



## Review

## Mercury methylation in rice paddy and accumulation in rice plant: A review

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## ABSTRACT:

The bioavailability and toxicity of mercury (Hg) are dependent on its chemical speciation, in which methylmercury (MeHg) is the most toxic compound. Inorganic Hg can be transformed into MeHg in anaerobic conditions. Subsequent accumulation and biomagnification in the food chain pose a potential threat to human health. Previous studies have confirmed that paddy soil is an important site for MeHg production, and rice fields are an important source of MeHg in terrestrial ecosystems. Rice (*Oryza sativa* L.) is recently confirmed as a potential bioaccumulator plant of MeHg. Understanding the behaviour of Hg in rice paddies is important, particularly the mechanisms involved in Hg sources, uptake, toxicity, detoxification, and accumulation in crops. This review highlights the issue of MeHg-contaminated rice, and presents the current understanding of the Hg cycling in the rice paddy ecosystem, including the mechanism and processes of Hg species accumulation in rice plants and Hg methylation/demethylation processes in rice paddies and the primary controlling factors. The review also identified various research gaps in previous studies and proposes future research objectives to reduce the impact of Hg-contamination in rice crops.

## 1. Introduction

Because of its high toxicity, mercury (Hg) has been listed as a priority pollutant by the United Nations Environment Program (UNEP), the World Health Organization (WHO), the European Union (EU), and the United States Environmental Protection Agency (US EPA). Due to its physicochemical properties, Hg is of global concern because it can exist in the natural environment in different oxidation states (Mergler et al., 2007). The mobility and toxicity of Hg are largely dependent on its chemical speciation, in which methylmercury (MeHg) is the most toxic compound (Hasegawa et al., 2005; Agency for Toxic Substances and Disease Registry (ATSDR), 2013). Inorganic Hg, which exhibits less toxicity, can be converted into MeHg in anaerobic biogeochemical conditions and enter food chains through biomagnification processes (Stein et al., 1996). Although fish consumption has been identified as one of the predominant exposure pathway of MeHg worldwide (Clarkson, 1993; Agency for Toxic Substances and Disease Registry (ATSDR), 2013), recent studies has confirmed rice consumption as another significant exposure route to residents in Hg-contaminated areas in Southwestern China (Feng et al., 2008; Zhang et al., 2010a; Li et al., 2015, 2017a, 2017b; Du et al., 2018).

Rice is the staple food for half of the global population; therefore,

research concerning Hg cycling in the paddy ecosystem has received considerable attention in past decades (Meng et al., 2018b). This review highlights the issue of methylmercury-contaminated rice, and then comprehensively and systematically summarises the current understanding of Hg methylation/demethylation processes in rice paddies and the mechanism of Hg species accumulation in rice plants. The review further commented on the shortcomings of previous studies and makes recommendations for future research.

## 2. Methylmercury-contaminated rice

Horvat et al. (2003) first announced the MeHg-contaminated rice seed (140 µg/kg) collected from Wanshan Hg mining sites in Guizhou province, China. Qiu et al. (2008) further demonstrated that the levels of MeHg detected in rice seed obtained at abandoned Hg mining sites can reach up to 180 µg/kg, which is approximately 100–1000 times higher than that in other local crops, including cabbage, tobacco, rape, and corn. MeHg-contaminated rice were also widely observed in other Hg mining sites in China, such as Xiushan in Chongqing, Wuchuan in Guizhou province, Xunyang in Shannxi province, and Xinhuang in Guangxi province (Qiu et al., 2006, 2012; Li et al., 2013; Xu et al., 2018). The environmental issue of Hg-contaminated rice also exists in

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industrial areas, such as the compact fluorescent lamp manufacturing area (Liang et al., 2015), electronic waste recycling area in Zhejiang province (Tang et al., 2015), chemical plant in Guizhou province (Horvat et al., 2003), and coal-fired power plant in Hunan province (Xu et al., 2017). Moreover, elevated levels of total Hg or MeHg in rice seeds were widely observed in other Hg-polluted areas around the world, including Ganjam in India (associated with a chlor-alkali plant, Lenka et al., 1992), California in the USA (gold and mercury mining activities, Windham-Myers et al., 2014), Phichit province in Thailand (gold mining activities, Pataranawat et al., 2007), Mindanao in the Philippines (artisanal gold mining, Appleton et al., 2006) and Lombok Island in Indonesia (artisanal gold mining, Krisnayanti et al., 2012).

Generally, Hg is considered to be existence as inorganic form in crop plants (WHO, 1991). Consequently, food safety standards are recommended based on total Hg levels, and therefore, safe guidelines of MeHg in foodstuff is unavailable. However, previous studies revealed that the MeHg level in rice grain exceeded the permissible limit of 20 µg/kg issued by the Chinese National Standard Agency (Meng et al., 2014; Qiu et al., 2008). More importantly, the percentage of MeHg to total Hg in rice grain can be very high and reach up to 72% (Meng et al., 2014). Thus, these studies have confirmed that rice plants can bioaccumulate MeHg and that MeHg contamination in rice is a common phenomenon in Hg-polluted areas. Brombach et al. (2017) and Cui et al. (2017) further reported the Hg concentrations in commercial market rice and rice-based products from the EU and USA. Both adults and infants in the EU and USA face potential health risks from Hg exposure through rice or rice-based product consumption (Brombach et al., 2017; Cui et al., 2017). Recently, a modelling simulation predicted that the Hg concentration in Chinese rice could increase by 13% if policies to regulate Hg emission are not introduced (Kwon et al., 2018).

As an important MeHg source in terrestrial ecosystems, paddy fields represent potential pathways for MeHg migrating to the terrestrial food chain. Rice is grown across the globe to serve as a primary nutrition source for nearly half of the world's population. Therefore, rice cultivation represents as an important position in agriculture throughout South and East Asia (FAO, 2002). The latest report from the Food and Agriculture Organization (FAO) of the United Nations indicated that the total area of land under rice cultivation is 163 million hectares globally, with ~50% produced in China (FAO, 2016). In China, the majority of Hg mining sites are distributed in Guizhou province, Hunan province, Chongqing, Yunna province, and Shanxi province, which are also the main rice-growing areas. With its rapidly developing economy, China holds the position in Hg producer, user, and emitter in the world. As a result, the concentrations of atmospheric Hg in China were significantly higher than those in developed regions and countries like Europe and America (Fu et al., 2012). The high levels of Hg in atmosphere may cause large-scale Hg pollution in rice once it is deposited into rice-planting areas.

It has been reported that rice consumption has become a predominant route of MeHg exposure to its consumers in Southwestern China (Feng et al., 2008; Zhang et al., 2010a; Li et al., 2015, 2017a, 2017b; Du et al., 2018). In Hg-polluted regions, the daily exposure to MeHg reaches up to 1.8 µg/kg of body weight, which is nearly 200 times higher than the edible standards recommended by the US EPA (Qiu et al., 2008). Therefore, the health risks of eating MeHg-contaminated rice need greater attention. Moreover, the problem of MeHg-contaminated rice requires urgent interventions in agriculture environments, especially in Hg-polluted areas. Therefore, understanding the mechanisms and processes of Hg conversion and accumulation in rice paddies is very important, which could potentially provide guidance on remediation of mercury-contaminated paddy soil, reduction of MeHg uptake in rice plant, and Hg exposure to local residents.

### 3. Mercury methylation/demethylation in rice paddies

Research on the biogeochemical cycle of Hg in paddy fields has

intensified since the discovery of MeHg accumulation in rice (Meng et al., 2018b). Methylation of inorganic Hg influences the MeHg content in paddy soil, which directly affects the MeHg levels in rice seeds (Zhao et al., 2016a, 2016b). With respect to the extreme lipophilicity, high neurotoxicity, and health risks of MeHg exposure, studies on the mechanism of Hg methylation in rice paddies have important practical significance to lower MeHg accumulation in rice and to reduce human health risks through contaminated rice consumption.

Generally, the net MeHg production in natural environments is largely controlled by the abundance and activity of bacteria that can methylate Hg (Hg-methylators) (Wang et al., 2014; Liu et al., 2014a, 2014b, 2018). In addition, Hg methylation is influenced by the organic content, electron acceptor ( $\text{SO}_4^{2-}$  and  $\text{Fe}^{3+}$ ), electron donor ( $\text{F}^{2+}$  and  $\text{S}^{2-}$ ), and bioavailable Hg content that can be used by Hg-methylators (Marvin-DiPasquale et al., 2009; Rothenberg and Feng, 2012; Ullrich et al., 2001; Zhao et al., 2016b). Due to the seasonable irrigation during the rice-growing periods, rice paddy fields are recognised as an intermittent wetland ecosystem. Previous studies revealed active Hg methylation in paddy fields, which resulted in MeHg accumulation in rice (Zhao et al., 2016a, b; Meng et al., 2010, 2011). The paddy field is a complicated ecosystem, in which atmosphere, rainfall, irrigation water, overlying water, soil (pore water), and rice are important components. The biogeochemical cycle of Hg in the rice paddy ecosystem involves multi-media/interface migration processes (e.g. overlying water-atmosphere, overlying water-surface soil layer, soil-pore water, and pore-water/soil-rice plant) and multiple biological, physical, and chemical transformation processes (e.g. reduction, oxidation, methylation and demethylation). Moreover, these processes are coordinated by multiple biological and abiotic factors.

#### 3.1. Microbiology

It is widely acceptable that the Hg methylation in the natural environment mainly occurs under anaerobic conditions with the participation of anaerobic microorganisms (e.g. iron-reducing bacteria (IRB), sulphate-reducing bacteria (SRB), and methanogens) (Ullrich et al., 2001; Peng et al., 2012; Rothenberg and Feng, 2012; Wang et al., 2014; Liu et al., 2014a, 2014b; 2018; Parks et al., 2013; Vishnivetskaya et al., 2018). However, the abiotic pathway of Hg methylation can be negligible (Ullrich et al., 2001). Nevertheless, not all strains of IRB, SRB, and methanogens participate in Hg methylation. The Hg-methylator strains are randomly distributed in phylogenetic trees (Ranchou-Peyruse et al., 2009; Gilmour et al., 2013). Researchers revealed a correlation between Hy methylation and a two-gene cluster composed of *hgcA* (which encodes a corrin protein) and *hgcB* (which encodes a ferro-reduced protein), in two species of bacteria (Parks et al. (2013)). The *hgcAB* genes are found in all currently known Hg-methylator strains, but they were not detected in non-methylator bacteria, indicating that *hgcAB* genes are required for the biotic pathway of Hg methylation (Parks et al., 2013). Recently, with the development of DNA sequencing technology and improvements in detecting Hg-methylation genes in different microorganism populations (Christensen et al., 2016; Liu et al., 2018), the *hgcAB* marker can not only be used as an effective method to discover new Hg-methylators, but also to evaluate Hg-methylation in specific environments.

Recent studies have demonstrated that there are significant differences in Hg-methylator bacteria in different ecosystems and even in different regions of the same ecosystem (Gilmour et al., 2013; Parks et al., 2013; Schaefer et al., 2002, 2004; 2014; Liu et al., 2018). As a typical seasonal waterflooding wetland ecosystem, the paddy field provides a favourable anaerobic condition for methylation of inorganic Hg (Zhao et al., 2016a, 2016b; Wang et al., 2014). Moreover, the microorganism population in the rice paddy ecosystem is very complex, and it includes various microorganism species, including SRB and IRB (Stubner and Meuser, 2000; Hori et al., 2010). Moreover, the paddy field is also an important habitat for methanogens (Grosskopf et al.,

1998; Bao et al., 2014). Paddy soil that has a Hg contamination gradient and different types of Hg pollution can influence the microbial community abundance, composition, diversity, and functional properties (e.g. abundance of functional genes) (Liu et al., 2018).

Liu et al. (2014a, 2014b) conducted a systematic study on methylation by microbial populations in rice paddy field at the Wanshan Hg mining area in Guizhou province, China. These studies indicated a significantly positive correlation between MeHg concentration with the *hgcA* gene abundance and SBR community in rice paddy soil, which implied that microbes carrying *hgcA* contributed to methylation of inorganic Hg (Liu et al., 2014a, b). Moreover, the microorganisms that participated in Hg methylation containing *hgcA* were mainly represented by *Euryarchaeota*, *Chloroflexi*, *Firmicutes*, *Deltaproteobacteria*, and two unidentified groups (Liu et al., 2014a, 2014b). Specifically, most of the *Deltaproteobacteria* and some unknown clusters were related to SBR and IRB. Moreover, researchers recently revealed that some *hgcAB* sequences were largely relevant to other Hg-methylators, including *Desulfovibrio* sp. J2, *Desulfuromonas* sp. DDH964, and *Geobacter anodireducens*, in which *Geobacter anodireducens* contributed to Hg methylation (Liu et al., 2018). Therefore, IRB, SBR, and methanogens were recently recognised as the predominant Hg-methylators in paddy fields (Zhao et al., 2016b; Liu et al., 2014a, 2014b; Wang et al., 2014).

According to previous studies, the syntrophism among microorganisms can significantly promote the capacity of Hg-methylators, and even non-methylation strains can participate in Hg methylation from syntrophism associations (Yu et al., 2012). Yu et al. (2012) developed a syntrophism model and further confirmed that the syntrophism (e.g. between SBR and methanogens, *Syntrophobacter* spp., *Desulfovibrio* spp., *Syntrophobacter* spp.) not only stimulated the activity of the Hg-methylator but also converted the non-methylator into a potential Hg-methylator. Sequencing analysis of a Hg-methylator carrying the *hgcA* gene revealed that most of the *hgcA* sequences belong to *Synergistes* (Bae et al., 2014, 2015). This study further speculated that the *Synergistes* might be dominant Hg-methylators in the wetland ecosystem. Therefore, it is generally accepted that the syntrophism among microorganisms can potentially facilitate Hg methylation. However, microorganisms might inhibit each other due to nutrient competition, consequentially offsetting the effect. Bravo et al. (2015) reported the competitive relationship between SBR and iron-reducing bacteria in Hg-polluted soil. This study showed that the addition of a specific SBR inhibitor could not inhibit Hg methylation in sediment; rather, the SBR inhibitor stimulated iron-reducing bacteria and thereby promoted Hg methylation in the sediment (Bravo et al., 2015). The role of syntrophism among microorganisms in rice paddy fields as an inhibitor or a promoter for Hg methylation is not well understood, and several key questions have yet to be answered. For example, does a common syntrophism association among microorganisms facilitate Hg methylation, or does syntrophism start a competitive relationship and further influence Hg methylation in rice paddy fields? What are the specific processes and mechanisms involved in how syntrophism influences Hg methylation in the rice paddy ecosystem? Further studies are required to address these questions.

Recently, a metagenomics study revealed that the bacterial abundance in paddy fields located in a Hg mining area was negatively correlated with total Hg and MeHg concentrations, while bacterial diversity was positively correlated with MeHg concentration (Liu et al., 2018). Interestingly, no relationship was obtained between the MeHg concentration and the abundance of the Hg-methylator *hgcA* gene (Liu et al., 2018). This study implied that the bacterial abundance and/or bacterial diversity cannot fully explain the Hg-methylation and the net production of MeHg in a natural environment. It is possible that the metagenomics technique can only estimate whether or not the Hg-methylator exists; however, it cannot detect the activity of Hg-methylators and thereby fails to determine the expression of corresponding functional genes as well as the enzyme activity. For this reason, researchers have begun to focus on the expression level of *hgcA* mRNA in

addition to the influence of microflora on the methylation of Hg (Goni-Urriza et al., 2015; Bravo et al., 2016). To date, no data is available on the expression of Hg-methylation genes in rice paddy fields.

### 3.2. The speciation and fraction of mercury

It is generally accepted that the bioavailable fractions of Hg were recognised as the most hazardous form in natural soil/sediment since it can participate in Hg methylation as the potential substrate of Hg (Bishop et al., 1998). A recent study reported that concentrations of total Hg and bioavailable Hg in paddy soil of an abandoned Hg mining site were approximately 100–200 times higher than those of an artisanal Hg mining site in the corresponding region in the Wanshan Hg mining area, Guizhou Province, China (Zhao et al., 2016a, 2016b). According to traditional thinking, MeHg concentration in the paddy soil of abandoned Hg mining sites should be logically higher than that of the artisanal Hg mining site. However, researches failed to observed any relationship between MeHg and total Hg concentrations in paddy soil, and the so-called “bioavailable Hg” is not the primary substrate which participates in Hg methylation in paddy soil. As such, the determination of total Hg and bioavailable Hg is insufficient to fully understand the transformation of Hg species in rice paddy ecosystem (Zhao et al., 2016b; Lin et al., 2012; Tessier et al., 1979).

Numerous studies confirmed that free Hg ion ( $Hg^{II}$ ),  $Hg^{II}$  bonded to natural organic matter (NOM-Hg),  $Hg^{II}$  reacted with mackinawite ( $\equiv FeS-Hg^{II}$ ), and nanoparticulate mercuric sulphide (nano-HgS) are distributed extensively in soils and participate in Hg methylation as potential Hg substrate species (Zhu et al., 2018; Jonsson et al., 2012; Zhang et al., 2012, 2014). There are several orders of magnitude difference in the methylation rate among these Hg compounds (Zhu et al., 2018; Jonsson et al., 2012; Zhang et al., 2012, 2014). However, the characteristics and the distribution patterns of Hg species/fractions in rice paddy fields in Hg mining areas, as well as key Hg species/fractions participating in methylation remain unknown.

Again, the levels MeHg in paddy soil and corresponding rice plants of an artisanal Hg mining area were 1–2 orders of magnitude higher than those of an abandoned Hg mining area (Meng et al., 2010). Accordingly, the authors suggested that the relatively higher MeHg levels in paddy soils at artisanal Hg mining site were linked to the new atmospheric deposition of Hg, and the bioavailable Hg received from newly deposited Hg appeared to be the key parameter controlling net Hg methylation in rice paddies. Recently, the *in situ* methylation/demethylation rate constants in rice paddy soil was further qualified by researchers (Zhao et al., 2016b). Results showed that the methylation rate constants were highly elevated in the paddy soil of the artisanal Hg mining area compared with that of the abandoned Hg mining area. This indicated that the new atmospheric deposition source of Hg was more easily methylated compared to the ambient Hg forms in rice paddy soil, and also that the older Hg in soil was not available for methylation (Zhao et al., 2016a, 2016b; Meng et al., 2010, 2011). Here, we speculate that the newly deposited Hg might be formed as simple adsorption-desorption interaction with soil particulates, and seems bioavailable to Hg methylators after originally goes into the rice paddy soils. Such Hg might act as the potential substrate of Hg methylation and be readily transformed into MeHg by microorganisms. In a different way, the old Hg could be tightly bound to organic material and form strong Hg-OM complexes, and/or react with sulphur to produce mercuric sulphide compounds ( $\beta$ -HgS) over time. Both Hg-OM and  $\beta$ -HgS complexes are more stable and exist as less available forms for methylation (Schuster, 1991). The authors further proposed that Hg concentration in atmosphere could be a useful monitoring indicator to possible evaluate and predict the relative levels of MeHg in the paddy soil and corresponding rice plant, which could help assess and reduce health risks associated with MeHg accumulation in rice grain (Zhao et al., 2016a, 2016b). More attention should be given to the control and reduction of the anthropogenic emissions of Hg from industrial and Hg

smelting activities, especially in rice planting areas, to reduce MeHg exposure risks from rice consumption.

### 3.3. Organic material

Organic matter plays an important role in regulating microbial Hg methylation in natural environments. This is generally attributed to the decomposition of organic matter, which may stimulate organic nutrients to affect microbial methylation of inorganic Hg to MeHg (Ullrich et al., 2001). Therefore, numerous studies reported the positive correlations between organic matter content and MeHg concentration in sediments/soil in aquatic ecosystems (Lucotte et al., 1999; Meng et al., 2016). Zhang et al. (2010b) observed that the organic content in paddy soil was positively correlated with MeHg levels in corresponding rice plants. However, they failed to observe a relationship between the bioaccumulation of MeHg in rice seeds and the organic content in the corresponding soil. Moreover, statistical analysis revealed that the influence of organic matter on the MeHg levels was less pronounced in paddy soil across the Wanshan Hg mining area (Zhao et al., 2016a, 2016b; Yin et al., 2018). Other studies have shown that the concentration of MeHg was significantly correlated with organic matter content as well as the abundance of Hg methylators in rice paddy soil (Liu et al., 2014a, 2016, 2018). It appears that MeHg production and accumulation in rice plants are related to the organic matter in rice paddy soil.

Reports have demonstrated that dissolved organic matter produced through rice residue return (a globally prevalent farming practice) can be an important factor controlling MeHg production in paddy fields (Zhu et al., 2015a, 2015b; 2016; Tang et al., 2019). Incorporation of rice residues into soils might promote Hg methylation in paddy soil (Zhu et al., 2015a, 2015b; 2016; Tang et al., 2019), and consequently increased MeHg concentrations in paddy soil and rice grain (Tang et al., 2019). Such increases were explained by the enhanced abundances and/or activities of microbial methylators and the formation of Hg-S-DOM complexes, which potentially stimulate microbial Hg methylation in soil. The authors further suggested that the organic matter derived from rice residue decomposition increased the accumulation of MeHg in rice through 1) enhancement of MeHg mobility in paddy soil, and 2) increase of MeHg uptake by rice roots and accumulation in rice seeds (Tang et al., 2019). Liu et al. (2016) specified that rice straw return can enhance methylation of inorganic Hg in serious Hg-contaminated paddy soil, but this promoting effect was less pronounced in paddy soil with relatively low Hg levels, which was explained by the differences in microbial communities associated with Hg methylation between Hg-contaminated and non-Hg-contaminated soil.

In summary, the role of organic matter in regulating Hg methylation in rice paddies is far to be fully understood. We propose that the absolute organic matter might be indirect and work together with other factors in Hg methylation process (Zhao et al., 2016a, 2016b; Zhang et al., 2010b; Yin et al., 2018). However, the fresh organic matter derived from rice residue decomposition can enhance the methylation of inorganic Hg in rice paddy soil and accumulation of MeHg in rice seeds. Therefore, rice residue return could enlarge the ecological risk of MeHg contamination in rice paddies, thus requiring carefully consideration on straw management, especially in Hg-polluted area.

### 3.4. pH

The pH can not only impact the bioavailability of Hg by influencing the exchange properties of soil particles but can also impact the dissolution of Hg by changing the composition of organic matter to control Hg species and Hg methylation processes in paddy soil (Ullrich et al., 2001). Steffan et al. (1988) and Ullrich et al. (2001) specified that acidic conditions are favourable for Hg dissolution in soil, thereby increasing the bioavailability of Hg, which could further promote the methylation of inorganic Hg. In contrast, alkaline conditions might

inhibit Hg methylation by decreasing the bioavailability of Hg in soil. Liu et al. (2015) reported that at an acidic pH, hydrogen ions ( $H^+$ ) compete for active adsorption sites with mercury ions ( $Hg^{2+}$ ) on the soil particles. Consequently, free  $H^+$  is combined with colloidal substances that have a negative charge and the  $Hg^{2+}$  will be released into the liquid phase of soil, which could potentially increase the bioavailability of Hg and stimulate Hg methylation. At an alkaline pH, soil particles might attract and retain  $Hg^{2+}$ , which can inhibit the bioavailability of Hg, thus decreasing the accumulation Hg in rice plants (Liu et al., 2015; Xu et al., 2014; Tang et al., 2018). Moreover, an acidic pH in paddy soil after straw amendment could promote the desorption of MeHg from soil particles to liquid phase, and then enhancing MeHg production and bioavailability, and facilitating the accumulation of MeHg in rice plant (Tang et al., 2019).

Generally, the methylation and demethylation processes are largely dependent on pH values (Ullrich et al., 2001). Stepwise regression analysis showed that the concentration of MeHg in paddy soil was controlled by numerous parameters, in which soil pH accounted for 52% (Tang et al., 2018). The overlying water and irrigation water in rice paddy of artisanal Hg mining area represented weak acidic conditions that are optimal for Hg methylation (Zhao et al., 2016a, 2016b); while for the rice paddies of an abandoned Hg mining area, they were shown to be more alkaline (Zhao et al., 2016a, 2016b). Thus, highly elevated methylation rates jointly with relatively low demethylation rates were observed in paddy soil of an artisanal Hg mining area compared with those of an abandoned Hg mining area (Zhao et al., 2016a, 2016b). The authors suggested that the relatively low pH, on one hand, stimulated the Hg methylation; on the other hand, inhibited the MeHg demethylation in the paddy soil of an artisanal Hg mining area. On the opposite, the alkaline conditions in rice paddy of abandoned Hg mining site may increase the degradation of MeHg. Similarly, Huang et al. (2018) revealed a negative correlation between MeHg concentrations in rice seeds and the values of soil pH, which was attributed to the stimulation of MeHg demethylation in soil porewater at higher pH. However, some other researchers failed to observe any relationship between pH values and MeHg concentration in rice paddy soil across the Wanshan Hg mining area, implying that individual pH cannot fully explain Hg methylation/demethylation processes in paddy soil (Yin et al., 2018; Zhao et al., 2016a, 2016b). Rothenberg and Feng (2012) reported an acidic pH in paddy soil porewater possibly attributed to the production of carbon dioxide ( $CO_2$ ) through microbial activities. However, the authors suggested that pH was not likely to be a primary driver for Hg methylation in the paddy soil since no relationship between pH and MeHg in soil porewater was observed within a 24-h timeframe.

### 3.5. Redox conditions

It is generally accepted that relatively low redox potential (Eh) in anaerobic conditions not only promotes Hg methylation but also inhibits MeHg demethylation (Gilmour and Henry, 1991). Hg methylation mainly occurs in anaerobic environments, which favour the growth of anaerobic methylators (e.g. SRB, IRB, and methanogens) in rice paddies. Generally, rice paddy ecosystems are typically vertically stratified with a 5–10 cm layer of overlying water. However, the paddy field is drained prior to harvest and thereafter kept dry during the ripening period, which lasts for more than 30 days. The redox conditions in paddy fields change with the alternating flooded and drying conditions during the rice-growing period and thus affect biogeochemistry cycling of Hg in the rice paddy ecosystem.

Zhao et al. (2016a) observed considerably elevated levels of MeHg in rice paddy soil during the flooded period. However, the MeHg levels in paddy soil decreased during the harvest period under aerobic conditions (Zhao et al., 2016a). This study implied that soil redox conditions could be an important factor in influencing the bioavailability and methylation/demethylation of Hg in rice paddy ecosystems. Changing

soil Eh by way of water management represents a method to effectively restrain the Hg accumulation in rice plant. Peng et al. (2012) revealed that the Eh in soil under flooded environment was significantly lower than in aerobic conditions. Consequently, the concentrations of total Hg and MeHg as well as the percentage of MeHg to total Hg in soil pore-water following flooding were much higher than those under the aerobic treatment. This study clearly showed that the bioavailability of Hg and the Hg methylation/demethylation were controlled by soil Eh with different treatments (flooding and aerobic conditions). Wang et al. (2014) reported that the bioavailability of Hg and net MeHg production in rice paddy soil were greatly reduced under aerobic conditions when compared with conventional continuously flooded conditions, which was explained by the decreased abundance of anaerobic microorganism involving in Hg methylation in paddy soil. Recently, Tanner et al. (2018) observed increased soil redox from an alternate wetting and drying treatment compared to a conventional continuously flooded condition, which resulted in a significant reduction of MeHg levels accompanied by oxidation of Fe(II) in soil. The authors further suggested that aerobic conditions through water management inhibit the activities of Hg-methylators, and could be a potential pathway to decrease MeHg production in rice paddy soils (Tanner et al., 2018).

In summary, low Eh condition in paddy soils can stimulate Hg mobilisation from solid phase into the liquid phase, which potentially increase the Hg bioavailability, and then favours the Hg methylation. Moreover, anaerobic conditions with low Eh could potentially promote activities of various anaerobic microbes involved in inorganic Hg methylation, together with the relatively weak MeHg degradation. In addition, the relatively high Eh under aerobic conditions could decrease the bioavailability of Hg in paddy soil, while potentially inhibiting the activities of microbial Hg methylation accompanied with accelerated MeHg demethylation.

### 3.6. Iron and sulphur

As summarised above, the Hg methylation is mainly mediated to some degree of SRB and IRB in rice paddies under anoxic conditions. Moreover, net MeHg production is largely facilitated by the activities of Hg-methylators, which are regulated by the bioavailability of electron donor (e.g.  $S^{2-}$  and  $Fe^{2+}$ ), electron acceptors (e.g.  $Fe^{3+}$  and  $SO_4^{2-}$ ), and  $Hg^{2+}$ . Therefore, iron (Fe) and sulphur (S) are essential elements for plant growth and their biogeochemical cycling impacts the Hg methylation in rice paddies (Ullrich et al., 2001; Marvin-DiPasquale et al., 2014).

Generally, mercury ion ( $Hg^{2+}$ ) can be transformed into insoluble mercuric sulphide ( $HgS$ ) in the presence of sulphide ( $S^{2-}$ ) under anoxic conditions. Consequently, Hg methylation is inhibited due to the reducing bioavailability of  $Hg^{2+}$  through the formation of insoluble  $HgS$ . However,  $HgS$  can also form soluble Hg-S chemical compounds (e.g.  $HgS_2^{2-}$ ) under excess sulphide conditions, which increase the bioavailability of Hg, and thus promote Hg methylation. Sulphate could also stimulate Hg methylation at low sulphate concentrations in aquatic ecosystems (Benoit et al., 1999, 2001; 2003; Jeremiason et al., 2006), whereby  $SO_4^{2-}$  can serve as an electron acceptor for SRB to potentially stimulate Hg methylation by enhancing the activities of microbial methylators (Gilmour et al., 1992; Ullrich et al., 2001). Therefore, significantly positive correlation between sulphate concentrations and net Hg methylation was observed in rice paddy soil (Zhao et al., 2016b). The authors suggested that sulphate could enhance microbial sulphur reduction and subsequently stimulate the Hg methylation in rice paddy soil (Zhao et al., 2016a, 2016b). Therefore, sulphate addition was reported to promote the net MeHg production in rice paddy, and then enhanced MeHg accumulation in rice seeds, probably attributed to the increased Hg bioavailability from the release of  $HgS$  to soluble  $Hg^{2+}$  together with the enhanced SRB activity (Li et al., 2019; Wang et al., 2015, 2016; Zhong et al., 2018). In contrast, sulphate was suggested to inhibit Hg methylation process in paddy soil especially under sulphate-

abundant condition (Wang et al., 2015; Li et al., 2017a, 2017b), which was explained by the reduced Hg availability to methylators under anoxic conditions caused by the strong affinity of Hg ions ( $Hg^{2+}$ ) to reduced sulphate species (e.g. polysulfides and organic thiols) (Wang et al., 2015) and conversion of RS-Hg-SR ( $Hg(GS)_2$ ) to the stable  $HgS$  (Li et al., 2017a, 2017b). Therefore, researchers reported that sulphate addition can effectively inhibit MeHg production in paddy soil and accumulation of MeHg in rice grain due to the reduction of Hg motility in rhizosphere soil (Wang et al., 2015). From the reviewed literature, we can summarise that sulphur plays a dual stimulation/inhibition role in influencing Hg methylation, depending on the speciation and concentration of sulphur in the paddy soil (Li et al., 2017a, 2017b). Consequently, conflicting results concerning the regulation of sulphur on the solubility, bioavailability, and methylation of Hg in rice paddy have been reported (Li et al., 2017a, 2017b, 2019; Wang et al., 2015, 2016). To date, the exact mechanisms of the interaction between sulphur and Hg in the rice paddy ecosystem are poorly understood.

Similar to sulphur, iron was also confirmed to be an important factor that controls Hg methylation/demethylation processes in wetland/aquatic ecosystems through iron oxide adsorption and reduction pathways (Ulrich and Sedlak, 2010; Ullrich et al., 2001; Kim et al., 2004; Wiatrowski et al., 2006; 2009). In addition, Hg adsorbed to iron oxides can be released into the liquid phase during the reduction process to impact Hg methylation (Slowey and Brown, 2007). Therefore, the Hg cycling is closely linked to the biogeochemical processes of Fe in rice paddies (Marvin-DiPasquale et al., 2014; Zhong et al., 2018). The reduction rate of  $Fe^{3+}$  as an electron acceptor was positively correlated with the Hg methylation rate, which further showed that IRB participated to some degree with Hg methylation (Marvin-DiPasquale et al., 2014). Therefore, the ferrous iron ( $Fe^{2+}$ ) concentration was recommended as a predictor of net MeHg production rates in rice paddy soil (Marvin-DiPasquale et al., 2014).

The biogeochemistry cycling of iron and sulphur co-occur and interact with each other, especially under alternate dry and wet conditions. For example, inorganic sulphur mainly exists as  $HS^-$ ,  $S^{2-}$  or S, and iron mainly exists as  $Fe^{2+}$ , allowing FeS or  $FeS_2$  to readily form. Therefore, the methylation of inorganic Hg is inhibited because solid FeS can effectively distract the bioavailable Hg by ways of co-precipitation and/or adsorption. A field study suggested that  $Fe^{2+}$  could restrain sulphur activates through the formation of FeS in rice paddy soil, and may consequently influence the bioavailability and methylation of inorganic Hg (Zhao et al., 2016b). Rothenberg and Feng (2012) further suggested that  $Fe^{3+}$  reduction can increase electron acceptors for SRB through oxidation of sulphide to sulphate instead of forming FeS, which potentially enhanced the methylation of inorganic Hg in rice paddy soil. Zhong et al. (2018) specified that the interaction of both iron and sulphate to influence Hg methylation in paddy soil was less pronounced compared with the separate treatment of iron or sulphate. Despite advances in the biogeochemical cycling of Hg in rice paddies, the individual and interactive influences of S and Fe on Hg methylation/demethylation processes in rice paddy soil still far to be fully understood. More detailed work is required to verify the relative importance of these two elements (S and Fe) in microbial Hg methylation of rice paddy ecosystems.

## 4. The mechanism of mercury accumulation in rice

### 4.1. Source of Hg species in rice plant

From analysing the publicly available data, we believe that rice seeds can accumulate high levels of MeHg, posing a potential threat to people and wildlife (Feng et al., 2008; Zhang et al., 2010a; Li et al., 2015, 2017a, 2017b; Du et al., 2018; Abeyasinghe et al., 2017a, 2017b; Xu et al., 2019). Therefore, understanding the bioaccumulation sources of inorganic Hg and MeHg in rice plants could provide environmental risk mitigation strategies. Horvat et al. (2003) attempted to show the

relative source of Hg in rice based on the relationship between the Hg concentration in rice and corresponding soil. However, this study failed to observe any relationship between total Hg or MeHg in rice and soil from the Wanshan Hg mining area. The authors suggested that the accumulation and retention of Hg in rice could be influenced by the concentration of total Hg in soil and other factors, such as air and soil Hg sources. However, a positive correlation between MeHg in soil and rice from the chemical industry area of Qingzhen was observed (Horvat et al., 2003). A study was subsequently conducted by Meng et al. (2010, 2011, 2012), which focused on the bioaccumulation pathways of inorganic Hg and MeHg in tissues rice plants. Results based on statistical analysis (e.g. regression analysis, principal components analysis, and factorial analysis) of the Hg concentrations in different parts of rice plants showed that the atmosphere is the principal source of inorganic Hg to the above-ground portions of rice plants. Specifically, both soil and ambient atmosphere provide inorganic Hg to the stalk, however, the soil source tends to be less pronounced. Differently, soil Hg pool was confirmed to be a predominant source of inorganic Hg in root, and a unique bioaccumulation pathway of MeHg for tissues of rice plants (Meng et al., 2010, 2011, 2012).

The diffusive gradient in thin films technique (Liu et al., 2012) and Hg isotope tracer technique (Strickman and Mitchell, 2017), have provided data to further confirm the relative bioaccumulation pathways of Hg species in the tissues of rice plants described above. However, the relative contribution of MeHg for rice plants from other sources is still in debate. Although planta demethylation of MeHg in rice plants was reported in previous studies (Strickman and Mitchell, 2017; Li et al., 2016), evidence of *in situ* methylation of inorganic Hg in rice plants is still unavailable. A recent study suggested that in addition to the soil MeHg source, the atmospheric source of dimethylmercury (DMeHg), which is originally derived from paddy soil, could be a potential contributor to a fraction of the MeHg in rice plants (Wang et al., 2018). However, more detailed studies are required to ascertain and confirm this alternative source.

#### 4.2. The distribution patterns and localisation of Hg species in rice plants

Based on field observations in different Hg-polluted areas, the distribution and localisation characteristics of inorganic Hg and MeHg in different parts of the rice plant have been previously studied (Meng et al., 2010, 2014). Generally, the distribution patterns of inorganic Hg concentration and content were largely dependent on the relative Hg levels in the soil compared to the atmosphere. From significant Hg contamination in the ambient air, the shoot regions of rice plants accumulated the majority of inorganic Hg, particularly in the leaves, at an artisanal Hg mining site (Meng et al., 2010). However, elevated inorganic Hg concentration and content in rice root tissues at abandoned Hg mining sites could be attributed to Hg-contaminated rice paddies (Meng et al., 2010). In contrast, the highest concentrations of MeHg were generally observed in rice grain regardless of sampling sites, and then root, hull, talk, and leaf. Moreover, the majority of the MeHg was stored in the rice seeds, which confirmed that the rice seed had the greatest ability to bioaccumulate MeHg compared to other tissues (Meng et al., 2010; Zhang et al., 2010b). The different distribution patterns between inorganic Hg and MeHg in the tissues of rice plants indicate varying sources and/or mechanisms of inorganic Hg and MeHg uptake into rice plant tissues (Meng et al., 2010, 2011, 2012).

The speciation and localisation of Hg species in different parts of rice grain (white rice, bran, and hull) have been studied (Rothenberg et al., 2011; Qiu et al., 2012; Meng et al., 2014). Results showed that rice bran represented the highest levels of inorganic Hg and MeHg followed by the hull and white rice (Meng et al., 2014). Moreover, most of the inorganic Hg in rice grain was observed in the bran and hull. Differently, most of the MeHg was detected in edible white rice. Consequently, the majority of inorganic Hg is removed, but most of the MeHg still remains in the edible parts (white rice) during grain

processing (Meng et al., 2014). Rothenberg et al. (2011) further suggested that MeHg as a possible organic nutrient could utilise membrane transport pathways in the endosperm more efficiently than inorganic Hg, which could explain the relative difference in the localisation of Hg species (MeHg vs. inorganic Hg) in different parts of brown rice grain. Using synchrotron radiation microscopic X-ray fluorescence (SR-uXRF), Meng et al. (2014) revealed the prominent localisation of Hg at the surface of the brown rice grain, which correspond to the pericarp and aleurone layers, and a gradient Hg levels from surface layers of rice grain (pericarp and aleurone) to the centre part (endosperm). The elevated levels of Hg located in the surface layers of the rice grain (pericarp and aleurone) could be due to the affinity of Hg ions to the protein-ligand side chains (Meng et al., 2014). Although some studies have investigated the relative localizations of Hg species in rice grains, the implication and mechanism of inorganic Hg and MeHg storage in different parts of rice grain is still required.

#### 4.3. The processes of Hg species accumulation in rice plants

The processes of inorganic Hg and MeHg accumulation in tissues of rice plants during the whole rice growing periods have been studied (Meng et al., 2010, 2012; 2014; Cui et al., 2014; Zhang et al., 2010b; Liu et al., 2012). A steady increase in the concentrations and mass of inorganic Hg in the above-ground parts of rice plants (e.g. the leaf and stalk) were generally observed during rice growing periods, especially for the rice plants grown in Hg mining areas that were exposed to highly elevated Hg vapour (Meng et al., 2010; Cui et al., 2014). Generally, Hg in atmosphere is dominated by  $Hg^0$  (>95%) (Schroeder and Munthe, 1998). The stomatal or nonstomatal routes were hypothesized to be the potential pathways of the atmospheric  $Hg^0$  vapour exchange and uptake into above-ground parts of rice plant (foliage and stalk) (Meng et al., 2012; Cui et al., 2014). Alternatively, atmospheric  $Hg^0$  can be physical removal through deposition usually preceded by oxidation to  $Hg^{2+}$  species (Shia et al., 1999). Deposited  $Hg^{2+}$  to leave surface from the atmosphere could traverse the epicuticular dissolution into the underlying cuticle and diffuse to epidermal cells (Moeckel et al., 2008). Therefore, the accumulated inorganic Hg from atmospheric source was fixed into leaf and stalk tissues, and cannot be re-emitted to atmosphere or translocated to other parts (e.g. seed, hull, and root) (Meng et al., 2012, Meng et al., 2018a, ; Cui et al., 2014). However, the influence of atmospheric Hg speciation on inorganic Hg accumulation in above-ground parts of rice plant is still unclear. Thus, to better understand this process, further work needs to be done. In contrast, the rice root (iron plaque) might represents as a potential barrier to inorganic Hg absorption and accumulation, which potentially sequesters inorganic Hg and consequently reduces the Hg uptake through the root to the above-ground parts (Meng et al., 2012; Cui et al., 2014; Zhang et al., 2010b). Moreover, the formation of Hg-phytochelatin complexes were observed in the rice root, which can effectively trap the inorganic Hg in rice root system but not translocate to above-ground parts (Krupp et al., 2009).

As described previously, the bioavailability and toxicity of Hg are largely dependent on its chemical speciation (Hasegawa et al., 2005). Therefore, the chemical speciation of Hg in rice grain was further studied based on X-ray absorption near-edge spectroscopy (XANES). Meng et al. (2014) proposed that inorganic Hg in rice grain was predominantly bound to cysteine and associated with phytochelatin, which possibly explained why the majority of Hg was located in surface layers of rice grain (pericarp and aleurone) but not in the centre part (endosperm) (Rothenberg et al., 2011). The localisation of inorganic Hg in different parts of rice grain and the chemical speciation of inorganic Hg in rice grain was recognised as mechanisms of self-detoxification or self-protection when the rice plant exposed to Hg-contaminated atmosphere (Meng et al., 2014; Rothenberg et al., 2011; Cui et al., 2014).

The mechanism and process of inorganic Hg accumulation in rice plants were shown to be different from that of MeHg (Meng et al., 2011, 2012, 2014). Generally, the bioaccumulation and translocation of

MeHg from the root systems to the above-ground parts was described as a dynamic process. Rice root can effectively absorb MeHg from soil, which suggested that these processes (absorption) within plant-soil interface occurred more easily than that of inorganic Hg (Meng et al., 2011; Krupp et al., 2009). Phytochelatin – small peptides that detoxify heavy metals in plants – were observed in rice plants and could sequester Hg<sup>2+</sup> but not MeHg (Krupp et al., 2009). Therefore, MeHg can pass through the physical barrier on the rice root surface (iron plaque). The authors speculated that the MeHg in roots behaves like a mobile plant nutrient, and then translocated to the above-ground parts. Consequently, very limited amount of MeHg was maintained in the rice root during the rice-growing season. During rice growing periods, most of the MeHg was stored in the leaf and stalk of the premature plant, however, the MeHg which temporarily located in leaf and stalk, was further transported and accumulated into ripen rice seeds when harvested (Meng et al., 2011). XANES and HPLC-ICP-MS observations confirmed that MeHg in rice ripen seeds was exclusively existence as MeHg-S compound (MeHg-cysteine), which is responsible for the transfer of MeHg across the blood-brain and placental barriers (Meng et al., 2014; Li et al., 2010). The authors, therefore, speculated that free MeHg-cysteine can be recognised as a movable compound and acts similar with free cysteine or amino acid. This MeHg complex might be associated with proteins in rice grain. The chemical speciation of MeHg explained the dynamic process and localisation of MeHg accumulation in rice plants (Meng et al., 2011, 2014).

## 5. Conclusions and future perspective

Most studies concerning MeHg in rice have been conducted in China. More data are urgently in rice growing areas at the regional level especially in other Asian countries, Africa, South America and North America. Such data is critical to further understand the global issue of Hg contaminated rice and related health risk. It is clear that the mechanism of Hg methylation/demethylation in rice paddies and accumulation in rice plant as well as their controlling factors are very complex and far from being fully understood. The key methylators in paddy soils were identified as SRB, methanogens, and iron-reducing bacteria. However, the relative contribution of microorganisms in Hg methylation in rice paddy soil is still unclear. More important, previous studies largely focused on the Hg methylation process in rice paddy soil. On the opposite, some other transformation processes of Hg such as the MeHg demethylation, Hg reduction/oxidation, and their controlling factors and how these processes impact Hg methylation were generally neglected; as such, further studies are required. Until now, no empirical model or mechanism model for Hg accumulation from soil to rice is available. The key challenges to develop model could be attributed to the unknown genetic factors associated with MeHg accumulation in rice plants and the unclear absorption and translocation pathways/channels of MeHg in rice root. Although previous studies proposed that newly deposited Hg is more easily methylated into MeHg in paddy soils, their conclusions were based on indirect evidence. In addition, previous studies failed to distinguish the newly deposited Hg and ambient Hg stored in paddy soil. Consequently, the relative contribution of these two Hg sources (newly deposited Hg VS. old Hg) to Hg methylation especially in Hg-polluted areas is still unknown. Therefore, further study is needed to uncover the Hg dynamics in rice paddy soils.

## Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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