

Main Controlling Factors and Empirical Models for Total Mercury and Methylmercury Accumulation in Rice (*Oryza Sativa* L.)

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Abstract

It has been reported that rice consumption is the main mercury (Hg) exposure pathway for humans, and soil properties could significantly affect the methylation and accumulation process of Hg in soil-rice system. In this study, 19 paddy soil with various properties were spiked with exogenous Hg(II) at three concentration levels to conduct a pot experiment after a 60-days aging period. Stepwise multiple linear regression was applied to determine the controlling soil factors and develop prediction models for the methylmercury (MeHg) concentration in soil and total Hg (THg) and MeHg in brown rice. The results showed that THg in brown rice was positively correlated with THg in soil, while it had a negative correlation with soil pH and clay content. Soil organic matter (OM) promoted Hg methylation and inhibited the accumulation of MeHg in brown rice. Soil pH and clay content were negative factors when soil MeHg was used to predict MeHg in brown rice. THg and MeHg in brown rice could be well predicted by soil THg, pH and clay content. These results may provide a theoretical basis for safe production of rice in Hg contaminated paddy field.

1. Introduction

Mercury (Hg) is a worldwide pollutant with various forms, and the most toxic form is methylmercury (MeHg) (Clarkson. 1998; Obrist et al. 2018). Rice consumption is reported to be the main Hg exposure pathway for humans in inland China (Zhang et al. 2010a). In Hg contaminated paddy soils, the production of MeHg in soil could result in high MeHg in rice grains, which could harm human health (Qiu et al. 2008). The source of MeHg in rice is mainly from soil (Meng et al. 2010), indicating that the Hg methylation process in soil should be studied. Zhang et al. (2010b) suggested that the bioaccumulation factor (BCF) of MeHg is 800 times higher than that of inorganic mercury (IHg), which means that rice can accumulate high levels of MeHg. Brown rice has the highest ability to accumulate MeHg compared to other rice tissues (Meng et al. 2010; Khanam et al. 2020). Taking current scenario into account, the Hg methylation process in soil and total mercury (THg) and MeHg accumulation processes in rice should be concerned.

The soil properties could influence the Hg methylation process in soil and THg and MeHg accumulation in rice plants (Zhao et al. 2020). It was reported that soil pH could increase the mobility and bioavailability of Hg in soil (Yin et al. 1996; Rózański et al. 2016). Meng et al (2016) reported that organic matter (OM) could promote the Hg methylation process in sediment in a hydroelectric reservoir, while an investigation by Yin et al. (2018) indicated that the OM content in soil had a negative relationship with MeHg in soil and rice grains in a mining area. Clay in soil could decrease the bioavailability of Hg because of the presence of higher exchangeable sites and thereby higher adsorption capacities (Malandrino et al. 2011; Wang et al. 2020a). The addition of sulfate could inhibit net MeHg production (Wang et al. 2016b). However, these studies focused on only one or two soil factors and ignored the interaction between soil properties. The major controlling factors to be identified are not representative because of limited geographic area and soil types.

Prediction models of the THg concentration in carrots have been developed previously (Ding et al. 2014). The Hg risk in paddy soil is higher than that in drylands because anaerobic conditions can promote the Hg methylation process (DeLaune et al. 2004). So far, the main soil controlling factors and the prediction models of THg and MeHg in brown rice have not been systematically studied and developed. Therefore, a study of prediction models of THg and MeHg accumulation in rice is necessary to provide theoretical support for the risk assessment of Hg in paddy soil.

The goals in this study are to (i) determine the main factors controlling Hg methylation and accumulation processes in soil-rice system, and (ii) build empirical models for THg and MeHg prediction in brown rice. The results of this experiment would provide data and theoretical basis for the safe production of rice in Hg contaminated paddy fields.

2. Materials And Methods

2.1 Soil collection

Nineteen paddy soils were sampled from the main rice producing areas in China. The soil samples were collected from the surface soil layer (0-20 cm topsoil) of uncontaminated farmland. The soils were air-dried at room temperature, and then passed through a 2 mm sieve. The soil pH (1:2.5 soil to water ratio), organic matter (OM, $K_2CrO_4:H_2SO_4$ Oil-Bath-Heating), clay content (pipette method), cation exchange capacity (CEC, 1 M ammonium acetate leaching method at pH 7.0), sulfate content (1:10 soil to water ratio, measured by ion chromatography) and free $FeO_x/ MnO_x/ AlO_x$ (dithionite-citrate-bicarbonate: DCB method) are measured according to Lu (2000). The results are displayed in Table S1.

2.2 Experimental design

The soil samples (6 kg) of each soil type were placed in separate plastic pots, and IHg (inorganic Hg, indicating $HgCl_2$ solution in this study) was added at four levels (0, 1, 2, 4 mg/kg). These treatments were named as CK, Hg1, Hg2 and Hg3. Each treatment had three replications. After a 60-day aging process under flooding conditions, the soils of all pots were sampled for the determination of THg and MeHg in the soil. These soil samples were stored in a $-20^\circ C$ refrigerator. The soil samples were freeze-dried and passed through a 0.149mm sieve for THg and MeHg analysis.

The rice cultivar in this study is Huiliangyou 996 (hybrid indica), which is widely planted in China. The rice seeds were pre-grown in a seeding tray for 20 days. Next, the rice seedlings were transplanted into the prepared pots after the soil aging process. The flooding conditions lasted during the whole rice growth period. Seeds from mature rice plants were harvested, washed with deionized water, and then freeze-dried at $-50^\circ C$. After that, rice grains were shelled, and the resulting brown rice was milled into powder for THg and MeHg determination.

The determination of THg in soil and brown rice was performed according to the following method: The soil samples were digested with (1+1) aqua regia in $100^\circ C$ water bath heating (Meng et al. 2010), while

brown rice was digested with HNO₃: H₂O₂ (4:3) by using the high pressure vessels according to Determination of total mercury and organic mercury in foods (GB5009.17-2014) (Ren et al. 2014; Ding et al. 2014). The digestion solutions were then measured by cold vapor atomic fluorescence spectrometer (CVAFS, AF-610D2, Beijing Rayleigh Analytical Instrument, China).

For MeHg determination, soil and brown rice samples were digested by 25% KOH/methanol (w/w) and kept in an incubation shaker at 60°C for 4 h (Bloom. 1989; Liang et al. 1994), and measured by cold vapor atomic fluorescence spectrometry (Brooks Rand, USA) according to the USEPA method 1630.

For QA and QC, blanks were excluded and standard materials (GBW07407 for total Hg in soil, GBW10049 for total Hg in brown rice, ERM-cc580 for MeHg in soil and TORT-2 for MeHg in brown rice) were included in the analysis. Analysis of these materials yielded total Hg concentrations of 780±66 µg/kg, 12.8±2.49 µg/kg, 71.2±4.07 µg/kg and 322±53 µg/kg, respectively, which were similar with the certified values of 740±110 µg/kg, 12.0±2.3 µg/kg, 75.5±4.0 µg/kg and 355±56 µg/kg. The LOD (limit of detection) and LOQ (limit of quantitation) of CVAFS for total Hg determination and GC-CVAFS for MeHg determination were 2 µg/kg and 0.2 pg respectively.

2.3 Data analysis

The maximum allowable limit of THg in brown rice is 20 µg/kg according to the National Food Safety Standard of China (GB 2762-2017). The maximum allowable limit of MeHg in rice grain has not been set by any countries or international organizations. The United States Environmental Protection Agency (USEPA) recommended 0.1 µg/kg/d as the probable daily intake (PDI) of MeHg from rice consumption. Equation 1 shows the correlation between the PDI and MeHg concentration in rice (Li et al. 2015):

$$PDI = \frac{C \times IR}{BW}$$

1

where the PDI is given in µg/kg/day, C is the MeHg concentration in rice (µg/kg), IR is the daily intake rate of rice (kg/d), and BW is the average weight of the exposed population (kg). The maximum allowable limit of MeHg in brown rice can be calculated by Equation 2:

$$C = \frac{PDI \times BW}{IR}$$

2

The BW and IR of adult (Duan et al. 2013), children aged 6-17 years old (Zhao et al. 2016), and children aged 0-5 years old (Wang et al. 2016a) were collected, and the maximum allowable limits of MeHg in brown rice were calculated. Accordingly, the strictest dose limit of MeHg in rice was 16.3 µg/kg (Table S2).

The data in this study were collected and calculated in Excel, and the figures were drawn by SigmaPlot 10.0. The prediction models in this study were built by linear stepwise regression using SPSS 19.0. In the model building process, more parameters could increase the accuracy and likelihood function of the models, which would result overfitting phenomenon. Therefore, the Akaike information criterion (AIC) and Bayesian information criterion (BIC) were used in this study. The calculated equations applied for calculations are shown in Equations 3 and 4:

$$AIC = 2k - 2\ln(L)$$

3

where k is the number of parameters, and L is the likelihood function. The model with the minimum AIC was chosen as the most suitable one.

$$BIC = k\ln(n) - \ln(L)$$

4

where k is the number of parameters, L is likelihood function. The model with minimum BIC was chosen as the most precise model. The BIC penalized more model parameters than the AIC, which led the BIC to prefer to choose simple models with fewer parameters. The column charts and linear regression equations were generated using SigmaPlot 10.0 and SPSS 19.0 respectively.

3. Results

3.1 Total Hg and methylmercury in soil and brown rice

The concentrations of THg and MeHg in soil (THg_{soil} and $MeHg_{soil}$) are displayed in Fig. 1. The MeHg concentration in the soil ranged from 0.501 to 114 $\mu\text{g}/\text{kg}$, with a mean of 24.3 $\mu\text{g}/\text{kg}$. The MeHg proportion of the THg in soil ranged from 0.272–3.47%, with an average of 1.41%. The THg_{soil} of all soils in the same Hg addition treatment were similar, while $MeHg_{soil}$ showed great differences. This phenomenon demonstrated that soil properties could significantly influence MeHg production.

The concentrations of THg and MeHg in brown rice (THg_{rice} and $MeHg_{rice}$) are shown in Fig. 2. In the CK treatment, the THg_{rice} and $MeHg_{rice}$ of these soils were under the maximum allowable limits for rice (ranging from 2.30 to 54.6 $\mu\text{g}/\text{kg}$, and 1.58 to 43.5 $\mu\text{g}/\text{kg}$, respectively), except for S2, S3, and S10. The THg and MeHg in brown rice of all soils in the Hg addition treatments (including Hg1, Hg2, and Hg3) exceeded the limits for brown rice (ranging from 113 to 1933 $\mu\text{g}/\text{kg}$, and 85.6 to 1622 $\mu\text{g}/\text{kg}$, respectively). The MeHg proportion of the THg in rice ranged from 50.5–95.2%, with a mean of 81.0%.

3.2 Main controlling factors and prediction models of mercury methylation and accumulation

3.2.1 The controlling factors and prediction models of Hg methylation process in soil

Scatter diagrams between THg_{soil} and $\text{MeHg}_{\text{soil}}$ are shown in Fig. S1(A) and exhibit positive relationships between THg_{soil} and $\text{MeHg}_{\text{soil}}$. The controlling soil factors of the Hg methylation process in paddy soil were studied and the prediction models of $\text{MeHg}_{\text{soil}}$ were calculated, as shown in Table 1.

Table 1
The prediction models of $\text{MeHg}_{\text{soil}}$ (n=76)

No.	Linear stepwise regression equation	R ²	p	AIC	BIC
1	$\log\text{MeHg}_{\text{soil}} = -1.93 + 1.00\log\text{THg}_{\text{soil}}$	0.861	<0.0001	11.0	18.0
2	$\log\text{MeHg}_{\text{soil}} = -2.58 + 0.999\log\text{THg}_{\text{soil}} + 0.437\log\text{OM}$	0.879	<0.0001	2.60	11.9

From the equations in Table 1, the soil OM content was nominated as an important factor among the soil properties. Except for THg_{soil} , the soils with a higher OM content showed a positive impact on the Hg methylation process. According to the AIC and BIC values, Equation 2 in Table 1 was a more precise model to predict $\text{MeHg}_{\text{soil}}$. The measured $\log\text{MeHg}_{\text{soil}}$ and predicted values were plotted to show the accuracy of the prediction models (Fig. 3). In Fig. 3, the coefficients of the regressions in the following figure were close to 1, which means that the prediction models are reliable. The coefficient of determination (R²) in Fig. 3(B) was higher than that in Fig. 3(A), and the root mean square error (RMSE) in Fig. 3(B) was lower than that in Fig. 3(A), which clearly indicated that Equation (2) in Table 1 was more accurate than Equation 1.

3.2.2 Controlling factors of the Hg accumulation process in rice and prediction models

During the Hg (including THg and MeHg) accumulation process in rice, soil properties could affect the Hg uptake process in rice by changing the bioavailability of Hg in soil. The prediction models of THg and MeHg in brown rice are displayed in Tables 2 and 3, respectively.

Table 2
The prediction models of THg_{rice} (n=76)

No.	Linear stepwise regression equation	R ²	p	AIC	BIC
1	$\log\text{THg}_{\text{rice}} = -1.12 + 1.14\log\text{THg}_{\text{soil}}$	0.851	<0.0001	36.0	43.0
2	$\log\text{THg}_{\text{rice}} = -0.419 + 1.14\log\text{THg}_{\text{soil}} - 0.122\text{pH}$	0.877	<0.0001	23.8	33.2
3	$\log\text{THg}_{\text{rice}} = 0.674 + 1.14\log\text{THg}_{\text{soil}} - 0.155\text{pH} - 0.697\log\text{clay}$	0.886	<0.0001	19.9	31.5

Table 3
The prediction models of MeHg_{rice} (n=76)

No.	Linear stepwise regression equation	R ²	p	AIC	BIC
1	$\log\text{MeHg}_{\text{rice}}=0.987+1.12\log\text{MeHg}_{\text{soil}}$	0.880	<0.0001	26.3	33.2
2	$\log\text{MeHg}_{\text{rice}}=1.99+1.15\log\text{MeHg}_{\text{soil}}-0.687\log\text{OM}$	0.910	<0.0001	6.29	15.6
3	$\log\text{MeHg}_{\text{rice}}=2.53+1.14\log\text{MeHg}_{\text{soil}}-0.716\log\text{OM}-0.084\text{pH}$	0.921	<0.0001	-1.66	9.99
4	$\log\text{MeHg}_{\text{rice}}=-1.37+1.19\log\text{THg}_{\text{soil}}$	0.852	<0.0001	41.9	48.9
5	$\log\text{MeHg}_{\text{rice}}=-0.603+1.19\log\text{THg}_{\text{soil}}-0.133\text{pH}$	0.880	<0.0001	27.9	37.3
6	$\log\text{MeHg}_{\text{rice}}=0.472+1.19\log\text{THg}_{\text{soil}}-0.166\text{pH}-0.686\log\text{clay}$	0.888	<0.0001	24.6	36.2

The positive relationship between THg_{soil} and THg_{rice} was shown in Fig S1(B). Besides that, according to the prediction models in Table 2, it was clear that soil pH and clay content appeared to be the main controlling factors in the prediction models of THg_{rice}. Lower pH and clay content could promote the THg accumulation process. The AIC and BIC values indicated that equation 3 in Table 2 was more accurate than other models. Linear graphs between the predicted values and measured of THg_{rice} were plotted and presented in Fig. 4. The coefficient determination (R²) values were increased and the root mean square error (RMSE) were decreased gradually from Fig. 4(A) to 4(C), which means that equation 3 in Table 2 is more accurate than equations 1 and 2.

The MeHg_{rice} was mainly originated from MeHg_{soil}. The MeHg_{soil} in this experiment mainly originated from THg_{soil}. Scatter diagrams between THg_{soil} and MeHg_{rice} are shown in Fig S1(D), which exhibit significant correlation between THg_{soil} and MeHg_{rice}. Hence, THg_{soil} was used to build the prediction models for MeHg_{rice}. The prediction models of MeHg_{rice} predicted by MeHg_{soil} and THg_{soil} are displayed in Table 3. From Equations 1 to 3 in Table 3, the soil with high MeHg_{soil}, low OM content and low soil pH was beneficial for MeHg uptake by rice. The AIC and BIC values in these three prediction models indicated that Equation 3 in Table 3 was more precise than Equations 1 and 2. Similarly, MeHg_{rice} can also be predicted by THg_{soil}, pH and clay content, and Equation 6 was suggested to be the most accurate model.

Meanwhile, the linear graphs between the measured log MeHg_{rice} and predicted log MeHg_{rice} predicted by MeHg_{soil} and THg_{soil} were drawn in Fig. 5 and Fig. 6, respectively. The coefficient of determination (R²) values in Fig. 5 were higher than those in Fig. 6, whereas the root mean square error (RMSE) in Fig. 5 was lower than that in Fig. 6, which suggested that the prediction accuracies of the prediction models in Fig. 5 were greater than those in Fig. 6. Considering that, the determination of THg in soil is simpler and more convenient than that of MeHg, Equation 6 in Table 3 can be more widely referenced than Equation 3.

4. Discussion

In this study, the prediction models for THg_{rice} and $\text{MeHg}_{\text{rice}}$ in Table 2 and 3, the soil pH, OM content and clay content were considered as the main soil factors controlling the Hg methylation and accumulation process. Soil with low pH could promote Hg methylation in soil and Hg accumulation in brown rice because soil with low pH could release more H^+ ion, which would increase the dissolution and bioavailability of Hg (Fairbrother et al. 2007; Huang et al. 2018). Therefore, the activation of Hg was higher in a soil with lower pH, and Hg in such a soil would be more available for microbes performing methylation (Golding et al. 2008; Pestana et al. 2019).

The effect of the OM content was controversial in Hg methylation and accumulation processes in soil-rice system. On the one hand, OM could enhance environmental Hg risks by promoting the Hg methylation process in paddy soil (Rothenberg et al. 2014; Windham-Myers et al. 2014; Zhu et al. 2015); on the other hand, OM could bind Hg to decrease Hg bioavailability in soil (Lin et al. 2010; Skyllberg et al. 2006). In our experiment, the influence of the OM content could be divided into two processes: Hg methylation in soil and Hg accumulation in rice. In the Hg methylation process, OM was positively related to the production of MeHg in paddy soil. The OM in soil could not only provide electrons, energy and nutrition for microbes performing methylation (Liu et al. 2014; Marvin-DiPasquale et al. 2014; Rothenberg and Feng. 2012), but also be beneficial for Hg microbial bioavailability in soil (Liem-Nguyen et al. 2017). Moreover, the OM content had a negative correlation with the MeHg accumulation process in brown rice. The abundant reduced sulfur sites on OM molecules provided strong binding sites for Hg, thus reducing Hg (including THg and MeHg) bioavailability (Hammerschmidt et al. 2008). Biochar amendment also reduced MeHg accumulation in rice (Shu et al. 2016). The OM content could facilitate the Hg methylation process in soil, but it could decrease MeHg accumulation in rice at a given $\text{MeHg}_{\text{soil}}$. The exact mechanism by which OM affects soil-rice systems is yet to be defined. Further studies should be conducted on the effects of OM in Hg methylation and accumulation processes in soil-rice systems.

The clay content in soil appeared to be a negative factor for THg and MeHg accumulation process when THg_{soil} was used in the prediction models. Because clay has a strong adsorption capacity and high cation exchange with heavy metals (Malandrino et al. 2011). Shi et al. (2019) described that higher clay content would reduce the available Hg in soil, because the migration rate of available Hg was in the order of sandy loam > loam > clay loam. In addition, higher clay content in soil correspond to the higher alkyl carbon, which was the main stable OM in soil (Mahieu et al. 1999; Li et al. 2019b).

This experiment was conducted in a pot experiment with exogenous Hg addition and flooding conditions lasted for the whole rice growth period, which was quite different than the actual field production. It has been reported that the methylation of exogenous Hg in paddy soils reached equilibrium after 30 days aging under the flooding conditions (Du et al. 2019), however, the bioavailability of Hg in the long-term contaminated field soil could be different from that in artificially spiked soil. Compared with persistent Hg in soil, newly deposited Hg more easily transforms to MeHg and accumulates in rice (Meng et al. 2011). In addition, flooding is an effective way to reduce cadmium (Cd) in brown rice (Arao et al. 2009; Liu et al. 2017). In contrast to Cd, Peng et al. (2012) found that the reduction rate of THg and MeHg in rice grain was enhanced up to 63.7% and 83.0% under the flooded-aerobic treatment than the flooded treatment,

which means that the flooding condition would increase the Hg (THg and MeHg) concentration in rice. Therefore, the prediction models in this study need to be further validated and improved under the field conditions.

5. Conclusion

In our study, the main soil controlling factors for Hg methylation in soil and accumulation in rice were identified and prediction models were nominated. The $\text{MeHg}_{\text{soil}}$ was positively related with THg_{soil} and OM content. For THg uptake in rice, soil pH and clay content appeared to be negative controlling factors. Soil pH and OM content were inversely correlated to MeHg in brown rice. This result indicated that the OM content promoted Hg methylation in paddy soil while inhibiting MeHg uptake in brown rice. Reliable empirical prediction models for THg and MeHg in brown rice were developed in incorporating soil THg concentration, pH and clay content. The obtained results could provide a theoretical basis for Hg risk assessment and scientific guide for the safe production of rice in Hg-contaminated paddy soil.

Declarations

Author contribution Shuyang Du: conceptualization, methodology, writing-review & editing. Xingxiang Wang: methodology, validation, formal analysis. Zhigao Zhou: methodology, data curation, writing-review & editing. Taolin Zhang: validation, supervision. Muhammad Kamran: resources, writing-review & editing. Changfeng Ding: writing- review & editing, supervision, project administration

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Availability of data and materials The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Ethical Approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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Figures

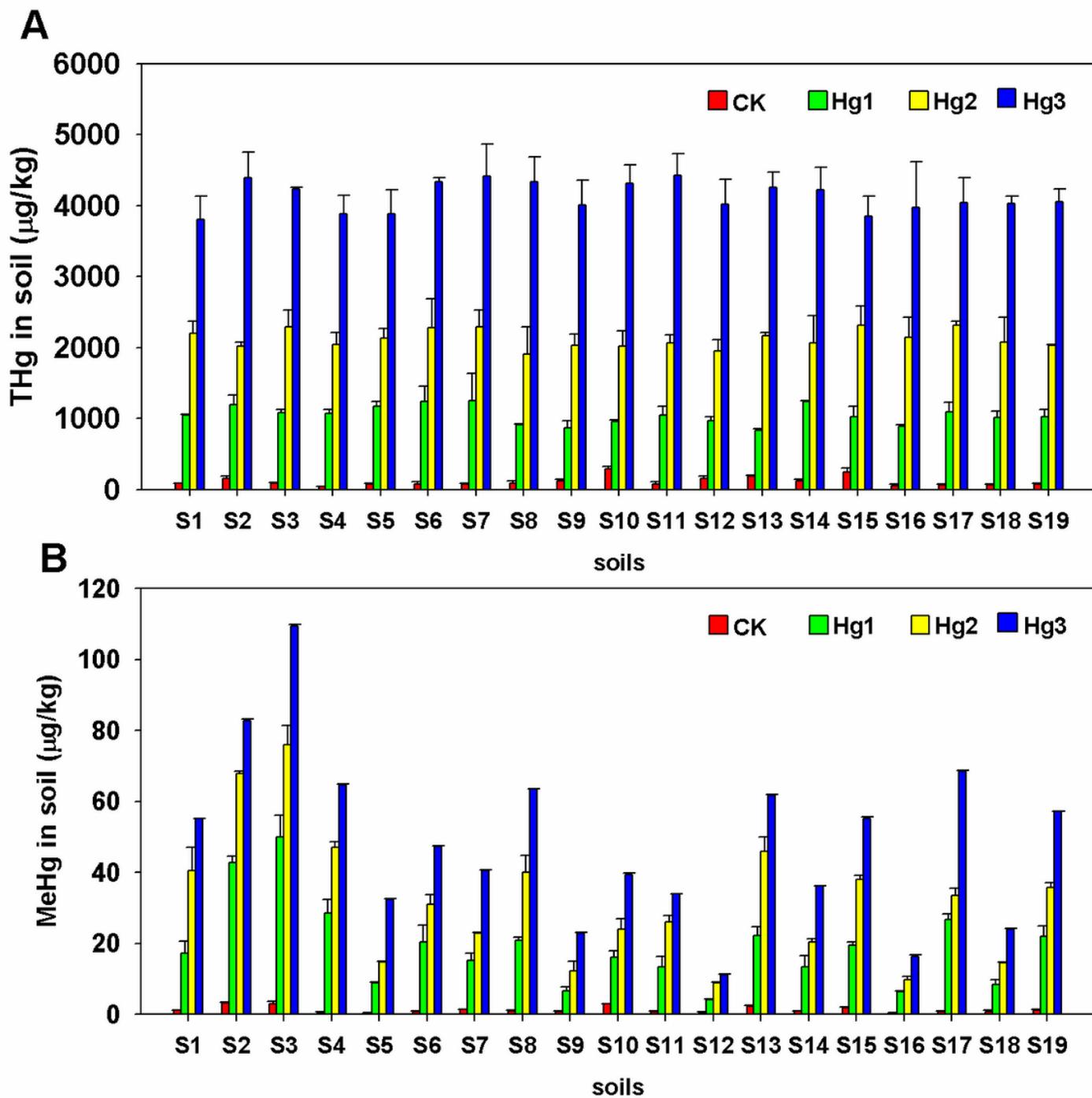


Figure 1

The THg (A) and MeHg (B) concentration in soils, CK means no Hg-added treatment, Hg1, Hg2 and Hg3 means Hg addition gradients 1, 2, 4 mg/kg.

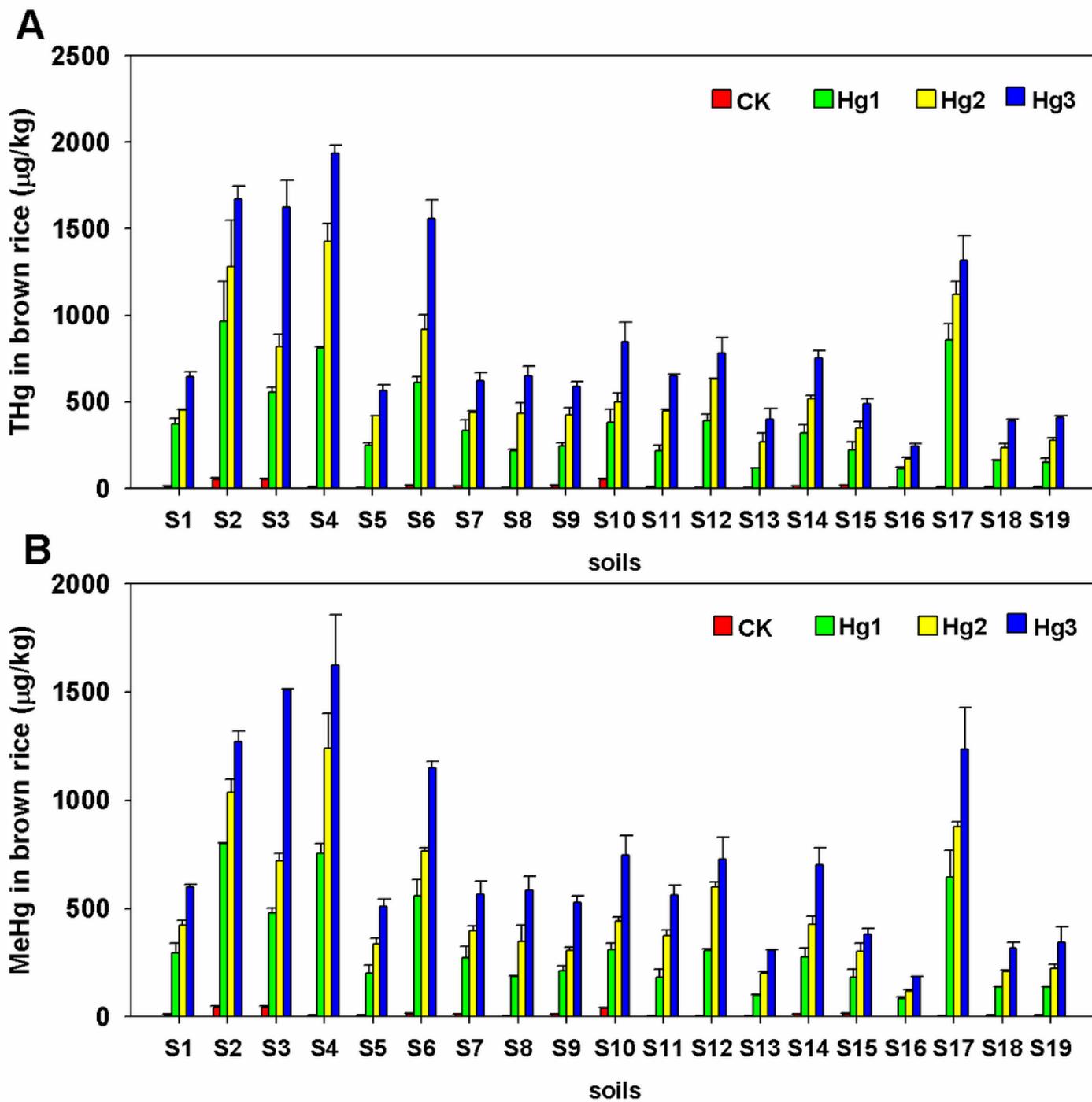


Figure 2

The THg (A) and MeHg (B) concentration in brown rice, CK means no Hg-added treatment, Hg1, Hg2 and Hg3 means Hg addition gradients 1, 2, 4 mg/kg.

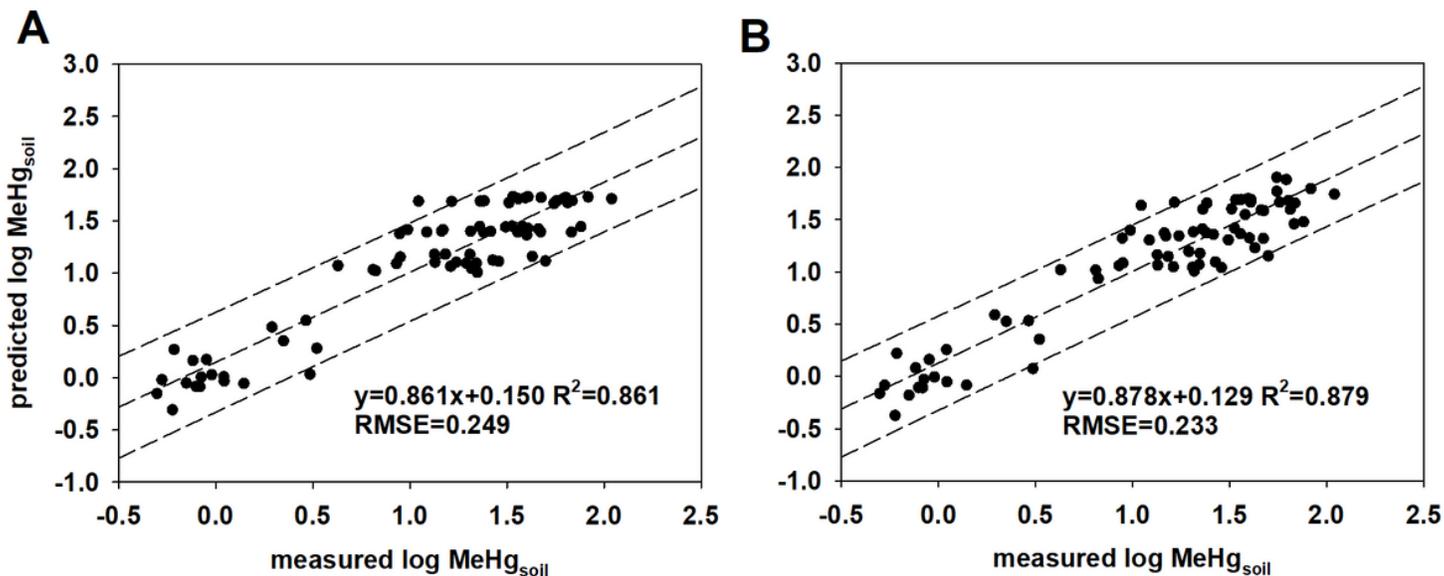


Figure 3

The linear graphs between measured log MeHg_{soil} and predicted log MeHg_{soil}, figure A represents the linear graph between measured log MeHg_{soil} and predicted log MeHg_{soil} from eq (1) in table 1, figure B represents eq (2) in table 1.

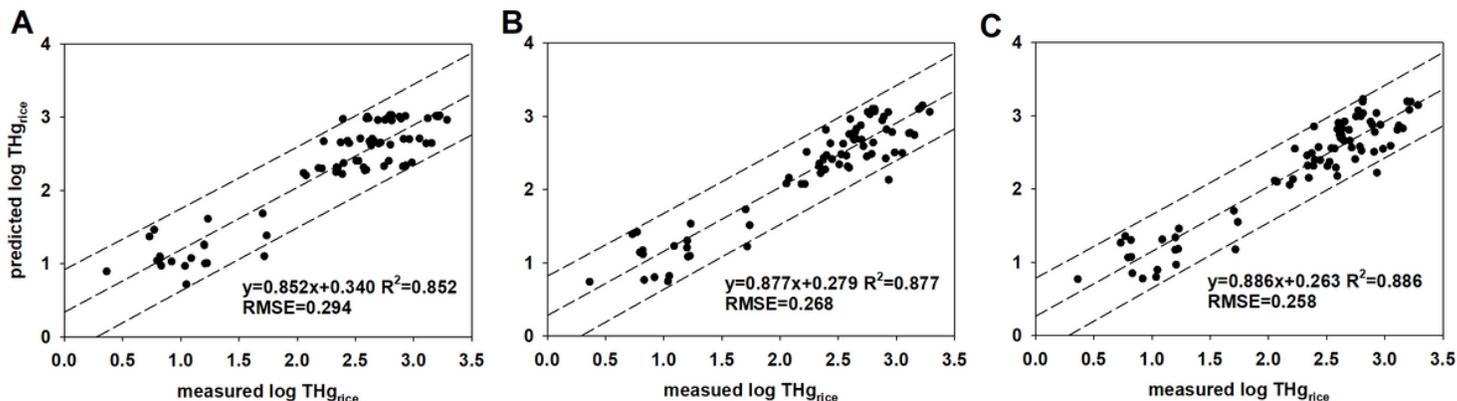


Figure 4

The linear graphs between predicted log THg_{rice} and measured log THg_{rice}, figure A represents the linear graph between measured log THg_{rice} and predicted log THg_{rice} from eq (1) in table 2, and figure B represents eq (2) in table 2, figure C represents eq (3) in table 2.

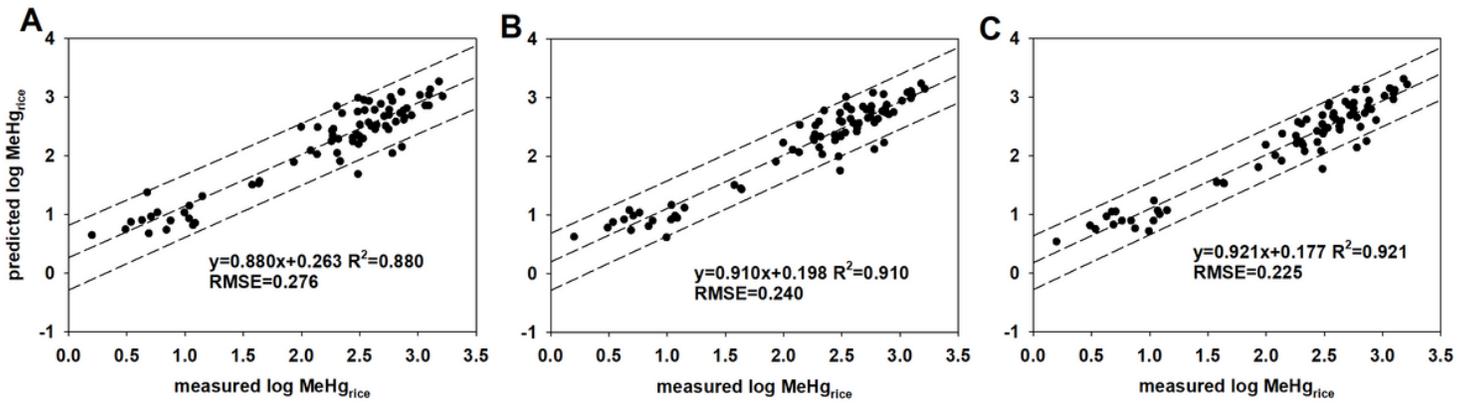


Figure 5

The linear graphs between measured log MeHg_{rice} and predicted log MeHg_{rice} which predicted by log MeHg_{soil}, figure A represents the linear graph between measured log MeHg_{rice} and predicted log MeHg_{rice} from eq (1) in table 3, and figure B represents eq (2) in table 3 and figure C represents eq (3) in table 3.

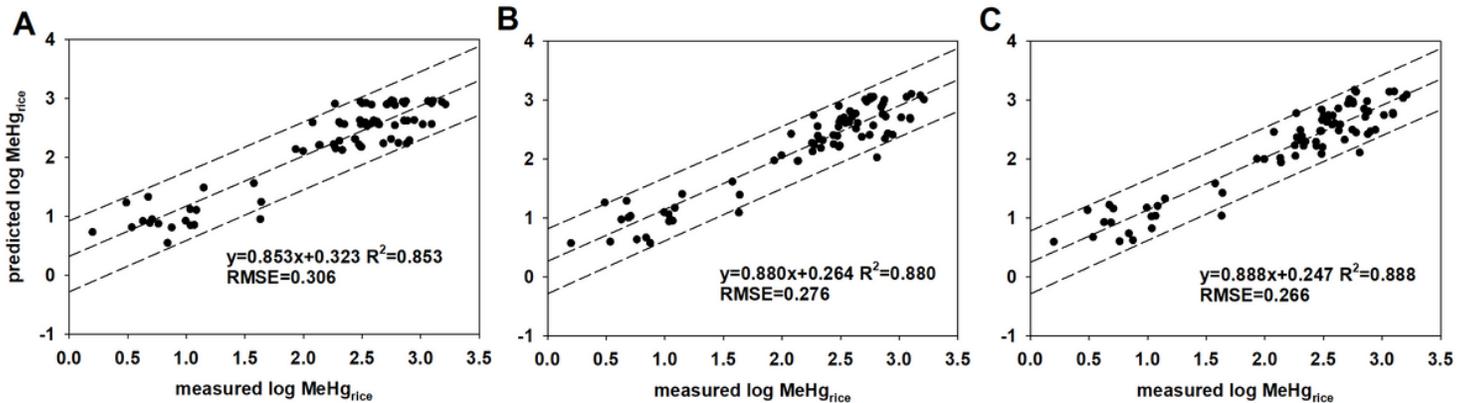


Figure 6

The linear graphs between measured log MeHg_{rice} and predicted log MeHg_{rice} which predicted by log THg_{soil}, figure A represents the linear graph between measured log MeHg_{rice} and predicted log MeHg_{rice} from eq (4) in table 3, figure B represents eq (5) in table 3, figure C represents eq (6) in table 3.

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