Science of the Total Environment 630 (2018) 222–230

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Bioaccessibility-corrected risk assessment of urban dietary methylmercury exposure via fish and rice consumption in China

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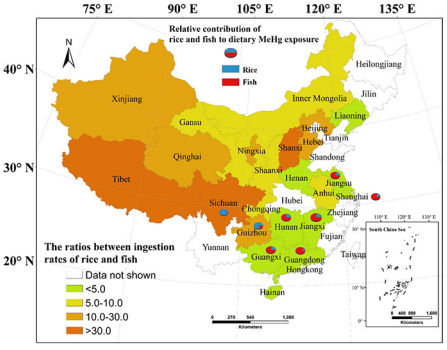
HIGHLIGHTS

- Relative importance of rice versus finfish in methylmercury exposure was assessed.
- Methylmercury bioaccessibility was quantified in market-collected rice and fish.
- Methylmercury bioaccessibility was lower in rice (40%) than fish (61%).
- Rice consumption contributed 7–96% of methylmercury exposure in China.
- Rice consumption was more important in methylmercury exposure in western China.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 November 2017
Received in revised form 18 February 2018
Accepted 18 February 2018
Available online xxxx

Editor: Mae Mae Sexauer Gustin

Keywords:
Mercury
Health risk
Dietary structure
Food safety
Asia

ABSTRACT

The role of seafood consumption for dietary methylmercury (MeHg) exposure is well established. Recent studies also reveal that rice consumption can be an important pathway for dietary MeHg exposure in some Hg-contaminated areas. However, little is known about the relative importance of rice versus finfish in MeHg exposure for urban residents in uncontaminated areas. Especially, the lack of data on MeHg bioaccessibility in rice hinders accurately assessing MeHg exposure via rice consumption, and its importance compared to fish. By correcting commonly used risk models with quantified MeHg bioaccessibility, we provide the first bioaccessibility-corrected comparison on MeHg risk in rice and fish for consumers in non-contaminated urban areas of China, on both city- and province-scales. Market-available fish and rice samples were cooked and quantified for MeHg bioaccessibility. Methylmercury bioaccessibility in rice (40.5 ± 9.4%) was significantly ($p < 0.05$) lower than in fish (61.4 ± 14.2%). This difference does not result from selenium content but may result from differences in protein or fiber content. Bioaccessibility-corrected hazard quotients (HQs) were calculated to evaluate consumption hazard of MeHg for consumers in Nanjing city, and Monte Carlo Simulations were employed to evaluate uncertainty and variability. Results indicate that MeHg HQs were 0.14 (P50) and 0.54 (P90). Rice consumption comprised 27.2% of the overall dietary exposure to MeHg in Nanjing, while fish comprised 72.8%. Employing our bioaccessibility data combined with literature parameters, calculated relative contribution to MeHg exposure from rice (versus fish) was high in western provinces of China, including Sichuan (95.6%) and Guizhou (81.5%), and low to moderate in

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https://doi.org/10.1016/j.scitotenv.2018.02.224

eastern and southern provinces (Guangdong: 6.6%, Jiangsu: 17.7%, Shanghai: 15.1%, Guangxi: 20.6%, Jiangxi: 22.8% and Hunan: 25.9%). This bioaccessibility-corrected comparison of rice versus fish indicates that rice consumption can substantively contribute to dietary MeHg exposure risk for urban populations in Asia, and should be regularly included in dietary MeHg exposure assessment.

1. Introduction

Recent studies demonstrate very high methylmercury (MeHg) production rates in contaminated soil-rice systems, resulting in elevated levels of MeHg in rice grains (Rothenberg et al., 2014; Strickman and Mitchell, 2017; Zhou et al., 2014). Because of the elevated MeHg concentrations in rice grains as well as high rice consumption rates by local residents, rice consumption could be an important route of MeHg exposure and risk for residents in some Hg-contaminated areas of Asian countries (Feng et al., 2008; Rothenberg et al., 2014). For instance in Wanshan, one of the biggest Hg mining areas in China, probable daily intake of MeHg via rice consumption for the local residents ranges from 0.015–0.45 μg/kg day, such that approximately 34% of the consumers exceed the reference dose for MeHg (0.10 μg/kg day, US NRC, 2000; Zhang et al., 2010). This results in elevated non-carcinogenic health hazard (hazard quotient, HQ: 0.15–4.5) in Wanshan. A recent study in Gaohong town, a compact fluorescent lamp manufacturing area in Zhejiang province, reported 2 out of 95 residents with HQ values higher than 1, which is mainly attributed to MeHg exposure via rice (47%) and fish (38%) consumption (Liang et al., 2015). Unfortunately, dietary exposure to MeHg in rice is less documented for general populations in areas that do not contain local Hg point sources. This is especially true for urban consumers, and hinders comprehensive understanding of risks of rice MeHg in Asian countries.

Rice and fish are the two food items generally recognized as the major sources of human exposure to MeHg in China. However, there are not many simultaneous comparisons of MeHg dietary exposure via rice versus fish in China or other Asian countries (Li et al., 2012; Rothenberg et al., 2014; Zhang et al., 2010). This may hinder accurate MeHg health risk assessment for residents in areas where both rice and fish are staple foods. In fact, queries to the UN Food and Agriculture Organization Website (FAO, 2017a; FAO, 2017b) indicated that production in Asia of rice and finfish in 2014 reached 667 and 84 million tonnes, respectively. The rice:finfish production ratio of 8:1 is higher than that in North America (1.7:1) or Europe (0.3:1), highlighting the importance of considering MeHg exposure through both rice and fish consumption for Asian consumers. Among the documented studies comparing MeHg in rice versus fish, most focus on areas heavily contaminated by local Hg sources, such as Hg mining areas or industrially contaminated areas. For example in Wanshan (mining-contaminated), Weining (zinc smelting), Qingzhen (coal-based industry) and Leigong (nature reserve) in Guizhou province, China, rice consumption could contribute to 94–96% of dietary exposure to MeHg, while fish consumption only accounts for 1–2% (Zhang et al., 2010). In Gaohong town (Zhejiang province) known for fluorescent lamp manufacturing, MeHg exposure through rice consumption for local residents is 1.2 times higher than that via fish (Liang et al., 2015). In Fengjiang town (Zhejiang province) a heavily contaminated area due to electronic waste recycling, rice and fish consumption account for 32% and 41% of estimated daily intakes of MeHg, respectively (Tang et al., 2015). These surveys in contaminated areas clearly indicate the need to consider MeHg exposure via rice consumption in addition to fish. However, studies comparing human exposure to MeHg in rice and fish are limited in uncontaminated areas (i.e., areas lacking documented Hg point sources). In order to accurately assess the health risk of MeHg for general populations in Asian countries, there is a need to compare dietary exposure to MeHg in rice versus fish for consumers in these relatively uncontaminated areas (Hong et al., 2016).

Dietary exposure to MeHg in rice and fish depends on MeHg concentrations in food, ingestion rates of food, and MeHg bioaccessibility in food. Recent studies reveal high variability of MeHg bioaccessibility in different food items, e.g., 4–20% in cooked crayfish (Peng et al., 2017), 16–68% in raw fish (He and Wang, 2011; Torres-Escribano et al., 2010; Wang et al., 2013) and 20–93% in boiled/steamed fish (Afonso et al., 2015a; He and Wang, 2011). These findings highlight the need to consider MeHg bioaccessibility when comparing dietary exposure risk among different food items. MeHg bioaccessibility in finfish has been widely reported, and is frequently observed to be high. For instance, MeHg bioaccessibility in meagre (boiled), tuna (boiled), and blue shark (raw) are 93 ± 1%, 57 ± 14% and 98 ± 5%, respectively (Afonso et al., 2015a; Afonso et al., 2015b; Matos et al., 2015). Surprisingly, there are no published data for MeHg bioaccessibility in rice to our knowledge. This hinders accurate comparison of dietary MeHg exposure from rice versus fish (or other food items, for that matter).

We hypothesize that rice, in addition to finfish, plays an important role in dietary MeHg exposure for urban consumers in non-contaminated areas of China. To evaluate this hypothesis, the present study answers three related questions: 1. What is bioaccessibility of MeHg in rice, as well as in local market fish; 2. What is the risk level from MeHg in food (rice and fish) for consumers in Chinese cities lacking documented local Hg point sources (i.e., uncontaminated cities). 3. What is the relative contribution of rice versus fish to dietary MeHg exposure for typical consumers in different Chinese provinces. To answer these questions, we performed original data collection on MeHg bioaccessibility, MeHg concentrations, and fish and rice consumption rates in Nanjing, a typical uncontaminated city of eastern China. We also compiled published data for eight different provinces in China (Fig. 1). We used these data in a bioaccessibility-corrected risk model, to compare MeHg hazard for urban consumers in Nanjing and the eight provinces. Finally, we used Monte Carlo Simulations to obtain statistical distributions for the estimated risk indices for Nanjing residents, and identify the key parameters in the risk model via sensitivity analysis. These results shed light on potential risk of MeHg associated with rice and fish consumption for Asian consumers in uncontaminated areas.

2. Materials and methods

2.1. Study design

Risks and relative contribution of fish and rice to dietary MeHg exposure were assessed on both the city- (Nanjing city) and province-scales (8 provinces in China). Bioaccessibility of MeHg was quantified in market-collected fish or rice. Concentrations of MeHg in fish or rice were quantified or obtained from literature. Their respective ingestion rates were assessed by a questionnaire survey or obtained from literature. The study design is summarized in Fig. 1.

2.2. Sampling and cooking of fish and rice

Rice and commonly consumed fish species were sampled from Nanjing, a typical city in eastern China, for quantifying MeHg bioaccessibility. The sampling in Nanjing city started in April 2016 and lasted for