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Public health risk of mercury in China through consumption of vegetables, a modelling study

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ABSTRACT

Sample measurement of mercury (Hg) contents is a common method for health risk assessment of Hg through vegetable consumption in China. In the present work, we undertook the first modelling study which produced consistent health-risk maps for the whole eastern China. Regional maps of Probable Daily Intake (PDI) of Total mercury (THg) and Methylmercury (MeHg) over the studied area were produced, which were important for the researchers and policy-makers to evaluate the risk and to propose mitigation measures if necessary. The model predictions of air-borne Hg(0) concentrations agreed well with the observations and simulated Hg distribution over China as reported elsewhere. Our calculated PDIs of THg in vegetables were also comparable to those reported in the literature. There was 19% of the studied area with PDIs > 0.08 μg kg^{-1} bw d^{-1} [half of the reference dose (RfD)]. The PDI for THg (MeHg) varied from 0.034 (0.007) to 0.162 (0.035) μg kg^{-1} bw d^{-1} with an average of 0.058 (0.013) μg kg^{-1} bw d^{-1}. The highest calculated PDIs of THg over China was equal to the RfD, while the calculated PDIs of MeHg were well below the RfD of 0.1 μg kg^{-1} bw d^{-1}. The health risk was of concern through consumption of THg in leafy vegetables, rice/wheat and fish in Liaoning Provinces, Hunan, Zhejiang and Guizhou Provinces, with the associated PDIs exceeding the RfD. Despite this, the heath risk of MeHg exposure for the general population in southern China from the same foodstuff consumption was not a concern. The contribution of consumption through leafy vegetation should be considered when THg and MeHg exposures to the population are evaluated. The results improve our understanding in managing public health risk in China especially in large cities with high population, and thus have important contribution to enhance sustainable urbanization as one of the principle goals under the framework of the Nature-Based Solution (NBS).

1. Introduction

Heavy metals are ubiquitous in the environment. Humans are exposed to them through various pathways including inhalation of contaminated air, food consumption, drinking water, and dermal contact of soil, etc. Mercury is one of the highly toxic heavy metals. Its organic form - Methylmercury (MeHg) is a potent toxicant and has received particular attention. Consumption of foodstuff including fish (Mergler et al., 2007; Díez, 2009; Yi et al., 2011; Kampalath and Jay, 2015), vegetables (Wang et al., 2005; Sipter et al., 2008) and crops (Zhang et al., 2010; Li et al., 2012) plays an important role in Hg exposure of humans. East Asia, including China, is the largest source region of global anthropogenic Hg emissions (Pan et al., 2010) and therefore it is critical to have a better understanding of the regional emission impacts on human health risk through foodstuff consumption.

To provide essential human nutrients, vegetables are an important source of food and an important part of the human diet in China (Wang et al., 2012). A few of studies on health risks of Hg through vegetable consumption in China have been undertaken. In Guizhou Province (southeastern China) where Hg-mining and artisanal smelting activities are located, vegetable consumption is the second most important contributor (22–42%) to THg exposure (Zhang et al., 2010). Near a zinc smelting plant in Liaoning Province (northeastern China), the maximum weekly intake of THg through vegetable consumption is 7% of the provisional tolerable weekly intake for adults (Zheng et al., 2007a). Wang et al. (2012) concluded that toxic metal (including Hg) contamination through consumption of vegetables grown around the industrial zones in the Guangdong Province of southern China imposed a high health risk on local inhabitants. Wang et al. (2011a, 2011b) analyzed multi-elemental contents in foodstuffs and evaluated the

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associated health risk in the Guizhou Province and found that Hg and cadmium were the most important contributors to contamination by potentially harmful elements there. The findings of these studies, however, were based on vegetables which were sampled from local supplies such as farmlands, markets and restaurants. Since typical life cycles for vegetables are less than a month, the variability of Hg in different environmental media which affect the Hg contents in vegetables has not been properly considered in these studies, which jeopardizes the health risk assessments. After all, the reliability of the data generated by the laboratory analysis of environmental samples is critical to the assessments (Hellmann and Cheatham, 1989).

In the current study, we investigated the human health risk of total mercury (THg) and MeHg exposure through consumption of leafy vegetables such as [Chinese white cabbage (Brassica chinensis L.), Flowering Chinese cabbage (Brassica parachinensis Bailey), Chinese kale (Brassica alboglabra Bailey), Chinese Amaranth (Amaranthus mangostanus L.), Garland chrysanthemum (Chrysanthemum coronarium L.), Chinese lettuce (Lactuca sativa L.) and Pea sprout (Pisum sativum L.)]. These vegetables are very common in the diet of Chinese people and possess higher contents of heavy metals (including Hg) compared with other kind of vegetables (Liu et al., 2013). The study hypothesis was whether the contribution of consumption through leafy vegetation should be considered when THg and MeHg exposures to the population are evaluated. Since uptake of atmospheric Hg contributed predominantly to the Hg content accumulated in the edible parts of the leafy vegetables (Mosbæk et al., 1988; Ericksen et al., 2003; Qiu et al., 2008; Li et al., 2008; Wang et al., 2011a, 2011b), our study started from investigating the atmospheric Hg over the whole eastern China. The study involved an atmospheric Hg transport model with Hg chemistry and known major emission sources. To increase the representativeness of our results, we used long-term (2007–2009) average air-borne Hg concentrations to calculate the vegetables uptake of Hg. Consistent regional health risk maps of Hg through consumption of leafy vegetables over the whole eastern China were finally obtained and assessed. We undertook the first modelling study which produced the consistent health risk maps for Hg, which are important for the researchers and policy-makers to propose mitigation measures. The detailed methodology involved in the present study is presented in the next section.

2. Material and methods

2.1. Source characterization and transport of Hg

To simulate the atmospheric Hg distribution over China, we used the GEOS-chem model v9-01-02 (http://acmg.seas.harvard.edu/geos) which was coupled to soil emission (Selin et al., 2008) and 2-dimensional slab ocean modules (Strode et al., 2007; Soerensen et al., 2010). The model simulated the behavior of the elemental Hg(0), soluble Hg (II), and particulate Hg(P). Since Hg(0) is the dominant atmospheric Hg species (> 95% of the total Hg concentration; Fitzgerald, 1986; Guo et al., 2008; Sheu et al., 2013), we assumed Hg(0) as total Hg (THg) in air and focused our discussion on Hg(0) here only. The model has been evaluated (Selin et al., 2007; Holmes et al., 2010) and extensively used (Selin et al., 2008; Soerensen et al., 2010; Corbitt et al., 2011; Fisher et al., 2012; Zhang et al., 2014a, 2014b) for Hg and other atmospheric chemical species (Wai et al., 2014; Wai and Tanner, 2014). It was driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS-5) with a horizontal resolution of 0.5° × 0.666° and 72 hybrid sigma pressure levels in vertical. Here, the horizontal resolution was re-gridded to 2° × 2.5° and reduced to 47 vertical levels. Simulations were performed for the period from 2007 to 2009 following 3 years (2004–2006) of model spin-up. Details of the Hg simulation were given by Selin et al. (2007) with updates from Holmes et al. (2010) and Amos et al. (2012). The model assumed that Hg(0) in the atmosphere was oxidized by Br atoms. Atmospheric Hg(II) was partitioned between the gas and aerosol phases, and was photo-reduced to Hg(0) in clouds. Wet deposition followed the scheme from Liu et al. (2001) with recent improvement by Wang et al. (2011a, 2011b). Wet scavenging processes included washout losses in convective updrafts and rainout losses in large-scale precipitation. Dry deposition followed the resistance-in-series scheme from Wesely (1989).

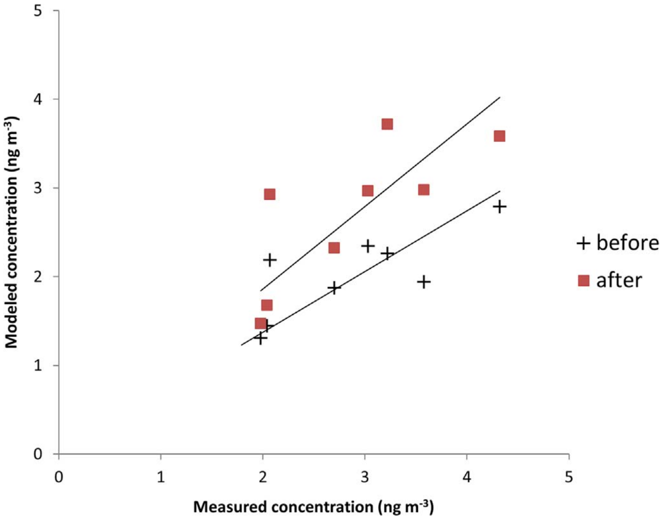

Fig. 1. Comparison of predicted and measured Hg(0) concentrations (ng m^{-3}) before and after the adjustment of emission strengths.

We used the emission inventory as described in detail in Selin et al. (2008). Briefly, the inventory included a modified GEIA global emissions of anthropogenic Hg(0) (from non-ferrous metal smelting and coal combustion) in 2000, biomass burning and artisanal mining emissions. When comparing with the Hg(0) measurements available in the literature (more discussion in next Section) especially at those hot-spot areas in south-western China (e.g., the Hunan and Guizhou Provinces and Chongqing), northeastern China (e.g., the Liaoning and Jilin Provinces) and Beijing, however, the predicted Hg(0) concentrations were under-estimated (Fig. 1). A likely reason for these low biases is that the emission factors used and in turn the emissions calculated in this study might be too low for China (Pacyna et al., 2006). We therefore scaled up the emission in northern China (≥ 40°N) by 3 times and those in southern China (< 40°N) by 1.5 times. The ground-level Hg(0) distributions over China is discussed in more detail in the following Section.

2.2. Risk assessment of leafy vegetable consumption

Table 1 summarizes the parameters used in the risk assessment. The leafy vegetables considered here have been mentioned in previous Section. After obtaining the atmospheric Hg(0) distribution over China, the THg and MeHg contents (mg per kg of dry vegetables) in leafy

Table 1
Summary of parameters used in the risk assessment.

Parameters	Values	References
Air-plant BCF		
THg	23,000	USEPA (1997)
MeHg	5000	USEPA (1997)
Dry to wet weight conversion factor	0.25	Baes et al. (1984) and USEPA (1997)
Average vegetable IR in China	360 g d^{-1}	Zhai (2008)
Average adult bw	60 kg	Zhang et al. (2010)
RfDs		
THg	0.16 μg kg^{-1} bw d^{-1}	OEHHA (2008)
MeHg	0.1 μg kg^{-1} bw d^{-1}	USEPA (2010)

vegetables due to air uptake were modelled by air-plant bio-concentration factors (BCF; USEPA, 1997 and references therein). The BCF approach has been used elsewhere (Giordano et al., 1994; Cangialosi et al., 2008). Since uptake of atmospheric Hg contributes predominantly to the Hg content accumulated in the edible parts of the leafy vegetables (Mosbæk et al., 1988; Ericksen et al., 2003; Qiu et al., 2008; Li et al., 2008; Wang et al., 2011a, 2011b) as mentioned earlier, the contribution from Hg uptake via soil is minimal for leafy vegetables. In fact various studies reported that poor or even negative correlation of metal concentrations existed between vegetable and the corresponding soil (Fytianos et al., 2001; Sharma et al., 2007; Liu et al., 2013). Therefore only the atmosphere-to-plant uptake of Hg was modelled but the soil-to-plant uptake of Hg was not modelled here. The Hg concentrations in vegetables were converted to wet weight according to Baes et al. (1984).

To assess the THg and MeHg exposure through consumption of leafy vegetables, we calculated Probable Daily Intake (PDI; Health Canada, 2008; Zhang et al., 2010; Li et al., 2012) values for the general adult population according to the following formula:

$$\mathrm{PDI}_i = \mathrm{C}_i \times \mathrm{IR/bw} \qquad (1)$$

where PDI is in micrograms per kilogram of body weight (bw) per day; *i* is THg or MeHg; C is the contaminant concentration in leafy vegetables and IR is the intake rate. The average vegetable IR used here is comparable to that used in other studies in China (e.g., Wang et al., 2005; Zhang et al., 2010). To evaluate if there is any potential heath risk of THg and MeHg through vegetables consumption, the PDI is compared to relevant RfD (Table 1). The health risk is not of concern when PDI < RfD.

3. Results and discussion

3.1. Airborne Hg(0) distribution

Fig. 2 shows the 3-year (2007–2009) annual average surface Hg(0) distributions over eastern China. Higher concentrations were predicted in southwestern, central, northeastern China and Beijing, which were consistent to those reported by Quan et al. (2009) and Pan et al. (2010). The high concentrations reflected non-ferrous smelting and coal combustion as the major contributors of Hg emissions in these areas. More inland rural areas in China such as those near the western and northern boundaries of the model domain had concentrations close to the background. Our simulated Hg(0) concentrations were evaluated by the recently available surface measurements in non-urban areas of China and Japan (Table 2 and Fig. 2). The predictions were in good agreement with the measurements with the ratio of modelled to observed Hg(0) concentration of 0.93 and with a correlation coefficient of 0.75 (n = 8), after the modification of emission strength as discussed. Our modelled Hg(0) distribution was also well agreed with the mean concentration of 2.0 ng m^{-3} obtained from a short-term measurement at Dongsha Island (Sheu et al., 2013).

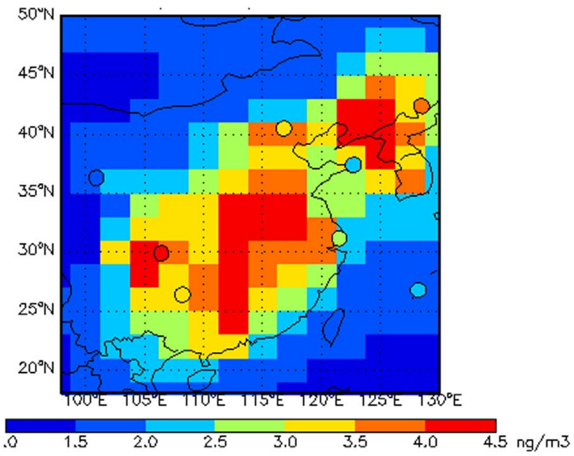

Fig. 2. Pixel map of simulated 3-year (2007–2009) annual average surface Hg(0) distributions over eastern China, with measurements (circles) overlaying on it.

3.2. PDI distribution and health risk assessment

The distributions of calculated PDIs of THg and MeHg (Fig. 3a and b) are similar to the Hg(0) distribution. Our calculated PDIs of THg are comparable to those available in the literature (Table 3). There was 19% of the studied area with PDI/RfD > 0.5. The highest calculated PDI of THg over China was equal to the RfD of 0.16 μg kg^{-1} bw d^{-1}, while the calculated PDIs of MeHg were well below the RfD of 0.1 μg kg^{-1} bw d^{-1}. The PDI for THg (MeHg) varied from 0.034 (0.007) to 0.162 (0.035) μg kg^{-1} bw d^{-1} with an average of 0.058 (0.013) μg kg^{-1} bw d^{-1} over the study area. The results suggested that the associated health risk of Hg through leafy vegetable consumption was in general not of concern over the whole eastern China, except for a small area in the Hunan Province. Higher PDIs were found in southwestern (Guizhou and Sichuan Provinces), central, northeastern China with elevated calculated PDIs at Liaoning and Hunan Provinces (Fig. 3a and b). They were attributed to the large contribution of Hg emissions from non-ferrous metals (zinc and lead) smelting activities there. It was noted that the PDIs for THg in rural areas of western and northern China were even higher than those in UK and Spain (Table 3), which reflected the impacts of high airborne Hg(0) concentrations due to the emissions in China compared to elsewhere of the world (Pan et al., 2010).

3.3. Implications to health risk of total diet in China

Since rice/wheat and fish are the two other important food groups in the Chinese diet, it is important to discuss the health risks from consumption of leafy vegetables, rice/wheat and fish and to understand the relative contribution from leafy vegetables. Table 4 summarizes the PDIs of THg in rice/wheat and fish in China available from the literature. The PDIs vary more than an order of magnitude when compared to one another.

Firstly, we discuss the spatial variations of PDIs of rice/wheat and fish based on the available literature. For the rice/wheat group, Huang et al. (2008) measured THg in wheat grain grown on contaminated soils in an industry city in the Jiangsu Province. The concentration in the wheat was minimal comparing with those in vegetables. It resulted in a low PDI of wheat with the value well below the relevant RfD (0.006 μg kg^{-1} bw d^{-1}, Table 4). To another extreme, the exposure of THg through rice posed a health risk (PDI = 0.23 μg kg^{-1} bw d^{-1}, Table 4) in a Hg polluted area in the Guizhou Province and rice was the staple food there.

As regards the fish group, it is understood that the population living in coastal areas or fisherman would have large daily consumption of fish [e.g. 96 g d^{-1} person^{-1} (Cheng et al., 2009)], which will result in large PDIs on THg (0.49 μg kg^{-1} bw d^{-1} in Table 4). The health risk was of great concern in the coastal areas since the PDI was higher than the RfD. The PDI was higher than but still comparable to that reported for Japan (0.32 μg kg^{-1} bw d^{-1}, Iwasaki et al., 2003) and for Norway (0.08 μg kg^{-1} bw d^{-1}, Mangerud, 2005) where fish consumption was high. However, exposure of THg through fish consumption became much less for the population living in inland China (0.002 μg kg^{-1} bw d^{-1}, Table 4) since fish was not a common diet with a daily consumption of 10 g d^{-1} person^{-1} or less (Li et al., 2012; Zhang et al., 2010). Furthermore, many of the commonly consumed fish species in inland China are fast-growing species and are produced by aquaculture. With a short food chain, they do not accumulate much Hg (Zhang et al., 2010). The low levels of PDI for fish consumption (Table 4) in the Yangtze River area confirmed this.

Table 2
Comparisons of model predictions with measurements.

Location		Sampling Year	Hg (0) Concentrations (ng m^{-3})		Reference
			Measured	Predicted	
Beijing	40.5°N, 116.8°E	2008–2009	3.22	3.72	Zhang et al. (2013)
Chongqing	29.9°N, 106.4°E	2006–2007	4.32	3.58	Yang et al. (2009)
Shanghai	31.2°N, 121.5°E	2009	2.70	2.33	Friedli et al. (2011)
Chengshantou	37.4°N, 122.7°E	2007–2008	2.07	2.93	Nguyen et al. (2011)
Mt. Leigong	26.4°N 108.2°E	2008–2009	3.03	2.97	Fu et al. (2010)
Mt. Waliguan	36.3°N, 100.9°E	2007–2008	1.98	1.47	Fu et al. (2012)
Mt. Changbai	42.4°N, 128.5°E	2005–2006	3.58	2.98	Wan et al. (2009)
Hedo, Japan	26.8°N, 128.2°E	2004	2.04	1.68	Jaffe et al. (2005)

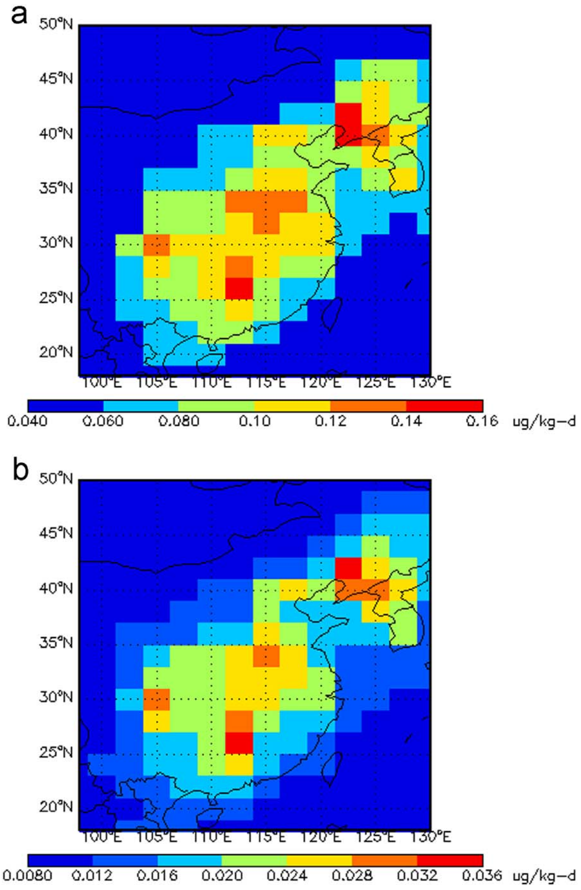

Fig. 3. PDI plots of (a) THg (upper panel) and (b) MeHg (lower panel).

Table 3
Comparison of PDIs from other studies.

Location	THg PDI	Reference
Liaoning Province, China	0.15	This study
	0.05	Zheng et al. (2007a)
Guizhou Province, China	0.11	This study
	0.21[a]	Zhang et al. (2010)
UK	0.05	FSA (2006)
Spain	0.01	Martorell et al. (2011)

[a] Average of PDIs from different areas.

Table 4
DIs of THg in rice/wheat and fish in China.

Location	THg PDI	Reference
Rice/wheat		
Zhejiang Province	0.10	Fu et al. (2008)
Henan Province	0.02[a]	Zhang et al. (2014a, 2014b)
Guizhou Province	0.23[b]	Zhang et al. (2010)
Jiangsu Province	0.006[c]	Huang et al. (2008)
Fish		
Liaoning Province	0.02[d]	Zheng et al. (2007b)
Yangtze River	0.03	Yi et al. (2011)
Guizhou Province	0.002	Zhang et al. (2010)
Zhejiang Province	0.49[e]	Cheng et al. (2009)

[a] Average of brown rice and milled rice.
[b] Average of PDIs from different areas.
[c] Average of urban and rural areas.
[d] As sea products.
[e] Average of PDIs from housewives and children.

We now evaluate the health risks based on the PDIs of THg through consumption of leafy vegetables, rice/wheat and fish. The health risk is of concern in the Liaoning, Hunan, Zhejiang and Guizhou Provinces. For the Liaoning and Hunan Provinces, the PDIs for vegetables were very close to the relevant RfD (i.e. PDI/RfD > 0.9) such that the combined exposure of THg through consumption of leafy vegetables, rice/wheat and fish would likely lead to exceeding of RfD. For the Zhejiang Province, the PDI for vegetable was calculated to be 0.1 μg kg^{-1} bw d^{-1}. With a high PDI for rice (Table 4) and due to its coastal location, the resulted PDI for leafy vegetables, rice/wheat and fish exceeded the relevant RfD and thus the health risk was of great concern. Although the PDI for fish consumption in Guizhou was very small, the human risk through consumption of vegetables and rice was a concern due to the associated high PDIs (Table 4). Unfortunately, the limited data available prevented us to extend our discussion to other Provinces. Nevertheless, we conclude that leafy vegetable consumption could play an important role on the health risk of THg exposure from the total diet in general since the minimum PDI over the whole eastern China contributed to 25% of the RfD.

For MeHg, Li et al. (2012) reported the PDIs through rice and fish consumption in six Provinces in southern China and found that the maximum PDI to be about 0.045 μg kg^{-1} bw d^{-1} in the Guangdong Province. Given the maximum PDI of 0.035 μg kg^{-1} bw d^{-1} for MeHg as mentioned above and the relevant RfD, there is no evidence that the PDI of MeHg through consumption of leafy vegetables, rice and fish in southern China should cause a great concern.

4. Concluding remarks

We introduced a novel method to assess the public health risk of Hg in China through consumption of leafy vegetables by mathematical models, which was not available in the literature to the best understanding of the authors. The modelled Hg(0) concentrations and PDIs were comparable to those reported in the literature. Our results provided the first consistent health-risk map over the whole eastern China, while previous studies were mainly based on local sample measurements within a short period of time. The combined health risks based on the THg/MeHg PDIs through consumption of leafy vegetables, rice/

wheat and fish in different areas of China were evaluated in the present work. It is recommended to include the soil uptake of Hg, although is believed to have minor contributions to leafy vegetables, in future relevant studies to make them more comprehensive. The results improve our understanding in public health risk management in China especially in large cities with high population. It contributes to enhance sustainable urbanization which is one of the principle goals addressed by the Nature-Based Solution (NBS; European Commission, 2015).

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