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Incorporating rice residues into paddy soils affects methylmercury accumulation in rice

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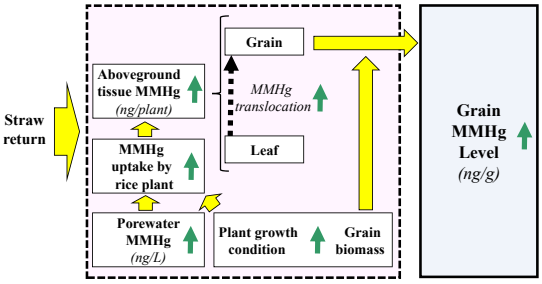
HIGHLIGHTS

- Incorporating rice residues into soils led to increased rice grain MMHg levels.
- Residue amendment may facilitate MMHg translocation to grain within rice plant.
- Increased grain biomass due to amendment would partly dilute MMHg in grain.
- Elevated porewater MMHg levels in amended soils may facilitate MMHg uptake by rice.
- Hg-organic interactions should be considered when planning straw return.

GRAPHICAL ABSTRACT



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ABSTRACT

Paddy fields are characterized by frequent organic input (e.g., fertilization and rice residue amendment), which may affect mercury biogeochemistry and bioaccumulation. To explore potential effects of rice residue amendment on methylmercury (MMHg) accumulation in rice, a mercury-contaminated paddy soil was amended with rice root (RR), rice straw (RS) or composted rice straw (CS), and planted with rice. Incorporating RS or CS increased grain MMHg concentration by 14% or 11%. The observed increases could be attributed to the elevated porewater MMHg levels and thus enhanced MMHg uptake by plants, as well as increased MMHg translocation to grain within plants. Our results indicated for the first time that rice residue amendment could significantly affect MMHg accumulation in rice grain, which should be considered in risk assessment of MMHg in contaminated areas.

1. Introduction

Recent research indicates that rice paddy field, a typical wetland environment, could favor methylmercury (MMHg) production and lead to high MMHg concentrations in soils (Meng et al., 2010). This could be partly attributed to the frequent organic input into paddy

Abbreviations: RR, rice root; RS, rice straw; CS, composted rice straw; MMHg, methylmercury; Hg_T, total mercury; IHg, inorganic mercury; SRB, sulfate-reducing bacteria; DOM, dissolved organic matter; DOC, dissolved organic carbon; WS, Wanshan; POC, particulate organic carbon; CK, the control; TF, translocation factor.

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soils (e.g., through fertilization, root exudation, and rice residue incorporation), which may facilitate growth of mercury methylation microbes (e.g., sulfate-reducing bacteria/SRB, Compeau and Bartha, 1985) and transformation of mercury into more bioavailable forms (e.g., Hg-S-DOM complexes, potentially available to mercury methylation bacteria, Graham et al., 2012; Zhang et al., 2012). And thus the potential effects of organic input on mercury methylation and MMHg bioaccumulation warrant investigation.

Rice residue (e.g., rice straw and root) amendment is a most common way of organic input into paddy soils, and may affect mercury methylation and MMHg bioaccumulation. For example, microbial decomposition of plant residues in paddy soils could result in high levels of dissolved organic matter (DOM, Cui et al., 2008; Yu et al., 2007). And elevated DOM levels may affect partitioning and mobility of MMHg, in view of the high affinity of mercury to organic matter (Yin et al., 1997; Zhong and Wang, 2008). Rice residue amendment could also affect plant physiology, e.g., growth (Eagle et al., 2000; Kaewpradit et al., 2009), which may consequently influence MMHg bioaccumulation.

However, to our knowledge, effects of rice residue amendment on MMHg production in paddy soils and its subsequent accumulation in rice were relatively unknown. A very recent study suggested that littering of plant residues on the surface of mercury-contaminated agricultural wetland and the succedent decomposition could possibly facilitate MMHg production in soils (Windham-Myers et al., 2014a, 2014b). This study provided initial evidence on the possible effects of rice residue amendment on mercury methylation in soils.

To explore the potential effects of rice residue amendment on MMHg production and accumulation in rice plants, three typical types of rice residues (rice root, rice straw, and commercially available composted rice straw), which are commonly incorporated into farming soils in China, were amended into a mercury-contaminated paddy soil. Concentrations of MMHg in soils, porewater and aboveground tissues of rice plant (i.e., straw, leaf and grain) were determined. Consequently, changes in grain MMHg levels after rice residue amendment were explained by changes in MMHg behaviors in soils and plant physiology.

2. Materials and methods

2.1. Soil, rice residues, chemicals and containers

Topsoil (0–20 cm) used in this study was collected from a drained paddy field in Wanshan mercury mining area, Guizhou Province, China (referred to as WS soil). The soil was air-dried, ground and sieved (150 μm mesh, the same below). Total mercury or MMHg concentration was 47.63 μg/g or 3.58 ng/g. Properties of WS soil were shown in Supplementary data, Table S1.

In order to minimize MMHg input into soil, which was not considered in this study, both rice root (RR) and rice straw (RS) were collected from a control site (Guiyang, Guizhou Province, China) free of any known mercury contamination. After washing, RR and RS were cut to approximately 1 cm pieces, soaked in deionized water (to facilitate their subsequent decomposition in soils, Pang et al., 2013; Yu et al., 2012) and air-dried. Commercially available composted rice straw (CS, as powders), which is being widely used as an organic fertilizer in China, was acquired from Jiang Yuan Agricultural Science & Technology Development Co., Ltd, China. Incorporating powders instead of intact rice residues is being popularized in China to facilitate decomposition of residues (Yang et al., 2012). Therefore, fine powders of residues (by grinding and sieving) were used, to ensure homogeneity of amended soils and facilitate decomposition. Particulate organic carbon (POC) and MMHg levels in residue powders were described in Supplementary data, Table S2.

Chemicals and containers used in this study were described in Supplementary data, text.

2.2. Setup of pot experiments

A total of 12 pots (3 replicates for each treatment) in 4 treatments (i.e., the control/CK, CS, RR and RS amended) were used. For each pot, 2.5 kg of WS soil was mixed with one type of rice residue (50.4 g CS, 29.0 g RR or 36.2 g RS), resulting in increases of POC levels in soils (i.e., 0.4%, comparable with those reported in straw amended soils, e.g., Yu et al., 2007; Ji et al., 2012). The consistent increase in POC levels under rice residue amendment would facilitate comparing effects of various types of rice residues on MMHg dynamics. Meanwhile, the application rate was comparable with that used in farming activities (Ji et al., 2012). Rice residue amendment resulted in an increase of less than 0.2% of MMHg in soils, and thus the changes could be ignored. Soil in each pot was added with basal fertilizers (167 mg N kg^{-1} soil as urea, 79 mg P kg^{-1} and 150 mg K kg^{-1} soil as KH_2PO_4 and K_2SO_4). On Day 0 (i.e., Jul 1st, 2013, 2 d after rice residue amendment), thirty-day-old rice seedlings of *indica* Eryou084 were transplanted into soil (two seedlings per pot), and all pots were placed in a greenhouse (Nanjing, Jiangsu Province, China) at ambient temperature (15–38 °C). During the entire rice growth period, soil was flooded with deionized water (3 cm above soil surface).

2.3. Sampling

Soil or porewater MMHg concentrations, mercury (mainly inorganic mercury or IHg) geochemical fractionation in soils (quantifying organo-complexed mercury levels in soils, Supplementary data, text), as well as MMHg levels in leaf were quantified through the rice growth period. On Day 4, Day 25 (tillering and jointing stage), Day 57 (heading and flowering period), Day 78 (filling stage) and Day 106 (mature stage), surface soils (1–11 cm, ~5 g, wet weight) were sampled into polypropylene centrifuge tubes, immediately vacuum-packed and transferred to the laboratory in an ice box within 3 h. To acquire soil porewater, soil samples were unpacked in an anaerobic glove box filled with nitrogen gas, capped, and then centrifuged at 4000 rpm for 20 min (Bufflap and Allen, 1995). After that, the tubes were transferred into the anaerobic glove box again, and the supernatants were filtered and stored at −20 °C (added with 0.2% HCl and 0.5% HAc) before determination of porewater MMHg levels. The remaining soils were then used for soil moisture and MMHg analysis (details below). On Day 25, Day 57, Day 78 and Day 106, two pieces of oldest leaves (flag leaf on Day 106) were collected from each plant following the same procedure. Leave cutting could have potential impacts on plant growth and grain biomass, while all treatments were subject to the same impacts (if any). On Day 109, the aboveground tissues of rice plants, i.e., grain and straw (including both stem and leaf), were harvested, washed by deionized water, oven-dried (40 °C) and weighed. According to our preliminary experiment, the drying process had minor effects on quantified MMHg concentrations. Rice straw or hulled grain (i.e. brown rice) were then ground into fine powders by an IKA basic analytical mill (IKA A11, Germany) and used for MMHg determination (described below). Schedule of all activities was available in Supplementary data, Table S3.

2.4. Analytical methods

Concentrations of Hg_T were analyzed by a DMA-80 direct mercury analyzer (Milestone, Italy). And MMHg concentrations were determined by an automatic Brooks Rand model III MMHg analyzer