Exposure and Health
https://doi.org/10.1007/s12403-024-00630-y

ORIGINAL PAPER

Influence of Selenium Supplementation on Mercury Levels in hair of Metropolitan Residents in China: Spatial Distribution, Impact Factors, and Antagonism

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Received: 3 September 2023 / Revised: 21 January 2024 / Accepted: 29 January 2024

Abstract
Selenium (Se) has an antagonistic effect on the toxicity of mercury (Hg). This study aimed to investigate the influence of Se supplementation on Hg levels in long-term chronically exposed metropolitan residents in China. The levels of Se and Hg in human hair samples from 1373 metropolitan residents in China were analyzed and differences in sex, age, and region were evaluated. Higher concentrations of Hg were found in the hair of males, 60–74 years of age, and South and East Chinese residents. Participants with Se supplementation exhibited statistical lower hair Hg levels ($p < 0.01$) compared to those without additional Se supplementation. The Se:Hg molar ratio was used to evaluate the detoxification ability of Se. The proportion of Se:Hg molar ratios of < 1 were 25.40% and 3.83% for participants belong to G1 (without additional Se supplementation) and G5 (Se supplementation for more than 24 months). These results indicate that long-term Se supplementation may enhance the body's ability to bind to or detoxify Hg, leading to a lower proportion of participants with imbalanced Se:Hg ratios. It is essential to consider targeted Se supplementation or dietary adjustments to optimize the Se:Hg balance and mitigate the risks associated with Hg exposure in Chinese residents.

Keywords Mercury · Selenium · Human hair · Se:Hg molar ratio · Antagonism effect

Introduction

Mercury (Hg) is a well-known hazardous substance that poses serious health risks to various organs of the human body (Driscoll et al. 2013; Zhang and Wong 2007). Human exposure to Hg occurs primarily via three routes: respiratory inhalation, dietary intake (water, fish, rice and other food items), and dermal contact (Sundseth et al. 2017; Zhang et al. 2010, 2014). Exposure to Hg, especially at high concentrations, can adversely affect the kidneys, lungs, nervous tissues, and reproductive system (Gonzalez et al. 2019; Ha et al. 2017; Henriques et al. 2019). Although acute high exposure to Hg can occur in specific occupational or accidental situations, the general population is more commonly exposed to low levels of Hg over an extended period than to acute high levels. Prolonged low-level exposure to Hg may not result in immediate or discernible symptoms but can have long-term health implications. Previous investigations have been conducted to assess the potential health effects of Hg exposure on workers or individuals living in heavily polluted environments (Gonzalez et al. 2019; Jia et al. 2018; Li et al. 2020); however, less attention has been paid to long-term chronic Hg-exposed metropolitan residents, especially in highly populated metropolitan cities in China.

Selenium (Se) is an essential trace element and antioxidant in humans and animals (Yang et al. 2022). It plays a vital role in the molecular mechanisms of Hg toxicity by being incorporated into certain antioxidant selenoenzymes such as glutathione peroxidase and thioredoxin reductase (Khan and Wang 2009). Many studies have confirmed that Se can counteract the toxicity of Hg (referred to as "Hg-Se antagonism") by forming less bioavailable Hg-Se particles, which are less likely to be absorbed and distributed in tissues and organs (Cabanero et al. 2005; Kalisinska et al. 2017; Tomza-Marciniak et al. 2023; Wang and Wang 2017).

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To comprehensively evaluate the potential risk of Hg toxicity, the Se:Hg molar ratio has been used as a biomarker to assess the detoxification ability of the human body. An Se/Hg molar ratio < 1 indicates enhanced Hg toxicity, whereas a Se/Hg molar ratio > 1 suggests possible detoxification (McCormack et al. 2020; Zheng et al. 2021). China has been recognized as one of the countries with Se deficiency in certain regions (Li et al. 2014). The World Health Organization (WHO) has established a recommended daily intake (RDI) of Se for healthy adults at 55 µg/day. According to a recent study by Dinh et al. (2018), a significant proportion of Chinese residents (39–61%) have a daily Se intake below the RDI value (55 µg/day). It is important to strengthen dietary Se supplementation and implement appropriate strategies to improve the Se status and the overall health of Chinese residents.

Human hair is widely used as an important biomarker in population-based studies to estimate environmental pollutant exposure and its potential health implications (Manceau et al. 2016; Wang et al. 2021b). Compared to other human biomarkers, such as blood, serum, and urine, scalp hair research offers the advantages of non-invasive and convenient collection, easy transport, simple storage, and long-term retrospective analysis (Packull-McCormick et al. 2022; Wang et al. 2009). Additionally, hair collection can be performed without any invasive procedures, making it a more comfortable experience for vulnerable groups, such as pregnant and parturient women, new-born infants, and senior citizens (Esplugas et al. 2019; Harley et al. 2019; Li et al. 2021; Llop et al. 2014). Most of the hair Hg is reported to be methylmercury (MeHg), accounting for more than 85% of total Hg (Dolbec et al. 2001). Moreover, owing to the slow growth rate of hair, it can provide a historical record of environmental pollutant exposure over a specific time window or long term (LeBeau et al. 2011).

Therefore, this study aimed to (a) investigate the levels and spatial distributions of hair Se and Hg levels in metropolitan residents in China, (b) analyze the effects of sex and age on hair Se and Hg levels, and (c) evaluate the detoxification potential of Se supplementation using Se:Hg molar ratios.

Materials and Methods

Participants and Ethical Considerations

We performed a cross-sectional study to investigate the influence of Se supplementation on Hg levels in hair of metropolitan residents in China. From July 2021 to May 2023, participants were recruited using convenience sampling from 16 provinces/municipalities in Central and Eastern China (Fig. 1). In this study, a total of 2,507 volunteers were initially recruited. Out of these volunteers, 2,125 agreed to participate in the investigation. The participants were given different types of questionnaires based on their recruitment method. For volunteers recruited online, they were provided with an electronic questionnaire that they could access and complete electronically. On the other hand, volunteers recruited offline were given paper copies of the questionnaire through local staff members. In total, 2,018 individuals successfully completed the questionnaire. Information in the questionnaire included sex, age, height, weight, address, hair-dyeing habits, and Se supplementation (Table S1). The questionnaires filled in by the participants were used to collect information regarding the duration and amount of Se supplementation. Due to several factors such as problems with delivery and manual errors, only 1,503 hair samples were ultimately received. These hair samples went through a screening process, which led to the identification of 1,373 residents who met the established standard criteria (Fig. S1).

The inclusion criteria consisted of three factors: (1) live in the local area for more than two years; (2) sign informed consent; and (3) some participants voluntarily consuming Se nutritional supplements. A total of 932 participants voluntarily supplemented with 1–3 packets (50 µg Se/packet) of Se-enriched supplements (e.g., corn flour, probiotics, and yeast) daily. The daily Se supplement amount for these participants ranged between 50 and 200 µg/d, as recommended by the Chinese Nutrition Association. This suggests that the Se supplemental dose for participants in this study is safe. On the other hand, the exclusion criteria were as follows: (1) pregnant women and those in the lactation period; (2) individuals with a history of exposure to related heavy metals; (3) individuals with hair permed and dyed in recent one year or using selenium-containing shampoo.

To ensure all participants fully understood the research purpose and characteristic, they received an explanation containing detailed information about the study objectives, procedures, potential risks, benefits, confidentiality protocols, and any other relevant information. Informed consent was obtained from all individual participants included in the study (Fig. S2). For children aged 2–18 years, written informed consent was obtained from their parents. This study was approved by the Medical Ethics Committee of Jiangnan University (approval number: JNU20211217IRB06).

Hair Sample Collection

To minimize potential contamination or interference from external factors, participants were instructed to wash their hair at home using a neutral, mild, and non-medical shampoo 1–2 days prior to collecting hair samples. In addition, hair samples were cut from the occipital region of the head

Fig. 1 Sampling cities in central and eastern regions of China

close to the scalp, using pre-cleaned stainless-steel scissors. Since hair grows at an average rate of approximately 1 cm per month (Harley et al. 2019), hair samples with a length of 3–5 cm allow the analysis of the previous 3–5 months of exposure. The approximate amount of hair sample collected from each participant for the determination of Se and Hg is approximately 1.0–2.0 g. After collecting samples from the participants, each sample is assigned a unique number for identification purposes. The collected hair samples were cut in small pieces samples and placed in the corresponding numbered polyethylene bags, quickly transported to the laboratory, and stored in a dry location at room temperature until further analysis.

Analytical Methods

The experimental procedure is illustrated in Fig. 2, which can be divided into the determination of Se and total Hg concentrations.

Se Determination

Dried hair samples (0.15–0.20 g) were digested by eight milliliter of nitric acid–perchloric acid mixed acid (4:1, v/v) using an automatic graphite digestion system (Gdana Inc., China). The digestion program involved heating the sample to specific temperatures for different durations (ramp to 120 °C in 22 min, hold for 80 min, ramp to 180 °C in 12 min, and hold for 32 min) to ensure complete digestion of the sample and absence of any residue in the solution. The reduction of Se from its hexavalent form (Se VI) to its tetravalent form (Se IV) was conducted using 4 mL of hydrochloric acid after the digests were cooled to room temperature. After digestion and reduction, the digests were diluted to 25 ml using Milli-Q water. Finally, the Se concentration was determined using an atomic fluorescence spectrometer with a detection limit of 0.32 μg/L (AFS-8220, Jitian Inc., China).

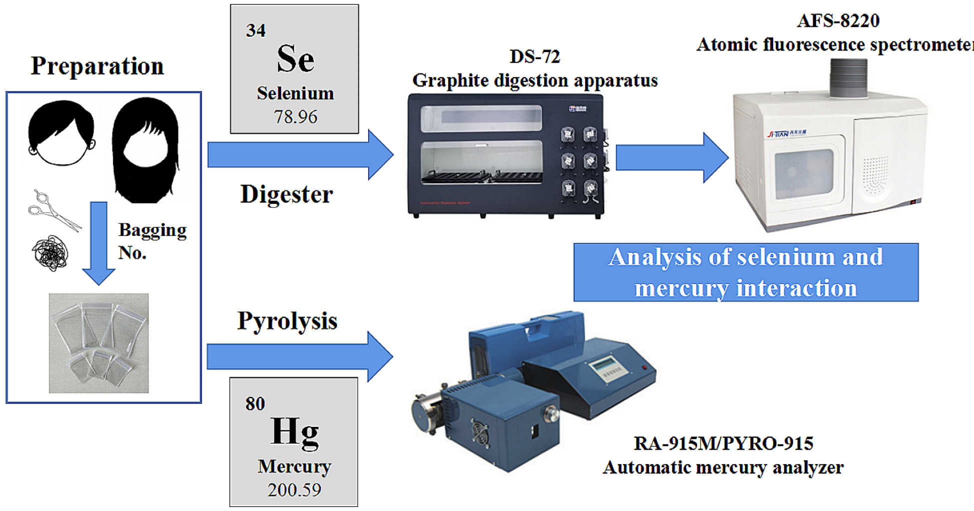

Fig. 2 Experimental flow chart of Se and Hg analysis in hair samples

Total Hg Determination

The total Hg concentration in each hair sample was determined directly using a cold vapor atomic absorption spectrophotometer following U.S. Environmental Protection Agency (EPA) method 7473, with a detection limit of 0.05 µg/kg. The combination of a pyrolysis furnace, amalgamation, and cold atomic absorption allows for a streamlined process from drying decomposition to analysis. Approximately 0.05–0.10 g of the dried hair samples were weighed into a quartz boat and introduced in an automatic Hg analyzer (RA-915 M/PYRO-915, Lumex Instruments, Canada).

Quality Assurance and Quality Control

Quality assurance and control consisted of method blanks, certified reference materials, sample duplicates, and matrix spikes. Blanks were prepared for each batch of hair samples. The measured level of Se in the certified reference material (GBW07601a(GSH-1a), Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences, China) was 0.58 ± 0.01 mg/kg ($n = 6$), which agreed with the certified value of 0.58 mg/kg. The measured Hg concentration was 0.67 ± 0.04 mg/kg ($n = 10$), which was comparable to the value of 0.67 mg/kg in the certified reference material. One hair sample was randomly selected for three parallel tests for every ten samples analyzed. The corresponding relative standard deviation values of duplicates sample were $\leq 10\%$. Analyte recoveries from the spiked hair samples ranged from 95 to 102% for Se and from 90 to 110% for Hg.

Statistical Analysis

Statistical analyses were performed using the IBM SPSS statistics software (version 22.0; IBM Inc., Chicago, IL, USA). Descriptive statistics, including the geometric mean (GM), geometric standard deviation (GSD), minimum, maximum, 95% confidence interval (95% CI), and the 10th, 25th, 50th, 75th, 90th percentile values, were used to summarize the characteristics of Se and Hg. Given the non-normality of the data, it was appropriate to use a non-parametric test (Mann-Whitney U test) to determine the statistically significant differences in Se and Hg levels between different groups. The Empirical Bayes Kriging method, available in the Geostatistical Analyst tool of ArcGIS version 10.3 (ESRI), was utilized to generate spatial maps of Hg and Se levels across the study area. Considering the antagonism of Se and Hg, the molar ratio of Se to Hg was used to evaluate the detoxification potential of Se supplementation (Zheng et al. 2021), which can be calculated using the following equation:

$$\mathrm{Se/Hg\ molar\ ratio} = \frac{\mathrm{C_{Se}} \times \mathrm{Ar_{Se}^{-1}}}{\mathrm{C_{Hg}} \times \mathrm{Ar_{Hg}^{-1}}} \quad (1)$$

where C_{Se} and C_{Hg} are the Se and Hg concentrations in the hair samples, respectively (mg/kg), and Ar_{Se} and Ar_{Hg} are the relative atomic masses of Se (78.96) and Hg (200.59), respectively.

Results

Demographics of Participants

The demographics of the 1373 participants are listed in Table 1, which can be influenced by various factors such as cultural practices, societal norms, and preferences towards

Table 1 Demographics of the 1373 participants who gave hair samples from 16 provinces/ municipalities across central and eastern China from June 2021 to April 2023

Region	Province/ Municipality	Sex		Age	Classification according to the duration of Se supplementation				
		Female	Male		G1	G2	G3	G4	G5
Northeast China	Liaoning	103	42	5–89	39	33	36	23	14
North China	Beijing	21	31	6–86	13	10	10	9	10
	Tianjin	62	27	2–80	21	16	18	17	17
	Hebei	61	26	4–83	26	10	12	16	23
	Shanxi	26	26	2–80	12	11	10	11	8
East China	Shanghai	66	19	10–84	27	10	12	26	10
	Jiangsu	26	16	15–88	10	6	10	7	9
	Zhejiang	24	7	32–72	9	7	4	9	2
	Anhui	13	10	55–75	19	3	0	0	1
	Fujian	17	2	47–88	5	6	1	2	5
	Shandong	62	43	3–79	44	21	13	20	7
Central China	Henan	28	22	2–86	14	10	7	6	13
	Hubei	14	5	35–81	0	3	6	3	7
	Hunan	28	17	33–84	10	14	7	6	8
South China	Guangdong	263	91	3–91	169	63	44	41	37
Northwest China	Shaanxi	128	47	8–93	23	32	42	40	38
Total	1373	942	431	2–93	441	255	232	236	209

Notes: G1 represents the group that participants without additional Se supplementation, G2, G3, G4, and G5 represent groups that participants supplemented with Se-enriched products for 3–6, 7–12, 13–24, and > 24 months, respectively

Table 2 Concentrations of Hg and Se in the hair of metropolitan residents for different groups that classified according to the duration of Se supplementation

Element	Group	N	Range	GM ± GSD	95% CI (lower limits, upper limits)	Percentile				
						10th	25th	50th	75th	90th
Se	G1	441	0.16–1.50	0.38 ± 1.27	0.39, 0.41	0.28	0.33	0.38	0.44	0.50
	G2	255	0.21–1.29	0.48 ± 1.31	0.48, 0.52	0.35	0.40	0.49	0.58	0.66
	G3	232	0.18–1.97	0.56 ± 1.34	0.56, 0.61	0.40	0.47	0.57	0.67	0.79
	G4	236	0.30–1.29	0.60 ± 1.28	0.60, 0.64	0.43	0.52	0.61	0.71	0.83
	G5	209	0.30–1.39	0.65 ± 1.33	0.65, 0.70	0.44	0.56	0.66	0.78	0.94
Hg	G1	441	0.02–3.65	0.25 ± 2.07	0.30, 0.36	0.10	0.16	0.25	0.41	0.59
	G2	255	0.02–1.79	0.21 ± 1.97	0.24, 0.29	0.09	0.14	0.22	0.32	0.51
	G3	232	0.02–0.86	0.20 ± 2.08	0.24, 0.28	0.07	0.12	0.21	0.34	0.53
	G4	236	0.02–1.22	0.21 ± 2.12	0.24, 0.29	0.08	0.12	0.21	0.34	0.54
	G5	209	0.02–1.12	0.19 ± 2.06	0.22, 0.27	0.07	0.11	0.19	0.31	0.46

Notes: GM ± GSD represents geometric mean ± geometric standard deviation; 95% CI represents 95% confidence interval

health and wellness among different age and gender groups. Participants were divided into six regions: Northeast China ($n = 145$), North China ($n = 280$), East China ($n = 305$), Central China ($n = 114$), South China ($n = 354$), and Southwest China ($n = 175$). There were 942 females (68.6%) and 431 males (31.4%) from 16 provinces/ municipalities across central and eastern China. These participants aged 2–93 years, with an average age of 58.64 ± 18.07 years. The 1373 participants were allocated to five groups according to the duration of Se supplementation. G1 ($n = 441$) represents the group that participants without additional Se supplementation. G2 ($n = 255$), G3 ($n = 232$), G4 ($n = 236$), and G5 ($n = 209$) represent participants who supplemented with Se-enriched supplements for a duration of 3–6 months, 7–12 months, 13–24 months, and > 24 months, respectively. Based on the national conditions of China, it is observed that the main consumers of Se-rich health care products are predominantly the elderly population. Additionally, women tend to be more willing to invest time and money in maintaining a healthy lifestyle compared to men. As a result, the surveyed population in this study reflects a higher representation of older individuals as compared to younger ones, as well as a higher proportion of women than men.

Hair Se Levels in Participants

As shown in Table 2; Fig. 3a, hair Se levels in different groups that classified according to the duration of Se

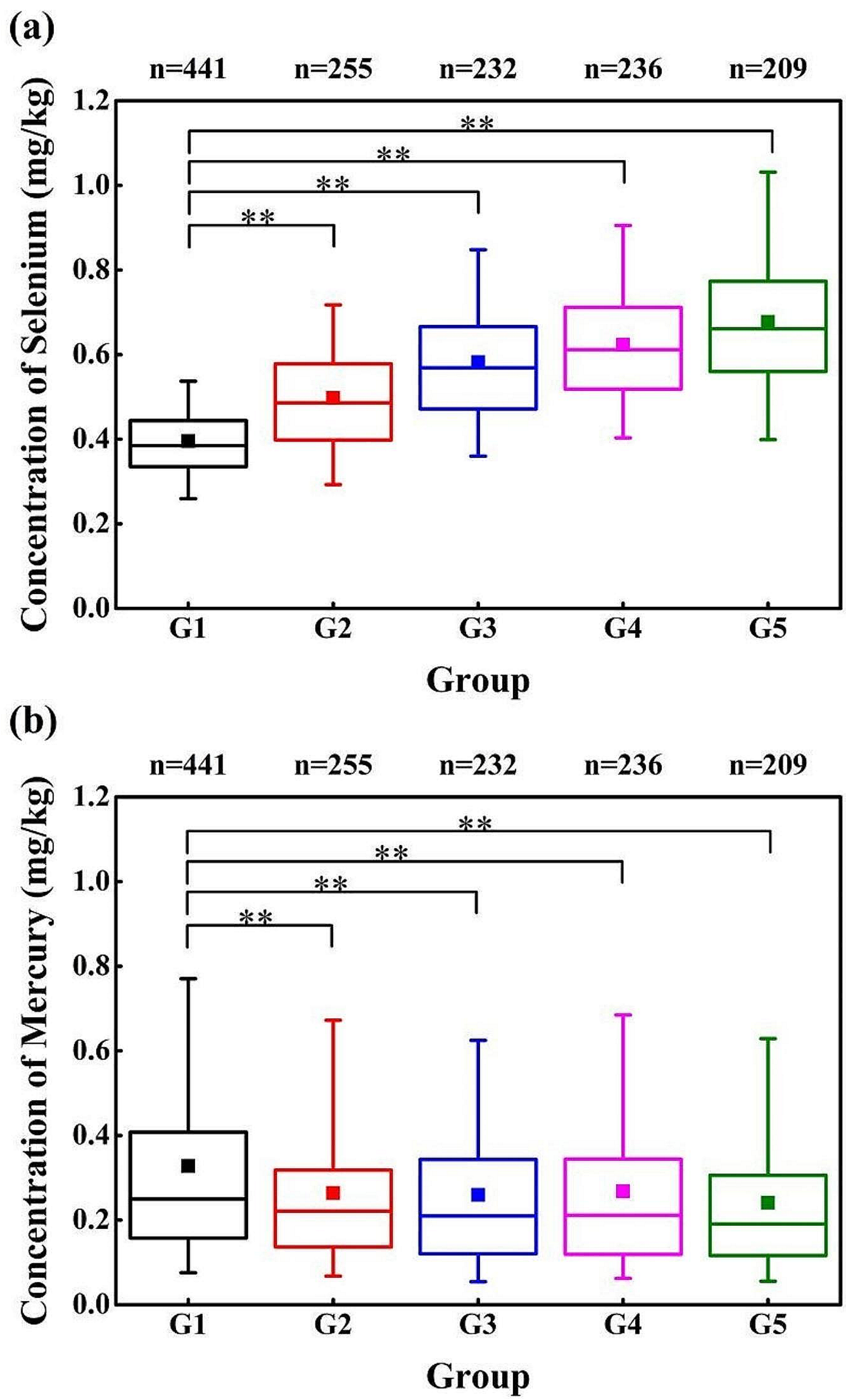

Fig. 3 (**a**) Se and (**b**) Hg concentrations in human hair samples for different groups that classified according to the duration of Se supplementation (*P <0.05, **P <0.01 compared with G1)

supplementation were compared. For participants belong to G1, hair Se levels ranged from 0.16 to 1.50 mg/kg, with a GM of 0.38 (GSD 1.27) mg/kg (95% CI: 0.39, 0.41). Based on the Chinese adult hair Se classification standard, the hair Se levels can be divided into five categories: Se-deficient (< 0.20 mg/kg), Se-marginal (0.20–0.25 mg/kg), Se-sufficient (0.25–0.50 mg/kg), Se-rich (0.50-3.00 mg/kg), and Se-excessive (> 3.00 mg/kg) (Dinh et al. 2018). Approximately 90% of the metropolitan residents' hair Se concentrations belongs to Se-sufficient status (Fig. S3). Hair Se concentrations in metropolitan residents without additional Se supplementation (G1) from different provinces and municipalities across mainland China are listed in Table 3. High hair Se levels were observed in Fujian (0.49 ± 1.29 mg/

Table 3 Hair Se and Hg concentrations for the participants without additional Se supplementation (G1) from different provinces/municipalities across mainland China (mg/kg)

Region	Province/ Municipality	N	Se			Hg		
			Range	Median	GM ± GSD	Range	Median	GM ± GSD
Northeast China	Liaoning	39	0.23–0.54	0.36	0.37 ± 1.21	0.03–0.54	0.18	0.16 ± 1.85
North China	Beijing	13	0.30–0.47	0.39	0.39 ± 1.16	0.02–0.45	0.13	0.12 ± 2.06
	Tianjin	21	0.24–0.61	0.40	0.41 ± 1.25	0.10–1.15	0.26	0.31 ± 2.08
	Hebei	26	0.25–0.50	0.38	0.38 ± 1.20	0.08–0.55	0.15	0.16 ± 1.46
	Shanxi	12	0.16–0.37	0.30	0.28 ± 1.30	0.03–0.25	0.12	0.10 ± 1.92
East China	Shanghai	27	0.21–0.85	0.36	0.36 ± 1.39	0.08–1.82	0.27	0.27 ± 2.07
	Jiangsu	10	0.26–0.48	0.32	0.34 ± 1.24	0.16–0.77	0.25	0.27 ± 1.65
	Zhejiang	9	0.26–0.47	0.34	0.36 ± 1.21	0.04–1.26	0.48	0.32 ± 2.86
	Anhui	19	0.27–0.47	0.36	0.36 ± 1.13	0.07–0.72	0.21	0.24 ± 1.79
	Fujian	5	0.40–0.75	0.44	0.49 ± 1.29	0.69–1.82	0.82	0.98 ± 1.47
	Shandong	44	0.32–0.63	0.39	0.39 ± 1.15	0.04–0.71	0.23	0.20 ± 2.07
Central China	Henan	14	0.20–0.34	0.30	0.28 ± 1.20	0.04–0.24	0.13	0.11 ± 1.75
	Hubei	0	NA	NA	NA	NA	NA	NA
	Hunan	10	0.31–0.44	0.35	0.35 ± 1.13	0.10–1.27	0.29	0.29 ± 2.04
South China	Guangdong	169	0.24–1.49	0.41	0.42 ± 1.27	0.08–3.65	0.34	0.33 ± 1.77
Northwest China	Shaanxi	23	0.22–0.49	0.35	0.34 ± 1.25	0.20–2.08	0.28	0.33 ± 1.68
Total		441	0.16–1.49	0.38	0.38 ± 1.27	0.02–3.65	0.25	0.25 ± 2.07

Notes: GM ± GSD represents geometric mean ± geometric standard deviation; NA: not available

kg) and Guangdong (0.42 ± 1.27 mg/kg), whereas low levels were observed in Shanxi (0.28 ± 1.30 mg/kg) and Henan (0.28 ± 1.20 mg/kg). For the participants without additional Se supplementation (belong to G1), there were no significant sex-related differences in hair Se levels among metropolitan residents from the six regions of China (Fig. 4a, Table S2). According to the Chinese age division standard, the participants in this investigation were classified into five groups: juvenile (≤ 18 years old), young adult (19–44 years old), middle-aged (45–59 years old), elderly (60–74 years old), and very elderly (≥ 75 years old) (Table S3). No significant correlations were found between the Se and Hg concentrations in human hair and age (Fig. 5a, Table S4).

The hair Se contents were 0.48 ± 1.31 (95% CI: 0.48, 0.52), 0.56 ± 1.34 (95% CI: 0.56, 0.61), 0.60 ± 1.28 (95% CI: 0.60, 0.64), and 0.65 ± 1.33 (95% CI: 0.65, 0.70) mg/kg for participants belong to G2, G3, G4, and G5, respectively (Table 2), which are similar to the levels found in Se-rich regions of China, ranging from 0.59 to 1.0 mg/kg (Dinh et al. 2018). For participants belong to G2, G3, G4, and G5, 47%, 66%, 80%, and 82% of metropolitan residents' hair Se concentrations belongs to Se-rich status (Fig. S3). For participants belong to Se supplementation groups (G2-G5), hair Se levels exhibited statistical significantly higher values ($p < 0.01$) compared to G1 (Fig. 3a), suggesting that Se supplementation can effectively increase Se levels in the body, as reflected in the hair.

Hair Hg Levels in Participants

For participants belong to G1, the Hg concentration in hair samples ranged from 0.02 to 3.65 mg/kg, with a GM of 0.25 (GSD 2.07) mg/kg (95% CI: 0.30, 0.36) (Table 2). Among these residents, 2.27% exceeded the safe limit of 1 mg/kg proposed by the U.S. EPA for hair Hg, whereas none exceeded the WHO tolerance limit of 10 mg/kg (Fig. S3). As listed in Table 3, high concentrations of hair Hg were found in Fujian (0.98 ± 1.47 mg/kg), Guangdong (0.33 ± 1.77 mg/kg) and Shaanxi (0.33 ± 1.68 mg/kg), whereas low concentrations were found in Shanxi (0.10 ± 1.92 mg/kg), Henan (0.11 ± 1.75 mg/kg), Beijing (0.12 ± 2.06 mg/kg), Hebei (0.16 ± 1.46 mg/kg), and Liaoning (0.16 ± 1.85 mg/kg). As shown in Fig. 4b, a significant difference ($p < 0.05$) was only found in hair Hg levels of southern region between males and females. For other regions, including the northeast, north, east, central, and northwest regions of China, there was no sex-specific accumulation of Hg in hair. There was no significant age difference in Hg content in G1 group among participants from six regions in China (Fig. 5b, Table S4).

The hair Hg contents were 0.21 ± 1.97 (95% CI: 0.24, 0.29), 0.20 ± 2.08 (95% CI: 0.24, 0.28), 0.21 ± 2.12 (95% CI: 0.24, 0.29), and 0.19 ± 2.06 (95% CI: 0.22, 0.27) mg/kg for participants belong to G2, G3, G4, and G5, respectively (Table 2). Participants with Se supplementation (G2-G5) exhibited statistical significantly lower hair Hg values ($p < 0.01$) compared to G1 (Fig. 3b), suggesting that Se supplementation had a positive effect on reducing Hg levels

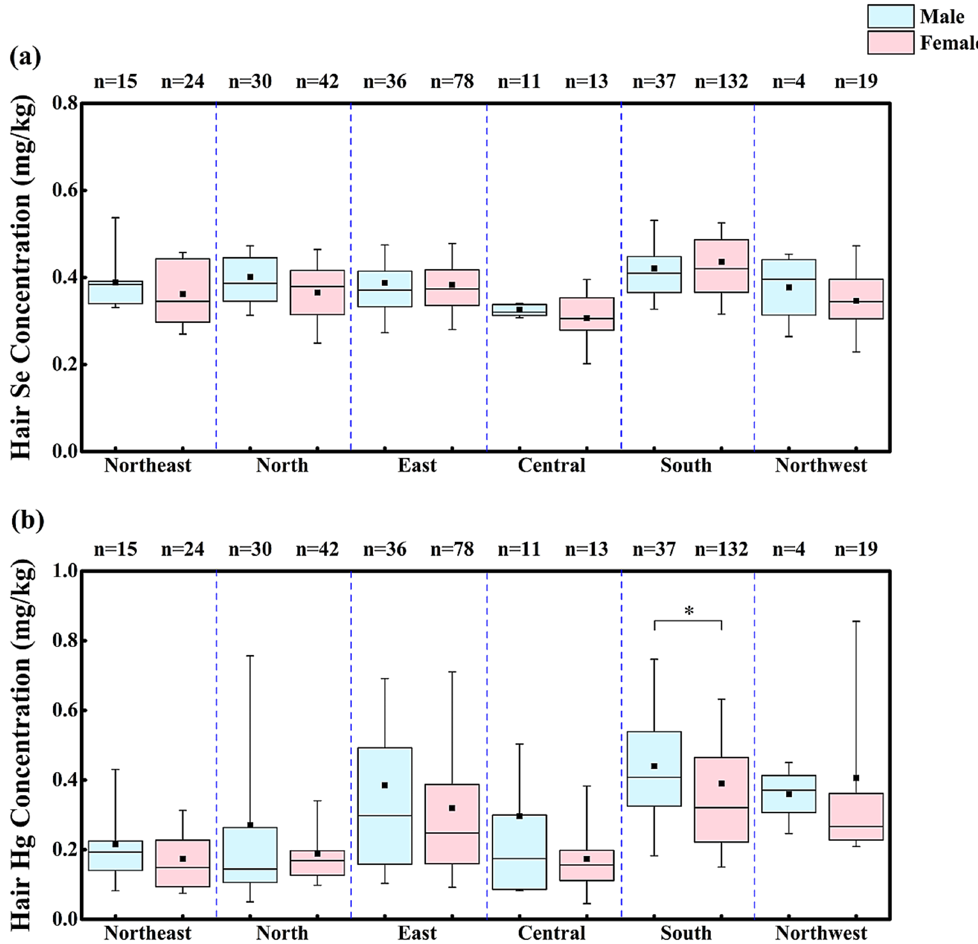

Fig. 4 Box plot displaying the concentration of (**a**) Se (mg/kg), (**b**) Hg (mg/kg) in hair by sex

in the hair of the resident. However, hair Hg concentrations did not significantly change with different duration of Se supplementation. This result suggests that the hair Hg levels of general population residents plateaued after 3–6 months supplemented with Se-enriched products. The significance of these findings is not yet clear; however, the plateau implies that the body has reached a balance in terms of Se and Hg interactions.

Discussion

Spatial Distributions of Hair Se and Hg Levels

Comparisons of Se and Hg levels in hair in this study with those reported in previous studies for different populations worldwide are shown in Table 4. Hair Se levels for general population residents without Se supplementation were far below those of Se-sufficient (Fang et al. 2012) and Se-rich regions (Li et al. 2012b; Zhao et al. 2023) in China. The mean hair Se concentration in this study was similar to that reported for the hair of metropolitan residents in China (Li et al. 2021; Zheng et al. 2021), indicating that there may be regional patterns in Se distribution within the Chinese population. The southern region of China has soils that are naturally rich in Se, leading to higher levels of Se uptake by plants, and subsequently higher levels of Se in the food chain (Yang et al. 2022). In addition, residents living in coastal provinces (e.g., Fujian and Guangdong) eat aquatic products (such as fish, shrimp, shellfish, and other seafood) more frequently, which are known dietary sources of Se (Li et al. 2014).

With respect to Hg, the mean hair Hg concentrations in this study were far below those of residents living around Hg mining areas (Li et al. 2012b; Rodriguez-Villamizar et al. 2023) or fishing villages (Fang et al. 2012), where Hg exposure occurs primarily through mining activities and fish consumption (Table 4). However, the average hair Hg level was higher than that in the hair of susceptible populations (e.g., children and students) worldwide (Astolfi et al. 2020; Izydorczyk et al. 2021; Singh et al. 2023). This may be due to several factors, including the Hg concentration in soils, coal combustion, gross domestic product (GDP), and per capita consumption of aquatic products. Zhou et al. (2018) found that the concentrations of Hg in agricultural soils were higher in East and South China, which is in

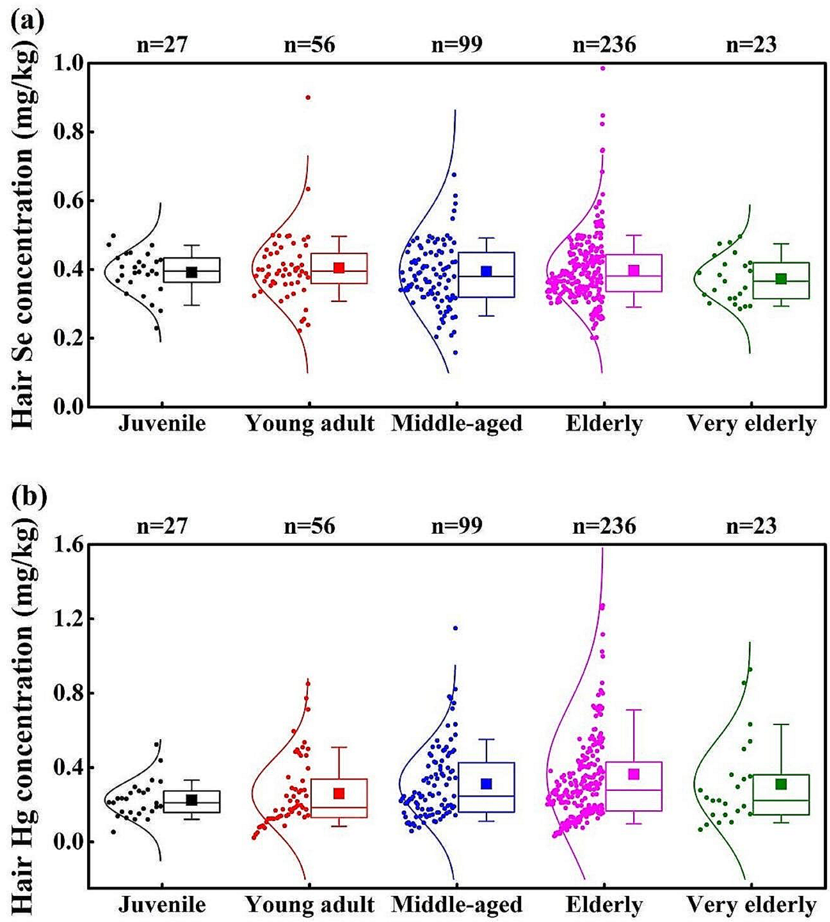

Fig. 5 Box plots for hair (**a**) Se, and (**b**) Hg concentrations among different age groups

Table 4 Comparisons of Se and Hg levels in hair in the present study with those reported in previous studies for different populations worldwide (mg/kg)

Study area	Description	Hair Se level (mg/kg)		Hair Hg level (mg/kg)		References
		range	mean ± S.D.	range	mean ± S.D.	
Hunan, China	Hg mining area	0.41–2.05	0.91 ± 0.43	0.30–7.62	3.04 ± 1.85	Li et al. (2012)
Zhejiang, China	Fishing village	0.76–1.58	1.0 ± 0.13	0.18–3.04	0.76 ± 0.51	Fang et al. (2012)
Anhui, China	Se-rich area	0.40–2.80	0.99 ± 0.40	0.27–1.10	0.45 ± 0.15	Zhao et al. (2023)
29 provincial capitals, China	General population residents	ND-200	0.37[a]	ND-12.5	0.22[a]	Zheng et al. (2021)
Bameza, Ethiopia	Children	ND-0.67	0.19 ± 0.13	0.01–0.48	0.06 ± 0.09	Astolfi et al. (2020)
Lower Silesia, Poland	Students	0.06–0.88	0.74 ± 1.42	0.03–0.34	0.15 ± 0.16	Izydorczyk et al. (2021)
10 provinces, Canada	General population residents	0.11–0.37	0.64 ± 0.02	0.14–0.25	0.19 ± 0.02	Singh et al. (2023)
Santander, Colombian	Gold-mining area	NA	0.60 ± 0.59	NA	0.80 ± 1.98	Rodriguez-Villamizar et al. (2023)
16 provincial capitals, China	General population residents without Se supplementation	0.16–1.50	0.38 ± 1.27	0.02–3.65	0.25 ± 2.07	This study
	General population residents with Se supplementation	0.18–1.97	0.57 ± 1.35	0.02–1.79	0.20 ± 2.06	This study

[a] reported as mean value; NA: not available

accordance with the high-value regions of hair Hg in this study. In addition, a recent study reported that Hg levels in human hair were significantly related to coal combustion and GDP (Zheng et al. 2021). For coastal residents in East and South China, the consumption of aquatic products is a major pathway for Hg exposure (Shao et al. 2013). According to the National Statistical Yearbook 2022, most provinces that exceed the national per-capita consumption of aquatic products are concentrated in East and South China, where fish and seafood constitute as part of the daily diet. In addition, we plotted the spatial distributions of hair Se and Hg levels throughout the country for different groups that classified according to the duration of Se supplementation. It should be noted that the 32 cities used in the Empirical Bayes Kriging method were concentrated in Central and Eastern China; thus, the modeled distributions in northwestern and southwestern China had large uncertainties. As presented in Fig. 6, the hotspots of hair Hg concentrations were concentrated in South and East China. Compared to G1, the distribution area of high value of hair Hg in the Se supplementation groups (G2-G5) has reduced gradually. This finding indicates that Se supplementation has a noticeable effect on reducing hair Hg concentration in coastal regions. Therefore, Se supplementation could be an effective strategy for protecting coastal residents in South and East China from the harmful effects of Hg.

Impacts of Sex and Age on Hair Se and Hg Levels

Box plots for the distribution of Se and Hg concentrations in hair from different sex groups are shown in Fig. 4 and Fig. S4. Participants in the G1 group studied in this study did not show gender differences in the six regions (Fig. 4a, Table S2), which is consistent with a previous investigation reported by Astolfi et al. (2020). A recent study by Zheng et al. (2021) evaluated the impact factors of heavy metals in the hair of general population residents from 29 provincial capitals in China and reported that males had higher concentrations of Se in their hair than females ($p < 0.05$). However, higher concentrations of Se in female hair (3.13 ± 1.91 mg/kg) than in male hair (2.21 ± 1.14 mg/kg) were observed in Enshi, China (Huang et al. 2013). Sex differences in Se content vary from place to place, which is probably due to various factors that may affect participants' responses to Se, such as dietary preferences, sexual hormones, metabolization, and lifestyle (Qin et al. 2021). For instance, people who prefer to eat staple foods or foods rich in Se can effectively increase the concentration of Se in their body (Li et al. 2014). With respect to Hg, there was a significant difference ($p < 0.05$) in hair Hg concentrations of southern region between males and females (Fig. 4b). Similar results have been reported in studies by Xie et al. (2021), Liu et al. (2014), and Qin et al. (2021). According to the mean and median values of hair Hg, male hair tended to have higher concentrations than female hair (Table S2), which is in agreement with the results of a previous study by Cegolon et al. (2023). Compared to females, a relatively higher daily diet intake (such as rice and fish), higher frequency and intensity of outdoor work, and hormonal control are possibly responsible for the higher hair Hg concentrations in males (Wang et al. 2021a).

Box plots of hair Se and Hg concentrations among the age groups are shown in Fig. 5. The hair Se concentrations (GM ± GSD) did not change much with age, with values varying from 0.39 ± 1.19 mg/kg in the juvenile group to 0.37 ± 1.19 mg/kg for the very elderly group. For Hg, the levels of hair Hg increased from the juvenile group to the elderly group before slightly decreasing, probably due to the capacity of human metabolism. The young adults group had the lowest mean value of hair Hg (0.20 ± 2.11 mg/kg), followed by juvenile (0.21 ± 1.58 mg/kg) and very elderly (0.24 ± 2.02 mg/kg), and the elderly group had the highest mean value (0.27 ± 2.20 mg/kg). Similar findings have also been reported in previous studies, indicating that the body burden of Hg increases with increasing age (Gibb et al. 2016; Liu et al. 2008). Compared with younger people, the elderly have a lower capacity for human metabolism and may be more susceptible to Hg because of the gradual lifelong accumulation of Hg in the human body (Wang et al. 2021a). We further compared Hg concentration in the hair of elderly residents ($n = 698$) for different groups that classified according to the duration of Se supplementation. The geometric mean hair Hg contents were 0.27 ± 2.20 mg/kg (95% CI: 0.32, 0.41) and 0.17 ± 2.07 mg/kg (95% CI: 0.19, 0.25) for participants belong to G1 (without Se supplementation) and G5 (Se supplementation for more than 24 months), respectively (Table S5). These results suggest that Se supplementation is an effective strategy for protecting elderly residents from the harmful effects of Hg.

Human Hg Exposure Assessment and Antagonism with Se

Traditional health risk assessments have primarily focused on the level of Hg exposure. However, it is important to consider the potential protective effects of Se against Hg toxicity (Zhang et al. 2014). Thus, we used two criteria to assess the health risks of Hg exposure in metropolitan residents in China for different groups that classified according to the duration of Se supplementation. For participants belong to G1, only 2.27% exceeded the recommended limit of 1.0 mg/kg for Hg concentrations established by the USEPA (Table 5, Table S6). For participants belong to G2, G3, G4, and G5, Hg levels in the hair of approximately 99%

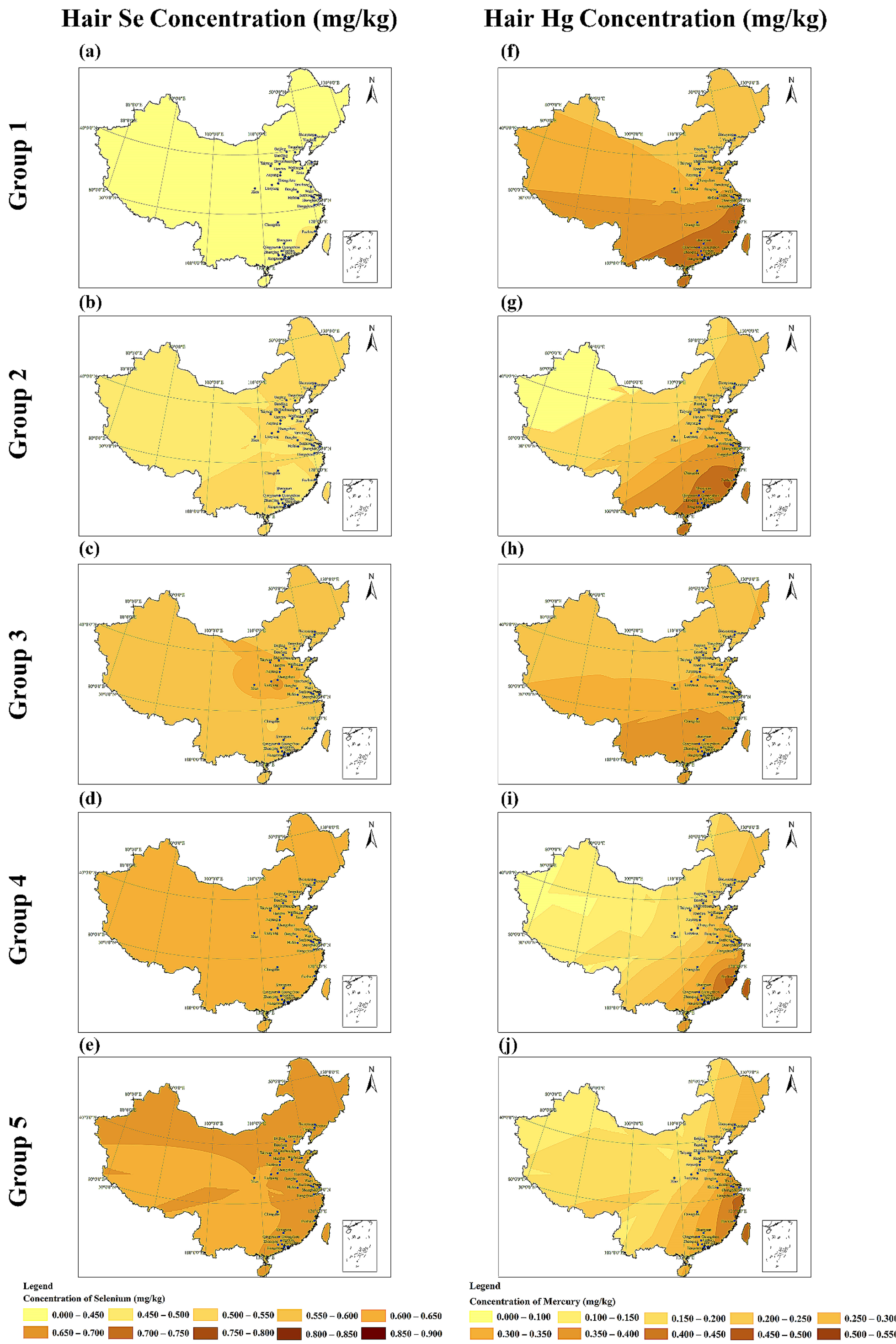

Fig. 6 Spatial distribution of hair (**a-e**) Se and (**f-j**) Hg concentrations in China for different groups that classified according to the duration of Se supplementation

of the participants were lower than the threshold exposure recommended by the USEPA. However, when considering the Se:Hg molar ratio, 25.40% of the participants had Se:Hg molar ratios below 1 (Fig. 7a; Table 5). This finding indicates that although only a small percentage of participants exceeded the recommended limit for Hg concentrations, a significant proportion exhibited imbalanced Se:Hg ratios, which could potentially increase their vulnerability to the toxic effects of Hg. Potential risks of Hg exposure were identified for the metropolitan residents of China, particularly those living in East China (30.70%) and South China (32.54%) (Table 5, Table S6). Many studies have confirmed that hair Hg levels are significantly correlated with the per capita consumption of aquatic products (Fang et al. 2012;

Table 5 Proportions (%) of participants with hair Hg levels exceeding recommended values

Group	Regions	Hg	Se/Hg
Recommended values		> 1 mg/kg (US EPA)	< 1
G1 (*n* = 441)	Northeast China (*n* = 39)	0	0.68
	North China (*n* = 72)	1.39	13.89
	East China (*n* = 114)	4.39	30.70
	Central China (*n* = 24)	4.17	12.50
	South China (*n* = 169)	1.18	32.54
	Northwest China (*n* = 23)	4.35	25.00
	Total	**2.27**	**25.40**
G2 (*n* = 255)	Northeast China (*n* = 33)	0	3.03
	North China (*n* = 47)	0	8.51
	East China (*n* = 53)	1.89	16.98
	Central China (*n* = 27)	3.7	18.52
	South China (*n* = 63)	0	15.87
	Northwest China (*n* = 32)	0	0
	Total	**0.78**	**11.37**
G3 (*n* = 232)	Northeast China (*n* = 36)	0	0
	North China (*n* = 50)	0	8.00
	East China (*n* = 40)	0	5.00
	Central China (*n* = 20)	0	20.00
	South China (*n* = 44)	0	13.64
	Northwest China (*n* = 42)	0	0
	Total	**0**	**6.90**
G4 (*n* = 236)	Northeast China (*n* = 23)	0	4.35
	North China (*n* = 53)	0	0
	East China (*n* = 64)	3.12	9.38
	Central China (*n* = 15)	0	13.33
	South China (*n* = 41)	2.44	14.63
	Northwest China (*n* = 40)	0	2.50
	Total	**1.27**	**6.78**
G5 (*n* = 209)	Northeast China (*n* = 14)	0	0
	North China (*n* = 58)	0	1.72
	East China (*n* = 34)	0	14.71
	Central China (*n* = 28)	0	3.57
	South China (*n* = 37)	2.70	2.70
	Northwest China (*n* = 38)	0	0
	Total	**0.48**	**3.83**

Liu et al. 2014). Therefore, metropolitan residents in East and South China need to establish a reasonable dietary structure, reduce their intake of large carnivorous fish at the top of the food chain (e.g., sardines, yellow croaker, hairtail, tuna, salmon, and codfish), and increase their intake of Se-rich foods.

For participants who supplemented with Se-enriched supplements (G2-G5), the majority of participants had Se:Hg molar ratios greater than 1 (Fig. 7b and e), suggesting that Se in the human body can potentially bind to or detoxify Hg. Considering the duration of Se supplementation, the proportion of participants with Se:Hg molar ratios < 1 were 11.37%, 6.90%, 6.78%, and 3.83% for G2, G3, G4, and G5, respectively, as indicated in Table 5. This suggests that long-term Se supplementation may enhance the body's ability to bind or detoxify Hg, leading to a lower proportion of participants with an imbalanced Se:Hg ratio. Based on the present mechanisms of the antagonistic reaction between Se and Hg, Hg has a high affinity for thiol (-SH) groups and this interaction is a significant factor contributing to its toxicity (Clarkson 2002; Zhang et al. 2013). In the human body, Se-enriched dietary supplements (e.g., corn flour, probiotics, and yeast) can be metabolized to selenide (Li et al. 2011). Because the binding affinity between Se and Hg (with a constant of 10^{45}) is one million-fold higher than that between sulfur (S) and Hg, the interaction between Se and Hg readily forms an Hg-Se complex ($[(Hg-Se)_n]_m SeP$) (Khan and Wang 2009). In general, two important Hg excretion mechanisms are involved in the human body: (1) Hg is deposited in hair along with the formation of nanoparticulate Hg sulfide (nano β-HgS) and an RS-Hg-SR complex (Manceau et al. 2016) and (2) demethylation and excretion of Hg(II) through urine (Queipo-Abad et al. 2019). For example, a 3-month organic Se supplementation trial in the Wanshan Hg mining area suggested that Se supplementation could increase urinary Hg excretion in long-term Hg-exposed populations (Li et al. 2012a). In contrast to urinary Hg excretion, scalp hair usually shows Hg exposure over a long period (months to years). For metropolitan residents exposed to long-term and low-dose Hg, Se may play a vital role in restoring the function of selenoenzymes and increasing Hg excretion after short-term (about 3–6 months) Se supplementation. Hair samples after 6 months of Se supplementation could reflect a steady level of body Hg in ordinary people in Chinese cities.

Certainly, it is important to understand and acknowledge the limitations of cross-sectional studies when examining changes in hair Se and Hg levels over a specific time window. Two important limitations associated with this study are as follows: (1) Individual variability: Cross-sectional studies assess participants at a specific point in time, which makes it difficult to account for individual variations. Factors such as initial mercury levels, dosages, bioavailability of Se-rich products used, and individual living and dietary habits can influence the results and make it challenging to establish clear cause-effect relationships. (2) Recall bias: In this study, recall bias refers to the potential distortion of results due to participants' ability to accurately remember and report their Se supplementation duration and dosage. The reliance on volunteers filling in questionnaires introduces the possibility of flawed information recall, which may lead to inaccuracies in assessing the relationship between Se supplementation and changes in hair Se and Hg levels. To assess the effects of Se on mercury detoxification, it is

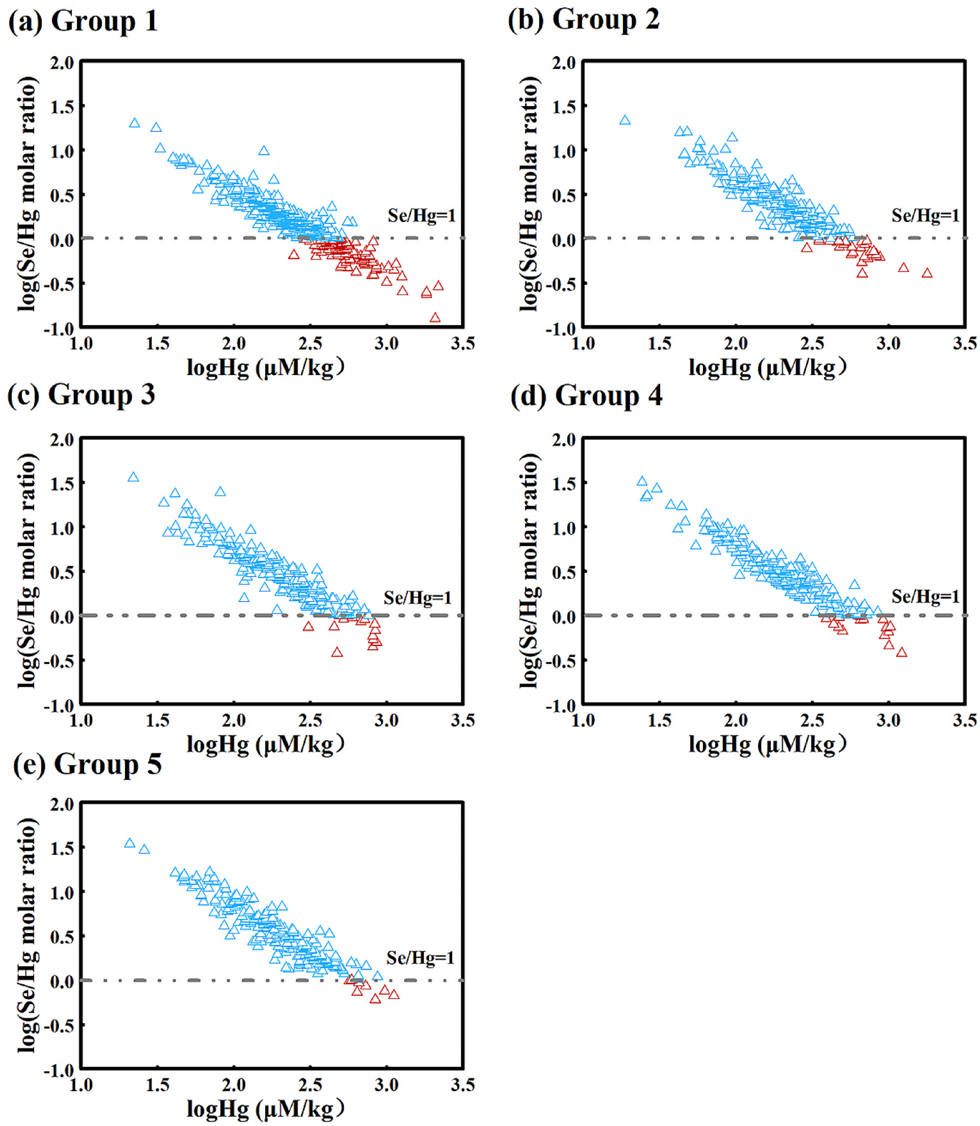

Fig. 7 Scatter plot of the logarithm of levels of Hg (μM/kg) and log (Se/Hg molar ratio) in human hair and its comparison with threshold 1 for different groups that classified according to the duration of Se supplementation

recommended that future research focus on the following aspects: (a) Intervention study in a controlled environment: Conducting an intervention study in a controlled environment can provide valuable insights into the direct effects of Se supplementation on Hg detoxification. (b) Longitudinal analysis with long hair samples: Long-term effects of Se supplementation on human Hg exposure can be explored through the collection of long hair samples and subsequent segmented analysis. (c) Incorporation of multiple biological indicators: To elucidate the antagonistic mechanism between Hg and Se after Se supplementation, it would be beneficial to incorporate multiple biological indicators such as blood, hair, and urine.

Conclusions

This study investigated the hair Se and Hg levels and evaluate the potential health impacts of Se supplementation on the detoxification of Hg for metropolitan residents in China. Participants with Se supplementation (G2-G5) exhibited statistical significantly lower hair Hg values ($p < 0.01$) compared to those without supplementation (G1), suggesting that Se supplementation had a positive effect on reducing Hg levels in the hair of metropolitan residents. Hotspots of hair Se and Hg concentrations were mainly concentrated in South and East China. A significant difference ($p < 0.05$) was only found in hair Hg levels of southern region between males and females. The hair Se concentrations did not change much with age, and hair Hg increased from the juvenile group to the elderly group before slightly decreasing, probably due to the capacity of human metabolism.

Considering the duration of Se supplementation, the proportion of participants with Se:Hg molar ratios < 1 were 25.40%, 11.37%, 6.90%, 6.78%, and 3.83% for G1, G2, G3, G4, and G5, respectively. This suggests that long-term Se supplementation may enhance the body's ability to bind or detoxify Hg, leading to a lower proportion of participants with an imbalanced Se:Hg ratio. By focusing on intervention studies, longitudinal analyses using long hair samples, and the inclusion of multiple biological indicators, future research can help advance the understanding of the effects of Se on Hg detoxification.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s12403-024-00630-y.

Acknowledgements This research was jointly supported by the National Natural Science Fund of China (Nos. 22176074, 42307135), the Natural Science Fund of Jiangsu Province (No. BK20210485), and the Project funded by China Postdoctoral Science Foundation (No. 2021M701453).

Author contributions Geting Wang: Investigation, Methodology, Data curation, Writing-original draft. Qianqian Hong: Conceptualization, Resources, Writing-review & editing, Supervision. Jian Mei: Conceptualization, Formal analysis. Jinyi Chen: Visualization, Formal analysis. Wansheng Wu: Investigation, Data curation. Feng Han: Investigation, Data curation. Jingchen Xing: Investigation, Data curation. Shijian Yang: Resources, Supervision.

Data Availability The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of Interest The authors declare that they have no conflict of interest.

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