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Methylmercury and inorganic mercury in Chinese commercial rice: Implications for overestimated human exposure and health risk[☆]

Xiaohang Xu [a, b, 1], Jialiang Han [a, b, 1], Jian Pang [c], Xun Wang [a], Yan Lin [d], Yajie Wang [e], Guangle Qiu [a, *]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China
[b] University of Chinese Academy of Sciences, Beijing, 100049, China
[c] Guizhou Normal University, Guiyang, 550001, China
[d] Norwegian Institute for Water Research, Oslo, 0349, Norway
[e] College of Food Safety, Guizhou Medical University, Guiyang, 550025, China

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ABSTRACT

China is the largest rice producer and consumer in the world, and mercury (Hg) levels, particularly methylmercury (MeHg), in rice and health exposure risks are public concerns. Total Hg (THg) and MeHg levels in 767 (domestic = 709 and abroad = 58) Chinese commercial rice were investigated to evaluate Hg pollution level, dietary exposures and risks of IHg and MeHg. The mean rice THg and MeHg levels were 3.97 ± 2.33 μg/kg and 1.37 ± 1.18 μg/kg, respectively. The highest daily intake of MeHg and IHg were obtained in younger groups, accounted for 6% of the reference dose-0.1 μg/kg bw/day for MeHg, 0.3% of the provisional tolerance week intake-0.571 μg/kg bw/day for IHg. Residents in Central China and Southern China meet the highest rice Hg exposure, which were more than 7 times of those in Northwest China. Lower concentrations than earlier studies were observed along the implementations of strict policies since 2007. This may indicate that a declining temporal trend of Hg in Chinese grown rice and associated exposures could be obtained with the implementations of strict policies. Though there exist Hg pollution in commercial rice, Hg levels in Chinese commercial rice is generally safe compared with Hg polluted sites. Populations dwelling in China have relatively a quite low and safe MeHg and IHg exposure via the intake of commercial rice. Strict policies contributed to the decrease in THg and MeHg levels in Chinese-grown rice. More attention should be paid to younger groups.

1. Introduction

Mercury (Hg) pollution in rice has become an emerging topic of concern since the beginning of this century. In 2003, Horvat and colleagues firstly observed high MeHg levels in rice collected from the Wanshan mercury mine in southwestern China, the world's third largest mercury mine (Horvat et al., 2003). Since that time, researchers have realized that rice paddies are hot spots of Hg methylation, and rice has a strong capability to bioaccumulate methylmercury (MeHg) from rice paddies (Qiu et al., 2008). Subsequent studies have also found that rice consumption constitutes >94% of the MeHg exposure for residents in Guizhou province, southwestern China, who seldom eat fish (Zhang et al., 2010a). With the knowledge that rice intake is an important human MeHg exposure source in polluted areas (Feng et al., 2008), public concern has risen in recent years, mainly because rice is the staple food of more than half of the world's population (FAOSTAT, 2019). Hg in rice has been highlighted by the United Nations Environment Programme (UNEP), World Health Organization (WHO) and a large number of other international and national organizations. Simultaneously, a large number of scientists have started to study Hg biogeochemistry in rice plants and rice paddies (Krupp et al., 2009; Liu et al., 2019a; Rothenberg and Feng, 2012; Rothenberg et al., 2014; Strickman and Mitchell, 2017; Windham-Myers et al., 2014; Xu et al., 2016).

The provisional tolerable weekly intake (PTWI) for inorganic Hg (IHg) is 4 μg/kg body weight (bw) (EFSA, 2012). The U.S.

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* Corresponding author. State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, 99 Lincheng West Rd, Guiyang, 550081, China.
E-mail address: qiuguangle@vip.skleg.cn (G. Qiu).
[1] Xiaohang Xu and Jialiang Han contributed equally to this study.

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Environmental Protection Agency (USEPA) proposed 0.1 μg/kg bw/day as a reference dose (RfD) for MeHg (Rice et al., 2000). The Joint FAO/WHO Expert Committee on Food Additives (JECFA) recommended a PTWI of 4 μg/kg bw for IHg (JECFA, 2010). Thus, the daily intake (DI) limit of IHg is 0.57 μg/kg bw/day. In China, the national standard limit of 20 μg/kg THg in rice is recommended (GB2762, 2017). Currently, no standard limit is set for rice MeHg.

Previous studies found more active Hg methylation in rice paddies than other kinds of farmlands (Qiu et al., 2008), and both MeHg and IHg in rice grains originate from soil (Rothenberg et al., 2014; Tang et al., 2017; Xing et al., 2019; Xu et al., 2019b; Zhang et al., 2010b). MeHg levels in rice were highly variable at different sites due to the differences in rice varieties, microorganisms, and factors influencing Hg methylation (Beckers and Rinklebe, 2017; Ma et al., 2019; Rothenberg et al., 2012; Rothenberg et al., 2014). Based on the daily rice consumption (620 g/day) for adults with a bw of 60 kg in Hg mining areas, the MeHg absorption rate (95%), and a RfD of 0.1 μg/kg bw proposed by the USEPA, we estimated that the maximum allowed MeHg limit in rice should be 10.2 μg/kg (Feng et al., 2008; Qiu et al., 2008; WHO-IPCS, 1990). Most studies on rice Hg were conducted in Hg mining areas, and the average THg levels were higher than Chinese national standard limit of 20 μg/kg, with a range of 7.1–1120 μg/kg (Table S2). And the mean rice MeHg in Hg mining areas can be 38.9 μg/kg, with a range of 1.97–174 μg/kg. As shown in Fig. 1 and Table S2, except for those collected from Hg polluted sites, the majority of rice samples had very low MeHg levels (<10.2 μg/kg). This leads to the hypothesis that the MeHg risk in rice may be overrated. Considering that rice is the dominant staple food for more than half of the world's population, it is necessary to evaluate the MeHg levels of rice in international/national markets to fully understand the risk of human exposure.

China is the world's leading rice producer and consumer, constituting 28.5% of the world's total production and 29.1% of the world's consumption in 2017 (FAOSTAT, 2019). Data indicated that the average rice consumption was 212 g/capita/day in 2013 in the country (FAOSTAT, 2019), approximately twice as high as that of world population (148 g/capita/day). To date, most studies on rice THg and MeHg have been conducted in China, mainly in Hg polluted areas (Rothenberg et al., 2014). These studies that reported some of the highest THg and MeHg levels in rice cannot be used to represent national levels. A nationwide investigation of THg and MeHg in commercial rice in China is therefore urgently needed.

In the present study, total Hg (THg) and MeHg levels in 767 rice samples throughout Chinese markets were investigated. The objectives of this study are to (1) elucidate both THg and MeHg levels in commercial rice in Chinese markets nationwide; (2) estimate the daily exposure of IHg and MeHg to Chinese populations associated with the rice ingestion; and (3) discuss the temporal trend in THg and MeHg levels in Chinese rice and associated risks compared to those in previously reported data. This investigation is basic and critical to the understanding of the risks of Hg via rice consumption by the Chinese population.

2. Materials and methods

2.1. Sampling and preparation

Rice samples (domestic: n = 709; imported: n = 58) were bought either from the local markets or online between August and November 2017. The producing areas of the samples were acquired from the information on each package. In summary, the samples were from 29 provinces of China (n = 709) and 10 countries (Cambodia: n = 8; India: n = 7; Laos: n = 6; Pakistan: n = 5; Spain: n = 2; Italy: n = 3; Japan: n = 6; Russia: n = 7; Vietnam: n = 7; and Thailand: n = 8) (Fig. S1 and Fig. S2). The samples were all polished, but their varieties were not known due to the lack of detailed information on the packages.

Each sample consisted of 0.5 kg–10 kg rice, depending on the package size. The samples were well mixed, and approximately 30 g of each sample was weighed, rinsed with distilled deionized water (DDW, 18.2 Ω cm water, Milli-Q, Millipore, USA), freeze-dried for 48 h (FDU-2110, EYELA, Japan), ground (IKA-A11 basic, IKA, Germany), passed through 80 mesh (size: 177 μm) (Xu et al., 2020), and stored in polyethylene bags prior to chemical analysis.

2.2. Total Hg and MeHg analysis

For THg analysis, approximately 0.5 g of sample was digested at 95 °C for 3 h with 5 mL HNO_3 and H_2SO_4 mixture (HNO_3:H_2SO_4 = 4:1; v:v) and measured by cold vapor atomic fluorescence spectroscopy (CVAFS, Model III, Brooksrand, USA) preceded by bromine chloride oxidation and stannous chloride reduction, according to USEPA Method 1631E (USEPA, 2002). For MeHg determination, approximately 0.5 g of sample was weighed and digested with 5 mL 25% KOH in methanol (m/m) at 75 °C for 3 h. The MeHg in the rice samples was leached with dichloromethane (CH_2Cl_2) and back-extracted into the water phase for determination by gas chromatographic cold vapor atomic fluorescence spectrometry (GC-CVAFS) based on USEPA Method 1630 (USEPA, 2001). All the acids used in the present study were ultrapure grade, and other reagents were analytical grade (Sinopharm Chemical Reagent Co., Ltd, China). The dichloromethane reagent was chromatographic grade (Tedia company, Inc., USA). The vials were rinsed with DDW water and preheated in a muffle oven (500 °C, 45 min) to ensure low Hg blanks. The IHg concentrations were calculated by THg minus MeHg (Xu et al., 2017).

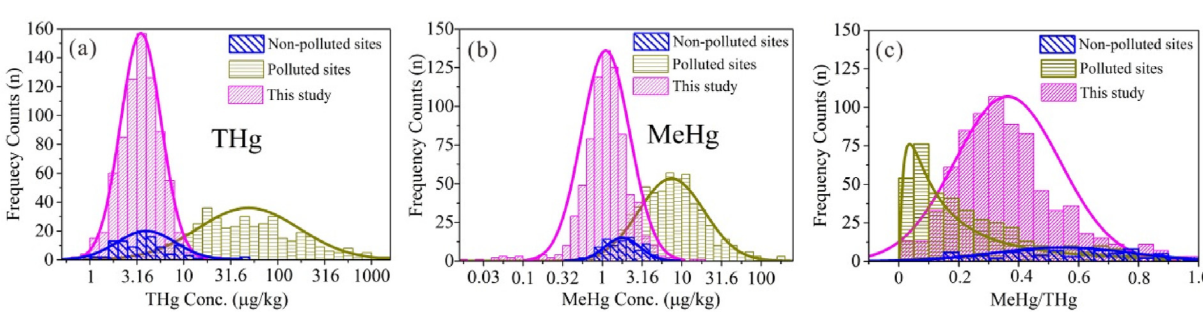

Fig. 1. Distribution of THg (a), MeHg (b), and MeHg/THg (c) in rice from polluted sites, non-polluted sites, and this study. (Rough data is derived from published literature in Table S2).

2.3. Quality assurance and quality control

The detection limits for THg and MeHg were 0.0120 μg/kg (3σ) and 0.00600 μg/kg (3σ), respectively. Quality assurance (QA) and quality control (QC) were implemented using duplicates, method blanks, matrix spikes and certified reference materials. The relative percentage difference in the duplicate samples for THg and MeHg were <9.5% and <16.4%, respectively. Recoveries from the matrix spikes were 102%–110% for THg and 90%–108% for MeHg.

GBW10020 (citrus leaves) was used as a certified reference material for THg determination. The obtained value of THg for GBW10020 was 149 ± 7 μg/kg (n = 30), with recoveries of 92%–107%, which was consistent with the certified value (150 ± 20 μg/kg). TORT-2 (lobster, *Hepatopancreas*) was used as a certified reference material for MeHg determination. The average MeHg value for TORT-2 was 150 ± 6.2 μg/kg (n = 30), with recoveries of 94%–104%, consistent with the recommended value (152 ± 13 μg/kg) (Fig. S3).

2.4. Statistical analysis

Statistical analysis was performed using SPSS 22 (Stanford, California, USA). Figures were obtained using Origin 9 (©OriginLab Corporation).

Dietary exposure to MeHg and IHg was assessed using the Monte Carlo method and bootstrap values. Monte Carlo simulation was employed to perform the analysis of sensitivity and uncertainty, using input probability distributions based on empirical data (Peng et al., 2016). Specifically, a Monte Carlo simulation is a statistical method that applies random statistical sampling techniques to acquire a probabilistic approximation to the solution of a mathematical equation or a model (Sofuoglu et al., 2014). The simulation of the frequency distribution in Crystal Ball© (Oracle, Redwood City, CA, USA) software was configured with 100000 iterations to guarantee the reliability of the results. The statistics of the mean values and percentiles (P50, P90, P95, P97.5, P99, and P99.9) were obtained using Monte-Carlo random distribution numbers.

2.5. Dietary exposure and risk estimates

The DIs (μg/kg bw/day) of MeHg and IHg were calculated using Eqs. (1)–(3):

$$DI = \frac{C_{MeHg\ or\ IHg} \times IR \times A \times 10^{-3}}{BW} \quad (1)$$

$$C_{bio\ of\ MeHg} = C_{MeHg} \times B_{bio\ of\ MeHg} \times 100\% \quad (2)$$

$$C_{bio\ of\ IHg} = C_{THg} \times B_{bio\ of\ THg} - C_{bio\ of\ MeHg} \quad (3)$$

where C_{MeHg} and C_{IHg} represent the MeHg and IHg concentrations (μg/kg) of rice, respectively; IR represents the rice intake rate (g/day); BW represents the bw (kg); A represents the absorption efficiency, which is assumed to be 8% for IHg and 95% for MeHg (WHO-IPCS, 1990, 1991). $B_{bio\ of\ MeHg}$ represents the bioaccessibility ratio of MeHg ($_{bio\ of\ MeHg}$ is 100% if there is without consideration of the bioaccessibility). The BW and IR values for different population groups (summarized in Table S1) were obtained from the Chinese National Health and Nutrition (Yang and Zhai, 2006; Zhai and Yang, 2006).

Since there existed large differences in rice consumption rates from different regions, rice intake rates in different provinces were obtained from published studies and provincial statistical yearbooks (Table S3). Due to the lack of data, some provinces were not included in our study. A bw of 60 kg was employed to estimate the rice MeHg and IHg exposure according to different intake rates.

The human health risks posed by chronic exposure to MeHg and IHg via rice consumption were estimated from the hazard quotient (HQ) (Eqs. (4) and (5)). The HQ is applied to express the risk of non-carcinogenic effects (when a single substance exposure level is higher than a reference dose, there may exist a risk of some expected negative health effects but not carcinogenic effects) of MeHg and IHg, and the HQ for residents was evaluated by comparing with the PTWI for IHg and the RfD for MeHg (Rothenberg et al., 2017; USEPA, 2000; Vieira et al., 2011; Zheng et al., 2007). Based on the additive effects, HQs can be summed to generate a hazard index (HI) for the combination pathway (Eq. (6)) (Qian et al., 2010). HQ or HI value > 1 indicates non-carcinogenic adverse health effects owing to both MeHg and IHg exposure from rice intake, and HQ or HI value < 1 denotes no adverse effects.

$$HQ_{IHg} = \frac{DI \times 7}{PTWI} \quad (4)$$

$$HQ_{MeHg} = \frac{DI}{RfD} \quad (5)$$

$$HI = HQ_{IHg} + HQ_{MeHg} \quad (6)$$

where the PTWI for IHg is 4 μg/kg bw/week from JECFA (JECFA, 2010); and the RfD is 0.1 μg/kg bw/day (Rothenberg et al., 2017).

3. Results and discussion

3.1. THg and MeHg in rice

The THg concentrations (0.640–31.7 μg/kg, n = 767) had lognormal distributions (Table 1, Fig. 1a); therefore, the geometric mean value was reported (3.97 ± 2.33 μg/kg). Only 3 samples had THg concentrations that exceeded the maximum limit of THg in food (20 μg/kg) recommended by the Chinese National Food Safety Standard (GB2762, 2017). Thus, > 99.5% of the rice from Chinese markets was safe for THg. A few studies have reported the THg levels in rice from Chinese markets, demonstrating a range of 0.860–47.2 μg/kg (Table S2). Other worldwide studies of rice have shown similar THg levels of 0.30–85 μg/kg (Table S2). In general, our results are within the range of the reported THg values of commercial rice. In the present study, these three samples, which exceeded the THg limit, were 31.7 μg/kg, 20.4 μg/kg, and 23.3 μg/kg from Fujian (Longyan), Guangxi (Nanning), and Zhejiang (Jiaxing), respectively. In the production areas of these three samples, soils or food were reported to have elevated Hg due to the mixed discharge of domestic and industrial sewage and the leaching of solid waste (Chen, 2013; Li, 1999; Pang et al., 2011; Qin et al., 2006; Zheng, 2003).

The MeHg concentrations of the rice samples were also lognormally distributed (Fig. 1b), with a mean value of 1.37 ± 1.18 μg/kg (range: 0.020–19.0 μg/kg). More than 99.5% of the samples had MeHg concentrations below 10.2 μg/kg, which is the maximum limit of MeHg in rice according to our earlier estimation in the Introduction Section. The MeHg levels of rice from Chinese markets have been investigated by only two previous studies. Shi et al. reported low MeHg levels of 1.90–10.5 μg/kg (mean: 4.70 μg/kg) for commercial rice from 15 provinces in China, but these data may not be representative due to the small sample sizes (n = 25) (Shi et al., 2005). Li et al. reported similar MeHg levels of 0.130–18.2 μg/kg (mean: 2.47 μg/kg) from 7 provinces in southern China with a sample size of n = 284 (Li et al., 2012). Generally, our

Table 1
THg, MeHg, and IHg variation in rice samples of this study.

Country of origin	Provinces	Number of brands (n)	THg (μg/kg) (mean ± SE)	Range	MeHg (μg/kg) (mean ± SE)	Range	IHg (μg/kg)	Range
China	Heilongjiang	68	4.04 ± 1.73	1.28–7.94	1.18 ± 0.500	0.317–2.89	2.86 ± 1.67	0.402–6.39
	Jilin	30	3.27 ± 0.900	1.96–5.14	1.37 ± 0.643	0.585–2.92	1.90 ± 0.648	1.17–3.62
	Liaoning	20	3.97 ± 1.11	2.25–6.24	1.04 ± 0.352	0.501–1.80	2.76 ± 1.01	1.72–5.00
	Hebei	6	5.13 ± 3.83	2.34–12.7	1.12 ± 0.0530	0.629–1.88	4.01 ± 4.00	1.34–12.0
	Shandong	15	5.27 ± 2.32	1.61–8.42	0.891 ± 0.254	0.467–1.28	4.38 ± 2.17	1.09–7.88
	Shanxi	5	1.66 ± 0.655	1.08–2.77	1.02 ± 0.384	0.495–1.44	0.635 ± 0.435	0.261–1.33
	Shaanxi	5	3.04 ± 1.26	0.970–4.32	1.21 ± 0.677	0.484–2.29	1.84 ± 1.10	0.486–3.06
	Tianjin	5	2.70 ± 0.593	2.33–3.75	0.739 ± 0.496	0.383–1.61	1.96 ± 0.173	1.73–2.13
	Ningxia	20	3.62 ± 0.946	1.59–5.94	1.36 ± 0.459	0.823–2.96	2.26 ± 1.08	0.663–4.69
	Gansu	3	3.058 ± 0.702	2.257–3.49	1.07 ± 0.0680	0.736–2.20	1.43 ± 0.123	1.29–1.52
	Xinjiang	20	2.72 ± 1.92	1.29–10.6	0.601 ± 0.270	0.169–1.50	2.12 ± 1.95	0.544–10.9
	Inner Mongolia	20	4.03 ± 1.49	2.12–8.29	2.11 ± 1.46	0.727–4.39	1.92 ± 0.447	0.959–2.46
	Jiangsu	36	1.56 ± 1.92	1.04–8.19	1.31 ± 0.869	0.0200–3.31	3.25 ± 1.86	0.250–7.46
	Anhui	35	3.56 ± 1.52	1.07–7.54	1.23 ± 0.549	0.536–3.20	2.32 ± 1.36	0.539–6.65
	Zhejiang	19	5.24 ± 4.71	1.38–23.3	1.16 ± 0.582	0.635–2.90	4.11 ± 4.28	0.399–20.4
	Hunan	53	4.17 ± 1.82	1.36–9.55	1.70 ± 1.17	0.435–6.80	2.48 ± 1.36	0.0730–6.90
	Hubei	35	5.14 ± 2.53	1.12–13.1	1.99 ± 1.09	0.648–5.62	3.16 ± 2.44	0.386–11.6
	Jiangxi	41	3.93 ± 2.09	1.01–8.59	1.31 ± 0.717	0.0430–3.82	2.62 ± 1.89	0.124–7.21
	Henan	21	3.86 ± 2.43	1.11–10.1	1.45 ± 0.804	0.541–4.11	2.42 ± 1.89	0.0320–7.02
	Shanghai	10	6.09 ± 1.93	1.61–8.71	1.41 ± 0.554	0.467–1.28	4.68 ± 1.61	1.09–7.88
	Guangdong	43	3.62 ± 1.91	0.638–12.0	1.40 ± 0.0730	0.150–5.38	2.22 ± 1.09	0.0120–6.69
	Guangxi	38	4.24 ± 3.13	1.89–20.4	2.14 ± 2.35	0.595–15.0	2.10 ± 1.14	0.617–6.45
	Fujian	20	3.11 ± 1.45	2.10–31.7	0.890 ± 0.428	0.375–19.0	2.18 ± 1.34	1.36–12.7
	Hainan	17	3.25 ± 2.16	0.769–6.98	0.898 ± 0.755	0.0470–2.45	2.35 ± 1.48	0.722–4.93
	Taiwan	14	4.72 ± 1.77	1.98–8.56	0.923 ± 0.280	0.508–1.59	3.80 ± 1.71	1.48–7.77
	Yunnan	27	4.37 ± 2.26	1.33–10.4	1.89 ± 1.40	0.0270–6.21	2.47 ± 1.18	0.278–5.63
	Guizhou	25	4.94 ± 1.81	1.65–7.86	1.52 ± 1.06	0.0830–4.95	3.42 ± 1.45	1.04–6.33
	Sichuan	39	3.01 ± 1.27	0.883–6.11	1.11 ± 0.588	0.178–2.93	1.90 ± 1.09	0.155–5.19
	Chongqing	19	2.56 ± 1.24	0.808–5.63	0.912 ± 0.453	0.298–1.99	1.65 ± 1.16	0.650–4.65
Total Chinese domestic		709	4.03 ± 2.37	0.638–31.7	1.40 ± 1.21	0.0200–18.6	2.63 ± 1.80	0.0123–20.4
Cambodia		8	3.92 ± 1.02	1.79–5.02	1.56 ± 0.580	1.11–2.86	2.36 ± 0.850	0.422–3.26
India		7	1.88 ± 0.180	1.73–2.16	0.680 ± 0.163	0.423–0.889	1.20 ± 0.194	1.01–1.57
Laos		6	5.26 ± 2.36	2.99–8.90	1.57 ± 0.863	1.04–3.29	3.68 ± 1.78	1.93–6.24
Pakistan		5	2.33 ± 0.455	1.63–2.85	0.738 ± 0.261	0.338–0.988	1.587 ± 0.230	1.29–1.86
Spain		2	2.30 ± 0.00900	2.29–2.30	0.680 ± 0.207	0.534–0.827	1.62 ± 0.199	1.48–1.76
Italy		3	4.29 ± 0.200	4.09–4.49	1.90 ± 0.600	1.40–2.57	2.39 ± 0.796	1.52–3.09
Japan		6	3.74 ± 1.36	1.88–6.02	1.21 ± 0.749	0.409–2.29	2.53 ± 0.846	1.47–3.73
Russia		7	2.59 ± 1.14	1.48–4.67	0.584 ± 0.360	0.184–1.10	2.01 ± 0.866	1.30–3.72
Vietnam		7	2.03 ± 0.780	1.12–3.09	0.662 ± 0.241	0.383-0.948	1.37 ± 0.699	0.197–2.22
Thailand		8	3.44 ± 1.59	1.74–5.81	0.820 ± 0.491	0.0690–1.47	2.62 ± 1.22	1.21–4.35
Total imported		58	3.82 ± 1.56	1.12–8.90	1.02 ± 0.639	0.0690–3.29	2.17 ± 1.13	0.197–6.24
Total average		767	3.97 ± 2.33	0.638–31.7	1.37 ± 1.18	0.0200–19.0	2.60 ± 1.14	0.0120–20.4

results of MeHg are within the range reported by previous studies. Considering the sample size and sampling locations, our results are currently representative of the MeHg level in rice from Chinese markets.

In general, our MeHg results are compatible or slightly lower than results from European markets (mean: 1.91 μg/kg, n = 87) and other markets (or non-polluted sites) worldwide but much lower than previous results on rice collected from polluted sites (e.g., mercury mines and coal-fired power plants) in China (Table S2). Fig. 1b shows that rice collected from polluted sites had both THg and MeHg levels that were approximately 1–3 orders of magnitude higher than those collected from non-polluted sites and Chinese markets. The samples with high Hg concentrations (n = 3 for THg; n = 2 for MeHg) were in a small scale compared to those of the whole sample size of this study (n = 767). We suggest that the Hg in rice from Chinese markets is at a safe level. Since rice Hg pollution does exist in Hg polluted sites in China, residents in Hg polluted areas may meet a high THg exposure. Though residents in polluted areas may consume rice from unpolluted areas, actions should also be taken in polluted areas to prevent Hg polluted rice flowing into the markets.

The THg and MeHg concentrations showed large spatial variations in China. Rice with relatively high THg and MeHg levels is mainly located in Shandong, Henan, Hebei, Hubei, Anhui, Jiangsu, Zhejiang, and Shanghai provinces in central and eastern China (Fig. 2). China's major heavy industries are located in these provinces. Furthermore, in southern China, Jiangxi, Hunan, Guangxi, Guizhou, and Yunnan provinces are non-ferrous metal production base (note that Hg is extremely enriched in hydrothermal ore minerals) in China. These provinces mentioned above account for approximately 50% of the total Chinese population, and consume >75% of the coal and oil in China (NBS, 2017). Anthropogenic activities, especially fossil fuel combustion and non-ferrous metal smelting release substantial amounts of Hg into the surrounding environment, which may be the main reason for the high Hg levels in rice in these provinces (Streets et al., 2005; Zhang et al., 2015).

The MeHg proportions (MeHg%) ranged from 0.5 to 98% (Fig. 1c) and were normally distributed, with a mean value of 36.3%. The MeHg proportions were much higher than previous proportions for on dry land crops such as corn and wheat, but were comparable to previous results for rice, suggesting that rice has a strong capability to accumulate MeHg (Qiu et al., 2008). Rice paddies have been shown to be hot spots of Hg methylation, and rice mainly receives MeHg from soil (Zhang et al., 2010b). Notably, 137 rice samples had MeHg% exceeding 50%, and 96 of them were grown in southern China. The remaining samples (n = 626) showed lower MeHg

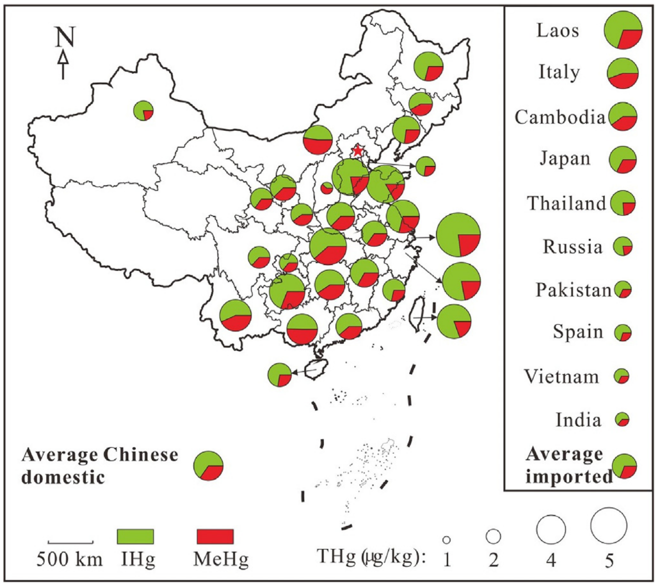

Fig. 2. Regional distribution of THg and MeHg in Chinese grown and imported rice.

proportions (29.8%). The high MeHg proportions in southern China were likely caused by high MeHg in soil, which may be due to higher soil Hg and temperature in southern China, since microbial Hg methylation can be promoted under high temperature and soil (Loseto et al., 2004; Ma et al., 2019; Xu et al., 2017). Notably, the MeHg% in rice from unpolluted sites may have been higher than those in polluted sites (Fig. 1) and may be influenced by atmospheric Hg deposition (Kwon et al., 2018). In comparison to unpolluted sites, at polluted sites lower rice MeHg% values were obtained (Fig. 1) because Hg in paddies from polluted sites is mostly in poorly solubilized forms (Beckers et al., 2019; Beckers and Rinklebe, 2017; O'Connor et al., 2019; Wang et al., 2019; Zhou et al., 2015), making Hg less bioavailable for MeHg methylation (Issaro et al., 2009). Meanwhile, MeHg% are also controlled by many factors, such as rice varieties, soil properties (N, S, pH, and organic matter), and microbial activities (Beckers and Rinklebe, 2017; Rothenberg et al., 2012; Wang et al., 2014; Xing et al., 2019; Yin et al., 2018).

3.2. Dietary exposure of MeHg and IHg via rice consumption

In the present study, the main factors of consumer bw, rice intake rate, rice Hg concentration, age, and gender were taken into account to evaluate exposure and risk. Notably, three widely used limits: RfD-0.1 μg/kg bw/day by USEPA, PTWI-1.3 μg/kg bw/week by the European Food Safety Authority, and PTWI-1.6 μg/kg bw/week by JECFA for MeHg were all obtained from the epidemiologic studies of fish intake (EFSA, 2012; FAO, 2007; Kjellstrom et al., 1986, 1989; Rice et al., 2000). Fish contain high levels of dososahexaenoic acid (DHA), which may counteract the adverse health effects associated with MeHg exposure; however, there is limited DHA in rice, which means that the same dose of MeHg from rice will be more harmful than that from fish (Rothenberg et al., 2011). The RfD for MeHg in rice should be warranted and be stricter than the RfD for fish. Thus, in this study, we employed the RfD of 0.1 μg/kg bw/day to calculate the HQ of MeHg. To avoid underestimating the exposure risk of Hg, in this study, we also considered both MeHg and IHg intake for the first time. The DI values of IHg and MeHg for male and female with different ages were calculated (Tables S4 and S5), using Eqs. (1)–(3).

For both male and female, the DI values of MeHg and IHg decrease as their ages increase, and younger group (4–7 years old for male; and 2–4 years old for female) have the highest DI values (Fig. 3a). In general, male showed higher DI values of MeHg than female of the same age, implying more MeHg exposure via rice consumption. Generally, the mean DI values of MeHg (range: 0.005–0.0109 μg/kg bw/day) and IHg (range: 0.0014–0.0017 μg/kg bw/day) in our study were two orders of magnitude lower than the RfD (0.1 μg/kg bw/day) recommended by the USEPA (Rice et al., 2000). Meanwhile, the DIs of both IHg and MeHg via rice were compatible or slightly lower than those obtained in non-polluted sites in China, but several magnitudes lower than those obtained in polluted sites (Table S2). This suggests that MeHg exposure via rice consumption is limited in Chinese populations, which is supported by the low MeHg levels in the hair of rice consumers in China (Du et al., 2018; Hong et al., 2016).

Residents in different regions showed regional differences in MeHg and IHg exposure levels via rice intake (Fig. S4). Rice MeHg and IHg exposure of residents was MeHg: 6.71E-3±4.29E-3 μg/kg bw/day and IHg: 6.75E-3±4.25E-4 μg/kg bw/day in Central China; and MeHg: 6.84E-3±4.38E-3 μg/kg bw/day; IHg: 6.89E-4±4.33E-4 μg/kg bw/day in Southern China, while residents in Northwest China had the lowest rice MeHg and IHg exposure levels (MeHg: 1.90E-4±5.88E-4 μg/kg bw/day; IHg: 9.30E-5±5.80E-5 μg/kg bw/day) (Fig. S4). The Hg exposure levels of residents in the highest rice consuming region were more than 7 times those in the lowest rice consumption areas. Correspondingly, the highest (Guangxi) and the lowest rice Hg exposure provinces (Inner Mongolia) were located in the highest and lowest exposure regions (Fig. S5), respectively. This is expected as the regional differences in diet patterns are influenced by economic and sociodemographic structure (Dong and Hu, 2010). Even in the highest rice MeHg exposure region-southern China, the rice MeHg exposure was only 17.2% of the average rice MeHg exposure (0.039 μg/kg bw/day) in previous studies (Liu et al., 2018; Liu et al., 2019b), suggesting a decline of rice MeHg exposure in China. In addition, since studies revealed that rice Hg pollution occurred mostly in southern China (Table S2), indicating that the special attention should be paid to residents in these areas.

High DI values of MeHg have been reported for residents from Europe (0.050 μg/kg bw/day), Japan (0.280 μg/kg bw/day) and Northern America (0.020 μg/kg bw/day), who are exposed to MeHg through fish consumption (Iwasaki et al., 2003; Mahaffey et al., 2004; Mangerud, 2005). Fish consumers from China and many other countries also have shown high DI values (Gong et al., 2018; Li et al., 2012). Low Hg exposure via rice consumption in this study was also consistent with the results of a recent study, which demonstrated that in China, in comparison to rice consumption (26%), fish intake (56%) plays a more important role in human MeHg exposure (Liu et al., 2018).

For both male and female, the HQ and HI values showed the same trends as the DI values (Fig. S6). The values at P50 (50th percentile) demonstrated the median risk exposure of rice consumers to the distribution, and the values at P95, P97.5, P99, and P99.9 demonstrated the higher exposure to both MeHg and IHg (Table S4). The Monte Carlo simulation derived the median P50, P95, P97.5 (97.5th percentile), P99, and P99.9. The HQ values of MeHg and IHg in all gender-age categories were all lower than 1, suggesting a low health hazard for MeHg and IHg. Furthermore, the corresponding HI values showed a decreasing trend similar to that of HQ (Fig. S6), and all the HI values were less than 1. This indicated that there is no non-carcinogenic risk of MeHg and IHg for the Chinese population via commercial rice consumption.

Additionally, only a part of ingested Hg in food can be released, which is defined as bioaccessibility (Bradley et al., 2017). The

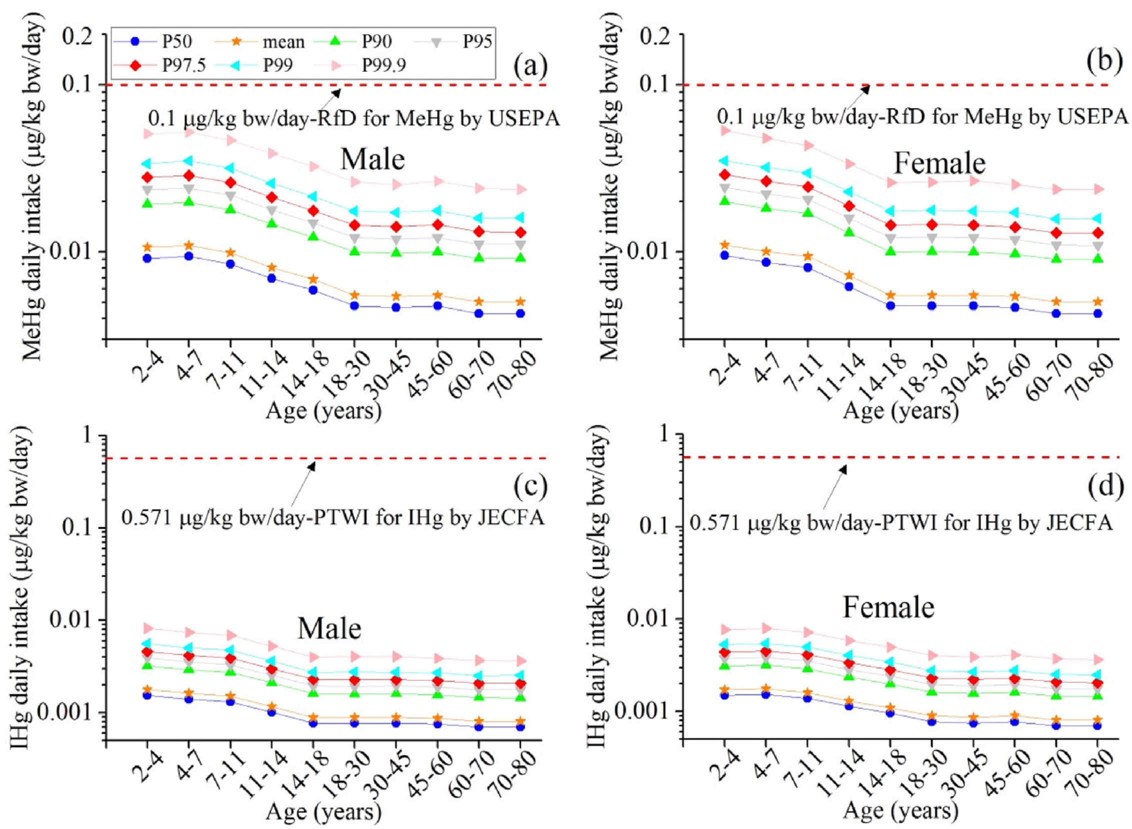

Fig. 3. Percentiles distribution of MeHg and IHg exposure via the intake of Chinese commercial rice.

bioavailability of Hg refers to the proportion of the ingested Hg in food that enters into the systematic circulation and exerts its toxic effects (Bradley et al., 2017). Bioaccessibility is dependent on soluble fraction by the end of the digestion processes and could be used as a conservative assessment of bioavailability since bioaccessiblity is a maximum value in theory (Lin et al., 2019).Thus, considering the THg and MeHg bioaccessibility of rice, the DIs of both MeHg and IHg were substantially low (Fig. S7). Studies have indicated that the bioaccessibility ratio of rice THg in rice ranges from 6.50% to 47.3% (Wu et al., 2017), and the bioaccessibility of MeHg in rice ranges from 15.9% to 56.3% (Gong et al., 2018). Even based on the high bioaccessibility ratios of 47.3% for THg and 56.3% for MeHg, for male and female, the DIs, HQs, and corresponding HI values were approximately 41.2%–56.3% of those values when without considering bioaccessibility, suggesting a much lower exposure risk of Hg with the consideration of bioaccessibility (Fig. 3b).

3.3. Temporal trend of Hg in Chinese rice

Compared with previous reported results, the results of this study showed a decreased temporal change from 2007 to 2017. The THg in Chinese-grown rice was 10.1 μg/kg in 2007, 5.80 μg/kg in 2008, 4.90 μg/kg in 2011, and 4.03 μg/kg in 2017, decreasing by rates of 42.5%, 15.5%, and 17.7%, respectively (Li et al., 2012; Qian et al., 2010; Zhang et al., 2014). Simultaneously, MeHg declined by approximately 43.3% from 2.47 μg/kg in 2007 to 1.40 μg/kg in 2017 (Li et al., 2012). Hence, these results suggested apparent downward trends in both THg and MeHg intakes via rice Hg exposure risk within the last decade and Hg exposure risk as well (Fig. 4, Fig. S8 and Fig. S9).

Corresponding to the decrease in THg and MeHg in Chinese commercial rice, a series of energy-saving and emissions-reduction policies have also been issued in China since 2005 (Fig. 4). Studies found that during the 11th Five-Year Plan, SO_2 emissions in China were decreased by 14% of emission level in 2005 (Schreifels et al., 2012), and NO_x emissions in China were reduced by 21% in 2010 during the 12th Five-Year Plan (Liu et al., 2017). Specifically, in 2011, the Chinese government announced a notice on developing pilot work for the control of atmospheric Hg pollution in CFPPs. With the implementation of these policies, a simultaneous decrease in atmospheric Hg emissions occurred in major Hg emissions sources, such as CFPPs (removal efficiency: 73%), non-ferrous smelting (removal efficiency: 79%), and coal-fired industrial boilers (removal efficiency: 42%) (Ancora et al., 2015; Kwon et al., 2018; Wang et al., 2012; Wu et al., 2016; Zhang et al., 2012).

Significant synergetic Hg removal, with efficiencies ranging between 42% and 79%, from those major anthropogenic atmospheric Hg sources with strict controls (Wang et al., 2012; Wu et al., 2016), might result in a decrease in Hg deposition into the environment. Recently, a study reported that a decreasing trend in atmospheric Hg has occurred since 2010 in background areas of China (Tong et al., 2016). Moreover, a decline in the temporal trend of Hg in water also occurred in the Pearl River and Yangtze River due to more stringent control measures that were strengthened in recent years (Duan et al., 2015; Liu et al., 2016a; Liu et al., 2016b; Xu et al., 2019a). Since soil MeHg is believed to be the major origin of MeHg in rice grains (Meng et al., 2011; Qiu et al., 2011; Zhang et al., 2010b), and evidences suggested that IHg in grain originates from soil (Tang et al., 2017; Xu et al., 2019b; Yin et al., 2013). Further studies have revealed that newly deposited Hg is more bioavailable and ready for methylation, and Hg methylation is regulated by newly

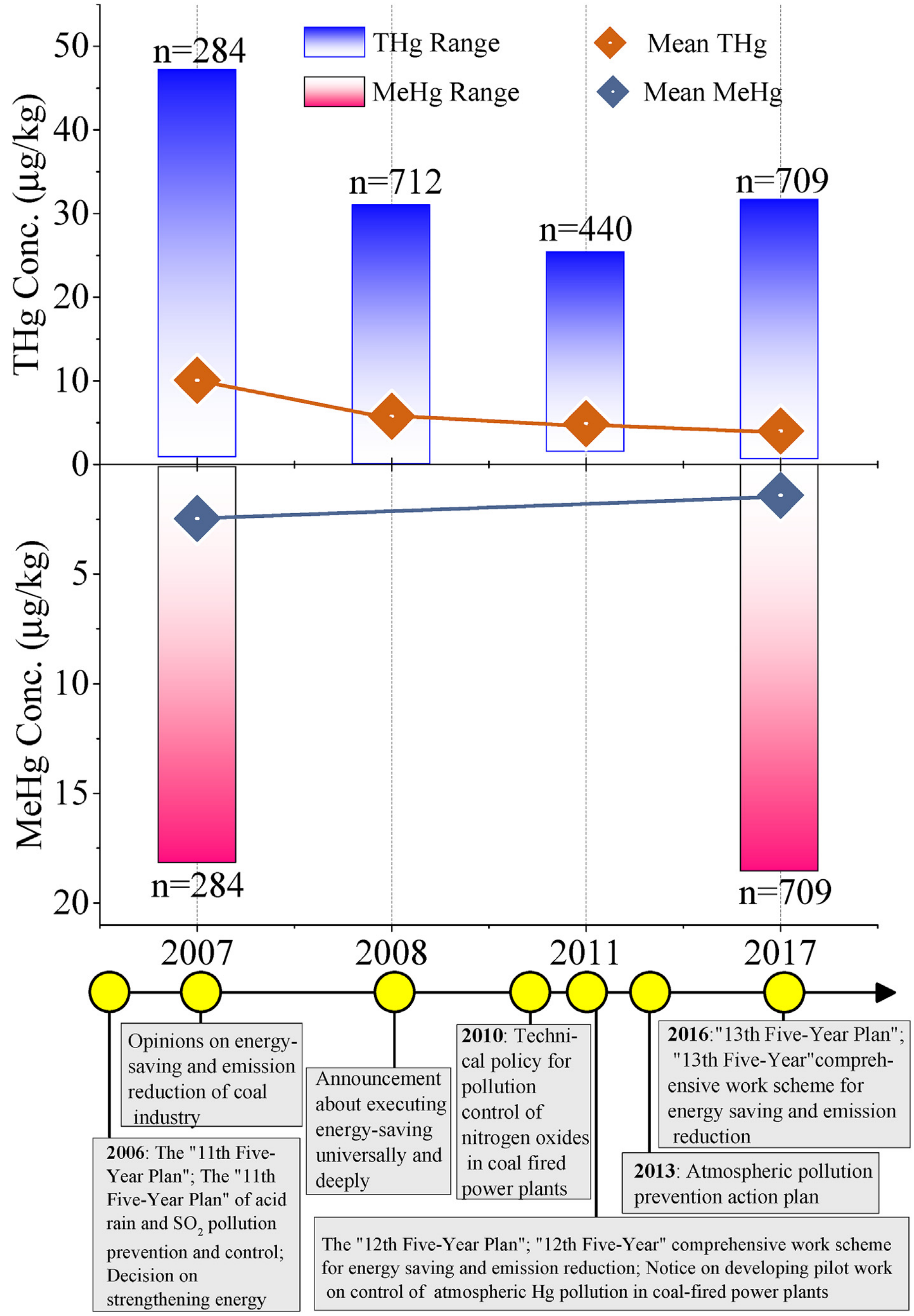

Fig. 4. Temporal trend of rice THg and MeHg in China from 2007 to 2017. Investigated data is derived from published literature and this study (Li et al., 2012; Qian et al., 2010; Zhang et al., 2014).

deposited Hg to soil (Kwon et al., 2018; Meng et al., 2011; Xu et al., 2017). The low input rate of the newly deposited Hg reduces the bioavailability of IHg and MeHg in paddy soils, resulting in a decrease in Hg in rice grains (Xu et al., 2017). Hence, a series of policies and regulations issued by the Chinese government since 2005 that aim to control air pollutants were a source of the decrease in both THg and MeHg in Chinese-grown rice. Our results are also in accordance with the results of a modeling study from a previous study (Kwon et al., 2018), which indicated that atmospheric deposition was the major source of Hg in rice for most regions of China except where soil was contaminated by point source Hg, and under strict policies, levels of rice Hg in China will show a sharp decrease. The decrease observed in the present study demonstrates a significant effect of the management of Hg reduction proposed by the Chinese government within the last decade, confirming the effectiveness of the adopted management.

It should be noted that the results in 2007 were only from 7 provinces of southern China (Li et al., 2012), which may lead to the bias when compared to the results in 2017. Considering that the rice yield of these provinces is 46.4% of the total yield in China (Li et al., 2012), we hypothesized that the MeHg trend might be representative to a certain extent. The rice production areas of the samples collected in 2008, 2011, and 2017 (this study) covered approximately 95.3%, 83.1%, and 100% of total rice production in China, respectively (NBS, 2017). Moreover, recent studies assessed rice Hg exposure across China with reported data (Liu et al., 2018; Liu et al., 2019b), and the employed data were 3.21 and 2.40 times of the measured THg and MeHg in this study, with ranges of 2.14−6.02 and 1.52−3.51 times in different regions (Fig. S10), respectively. Correspondingly, this indicated rice THg and MeHg exposure wre overestimated, and also suggested that rice THg and MeHg showed a downward trend. Therefore, we believe that the results of these studies could fully represent the rice Hg levels in China. The THg results with a large sample size from 2008 to 2017 exhibited a steadily declining trend, even when the results from 2007 were not included.

4. Conclusion

We carried a nationalwide survey of both THg and MeHg in Chinese commercial rice. Both THg and MeHg levels in Chinese commercial rice are generally low and safe, though there exist Hg pollution in commercial rice. Correspondingly, rice IHg and MeHg exposures in different Chinese age-gender groups were quite lower compared to RfD-0.1 μg/kg bw/day and PTWI-0.57 μg/kg bw/day. Residents in Central China and Southern China meet the highest rice Hg exposure, which were more than 7 times of those in Northwest China. While rice Hg pollution a valid concern for Hg-contaminated sites, there does not to be much worry about Hg exposure from Chinese markets. With the efforts of strict policies to control Hg implemented by the Chinese government, Hg emissions to the environment have declined, and Hg concentrations in rice are expected to decrease in the future. Hence, concerns related to Hg contamination in rice should not be overemphasized, considering the fact that >99.5% of rice in the market is low in Hg. However, the high ratios of MeHg to THg observed in rice might cause chronic low-dose exposure to humans, particularly sensitive populations of pregnant women and children, and should be given more attention in the future. Moreover, MeHg bioaccumulation is an issue at Hg-contaminated sites, and this situation should be resolved with proper controls, such as phytoremediation of soils and rice planting bans.

Declaration of competing interest

The authors declare they have no actual or potential competing financial interests.

CRediT authorship contribution statement

Xiaohang Xu: Conceptualization, Formal analysis, Methodology, Writing - original draft, Visualization, Data curation, Writing - review & editing, Funding acquisition. **Jialiang Han:** Validation, Formal analysis, Investigation, Resources, Visualization, Data curation, Writing - review & editing. **Jian Pang:** Validation, Formal analysis. **Xun Wang:** Visualization. **Yan Lin:** Visualization. **Yajie Wang:** Investigation. **Guangle Qiu:** Supervision, Project administration, Funding acquisition.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.113706.

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