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Monthly variations in mercury exposure of school children and adults in an industrial area of southwestern China

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ABSTRACT

Recent studies have shown that rice consumption can be the major pathway for human methylmercury (MeHg) exposure in inland China. However, few studies have considered the susceptible population of school children's exposure through rice ingestion. In this study, monthly variations in total Hg (THg)/MeHg concentrations in rice, fish, hair, and urine samples were studied to evaluate the Hg (both THg and MeHg) exposure in Guiyang, a typical industrial area with high anthropogenic emission of Hg. A total of 17 primary school (school A) students, 29 middle school (school B) students, and 46 guardians participated in this study for one year. Hair THg, hair MeHg, and urine THg concentrations ranged from 355–413 ng g^{-1}, 213–236 ng g^{-1}, and 469–518 ng g^{-1} Creatinine (ng·g^{-1} Cr), respectively, and no significant differences were observed between different genders and age groups. Hair and urine Hg concentrations showed slightly higher values in the cold season (October to February) than the hot season (March to September), but without significant difference. High monthly variability of individual hair and urine Hg concentrations suggested that long-term study could effectively decrease the uncertainty. The school students showed significantly higher urine THg concentrations than adults due to children's unique physiological structure and behaviors. Probable daily intake (PDI) of MeHg via rice and fish ingestion averaged at 0.0091, 0.0090, and 0.0079 μg kg^{-1} d^{-1} for school A students, school B students, and their guardians, respectively, which means that 86%, 84%, and 87% of the PDI were originated from rice ingestion, respectively. Therefore, more attention should be paid to children as a susceptible population. The results indicated low risk of Hg exposure via rice and fish consumption for urban residents in a Chinese industrial city.

1. Introduction

Mercury (Hg) is a global pollutant and human Hg exposure is a global issue in the public health perspective. Mercury is emitted substantially by industrial activities (e.g. coal combustion, nonferrous metal smelting, mining, and cement production) and from natural sources and processes (e.g. volcanic eruptions and rock weathering) (Amos et al., 2013; Bourtsalas et al., 2019). Global Hg models estimate that annual anthropogenic emission of Hg to the atmosphere is 2500 ± 500 Mg (Outridge et al., 2018; Obrist et al., 2018), and approximate 3200 Mg of atmospheric Hg is deposited to terrestrial surfaces per year (Amos et al., 2013; Mason et al., 2012). The increase in atmospheric Hg deposition has driven a 15–230% increase in Hg in marine waters during the last 100 years (Outridge et al., 2018). The increased Hg loading on the surface environment increases the risks of Hg entering food chains and potentially posing a health risk to humans.

Mercury emitted from anthropogenic sources is mainly in elemental or inorganic forms, but the non-occupational human exposure is mainly via its organic compounds from food consumption, such as methylmercury (MeHg), which is potent toxicant (USEPA., 1997; Tatsuta et al., 2012; Pollard et al., 2019; López et al., 2019). Previous studies have shown that sulfate-reducing bacteria and iron-reducing bacteria with *hgcA* and *hgcB* genes facilitate the transformation of inorganic Hg (IHg) into MeHg due to their high potential for Hg methylation (Parks et al., 2013; Ma et al., 2019). The main route of human exposure to elemental mercury vapor is through inhalation of ambient air from occupational

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working environments and through dental amalgams. The main route of human exposure to MeHg is dietary intake (Kobal et al., 2017; Li et al., 2015a), while ingestion of fish and seafood is considered to be the most common source (Perez et al., 2019; Wells et al., 2020). However, it should be noted that MeHg exposure also occurs through rice ingestion (Feng et al., 2008; Zhang et al., 2010; Du et al., 2016). Davis et al. (2014) confirmed a positive relationship between rice Hg consumption and blood Hg concentration in a US population (n = 16,236). In an Hg-mining area of China, hair and blood Hg concentrations were positively correlated with MeHg intake through rice ingestion (n = 237, Du et al., 2016; n = 98, Feng et al., 2008; n = 168, Li et al., 2015b). Many studies focused on MeHg exposure of children in fish/seafood-consumption populations (Chan et al., 2018; Diez et al., 2008; Gibb et al., 2016; Kobal et al., 2017; Pedro et al., 2019; Wells et al., 2020), but few studies have considered the health risks of MeHg exposure in children via rice consumption.

MeHg is well known as a notorious neurotoxicant to humans even at low levels of exposure (Carocci et al., 2014). Although Hg and its compounds can result in toxicity to the respiratory, immune and reproductive systems, blood cells, the kidneys, and the central nervous system are also important targets of Hg toxicity (Carocci et al., 2014; Officioso et al., 2018; López et al., 2019; Yang et al., 2020). More than 95% of MeHg ingested from the diet is absorbed by the gastrointestinal tract, where MeHg can easily enter the bloodstream during gestation. Additionally, MeHg can readily cross the placental barrier and accumulate in fetal blood, and cross the blood-brain barrier; both of these outcomes pose key risks for developmental neurotoxicity (Grandjean et al., 2014; Pino et al., 2018; López et al., 2019). Therefore, fetal, neonatal and early age Hg exposure is of particular concern, and it can affect the health of future generations (Llop et al., 2020). It is important to evaluate the Hg exposure in the susceptible population, such as children, in order to decrease the health risks of Hg exposure (Akrout et al., 2015; Ruggieri et al., 2017; Perez et al., 2019; Papadopoulou et al., 2019).

Biological indicators are important tools for the evaluation of an internal dose as well as for the evaluation of temporal trends in the fields of public and environmental health (Cryderman et al., 2016; Du et al., 2018b; Tratnik et al., 2013). Therefore, the selection of suitable biological indicators of environmental pollutants is of particular importance to inform decision making in public health. Blood, urine and hair are widely used matrixes in human biological monitoring (Jain, 2017; Pino et al., 2018). Total Hg in urine can reflect short-term exposure to IHg, while Hg in blood reflects relatively short-term exposures to MeHg. Hair THg or MeHg can reflect temporal variations in MeHg exposure by measuring segmental Hg concentrations of hair (WHO, 2003; Barregard et al., 1993; Li et al., 2011a).

With rapid economic growth over the last four decades in China, Hg emissions have increased from 147 to 530 Mg during the 1978–2014 period and the major emission sources were coal-fired industrial boiler, coal-fired power plant, nonferrous metals smelting, and cement production (Wu et al., 2016). Guiyang City in southwestern China is considered as a hotspot due to its high Hg concentrations in the atmosphere, soil, water, fish, and vegetables (Chen et al., 2016; Du et al., 2018a; Fu et al., 2011; Feng et al., 2005, 2011). Mercury in environmental substrates can be transferred to urban residents' bodies though food chain, posing particularly high risks to susceptible populations, like children. Therefore, it was necessary to evaluate the Hg exposure level for the local residents in Chinese urban areas.

This study presented a one-year continuous study of THg and MeHg in daily food (rice and fish), and in hair and urine samples collected monthly from children and their guardians from a primary school (school A) and a middle school (school B) in Guiyang City. The objectives were: (1) to examine the exposure levels of MeHg via rice and fish consumption in school-age children and adults; (2) to assess internal Hg exposure using hair and urine THg and MeHg concentrations; and, 3) to evaluate monthly variations in food Hg, and hair and urine THg and MeHg concentrations of children living in inland industrial cities in China.

2. Materials and methods

2.1. Study area

This study was conducted in Guiyang City, the capital of Guizhou Province. Guiyang is an ecological tourist city in the east of the Yunnan-Guizhou Plateau, with an elevation of about 1100 m. The climate is influenced by the Southwest Monsoon. Annual means of air temperature and rainfall are 15.3 °C and 1200 mm, respectively. The area stretches from 106°07′ to 107°17′ E longitude and from 26°11′ to 26°55′ N latitude. The Guiyang City is consisted of 10 administrative districts (counties), while two districts (Yunyan and Nanming) constitute the urban area (220 km^2). The population was about 4.3 million in 2014, with the central district having more than 2 million urban residents and a density of 30 thousand people per km^2. Industry is the main category of the Guiyang economy, including coal-fired power plants, coal mines, nonferrous metal smelting, cement production, tobacco, and rubber industry. Soil THg concentrations in the urban areas averaged at 120 ng g^{-1} (with a range of 20–460 ng g^{-1}), which showed a moderate level of ecological risk (Duan et al., 2018). The gaseous elemental mercury (GEM), particulate bound mercury (PBM), and gaseous oxidized mercury (GOM) ranged from 6.1 to 22.2 ng m^{-3}, 55.9–1984.9 pg m^{-3}, and 20.5–68.8 pg m^{-3}, respectively (Xu et al., 2016).

2.2. Sample collection

One primary school (Guiyang Nanming Primary School, school A) and one middle school (Guiyang No. 18 Middle School, school B) were selected for the current study. Both schools were located in the urban center (Figure S1), surrounded by twelve Hg sources (including power plant, cement production, iron and steel production, etc.) within 1–24 km. Total gaseous mercury at the studying sites averaged at 9.7 ng m^{-1}, which showed a mixture of both residential coal burning and large point sources (Fu et al., 2011). Sampling campaign was conducted from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B. A total of 17 students (8–9 years old) and 17 corresponding family members (33–56 years old) participated in the sampling campaign in school A, and 29 students (13–15 years old) and 29 corresponding family members (36–76 years old) participated in the sampling campaign in school B. The students were chosen randomly in one grade of each school. Details of the basic information of the participants are shown in Table S2. All of the students and their custodial relative signed the informed consent before sampling. This study was conducted in accordance with the Declaration of Helsinki, and obtained ethics approval from the Institute of Geochemistry, Chinese Academy of Sciences (20111201).

All of the subjects were required to complete a questionnaire. The questionnaire included social-demographic and occupational information regarding dietary habits, food consumption frequency, and health status (including height, weight, health, medical reports, smoking, drinking, etc). Hair and urine samples from each participant were collected each month, as well as rice and fish samples (if they ingested fish during the sampling month) taken from their home and the school canteens. Hair samples with a length of 1 cm, which reflected the last month's exposure, were cut with stainless steel scissors from the occipital region of the scalp, stapled together, packed in polythene bags, and then transported to the laboratory. The morning urine samples were collected in 50 mL pre-cleaned polyethylene bottles and were preserved in an icebox during transport to the laboratory. A total of 552 rice samples and 126 fish samples were collected during the whole sampling campaign.

2.3. Sample preparation and analysis

The collected hair samples were carefully cleaned with nonionic detergent, distilled water, and acetone, and then dried in an oven at 50 °C prior to analysis (Li et al., 2011a). The urine samples were added with concentrated HNO_3 with 10% of the total volume and then stored at −20 °C until analysis. Rice samples were air-dried, ground, poured through a 100-mesh screen and stored in polythene bags. Fresh fish samples were stored at −20 °C.

The THg concentrations in hair samples were measured by a Lumex RA-915+ Hg analyzer (Lumex Ltd, Russia) coupled with a pyrolysis attachment (Wang et al., 2010). Each sample was analyzed at least twice and the average value was obtained for statistical analysis. Urine, fish, and rice samples were digested with a mixture of HNO_3/H_2SO_4 (v:v = 4:1, 5 mL) for 3 h in a water bath at 95 °C. The THg concentrations in the digestion solutions were determined by BrCl oxidation, $SnCl_2$ reduction, dual-stage gold amalgamation, and cold vapor atomic fluorescence spectrometry (CVAFS, Tekran 2500, Tekran Inc., Toronto, ON, Canada) following Method 1631 (USEPA, 2002).

Rice samples were digested using the KOH-methanol/solvent extraction technique (Liang et al., 1996). Hair and fish were digested with KOH-methanol directly. The MeHg in prepared solutions were ethylated and purged with N_2 onto a Tenax trap, and analyzed by GC-CVAFS (Brooks Rand Model III, Seattle, WA, USA) following Method 1630 (USEPA, 2001a).

To take hydration and urinary flow rate into account, urine THg concentrations were calibrated by Creatinine (Cr) excretion. A HITACHI 7170 A automatic biochemical analyzer (HITACHI, Tokyo, Japan) was used to measure urine Cr concentration within 24 h after sampling. Urine THg concentration is presented with unit of $ng \cdot g^{-1}$ Cr.

2.4. Quality assurance and quality control

Quality assurance and quality control were consisted of method blanks, certified reference materials (CRMs), and duplicate analysis. The limits of detection for THg were 1 ng g^{-1} using the Lumex RA-915+ and 0.03 ng g^{-1} using the Tekran 2500, and the limit of detection for MeHg was 0.003 ng g^{-1}. Recoveries on THg and MeHg in CRMs averaged at 98 ± 3.9%, 94 ± 2.8%, 105 ± 4.5%, 91 ± 1.1%, and 94 ± 3.1%, respectively. The accuracy obtained from duplicate measurements were <5.0% for both THg and MeHg. Results on quality assurance and quality control are shown in Table S1 of supporting information (SI).

2.5. Calculation of MeHg probable daily intakes (PDIs)

The PDIs MeHg via rice and fish consumption were calculated according to equation (1):

$$PDI = \frac{\Sigma(C_i \times IR_{i)}}{bw} \tag{1}$$

where PDI ($\mu g \cdot kg^{-1}$ d^{-1}) is the probable daily intake of MeHg; *i* is the potential MeHg-containing substance (fish or rice). C represents the MeHg concentration of the substance, which was measured in this study. *IR* is the ingestion rate of the substance; *bw* is the body weight of the subject. Both *IR* and *bw* are obtained from the questionnaires, and the detailed data are shown in Table S3. The calculation is based on the assumption that MeHg exposure from other routes (i.e., other foods) are negligible.

2.6. Data analysis

All data were analyzed by SPSS (Version 25, IBM, USA). The data were tested for normal distribution by the Kolmogorov-Smirnov test. If they were not normally distributed, the data were log transformed for further statistical analysis. A total of 40 hair THg and MeHg concentrations (3.8% of hair data) and 77 urine THg concentrations (7.5% of urine data) were removed for regression analyses, because these were outliers based on the 95% confidence interval for the variable. The characteristics of the data are described in Mean ± Standard Deviation (SD) for normal distribution and Geometric mean for log normal distribution. Mean values of hair and urine THg and MeHg concentrations were compared between children and adults using multiple-way ANOVA (S–N–K test). The correlation coefficients between different parameters were determined by the Pearson correlation analysis, and, the relationship between hair MeHg and PDI of MeHg and between urine THg and PDI of IHg were studied by Linear Regression analysis. The significance level was set at $p < 0.05$.

3. Results and discussion

3.1. Diet Hg

THg concentrations in rice and fish samples collected from students' homes averaged at 3.7 ± 1.6 ng g^{-1} (with a range of 1.1–9.4 ng g^{-1}) and 10.4 ± 12.5 ng g^{-1} (with a range of 0.2–96.1 ng g^{-1}), respectively. THg concentrations in rice samples collected from school dining rooms averaged at 3.1 ± 1.0 ng g^{-1} (with a range of 1.4–5.9 ng g^{-1}). The means of MeHg concentrations in rice and fish samples collected from students' homes were 2.0 ± 0.9 ng g^{-1} (with a range of 0.4–3.6 ng g^{-1}) and 4.4 ± 5.3 ng g^{-1} (with a range of 0.1–43.3 ng g^{-1}), respectively. MeHg concentrations in rice samples collected from school dining rooms averaged at 2.3 ± 0.9 ng g^{-1} with a range of 0.6–3.4 ng g^{-1} (Fig. 1). No significant difference in rice THg and MeHg concentrations was observed between students' homes and school dining rooms.

MeHg was the main form of Hg in collected rice and fish samples. MeHg accounted for 54% (7–96%) and 69% (16–92%) of THg in the rice and fish samples collected from home kitchens on average, respectively, and 52% (31–73%) of THg in the rice samples collected from the school dining rooms on average. A significant relationship ($r^2 = 0.14$, $p < 0.05$, $n = 224$) between THg and MeHg concentrations in all rice samples was observed. No significant monthly variation in THg and MeHg in rice samples was observed throughout the monitoring year, but THg and MeHg concentrations in fish samples showed high monthly variability due to large differences between fish species.

None of the rice or fish samples exceeded the national limits of 20 ng g^{-1} for rice THg and 500 ng g^{-1} for fish MeHg recommended by the Standardization Administration of China (SAC, 2017). The THg concentrations in rice samples collected from students' homes were comparable with those from the local markets (2.7 ng g^{-1}; Du et al., 2016), but much lower than the mean concentration of 5.8 ng g^{-1} in rice from sold market of 20 provinces in China (Qian et al., 2010) and 4.74 ng g^{-1} in rice from 15 major rice growing provinces in China (Zhao et al., 2019). However, rice MeHg concentrations were comparable with results from 7 provinces in south China (2.47 ng g^{-1}; Li et al., 2012), but much higher than results from 15 provinces in China (0.682 ng g^{-1}; Zhao et al., 2019). The fish MeHg concentrations obtained in this study were much lower than these reported in Europe and North America for freshwater fish, although serious Hg contamination has been found in reservoirs of Guizhou Province (Feng and Qiu, 2008). A previous study suggested that the short food chains in aquatic environments do not promote the accumulation of MeHg in fish, and Chinese populations usually eat herbivorous or omnivorous fish produced by aquaculture, which have short lifetimes and therefore do not bioaccumulate MeHg as wild species in natural environment (Li et al., 2013).

3.2. Hair Hg

The monthly variations of THg and MeHg concentrations in the pupils and their guardians' hair are shown in Figs. 2 and 3, respectively. The annual averages of THg concentrations (±1SD) in hair of school A students, their guardians, school B students, and their guardians were

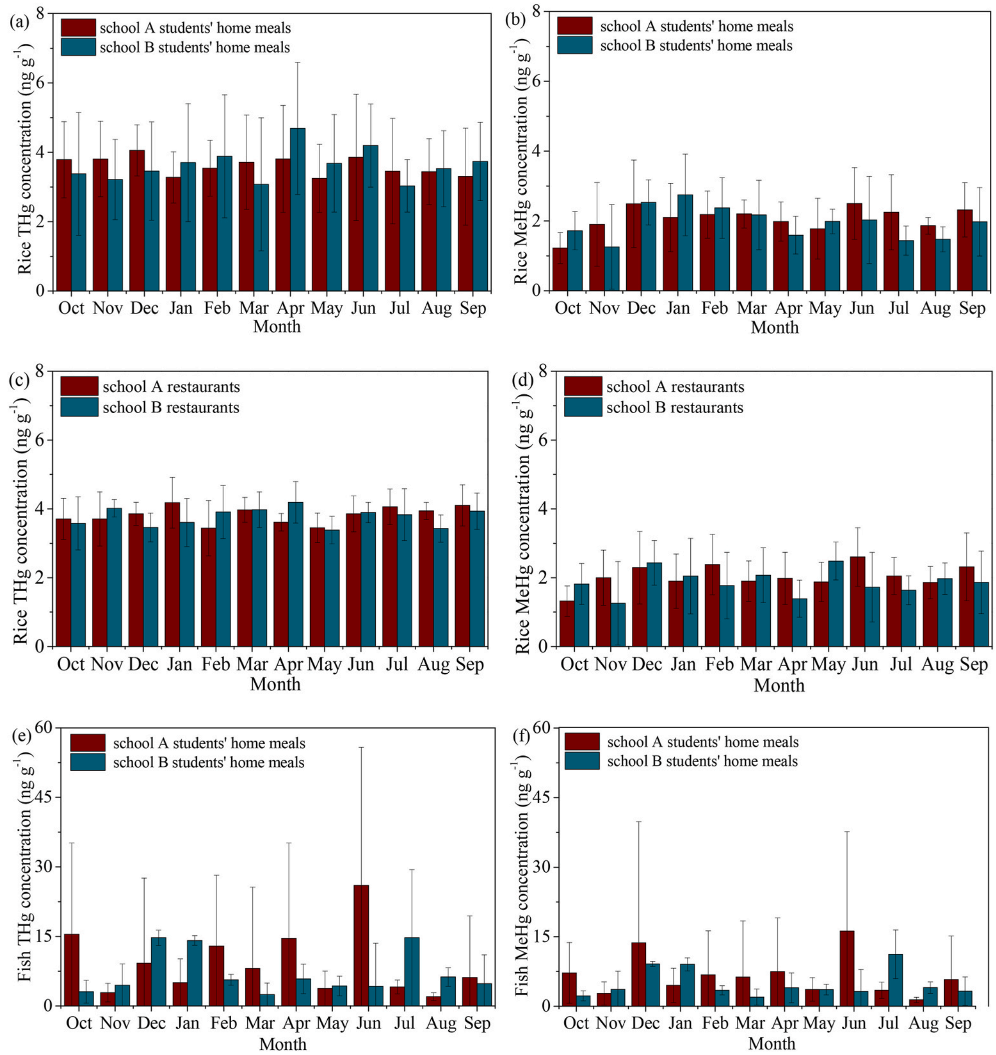

Fig. 1. Monthly averages (±2SD) of THg and MeHg concentrations in rice and fish samples from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B.

388 ± 166, 409 ± 167, 356 ± 151, and 413 ± 168 ng g^{-1}, respectively. The annually averages of hair MeHg concentrations in school A students, their guardians, school B students, and their guardians were 222 ± 104, 231 ± 98, 213 ± 90, and 236 ± 95 ng g^{-1}, respectively. A significant relationship was observed between hair MeHg concentrations and THg concentrations ($r = 0.83$, $p < 0.001$, Figure S2). Hair MeHg concentration accounted for 55% of THg for all studied subjects on average, which was much lower than the ratio of 90% in other populations exposed via fish consumption (NRC, 2000), and no significant difference was found among the four studied groups.

Monthly patterns of hair THg, hair MeHg, and urine THg concentrations are shown in Figs. 2 and 3. Generally, the cold season (October to February) showed slightly higher hair THg, hair MeHg concentrations than the warm season (March to September), but without statistically significant difference ($p > 0.01$). Due to higher energy/food intake in the cold season (Malisova et al., 2015), more MeHg and THg would be ingested by human body, which may result in higher hair Hg concentrations. Additionally, atmospheric GEM and PBM are significantly elevated in the cold season due to residential coal burning (Fu et al., 2011), which would also increase urinary Hg exposure via inhalation for local residents.

In this study, students' hair THg and MeHg concentrations were slightly lower compared to their guardians. Significant differences of hair THg and MeHg were observed between students and guardians in school B ($p < 0.05$), while no significant difference was observed in school A. However, ratios of hair THg and MeHg between each pair of student/guardian from school A and school B showed no obvious difference and varied in the vicinity of 1 (Fig. 4). These results indicated that dietary and nutrition patterns may change with age (Gutierrez--Mosquera et al., 2018; Liu et al., 2014), though family dietary habits may mask this phenomenon.

Generally, the ratio of hair MeHg/THg is relatively low in Chinese populations; for example, 67% for pregnant mothers in a rural area (Hong et al., 2016), 0.62–78% in a Wanshan Hg mining area (Du et al., 2016, 2018a; Jia et al., 2018; Li et al., 2009, 2011b, 2012, 2015b, 2017), 14% in a compact fluorescent lamp factory area (Liang et al., 2015), and 53–78% for fish consumption population in coastal areas (Cheng et al., 2009; Shao et al., 2013). These results suggested that the current recommended Reference Dose (RfD) levels of hair THg set by the EPA or NRC may not suitable for Chinese populations, and more research is urgently needed to understand potential demethylation of MeHg in human body.

3.3. Urine Hg

The monthly variations in urine THg concentrations in school A students, school B students, and their guardians are shown in Fig. 5. The annual means of urine THg concentrations in school A students, school B

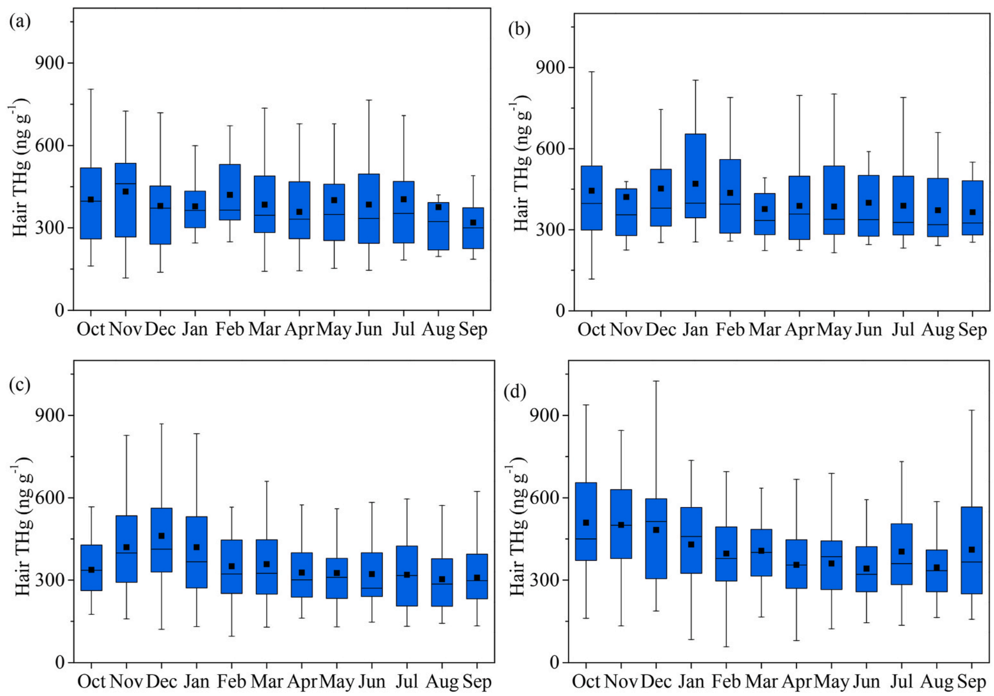

Fig. 2. Monthly variation in hair THg concentrations in different study populations from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B. (a) school A students; (b) guardians of the school A students; (c) school B students; (d) guardians of the school B students. Each box plot represents interquartile range (25th and 75th percentiles), the band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentiles.

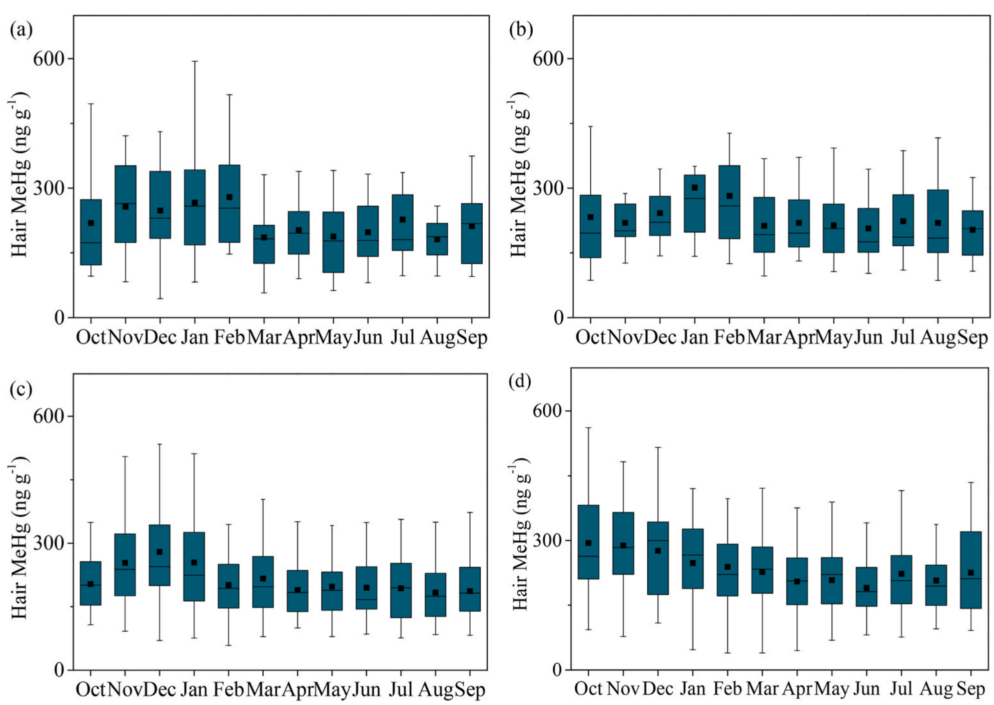

Fig. 3. Monthly variation in hair MeHg concentrations in different study populations from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B (a) school A students; (b) guardians of the school A students; (c) school B students; (d) guardians of the school B students. Each box represents interquartile range (25th and 75th percentiles), the band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentiles.

students, and their guardians, were 532 ± 317, 407 ± 263, 518 ± 300, and 452 ± 289 ng g^{-1} Cr, respectively, and no significant difference was observed among different groups. However, high monthly variability of individual hair/urine Hg was observed and the ratios of SD/mean ranged from 5–55% for hair THg, 6–76% for hair MeHg, and 23–118% for urine THg, respectively. These results suggested that long-term study is needed to decrease the temporal uncertainty.

Monthly patterns of urine THg concentrations is comparable with hair Hg (Fig. 5) as the cold season (October to February) showed slightly higher urine THg concentrations than the warm season (March to September), but without statistically significant difference ($p > 0.01$). Higher energy/food intake, and higher concentrations of atmospheric Hg in the cold season (Fu et al., 2011) may increase urinary Hg exposure for local residents.

Urine THg concentrations in the students from both schools A and B were slightly higher than those of their guardians but without significant difference ($p > 0.05$). The average ratios of urine THg between each pair of student/guardian from schools A and B were greater than 1 (1.21 ± 0.87 in school A; 1.21 ± 1.08 in school B; Fig. 6), which was consistent with previous finding that children are more easily exposed to high levels of IHg than adults (Du et al., 2016). This may arise from children's unique physiology–including higher breathing rate, smaller airways,

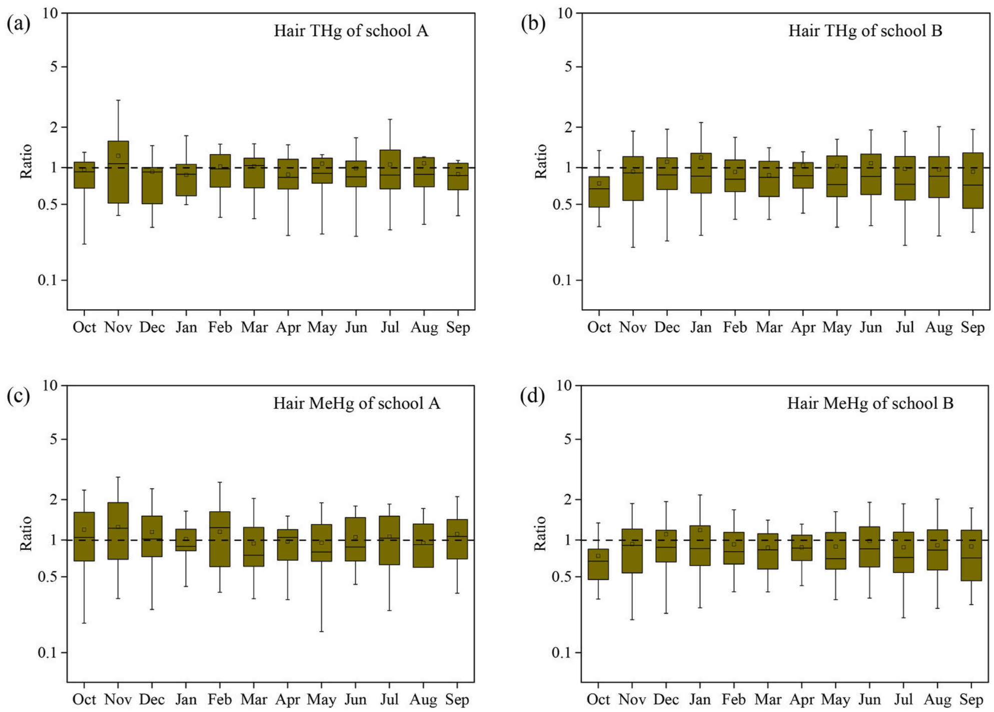

Fig. 4. Monthly variation of the ratio of hair THg, and hair MeHg for students and their guardians from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B. (a) hair THg of school A; (b) hair THg of school B; (c) hair MeHg of school A; (d) hair MeHg of school B. Each box represents interquartile range (25th and 75th percentiles), the band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentiles.

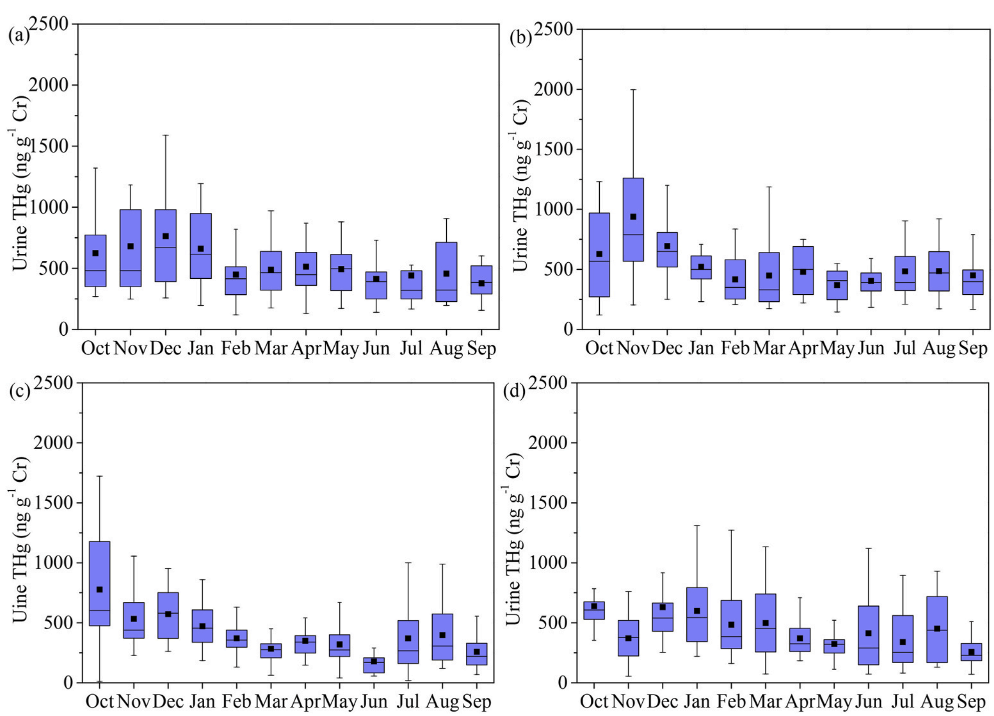

Fig. 5. Monthly variation in urine THg concentrations in different study populations from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B (a) school A students; (b) guardians of the school A students; (c) school B students; (d) guardians of the school B students. Each box represents interquartile range (25th and 75th percentiles), the band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentiles.

and lower breathing zone, and behavior–such as regular hand to mouth contact, causing the accidental or deliberate ingestion of contaminated soil during play (Davis et al., 2006; Ljung et al., 2007; Guney et al., 2010; Du et al., 2016). Compared with urine Hg concentrations of school students and adults, hair Hg concentrations showed the opposite results shown in section 3.2, which is consistent with our previous research (Du et al., 2016). The main reason is that specific metabolic reactions of children affected the behavior of MeHg in children's bodies. For instance, the mean breathing rate over the first 12 years of life is almost twice as fast compared with adult breathing rates (452 vs. 232 L/kg/day, Layton et al., 1993). The metabolic reactions of children are much more active than the adults, which can affect Hg transfer and speciation in children's bodies. Additionally, the enzymatic activity of children may modify different reactions kinetics such as absorption, methylation/demethylation, distribution, and excretion compared with adults (Smith et al., 2009).

A significant relationship was found between THg and MeHg concentrations in human hair (Figure S2), but both of hair THg and MeHg concentrations were not positively correlated with urine THg concentrations. This can be illustrated by different metabolic pathways for each

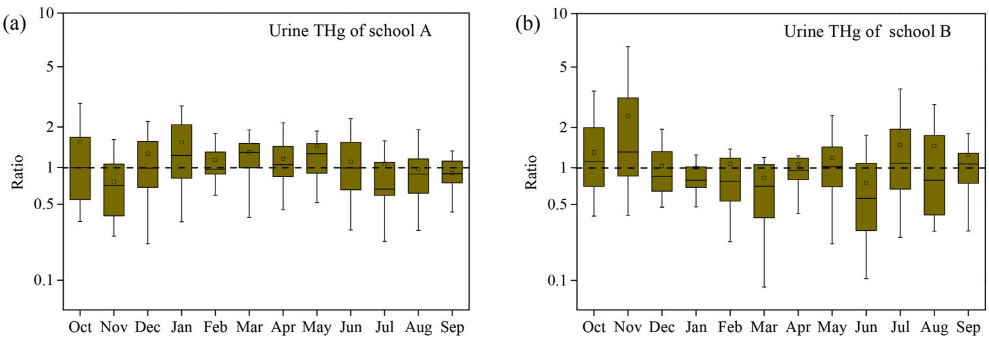

Fig. 6. Monthly variation of the ratio of urine THg, for students and their guardians from October 2013 to September 2014 in school A, and from October 2012 to September 2013 in school B. (a) urine THg of school A; (b) urine THg of school B. Each box represents interquartile range (25th and 75th percentiles), the band near the middle of the box is the 50th percentile (the median), and the whisker represents 5th and 95th percentiles.

Hg species: hair THg and MeHg concentrations are mainly related to dietary intake, while urine THg mainly reflects both atmospheric exposure to IHg and diet exposure. Previous studies have proved that a small proportion of MeHg demethylated within the liver in mammals. However in this study, our results can not demonstrate that methylmercury's demethylation from dietary exposure are the dominate source of urine Hg.

Other factors, such as age ($p = 0.53$), gender ($p = 0.25$), and smoking habit ($p = 0.89$), had no significant effect on hair THg, hair MeHg, or urine THg concentrations (Figures S3–S5) even though adults showed 16%, 5.6%, and 11% higher values than pupils in school B, respectively. Some previous studies showed that gender is unlikely to be a key factor that regulates Hg accumulation in hair (Gibb et al., 2016; Michalak et al., 2014; Okati and Esmaili-sari, 2018) or in urine (Du et al., 2016; Li et al., 2015a; McKelvey et al., 2018). However, other studies reported that males showed elevated mean values of THg concentration in hair than females in Western Colombia (Gutierrez-Mosquera et al., 2018), Wisconsin (Knobeloch et al., 2007), and Western Europe (Diez et al., 2008). The gender-related differences were explained as being due to males ingesting more fish per kg body weight than females (Diez et al., 2008), and the metabolization of MeHg, as well as hair treatments resulting in the elimination of MeHg in females (Knobeloch et al., 2007).

In this study, we found that the study population had low-level Hg exposure compared with the recommended value for hair Hg (1 μg g^{-1}) suggested by the National Research Council (NRC, 2000) and for urine THg (5 μg g^{-1} Cr) set by the United Nations Industrial Development Organization (UNIDO, 2003). The comparison of hair and urine THg from different studies with low-level Hg exposure is shown in Table 1. Generally, the amount of fish/seafood consumption in this population was much lower than those in other studies (1/5); but the population in this study consumed more rice than other previously study populations. The averages (median or geometric mean) of hair THg ranged from 145 to 790 ng g^{-1}, and urine THg ranged from 0.22 to 1.28 μg g^{-1} Cr. Although significant differences in dietary (rice and fish consumption), hair THg and urine THg in the this study were comparable with those for other residents with low-level MeHg exposure (Kobal et al., 2017; Gibb et al., 2016; Perez et al., 2019; Pino et al., 2018; Schwedler et al., 2017; Puklova et al., 2010; Tratnik et al., 2013; Imo et al., 2018; Cryderman et al., 2016; Rogers et al., 2008).

3.4. PDIs of MeHg and hair MeHg

Details of the calculation method of MeHg PDIs are shown in supplementary information (SI), and the results are shown in Table S3. Generally, the school students had slightly higher PDIs of MeHg than the adults but without any significant difference, which may due to the relatively lower body weight of young pupils. The average PDIs of MeHg via rice consumption for school A and school B students were 0.0079 and 0.0075 μg kg^{-1} d^{-1}, respectively. Their guardians had lower PDIs of MeHg via rice consumption (0.0069 and 0.0067 μg kg^{-1} d^{-1}, for school

Table 1
Low-level hair and urine Hg levels in residents around the world.

Study Site	Cohort	Average rice ingestion (g day^{-1})	Average fish/seafood ingestion (g day^{-1})	Median/mean Hair THg (ng g^{-1})	Median urine THg (μg L^{-1} Cr)	Reference
Idrija, Slovenia	Mother, children	11[a]	25	236	1.28	Kobal et al. (2017)
Sundarban and Calcutta, India	Residents	150[a]	33	725		Gibb et al. (2016)
Valencian, Spain	Children	0	218	790		Perez et al. (2019)
Italy	Children	16[a]	95[a]	538		Pino et al. (2018)
17 European countries	Mother/children	16[a]	95[a]	225/145		Schwedler et al. (2017)
Czech Republic	Children	30[a]	97[a]	180		Puklova et al. (2010)
Slovenia	Residents	11[a]	25	241	0.61	Tratnik et al. (2013)
Switzerland	Mother, children	17[a]	25[a]	160	0.22	Imo et al. (2018)
Canada	Mother/children	20[a]	137/88	180	0.5[b]	Cryderman et al. (2016)
New York, USA	Children	20[a]	29[a]		0.31	Rogers et al. (2008)
Guiyang, China	Primary student	120	8	404 ± 190	0.63 ± 0.57	This study
Guiyang, China	Middle school student	210	15	394 ± 235	0.53 ± 0.50	This study
Guiyang, China	Adult	290	18	461 ± 255	0.67 ± 0.71	This study

[a] Average per capita consumption of rice and fish/seafood were obtained from FAO (2015).
[b] Unit was μg L^{-1}.

A and school B guardians, respectively). For the PDIs of MeHg via fish consumption, no significant difference was observed among different study groups. The average PDIs of MeHg via fish consumption were 0.0013, 0.0014, 0.0015, and 0.0017 μg kg^{-1} d^{-1} for school A students, school B students, and the guardians of the school A students and school B students, respectively. Having lunch at school dining rooms did not affect PDIs of MeHg in the children via rice consumption, because there was no significant difference in rice Hg concentration between school dining rooms and students' homes. However, the lack of fish consumption in the dining rooms resulted in lower PDIs of THg/MeHg for the students compared with the adults.

The PDIs of MeHg via rice and fish consumption in this study were much lower than total diet MeHg exposure in the Chinese population (0.057 μg kg^{-1} d^{-1}; Liu et al., 2018), children (0.041 μg kg^{-1} d^{-1}) and women at childbearing age (0.031 μg kg^{-1} d^{-1}) in Hong Kong via fish consumption (Chan et al., 2018), children in Taiwan (0.007–0.03 μg kg^{-1} d^{-1}) via fish and seafood consumption (You et al., 2018), the Japanese population (0.14 μg kg^{-1} d^{-1}) via fish consumption (Zhang et al., 2009), and in adult women (0.02 μg kg^{-1} d^{-1}) in America (Mahaffey et al., 2004). This indicated a low risk of MeHg exposure via rice and fish consumption in urban residents of inland China even in an industrial area, despite serious environmental Hg contamination having been reported (Feng and Qiu, 2008).

Feng et al. (2008), Zhang et al. (2010) and Li et al. (2012) confirmed that rice consumption was the major pathway of MeHg exposure for rural residents in inland China, and that fish consumption only contributed 1–28% of the total MeHg intake due to low fish MeHg concentrations and low daily consumption amount. In this study, > 85% of PDIs of MeHg were originated from rice consumption. Therefore, this study also confirmed that even though the MeHg concentrations in fish are much higher than those in rice, rice consumption was the major route of MeHg exposure for urban residents in Guizhou province due to high daily rice intake.

The average values of total MeHg PDIs from the consumption of fish and rice were 0.0091, 0.0076, 0.0090, and 0.0083 μg kg^{-1} d^{-1} for school A students, guardians of school A students, school B students, and guardians of school B students, respectively, and none exceeded the RfD of 0.1 μg kg^{-1} d^{-1} (USEPA, 2001b). The results indicated low risk of MeHg exposure for the urban residents in Guiyang. However, it is worth mentioning that the current RfD for MeHg exposure was established based on fish consumption. Fish contains higher levels of beneficial micronutrients than rice, such as long-chain polyunsaturated omega-3 fatty acids, essential amino acids, and selenium (Li et al., 2012), which can protect against Hg toxicity (Zhang et al., 2014). A study in the Canadian Arctic showed that Se: Hg molar ratios averaged 39:1 in fish and northern shrimp (Pedro et al., 2019). Although Se concentrations in the rice and fish samples were not determined in this study, using the mean value of rice Se concentration in China (25 ± 0.011 ng g^{-1}; Chen et al., 2002) gave a calculated Se:Hg ratio of 34:1, which is lower than that in fish. Additionally, essential polyunsaturated fatty acids can also effectively decrease the bioavailability of MeHg in organisms, but the concentrations are much lower in rice (Li et al., 2012; Pedro et al., 2019). Therefore, using the current RfD of MeHg from fish consumption would underestimate the adverse effects of MeHg exposure in rice consumption populations.

No significant relationship was observed between human hair MeHg concentrations and PDIs of MeHg. However, the regression slope was 19 in this study (Fig. 7a), which was comparable with results obtained by previous studies on residents living in the Wanshan Hg mining area (Li et al., 2015b; Du et al., 2016). Li (2015b) and Du (2016) found significant positive relationships between human hair MeHg concentrations and PDIs of MeHg, with regression coefficients of 23 and 15, respectively. Rice THg concentrations in the Wanshan area were relatively high (with an average of 80 ng g^{-1} and 25% of MeHg), and rice ingestion contributed to 95% of MeHg exposure for Wanshan populations. However, rice Hg concentrations were much lower (with an average of 3.6 ng g^{-1}) and fish consumption showed higher contribution of Hg exposure in Guiyang population (Du et al., 2018a).

4. Conclusions

Although the most common pathway for human MeHg exposure is considered to be via consumption of fish and seafood, rice consumption was found to contribute significantly to human MeHg exposure in industrial and urban area of China. More than 85% of PDIs of MeHg were originated from rice consumption due to the high rice ingestion rate in inland China. It is likely that a similar phenomenon occurs in other populations around the world where rice is consumed as a staple food.

Hair and urine Hg concentrations in the study populations were slightly higher in the cold season (October to February) than the warm season (March to September), but without significant difference. This suggested that higher food intake Hg levels in the cold season may cause higher Hg exposure level. In this study, hair THg and MeHg concentrations in school B students were significantly lower than those of their guardians. The school students showed significantly higher urine THg concentrations than adults, which is due to children's unique physiology structure and behavior. Therefore, more attentions should be paid on children as a susceptible population. Both the PDIs and hair/urine Hg concentrations indicated low risks of Hg exposure for urban residents in inland China, although serious environmental Hg contamination has been reported in China.

Credit author statement

Buyun Du: Methodology, Software, Data curation, Writing- Original draft preparation, Writing- Reviewing and Editing. **Ping Li:** Conceptualization, Methodology, Supervision, Writing- Reviewing and Editing. **Xinbin Feng:** Conceptualization, Supervision, Writing- Reviewing and

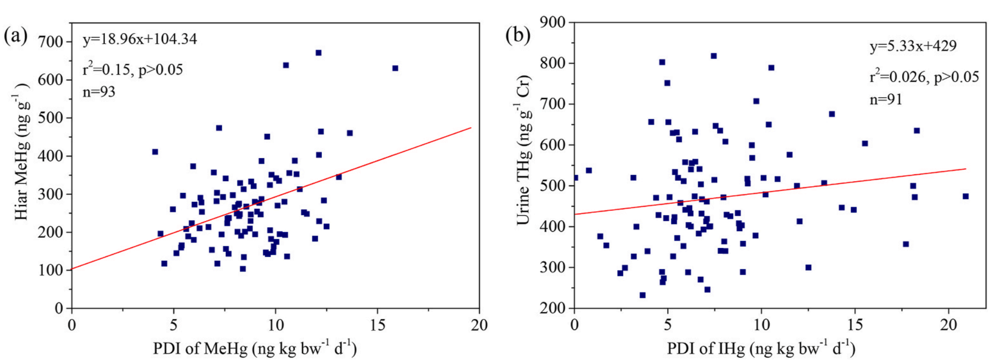

Fig. 7. Relationship between hair MeHg concentrations and PDI of MeHg (a) and urine THg and PDI of IHg (b) through rice and fish consumption. a, hair MeHg VS total MeHg intake; b, Urine THg VS IHg intake.€. including p-values for Pearson's correlation (n = 91) (each scatterplot was obtained from the annual average).

Editing. **Runsheng Yin:** Supervision, Writing- Reviewing and Editing. **Jun Zhou:** Writing- Reviewing and Editing. **Laurence Maurice:** Conceptualization, Methodology, Supervision, Writing- Reviewing and 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

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