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Rice Methylmercury Exposure and Mitigation: A Comprehensive Review

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Abstract

Rice cultivation practices from field preparation to post-harvest transform rice paddies into hot spots for microbial mercury methylation, converting less-toxic inorganic mercury to more-toxic methylmercury, which is likely translocated to rice grain. This review includes 51 studies reporting rice total mercury and/or methylmercury concentrations, based on rice cultivated or purchased in 15 countries. Not surprisingly, both rice total mercury and methylmercury levels were significantly higher in polluted sites compared to non-polluted sites (Wilcoxon rank sum, $p < 0.001$). However, rice percent methylmercury (of total mercury) did not differ statistically between polluted and non-polluted sites (Wilcoxon rank sum, $p = 0.35$), suggesting comparable mercury methylation rates in paddy soil across these sites and/or similar accumulation of mercury species for these rice cultivars. Studies characterizing the effect of rice cultivation under more aerobic conditions were reviewed to determine the mitigation potential of this practice. Rice management practices utilizing alternating wetting and drying (instead of continuous flooding) caused soil methylmercury levels to spike, resulting in a strong methylmercury pulse after fields were dried and reflooded; however, it is uncertain whether this led to increased translocation of methylmercury from paddy soil to rice grain. Due to the potential health risks, it is advisable to investigate this issue further, and to develop separate water management strategies for mercury polluted and non-polluted sites, which minimize methylmercury exposure through rice ingestion.

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Keywords

rice; *Oryza sativa*; methylmercury; alternating wetting and drying; carbon exudates

1. Introduction

Mercury (Hg) is a global pollutant and potent neurotoxin. Methylmercury (MeHg) is one of the most toxic forms of Hg, which can severely afflict the unborn fetus (Clarkson and Magos, 2006). Fish consumption is considered the primary human MeHg exposure pathway due to efficient biomagnification of MeHg in aquatic food chains, especially among piscivorous fish (Cabana et al. 1994; Morel et al. 1998; Mahaffey et al. 2004). This assumption is currently challenged by recent research in Guizhou province, China, where elevated rice grain MeHg levels were reported in some villages near the former Wanshan Hg Mine (e.g., see Table 1 and references therein). In this region, median rice MeHg concentrations were up to 10 times lower than those typically measured for fish tissue (e.g., Feng et al., 2008; Horvat et al. 2003; Rothenberg et al. 2012; Zhang et al., 2010a). However, rice is a staple food, resulting in daily rice-based meals (without fish) containing MeHg exposure levels comparable to a fish meal (Zhang et al., 2010b), without the same beneficial micronutrients associated with fish ingestion (e.g., docosahexaenoic acid, DHA), potentially increasing neurodevelopmental risk to the unborn fetus (Rothenberg et al. 2011a; 2013).

In 2012, the total amount of land in rice cultivation globally was 163 million hectares (1.63 million km²) and the global production of rice was 729 million tons, of which 90% was produced in Asia (FAO, 2013). Despite the importance of rice as a staple food for half the global population, MeHg exposure through rice ingestion has received relatively little comprehensive study to date, particularly in geographic regions outside of Guizhou province, China, making it difficult to assess the global extent of MeHg exposure through rice ingestion, and to provide recommendations to communities depending on rice as a staple food.

This review is motivated by 1) the need to characterize the extent of MeHg exposure through rice ingestion and to highlight regions where more research is needed, and 2) to determine whether rice cultivation methods exist, which may reduce MeHg accumulation in rice grain. Both areas represent critical knowledge gaps. To address the first aim, a global inventory of studies reporting total Hg (THg) and/or MeHg concentrations for rice was developed, and the spatial distributions for rice THg and MeHg concentrations were mapped. Most studies reporting rice THg and/or MeHg concentrations are monitoring studies, and do not include paddy soil or pore water biogeochemical data. For the second aim, studies addressing Hg cycling in rice paddies were synthesized, including specific biogeochemical factors distinguishing MeHg cycling in rice paddies from other wetlands, as well as factors controlling the translocation of MeHg from the paddy soil to the rice grain. Additionally, the effect of reduced water usage for rice cultivation was considered as a mitigation tool for reducing rice MeHg levels. The topic of transgenic rice, engineered to hyperaccumulate Hg from contaminated soil for bioremediation, was not reviewed (e.g., Heaton et al., 2003). The scope of this review complements other recent reports on Hg

contamination and exposure within China (Feng and Qiu 2008; Lin et al. 2012; Qiu et al. 2012a; Zhang and Wong, 2007).

2. Methods

The first part of this review characterizes rice THg and MeHg levels. A global inventory of rice total THg and MeHg levels was compiled, which included peer-reviewed studies accessed through Thomson Reuters (ISI) Web of Knowledge and the National Library of Medicine PubMed using the search terms “rice and mercury” (last accessed November 13, 2013) (see Figure 1). The terms “rice and methylmercury” resulted in a subset of citations, and therefore not used. Studies were included if manuscripts were published in English, and rice grain analyses were completed after 1980 (when more reliable quantification methods for THg and MeHg were developed). Data were considered relevant if the authors reported rice THg and/or MeHg concentrations and results were for the rice species, *Oryza sativa*. For the inventory, experimental studies utilizing treated/untreated paddy soil were not included; however, relevant findings were discussed within this review. Likewise, rice Hg studies completed before 1980 are summarized within this review for historical perspective. Results of the global inventory are listed in Table 1, and categorized in Table 2 according to whether sampling sites were considered *a priori* Hg-polluted or non-polluted.

Rice THg and/or MeHg levels were mapped along with rice ingestion rates (Figures 2a and 2b). Among the 51 studies reporting rice THg and/or MeHg levels, there were 85 sites for rice THg and 65 sites for rice MeHg (see Table 1); however many sites were sampled more than once. To construct the maps, rice THg (or rice MeHg) concentrations were averaged for the same region or province, reducing the number of separate locations to 42 and 36 for rice THg and MeHg, respectively. There were two exceptions. First, in some cases researchers paired highly contaminated sites with background sites from the same region or province to emphasize localized pollution sources (e.g., in Indonesia, Krisnayanti et al., 2012). Second, researchers from the same region focused on different questions (e.g., Hg pollution from gold mining in Thailand, from Pataranawat et al., 2007; and market-basket survey in Thailand, from Zarcinas et al., 2004), producing variable results. For these exceptions, both maps included one value for the Hg-contaminated area and one for the background area. For Guizhou province, China, two values were plotted, including one for the town of Wanshan, and one for other sites in Guizhou province.

Statistical analyses of compiled rice THg and MeHg data included comparisons between polluted and non-polluted sites using Wilcoxon rank-sum nonparametric test, and regression analysis between rice THg and MeHg concentrations. The latter was determined with weighted least squares regression, using the inverse of the study sample size for the analytical weight. Assumptions for regression residuals (ϵ) were checked; i.e., mean=0 and constant variance. Distributions for rice THg and MeHg were highly-skewed (mean $\gg$ median); therefore a log₁₀-transformation was applied, which improved normality of residuals. Descriptive statistics, including Cook's Distance, were used to assess the fit of the regression model. An alpha level of less than 0.05 was chosen as a guide for significance where appropriate. Data analyses were completed using Stata (Version 9.2, College Station, TX, USA).

3. Comprehensive inventory of rice grain THg and MeHg concentrations

3.1. Rice Hg, 1960s to 1980s

Rice Hg concentrations were a concern beginning in the 1960s due to fungicidal use of phenylmercuric acetate on agricultural fields in Louisiana, USA (Epps, 1966). Despite high detection limits available at that time (i.e., 100 ng/g), the maximum THg concentration for polished rice from treated fields measured 200 ng/g, compared to non-detectable levels in control fields (Epps, 1966). In 1968, a market-basket survey in the UK reported THg content in rice imported from eleven countries, with concentrations ranging from less than 5 ng/g (detection limit) to 95 ng/g (Smart and Hill, 1968). Although organo-Hg fungicides, including phenylmercuric acetate, were banned in Japan after the discovery of Minamata disease in the 1950s–1960s, residual Hg levels in paddy soil were a potential concern. However, THg concentrations in unhulled rice (i.e., inedible rice) from paddy fields in Kyushu, Japan averaged 11.3 ng/g (range: 8.5–29.4 ng/g), and the authors concluded rice played a “minor role in the food chain” with respect to MeHg exposure (Gotoh and Koga, 1977).

3.2. Rice Hg, 1980s to present

Beginning in the 1980s, 51 studies from 15 countries documented rice THg and/or MeHg concentrations, including nationwide sampling in Taiwan (n=407 samples, Lin et al., 2004), Thailand (n=108 samples, Zarcinas et al., 2004), and 20 provinces in China (n=712, Qian et al., 2010). Table 1 includes summary data, as well as background information on where the rice was cultivated or purchased, whether sampling sites were considered polluted, whether rice data were part of a market basket survey, and whether rice samples were polished or unpolished (i.e., white and brown rice, respectively). The latter category was considered important because of potential differences in accumulation of inorganic Hg(II) and MeHg between brown rice and polished white rice (discussed in Section 5) (Rothenberg et al., 2011b).

The spatial distributions of rice THg and MeHg concentrations were overlaid onto a global map with rice ingestion rates for all countries, with all data subdivided into quintiles (FAO, 2013) (Figures 2a and 2b). Data were included for all sites, except where rice samples were imported or when provincial-level data were not provided (see Figure 2 caption for details). For both rice THg and MeHg concentrations, the highest quintiles reflect a wide range of values due to the highly skewed distributions for samples from polluted sites (Table 3). For example, the highest quintile for rice THg ranges from 27.7 ng/g and 510 ng/g, which differs by a factor of 18. The range for rice MeHg is narrower; values for the highest quintile differ by a factor of 5.5.

One of the motivations for reviewing rice THg and MeHg studies was to identify sites where studies were needed. Figures 2a and 2b provide evidence that studies are lacking in countries where the rice ingestion rates are highest, including Myanmar (386 g/capita/day), Vietnam (387 g/capita/day), Laos (454 g/capita/day), and Bangladesh (475 g/capita/day), while more studies are needed in India (187 g/capita/day) (ingestion rates from FAO, 2013).

3.3. Comparison of polluted and non-polluted sites

For this review, studies are categorized as polluted and non-polluted sites to compare parameters (Table 2, Figure 3), and summary statistics are in Table 3. From Table 1, Hg pollution sources included Hg mining (n=23 sites), historical and present-day artisanal and small-scale gold mining (n=9 sites), contamination of irrigation water from nearby chlor-alkali plants (n=2 sites), e-waste (n=2), and miscellaneous sources (n=9 sites) (e.g., zinc and lead mining, proximity to coal-fired power utilities, and other industrial pollution). Highest rice THg levels were cultivated in polluted sites in Ganjam, India (530 ng/g, from Lenka et al., 1992) and Guizhou province, China (569 ng/g, from Horvat et al., 2003), while highest rice MeHg levels were observed in polluted sites in Guizhou province, China (144 ng/g from Horvat et al., 2003; 174 ng/g, from Qiu et al., 2008) and Indonesia (115 ng/g, from Krisnayanti et al., 2012). Not surprisingly, both rice THg and MeHg levels were significantly higher in polluted sites compared to non-polluted sites (Wilcoxon rank-sum test, $p < 0.001$). However, rice percent methylmercury (of total mercury) did not differ statistically between polluted and non-polluted sites (Wilcoxon rank sum, $p = 0.35$), suggesting comparable mercury methylation rates in paddy soil across these sites and/or similar accumulation of mercury species for these rice cultivars.

Rice THg and MeHg concentrations associated with each pollution source are shown in Figure 3. Rice cultivated in paddies near Hg mining activities contained the highest MeHg levels. Artisanal Hg mining uses crude, inefficient smelters that release copious amounts of Hg to the atmosphere, which is then deposited to nearby rice paddies through wet or dry deposition (Meng et al., 2011; Rothenberg et al., 2012). Highest rice MeHg levels near Hg mining sites possibly reflected higher bioavailability of atmospheric Hg for microbial Hg methylation (Hintelmann et al., 2002; Meng et al., 2011), or other environmental factors not measured, or greater replication of results due to the number of studies investigating rice paddies in sites near Hg mining sources in China. From Table 2, more than half the studies (59%) were completed in China, including 13 (25%) in Guizhou province, within the vicinity of former Wanshan Hg mine. There are no other sites globally, which report rice THg and/or MeHg levels, with such a high degree of replication.

When investigating polluted sites, researchers often analyzed rice samples from nearby towns, where lower rice THg and/or MeHg levels contrasted rice data from polluted areas (e.g., Cheng et al., 2013, Krisnayanti et al., 2012, Qiu et al., 2013, from Table 1). Results from side-by-side analyses of neighboring polluted and non-polluted sites suggest the extent (or severity) of rice as a MeHg exposure pathway is more likely a function of proximity to active Hg pollution sources (e.g., Qiu et al., 2013; Zhang et al., 2010a), rather than attributable to diffuse regional Hg sources. This suggests Hg pollution within rice paddies is a localized issue, more dependent on point sources of pollution. Therefore, future mitigation strategies should target cleanup and remediation of specific Hg-polluted sites, which will likely be more effective than blanket regional recommendations.

3.4. Association between rice THg and MeHg concentrations

Paired rice THg and MeHg concentrations were reported in 19 studies, including 17 studies in China, one in Cambodia, and one in the USA (see Table 1 for references). For studies

reporting both rice THg and MeHg levels, a strong association was observed (when variables were log₁₀-transformed) ($r^2=0.74$, $p<0.0001$, $n=43$ sites) (Figure 4). An indicator variable (coded 0 or 1) for polluted sites was positively correlated with rice MeHg levels, but not significantly ($p=0.083$). The strength of the relationship between rice THg and MeHg levels may reflect the number of replicated studies from the same area (i.e., Guizhou province, China). As already noted, to more accurately characterize the association between rice THg and MeHg levels, more monitoring is needed in other regions where rice is a staple food (Figures 2a and 2b), especially where point sources for Hg pollution were previously identified (e.g., Vietnam: Noh et al., 2013).

3.5. International MeHg Reference Doses (RfDs) for MeHg

A reference dose (RfD) for MeHg is defined as the daily tolerable MeHg exposure level for the human population (including sensitive subpopulations), which is likely to result in no risk of adverse effects when experienced over a lifetime (USEPA, 1993). Established RfDs for MeHg were based on epidemiologic studies where fish consumption was the primary MeHg exposure pathway, including studies in New Zealand (Kjellstrom, 1986, 1989), the Faroe Islands (Debes et al., 2006; Grandjean et al., 1997, 2001) and the Republic of Seychelles (Davidson et al., 1998, 2008; Myers et al., 1995, 2003).

Researchers often assume MeHg bioavailability from rice ingestion is the same as for fish consumption, and adverse neurodevelopmental outcomes are equivalent (e.g., Qiu et al., 2008; 2012b, 2012c, 2013; Rothenberg et al., 2012, 2013; Zhang et al., 2010b). However, there are two caveats, which may render MeHg RfDs for fish consumers not applicable for rice consumers. First, fish tissue contains MeHg, which harms offspring neurodevelopment, but fish tissue is also a rich source of micronutrients including DHA, which benefits fetal neurodevelopment (Choi et al., 2008). Fish consumers, especially pregnant women, are advised to consume fish tissue low in MeHg but high in DHA, which may minimize adverse health effects associated with prenatal MeHg exposure (Davidson et al., 2008; Mahaffey et al., 2011; Oken et al., 2005, 2012). Rice does not contain the same micronutrients as fish tissue (e.g., DHA), and therefore a dose of MeHg from maternal rice ingestion may be more harmful to the unborn fetus (Rothenberg et al., 2011a). Second, differences in fiber content between rice and fish (cooked white rice: 0.6 g fiber/cup, fish: 0 g fiber/cup, from USDA, 2012) may alter enterohepatic cycling of MeHg in the gut, resulting in differential rates of MeHg absorption. Diets are important modulators of MeHg absorption (Chapman and Chan, 2000). Mice fed a high fiber diet (as 30% wheat bran) accumulated less THg in the small intestine, brain and blood compared to mice fed fiber-free and low-fiber diets, and the authors attributed this to differences in the metabolic activity of gut microbiota (Rowland et al., 1986). Among Brazilian fishers, the strength of the relationship between average hair Hg levels and fish consumption was weaker for those consuming at least one piece of fruit per day, and the authors suggested this may be due to the soluble dietary fiber content or other prebiotic nutrients of fruits, which modified the absorption of MeHg from the gut (Passos et al., 2003, 2007).

Human health studies are lacking among populations, where rice ingestion is the primary MeHg exposure pathway. In China, MeHg exposure through rice ingestion was investigated

among children and adults living near Hg mines (e.g., Li et al., 2008), while another study reported a positive relationship between hair and rice MeHg concentrations among village residents (Feng et al., 2008). Aside from one feasibility pilot among 17 mothers in Wanshan, China (Rothenberg et al., 2013), to the best of our knowledge there are no reports concerning the impacts of prenatal MeHg exposure through maternal rice ingestion. New MeHg RfDs for rice consumers may be warranted due to differences in beneficial micronutrients and fiber content between rice and fish; however, more research relating prenatal MeHg exposure to offspring development is needed to guide this process.

4. The influence of rice cultivation practices on Hg cycling

Rice cultivation practices, from planting to post-harvest, help transform rice paddies into hot spots for Hg(II)-methylation. Microbial Hg(II)-methylation rates are a function of factors influencing 1) anaerobic microbial activity (e.g., temperature, anoxia and labile organic carbon (C), reviewed by Ullrich et al., 2001), as well as 2) factors affecting the bioavailability of inorganic Hg(II) (e.g., organic C, and sulfur and iron speciation and biogeochemistry, Benoit et al., 1999, 2003; Marvin-DiPasquale et al. 2009, in press). Table 4 includes rice management practices, which influence these factors and thus rates of Hg(II)-methylation, while Figure 5 shows linkage between all processes. Each phase of rice management is discussed in the sections below.

4.1. Flooded rice paddies as Hg(II)-methylation sites

Unlike tidal or freshwater wetlands, which are flooded daily or intermittently, 95% of temperate and tropical rice acreage is cultivated under irrigated or rainfed flooded conditions, with limited advective flow of surface water (Kirk, 2004). Standing water depth may range from a few cm to a few meters, producing anoxic conditions throughout most of the rice growing season (Kirk, 2004).

In freshwater anoxic sediment, sulfate-reducing bacteria are considered the primary methylators of inorganic Hg(II), while iron-reducing bacteria also methylate Hg (Fleming et al. 2006; Gilmour et al., 1992; Kerin et al. 2006; Parks et al., 2013; Schaefer et al., 2011; Warner et al. 2003; Yu et al., 2011), and both compete for electron donors in rice paddy soil (Acht nich et al., 1995a, 1995b, Marvin-DiPasquale et al., in press). Like other wetlands (Bouchet et al. 2011; Creswell et al., 2008; Drott et al., 2007; Gilmour et al., 1992, 1998; Langer et al. 2001; Marvin-DiPasquale et al., 2003; Mitchell et al., 2008; Rothenberg et al., 2008; St Louis et al., 1994), rice paddies are active sites for Hg(II)-methylation, converting less-toxic inorganic Hg(II) to more toxic MeHg, which is likely translocated from paddy soil to rice grain (Table 1).

In a comparison between rice and other agricultural crops cultivated in upland soil (e.g., corn and tobacco), average rice THg levels were similar to the other crops, while average rice MeHg levels were more than 40 times higher, which was attributed to cultivation of rice in standing water (Qiu et al., 2008).

4.2. Oxidation of the rice rhizosphere

Rice is not a native wetland plant but is adapted morphologically to withstand cultivation under flooded conditions (Kirk, 2004). To survive waterlogged soil, rice roots contain sponge-like aerenchyma tissue within xylem transport cells, which enhances diffusive gas transport between relatively oxygen-rich aboveground tissues and carbon dioxide (CO₂)-rich belowground tissues (Armstrong, 1967; Armstrong et al., 1991; Colmer, 2003; Justin and Armstrong, 1991; Jackson and Armstrong, 1999). Oxygen transport and radial oxygen loss through aerenchyma tissue in rice roots, as well as transpiration-induced oxidation at root surfaces, are capable of altering the chemistry of the surrounding rhizosphere, operationally defined as the narrow (mm-scale) band of soil influenced by proximity to the root surface (Farrar et al. 2003; Liesack et al., 2000). The oxidized rhizosphere supports a rich diversity of microbial functional groups (Liesack et al., 2000), and higher rates of both sulfate reduction and sulfide reoxidation were observed in the rice-planted rhizosphere compared to unplanted paddy soil (Stubner et al., 1998; Wind and Conrad, 1997; Windham-Myers et al., 2009, in press, b). Oxygen leakage from root tips produces acidity, primarily due to oxidation of iron(II), as well as uptake of nitrogen (NH₄⁺), and CO₂ exchange between roots and soil (Begg et al., 1994). Higher sulfate reduction rates are associated with higher Hg(II)-methylation rates (King et al., 2000), while lower pH may enhance rhizosphere Hg(II) bioavailability and bacterial uptake, increasing microbial Hg(II)-methylation (Kelly et al., 2003; Miskimmin et al., 1992; Winfrey and Rudd, 1990). Differences between the rice-planted and fallow regions of the same flooded rice paddy included increased acidity in the rice-planted region, which altered iron and sulfur cycling and resulted in consistently higher pore water MeHg levels between 10–18 cm depth compared to the fallow area (Rothenberg and Feng, 2012). Results supported the hypothesis that oxygenation of the rhizosphere due to rice roots contributed to microbial Hg(II)-methylation.

Of the 51 citations in Table 1, only three studies reported biogeochemical data, including soil or surface water pH, and organic content (Li, 2013; Qiu et al., 2013; Rothenberg et al., 2012; Windham-Myers et al., in press, a). Li (2013) did not correlate soil parameters with rice THg levels, while Qiu et al. (2013) reported rice MeHg levels were most correlated with soil MeHg levels. In one study comparing 50 rice genotypes along a contamination gradient, environmental factors (e.g., elevated pH) played an important role in limiting the bioavailability of inorganic Hg(II) and/or the micronutrients needed for plant growth, resulting in lower rice THg and MeHg levels despite elevated soil THg and MeHg levels (Rothenberg et al., 2012).

4.3. The influence of carbon (C) dynamics on Hg(II)-methylation

Microbial activity (and hence Hg(II)-methylation) is driven by both bioavailability of redox-sensitive trace elements (see previous section) and root-derived C inputs. Rice has been genetically driven to deliver some of the greatest net primary productivity rates observed for a grass-dominated ecosystem (Cramer and Field, 1999; Lobell et al., 2002). Estimated daily net primary productivity rates of up to 4 g C m⁻² d⁻¹ were calculated from satellite data in tropical rice fields (Panigrahy et al, 2004), while aboveground biomass harvests in California rice fields averaged up to 10 g C m⁻² d⁻¹ (Windham-Myers et al., in press, a). Belowground, roughly an equivalent amount of structural biomass (e.g. 400–800 g C m⁻²

during the 3-month rice growing season) is accumulated in root tissues, plus an additional supply of C is lost through root exudation (Farrar et al. 2003), which are both unaccounted for in net primary productivity estimates. All of this non-harvested productivity, both structural and exuded C, is subject to decay and secondary consumption. Thus, the large C pool fixed in rice fields is a source of energy for microbial activity, both through root exudates and through litter decomposition post-harvest (discussed further in Section 4.5). Importantly, a wide variety of organic C compounds exuded by plant roots serve as electron donors for sulfate-reducing bacteria, including hydrogen, aliphatic hydrocarbons, and simple aromatic compounds (Widdel and Bak, 1992; Devereux et al. 1996; King et al. 2000, 2002; Achá et al. 2005).

Availability of C was an important driver of MeHg production and bioaccumulation in rice plants in a year long study of agricultural and non-agricultural wetlands (Marvin-DiPasquale et al., in press). In flooded rice fields, experimental devegetation reduced pore water concentrations of acetate (an index of labile C pools) by 63% and simultaneously reduced Hg(II)-methylation rates by 64% (Windham-Myers et al., 2009). Further, root density was high in rice fields (up to 10% of soil volume) and positively correlated with both pore water acetate concentrations and microbial Hg(II)-methylation rates during the growing season (Windham-Myers et al., in press, b).

Rice plant tissues function as more than a microbial food source for Hg(II)-methylation. High concentrations of pore water dissolved organic C are generated from tissue leaching and decay, which promotes partitioning of MeHg into the dissolved phase, enhancing its bioavailability for uptake in biota and rice plants (Fleck et al, in press). Rice plant density also influences net MeHg yields in surface water and pore water. For example, photodegradation of MeHg is important in lakes, abiotically reducing MeHg concentrations (Hammerschmidt and Fitzgerald, 2006; Sellers et al., 1996). However, dense leaf canopies in rice fields (Leaf Area Index: up to 7 (unitless); from Hatala et al., 2012) limit the amount of incident radiation on the water surface, thus lowering photodemethylation of MeHg in the shallow surface waters of rice fields (Fleck et al., in press; Windham-Myers et al., 2010). In rice fields, photodemethylation reduced surface water MeHg loads by only ~6% over the growing season, while surface water MeHg concentrations increased 3–5 fold from inlet to outlet due to evapotranspiration (Alpers et al., in press). The dense leaf canopies also support significant rates of transpiration (Bouman et al., 1994), which is the dominant form of atmospheric flux, and results in increased pore water concentrations and root zone storage of conservative constituents, such as chloride, and nonconservative constituents, including MeHg (Bachand et al., in press, a; Windham-Myers et al., in press, c).

When rice paddy surface water MeHg concentrations are elevated, *in situ* biotic responses may be significant, which has direct implications for human and wildlife health. For example, both invertebrates and fish cultivated within flooded rice paddies accumulated MeHg to toxic threshold levels in less than 30 days (>0.2 ppm wet weight, from Ackerman et al., 2010; Ackerman and Eagles-Smith, 2010). Elevated MeHg concentrations are of concern to resident and migrating waterfowl, which visit rice fields due to their high productivity and extent (Elphick et al., 2000). More importantly, in tropical rice paddies, which combine rice and fish cultivation (e.g., Lansing and Kremer, 2011), potential co-

exposure of MeHg through rice and fish consumption may be another concern, and requires further research.

4.4. Alternating wetting and drying (AWD) and Hg cycling in rice paddies

Throughout Asia, fresh water resources are stressed due to increased water demand, shrinkage of lakes, decline in the ground water table, and contamination of existing water resources (Li and Barker, 2004; Li, 2006), resulting in increased pressure on water managers to transfer high quality fresh water from agricultural use to industrial, residential and municipal uses, while increasing rice production (Bindraban et al., 2006; Bouman and Tuong, 2001; Li, 2006). Water use efficiency in continuously flooded rice agriculture is generally low; approximately 80% of the water applied to rice paddies is lost through evaporation, surface runoff, and percolation (Bindraban et al. 2006; Bachand et al., in press, b). Global climate change further heightens the need to reduce fresh water use for rice cultivation (Bates et al., 2008).

To address impending reductions in water resources, in the 1970s, water managers initiated research into more efficient irrigation regimes for rice cultivation, including the development of aerobic rice varieties (Bouman et al., 2005; Wang et al., 2002; Yang et al., 2005), the use of straw mulch and/or plastic covers to minimize evaporation (Fan et al., 2002; Tao et al., 2006–2007), and alternating wetting and drying (AWD) cycles in rice fields (Bouman et al., 2001; Li and Barker, 2004). AWD replaces continuous storage of water on the rice field with carefully timed periods of nonsubmergence, including mid-season drainage after seedlings are transplanted, then intermittent wetting and drying until the milk ripening stage, allowing the paddy water to drain below the soil subsurface between wetting periods (Belder et al., 2004; Dong et al., 2004). In the 1990s, a major impediment to farmer adoption of AWD was overcome when researchers and farmers implemented plastic tubes with holes, which measured field water depth, and signaled to farmers when it was time to re-irrigate (Bouman et al., 2007). The approach was termed “safe AWD” because rice plants were not stressed and yield was maintained, while freshwater use was reduced by ~15% (Bouman et al., 2007). Presently, AWD is the most widely employed water saving technology applied in lowland areas with sufficient rainfall. In response to global warming, the Intergovernmental Panel on Climate Change recommended rice paddy drainage at least once during the rice growing season to reduce methane emissions (Smith et al., 2008; Yan et al., 2009). Therefore, the application of AWD for rice cultivation is expected to increase in the near future, in response to both freshwater shortages and global climate change.

The effect of AWD cultivation practices on rice grain MeHg levels remains a critical knowledge gap. Of the 51 studies in Table 1, rice was cultivated under continuously flooded conditions except for one study from Hubei province, China (Rothenberg et al., 2011b). However, this study did not compare rice grain THg and MeHg levels between AWD and flooded paddies. Instead, the potential effect of intermittent wetting and drying on soil MeHg levels was inadvertently captured when the fallow area of the paddy dried, while the rice-planted area remained saturated due to the dense leaf canopy (Rothenberg and Feng, 2012). The paddy was re-irrigated just before sediment and pore water samples were collected for analysis; in the fallow area, sediment MeHg and pore water sulfate levels were

4.6 and 2.5 standard deviations above the mean, respectively, while the same parameters were 0.72 and -0.73 standard deviations from the mean in the saturated rice-planted area (Rothenberg and Feng, 2012). Results suggested anaerobes were dormant when the paddy was completely dried, but recovered within hours after flooding due to a return to more anoxic conditions and a spike in sulfate levels, causing soil MeHg levels to also spike (Rothenberg and Feng, 2012).

Similar results were also observed in California rice fields, whereby one replicate field was drained mid-season, and its estimated MeHg production was enhanced 3-fold due to both increased availability of inorganic Hg(II) and increased microbial methylation (Marvin-DiPasquale, et al. in press). MeHg production and release was associated with wetting and drying events via hydrologic flows (Marvin-DiPasquale et al., in press). Specifically, during wet-dry-wet events, pulsed production of MeHg was observed during flooding, then soil storage of MeHg during drydown, then later release of MeHg during flood-up, causing MeHg spikes during the summer (crop season) as well as the winter (post harvest) (Alpers et al., in press). No differences were found in rice tissue THg or MeHg concentrations between these fields (Windham-Myers et al., in press, a).

Results from rice paddies were consistent with studies in newly-flooded reservoirs, where spikes in surface water MeHg concentrations were observed immediately after flooding, and were attributed to decomposition of organic matter, and stimulation of anaerobic bacteria shortly after submergence, resulting in a three-fold increase in fish tissue MeHg levels (Kelly et al., 1997). However, it is unknown whether soil MeHg spikes due to frequent periods of wetting and drying under AWD result in higher rice grain MeHg levels compared to rice cultivated under continuous flooded conditions. There may be specific periods during rice plant development, when rice plants are less likely to translocate MeHg from paddy soil under AWD (Liu et al., 2012), which should be further investigated.

Findings from field studies cited above (Alpers et al., in press; Marvin-DiPasquale et al., in press; Rothenberg and Feng, 2012; Windham-Myers et al., in press, a) differed from a greenhouse study using pots with holes to replicate AWD, where the authors reported rice grain MeHg and THg concentrations decreased in AWD pots compared to continuously flooded pots (Peng et al, 2012). Differences were possibly an artifact of the methods; e.g., inorganic Hg(II) was leached from the pots, thus reducing available substrate for Hg(II)-methylation, lowering both rice THg and MeHg levels. To replicate AWD field conditions, surface water in pot experiments should naturally evaporate, which is more similar to field conditions, and prevents loss of inorganic Hg(II) substrate (Sarah Johnson-Beebout, IRRI, personal communication). In another report, rice MeHg levels were lower when cultivated under completely aerobic conditions compared to continuously flooded conditions in both greenhouse and field studies (Wang et al., in press). However, rice cultivation under aerobic conditions is not considered a viable alternative due to severe reductions in rice yields; instead, “safe AWD” is recommended, as noted above (Bouman et al., 2007).

Although MeHg data were limited, results from field studies for MeHg differed from those for arsenic, where lower accumulation of arsenic in rice grain was observed under non-flooded regimes compared to continuously flooded regimes in field studies (Norton et al.,

2012) and pot experiments (Xu et al., 2008). More research is needed to address the effects of AWD on multiple trace elements because less flooding may increase rice MeHg levels, while simultaneously decreasing the bioavailability of other trace elements. Thus, water management practices may need to differ, depending on the level and type of contamination.

4.5. Post-harvest litter management and Hg cycling

Post-harvest litter management (rice crop residue) may be a means of limiting available C pools and thus reduce microbial MeHg production. Whereas half of the aboveground C pool is the seed biomass yield and may be removed at harvest, the other half is represented in leaves and structural tissues that become litter or “residue” following harvest. The fate of this residue has implications for Hg cycling.

Despite laws prohibiting biomass burning in Asia, this is one of the most common methods of disposing crop residue, which is partially attributed to limited time between crops and lack of machinery to manage residues (Singh et al., 2008). Biomass burning results in poor air quality, higher greenhouse gas emissions, and increased hospital admissions due to respiratory complaints (reviewed by Singh et al., 2008). Along with wildfires (Friedli et al. 2001, 2003a, 2003b; Rothenberg et al., 2010), agricultural waste fires are also significant source of Hg emissions (Streets et al., 2009). In 2006, biomass burning, including rice residues, contributed 24% of the global atmospheric Hg emissions, which was greater than power plants (18%) but less than industrial emissions (41%) (Streets et al., 2009). Although most Hg is released as elemental Hg (Hg^0), higher soil MeHg levels and Hg(II)-methylation rates were reported within the vicinity following a wildfire, which corresponded to increased availability of inorganic Hg(II) and higher soil total organic C (Caldwell et al., 2000). Therefore, Hg(II)-methylation in paddy soil may be enhanced in regions where biomass burning is more widely employed to destroy rice residue, another reason why biomass burning should be discouraged.

During field preparation, both burned and unburned residues are incorporated into paddy soil as mulch, depending on the type of cropping system (e.g., rice-rice, rice-wheat), resulting in higher soil C content (Kalbitz et al., 2013). Over an annual cycle, highest concentrations of pore water acetate and associated MeHg production rates were observed during winter post-harvest, which likely reflected post-harvest litter decay (Marvin-DiPasquale et al., in press; Windham-Myers et al. in press, a). This practice of incorporating rice residues into the paddy soil may be important in rice-rice-rice systems, where there is insufficient time to allow the paddy to dry between crops, resulting in continuously flooded conditions throughout the year and more tissue decomposition (Singh et al., 2008).

Decreasing post-harvest aboveground biomass by baling and removing rice straw may provide a mechanism for reducing residue C, thus minimizing the potential to increase MeHg production. An alternative is tilling (disking) the rice straw into the soil, thus altering the density of C in the profile and either burying or redistributing the C pool and any associated microbial activity. Both methods are being tested in California rice fields currently (Windham-Myers et al., unpublished data). Results appear to differ between rice fields based on initial conditions (e.g. soil %Hg and %organic matter) and effectiveness of

the straw removal. However, disking of rice straw depends on the availability of machinery, and may not be a suitable option in small land-holding farms.

5. Translocation of MeHg from paddy soil to rice grain

Several lines of evidence confirm MeHg and inorganic Hg(II) are accumulated in different proportions in rice plant tissue. Compared to roots, stalks, and leaves, MeHg is more concentrated in rice grain, while concentrations of THg (or inorganic Hg(II)) are highest in the roots, suggesting more efficient uptake and loading of MeHg to the grain compared to inorganic Hg(II) (Rothenberg et al., 2011b; Zhang et al., 2011a). Similar results were observed for other plant species, including plants ingested by deer (Gnamus et al., 2000) and typical plant varieties from a coniferous forest (Schwesig and Krebs 2003). Bioaccumulation factors (BAFs) for paired rice grain and soil concentrations from 11 studies are summarized in Table 5. Most MeHg BAFs were >1, indicating MeHg was more concentrated in rice grain compared to soil, which was consistent with BAFs reported by Zhang et al. (2010a). In addition, the ratio between MeHg and THg BAFs averaged 11,000–47,000, suggesting higher accumulation of MeHg in rice grain compared to THg (or inorganic Hg(II)) from soil.

Differential accumulation from soil of inorganic Hg(II) and MeHg in rice grain may be related to phytochelatin. Unlike other metal cations, inorganic Hg(II) is not an essential micronutrient (Patra and Sharma, 2000); however, inorganic Hg(II) is phytotoxic at high concentrations (> 50 μ M), causing water stress, reduced plant growth, blockage of nutrients in the roots, and decreased levels of chlorophyll and proteins (see Zhou et al., 2007 and references therein). To control the uptake of metals, plants up-regulate cysteine-rich phytochelatin, which chelate inorganic Hg(II) and inhibit the translocation of inorganic Hg(II) from the roots to the aerial portions of the plant (Cobbett and Goldsbrough, 2002). Krupp et al. (2009) investigated phytochelatin in rice (*Oryza sativa*) and horehound (*Marrubium vulgare*), and identified inorganic Hg(II)-phytochelatin but no MeHg-phytochelatin complexed in the roots, suggesting binding to phytochelatin may limit the translocation of inorganic Hg(II) from rice roots to stems, but not MeHg.

Although rice grain accumulates MeHg more efficiently than inorganic Hg(II), there are significant differences between rice varieties (Li et al., 2013; Zhu et al., 2008). For example, 50 varieties of rice commonly cultivated in Guizhou province, China, were grown in three sites along a contamination gradient within the province (Rothenberg et al., 2012). Polished rice inorganic Hg(II) and MeHg concentrations were standardized for each site, and the association with rice variety (as a categorical variable) was assessed using regression. Rice MeHg levels were significantly associated with rice varieties, but rice IHg levels were not (MeHg: $p < 0.001$, IHg: $p = 0.44$), suggesting there were possibly genetic markers controlling the accumulation of MeHg in the filial tissue (Rothenberg et al., 2012). In addition, within the most highly contaminated site, MeHg levels in some rice varieties were up to 69% lower compared to high-accumulating MeHg varieties, indicating there were safer rice varieties to plant, which were immediately available to rice farmers (Rothenberg et al., 2012). Within rice-growing regions where Hg contamination is serious, it may be possible to significantly reduce MeHg exposure merely by recommending different varieties, which are adapted to the region's growing conditions.

Inorganic Hg(II) and MeHg are distributed differently in polished and unpolished rice grain. To the best of our knowledge, only one study compared rice grain THg and MeHg concentrations between polished and unpolished rice (Rothenberg et al., 2011b). Rice grain THg levels were significantly higher in unpolished rice compared to polished rice, while MeHg levels did not differ, suggesting MeHg was more evenly distributed within maternal and filial tissues, while inorganic Hg(II) was more concentrated in the bran (i.e., maternal tissue). Results indicated polishing rice, which often decreases rice nutrient levels (Heinemann et al., 2005; Sellappan et al., 2009; Villareal et al., 1991), will not likely reduce rice MeHg concentrations. The ability of MeHg to bypass the bottleneck between the maternal and filial tissue (i.e., the ovular vascular trace, Krishnan and Dayanandan, 2003) was also reported for organo-selenium and organo-arsenic species (Selenium: Li et al., 2010; Sun et al., 2010; Williams et al., 2009; Arsenic: Carey et al., 2010; Lombi et al., 2009; Meharg et al., 2008). However, most organo-arsenic species are less toxic than methylated arsenic species (Rosen and Liu, 2009), and some methylated selenium species are anticarcinogenic (Sun et al., 2010). Efforts to breed rice lines, which potentially accumulate more methylated arsenic in the filial tissue and less inorganic arsenic (e.g., Norton et al., 2012), may inadvertently increase accumulation of MeHg, potentially increasing adverse health effects. Measures to mitigate rice arsenic levels need to be assessed in conjunction with other trace elements, including MeHg.

6. Conclusions and research needs

MeHg exposure through rice ingestion is an emerging global health issue, although its geographic extent and the risks posed to rice consumers are largely unknown. The vast majority of the studies on this topic were conducted in China (59%), with nearly a quarter (25%) in a single province. Data from other rice-growing regions of the world are needed to determine the extent of the health threat posed by MeHg in rice. More research is also needed to determine the risks from MeHg exposure through rice ingestion, which may differ from exposure through the much better-studied pathway of fish ingestion.

This review provides an inventory of studies reporting rice THg and MeHg concentrations, including the range of data from known polluted and non-polluted sites, which may be used by rice farmers and managers to aid in the selection of management practices. Rice farmers are faced with two problems: rice paddies are hot spots of Hg(II)-methylation, and rice plants are efficient accumulators of MeHg. Several features associated with rice cultivation, including flooded conditions, anoxic soils, and high net primary productivity, make rice paddies conducive sites for Hg(II)-methylation. This is due to enhanced cycling of redox-sensitive elements and availability of organic C, both of which promote microbial activity and inorganic Hg(II) bioavailability.

Certain features associated with accumulation of Hg species in rice grain are known. First, MeHg appears to be more efficiently accumulated in the filial tissue (i.e., polished rice) compared to inorganic Hg(II). Second, there may be genetic markers associated with uptake and translocation of MeHg in rice grain. Third, post-harvest management practices, including removal of rice straw before re-flooding, may be employed to mitigate MeHg production. However, to better understand the specific mechanisms through which Hg

species are translocated, stable isotopes labeled for inorganic Hg(II) and MeHg can be applied to paddy soil as tracers, to test whether rice grain MeHg originates from the soil or inorganic Hg(II) is methylated *in planta*. These studies are needed to better mitigate rice MeHg levels.

Management practices for rice production must be carefully selected after taking into account a wide range of goals including lowering contaminant uptake, minimizing water use, and reducing methane emissions. Rice cultivation methods employing intermittent wetting and drying will lower water use and methane emissions but may increase rice MeHg levels compared to rice grain cultivated under continuously flooded conditions. Given the pressure on rice farmers to adapt water-saving rice cultivation methods, there is an urgent need to develop management practices that take multiple contaminants into account due to potential differences in toxicity and bioavailability between Hg species and other trace elements (e.g., arsenic). These practices should be highly tailored to the geology and pollution levels at the village level, with separate recommendations for polluted and non-polluted sites, and less reliance on blanket regional recommendations. The research community needs to broaden its focus beyond individual pollutants, such as MeHg or arsenic, and take into account a variety of contaminants when studying rice agriculture.

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Highlights

- Found 51 studies from 15 countries concerning rice and mercury/methylmercury.
- Highest rice mercury/methylmercury in China, India, and Indonesia.
- Flooding and intermittent flooding in rice paddies promoted mercury methylation.
- Mitigation should be tailored for mercury polluted and non-polluted sites.

Figure 1. Selection process for studies included in the comprehensive inventory of rice total Hg (THg) and methylmercury (MeHg) concentrations.

Figure 2.

Global inventory of rice ingestion rates (g rice/capita/day) (FAO, 2013), and spatial distribution of a) rice total mercury (THg) (ng/g) concentrations and b) rice methylmercury (MeHg) (ng/g) concentrations, separated into quintiles. From Table 1 references, data were excluded for rice purchased in Saudi Arabia (reference #1, Al-Saleh and Shinwari, 2001), Spain (from Japan and Thailand, reference #8, Da Silva et al., 2010), France (reference #18,

Leblanc et al., 2004), and rice cultivated or purchased in multiple provinces in China (reference # 10, Fang et al., 2014; reference #23, Li P., et al., 2012; reference #32, Qian et al., 2010). All other data from studies in Table 1 were mapped.

Figure 3.

Boxplots for concentrations of rice grain a) total mercury (THg) (ng/g), b) methylmercury (MeHg) (ng/g), and c) rice %MeHg (of THg) categorized by pollution sources, including BG (background levels from non-polluted sources), Hg (mercury mining), Gold (artisanal and small scale gold mining), Chlor (contamination of irrigation water from chlor-alkali plants) and Other (zinc and lead mining, proximity to coal-fired power utilities and e-waste incineration, and other industrial pollution).

Figure 4.

Two-way scatterplot for rice grain total mercury (THg) versus methylmercury (MeHg) (both variables log_{10} -transformed) for polluted (black circle) and non-polluted (open circle) sites. The dotted line represents the theoretical 1:1 curve, and the solid line represents the regression line. Results listed are for weighted least squares regression, using the study sample size as the analytical weight.

Figure 5. Biogeochemical factors influencing methylmercury production in rice paddies.

Global inventory of 43 studies reporting Hg concentrations for rice grain, including total mercury (THg), methylmercury (MeHg) and/or %MeHg (of THg). In addition to background information (e.g., country of origin), summary statistics include the mean and parenthetical range. NA indicates data were not available.

Table 1

Rice country of origin	Sampling/purchasing site	Polluted site?	Market-basket survey?	Polished rice grain?	Sample size	THg (ng/g)	MeHg (ng/g)	%MeHg (of THg)	Method ¹	Reference	Ref #
India	Riyadh, Saudi Arabia	No	Yes	Yes	17	1.6 (<3.0–3.309)	NA	NA	AAS	Al-Saleh and Shinwari, 2001 ²	1
		No	Yes	Yes	4	1.8 (<3.0–3.5)	NA	NA			
		No	Yes	Yes	2	1.631 (0.513–2.75)	NA	NA			
		No	Yes	Yes	2	23.7 (3.8–43.5)	NA	NA			
		No	Yes	Yes	2	<3.0	NA	NA			
Philippines	Mindanao, Philippines	Yes, gold mining	No	No	NA	20 (1–43)	NA	NA	Flame-AAS	Appleton et al., 2006 ²	2
Philippines	Mindanao, Philippines	Yes, gold mining	No	Yes	NA	18 (8–50)	NA	NA	ICP-MS	Batista et al., 2012	3
Brazil	Brazil	No	Yes	Yes	23	2.3 (0.3–10.4)	NA	NA			4
China	Jiangsu province, China	Yes, industrial runoff	No	No	23	5.7 (1.0–13)	NA	NA			5
China	Zhoushan Island, China	No	No	No	6	9	4	44.4			6
China	Guizhou province, China	Yes, chemical plant	No	No	13	30.7	18.7	NA	CV-AAS (THg), Electron capture (MeHg)	Cheng J. et al., 2013	7
China	Shanghai, China	No	No	No	NA	8.1	6.0	NA	AAS (THg), CVAFS (MeHg)	Cheng Z. et al., 2013	
Cambodia	Kampong, Cambodia	No	Yes	Yes	6	8.14 (6.16–11.7)	1.44 (1.17–1.96)	NA			
	Kratie, Cambodia	Yes, gold mining	Yes	Yes	6	12.7 (9.90–16.7)	1.54 (1.06–2.31)	NA			
	Kandal, Cambodia	No	Yes	Yes	6	10.2 (5.91–15.1)	2.34 (0.48–5.23)	NA			
	Recife and Sao Paulo, Brazil	No	Yes	Yes	9	3.1 (2.1–4.4)	NA	NA			
Brazil	Recife and Sao Paulo, Brazil	No	Yes	Yes	9	3.1 (2.1–4.4)	NA	NA	CV-AFS	Da Silva et al., 2010	8
Spain	Valencia, Spain	No	Yes	Yes	6	2.1 (1.6–3.3)	NA	NA	CV-AFS	Da Silva et al., 2013	
Japan	Valencia, Spain	No	Yes	Yes	5	3.1 (1.2–7.8)	NA	NA			
Thailand	Valencia, Spain	No	Yes	Yes	4	2.6 (1.3–3.7)	NA	NA			
Spain	Palma de Mallorca, Spain	No	Yes	Yes	12	4.48 (2.15–7.25)	NA	NA			
China	22 provinces, China	No	No	Yes	92	2 (trace–19)	NA	NA	AFS	Fang et al., 2014 ³	10
China	Guizhou province, China	Yes, Hg mining	No	Yes	25	58.5 (21.1–191.9)	14.6 (7.5–27.6)	27.2 (7.9–65.9)	CV-AFS	Feng et al., 2008 ⁴	11
China	Guizhou province, China	Yes, Hg mining	No	Yes	18	21.3 (10–66.9)	5.7 (3.3–10.2)	30.8 (6.1–72.3)			
China	Guizhou province, China	Yes, Hg mining	No	Yes	27	33.1 (4.9–214.7)	4.0 (1.9–14.7)	17.7 (2.4–75.1)			

Rice country of origin	Sampling/purchasing site	Polluted site?	Market-basket survey?	Polished rice grain?	Sample size	THg (ng/g)	MeHg (ng/g)	%MeHg (of THg)	Method ^l	Reference	Ref #
China	Guizhou province, China	No	No	Yes	24	7 (3.2–15.1)	2.5 (–8–4.3)	40.8 (9.6–88.3)			
China	Zhejiang et al., China	Yes, e-waste	No	Yes	13	22 (15.6–68.4)	NA	NA	Hydride generation-AFS	Fu et al., 2008	12
China	Jiangsu province, China	Yes, industrial pollution	No	Yes	155	14.5	NA	NA	Hydride generation-AFS	Hang et al., 2009	13
China	Guizhou province, China	Yes, Hg mining	No	No	10	149 (11.1–569)	38.9 (8.03–144)	42.7 (5.46–72.3)	CV-AFS	Horvat et al., 2003	14
China	Guizhou province, China	Yes, power plant, and chemical plant	No	No	4	14.5 (2.53–33.4)	11.3 (071–28)	59.0 (28.1–83.8)			
Italy	Italy	No	Yes	Yes	1	5.21	0.86	16.5			
China	Zhejiang province, China	No	Yes	Yes	224	5.0 (<5.0–88)	NA	NA	HG-AFS	Huang et al., 2013	15
China	Zhejiang province, China	No	No	Yes	216	22.4 (2.46–65.85)	NA	NA	AFS	Jiang et al., 2012	16
Indonesia	Indonesia	Yes, gold mining	No	No	6	NA	57.7 (10.6–115)	NA	CV-AFS	Krisnayanti et al., 2012	17
Indonesia	Indonesia	Yes, gold mining	No	Yes	1	NA	1.02	NA			
NA	Paris, France	No	Yes	Yes	3	5	NA	NA	ICP-MS	Leblanc et al., 2005 ⁵	18
Korea	Korea	No	Yes	Yes	1	2	NA	NA	CV-AAS	Lee et al., 2006 ⁶	19
India	Ganjam, India	Yes, chloralkali facility	No	No	6	510 (470–530)	NA	NA	CV-AAS	Lenka et al., 1992	20
China	Guizhou province, China	Yes, Hg mining	No	No	26	26 (13–52)	9.4 (3.5–23)	39.2 (17.7–89)	AFS (THg) CV-AFS (MeHg)	Li B. et al., 2013	21
China	Hunan province, China	Yes, Hg mining	No	No	26	29 (11–58)	11 (6.5–24)	45.8 (16.3–96.4)			
China	Guangdong province, China	Yes, zinc and lead mining	No	No	26	15 (1.5–52)	1.2 (0.28–3.5)	16.2 (1.0–72)			
China	Guizhou province, China	Yes, Hg mining	No	Yes	17	26.8 (6–113)	7.8 (3.8–12.3)	40.2 (6.0–83.6)	CV-AFS	Li P. et al., 2008	22
China	Guizhou province, China	No	No	Yes	2	2.75 (2.1–3.4)	1.3 (0.6–2.0)	44 (28.8–59.3)			
China	7 provinces, China	No	Yes	Yes	284	10.1 (0.86–47.2)	2.47 (0.13–18.2)	NA	CV-AFS	Li P. et al., 2012 ⁷	23
China	Hunan province, China	Yes, Hg mining	Both	Both	33	30.6 (10–150)	NA	NA	CV-AAS	Li Y., 2013	24
Taiwan	Taiwan	No	Yes	Yes	407	1 (<10–40)	NA	NA	AAS	Lin et al., 2004	25
Philippines	Honda Bay, Philippines	Yes, historical gold mining	No	No	2	3.345 (3.08–3.61)	NA	NA	NA	Maramba et al., 2006	26
China	Guizhou province, China	No	No	No	11	6.2 (3.8–10.3)	2.9 (1.8–4.5)	47 (35–64)	CV-AFS	Meng et al., 2010 ⁸	27
China	Guizhou province, China	Yes, Hg mining	No	No	22	187 (24.8–548)	7.0 (3.8–18)	8.1 (1.6–23)			
China	Guizhou province, China	Yes, Hg mining	No	No	13	302 (128–622)	32 (18–62)	11 (7.4–15)			
China	Guizhou province, China	Yes, Hg mining	No	No	21	93.6	30.4	NA	CV-AFS	Meng et al., 2014	28
China	Guizhou province, China	Yes, Hg mining	No	No	11	85.8	20.8	NA			
China	Guizhou province, China	Yes, Hg mining	No	No	15	22.0	12.3	NA			

Rice country of origin	Sampling/purchasing site	Polluted site?	Market-basket survey?	Polished rice grain?	Sample size	THg (ng/g)	MeHg (ng/g)	%MeHg (of THg)	Method ¹	Reference	Ref #
China	Guangdong province, China	Yes, zinc and lead mining	No	No	18	16.0	2.0	NA			
Japan	Mie prefecture, Japan	Yes, chloralkali facility	No	No	25	22.5 (3–60)	NA	NA	AAS	Morishita et al., 1982	29
Japan	Niigata prefecture, Japan	No	No	No	11	3 (trace-11)	NA	NA	AAS	Nakagawa et al., 1998	30
Japan	Niigata prefecture, Japan	No	No	Yes	19	1.0 (trace-8.0)	NA	NA			
Thailand	Phichit Province, Thailand	Yes, gold mining	No	Yes	4	212 (172–268)	NA	NA	CV-AAS	Patanawat et al., 2007	31
China	20 provinces, China	No	Yes	Yes	712	5.8 (0.02–31)	NA	NA	Hydride generation-AFS	Qian et al., 2010 ⁹	32
China	Guizhou province, China	Yes, Hg mining	No	No	70	228 (10.3–1120)	25.3 (1.61–174)	NA (1.4–93)	CV-AFS	Qiu et al., 2008	33
China	Shaanxi province, China	Yes, Hg mining	No	Yes	10	103 (51–200)	22 (8.2–80)	19 (9.9–40)	CV-AFS	Qiu et al., 2012b	34
China	Shaanxi province, China	Yes, Hg mining	No	Yes	42	NA	17 (4.0–78)	NA	CV-AFS	Qiu et al., 2012c	35
China	Guizhou province, China	Yes, Hg mining	No	Yes	26	22 (10–45)	11 (3.2–39)	NA	AAS (THg), CV-AFS (MeHg)	Qiu et al., 2013	36
China	Hubei province, China	No	No	No	12	3.7 (1.9–6.8)	1.2 (0.33–3.0)	31 (9.9–54)	AAS (THg), CV-AFS (MeHg)	Rothenberg et al., 2011	37
China	Hubei province, China	No	No	Yes	12	3.3 (1.7–6.5)	1.2 (0.57–3.1)	36 (17–52)			
China	Guizhou province, China	Yes, Hg mining	No	Yes	50	120 (72–190)	63 (31–100)	54 (37–79)	AAS (THg), CV-AFS (MeHg)	Rothenberg et al., 2012	38
China	Guizhou province, China	Yes, Hg mining	No	Yes	50	18 (11–34)	9.9 (1.5–18)	56 (8.6–120)			
China	Guizhou province, China	No	No	Yes	50	2.8 (1.0–5.5)	2.0 (0.37–3.3)	75 (12–140)			
China	Guizhou province, China	Yes, Hg mining	No	Yes	17	12 (2.4–38)	5.0 (1.2–13)	46 (23–72)	AAS (THg), CV-AFS (MeHg)	Rothenberg et al., 2013	39
India	Gaganvati, India	No	No	NA	7	38 (26–58)	NA	NA	CV-AAS	Sarkar et al., 2012	40
China	15 provinces, China	No	Yes	Yes	25	23 (6.3–39.3)	4.7 (1.9–10.5)	22.2 (7–38)	GC-AFS	Shi et al., 2005 ¹⁰	41
Brazil	Salavador City, Brazil	No	Yes	Yes	7	8.36 (4.10–13.72)	NA	NA	CV-AAS	Silva et al., 2012	42
India	Southern India	No	No	Yes	6	7.4	NA	NA	CV-AAS	Srikumar, 1993	43
India	Southern India	No	No	No	6	3.1	NA	NA			
Tanzania	Rwagagasa, Tanzania	Yes, gold mining	Both	Both	3	26 (11–35)	NA	NA	AAS	Taylor et al., 2005	44
China	Hunan province, China	Yes, industrial pollution	No	No	30	10 (1.0–60)	NA	NA	AAS	Wang and Suanes, 2003	45
China	Guizhou province, China	Yes, Hg mining	No	No	15	7	NA	NA	ICP-MS	Wang et al., 2011	46
USA	California, USA	Yes, historical gold and Hg mining	No	No	14	50.4 (50.0–50.9)	4.6 (4.1–5.0)	9.1 (8.1–10.0)	CV-AFS	Windham-Meyers et al., in press, a	47
Thailand	Thailand	No	No	No	108	4.8 (3.6–22.3)	NA	NA	CV-AAS	Zarcinas et al., 2004	48
China	Guizhou province, China	Yes, Hg mining	No	No	32	105 (8.6–504)	11 (1.2–44)	17 (3.5–40)	CV-AFS	Zhang et al., 2010a ⁸	49
China	Guizhou province, China	No	No	No	19	50.8 (7.4–194)	5.8 (1.4–24)	21 (3.3–40)			

Rice country of origin	Sampling/purchasing site	Polluted site?	Market-basket survey?	Polished rice grain?	Sample size	THg (ng/g)	MeHg (ng/g)	%MeHg (of THg)	Method ¹	Reference	Ref #
China	Guizhou province, China	No	No	No	8	29.7 (9.6–79)	4.7 (1.6–13)	18 (6.9–40)			
China	Guizhou province, China	Yes, Hg mining	No	No	59	78	9.3	NA	CV-AFS	Zhang et al., 2010b ²	50
China	Guizhou province, China	Yes, power plant	No	No	65	5.5	2.2	NA			
China	Guizhou province, China	Yes, zinc smelting	No	No	16	2.3	1.6	NA			
China	Guizhou province, China	No	No	No	10	3.2	2.1	NA			
China	Zhejiang province, China	Yes, e-waste	No	Yes	12	37.5 (20–60)	NA	NA	ICP-MS	Zhao et al., 2010 ⁴	51
China	Zhejiang province, China	No	No	Yes	3	10	NA	NA			

¹ Method abbreviations include flame atomic absorption spectrophotometry (Flame-AAS), inductively coupled-mass spectrometry (ICP-MS), cold vapor atomic absorption spectrometry (CV-AAS), cold vapor atomic fluorescence spectrometry (CV-AFS), hydride generation atomic fluorescence spectrometry (hydride generation-AFS), atomic fluorescence spectrometry (AFS), atomic absorption spectrophotometry (AAS), and gas chromatography atomic fluorescence spectrophotometry (GC-AFS).

² Rice samples were oven-dried prior to THg or MeHg analysis.

³ 22 provinces in China include Heilongjiang, Jilin, Liaoning, Hebei, Shandong, Shanxi, Ningxia, Gansu, Qunghai, Xinjiang, Jiangsu, Anhui, Zhejiang, Hunan, Hubei, Jiangxi, Guangdong, Guangxi, Fujian, Yunnan, Guizhou, Sichuan. Data for each province are not provided.

⁴ Reported the geometric mean, not arithmetic mean.

⁵ Data are for rice and semolina.

⁶ The sample from Korea is cooked rice.

⁷ 7 provinces in China include Guangdong, Shanghai, Jiangsu, Guangxi, Hunan, Jiangxi, and Guizhou. Data for each province are not provided.

⁸ To obtain THg concentrations, values for inorganic Hg and MeHg were summed.

⁹ 20 provinces not specifically identified.

¹⁰ 15 provinces in China include Jiangsu, Fujian, Liaoning, Hubei, Heilongjiang, Neimenggu, Guangxi, Henan, Sichuan, Ningxia, Shandong, Guizhou, Tianjin, Anhui, and Hebei. Data for each province were provided.

Table 2

Breakdown by country for 51 studies measuring rice THg and/or MeHg concentrations, including rice from mercury (Hg)-polluted or non-polluted sites, and references.

#	Sampling or purchasing country	Region	# of studies in non-polluted sites	# of studies in polluted sites	References
1	Brazil/Spain	Non-polluted: 1 Different regions of Brazil 2 Salvador City, Bahia State, Brazil 3 Samples purchased in Valencia (Spain), and Recife and Sao Paulo (Brazil), and were originally cultivated in Brazil, Japan, Spain and Thailand	3	0	Batista et al., 2012; Da Silva et al., 2010; Silva et al., 2012
2	Cambodia	Polluted: Kratie, Kampong, and Kandal	0	1	Cheng et al., 2013
3	China	Non-polluted: Hubei province (1), Zhejiang province (1), Zhoushan Island (1), multiple provinces (4) Polluted: Guizhou province (13), Zhejiang province (3), Jiangsu province (2), Shaanxi province (2), Hunan province (2), multiple provinces (1)	7	23	Cao et al., 2010; Cheng et al., 2009, 2013; Fang et al., 2014; Feng et al., 2008; Fu et al., 2008; Hang et al., 2009; Horvat et al., 2003; Huang et al., 2013; Jiang et al., 2012; Li B. et al., 2013; Li P. et al., 2008, 2012; Li, Y. et al., 2013; Meng et al., 2010; Meng et al., 2014; Qian et al., 2010; Qiu et al., 2008, 2012a, 2012b, 2013; Rothenberg et al., 2011, 2012, 2013; Shi et al., 2005; Wang and Stuanes, 2003; Wang et al., 2011; Zhang et al., 2010a, 2010b; Zhao et al., 2010
4	France	Non-polluted: Purchased in Paris	1	0	Leblanc et al., 2005
5	India	Non-polluted: 1 Gaganvati, India 2 Southern Indian markets Polluted: 1 Ganjam, India	2	1	Lenka et al., 1992; Sarkar et al., 2012; Srikumar, T.S., 1993
6	Indonesia	Polluted: Sekotong artisanal mining area	0	1	Krisnayanti et al., 2012
7	Japan	Non-polluted: 1 Niigata prefecture, Japan Polluted: 1 Mie prefecture, Japan	1	1	Morishita et al., 1982; Nakagawa et al., 1998

#	Sampling or purchasing country	Region	# of studies in non-polluted sites	# of studies in polluted sites	References
8	Republic of Korea	Non-polluted: NA	1	0	Lee et al., 2006
9	Philippines	Polluted: 1 Mindanao 2 Honda Bay	0	2	Appleton et al., 2006; Maramba et al., 2006
10	Saudi Arabia	Non-polluted: Purchased in Riyadh, cultivated in India, Egypt, Thailand, Australia, and USA.	1	0	Al-Saleh and Shinwari, 2001.
11	Spain	Non-polluted: 1 Palma de Mallorca, Spain	1	0	Da Silva et al., 2013
12	Taiwan	Non-polluted: Nationwide	1	0	Lin et al., 2004
13	United Republic of Tanzania	Polluted: Rwamagasa artisanal gold mining area, northwest Tanzania	0	1	Taylor et al., 2005
14	Thailand	Non-polluted: 1 Nationwide Polluted: 1 Phichit Province, Thailand	1	1	Pataranawat et al., 2007; Zarcinas et al., 2004
15	USA	Polluted: Sacramento River region, California	0	1	Windham-Meyers et al., in press, a

Table 3

Summary statistics, including mean, median and range, for studies reporting average rice total mercury (THg) and methylmercury (MeHg) concentrations for polluted sites and for non-polluted sites (the latter including market-basket surveys). See Tables 1 and 2 for references.

	Parameter	Mean (ng/g)	Median (ng/g)	Range (ng/g)	Sample Size (n)
Non-polluted	THg (ng/g)	8.2	4.1	1.0–45	40
	MeHg (ng/g)	2.5	2.2	0.86–5.8	16
	% MeHg (of THg)	36	36	17–75	11
Polluted	THg (ng)	65	26	2.3–510	42
	MeHg (ng/g)	16	11	1.2–63	30
	% MeHg (of THg)	30	29	8.1–56	16

Summary of rice cultivation practices, including hydrology, planting and post-harvest treatment of rice residue, and their influence on mercury (Hg) cycling and methylmercury (MeHg) production in rice paddies.

Table 4

	Hydrology		Planting	Post-harvest Treatment of Rice Residue	
	Continuous flooding of paddy soil	Alternating wetting and drying		Burning rice residue	Allow residue to degrade in the field
Promotes the following:	Anoxia in soil subsurface, increased anaerobic activity, attenuation of pH	Anaerobes are dormant, then revived, electron acceptors replenished	Increased exudates from rice roots, increased acidity, increased rhizosphere concentration and rhizooxidation promoting replenishment of sulfur and iron electron acceptors, increased transpiration and shading through leaf area	Increased atmospheric Hg levels and particulate matter	Degradation of rice residue increases available carbon
Links to Hg(II)-methylation	Hg(II)-methylation by anaerobes (sulfate-reducing bacteria and iron-reducing bacteria, methanogens)	Causes periodic spikes in net MeHg production in paddy soil	Hg(II)-methylation by anaerobes, rice canopy decreases photo-demethylation of MeHg in surface water	Increased deposition of carbon and atmospheric Hg(II) to rice paddies	Readily available carbon increases Hg(II)-methylation rates

Table 5

Bioaccumulation factors (BAFs) (unitless) for total mercury (THg) (n=11 studies) and methylmercury (MeHg) (n=6 studies), where BAF is defined as the ratio between the concentration in rice grain and soil for THg and MeHg.

	Parameter	BAF Mean $\pm$ 1 SD	BAF Median	BAF Range
Overall	THg	0.32 $\pm$ 1.9	0.0089	0.00012–15
	MeHg	5.5 $\pm$ 6.7	3.2	0.20–34
	MeHg/THg	2900 $\pm$ 4700	930	18–21,000
Polished rice	THg	0.87 $\pm$ 3.2	0.014	0.00037–15
	MeHg	4.3 $\pm$ 4.2	2.7	0.69–17
	MeHg/THg	1100 $\pm$ 1600	550	18–6300
Unpolished rice	THg	0.024 $\pm$ 0.53	0.0052	0.00012–0.16
	MeHg	6.6 $\pm$ 8.5	4.9	0.20–34
	MeHg/THg	4700 $\pm$ 6200	2900	21–21,000

References: Appleton et al., 2006; Horvat et al., 2003; Jiang et al., 2012; Morishita et al., 1982; Pataranawat et al., 2007; Qiu et al., 2012b; Rothenberg et al., 2011b, 2012; Taylor et al., 2005; Windham-Meyers et al., in press, a; Zarcinas et al., 2004