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Low-level methylmercury exposure through rice ingestion in a cohort of pregnant mothers in rural China

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ABSTRACT

Background: Rice ingestion is an important dietary exposure pathway for methylmercury. There are few studies concerning prenatal methylmercury exposure through rice ingestion, yet the health risks are greatest to the developing fetus, and thus should be investigated.
Objectives: Our main objective was to quantify dietary methylmercury intake through rice and fish/shellfish ingestion among pregnant mothers living in southern China, where rice was a staple food and mercury contamination was considered minimal.
Methods: A total of 398 mothers were recruited at parturition, who donated scalp hair and blood samples. Total mercury and/or methylmercury concentrations were measured in biomarkers, in rice samples from each participant's home, and in fish tissue purchased from local markets. Additional fish/shellfish mercury concentrations were obtained from a literature search. Dietary methylmercury intake during the third trimester was equivalent to the ingestion rate for rice (or fish/shellfish) × the respective methylmercury concentration.
Results: Dietary methylmercury intake from both rice and fish/shellfish ingestion averaged 1.2 ± 1.8 μg/day (median = 0.79 μg/day, range = 0–22 μg/day), including on average 71% from rice ingestion (median: 87%, range: 0–100%), and 29% from fish/shellfish consumption (median 13%, range: 0–100%). Median concentrations of hair total mercury, hair methylmercury, and blood total mercury were 0.40 μg/g (range: 0.08–1.7 μg/g), 0.28 μg/g (range: 0.01–1.4 μg/g), and 1.2 μg/L (range: 0.29–8.6 μg/L), respectively, and all three biomarkers were positively correlated with dietary methylmercury intake through rice ingestion (Spearman's rho = 0.18–0.21, $p \leq 0.0005$), although the correlations were weak. In contrast, biomarkers were not correlated with fish/shellfish methylmercury intake (Spearman's rho = 0.04–0.08, p = 0.11–0.46).
Conclusions: Among pregnant mothers living in rural inland China, rice ingestion contributed to prenatal methylmercury exposure, more so than fish/shellfish ingestion.

1. Introduction

Fish consumption is the main dietary source for methylmercury (MeHg), a potent neurotoxin (National Research Council (NRC), 2000); however MeHg exposure also occurs through rice ingestion (Rothenberg et al., 2014). Most rice is cultivated under 5–10 cm of standing water (unlike other terrestrial crops), providing optimal conditions for microbial mercury (Hg) methylation (Qiu et al., 2008; Rothenberg et al., 2014; Windham-Myers et al., 2014). In submerged rice paddy fields, anaerobic microorganisms convert less toxic inorganic Hg to MeHg (Rothenberg and Feng, 2012), which is efficiently translocated from paddy soil to the rice endosperm (i.e., polished rice grain) (Rothenberg et al., 2011a, 2014).

The prenatal period is the most vulnerable exposure window for MeHg (Clarkson and Magos, 2006), yet there are few studies reporting dietary MeHg intake through rice consumption among pregnant mothers. We searched Thomas Reuters (ISI) Web of Science using the phrase "rice and mercury" combined with "hair", "blood", or "urine", which yielded a total of 101 studies. Hair Hg is a biomarker for dietary MeHg intake, blood Hg reflects both MeHg and inorganic Hg exposure, and urine Hg mostly represents elemental and inorganic Hg (reviewed by Clarkson and Magos (2006)). Overlapping studies were excluded, as well as those not reporting data for human health or for rice ingestion and/or rice Hg, resulting in 17 studies (Table A1).

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In summary, rice consumption was positively correlated with blood Hg in a U.S. population (n=16,236) (Davis et al., 2014). In a Hg-contaminated area of China, hair and blood Hg were positively correlated with rice MeHg (n=17) (Rothenberg et al., 2013), and one or both biomarkers were correlated with MeHg intake through rice ingestion (μg/kg bw/day) (n=98, Feng et al., 2008; n=168, Li et al., 2015). Only two studies reported data for pregnant mothers (Maramba et al., 2006; Rothenberg et al., 2013); both had small sample sizes (n=35 and n=17, respectively), and pregnant mothers were recruited from Hg-contaminated sites.

In 2013, we initiated a birth and child cohort study in rural China, where rice is a staple food. In this report, our main objective was to quantify dietary MeHg intake through rice and fish/shellfish ingestion, and correlate with biomarkers for prenatal MeHg exposure. We also queried mothers concerning other potential sources of inorganic Hg, including proximity to a mine or working in a mine, whether mothers had dental fillings, and the use of skin whiteners during pregnancy; the latter often contain Hg and are routinely used in Asia (Murphy et al., 2009). We did not specify the kind of mine because Hg often co-occurs with other minerals, which are simultaneously emitted into the environment (Feng et al., 2006). We chose a background site for this investigation rather than a Hg-contaminated site because findings would be relevant to a wider range of rice-eating communities, and to minimize confounding from other Hg sources (e.g., atmospheric Hg). In a companion report, we assessed the impacts of prenatal MeHg exposure on offspring neurodevelopment at 12 months, including maternal hair total Hg (THg), hair MeHg, and blood THg also reported in the present study (Rothenberg et al., submitted for publication).

Rice is a staple food for half the global population (Food and Agriculture Association (FAO), 2015). Compared to fish, rice does not contain the same beneficial micronutrients (Rothenberg et al., 2011b). Thus maternal rice ingestion may represent a potentially important exposure pathway for MeHg.

2. Methods

2.1. Recruitment and data collection

The study site was located inland, in Daxin county, Guangxi Zhuang Autonomous Region, near the China-Vietnam border. Daxin county is 2742 km^2 with a population of 359,800 (Guangxi Daxin County Government, 2011), including ~50,000 residents (14%) living in the town of Daxin. There were no significant sources of Hg nearby (e.g., coal-fired power plants). Rice THg from the surrounding villages was low, averaging 1.7 ng/g (range: 0.30–3.7 ng/g, n=15, unpublished data), which was 4.8 times lower compared to other non-contaminated sites (average rice THg: 8.2 ng/g, reviewed by Rothenberg et al. (2014)), suggesting the area was relatively non-contaminated for Hg.

Between May 2013 and March 2014, adult women were recruited at parturition at the Maternal and Child Health Hospital in Daxin county. Eligible mothers were in good general health, resided in Daxin county during the three previous months, and planned to remain for the next year. Protocols were reviewed and approved by the Institutional Review Boards at the University of South Carolina (USA) and XinHua Hospital (China). Mothers provided written informed consent prior to enrollment in the study.

Upon enrollment, a hair sample was collected form the occipital region using stainless steel scissors, the proximal end was tied with dental floss, and the hair sample was stored in a plastic bag at room temperature. A blood sample was collected by venipuncture (6 mL) into two vials, including one with lithium heparin anticoagulant, and a second vial for separation of serum by centrifugation (Yu et al., 2011). Whole blood and aliquots of serum were stored frozen at −26 °C for up to 10 months, then stored at −80 °C until analysis. A family member brought a ~100 g polished rice sample from home (all participants donated a rice sample), which was stored at −26 °C, and then archived at −80 °C until analysis.

While in the hospital mothers filled out a questionnaire, including maternal health, maternal/paternal education, occupation, and household income, and neonatal statistics (e.g., gender, length, and weight). Mothers also filled out a modified 102-item semi-quantitative food frequency questionnaire (FFQ) (Cheng et al., 2009) about their food intake during the third trimester, including rice, fish/shellfish, pork, other meat, eggs, fruits and vegetables. The FFQ asked mothers to chose from eight options ranging from "never or rarely" to "≥ twice/day", and frequencies were converted to servings per day as follows: 0=never or rarely, 1/30.5=monthly, 2.5/30.5=two to three times/month, 1/7=once per week, 2.5/7=two to three times/week, 5/7=four to six times/week, 1=once per day, and 2.5=at least twice per day. For rice, mothers were asked to select portion size (grams per serving) from one of three bowls using a picture and/or actual bowls. The daily rice ingestion rate (grams per day) was calculated by multiplying frequency (servings per day) × the portion size (grams per serving). Mothers also reported the consumption frequency for seven categories of fish/shellfish (including freshwater fish, ocean fish, shrimp, eel, shrimp, crab, and other shellfish) using the same eight options (from "never or rarely" to "≥ twice/day") and frequencies were converted to servings per day as described above. To calculate fish/shellfish ingestion (grams per day), we assumed 170 g/serving for ocean fish and freshwater fish [170 g=6 oz, the recommended serving size from the U.S. Food and Drug Administration (USFDA) (2001)], and 100 g/serving for other categories (Cheng et al., 2009).

Dietary MeHg intake from rice and fish/shellfish consumption (micrograms per day) was quantified by multiplying the ingestion rate (grams per day) × the concentration of rice MeHg (micrograms per gram) or fish/shellfish THg (micrograms per gram). Rice MeHg was measured in each participant's rice sample brought from home. We measured rice MeHg (not THg) because average rice %MeHg (of THg) ranged from 17 to 75% (reviewed by Rothenberg et al., 2014), while fish THg was >90% MeHg (NRC, 2000). THg concentrations for freshwater fish were analyzed in seven commonly consumed varieties purchased in Daxin markets in 2014 (n=13); however other fish/shellfish varieties were not available for purchase. Therefore a comprehensive literature search was conducted using Thomas Reuters (ISI) Web of Science and the phrase "mercury and China", which was combined with "seafood", "fish", "eel", "shrimp", "crab", "mollusk", "shellfish", "snail", "scallop", "oyster", "lobster", "spiral shell", or "bivalve", resulting in 209 studies. Data were included if most or all fish/shellfish samples were collected after 1 January 2011, studies were conducted in non-contaminated sites in China, and THg (or MeHg) concentrations were reported in wet weight (n=11 studies). For "eel", just one study for China was published in 2006 and was included, resulting in a total of 12 studies (Table 1 and Table A2). For fish/shellfish varieties from the literature search, summary statistics were calculated if possible (mean, median and standard deviation); for studies reporting the sample size, summary statistics were calculated using the sample size as the analytical weight. Daily MeHg intake through fish/shellfish (micrograms per day) was quantified by multiplying the ingestion rate (grams per day) × the average THg concentration (micrograms per gram). The distribution for fish/shellfish THg was highly skewed; therefore the daily MeHg intake was also calculated using the median fish/shellfish THg concentration.

Total dietary MeHg intake (micrograms per day) was equivalent