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Impacts of farmed fish consumption and food trade on methylmercury exposure in China

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ABSTRACT

The global pollutant mercury (Hg), especially as methylmercury (MeHg), threatens human and ecosystem health. But major contributors of MeHg exposure to people in China remain highly debated. We developed the China Mercury Exposure Assessment (CMEA) model, which incorporates human exposure pathways for MeHg and total Hg (THg), the interregional, including international and interprovincial, food trading as well as human physiology to provide a comprehensive system that can evaluate the pathway of Hg forms to human consumers in China. Based on the CMEA model that employed the most comprehensive and recent data, we have found that the Probable Daily Intake (PDI) of MeHg for the Chinese population was 0.057 (range: 0.036–0.091 as 60% confidence interval) $\mu g \cdot kg^{-1} \cdot day^{-1}$, while that of THg was 0.35 (range: 0.22–0.55) $\mu g \cdot kg^{-1} \cdot day^{-1}$. MeHg exposure was dominated by fish intake, especially by farm-raised freshwater fish due to higher consumption of these fish. In 2011, fish intake contributed to 56% to the total MeHg exposure, followed by rice (26%). Consumption of farm-raised fish reduced human exposure to MeHg by 33%. On the other hand, interregional food trading increased MeHg exposure of the Chinese population, as a whole, by 7.6%. The international and interprovincial food trades contributed to 5.1% and 22% of MeHg intake, respectively. For the whole China, fish intake related exposure to MeHg was highest for the Eastern and Northeastern populations, while Tibetans were chronically exposed to the highest MeHg from other sources. Our findings highlight the importance of farmed fish and food trade for MeHg exposure.

1. Introduction

The toxic element mercury (Hg) biomagnifies through food webs and poses health risks to wildlife and people (Clarkson et al., 2003; Grandjean et al., 1997; Harris et al., 2007). Although Hg occurs naturally, human activities have altered its global biogeochemical cycle in the atmosphere, hydrosphere and pedosphere (Driscoll et al., 2013; Mason and Sheu, 2002; Obrist et al., 2018). Hence, extensive research efforts on global inventories and fluxes of anthropogenic Hg among various environmental media have been carried out over the past several decades (Amos et al., 2015; Lamborg et al., 2014; Mason and Sheu, 2002; Nriagu and Pacyna, 1988; Streets et al., 2017). As one of the most toxic forms of Hg, methylmercury (MeHg) can cause developmental delays and decrement in the intelligence quotient (IQ) in children when it crosses the placenta and passes the blood–brain barrier (Clarkson et al., 2003; Debes et al., 2006; Grandjean et al., 1997; NRC, 2000). Adult exposure to MeHg may result in cardiovascular impairment (Roman et al., 2011). As a result, human exposure to MeHg has been related to a loss of workforce productivity and human impairment (Rice et al., 2010; Trasande et al., 2005).

MeHg biomagnifies in aquatic food webs, where fish are at higher trophic levels, resulting in muscle tissue concentrations that are several

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orders of magnitude higher than the water in their ecosystem (Stein et al., 1996). Therefore, in most cases significant human exposure to MeHg occurs via fish intake (FDA, 2000), and has been considered as the single significant dietary source of MeHg in most studies (Giang and Selin, 2016; Selin et al., 2010). For example, intake of fish products accounted for 87–95% of the total Hg (THg) exposure of adults in the U.S. and the U.S. Food and Drug Administration (U.S. FDA) stated that fish were the exclusive source of Hg to people living in the U.S. (FDA, 2000; Haxton et al., 1979; MacIntosh et al., 1996). In the 1950s and 60s, intake of MeHg contaminated fish caught in the Minamata Bay in Japan resulted in the spread of neurological disease that is now known as the Minamata disease (Harada, 1995). Since, there has been a common understanding that fish intake, especially of marine species, acts as the critical exposure pathway (dietary exposure through different food sources) to MeHg and THg (Björnberg et al., 2003; FDA, 2000; Mahaffey et al., 2009).

As an industrial center with extensive coal powered energy production plants, China is the largest contributor to the global anthropogenic THg release (AMAP/UNEP, 2013). Overall, large quantities of THg are released to both the atmosphere (530 Mg in 2014) and aquatic ecosystems (100 Mg in 2012) (Liu et al., 2016b, 2017; Wu et al., 2016), accounting for 25% and 9.1% of the global releases, respectively (Kocman et al., 2017; Pirrone et al., 2010; Streets et al., 2011). While the atmospheric THg emissions in China have continued to increase (Wu et al., 2016), the direct release of THg from wastewater to aquatic environment decreased following 2004, reflecting major improvements in wastewater management (Liu et al., 2016b). However, the application of sewage sludge as a fertilizer in farmland may be a source of direct pollution to rice (Liu et al., 2017). Given the ongoing Hg pollution in China as well as the implementation of the Minamata Convention, progress towards a comprehensive assessment of Hg cycling and human health implications is needed (Hsu-Kim et al., 2018).

Despite severe environmental Hg contamination in China (Feng et al., 2007; Zhang and Wong, 2007), an independent assessment system for Hg (including MeHg and THg) in the human-environment system has not been developed. This is because in China Hg exposure pathways are diverse (Hong et al., 2016; Li et al., 2012; Zhang et al., 2010), and the required data is insufficient. Zhang et al. established the Hg exposure inventory in rural areas of Guizhou province, which included fish, grain, vegetables, livestock and water as exposure pathways, and indicated that rice was the most significant dietary contributor of MeHg exposure (Zhang et al., 2010). The study of Hong et al. and Li et al. also verified this result in the rural areas of Southwest China (Hong et al., 2016; Li et al., 2012). Other MeHg exposure sources such as poultry were also significant in some inland regions of China i.e. Guizhou (Yin et al., 2017). High Hg concentrations found in some traditional medicinal products used in China can also be exposure sources of Hg to the Chinese public (Liu et al., 2018a). Therefore, MeHg exposure pathways in China might be more intricate than in western countries such as the U.S. (FDA, 2000; MacIntosh et al., 1996; Mahaffey et al., 2009; Sunderland et al., 2018). To our knowledge, previous studies in China have ignored the potential importance of the food trade in contributing to Hg exposure. Further, despite severe Hg pollution in waters and soils (Liu et al., 2016a; Zhang et al., 2010), the environmental protection administration in China has not paid sufficient attention to the long-term human exposure to Hg (Zhang and Wong, 2007). Hence, research assessing exposure pathways, including data statistics such as per capita diet intake and food trade are very limited. Thus, it has been difficult to provide a quantitative assessment of human exposure to Hg in relation to consumption patterns, therefore hampering efforts to effectively manage Hg risks for the Chinese public. Notably, the total consumption of both marine and freshwater fish species has been increasing in China mainly due to the improved economic situation and living standards, which enables more people to afford purchasing of fish on regular basis (Cao et al., 2007; NBS, 2012). For example, in total, in 2011, freshwater fish accounted for 78% of all China-consumed freshwater and marine edible products including fish, shellfish etc., and has more than doubled since 1980 (35%; Food and Agricultural Organization, http://www.fao.org/home/en/). In parallel, consumption of rice has been gradually decreasing recently as it is partly replaced by other foods e.g. pork and poultry, due to the improvement of living standards (NBS, 2012).

Therefore, the factors and trends highlighted above provide motivation for the present study. The main objective here was to establish validity of the China Mercury Exposure Assessment (CMEA) model as an effective tool to evaluate the connections between environmental Hg pollution and human exposure and to determine exposure of the Chinese public as a whole, as well as on a region specific basis. The model includes 14 main pathways and 8 regions of China and considers the impact of international and interprovincial food trade. As an effective method to quantify the transfer of a chemical in human body, we have also included the Physiologically Based Pharmacokinetic (PBPK) model into the CMEA framework to verify the validity of the modeled Hg, including MeHg and THg, exposure intake and demonstrate the relationship between concentrations in human blood Hg and exposure of the Chinese population. In the present study we have evaluated the potential exposure risk to Hg for the general population of China and provided a new tool that can be used to understand Hg exposure pathways for humans worldwide. To our knowledge, this work is the first comprehensive nationwide assessment of Hg exposure for Chinese populations, which accounts for 20% of the global human population. This novel assessment evaluates the influence of farmed fish consumption and food trade, as well as the consumption of wild fish, and other products.

2. Materials and methods

2.1. China Mercury Exposure Assessment (CMEA) model

The CMEA model connects the environmental Hg system, including MeHg and THg (Note: in this manuscript Hg represents both MeHg and THg), and human exposure to this Hg in China. The model includes three submodules: I. a MeHg and THg intake model based on Hg monitoring data for different kinds of foods from published peer-reviewed literature and interregional food trade information; II. international and interprovincial trades of food (Supplementary Table S1); and III. a Physiologically Based Pharmacokinetic (PBPK) model, which was used to verify Hg intake modeling results, and simulate blood Hg levels (Fig. 1).

2.2. THg and MeHg intake modeling

The Probable Daily Intake (PDI) of MeHg or THg was calculated to evaluate Hg exposure on the Chinese population (EPA, 1997). A Monte Carlo simulation was incorporated to calculate the probabilistic distribution of PDI following the method described in previous studies (Liu et al., 2016a,b). The calculation method is described below:

$$PDI(x)_{ij} = \sum_{k} (I_{jk} \times C(x)_{ijk})/bw$$

where $PDI(x)_{ij}$ is the probabilistic distribution of Probable Daily Intake of MeHg or THg in different regions in China (μg·kg^{-1}·day^{-1}, Supplementary Fig. S1); I is intake rate (ingestion rate or inhalation rate, g/day) of different exposure pathways in 2011 (Supplementary Tables S2 and S3); $C(x)$ is the probabilistic distribution of MeHg or THg concentrations (ng/g) of different exposure pathways in recent years (Supplementary Material, MeHg and THg concentrations, Table S4). The following abbreviations indicate the following: bw - body weight (kg); i - MeHg or THg; j - different regions in China; k - exposure pathways, including marine fish and other seafood, freshwater fish, rice, wheat, bean, vegetables, pork, poultry, milk, eggs, water, air (through inhalation), soil (through ingestion) and traditional Tibetan