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Original Research

The Bioaccumulation and Migration of Inorganic Mercury and Methylmercury in the Rice Plants

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Abstract

In the present study, $(NH_4)_2S_2O_3$ extraction methods were applied to assess bioaccumulation of methylmercury (MeHg) in rice grains, and inorganic mercury (IHg) concentrations in rice leaves were analyzed during the rice growing time to access the bioaccumulation of IHg in rice leaves. The results show that the IHg concentrations in leaves increased in the rice harvest stage, indicating that the limit or no IHg was migrated to the rice grain. Also, the Hg-contaminated leaves may potentially cause the input of 'new Hg' into soil, leading to a vicious Hg pollution cycle in a rice paddy system. Our results indicated that MeHg concentrations in leaves could not be used to predict the MeHg bioaccumulation in rice grain. Meanwhile, MeHg transferred capability from soil to leaves decreased with time, which could be the common effect of the decreased soil MeHg bioavailability and translocation of MeHg from leaves to rice grains.

Keywords: methylmercury, inorganic mercury, bioaccumulation, rice leaves

Introduction

Mercury (Hg) is a global pollutant that can be methylated to methylmercury (MeHg), which is of great concern due to its biomagnification through the food chain. Recently, there has been an increasing awareness of food safety problems resulting from Hg-contaminated farmland [1-2]. It has been reported that extremely high concentrations of total mercury (THg, e.g., 330 -790,000 ng/g) and MeHg (e.g., 0.13-23 ng/g) have been found in contaminated farmland soils [3-4], and rice plants grown in Hg-contaminated areas could accumulate high levels of MeHg in the grain [5], leading to MeHg exposure for people. This could be because flooded conditions in Hg-polluted rice paddy fields favor MeHg yield and could possibly result in extremely high MeHg bioaccumulation in rice plants [6]. Considering that rice is a staple food in Asia, consumption of rice could be an important MeHg exposure route for residents in some mercury-polluted areas [7]. Therefore, it is of great importance to explore the underlying mechanisms of bioaccumulation of inorganic mercury (IHg) and MeHg in the rice plants, thus helping to predict the bioaccumulation and risk of Hg in

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rice plants.

MeHg uptake from soil is considered the major pathway of MeHg accumulation in rice plants [8]. MeHg in soil was first absorbed by roots and then translocated to the above-ground parts (leaves and stalk, [9]). However, IHg accumulated in rice plants could mainly come from the atmosphere through leaf uptake, but not from soil [10-11], because the root surface, acting as IHg barrier, could inhibit the translocation of IHg through the root system to the aboveground parts, but could not inhibit MeHg transfer [12]. Meanwhile, recently rice leaves and stalk burial is being encouraged all over the world as an economical and environmentally friendly method to get rid of rice residuals. However, the Hg-contaminated leaves may potentially cause Hg pollution in soil, resulting in the high MeHg concentrations in rice plants, which cause a vicious Hg pollution cycle in the rice paddy system. Therefore, a better understanding of different bioaccumulation of MeHg and IHg in rice plants and leaves could be critical in risk assessment of human mercury exposure.

To our knowledge, the Hg bioaccumulation in the rice plants is generally believed to be a dynamic process of uptake-translocation-accumulation [13]. However, the mechanism of Hg migration within the rice plant and the difference between the MeHg and IHg in the translocation mechanism was unclear. Besides, lots of research mainly focused on MeHg accumulation in the rice grain [14-15], given its potential MeHg exposure to humans. Limited studies have focused on Hg pollution in rice leaves. Since after harvest the rice leaves would be buried in soils in order to increase soil fertility in rice paddy fields [16]. Consequently, the Hg-contaminated rice leaves may potentially lead to Hg pollution in soil, further contaminating the rice plants in the second year. Therefore, studying MeHg and IHg accumulation in rice leaves was considered necessary.

In the present study, we analyzed farmland soils amended with IHg that were planted with rice plants as well as IHg and MeHg concentrations in rice leaves during the rice growing time to assess its bioaccumulation in leaves. Meanwhile, the $(NH_4)_2S_2O_3$ chemical extraction method was used in this experiment to evaluate the potential bioaccumulation of MeHg in rice grains [17]. The MeHg transfer capability from soil to rice plants was also investigated. Our main objectives were to investigate the bioaccumulation and migration of IHg and MeHg mechanisms in rice leaves and plants.

Materials and Methods

Soil samples were collected from Heilongjiang (HLJ), Sichuan (SC), Shanxi (SX), Hubei (HB), Jiangxi (JX), Henan (HN), Jiangsu (JS), and Guizhou (GZ) provinces in China, dried, ground, and sieved through a 150 μm mesh.

A total of 24 pots (three replicates for each treatment) in eight treatments (eight soils) were used. For each pot, 3 kg soil was amended with Hg (5 mg/kg, as $HgCl_2$ solution) at the start of the experiment (day 0, [18]). Soil samples were placed into pots (2.8 kg soil/pot) and equilibrated for 20 days under flooded conditions. Seeds of indica Wufengyou2168 (*indica* WFY2168) were germinated in soil for one month in a growth chamber prior to transplantation into pots (two seeds per pot). After the seeds were transplanted into soil, granulated fertilizer (0.32 $gkg^{-1}Ca(HPO_4)_2 \cdot H_2O$,0.18 g $kg^{-1}KCl$, and 0.21 g kg^{-1} $CO(NH_2)_2$) [19] was added as basal application, which was repeated on days 60 (panicle stage) and 90 (flowering stage) to ensure adequate nutrition for the growth of rice seedlings. And plants were grown in the greenhouse from 1 July to 24 October 2013 (115d) at ambient temperature (15-38ºC). The flooded condition was maintained with deionized water (3 cm above soil surface) during the entire rice growth period.

Soils and plant leaves were sampled at day 54 (i.e., panicle initiation stage), day 77 (i.e., heading stage), day 105 (i.e., milk-ripe stage) and day 132 (i.e., harvest stage) of the growing period, respectively. Soil samples were sampled in the soil surface (1-11 cm) and then collected into polypropylene centrifuge tubes, immediately vacuum-packed to remove oxygen *in situ*, and transferred to the laboratory in an ice box within 3 h. The leaf samples with a composition of two leaves per pot were cut at the bottom of the rice plant each time and then were cleaned first with tap water to remove the remaining soil, and cysteine solution (8 mm/L) were subsequently used to remove the mercury adsorbed on the surface of leaves [20]. After that, leaf samples were rinsed thoroughly in deionized water and freeze-dried. Then leaves were ground into fine powders by an IKA basic analytical mill (IKA A11, Germany) and used for mercury determination (described below).

The soil samples were centrifuged at 3,000 rpm for 30 min. to remove pore water and amended with $(NH_4)_2S_2O_3$ solution (0.0135 mol/L) at a 1:2 ratio, and then were rotated at 250 rpm per minute overnight in a shaker [17]. The mixtures were centrifuged at 3,000 rpm for 30 min. and the supernatants were filtered. HCl was added to the supernatant to obtain 0.5% HCl before determining MeHg [7]. All the process was employed in the anaerobic chamber in the dark, and the extractants were prepared with deoxygenated water to avoid soil oxidation. Meanwhile, the soil property including pH (HACH HQ30d) as well as particulate organic carbon (POC) levels (vario TOC cube, Elementar, Germany) in the soils were determined.

Total Hg was determined by a DMA-80 direct mercury analyzer (Milestone, Italy). MeHg concentrations were analyzed by an automatic Brooks Rand model III MeHg analyzer (CVAFS, Brooks Rand, USA) according to EPA method 1630. The soil, liquid, and plant samples were digested using KOH-methanol solution, and incubated in the shaker for four hours at 60°C for MeHg analysis [21-22]. The minimum detection levels for THg and MeHg is 0.2ng/g and 0.002ng/L, respectively. The recoveries of the standard samples for MeHg (ERM-cc58) and for THg (soil standard GSS-9 and mercury standard solutions)

were between 80-120% and 85-110%. IHg concentrations of leaves were calculated by subtracting the MeHg concentrations from the THg concentrations.

Background mercury levels in all chemicals were extremely low. Polypropylene centrifuge tubes were used in this study. And the tubes were considered relatively mercury-free because mercury levels in 2% nitric acid after rinsing the tubes were below detection limits. Quality control was assured by method/reagent blanks, matrix spikes, and certified reference materials as well as duplicate analysis. Any change of mercury extraction by $(NH_4)_2S_2O_3$, or mercury concentrations in soils and in leaves was tested using one-way analysis of variance (ANOVA).

Results and Discussion

Properties of Different Soils and Concentrations of MeHg in Soils

Background MeHg and THg levels in soil ranged from 0.025-0.132 ng/g and 27.8-130.7 ng/g, respectively. Compared to the spiked THg (5 mg/kg), the background Hg levels could be ignored. MeHg concentrations in various soils on four stages are shown in Fig. 1. Soil MeHg levels on panicle initiation stage, heading stage, milk-ripe stage, and harvest stage ranged from 12.67 to 75.87 ng/g, 8.03 to 91.37 ng/g, 8.15 to 14.24 ng/g, and 8.54 to 89.62 ng/g, respectively.

Most soils were weakly alkaline or near neutral (6.9-8.0), except JX soil (pH = 4.7). Clay contents (15.5-45.7%) and soil organic contents (0.3-4.3%) varied a lot in different soils (Table 1). The alteration of soil properties could have an important effect on MeHg production, which could cause the different concentrations of soil MeHg. For instance, soil organic matter could increase MeHg production [23] or inhibit the process of Hg methylation by binding strongly with IHg [24]. Also, it has been reported that clay could bind the IHg itself or form clay-OM to adsorb IHg [25], which could result in

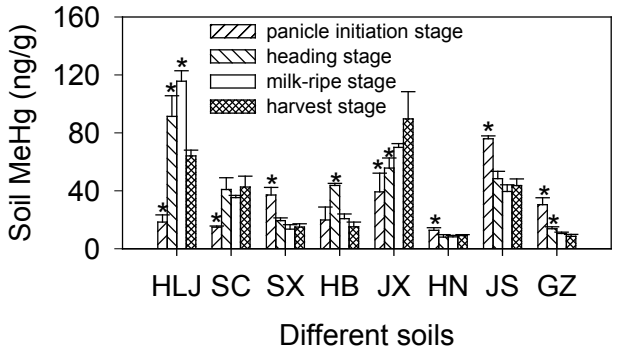

Fig. 1. MeHg concentrations in different soils at panicle initiation, heading, milk-ripe, and harvest stages. Mean ± SD (n = 3). Asterisk (*) indicates significant difference from harvest stage (p<0.05, one-way ANOVA).

Table 1. Soil properties and sampling inf ormation.

Soil	Sampling site	Sampling depth	pH	POC (%)	Clay (%)
HLJ	Heilongjiang Province	0-30 cm	7.5	2.0	37.6
SC	Sichuan Province	0-30 cm	7.8	1.6	41.9
SX	Shanxi Province	0-30 cm	8.0	0.7	41.4
HB	Hubei Province	0-30 cm	7.8	0.5	35.3
JX	Jiangxi Province	0-30 cm	4.7	0.3	15.5
HN	Henan Province	0-30 cm	7.7	0.9	30.2
JS	Jiangsu Province	0-30 cm	6.9	0.9	45.7
GZ	Guizhou Province	0-30 cm	7.9	4.3	41.3

the decrease of bioavailable IHg in soil, thus inhibiting the production of MeHg. Soil pH also plays a role in the process of Hg methylation [26]. For instance, low pH caused an increase of MeHg production [27]. Besides, the MeHg concentrations in soil were the common effect of the process of Hg methylation and MeHg demethylation. And soil properties including organic matter [28] and pH [29] could also alter the MeHg concentrations in soil by affecting the process of MeHg demethylation. We found that average soil MeHg concentrations were significant related to the soil properties by multiple regression analysis (averaged soil MeHg concentrations = f(pH, POC, clay), p = 0.006). Therefore, in this study properties variation of eight soils may affect variation in soil MeHg concentrations to different extents by affecting the process of Hg methylation and MeHg demethylation.

Meanwhile, the fluctuation of MeHg concentrations in four stages could be derived from the common effect of net production of MeHg in soil and the transportation of MeHg from soil to the rice plants. MeHg in soil was absorbed by roots and then transferred to the stem and leaf during the whole rice growth time [30]. This may imply that the difference of fluctuation of MeHg concentrations among eight soils was because the variation was controlled by the combined effects of soil properties and the MeHg migration capability of rice plants. And we suspected that rice growth was quite different in eight treatments since the biomass per pot of rice plant varied, ranging from 21.8 to 81.6g , which may lead to the change of MeHg migration capability of rice plants.

Bioaccumulation of Inorganic Mercury and MeHg in Rice Plants

IHg concentrations in leaves are shown in Fig. 2. IHg concentrations in leaves were quite different, but the IHg

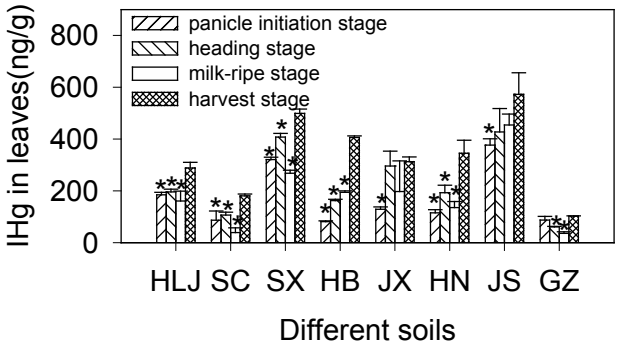

Fig. 2. IHg concentrations in rice leaves at panicle initiation, heading, milk-ripe, and harvest stages. Mean ± SD (n = 3). Asterisk (*) indicates significant difference from harvest stage (p<0.05, one-way ANOVA).

concentrations of leaves for all the treatments in harvest stage were the highest compared to that in other stages. The increase of IHg concentrations of leaves in this stage could be due to the fact that little or no IHg in rice leaves could be transferred to rice grains in the harvest stage. Similarly, Meng et al. found that accumulated IHg in aboveground parts of rice plants cannot be transported to seeds [12]. And the decrease of IHg concentrations in leaves in some stage could be mainly due to the emissions of IHg in leaves to the air through leaf pores or the translocation of IHg from leaves to the stalk during the rice-growing season [13].

MeHg concentrations and MeHg rates of leaves (%, MeHg concentrations in leaves / THg concentrations in leaves) are shown in Fig. 3. Large variations in MeHg concentrations (137.46-2,067.55 ng/g for panicle initiation stage, 85.51-666.84 ng/g for heading stage, 49.86-365.39 ng/g for milk-ripe stage, 15.33-117.91 ng/g for harvest stage), and MeHg rates (42.53-88.15% for panicle initiation stage, 30.36-60.25% for heading stage, 24.31-75.88% for milk-ripe stage, 7.08-20.55% for harvest stage) in leaves among different soils at different stages were observed. Generally speaking, MeHg concentrations and MeHg rates in leaves decreased (73.58 to 96.91% and 74.41 to 91.97%) with time for all soil, respectively. And the gradually decreasing MeHg concentrations and MeHg rates of leaves during the rice-growing season may indicate that most MeHg was translocated from leaves to the rice grains in the harvest stage.

In the harvest stage, the IHg and MeHg concentration in leaves ranged from 102.35 to 572.82 ng/g and 15.33 to 117.91 ng/g, respectively. After harvesting, rice straw and leaves were often buried into soils in order to increase soil organic matter in rice paddy fields [16]. Therefore, the mercury-contaminated rice leaves may result in the input of 'new mercury' into soils, leading to Hg pollution circulation in the rice paddy system.

MeHg concentrations extracted by $(NH_4)_2S_2O_3$ and MeHg extraction rates (%, MeHg concentrations extracted from soil / soil MeHg concentrations) are shown in Fig. 4. Significant relationships between rice grain MeHg levels and $(NH_4)_2S_2O_3$-extracted MeHg of soil have been reported, and $(NH_4)_2S_2O_3$ extraction may mimic MeHg uptake by roots and its accumulation in rice plants [17]. Therefore, MeHg concentrations extracted from soils were used to indicate potential

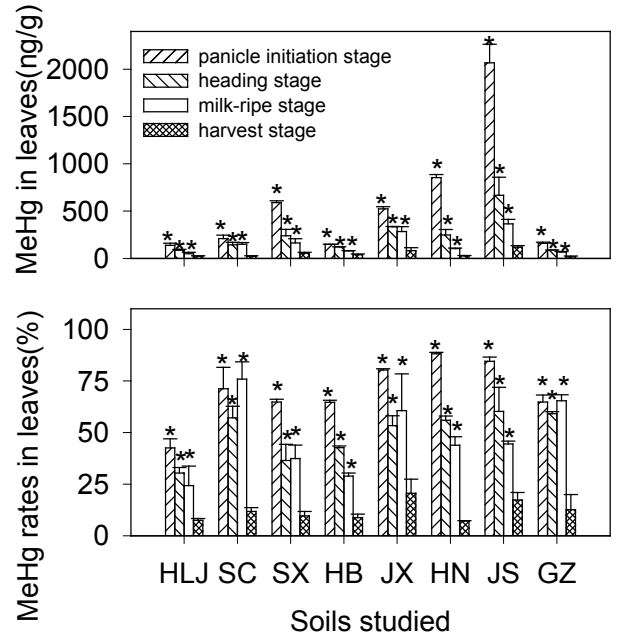

Fig. 3. MeHg concentrations in rice leaves at panicle initiation, heading, milk-ripe, and harvest stages (upper panel); MeHg rates in rice leaves at different stages (lower panel). Mean ± SD (n = 3). Asterisk (*) indicates significant difference from harvest stage (p<0.05, one-way ANOVA).

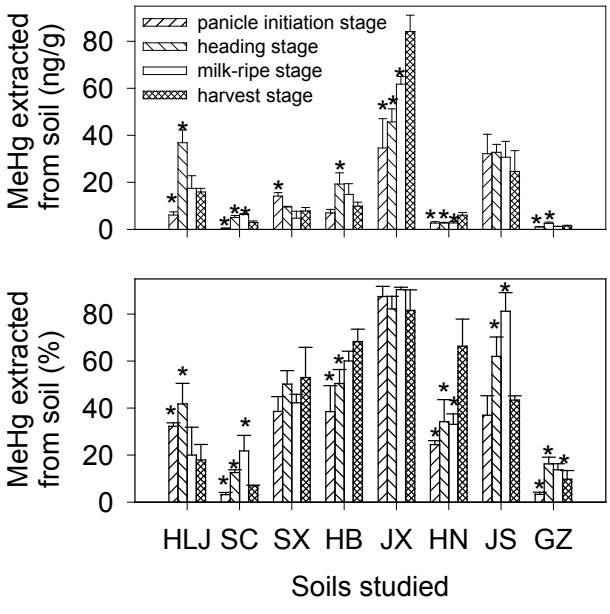

Fig.4. Concentrations of MeHg extracted by $(NH_4)_2S_2O_3$ from different soils (upper panel), as well as MeHg extraction rates (lower panel) at panicle initiation, heading, milk-ripe, and harvest stages. Mean ± SD (n = 3). Asterisk (*) indicates significant difference from harvest stage (p<0.05, one-way ANOVA).

MeHg bioaccumulation, and the MeHg extraction rate was used to indicate phytoavailable MeHg in this study. Unlike the MeHg concentrations in leaves, the MeHg concentrations extracted from soils and MeHg extraction rates by $(NH_4)_2S_2O_3$ fluctuated during the whole rice growth season, which may indicate the MeHg potential phytoavailable and bioaccumulation changing all the time. This could be mainly due to the variance of MeHg bioavailability in soil as affected by soil properties and rice plant physiology conditions, which varied in eight treatments. And the MeHg concentrations extracted from soils varied from the MeHg concentrations in leaves, suggesting that MeHg concentrations in leaves could not indicate the bioaccumulation of MeHg in rice grains. This could be attributed to the fact that the proportion of MeHg in leaves transported to the rice grain was different among different treatments.

Migration of MeHg in the Rice Plants

MeHg migration rates (%, MeHg concentration in leaves / MeHg concentration in soil) are shown in Fig. 5, which was used to indicate the MeHg migration capability of MeHg from soil to rice plants. Compared to the MeHg migration rates in panicle initiation stages, its migration rates in harvest stage significantly decreased (61.85-95.95%) for all the treatments, indicating that the translocation of MeHg from soil to rice plant continuously declined with the growth of rice plants. This may be because the bioavailability of MeHg generated from the IHg decreased with time. It was been documented that mobility and bioavailability of mercury may decrease with time, which has been mentioned as the aging effect [31]. The decrease in MeHg bioavailability may be attributed to the transfer of MeHg to stronger binding sites in soils [31]. We suspected that the decrease in MeHg migration rates in leaves may also be attributed to the translocation of MeHg to the rice grains in harvest time. It has been reported that most MeHg accumulates in the rice grain compared to other rice plant tissues [32-33]. And MeHg in soil was first absorbed by roots and then transferred to the stem and leaf, and finally translocated to the grain during the harvest period [34].

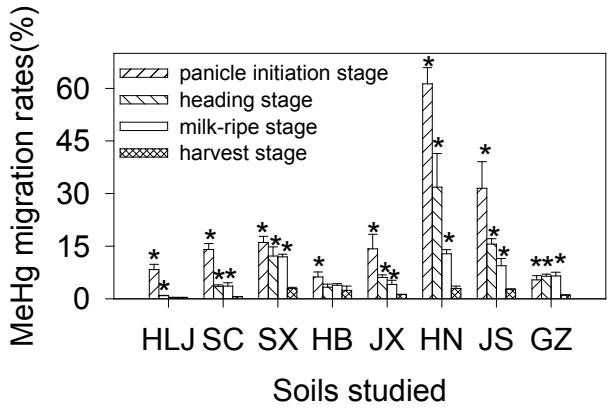

Fig. 5. MeHg migration rates at panicle initiation, heading, milk-ripe, and harvest stages. Mean ± SD (n=3). Asterisk (*) indicates significant difference from harvest stage (p<0.05, one-way ANOVA).

Conclusion and Implications

In conclusion, the fluctuation of MeHg concentrations in soil could be because MeHg production in soil was affect by the two processes (Hg methylation and MeHg demethylation), and Hg methylation and MeHg demethylation were dynamic during the whole rice growth season. Meanwhile, the translocation of MeHg from soil to the rice plants could also lead to the variation of MeHg concentrations in soil. Our study also reported that the limit or no IHg was migrated to the rice grains in harvest stage, since the IHg concentrations in leaves were the highest in the harvest stage compared to that in other stages.

Based on our results, MeHg concentrations in leaves could not be used to indicate MeHg bioaccumulation in rice grain, for there was a difference between the MeHg concentrations in leaves and the MeHg concentrations extracted from soils by $(NH_4)_2S_2O_3$, which could potentially simulate MeHg accumulation in rice plants. We suspected that this could be attributed to the different proportions of MeHg translocation from leaves to rice grains. More studies are needed to fully understand the mechanism of MeHg migration in the rice plants.

In this study, the decrease of MeHg transferred capability from soil to leaves in the harvest stage could be due to the lower bioavailability of MeHg in soil. Another explanation of the lower MeHg transferred capability to leaves could be the MeHg migration from leaves to rice grains, which could be due to the preferential partitioning of MeHg into rice grains.

In view of the scarcity of arable lands in China, large areas of mercury-contaminated lands are still being farmed, e.g., for rice cultivation. Therefore, it is of significance to study the MeHg and IHg bioaccumulation and migration mechanism in the rice plant. It should be noted that we preliminarily focused on the soil that was newly contaminated by mercury. Further studies are necessary on mercury contacting soil in the long term to investigate the mechanism of the difference of bioaccumulation and migration between MeHg and IHg.

Acknowledgements

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