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Dietary exposure assessment of total mercury and methylmercury in commercial rice in Sri Lanka

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HIGHLIGHTS

- We firstly carried a nationwide investigation of Hg in Sri Lankan commercial rice.
- THg and MeHg exposures in different rice and fish eating populations were assessed.
- Rice consumption plays less important role in MeHg exposure in Sri Lanka.

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ABSTRACT

Methylmercury (MeHg) in rice has attracted growing health concern over the past decade, due to the accumulation of high MeHg levels, which may pose potential health risk to humans. Rice is the staple food in Sri Lanka; nevertheless, the presence of micro pollutants, such as MeHg has been not investigated. Therefore, commercial rice samples from the Sri Lankan market ($n = 163$) were measured to reveal the total mercury (THg) and MeHg levels. THg (mean: 1.73 ± 0.89 ng/g, range: 0.21–6.13 ng/g) and MeHg concentrations (mean: 0.51 ± 0.37 ng/g; range: 0.03–3.81 ng/g) were low. Compared to the fish MeHg exposure, the rice MeHg exposure was generally lower in different consumption groups, suggesting that rice plays a less role than fish in MeHg exposure in Sri Lanka. Babies (infants and toddlers) at one year old may face fish MeHg exposure (0.17 μg/kg bw/day) higher than the reference dose for MeHg (RfD)-0.1 μg/kg bw/day, which was more than 5 times that of rice MeHg exposure (0.031 μg/kg bw/day). Future studies in Sri Lanka should focus on health impacts under long-term overexposure of MeHg, especially in vulnerable populations. Some diet changes should be made to mitigate MeHg exposure levels in Sri Lankans.

1. Introduction

Mercury (Hg) is one of the poisonous pollutants worldwide, due to its potential neurotoxicity (Karagas et al., 2012; NRC, 2000), and it exists as inorganic Hg (IHg) and organic Hg (mostly methylmercury, MeHg) in different environmental media. Hg can easily be bioaccumulated through food chains posing a potential threat to humans (Beckers and Rinklebe, 2017). After ingestion, approximately 8% of IHg and 95% of MeHg will be absorbed through the digestive tract and transported into the bloodstream (Ceccatelli et al., 2010; Clarkson and Magos, 2006). MeHg can cross the blood-brain and placenta barriers, which may lead to neurotoxicity and teratogenicity (Ceccatelli et al., 2010). The toxicity of IHg can cause cardiovascular disorders and kidney damage (EFSA, 2012). The biological half-lives of Hg fractions are approximately 1–3 months, and the excretion pathways are bile and feces for MeHg and urine

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and feces for IHg (Akerstrom et al., 2017; Guérin et al., 2018).

Elevated THg and MeHg were found in rice grain from Hg mining areas (Horvat et al., 2003; Qiu et al. 2006, 2008), and rice intake was considered the major pathway of MeHg exposure for native residents in the region (Feng et al., 2008), attracting researchers attention to rice THg and MeHg exposure in Hg polluted areas, particularly Hg mining regions (Liang et al., 2015; Qiu et al., 2013; Rothenberg et al. 2011, 2014; Shao, 2014; Xu et al. 2017, 2018; Zhang et al., 2010a). However, until now, only a few studies on Hg in commercial ricedirectly consumed by residents have been reported (Al-Saleh and Abduljabbar, 2017; Antoine et al., 2012; Brombach et al., 2017; Qian et al., 2010; Zhang et al., 2014), and some high Hg levels were also found in commercial rice, exceeding the maximum limit of 20 ng/g in the Chinese National Food Safety Standards (GB2762 2017).

Rice is the dominant staple food for Sri Lanka and provides 42% of the total calories for Sri Lankans. Sri Lanka produced 3.08 million tonnes of rice and approximately 2.33 million tonnes were consumed in 2013 (FAOSTAT, 2019), suggesting that the produced rice can meet the domestic demand. Studies revealed that pollution of paddy soils was mainly due to the fertilizer applications in Sri Lanka (Chandrajith et al., 2005). In Sri Lanka, most paddies were cultivated in two seasons, dry (March to September) (*yala*) and wet (October to February) (*mala*) seasons (De Silva et al., 2007). The total rice consumption in Sri Lanka was approximately equal to the total consumption amount (2.51 million tonnes) in Europe (FAOSTAT, 2019). The daily rice consumption was 300.6 g/capita/day in Sri Lanka, ranking 9th highest in the world, which was more than 22 times the European consumption of 13.49 g/capita/day and more than 2 times the world daily consumption of 148 g/capita/day (FAOSTAT, 2019). With such a high ratio diet of rice in Sri Lankans (Hu et al., 2016), it is vital to reveal the Hg levels, especially MeHg, in Sri Lankan rice and estimate the associated exposure levels. On the other hand, fish were considered the main total Hg (THg) and MeHg source in Europe and North America (Beckers and Rinklebe, 2017; Kim et al., 2016). Fish are widely consumed by the Sri Lankan population with the consumption amount ranging from 43 to 265 g/day (Jayasinghe et al., 2018; Jinadasa et al., 2014). Thus, it is also important to identify the dominant role and major pathway of MeHg exposure through the consumption of fish in Sri Lanka.

Nevertheless, to the best of our knowledge, no studies have been conducted on rice Hg (both THg and MeHg) in Sri Lanka. Thus, a systematic investigation on commercial rice Hg, particularly MeHg, to evaluate the Hg exposure risk via rice ingestion is urgently needed. The objectives of this study were (1) to reveal the rice THg and MeHg levels in commercial rice from different areas of Sri Lanka, (2) to assess the exposure to THg and MeHg by the public in Sri Lanka, and (3) to compare and identify the dominant pathway of Hg exposure via rice and fish in Sri Lanka since both are found to be the primary Hg dietary sources (Rothenberg et al., 2014).

Our study can not only reveal the rice Hg levels and exposure of association populations in Sri Lanka but also provide data for the worldwide estimation of Hg (especially MeHg) exposure. Additionally, the relative significance of Hg exposure via rice and fish consumption was identified to protect the populations from Hg exposure via diet, which is also the goal of the Minamata Convention (Basu et al., 2018).

2. Materials and methods

2.1. Sample collection

During August to October 2018, a total of 163 rice samples were collected from different provincial markets in Sri Lanka. The provinces of origin for the samples were North Central (n = 28), Western (n = 28), Central (n = 17), Northern (n = 15), Eastern (n = 20), Southern (n = 11), Northern Western (n = 12), Uva (n = 15), and Sabaragamuwa (n = 17). For each sample, 0.5 kg rice was purchased and transported into the laboratory.

2.2. Sample preparation

In the laboratory, rice samples were completely homogenized. Then, approximately 30 g were weighed and rinsed with ultrapure water three times, and then freeze dried with a freeze drier (FDU-2110, EYELA, Japan). The sample was crushed into powders with a grinder (IKA-A11 basic, IKA, Germany), and passed through an #80 mesh screen (size: 177 μm), and then stored in zip-locked plastic bags until analysis (Xu et al., 2017).

2.3. THg and MeHg analysis

For rice THg, approximately 0.5 g of dry sample was digested using a mixture of HNO_3 and H_2SO_4 (4:1, v/v) for 2 h in a water bath at 95 °C. After dilution with ultrapure water, THg was detected with cold vapor atomic fluorescence spectroscopy (CVAFS) (USEPA, 2002).

For rice MeHg, approximately 0.5 g of samples was digested using 25% KOH in methanol for 3 h in a water bath at 75–80 °C. MeHg in the samples was then extracted with dichloromethane and back-extracted into water, and measured according to USEPA method 1630 (Liang et al., 1994; USEPA, 2001). The limits of determination were 0.013 μg/kg for THg and 0.005 μg/kg for MeHg.

2.4. QA & QC

Both the THg and MeHg analyses were validated using duplicates, method blanks, matrix spikes, and certified reference materials. For THg, GBW10020 (*Citrus* leaves) was used, and the measured value of THg was 148 ± 7.0 ng/g, in accordance with the certified value of 150 ± 20 ng/g. For MeHg, TORT-2 (Lobster, *Hepatopancreas*) was employed. The obtained values were 151 ± 7.1 ng/g (n = 5) for TORT-2, comparable to the reference value of 152 ± 13 ng/g for TORT-2. The relative standard deviations of the duplicates for THg and MeHg ranged from 2.2 to 14% and 3–13%, respectively. Meanwhile, recoveries of the matrix spikes of THg and MeHg ranged from 90 to 110% and 93–98%, respectively.

2.5. Data analysis

One-way ANOVA and Student's T-test were performed with SPSS 22 (Standard, California, USA) to analyze the significant differences in rice THg and MeHg in different provinces. All figures were plotted with Origin 9 (©OriginLab Corporation). Monte Carlo simulation was used to perform the uncertainty and sensitivity analyses with Crystal ball© (Oracle, Redwood City, CA, USA).

2.6. Dietary Hg exposure

THg and MeHg intake (μg/kg bw/day) were calculated by the following equation.

$$DI = \frac{C_{\text{MeHg or THg}} \times IR \times 10^{-3}}{BW}$$

where C is the THg or MeHg concentration (ng/g) in food stuff and IR is the daily consumption rate of food stuff (g/day). BW is 60 kg, which is the average body weight of the adult individuals in Sri Lanka (Diyabalanage et al. 2016, 2017; Jinadasa et al., 2018). The rice intake rate of different provinces in Sri Lanka is obtained from questionnaires.

3. Results and discussion

3.1. Hg concentration in rice

THg in commercial rice from Sri Lanka exhibited a lognormal distribution (Fig. 1). The geometric mean concentration of THg in all samples (n = 163) was 1.73 ± 0.89 ng/g, ranging from 0.21 to 6.13 ng/g. No sample exceeded the maximum limit of 20 ng/g recommended by the Chinese Government (GB2762 2017), indicating a safe Hg level in Sri Lanka. Frequency counts of MeHg showed a lognormal distribution (Fig. 2). MeHg concentrations in samples range from 0.03 to 3.81 ng/g, with a geometric mean of 0.51 ± 0.37 ng/g.

Generally, THg in Sri Lankan rice is at a low level. The mean THg

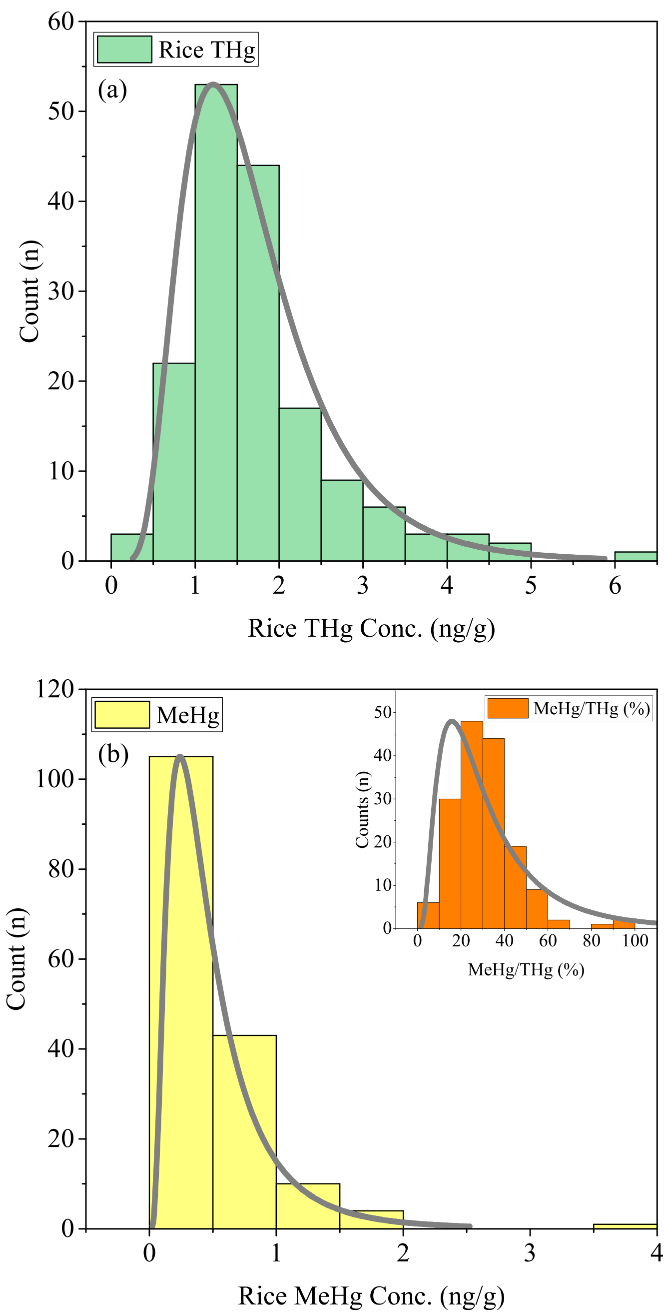

Fig. 1. Distribution of THg (**a**), MeHg, and MeHg/THg (**b**) in rice from Sri Lanka.

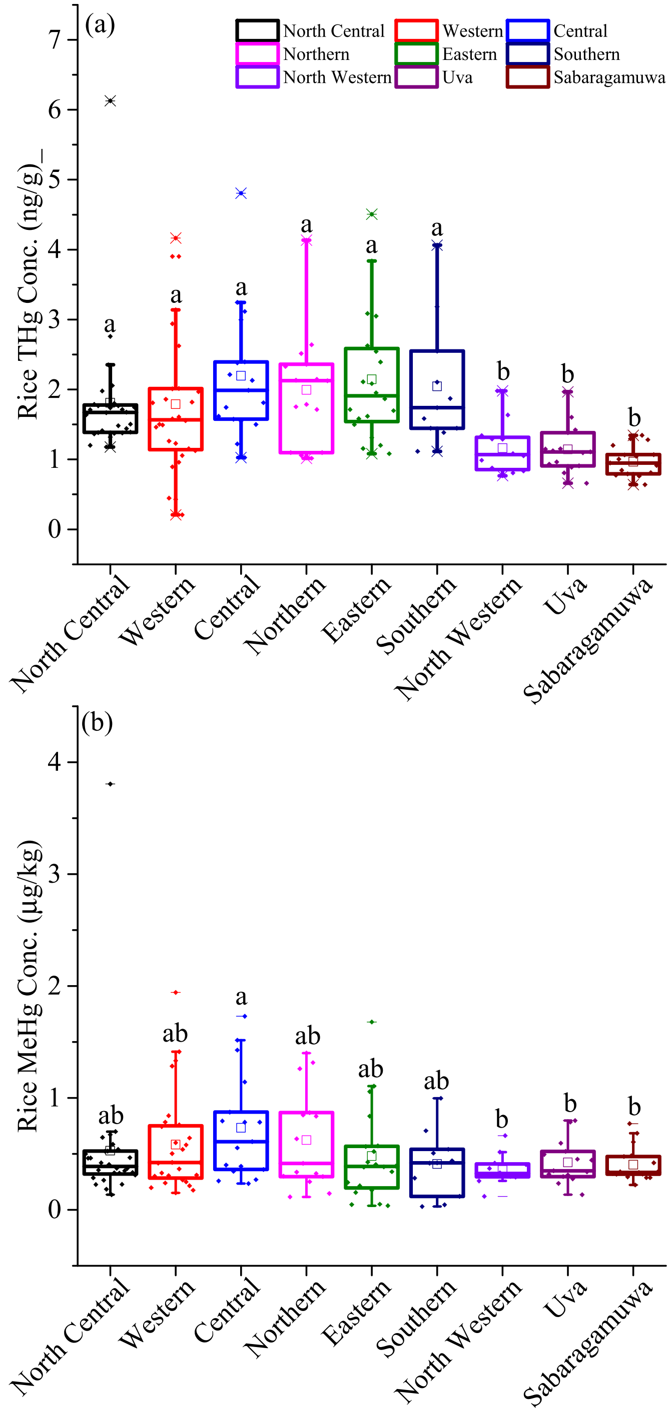

Fig. 2. Comparison of THg (**a**) and MeHg (**b**) in rice from different provinces in Sri Lanka.

in Sri Lankan rice was compatible with that in Madagascar (mean: 1.1 ng/g, range: 0.36–4.7 ng/g), the lowest reported THg in the world (Rothenberg et al., 2015), suggesting the low Hg geological background of rice producing areas in Sri Lanka. Additionally, the mean rice THg in Sri Lanka was lower than those collected in worldwide markets: China (mean: 3.4 ng/g, range: undetected to 17.8 ng/g (Zhang et al., 2014), Europe (mean: 3.04 ± 2.7 ng/g, range: 0.53–11.1 ng/g) (Brombach et al., 2017), Spain (mean: 4.48 ng/g, range: 2.15–7.25 ng/g) (da Silva et al., 2013), Kuwait (6.4 ± 2.4 ng/g, range: 4–14 ng/g) (Jallad, 2015), and Cambodia (mean: 8.14 ng/g, range: 6.16–11.7 ng/g) (Cheng et al., 2013b). The mean THg levels of rice in Sri Lanka are definitely several times lower than those in Hg mining areas (Qiu et al., 2013; Xu et al., 2018), coal fired power plants (Xu et al., 2017), chemical plants (Cheng et al., 2013a), compact fluorescent lamp manufacturing areas (Liang et al., 2015), e-waste areas (Zhao et al., 2010), and industrial runoff areas (Cao et al., 2010).

The mean rice MeHg in this study was slightly higher than that obtained from non-polluted sites in Madagascar (mean: 0.12 ng/g, range: 0.015–1.1 ng/g) but lower than that collected in commercial rice in worldwide markets: Europe (mean: 1.91 ± 1.07 ng/g, range: 0.110–6.45 ng/g) (Brombach et al., 2017), China (mean: 2.47 ng/g, range: 0.13–18.2 ng/g) (Li et al., 2012), and Cambodia (mean: 1.44 ng/g, 1.17–1.96 ng/g) (Cheng et al., 2013b). The rice MeHg in Sri Lanka can be more than 31 times lower than that collected in Hg mining areas with the mean MeHg of 16 ng/g in (Rothenberg et al., 2014). Since MeHg in rice grain originates from soil (Zhang et al., 2010b), low levels of MeHg in rice suggest the low MeHg production in paddy soils in Sri Lanka.

The average MeHg% (MeHg/THg%) of all the 163 rice samples were 31.8 ± 18%, ranging from 0.7% to 100%. However, the mean MeHg% in this study was lower than that obtained in worldwide markets (Brombach et al., 2017; Cui et al., 2017; Li et al., 2012). Nevertheless, compared with the MeHg% of other plant like corn (0.10–12%), rape (0.1–0.3%), and cabbages (0.031–3.3%) (Qiu et al., 2005), the MeHg% of rice should be emphasized since rice is consumed as the staple food in Sri Lanka. Further study should be carried out on revealing the factors influencing low MeHg production in Sri Lankan paddy soil and on long-term rice MeHg exposure in Sri Lanka. MeHg% is also influenced by the MeHg production rate in soil, which may be due to the lower atmosphere Hg deposition (Zhao et al., 2016), low MeHg methylation microorganisms and gene abundance (Ma et al., 2019; Vishnivetskaya et al., 2018), flooding time (Wang et al., 2014), Fe and sulfur (Marvin-DiPasquale et al., 2014), soil pH (Rothenberg and Feng, 2012), the source of MeHg in organisms (Xu et al., 2019b), and other factors influencing paddy soil (Li et al., 2019; Wang et al., 2016; Windham-Myers et al., 2014; Xu et al., 2019a).

Significant provincial differences in rice THg can be observed in this study (Fig. 3). The mean rice THg concentrations in North Western (1.16 ± 0.37 ng/g), Uva (1.14 ± 0.34 ng/g), and Sabaragamuwa (0.97 ± 0.22 ng/g) Provinces were remarkably ($p < 0.05$) lower than those in other regions, which may be due to the low Hg emissions from Hg pollution sources and the low Hg accumulating rice varieties in these three provinces (Rothenberg et al., 2014; Xu et al., 2017). Regional differences in MeHg can also be found, and the average rice MeHg from Central (0.73 ± 0.47 ng/g) was significantly higher ($p < 0.05$) than that those from North Western (0.36 ± 0.14 ng/g), Uva (0.42 ± 0.19 ng/g), and Sabaragamuwa (0.40 ± 0.15 ng/g) Provinces (see Fig. 4).

According to the pericarp color, the rice samples from Sri Lanka markets were categorized into red and white rice, and no significant difference ($p = 0.86$) in THg between red (mean: 1.74 ± 0.87 ng/g, range: 0.43–4.51 ng/g) and white rice (mean: 1.71 ± 0.92 ng/g, range: 0.21–6.13 ng/g) was observed. There was no significant difference (*t*-test, $p = 0.19$) in MeHg between red (mean: 0.47 ± 0.32 ng/g, range: 0.03–1.94 ng/g) and white rice (mean: 0.55 ± 0.49 ng/g, range: 0.05–3.81 ng/g). On the basis of rice shape, rice from Sri Lanka was roughly divided into three types (intermediate bold, short round, and others). Although there were no obvious differences in THg, MeHg among the three types, levels of intermediate bold and short round types were significantly ($p < 0.01$) higher levels than those of others types (Fig. S1). Furthermore, commercial rice in this study can be categorized into 35 brands (Fig. S2), and the average THg concentrations of different brands ranged from 0.85 ng/g (Nipuna rathu kakulu) to 4.81 ng/g (Kosala white kakulu); meanwhile, the mean MeHg concentrations of different brands varied from 0.13 ng/g (Lake Nadu) to 1.43 ng/g (Kosala white kakulu).

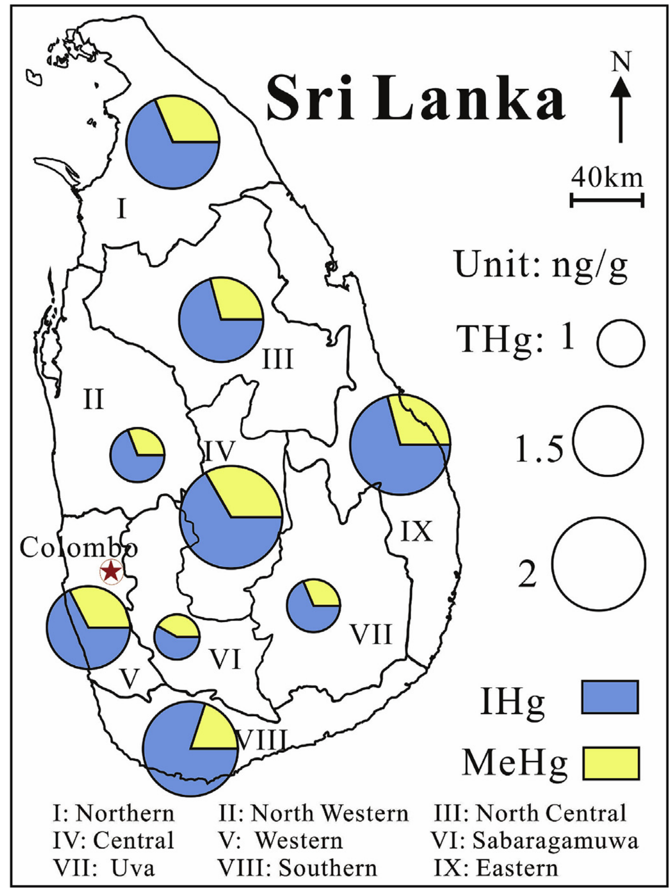

Fig. 3. Geographical distributions of THg, MeHg, and IHg in rice from different provinces in Sri Lanka.

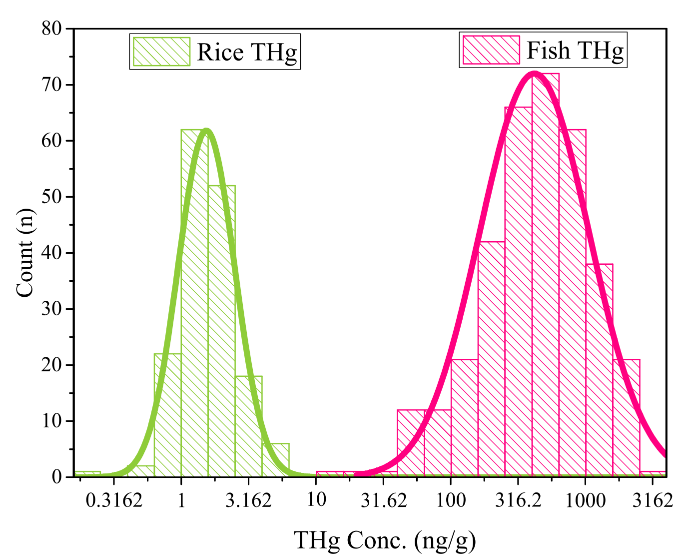

Fig. 4. Distribution characteristics of THg in rice and fish in Sri Lanka.

3.2. Dietary intake of Hg via rice and fish

For Sri Lankans, who have been consuming locally produced rice, provincial THg and MeHg levels, together with the different rice intake rates, lead to regional differences in rice THg and MeHg exposure levels (Fig. 5a and b). Since inorganic Hg is hundreds times less toxic than MeHg (Chen et al., 2009; Kungolos et al., 1999), low dietary intake of THg rice may not be a potential risk for Sri Lankan rice consumers under this low level of THg; thus, IHg intake will not be considered in this study.

For rice THg exposure, residents in Northern Central (0.013 ± 0.0065 μg/kg bw/day), Central (0.015 ± 0.0062 μg/kg bw/day), Eastern (0.014 ± 0.0058 μg/kg bw/day), and Southern (0.015 ± 0.0064 μg/kg bw/day) Provinces met notably higher (one-way ANOVA, $p < 0.05$) levels than residents in other provinces. Rice THg exposure in all provinces was lower than the provisional tolerable weekly intake (PTWI) of 4 μg/kg bw/day (equal to 0.57 μg/kg bw/day) for inorganic Hg set by the joint FAO/WHO Expert Committee on Food Additives (JECFA) (JECFA, 2010).

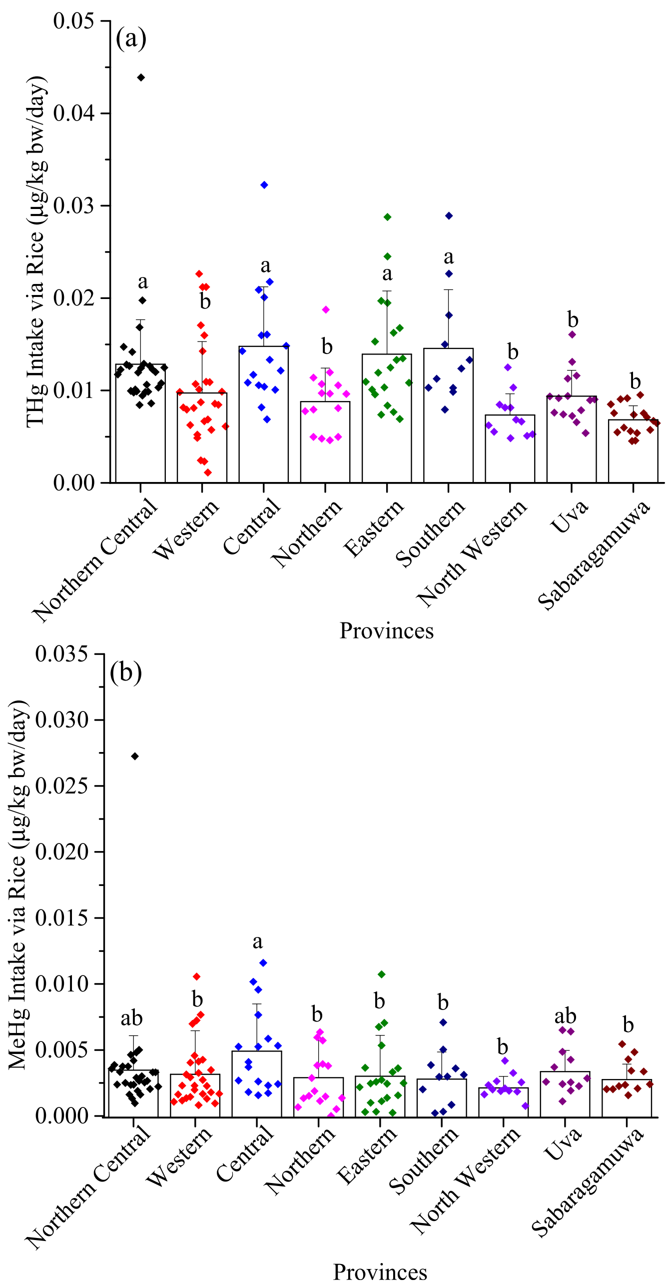

Fig. 5. Comparisons of THg (**a**) and MeHg exposure (**b**) via commercial rice intake for Sri Lankans from different provinces.

For rice MeHg exposure, the highest exposure levels can be found in Central Province (0.0049 ± 0.0032 μg/kg bw/day) and are obviously higher (one-way ANOVA, $p < 0.05$) than those in Western (0.032 ± 0.024 μg/kg bw/day), Northern (0.0027 ± 0.0021 μg/kg bw/day), Eastern (0.0031 ± 0.0026 μg/kg bw/day), Southern (0.0029 ± 0.0021 μg/kg bw/day), Northern Western (0.0023 ± 0.00086 μg/kg bw/day), and Sabaragamuwa Provinces (0.0030 ± 0.0013 μg/kg bw/day) (see Table 1). Rice MeHg exposures in all provinces were lower than the reference dose (RfD) of 0.1 μg/kg bw/day for MeHg by the USEPA (USEPA, 2000). Although the rice intake rate was higher than that in China, due to the lower MeHg levels in Sri Lankan rice, the rice MeHg exposure levels of Sri Lankans were compatible with that of Chinese (0.005 μg/kg bw/day), however, much higher than that of Europeans (0.0004 calculated on data from FAO and Bromach et al.) (Brombach et al., 2017; FAOSTAT, 2019; Li et al., 2012).

To identify the dominant pathway of Hg exposure, both rice and fish intake rates were taken into consideration. We categorized both fish and rice eating populations into three subgroups (low, average, and high intake groups), and their associated THg and MeHg exposure were estimated (Table 2). In the present study, concentrations of THg in fish were derived from data in publications (Jayasinghe et al., 2018; Jinadasa et al., 2014). The average THg in fish was 581 ± 491 ng/g, ranging from 10 to 2580 ng/g. The fish THg was more than two magnitudes higher than that in rice (Fig. 4). It is well known that more than 90% of THg is in MeHg form (ATSDR, 1999; Borum et al., 2001; Hall et al., 1997; Spry and Wiener, 1991), thus, MeHg was quantified as 90% of THg in fish. Correspondingly, MeHg in Sri Lankan fish was 523 ± 442 ng/g, ranging from 9 to 2322 ng/g.

The results showed that rice THg exposures were 0.0078 ± 0.0041, 0.0086 ± 0.0045, and 0.014 ± 0.0073 μg/kg bw/day for low (272 g/day), average (300.6 g/day), and high rice consumption (490 g/day) populations, respectively. Meanwhile, fish THg exposure was 0.42 ± 0.35, 0.66 ± 0.56, and 2.57 ± 2.17 μg/kg bw/day for low (43 g/day), average (265 g/day), and high fish consumption (265 g/day) populations, respectively. Comparisons of rice and fish have shown that the average THg exposure via rice consumption was approximately 75 times lower than those of fish consumption. Even in the high rice consumption and low fish consumption populations, rice THg exposure is approximately 30 times lower than fish THg exposure, suggesting that fish play a more important role in dietary THg exposure in Sri Lanka.

Rice MeHg exposures were 0.0024 ± 0.0020, 0.0026 ± 0.0022, and 0.0042 ± 0.0035 μg/kg bw/day for low, average, and high rice eating populations, respectively. Generally, rice MeHg can contribute to 4.2% of RfD (0.1 μg/kg bw/day) even in high rice consumption populations (490 g/day), suggesting low MeHg exposure via rice consumption. Moreover, fish MeHg exposures were 0.37 ± 0.32, 0.60 ± 0.50, and 2.31 ± 1.95 μg/kg bw/day in low, average, and high fish consumption populations, respectively. Even in low fish consumption populations, fish MeHg exposure can be 3.7 times that of RfD, which may pose a potential risk to human health. Dietary MeHg exposure through fish intake is 71 times higher than that through rice intake. Even in high rice and low fish intake populations, MeHg exposure from rice was approximately 1.1% of that in low fish intake populations (Fig. 6), suggesting that fish, not rice, plays the dominant role in Sri Lankan's dietary MeHg exposure.

Recently, studies revealed that only a part of ingested Hg can enter into the bloodstream, which is considered as the

Table 1
THg and MeHg in rice from different provinces in Sri Lanka and associated provincial Hg exposure.

Provinces	Rice intake (g/day)	THg (ng/g)	Range	MeHg (ng/g)	Range (ng/g)	THg intake (μg/kg bw/day)	Range (μg/kg bw/day)	MeHg intake (μg/kg bw/day)	Range (μg/kg bw/day)
North Central	430	1.79 ± 0.67	1.17 6.13	0.49 ± 0.36	0.13−3.81	0.013 ± 0.0065	0.0084−0.044	0.0038 ± 0.0047	0.00097−0.027
Western	326	1.79 ± 1.03	0.21−4.16	0.61 ± 0.61	0.15−1.94	0.0097 ± 0.0056	0.0011−0.023	0.0032 ± 0.0024	0.00082−0.01056
Central	402	2.21 ± 0.98	1.03−4.81	0.74 ± 0.54	0.23−1.73	0.0015 ± 0.0062	0.0069−0.032	0.0049 ± 0.0032	0.0016−0.012
Northern	272	2 ± 0.81	1.02−4.14	0.66 ± 0.66	0.11−1.40	0.0090 ± 0.0037	0.0046−0.01874	0.0028 ± 0.0020	0.00052−0.00635
Eastern	383	2.18 ± 1.12	1.08−4.51	0.5 ± 0.49	0.037−1.68	0.014 ± 0.0058	0.0069−0.029	0.0031 ± 0.0026	0.00023−0.011
Southern	427	2.04 ± 0.89	1.11−4.06	0.41 ± 0.29	0.029−1.00	0.015 ± 0.0064	0.0079−0.029	0.0029 ± 0.0021	0.00020−0.00709
North Western	379	1.16 ± 0.37	0.77−1.98	0.36 ± 0.14	0.12−0.66	0.0073 ± 0.0023	0.0048−0.013	0.0023 ± 0.00086	0.00076−0.0042
Uva	490	1.15 ± 0.34	0.66−1.97	0.43 ± 0.2	0.13−0.80	0.0094 ± 0.0027	0.0054−0.016	0.0035 ± 0.0173	0.0011−0.0065
Sabaragamuwa	425	0.97 ± 0.22	0.64−1.34	0.4 ± 0.17	0.22−0.77	0.0068 ± 0.0016	0.0045−0.0095	0.0030 ± 0.0013	0.0016−0.055

Table 2
Dietary THg and MeHg exposure via rice and fish consumption, and MeHg exposure of rice and fish under consideration of bioaccessibility.

Categories	Intake rate	THg (μg/kg bw/day)	Range (μg/kg bw/day)	Without consideration of bioaccessibility		Consideration of bioaccessiblity	
				MeHg (μg/kg bw/day)	Range (μg/kg bw/day)	MeHg (μg/kg bw/day)	Range (μg/kg bw/day)
Rice intake	Low (272 g/day)[a]	0.0078 ± 0.0041	0.00094−0.028	0.0024 ± 0.0020	0.00013−0.017	0.0010 ± 0.000836	0.000054−0.0007106
	Average (300.6 g/day)[b]	0.0086 ± 0.0045	0.0010−0.031	0.0026 ± 0.0022	0.00014−0.019	0.0011 ± 0.00092	0.000059−0.0079
	High (490 g/day)[a]	0.014 ± 0.0073	0.0017−0.050	0.0042 ± 0.0035	0.00024−0.031	0.0018 ± 0.0015	0.00010−0.013
Fish intake	Low (43 g/day)[c]	0.42 ± 0.35	0.0072−1.85	0.37 ± 0.32	0.0065−1.66	0.15 ± 0.13	0.0027−0.69
	Average (68.3 g/day)[b]	0.66 ± 0.56	0.011−2.93	0.60 ± 0.50	0.010−2.64	0.25 ± 0.21	0.0042−1.1
	High (265 g/day)[d]	2.57 ± 2.17	0.044−11.4	2.31 ± 1.95	0.040−10.3	0.97 ± 0.82	0.017−4.31

[a] Data were collected in this study.
[b] (FAOSTAT, 2019).
[c] (Jinadasa et al., 2018).
[d] (Rathnasuriya et al., 2018).

bioaccessibility (Bradley et al., 2017; Moreda-Piñeiro et al., 2011). Bioaccessibility depends on the soluble fractionation by the end of the digestion processes and could be applied as a conservative assessment of bioavailability (Lin et al., 2019). Bioaccessibility of MeHg in rice and fish is the important sources that substantially influences dietary fish and rice exposure (Gong et al., 2018). Based on the Hg speciation results in this study and MeHg bioaccessibility of rice (41.8%) by Gong et al., (2018) and fish (57%) reported by Afonso et al., (2015), dietary intake of MeHg among different rice and fish consumption populations were evaluated under the consideration of bioaccessibility. The mean rice MeHg exposures of the low, average, and high rice consumption groups were 0.0010 ± 0.00084, 0.0011 ± 0.00092, and 0.0018 ± 0.0015 μg/kg bw/day, respectively. This result means that the actual MeHg exposure via rice, even high rice consumption, contributed to only 1.8% of RfD (0.1 μg/kg bw/day). Meanwhile, the mean fish MeHg exposures in the low, average, and high fish consumption populations were 0.15 ± 0.13, 0.25 ± 0.21, and 0.97 ± 0.82 μg/kg bw/day, respectively. Considering the bioaccessibility of MeHg in fish, even the low fish consumption groups meet an exposure level higher than RfD, which means fish consumption may pose a potential threat, while rice consumption plays a less significant role in MeHg exposure.

Because of their lack of a mature blood brain barrier (Keating, 1997), children may have continued susceptibility to Hg. Thus, the evaluation of MeHg exposure risk is especially important in children. We assumed that one baby at one year eats 75 g rice (Brombach et al., 2017) and 0.66 g fish per day (Xue et al., 2012), and 9.25 kg (WHO, 2006). For a one-year-old baby, the average MeHg intake via rice and fish was 0.0041 ± 0.003 μg/kg bw/day and 0.037 ± 0.0031 μg/kg bw/day, respectively. When consuming the rice and fish with the highest MeHg concentrations (3.81 ng/g for rice and 2322 ng/g for fish), fish MeHg exposure was (0.17 μg/kg bw/day) higher than RfD (0.1 μg/kg bw/day), which was also more than 5 times that of rice MeHg exposure (0.031 μg/kg bw/day). Even under the consideration of MeHg bioaccessibility in rice (41.8%) and fish (57%), fish MeHg exposure (0.094 μg/kg bw/day) was more than 7 times that via rice (0.013 μg/kg bw/day). Although fish MeHg exposure is lower than the RfD value (approximately 94% of RfD) under the consideration of MeHg bioaccessibility, other food sources may also contribute to MeHg exposure, adding up to exposure values higher than RfD.

4. Conclusion

We first conducted a nationwide survey of rice THg and MeHg in Sri Lanka. It can be concluded that the concentrations of THg and MeHg in rice grain were low, resulting in lower Hg exposure through rice intake. In future study, it is worthwhile to conduct work on THg and MeHg concentrations in the paddy soil of Sri Lanka, and revealing the dominant Hg emission source and Hg deposition in Sri Lanka. Due to the higher MeHg% in rice than in other crops, attention should also be paid to long-term MeHg exposure via rice intake. Compared to the average MeHg exposure from fish, that from rice was two magnitudes higher. Hence, fish intake contributed to the dominant MeHg exposure in Sri Lanka. Future studies in Sri Lanka should focus on fish MeHg exposure and

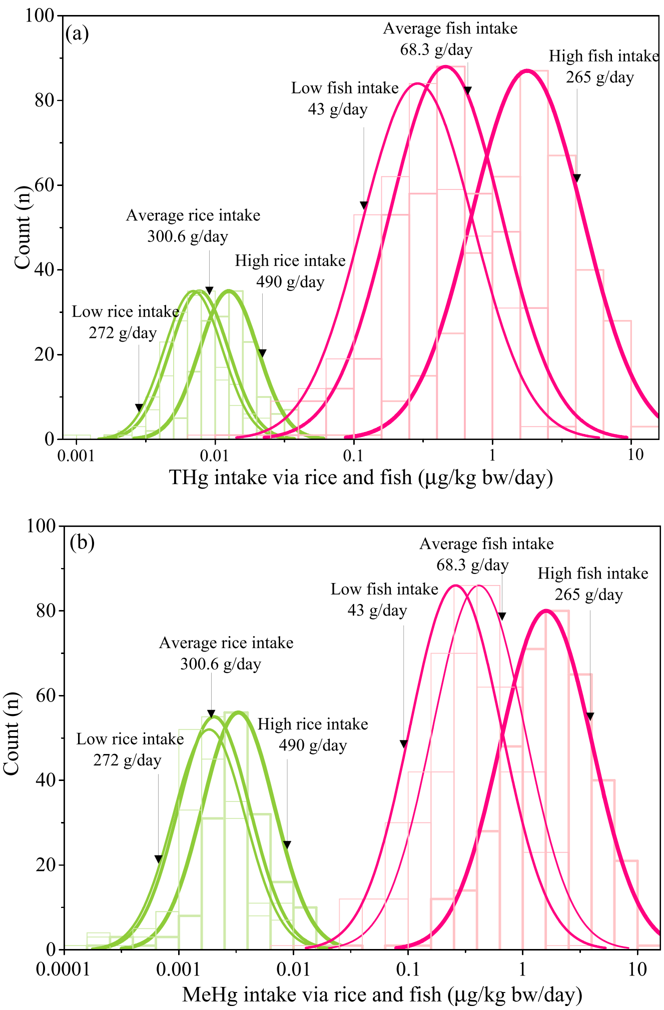

Fig. 6. Comparisons of THg exposure in rice and fish consumption populations (**a**), and Comparisons of MeHg exposure in rice and fish consumption populations (**b**).

the health impacts after long-term fish MeHg exposure, especially in vulnerable populations, such as babies or pregnant women. In addition, some diet strategies should be studied and proposed to mitigate MeHg exposure levels.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2019.124749.

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