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Home-produced eggs: An important pathway of methylmercury exposure for residents in mercury mining areas, southwest China

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ABSTRACT

In light of the documented elevated concentrations of total mercury (Hg) and methylmercury (MeHg) in poultry originating from Hg-contaminated sites, a knowledge gap persists regarding the levels of Hg found in home-produced eggs (HPEs) and the associated dietary exposure risks in regions affected by Hg mining. To address this knowledge gap, a comprehensive investigation was undertaken with the primary objectives of ascertaining the concentrations of THg and MeHg in HPEs and evaluating the potential hazards associated with the consumption of eggs from the Wanshan Hg mining area in Southwest China. The results showed that THg concentrations in HPEs varied within a range of 10.5–809 ng/g (with a geometric mean (GM) of 64.1 ± 2.7 ng/g), whereas MeHg levels spanned from 1.3 to 291 ng/g (GM, 23.1 ± 3.4 ng/g). Remarkably, in half of all eggs, as well as those collected from regions significantly impacted by mining activities, THg concentrations exceeded the permissible maximum allowable value for fresh eggs (50 ng/g). Consumption of these eggs resulted in increased exposure risks associated with THg and MeHg, with GM values ranging from 0.024 to 0.17 μg/kg BW/day and 0.0089–0.066 μg/kg BW/day, respectively. Notably, the most substantial daily dosage was observed among children aged 2–3 years. The study found that consuming HPEs could result in a significant IQ reduction of 34.0 points for the whole mining area in a year. These findings highlight the potential exposure risk, particularly concerning MeHg, stemming from the consumption of local HPEs by residents in mining areas, thereby warranting serious consideration within the framework of Hg exposure risk assessment in mining locales.

1. Introduction

Mercury (Hg) is a metallic chemical element existing in liquid form at room temperature and possessing a high degree of toxicity. Methylmercury (MeHg), an organic compound, represents a toxic variant of Hg and can be formed through anaerobic microbial methylation (Gilmour et al., 1992; Stein et al., 1996; Selin, 2009). MeHg has the propensity to biomagnify along with the food chain, leading to substantial adverse impacts on the health of both humans and wildlife (Mergler et al., 2007; Scheuhammer et al., 2007; Eagles-Smith et al., 2016). Even at low exposure levels, MeHg exposure during pregnancy can significantly impair fetal cognitive development. Research has demonstrated that a 1 μg/g increase in maternal hair Hg content results in a decrease of 0.18 intelligence quotient (IQ) point in the developing fetus (Axelrad et al., 2007). Given its toxicological profile, the Minamata Convention on Mercury was formulated and entered into force in 2017 (UNEP, 2017), thereby necessitating the urgent implementation of health risk assessments pertaining to Hg-contaminated food and other environmental media worldwide, as dictated by the provisions of this convention.

Home produced eggs (HPEs) constitute a vital component of the diet for both urban and rural residents, owing to their robust nutritional profile, including a high content of proteins, lipids, amino acids, and other beneficial elements like selenium (Iddamalgoda et al., 2001; Ruxton et al., 2010; Aendo et al., 2019; Zhao et al., 2021). Owing to the consumption of cereals, invertebrates, and even small animals from contaminated regions, poultry can accumulate various contaminants,

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attaining elevated concentrations in their blood and tissues, including muscles, liver, and kidney. Consequently, the consumption of HPEs derived from such poultry becomes a noteworthy pathway for exposure to many contaminants, such as perfluoroalkylated substances (Su et al., 2017; Lasters et al., 2022), dioxins and polychlorinated biphenyls (Hoogenboom et al., 2016; Polder et al., 2016), brominated flame retardants (Covaci et al., 2009), and potentially toxic elements (Ji et al., 2006; Wang et al., 2023b), thereby raising significant concerns for human health.

In the context of the Wanshan Hg mining area (WSMM), extensive Hg mining and retorting activities have engendered substantial Hg and MeHg contamination across various environmental matrices, including soil, water, ambient total gaseous Hg, rice, vegetables, and other foodstuffs (Feng et al., 2008; Qiu et al., 2009; Zhang et al., 2010; Tang et al., 2021; Xu et al., 2020a; Xu et al., 2020). In this locale, poultry accumulates Hg through the consumption of cereals, invertebrates, and even soil. As documented by Yin et al. (2017), the mean concentration of MeHg in poultry liver and blood in WSMM reaches as high as 19.6 ng/g and 8.6 ng/g, respectively. Previous investigations have also elucidated the transfer of Hg from parents to eggs, substantiated by the significant positive correlation between Hg content in eggs and maternal tissues, including blood, muscle, liver, and kidney (Ackerman et al., 2017). Moreover, elevated Hg levels, with a substantial proportion of MeHg (approximately 90%) as a fraction of total Hg (MeHg%), have been documented in eggs of both free-ranging and captive birds (Ackerman et al., 2013; Bond and Diamond, 2009; Evers et al., 2003; Scheuhammer et al., 2001). Due to the similar physiological metabolic characteristics and dietary types with birds, this raises concerns regarding increased MeHg levels in HPEs of free-living poultry. Nevertheless, within the typical context of Hg-contaminated sites within Hg mining areas, an important food source, information regarding the occurrence, levels, and associated dietary exposure risks of Hg, particularly MeHg, in HPEs, remains substantially unreported. Consequently, a comprehensive study is necessitated to delineate the concentrations of THg and MeHg in HPEs and to evaluate their potential health risks to humans.

Within the confines of this present study, the typical WSMM region was selected as the study area, and HPEs from free-ranging poultry were collected. The primary objectives included: 1) the investigation of THg and MeHg levels in HPEs within the Hg mining areas; 2) the assessment of daily dietary exposure to THg and MeHg across various age groups within the population, attributable to HPE consumption; and 3) the evaluation of the impact of MeHg exposure on children's cognitive development following egg ingestion. This study aspires to fill the gap in research concerning pollution levels and daily intake values of THg, particularly MeHg, in HPEs originating from Hg mining areas. Additionally, it endeavors to offer recommendations aimed at mitigating the significant risks associated with MeHg exposure among local residents.

2. Materials and methods

2.1. Study area and sample collection

The WSMM (109°07′–23′ E, 27°24′–38′ N) is situated in eastern Guizhou Province, Southwest China. A total of 194 HPEs were meticulously collected from various locations within the Aozhai-Gouxi river watershed (Meizixi (MZX), Supeng (SP), Gouxi (GX), Shenchong (SC), and Daping (DP)), Xiaxi river watershed (Zhongjiapo (ZJP), Xiachangxi (XCX), Baoxi (BX), and Huangcha (HC)), and Gaolouping-Huangdao River watershed (Liukeng (LK), Sanmudong (SMD), and Yangqiao (YQ)), as depicted in Fig. 1.

At each designated sampling site, about 5–10 HPEs, comprising free-range ducks and chickens, were systematically procured and ensured from different hens, resulting in a total of 194 egg samples. To facilitate the classification of Hg distribution across various egg components, 17 eggs were chosen at random to separate egg yolk from egg whites. Furthermore, 30 free-range chicken eggs were collected from Jinping County (JP) in the east of Guizhou Province, serving as a control area devoid of Hg contamination sources. All samples were judiciously arranged within pre-prepared foam egg cartons, transported to the laboratory, and subsequently preserved in a refrigerator at a temperature of 4 °C for subsequent analytical endeavors. Within the laboratory setting, the egg samples were opened carefully via stainless scissors, followed by a process of homogenization. These samples were subsequently transferred into 50 mL centrifuge tubes and preserved at −20 °C until further

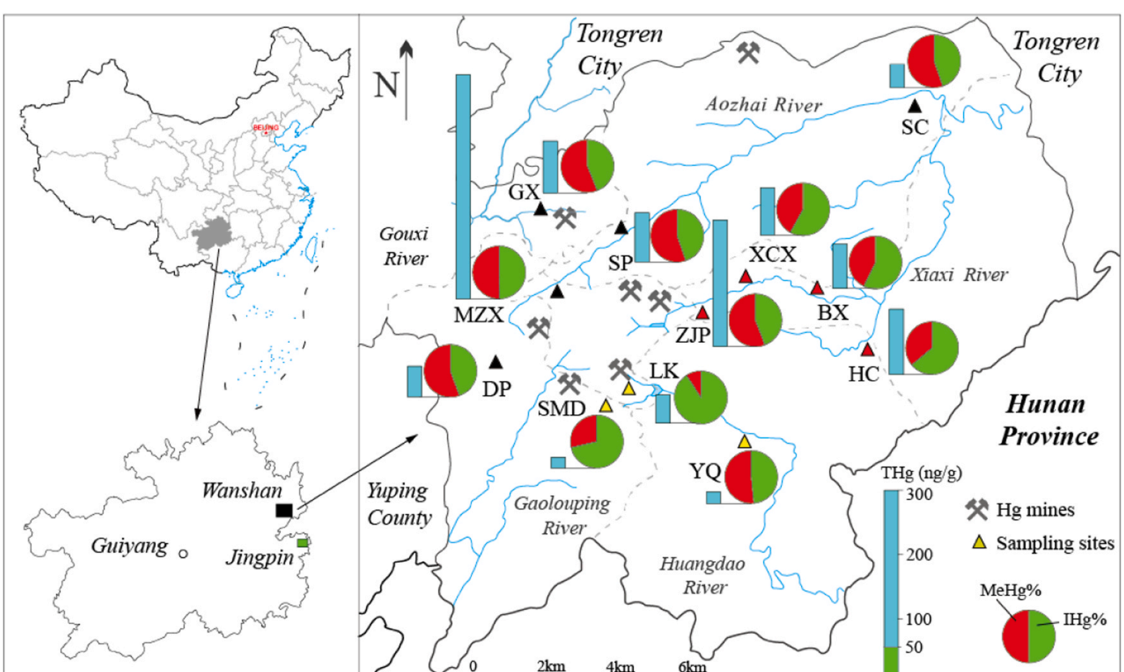

Fig. 1. Study area and sampling sites. Sampling sites colored in black, red, and yellow are situated within the Gouxi-Aozhai River, Xiaxi River, and Gaolouping-Huangdao River watersheds, respectively. The rectangle and pie chart represent the THg concentrations and percentages of MeHg.

analysis.

2.2. Sample analysis

For the analysis of total Hg, approximately 0.2 g of egg samples were weighed and placed in 15 mL centrifuge tubes. These samples were digested with 3 mL of mixed acid (HNO_3: H_2SO_4 = 4:1, v/v) at 95 °C in a water bath for three hours. The digests were subjected to oxidation through BrCl, followed by neutralization with NH_4OH HCl, culminating in their subsequent reduction employing $SnCl_2$. The quantification of Hg concentration within these digests was carried out through cold vapor atomic fluorescence spectrometry (CVAFS; Model III, Brooks Rand Co. Ltd., USA) following EPA Method 1631e (USEPA, 2002), Liang et al. (1996), and Liu et al. (2014). In MeHg analysis, approximately 0.1 g of egg samples was subjected to digestion utilizing a 5 mL solution comprising 25% KOH methanol solvent. Subsequent analysis was executed via gas chromatography cold vapor atomic fluorescence spectrometry (GC–CVAFS; Model III, Brooks Rand Co. Ltd., USA), in accordance with the methodology outlined in method 1630 (USEPA, 1998) and as expounded upon by Liang et al. (1996).

The validation of Hg measurements included the incorporation of method blanks, sample blanks, duplicates (10%), spiked samples (THg and Hg standards), and certified reference material (fish, GBW10029). These measures served to affirm the precision and accuracy of the Hg measurements. Notably, the recoveries of THg and MeHg in fish were found to be 91.3 ± 3.3% (within a range of 86.1–95.1%, n = 5) and 103 ± 10% (ranging from 92.4% to 117%, n = 5), respectively. Similarly, the recoveries associated with spiked THg and MeHg samples were consistent with the ranges of 106.0 ± 3.2% (ranging from 100.9% to 109.8%, n = 7) and 105.5 ± 5.5% (within the range of 96.9–113.0%, n = 5), respectively. The relative standard deviations (RSD) for these measures remained below the 10%.

2.3. Health risk assessment

Within the purview of this study, the Human Health Risk Assessment (HHRA) model, as recommended by USEPA (2023), was employed to measure the health risk posed by Hg originating from egg consumption among the residents of the Wanshan region. The estimated daily intake (*EDI*) of Hg via egg consumption across various age groups was computed utilizing the following equation (Hashemi et al., 2019):

$$EDI = C \times IR / (BW \times 10^3)$$

where *C* is the Hg concentration (ng/g), *IR* denotes the food intake rate (g/day), *EF* represents the frequency of exposure (1 time/day), *ED* indicates the exposure duration (365 days/year), *BW* denotes the mean body weight (kg), and *AT* corresponds to the average exposure time to non-carcinogens, with *AT* calculated as $AT = EF \times ED$. Specific values for *IR* and *BW* corresponding to different age populations are detailed in Table S1.

2.4. Intelligence quotient loss calculation

Based on the previous epidemiologic studies (Crump et al., 1998; Myers et al., 2003; Grandjean et al., 1997), a linear dose-response relationship between maternal intake of MeHg and the decrease in the foetal intelligence quotient (*IQ*) was established by Axelrad et al. (2007), making it possible to estimate the IQ loss points of children caused by MeHg exposure. In this study, an assessment was conducted concerning the adverse effects of MeHg derived from HPE consumption on the *IQ* of children. The methodology for calculating *IQ* point changes, widely employed in populations associated with fish and rice consumption and as reported by Zhang et al. (2018) and Zhang et al. (2021), was adopted. Nonetheless, different with fish consumption population, it is important to note that no specific parameter values are available for populations engaged in rice consumption within inland areas. In this study, the linear fitting of MeHg EDI with MeHg concentration in human hair ($MeHg_{hair} = 22.9 \times EDI_{MeHg}$, as detailed by Li et al., 2015) was tentatively utilized to estimate the hair MeHg concentration in pregnant women. This approach has been adopted in prior works by Wang et al. (2020) and Wang et al. (2021b). The calculation of *IQ* loss points resulting from egg consumption followed these equations:

$$\text{IQ loss} = TN \times \Delta IQ$$

$$\Delta IQ = \gamma \times \Delta M_{p\text{-}hair}$$

$$\Delta M_{p\text{-}hair} = \varphi \times \Delta EDI$$

where *TN* represents the number of annual newborn children in the study area (905; Feng et al., 2020); ΔIQ signifies the total change in *IQ* points; $\Delta M_{p\text{-}hair}$ (μg/g) reflects the hair Hg content in pregnant women; γ (points per 1 μg/g of maternal hair Hg) denotes the decrement in *IQ* points for each μg/g of maternal hair Hg (0.18, as reported by Axelrad et al., 2007); $\Delta M_{p\text{-}hair}$ (μg/g) represents the alteration in Hg concentration within the hair of pregnant women; ΔM_b designates the alteration in Hg concentration within the blood of pregnant women; ΔEDI embodies the alteration in daily Hg intake attributable to HPE consumption; and φ (μg/g per μg/kg/d) signifies the slope characterizing the relationship between $\Delta M_{p\text{-}hair}$ and ΔEDI (22.9, Li et al., 2015).

2.5. Statistical analysis

Descriptive statistical analyses, linear fitting, and significance assessments were performed through Origin 2021 (Origin Lab, USA). Comparisons between the data were made using the one-way ANOVA. To evaluate Hg exposure risk levels, the Monte Carlo method was applied with Crystal Ball (Oracle, USA) at 10,000 simulated iterations.

3. Results and discussion

3.1. THg and MeHg concentrations

In Table 1, the concentrations of THg and MeHg in HPEs collected from WSMM ranged from 10.5–809 ng/g (64.1 ± 2.7 ng/g, Geometric mean, GM) and 1.3–291 ng/g (23.1 ± 3.4 ng/g, GM), respectively. As depicted in Fig. 2, THg concentrations in 52% of HPE samples exceeded the maximum allowable value of 50 ng/g in eggs (NHC, 2022). The maximum THg value (809 ng/g) was approximately 16 times higher than the limit. Moreover, MeHg levels in HPEs ranged from 1.3–291 ng/g, with an average of 23.1 ng/g, surpassing the levels found in rice grown in WSMM (mean: 8.5 ng/g, ranging from 1.9 to 27.6 ng/g). Based on the average values, duck eggs (n = 50) exhibited relatively higher THg and MeHg concentrations than chicken eggs. This difference could be attributed to the special dietary composition of ducks, allowing them to prey on small aquatic invertebrates in rivers or flooded rice paddies. This finding aligns with the results of Yin et al. (2017), who observed similar trends in the tissues and organs of ducks and chickens. The distribution of THg and MeHg concentrations in eggs generally follows a log-normal distribution, as shown in Fig. 2. The ratio of MeHg to THg (MeHg%) ranged from 3.2% to 98%, with an average value of 44 ± 22%, which is much lower than the 89–95% found in 171 caged chicken eggs and the 82–111% in 217 wild bird eggs reviewed by Ackerman et al. (2013), which may relate to the feed types (e.g., cage feeding and free living) and dietary types (e.g., piscivorous, insectivorous, omnivorous, and granivorous).

In this study, the elevated levels of THg and MeHg found in HPEs from WSMM were higher compared to duck eggs from the Qingzhen area with Hg contamination due to chemical plants (THg: 46.9 ± 7.5 ng/g, MeHg: 19.3 ± 7.7 ng/g; Cheng et al., 2013), which may relate to the much lower Hg concentration in rice from Qingzhen (Horvat et al., 2003; Cheng et al., 2013) compared to Wanshan (Zhang et al., 2010; Xu

Table 1
THg and MeHg concentrations (wet weight) in home-produced eggs from Hg mining areas and other studies.

Watersheds	Sites	No.	THg ng/g		MeHg ng/g		MeHg%	Exceeding rates	Sources
			GM ± SD	Range	GM ± SD	Range	%	%	
Xiaxi- river, Wanshan, China	ZJP	16	200 ± 86.9	53.4 ~ 352	105 ± 59.6	35.4 ~ 290	55.7 ± 22.1	100	This Study
	XCX	7	76.3 ± 29.6	31.8 ~ 132	32.6 ± 19.4	11.8 ~ 63.8	41.8 ± 18.2	85.8	
	BX	37	70.6 ± 48.6	18.6 ~ 184	24.6 ± 12.4	3.29 ~ 68.5	42.7 ± 22.1	47.2	
	HC	10	104 ± 94.9	23.2 ~ 327	32.7 ± 12.9	17.6 ~ 58.4	36.1 ± 19.9	75	
Aozhai- Gouxi river, Wanshan, China	MZX	21	357 ± 165	294 ~ 809	141 ± 45.4	92.2 ~ 241	50.2 ± 28.5	100	
	SP	17	76.7 ± 29.2	36.8 ~ 136	49.6 ± 19.2	28.9 ~ 78.6	63.0 ± 13.7	77.8	
	GX	8	81.4 ± 37.0	25.6 ~ 125	42.8 ± 33.3	25.6 ~ 125	55.9 ± 22.2	80	
	SC	12	37.9 ± 23.6	11.9 ~ 97.3	16.1 ± 6.00	7.68 ~ 28.4	50.1 ± 20.3	25	
	DP	10	49.5 ± 12.7	31.7 ~ 65.9	27.9 ± 8.05	17.4 ~ 38.1	55.3 ± 13.1	50	
Wanshan-Huangdao river, Wanshan, China	SMD	17	16.7 ± 3.6	10.5 ~ 24.0	4.72 ± 1.71	2.28 ~ 8.03	28.5 ± 9.8	0	
	LK	14	45.1 ± 16.2	22.3 ~ 77.1	4.14 ± 3.22	1.33 ~ 12.7	9.1 ± 5.3	42.9	
	YQ	11	17.9 ± 2.8	14.7 ~ 23.4	9.19 ± 2.83	4.82 ~ 13.2	51.4 ± 15.5	0	
Wanshan, China	Whole	177	108 ± 127	10.5 ~ 809	44.7 ± 52.3	1.33 ~ 290	43.9 ± 22.2	52	
	Chicken eggs	127	99.2 ± 138	10.5 ~ 809	43.5 ± 52.8	1.50 ~ 241	47.4 ± 22.7	50.7	
	Duck eggs	50	133 ± 83.7	23.1 ~ 352	51.2 ± 50.2	1.33 ~ 290	36.7 ± 25.8	46.0	
Jinping, China	Chicken eggs	30	23.0 ± 7.8	14.7 ~ 37.0	3.33 ± 1.40	1.14 ~ 6.11	16.6 ± 8.2	0	
Wawu, Wanshan, Guizhou	Chicken eggs	5	61.7 ± 41.0	27.4 ~ 108	—	—	—	—	Zhan et al. (2017)
Hezhang, Guizhou	Chicken eggs	—	3.0 ± 1.1 *	1.6 ~ 5.7	0.14 ± 0.44 *	0.02 ~ 1.7	—	—	Wang et al. (2023a)
Guiyang, Guizhou	Chicken eggs	—	0.37 ± 0.43	0.18 ~ 2.1	—	—	—	—	Wang et al. (2023a)
Qingzhen, Guizhou	Duck eggs	—	46.9 ± 7.5	—	19.3 ± 7.7	—	—	—	Cheng et al. (2013)
Shanghai rural area	Duck eggs	—	11.5 ± 5.3	—	5.9 ± 2.6	—	—	—	Cheng et al. (2013)
China	Poultry eggs	5194	7.6	—	—	—	—	—	Qing et al. (2022)

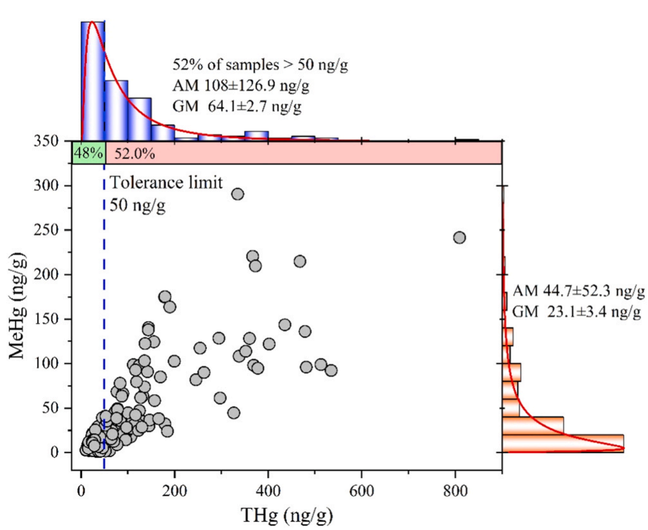

Fig. 2. Concentrations of THg and MeHg in HPEs from the Hg mining area.

et al., 2020b; Wang et al., 2021). However, the THg and MeHg levels in HPEs from the control area of Jinping were much lower, at 23.3 ± 8.7 ng/g (arithmetic mean, AM) and 3.5 ± 1.4 ng/g (AM), respectively. Furthermore, in the non-Hg mining area, duck eggs from the Shanghai rural area had average THg and MeHg levels of 11.5 ± 5.3 ng/g and 5.9 ± 2.6 ng/g, respectively (Cheng et al., 2013). Another study reported an average THg concentration of 8 ng/g in eggs from 295,688 food samples in China (Qing et al., 2022). The higher levels of Hg observed in the HPEs in this study can be attributed to the consumption of Hg-rich foods by poultry, such as corn, rice, and even invertebrates, which can accumulate Hg from polluted soil, water, and atmosphere (Hashemi et al., 2019; Wang et al., 2018).

3.2. Characterization of Hg distribution in eggs

3.2.1. Spatial Hg distribution in eggs

The spatial distribution of THg and MeHg in HPEs on a regional scale is depicted in Fig. 1. Within the Xiaxi River watershed, eggs from ZJP exhibited the highest levels of THg and MeHg, with 200 ± 86.9 and 105 ± 59.6 ng/g, respectively. This was followed by HC (104 ± 94.9 ng/g, THg; 32.7 ± 12.9 ng/g, MeHg), XCX (72.8 ± 39.1 ng/g, THg; 22.5 ± 13.6 ng/g, MeHg), and BX) (70.6 ± 48.6 ng/g, THg; 24.6 ± 12.4 ng/g, MeHg). In the Aozhai/Gouxi river watershed, eggs from MZX exhibited the highest THg and MeHg levels of 357 ± 165.2 and 141 ± 45.4 ng/g, respectively, which were also the highest values observed throughout the entire area. In SC and YQ, located downstream of the Aozhai River and in the midstream of the Huangdao River, respectively, the collected eggs showed lower values of THg (37.9 ± 23.6 ng/g and 17.9 ± 2.8 ng/g) and MeHg (16.1 ± 6.0 ng/g and 9.2 ± 2.8 ng/g) compared to other sampling sites. This is because the food source (cereals and fodder) of the chickens and ducks that produced these eggs are purchased from non-contaminated sources of Hg. In other sites located between the Hg mines and the farthest sites, the free-range HPEs exhibited moderate concentrations of THg and MeHg, as shown in Fig. 1.

Among these sites, large-scale Hg mining activities were conducted in the upstream areas of MZX, ZJP, and GX, resulting in serious Hg contamination of local free-range poultry. The Hg concentrations in all HPEs (100%) collected from MZX and ZJP, which are directly affected by Hg mining, exceeded the maximum allowable value of Hg concentration in eggs (50 ng/g). Similarly, in other heavily Hg-contaminated sites, such as XCX and GX, 85.8% and 80% of egg samples contained Hg concentrations exceeding the maximum allowable value, respectively. Therefore, more attention should be paid to these seriously contaminated sites. However, in SC, which is the farthest site from WSMM, a higher Hg concentration of 37.9 ± 23.6 ng/g was also observed compared to the control site, and approximately 25% of the eggs exceeded the threshold. Therefore, the results indicate that the Hg concentration in HPEs can serve as an indicator to assess the degree of Hg contamination in the surrounding environment.

3.2.2. Hg distribution in different egg parts

The THg and MeHg concentrations in egg whites and yolks are presented in Fig. 3(a). The results showed that egg whites had significantly higher concentrations of THg (265.4 ± 90.1 ng/g) and MeHg (35.3 ± 14.0 ng/g) compared to the yolk, which had concentrations of THg (67.4 ± 27.3 ng/g) and MeHg (27.4 ± 8.2 ng/g), respectively. These findings align with previous research by Magat and Sell (1979), who also reported higher THg concentrations in egg whites compared to other parts of the egg.

Fig. 3(b) displays the net content (mass × concentration) of Hg, revealing that the egg yolk contained a lower amount of THg (726 ± 260 ng) but a higher amount of MeHg (303 ± 112 ng) compared to the egg white (THg 933 ± 652 ng; MeHg 125 ± 89 ng). This indicates a clear difference in inorganic Hg (IHg, calculated as IHg = THg - MeHg) concentrations between the egg white and yolk, as shown in Fig. 3(a). Thus, the elevated concentration of IHg in egg white is the reason why THg in egg white showed much higher concentrations and net amounts of THg but lower concentrations and net amounts of MeHg compared to egg yolk. Egg white is rich in protein, mainly ovalbumin (Li-Chan et al., 1995, 2007), which facilitates the combination of Hg, resulting in the elevated concentration and net amount of THg and IHg in egg white compared to egg yolk.

3.3. Health risk assessment

The estimated daily intakes (EDIs) of THg and MeHg for residents through the consumption of HPEs are presented in Fig. 4. In WSMM, the THg EDI and MeHg EDI via egg consumption ranged from 0.024 to 0.17 μg/kg BW/day and 0.0089–0.066 μg/kg BW/day, respectively. These values were significantly higher compared to the control site in JP (control site, 0.0081–0.059 μg/kg BW/day, THg; 0.0012–0.0088 μg/kg BW/day, MeHg). As shown in Fig. 4, the P95 values, which indicate a high risk of exposure for consumers, exceeded 0.3 μg/kg BW/day for adolescents under 18 years old, ranging from 0.34 to 1.1 μg/kg BW/day. In addition, the EDIs of THg and MeHg tended to decrease with age. Specifically, among children aged 2–3 years, the EDIs for this age group showed the highest p95 values of 1.1 μg/kg BW/day for THg and 0.51 μg/kg BW/day for MeHg.

These values exceeded the threshold by 4–5 times, surpassing the provisional tolerable weekly intakes (PTWIs) for THg and MeHg as recommended by JECFA (2010). Consequently, the consumption of HPEs may present Hg exposure risks for both adults and children in WSMM. This risk is especially significant for toddlers aged 2–3 years. It is worth noting that children are more vulnerable to the health effects of Hg due to their immature organs (Järup, 2003). Hence, special attention should be given to the Hg exposure risk through HPE consumption. In earlier research, the overall EDIs, also known as PDI, of MeHg through dietary routes were documented as 0.096 μg/kg BW/day (Zhang et al., 2010) and 0.034 μg/kg BW/day (Xu et al., 2020). Therefore, the EDI via the consumption of HPEs (0.0089–0.010 μg/kg BW/day) by over 18-year-old adults can contribute to approximately 10% and 30% of the total EDIs of MeHg. Moreover, in heavily Hg-contaminated areas (within 4 km) (Xu et al., 2020), such as MZX and ZJP, where the average MeHg concentrations in HPEs are three times and twice as high as the average levels in the entire area, consuming eggs would have a higher contribution rate to the total EDI of MeHg. Additionally, considering that rice consumption is the major exposure pathway to MeHg in inland Hg mining areas, egg consumption accounts for nearly one-third of the MeHg EDI compared to rice consumption.

In comparison to the average MeHg daily intake of children via rice consumption in WSMM (0.052 μg/kg BW/day; Du et al., 2016), the GM

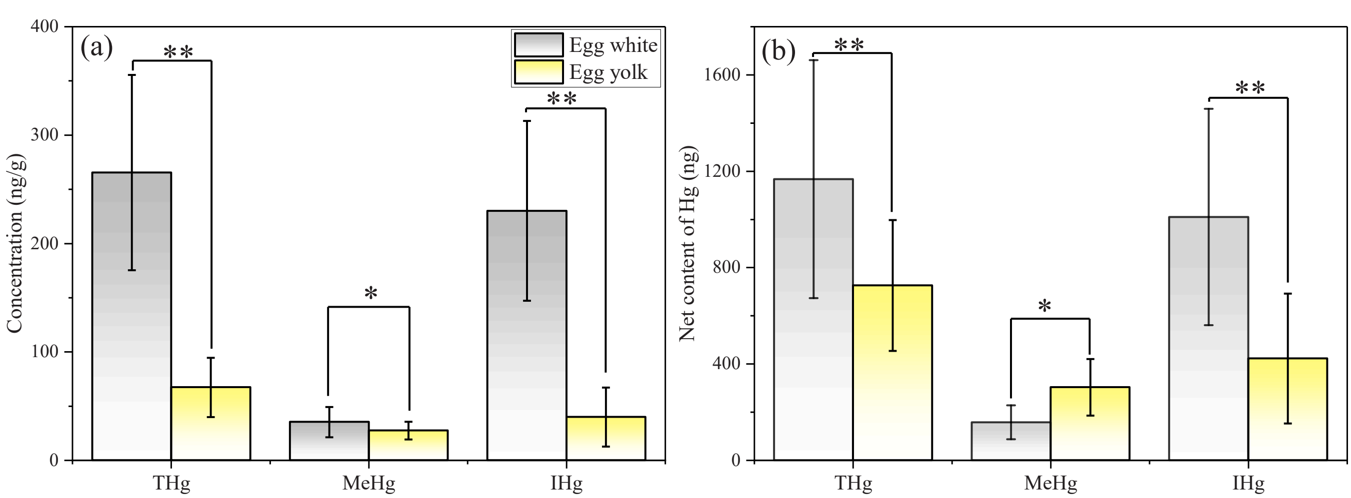

Fig. 3. THg, MeHg, and IHg concentrations (a) and net content (b) in egg white (white) and yolk (yellow) from HPEs collected in Hg mining areas. The net content of Hg is calculated as net content= mass × concentration. ** denotes $p < 0.01$, * denotes $p < 0.05$.

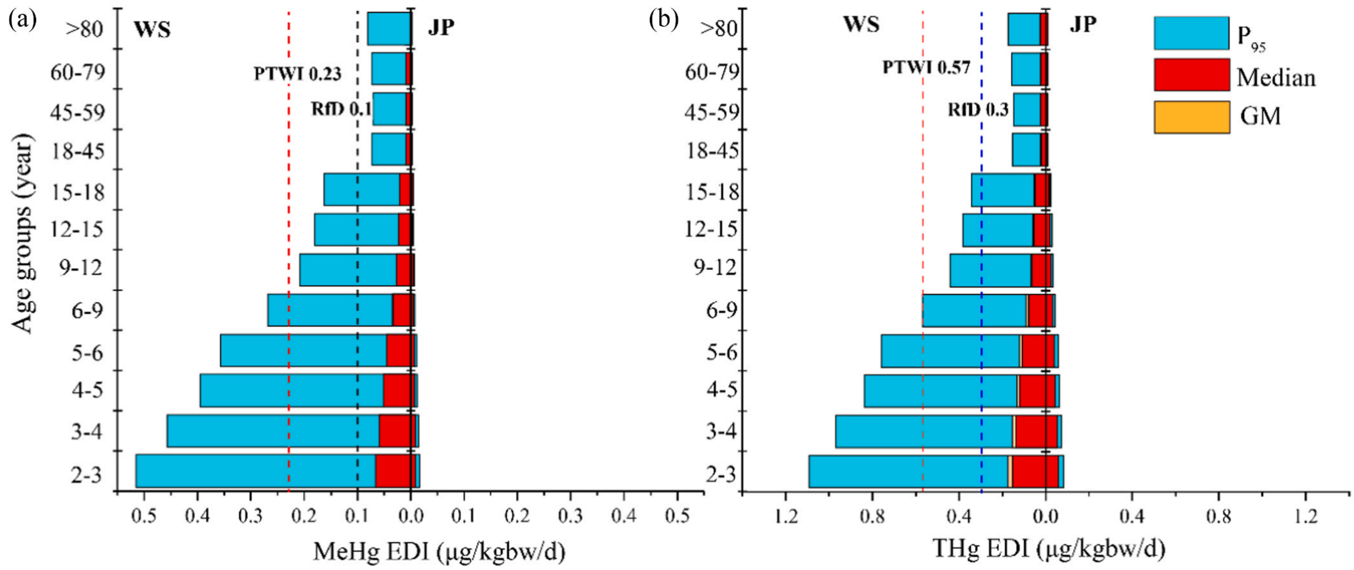

Fig. 4. Estimated EDIs of MeHg (a) and THg (b) in various age groups through the consumption of HPEs from Hg mining areas (WS, left part) and control site (JP, right part).

of MeHg EDI via egg consumption for children aged 2–3 years and 3–4 years showed comparable values of 0.066 and 0.058 μg/kg BW/day, respectively. Previous studies have reported that MeHg EDI via fish consumption by residents in Europe and North America is 0.05 μg/kg BW/day and 0.02 μg/kg BW/day, respectively (Mahaffey et al., 2004; Mangerud, 2005). Therefore, the MeHg EDI through egg consumption cannot be considered negligible for both adults (GM: 0.009–0.010 μg/kg BW/day) and children (GM: 0.021–0.066 μg/kg BW/day). Although the consumption of HPEs was not considered in previous studies, our findings show that free-range egg consumption is an important MeHg exposure pathway for both adults and children, which should not be ignored.

3.4. IQ loss caused by egg consumption

The loss of IQ points attributed to egg consumption in each sampling site and across the entire WSMM region is depicted in Fig. 5. Across the entire study area, the average IQ point reduction for each newborn child is 0.07 ± 0.09 (0.04, GM; 0.23, 95th percentile, p95). Considering the recent number of newborns in Wanshan (905, Feng et al., 2020), this translates to an annual average IQ point reduction of 65.8 ± 77.0 (AM; 34.0, GM; 211.2, p95) due to the consumption of local HPEs. Among the 11 sampling sites, MZX from Aozhai-Gouxi River Watershed and ZJP from the Xiaxi River watershed recorded the highest IQ point reductions, ranging from 0.15 to 0.39 (average: 0.23 ± 0.08) and 0.06–0.47 (average: 0.17 ± 0.10), respectively. Conversely, LK, SMD, and YQ from the Huangdao River watershed, SC from the Gouxi-Aozhai River watershed, and HC from the Xiaxi River watershed exhibited the lowest IQ point reductions, indicating a lower risk of intellectual impairment associated with egg consumption in these areas.

When compared to other studies, it is evident that the Hg concentration in free-range eggs is significantly higher than that in rice and fish. However, the IQ point reduction per child caused by rice and fish consumption in Wanshan (0.62, Wang et al., 2021a) is considerably higher than that resulting from HPE consumption. This discrepancy may be due to variations in intake rates of these foods, even with elevated MeHg concentrations. In this study, the IQ point reduction (0.07 ± 0.09, Arithmetic mean, AM) through HPE consumption is similar or slightly lower than the reported average IQ point reductions near coal-fired power plants (0.07), gold mining areas (0.10), zinc smelter areas

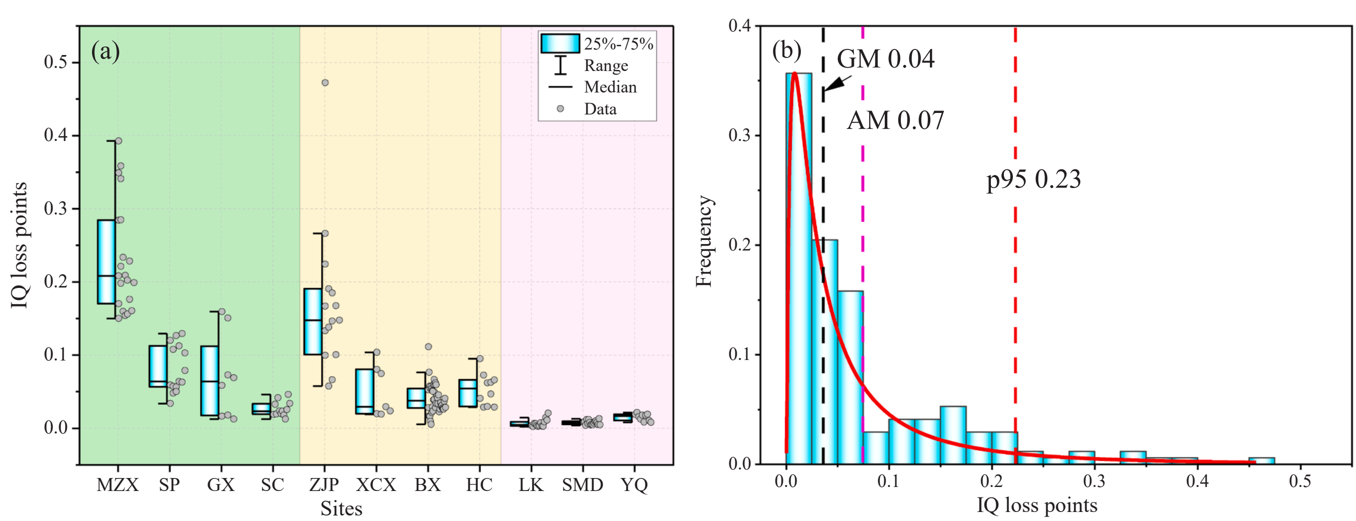

Fig. 5. IQ loss points due to the consumption of free-range HPEs at each sampling site (a) and across the entire area (b).

(0.16), as well as in the inland city of Wuhan (0.08), and coastal cities of Qingdao (0.03), Xiamen (0.08), and Zhoushan (0.15) by Wang et al. (2021a). Given that rice consumption is a major pathway for MeHg exposure, especially in inland mining areas (Zhang et al., 2010), the IQ point reduction caused by HPE consumption is much lower than that caused by rice and fish consumption in Hg mining areas. However, it is higher than that in other rice-consuming countries such as Bangladesh (0.038; Wang et al., 2020) and Nepal (0.018; Wang et al., 2021b). Thus, these findings indicated that intellectual impairment resulting from the consumption of free-range poultry eggs is still a noteworthy concern for local residents, which has been previously overlooked. We did not consider the effect of other elements in this study. For example, selenium can mitigate potential toxic effects of MeHg (Horvat et al., 2003). Therefore, the actual intellectual impairment may be lower than that we estimated.

3.5. Future perspectives and limitations

To the best of our knowledge, this is the first study to comprehensively investigate the levels, dietary exposure risks, and associated adverse effects of Hg, particularly MeHg, in free-range HPEs in Hg mining areas. The variability in MeHg% in HPEs, with some data reaching levels comparable to those found in bird eggs, highlights the significance of HPEs consumption as a MeHg exposure risk, similar to rice consumption which is a major exposure pathway for MeHg in residents of inland mining areas and is known to cause significant intellectual impairment. Egg consumption should be recognized as a crucial MeHg source for residents in Hg mining areas and other Hg-contaminated regions. However, this pathway has been largely ignored in previous studies of MeHg sources in the human body using exposure dose (Zhang et al., 2010; Xu et al., 2020b; Wang et al., 2021b) and Hg isotopes (Du et al., 2018). This study sheds light on the need to consider egg consumption when evaluating MeHg exposure risks in Hg mining areas and other Hg-contaminated sites, providing a more comprehensive understanding of overall MeHg exposure risks and identifying dietary sources of MeHg for residents in Hg-contaminated areas, beyond rice and fish/aquatic products.

Although this study extensively investigated Hg levels and associated exposure risks, other potential factors such as variations among different poultry species (e.g., various species of chicken and ducks), egg laying times (Brasso et al., 2010), life stages or feeding times (e.g., young or old female poultry), and feeding bait or patterns (e.g., natural feeding in the wild or manual feeding with other cereals, Brasso et al., 2012; Aendo et al., 2018) should be considered. Confirming the consistency of these factors during fieldwork is often challenging. Moreover, due to the lack of a questionnaire related to the exposure factors including body weight, daily intake, and other specific parameters, as well as the difference nutrients in fish products (Tilami and Sampels, 2018), rice (Rohman et al., 2014), and eggs (Ruxton et al., 2010), certain uncertainty would affect the final estimates of EDIs and associated IQ point reductions. Therefore, future studies should implement a more detailed sampling design to obtain more accurate Hg levels in HPEs and assess Hg exposure risks.

4. Conclusions

This study reports elevated levels of THg and MeHg in free-range HPEs in Hg mining areas in Southwest China. Nearly half of the egg samples showed higher Hg concentrations than the national maximum allowable value for eggs. Notably, THg levels in nearly all HPEs from areas directly affected by Hg mining exceeded this threshold. Egg whites exhibited significantly higher THg concentrations and net Hg content compared to yolks. However, the levels of MeHg and net MeHg content in egg whites were comparable to or even lower than those in yolks, primarily due to the elevated levels of IHg in egg whites. This study reveals that HPEs consumption poses a MeHg exposure risk comparable to rice consumption and can lead to significant intellectual impairment in newborn children in Hg mining areas. The consumption of local free-range HPEs is a vital pathway for MeHg exposure among residents in Hg mining areas, particularly those people who consume more than average amount of eggs and pregnant women and children (especially those aged 2–3 years old) who are more susceptible to the effects of MeHg. Given this substantial MeHg exposure risk through HPE consumption, it is essential to clarify updated comprehensive MeHg exposure risks and identify dietary sources of MeHg for residents in Hg-contaminated areas rather than restricting assessments to rice and fish/aquatic products.

CRediT authorship contribution statement

Zhidong Xu: Writing and revise draft, Conceptualization, Formal analysis, Funding acquisition. **Yuhua Yang**: Writing draft, Investigation. **Jun Li**: Editing and Revise draft. **Na Yang**: Methodology. **Qinghai Zhang**: Reviewing and editing. **Guangle Qiu**: Reviewing and editing. **Qinhui Lu**: Writing-reviewing and editing, Supervision, Funding acquisition.

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.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.ecoenv.2023.115678.

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