **Ping Li:** Conceptualization, Investigation, Methodology, Data curation, Writing - review & editing. **Leiming Zhang:** Data curation, Writing - review & editing. **Xinbin Feng:** Conceptualization, Methodology, Data curation, Writing - review & editing.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2021.106561.

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