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Mercury contents in rice and potential health risks across China

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ABSTRACT

Rice samples were collected at 560 sites in 15 provinces across China in areas without known point mercury (Hg) sources. Total mercury (THg) and methylmercury (MeHg) concentrations were analyzed in these rice samples for risk assessment. Relatively low THg and MeHg concentrations were found in the majority of the white rice samples with an overall mean of 4.74 (1.06–22.7) μg kg^{-1} and 0.682 (0.03–8.71) μg kg^{-1}, respectively. The means (range of) THg concentration of rice in each geographical region were 5.23 (1.07–19.5) μg kg^{-1}, 5.14 (1.06–17.2) μg kg^{-1}, 4.45 (1.41–17.2) μg kg^{-1}, 4.20 (1.48–19.4) μg kg^{-1}, 3.49 (1.49–10.7) μg kg^{-1}, and 4.53 (1.30–19.4) μg kg^{-1} in east, centre, south, southwest, northwest and northeast, China, respectively, and the corresponding values for MeHg concentrations were 0.898 (0.127–8.35) μg kg^{-1}, 0.603 (0.207–2.48) μg kg^{-1}, 0.516 (0.032–1.50) μg kg^{-1}, 0.615 (0.050–5.03) μg kg^{-1}, 0.704 (0.148–2.41) μg kg^{-1} and 0.565 (0.035–8.71) μg kg^{-1}, respectively. Hg contents in rice across China were found to be at background levels. Both the probable daily intakes (PDIs) of inorganic Hg (IHg) and MeHg from rice consumption showed low risks for general population in the investigated regions.

1. Introduction

Mercury (Hg) is a toxic metal characterized by its persistence, bioaccumulation and neurotoxicity, which even at low exposure may cause irreversible damage to human and mammalian central nervous systems (Clarkson and Magos, 2006). In many geographical regions globally, consumption of fish and other aquatic products is the major exposure of Hg to humans (Zahir et al., 2005; Mergler et al., 2007). However, consumption of other food products containing Hg may also induce health risks to humans especially in areas with high air and soil Hg concentrations. For example, high Hg concentrations (up to 1280 μg kg^{-1}) were found in rice grown in Hg mining areas of southwest China (Qiu et al., 2008), which may pose a great risk of Hg exposure to the populations using the locally produced rice as the major food supply in south China (Zhang et al., 2010a).

China is currently the largest emitter of anthropogenic atmospheric Hg worldwide. Such emissions include a substantial fraction of oxidized and particle-bound Hg, which can deposit fast to nearby areas of the sources regions, causing elevated Hg loading in many ecosystems (Streets et al., 2009, 2011; Zhang et al., 2015; Zhang et al., 2016). Since the elevated Hg deposition from anthropogenic sources tends to concentrate in labile pools (Fu et al., 2012; Smith-Downey et al., 2010), the potential for chemical transformation (e.g., methylation) of deposited Hg has increased. In China, rice production accounts for 40% of total grain production and > 60% of Chinese residents feed on rice (NBS, 2015). Therefore, investigating Hg contents in rice has important implications for human health risk assessments in China as well as in the other parts of the world having similar situations.

Existing studies on rice contents of Hg were mostly focused on mining or contaminated areas (Feng et al., 2008; Qiu et al., 2008; Meng et al., 2010; Rothenberg et al., 2017; Zhao et al., 2016; Yin et al., 2013). Horvat et al. (2003) reported that rice seeds collected from Wanshan Hg mining area contained high levels of total Hg (THg) and methylmercury (MeHg) (THg, 569 μg kg^{-1}; MeHg, 145 μg kg^{-1}). Zhang et al. (2010b) specified that rice grain accumulates MeHg > 800 times higher than inorganic mercury (IHg). Xu et al. (2017) concluded that Hg emissions from coal-fired power plants may affect Hg accumulation in rice grown in the vicinity of the plants. Li et al. (2017) used MeHg compound specific stable isotope analysis to trace human dietary exposure to MeHg in the Wanshan Hg mining area in southwest China. To date, very limited studies have focused on THg and MeHg in rice in non-polluted areas (Li et al., 2012; Shi et al., 2005). However, it should be noted that elemental Hg occupies the majority of the mass of atmospheric Hg, has a long life time of six months or longer, and can thus be transported on global scale, which has potential to affect areas far away from point sources.

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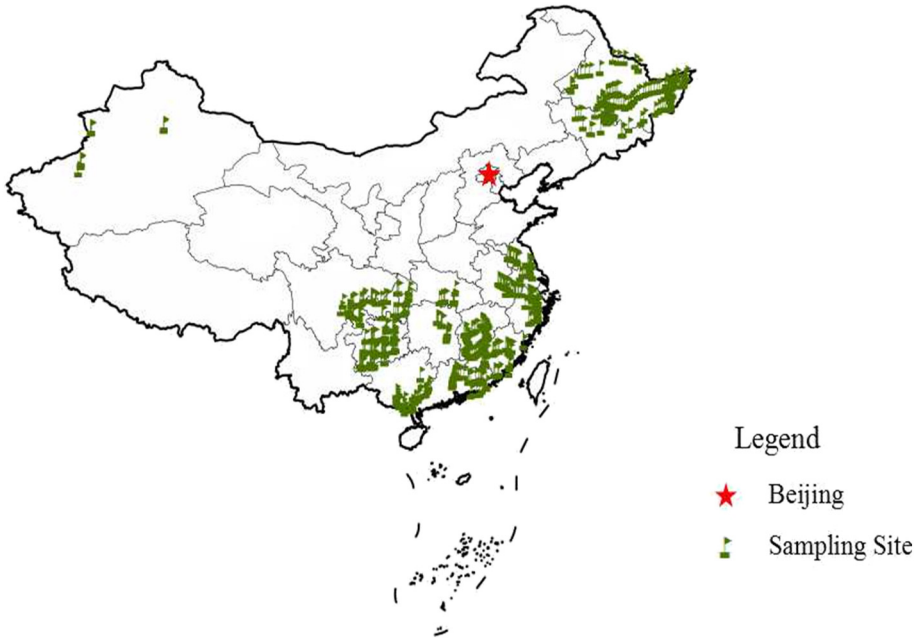

Fig. 1. Spatial distribution of sampling sites in this study.
XJ - Xinjiang, HLJ - Heilongjiang, JL - Jilin, ZJ - Zhejiang, JS - Jiangsu, JX - Jiangxi, AH - Anhui, HB - Hubei, HN - Hunan, FJ - Fujian, GX - Guangxi, GD - Guangdong, GZ - Guizhou, SC - Sichuan, and CQ - Chongqing.

The aim of the present study is to conduct a large-scale survey of Hg contents in rice across most major rice growing areas in China and to use these data to evaluate health risks of MeHg exposure via rice consumption for those populations taking rice as the main food supply.

2. Materials and method

2.1. Sample collection

A total of 560 rice samples were collected on-site from major rice growing areas in 15 provinces of China during the harvest time (September–October) of either 2014 or 2015 (Fig. 1). These provinces were geographically categorized into east: Zhejiang (ZJ), Jiangsu (JS), Anhui (AH), Jiangxi (JX) and Fujian (FJ); centre: Hubei (HB), and Hunan (HN); south: Guangxi (GX), and Guangdong (GD); southwest: Guizhou (GZ), Sichuan (SC), and Chongqing (CQ); northwest: Xinjiang (XJ); northeast: Heilongjiang (HLJ) and Jilin (JL) regions. The sampling areas covered in this study accounted for 89% of the total rice growing areas in China, and 88.6% of rice yield. Detailed information is shown in Table 1.

There is one sample at each site and three strains of rice grain were randomly collected from the same field and were pooled together as one sample, which was washed immediately with tap water for several times before being double-bagged. All the rice grain samples were rigorously rinsed with deionized water when brought back to the laboratory and left to dry in a clean environment. Rice grain samples were divided into three fractions: hull, bran, and white rice. Subsequently, the white rice samples were crushed into 150 meshes and stored double-bagged in the dark for further Hg analysis.

2.2. Analytical method

For THg analysis, 0.2–0.3 g of a milled sample was digested by HNO_3 in a water bath held (95 °C) for 3 h. An aliquot was analyzed for Hg by cold air atomic fluorescence spectrometry (CVAFS) on a Hg analyzer (Tekran Model 2500) following BrCl oxidation, $SnCl_2$

Table 1
Basic information of rice sampling sites in different provinces.

Province	Sampling Date	Sampling site	Rice producing area/Hectare	Percentage in national producing area/%	Rice Yield/Mt	Percentage in national rice yield/%
HLJ	2015/10	78	3205.5	10.6	2251	10.8
JL	2015/10	15	747.1	2.5	587.6	2.8
XJ	2015/10	21	75.1	0.3	76.2	0.4
ZJ	2015/10	57	824.2	2.7	590.1	2.8
JS	2015/10	44	2271.7	7.5	1912	9.2
JX	2015/10	39	3339.5	11	2025.2	9.7
AH	2014/09	25	2214.1	7.3	1394.6	6.8
HB	2014/09	21	4085	13.5	2634	12.8
HN	2014/09	41	2101.2	6.9	1729.5	8.4
FJ	2015/10	36	804.5	2.7	497.1	2.4
GX	2014/10	28	2046.6	6.8	1166.1	5.6
GD	2015/10	71	1893.3	6.3	1091.6	5.2
GZ	2015/09	30	682	2.3	403.2	1.9
SC	2015/09	36	1991.8	6.6	1526.5	7.3
CQ	2015/09	18	689.7	2.3	503.2	2.4
Total	2014/09, 2015/10	560	26,971.3	89	18,387.9	88.6

Data were obtained from the website: http://www.stats.gov.cn.

reduction, purge-and-trapping, and eventually thermal desorption (USEPA Method 1631 (2002)).

For MeHg analysis, approximately 0.5 g of a rice sample was digested in a 25% KOH-CH_3OH solution at 75 °C for 3 h. Upon completion, the digestions were acidified by addition of concentrated HCl. MeHg in the digestion was leached with dichloromethane (CH_2Cl_2) and back-extracted into the water phase. After derivatization by an ethylating agent ($NaBEt_4$), volatile Hg species were separated from solution by purging with N_2 onto a Tenax trap. The trapped methylethyl was then thermally desorbed, separated from other Hg species by an isothermal gas chromatography (GC) column, decomposed to Hg^0 in a pyrolytic decomposition column (800 °C) and analyzed by CVAFS (Brooks Rand Model III, Brooks Rand Laboratories, Seattle, WA) (USEPA Method 1630 (2001a)).

2.3. Quality assurance and quality control

Quality control measures consisted of method blanks, triplicates, matrix spikes, and several certified reference materials. The limits of detection were 0.01 μg kg^{-1} for THg and 0.002 μg kg^{-1} for MeHg in rice samples, respectively. The certified reference materials of GBW10020 (Orange foliage) and TORT-2 (Lobster Hepatopancreas) were used in this study for THg and MeHg analysis and the average recoveries were 103.5% and 90.5%, respectively. The recoveries for matrix spikes averaged at 103% (with a range of 96–110%) for THg analysis, and 109% (with a range of 83–120%) for MeHg. The relative standard deviation (RSD) of duplicate analysis for Hg concentration data averaged at 6.1% with a range of 2.6%–8.7%.

2.4. Calculation of probable daily intake (PDI)

To estimate MeHg intake from rice consumption, we calculated PDI values of MeHg for the general adult population in the investigated 15 provinces according to the following equation:

$$PDI = (C \times IR \times A)/bw \quad (1)$$

where PDI is given in μg kg^{-1} d^{-1}, bw is the body weight set at 60 kg, C is MeHg or IHg concentration in rice (μg kg^{-1}), IR is daily intake rate (kg d^{-1}), and A is the absorption rate by human body, which is 7% for IHg and 95% for MeHg, respectively (WHO, 1990). IHg concentration in rice is its THg concentration minus MeHg concentration. IR was estimated for each province from the Survey on Nutrition and Health Status of Chinese Citizens in Different Provinces (Municipalities and Autonomous Regions) in 2002. The PDIs of Hg via rice consumption presented in the study based on provincial average. Hg exposure via fish intake for local residents were not included in this study.

2.5. Statistical analysis

Statistical analysis was performed using SPSS 22.0 software and graphs of Hg contents were plotted using Origin (Version 9.0). The Hg measurements in samples are generally described by giving the mean ± standard deviation (SD) or geometric mean, which depends on whether the data was in a normal distribution. Relationships between covariant sets of data were subjected to regression analysis. Correlation coefficients (r) and significance probabilities (p) were computed for the linear regression fits. Differences are declared as significant in case that $p < 0.05$.

3. Results and discussion

3.1. THg concentration in rice

The mean THg concentration in the rice samples from each province is shown in Fig. 2. A log-normal distribution was found from the 560 samples with the geometric mean of 4.74 μg kg^{-1} and a range of 1.06–22.7 μg kg^{-1}. 85% of rice samples had THg concentration in the range of 1.0–10.0 μg kg^{-1}. The geometric mean concentration for individual provinces was in the range of 3.44–6.11 μg kg^{-1} whilst the corresponding median value in the range of 2.69–6.39 μg kg^{-1}. Analysis of variance showed that there were no significant differences in THg concentrations between different provinces ($p > 0.05$, n = 560). Only two out of the 560 rice samples (0.3%) had THg concentration exceeding national limit for THg in cereals (20 ng·g^{-1}) set by the Standardization Administration of China. However, these two values were significantly higher than rice THg concentrations found in the adjacent areas, which indicated serious Hg pollution. In order to facilitate statistical analysis, these two points were treated as outliers and do not participate in the following discussion.

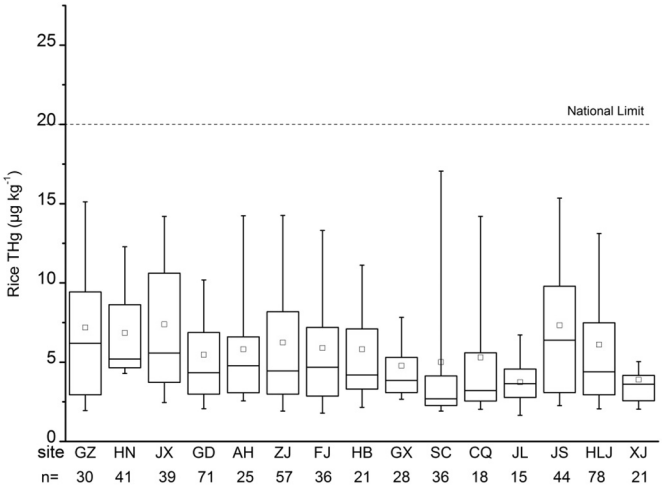

Fig. 2. Comparison of THg concentrations in rice samples collected from different provinces in China.
Each box represents the interquartile range (25th to 75th quantiles), the band near the middle of the box is the 50th percentile (the median), the whisker spans from the 10th to 90th quartiles, and open squares indicate average values.

Comparisons of rice Hg concentrations between this and previous studies were shown in Table 2. To date, the majority of the studies reporting Hg concentrations in rice were conducted in China, especially in Hg mining areas. Results of THg concentrations found in the present study are comparable to several earlier studies (3.4–7.0 μg kg^{-1}) conducted in non-polluted areas in China (Song et al., 2011; Zhang et al., 2010b, 2014), but much lower than those (10–569 μg kg^{-1}) found in the Hg mining areas in China (Feng et al., 2008; Horvat et al., 2003; Li et al., 2012; Qiu et al., 2012, 2013). Note that the study by Song et al. (2011) covered six provinces of south China (THg concentration of 4.8 μg·kg^{-1}) while that by Zhang et al. (2014) covered 15 provinces of China (3.4 μg kg^{-1}). However, Yuan et al. (2011) collected the rice in the market in 2010 instead of from direct field sampling and found THg concentration of 9.5 ± 8.7 μg kg^{-1}, which had no significant difference from the present study. Overall, the THg contents in rice in non-polluted areas of China have not changed much when comparing studies conducted in different years. Compared to several limited studies outside China, THg concentrations in rice were slightly higher in China than in the other countries, e.g., 3.04 ± 2.07 μg kg^{-1} in Europe (Brombach et al., 2017) and 2.91 ± 0.86 μg kg^{-1} in Republic of Korea (Eom et al., 2014).

The means (range of) THg concentration of rice in each geographical region 5.23 (1.07–19.5) μg kg^{-1}, 5.14 (1.06–17.2) μg kg^{-1}, 4.45 (1.41–17.2) μg kg^{-1}, 4.20 (1.48–19.4) μg kg^{-1}, 3.49 (1.49–10.7) μg kg^{-1}, and 4.53 (1.30–19.4) μg kg^{-1} in east, centre, south, southwest, northwest and northeast region, respectively (Fig. 3). The significant differences in the THg concentration in rice was found between the two group regions (east and northwest; centre and northwest) ($p < 0.05$, n = 560). The average THg concentration in rice grain was

Table 2
Comparison of THg and MeHg concentrations in rice from different studies.

Locations	Hg Pollution?	THg (μg kg^{-1}) mean (range of)	MeHg (μg kg^{-1}) mean (range of)	Reference
China (background)	No	4.74(1.06–22.7)	0.682 (0.03–8.71)	This Study
Various sources (83 rice products sold in EU)	No	3.04(0.53–11.1)	1.91(0.11–6.45)	Brombach et al. (2017)
Hubei, China	No	3.7(1.9–6.8)	1.2(0.33–3.0)	Rothenberg et al. (2011)
Guizhou, China	No	6.2(3.8–10.3)	2.9(1.8–4.5)	Meng et al. (2010)
Guizhou, China	No	50.8(7.4–194)	5.8(1.4–24)	Zhang et al. (2010a)
Guizhou, China	No	29.7(9.6–79)	4.7(1.6–13)	Zhang et al. (2010a)
Kampong, Cambodia	No	8.14(6.16–11.7)	1.44(1.17–1.96)	Cheng et al. (2013)
Kandal, Cambodia	No	10.2(5.91–15.1)	2.34(0.48–5.23)	Cheng et al. (2013)
Hubei, China	No	3.3(1.7–6.5)	1.2(0.57–3.1)	Rothenberg et al. (2011)
Guizhou, China	No	7(3.2–15.1)	2.5(0.8–4.3)	Feng et al. (2008)
Guizhou, China	No	2.75(2.1–3.4)	1.3(0.6–2.0)	Li et al. (2008)
Guizhou, China	No	2.8(1.0–5.5)	2.0(0.37–3.3)	Rothenberg et al. (2012)
7 provinces , China	No	10.1(0.86–47.2)	2.47(0.13–18.2)	Li et al. (2012)
15 provinces , China	No	23(6.3–39.3)	4.7(1.9–10.5)	Shi et al. (2005)
Wanshan, China	Yes, Hg mining	50.81(2. 83–295. 53)	13.20(2.3–43.18)	Yin et al. (2013)
Guizhou, China	Yes, Hg mining	149(11.1–569)	38.9(8.03–144)	Horvat et al. (2003)
Guizhou, China	Yes, Hg mining	26(13–52)	9.4(3.5–23)	Li et al. (2013)
Hunan, China	Yes, Hg mining	29(11–58)	11(6.5–24)	Li et al. (2013)
Guizhou, China	Yes, Hg mining	187(24.8–548)	7.0(3.8–18)	Meng et al. (2010)
Guizhou, China	Yes, Hg mining	302(128–622)	32(18–62)	Meng et al. (2010)
Wanshan, China	Yes, Hg mining	96.3 ± 14.2	30.4 ± 26.5	Meng et al. (2014)
Danzhai, China	Yes, Hg mining	85.8 ± 43.6	20.8 ± 9.3	Meng et al. (2014)
LMC. China	Yes, Hg mining	22.0 ± 5.3	12.3 ± 4.9	Meng et al. (2014)
Guizhou, China	Yes, Hg mining	228(10.3–1120)	25.3(1.61–174)	Qiu et al. (2008)
Guizhou, China	Yes, Hg mining	105(8.6–504)	11(1.2–44)	Zhang et al. (2010a)
Guangdong, China	Yes, Pb–Zn mining	15(1.5–52)	1.2(0.28–3.5)	Li et al. (2013)
Guangdong, China	Yes, Pb–Zn mining	16.0 ± 4.1	2.0 ± 0.5	Meng et al. (2014)
Guizhou, China	Yes, wastewater irrigation	14.5(2.53–33.4)	11.3(0.71–28)	Horvat et al. (2003)
Tianjin, China	Yes, wastewater irrigation	12.80 ± 5.14	2.09 ± 1.20	Zheng et al. (2015)
Guizhou, China	Yes, Hg mining	58.5(21.1–191.9)	14.6(7.5–27.6)	Feng et al. (2008)
Guizhou, China	Yes, Hg mining	21.3(10–66.9)	5.7(3.3–10.2)	Feng et al. (2008)
Guizhou, China	Yes, Hg mining	33.1(4.9–214.7)	4.0(1.9–14.7)	Feng et al. (2008)
Wanshan, China	Yes, Hg mining	42.4 ± 52.7	11.7 ± 8.84	Li et al. (2015)
Guizhou, China	Yes, Hg mining	26.8(6–113)	7.8(3.8–12.3)	Li et al. (2008)
Shanxi, China	Yes, Hg mining	103(51–200)	22(8.2–80)	Qiu et al.,(2012)
Guizhou, China	Yes, Hg mining	22(10–45)	11(3.2–39)	Qiu et al. (2013)
Guizhou, China	Yes, Hg mining	120(72–190)	63(31 – 100)	Rothenberg et al. (2012)
Guizhou, China	Yes, Hg mining	18(11–34)	9.9(1.5–18)	Rothenberg et al. (2012)
Guizhou, China	Yes, Hg mining	12(2.4–3.8)	5.0(1.2–13)	Rothenberg et al. (2013)
Kratie, Cambodia	Yes, Gold mining	12.7(9.90–16.7)	1.54(1.06–2.31)	Cheng et al. (2013)

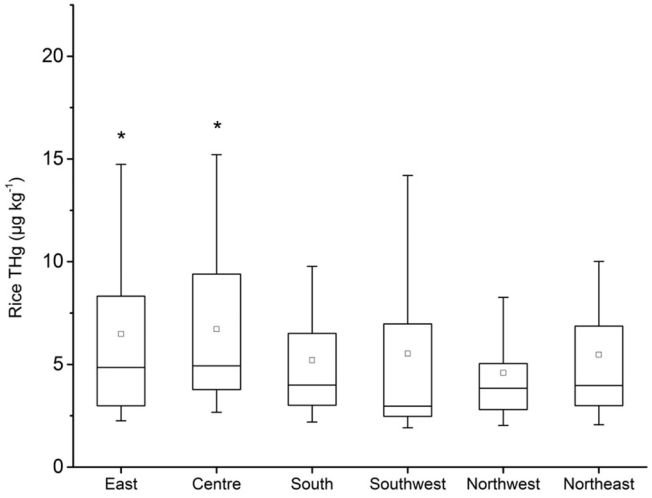

Fig. 3. Comparison of THg concentrations in rice among different regions.
*, $p < 0.05$, compared with northwest.
Each box represents the interquartile range (25th to 75th quantiles), the band near the middle of the box is the 50th percentile (the median), the whisker spans from the 10th to 90th quartiles, and open squares indicate average values.

the lowest in the northwest region (3.89 ± 2.11 μg kg^{-1}, geometric mean 3.49 μg kg^{-1}) and the highest (6.60 ± 4.72 μg kg^{-1}, geometric mean 5.23 μg kg^{-1}) in the east. Hg soil contents derived from parent rock weathering showed increasing gradients from north to south and from west to east in China (Wen, 2007). In particular, the arid soils of northwestern China displayed lower THg content than the background level elsewhere. On the other hand, ZJ and AH are located in east China that has gone through rapid industrialization and urbanization. As such, the amount of Hg emissions was higher, which was reflected in higher annual Hg dry deposition rate of 35 mg m^{-2} yr^{-1} in eastern China (Wang et al., 2014). This could have contributed to higher Hg concentrations in rice from this region.

In the study of Qian et al. (2010), the rice areas of China were divided into north, south, centre and southwest regions, and the geometric mean of THg concentrations was 2.5 μg kg^{-1}, 2.7 μg kg^{-1}, 3.8 μg kg^{-1} and 3.8 μg kg^{-1}, respectively in these four regions, which showed no significant regional differences ($p < 0.05$).

3.2. MeHg concentration in rice

In this study, we selected 115 rice samples for MeHg analysis, and these rice samples covered low, medium, and high THg contents in each provinces to ensure the representativeness (Fig. 4a). The overall MeHg concentration geometric mean was 0.682 (range of 0.035–8.71) μg kg^{-1}, similar to that (1.9–10.5 μg kg^{-1}) reported by Shi et al. (2005) for 15 provinces, although the overall mean value in the present study

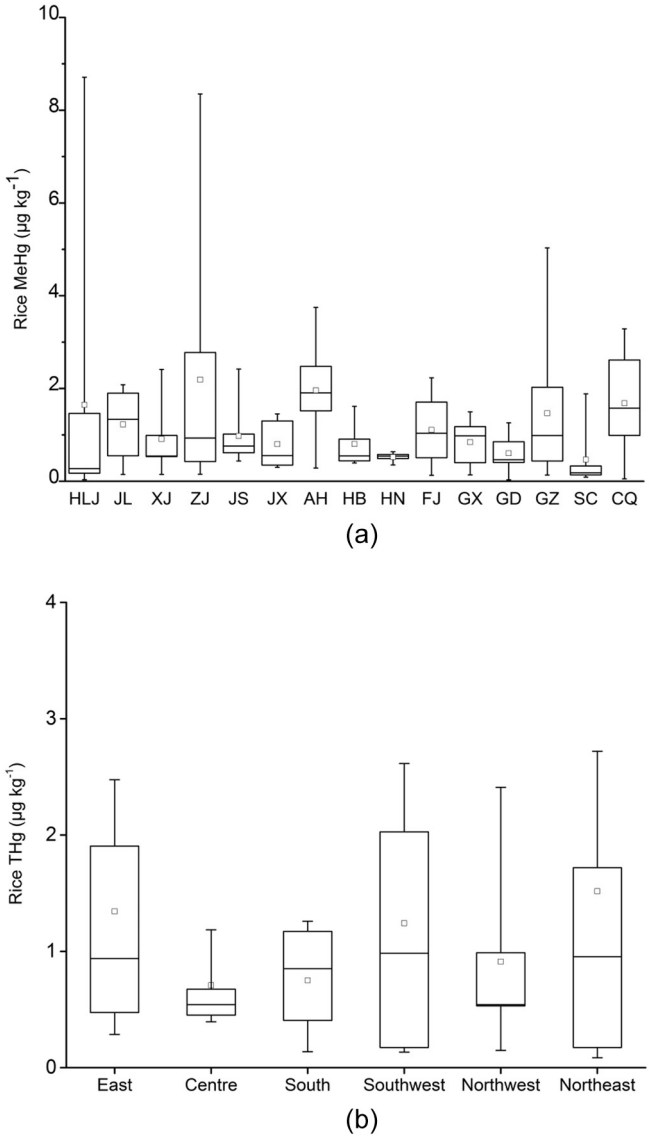

Fig. 4. Comparison of MeHg concentrations in rice collected from different provinces (a) and regions (b).
Each box represents the interquartile range (25th to 75th quantiles), the band near the middle of the box is the 50th percentile (the median), the whisker spans from the 10th to 90th quartiles, and open squares indicate average values.

(1.06 μg kg^{-1}) was three times lower than that reported in Shi et al. (2005) (4.7 μg kg^{-1}). Statistical results demonstrated that MeHg in rice had no significant regional differences ($p > 0.05$).

Compared with earlier studies (Table 2), the mean rice MeHg concentrations found in the present study were in a similar range of that reported for the Hubei province where without known Hg point sources (Rothenberg et al., 2011), and those reported in an area of gold mine (Kratie, Cheng et al., 2013) and Pb−Zn mine (Guangdong, Li et al., 2013).

Observed MeHg concentrations in the main rice growing areas in the east, centre, south, southwest, northwest and northeast China were 0.898 (0.127–8.35) μg kg^{-1}, 0.603 (0.207–2.48) μg kg^{-1}, 0.516 (0.032–1.50) μg kg^{-1}, 0.615 (0.050–5.03) μg kg^{-1}, 0.704 (0.148–2.41) μg kg^{-1} and 0.565 (0.035–8.71) μg kg^{-1}, respectively. (Fig. 4b).

According to the previous research, the main source of Hg in rice is atmosphere and soil. Serious soil Hg pollution was confirmed in China, and THg concentrations in 16.1% of surveyed sites exceeded national limit. However, in this study, it showed relative level of Hg content in rice samples across China. Li et al. (2017) confirmed that 30% of IHg in rice grain originated from the atmosphere. Unlike IHg, Meng et al. (2011) found that MeHg in soil can easily penetrate the iron membrane "barrier" of rice roots into the rice and can be transported to the aerial parts. Zhao et al. (2016) found that MeHg in paddy soils in Hg mining areas is mainly derived from methylation of IHg in the soils, while MeHg from external inputs (atmospheric dry/wet deposition, irrigation water) is negligible.

Table 3
Rice Hg contents, daily intake, and PDIs of MeHg and IHg through rice consumption.

Province	C (μg kg^{-1})		Daily intake rate (kg d^{-1})	PDI (μg kg^{-1} d^{-1})	
	MeHg	IHg		MeHg	IHg
HLJ	1.65	4.68	0.217	0.0057	0.0012
JL	1.22	3.70	0.319	0.0062	0.0014
XJ	0.91	4.05	0.081	0.0012	0.0004
ZJ	2.19	8.07	0.303	0.0105	0.0029
JS	0.98	8.14	0.253	0.0039	0.0024
JX	0.80	7.31	0.410	0.0052	0.0035
AH	1.96	7.53	0.431	0.0134	0.0038
HB	0.80	6.78	0.307	0.0039	0.0024
HN	0.52	8.25	0.371	0.0031	0.0036
FJ	1.11	6.21	0.337	0.0059	0.0024
GX	0.84	4.06	0.363	0.0048	0.0017
GD	0.60	10.85	0.348	0.0033	0.0044
GZ	1.46	8.41	0.334	0.0078	0.0033
SC	0.47	7.70	0.252	0.0019	0.0023
CQ	1.68	5.86	0.294	0.0078	0.0020

3.3. Human health assessment

Li et al. (2006) used the provisional tolerable weekly intake (PTWI) value recommended by the World Health Organization to evaluate the THg intake in men's diets in four regions of China and the results showed that the THg intake in Chinese people are safe. In China, the daily intake of rice is relatively high, as well rice is more likely to be enriched in MeHg than other crops, which is generally 10 to 100 times higher (Meng et al., 2011). As we know, IHg and MeHg have completely different exposure pathway and metabolic process uptake in human body and MeHg is more toxic than IHg for human beings, therefore we use rice MeHg and IHg data for risk assessment.

The average PDI of MeHg and IHg through rice consumption in these 15 provinces is shown in Table 3. The provincial average PDI of IHg from rice consumption for adult population averaged at 0.0025 ± 0.0011 μg kg^{-1} d^{-1} (with a range of 0.0004–0.0044 μg kg^{-1} d^{-1}). The maximum value was 0.0044 μg kg^{-1} d^{-1}, which was 200 times lower than limit of 0.57 μg kg^{-1} d^{-1} set by World Health Organization (JECFA, 2003). And the provincial average PDI of MeHg from rice consumption for adult population ranged from 0.0012 to 0.0134 μg kg^{-1} d^{-1} with a 15 province overall average value of 0.0056 μg kg^{-1} d^{-1}, which was far below the limits set by World Health Organization (JECFA, 2003) and USEPA (USEPA, 2001b) of 0.23 and 0.10 μg kg^{-1} d^{-1}, respectively. It can be concluded that the rice samples in this study showed relative low Hg levels and human health risks.

Some researchers have conducted risk assessment of Hg exposure via rice consumption in non-polluted areas. Huang (2012) found that MeHg in rice collected from Henan Province is very low and rice consumption will not affect residents' health. Yuan et al. (2011) assessed health risks of Hg exposure via rice consumption in nine different provinces and cities in China, where rice consumption only contributed 1.1–6.9% of PTWI. Guo et al. (2018) analyzed the risk of dietary intake of Hg in Jilin Province and MeHg concentrations in rice were under the LOD (0.002 mg kg^{-1}). Shi et al. (2005) showed that rice MeHg concentrations ranging from 1.9 to 10.5 μg kg^{-1} in 15 provinces of China

and calculated the low risks for human MeHg exposure. Zhang et al. (2014) assessed the mean values of THg weekly intakes via milled rice and brown rice consumption for different age categories were in the range of 0.09–0.19 μg kg^{-1} bw^{-1} and 0.14–0.27 μg kg^{-1} bw^{-1}, respectively, suggesting relatively low health risks. Even in Hg polluted areas, the weekly intakes of MeHg via rice consumption in a sewage irrigation area in Tianjin were 0.0095–0.49 μg kg^{-1} bw^{-1}, which were still at a safety threshold for residents' health risks (Zheng et al., 2015). In many geographical regions globally, seafood is a major human exposure pathway of Hg. Seafood consumption accounts for about 95% of MeHg exposure to humans (Houserová et al., 2007). Tian (2011) found that the mean PDI of fish in the Pearl River Delta Region was 0.046 μg kg^{-1} d^{-1}, accounting for 4.2% of the MeHg exposure RfD (reference dose) value set by the USEPA, and is equivalent to 20% of the PTWI (provisional tolerable weekly intake) value set by the WHO/FAO. Daily exposure to MeHg is within the safety limits so the health risk of MeHg exposure is low. Liu et al. (2013) conducted an assessment of MeHg exposure among four marine species in Yongxing Island. The results showed that long-term consumption of carnivorous fish may pose some health risks to avid consumers. Zhang et al. (2010a) found that MeHg intake did not exceed the dietary reference dose of 0.23 μg kg^{-1} d^{-1} as recommended by JECFA (2003), but there were 34% PDIs of samples exceeding the USEPA recommended values of 0.1 μg kg^{-1} d^{-1} for MeHg (USEPA, 1997). In addition, Li et al. (2012) reported the PDIs through rice and fish consumption in six provinces in south China and found general populations being at low risk of MeHg exposure with rice as the dominant source for inland communities. Li et al. (2015) calculated the MeHg exposure of rice and fish on the premise of the same intake amount, and the health risk of eating rice was greater.

MeHg is more soluble than IHg and elemental Hg in lipids and can be easily absorbed by mammals and has a long half-life in the organism. Using a rice paddy biogeochemical cycle model of Hg together with a global scale atmospheric chemistry transport model for Hg, Kwon et al. (2018) revealed the link between industrial activity and coal combustion and rice Hg pollution and pointed out that different policy options will affect the future of rice Hg content. And World's atmospheric Hg emissions and atmospheric Hg levels are decreasing (Zhang and Jaeglé, 2013; Wu et al., 2016; Fu et al., 2015). Kwon et al. (2018) predicted that IHg and MeHg concentrations in China rice will increase by 13% under future scenarios of no policy of Hg emission control. However, the human mercury exposure via rice consumption in non-polluted areas in China is still within the safety threshold. It should be noted that even if the concentration of MeHg in food meets the safety standard, it might be hazardous to the human central nervous systems in the long run (NRC, 2000).

4. Conclusions

This study provides the first large-scale survey and potential risk assessment of Hg and MeHg in rice harvested and consumed in China. The measured levels of THg and MeHg in rice samples are generally low. It can be concluded that the general population of China is at low risks both for MeHg and IHg exposures via the rice based diet. Future studies should take into account the contributions to PDIs from other pathway, such as consumption of fish and other aquatic food.

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