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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

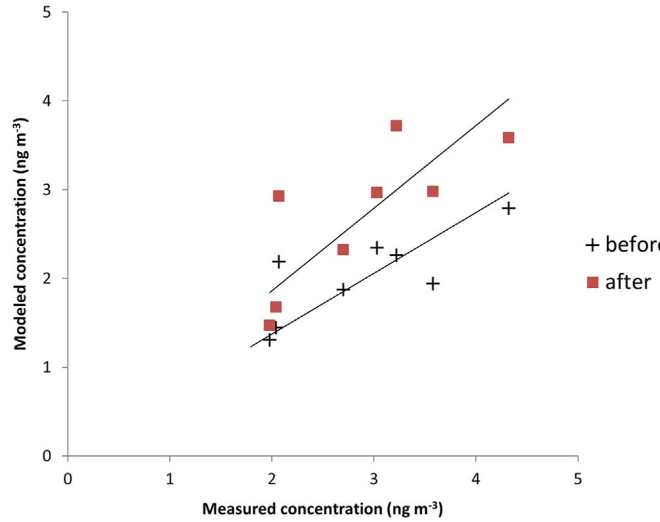

Fig. 1. Comparison of predicted and measured Hg(0) concentrations (ng m^{-3}) before and after the adjustment of emission strengths.

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).

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)