

Effect of Humic Acid Fertilizer on Mercury Release from Greenhouse Soils

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ABSTRACT

The elemental mercury (Hg⁰) release characteristics from the Hg-contaminated soil applied with Humic acid fertilizer (HAF) in the greenhouse were identified. The adsorption features of mercuric ion (Hg²⁺) on HAF under different reaction times and pH were investigated to elucidate the influencing mechanism of HAF on soil Hg⁰ release. Besides, the microstructure of HAF loading with Hg²⁺ was characterized by using Fourier transform infrared spectroscopy (FTIR) and scanning electron micrograph-energy dispersive spectrometry–EDS). The results showed that with the increasing HAF dosage, soil oxidation-reduction potential (Eh), and organic matter (SOM) content, as well as the decreasing soil pH, the soil Hg⁰ release fluxes showed a decreasing tendency. The soil pH, Eh, SOM, and total Hg content are the key factors that can affect the soil Hg⁰ release fluxes. The interior air temperature, light intensity, soil moisture, and soil temperature have little impact on soil Hg⁰ release fluxes when the greenhouse soil is applied with HAF. The HAF can immobilize Hg²⁺ and reduce its activity by surface precipitation and specific adsorption, then affecting the soil Hg⁰ release fluxes. The results of this study provide a basis for the application of HAF to reduce soil Hg⁰ release fluxes in the greenhouse of Hg-contaminated areas.

INTRODUCTION

The mercury (Hg) is a highly toxic element to the environment and has caused many public health hazards. It is one of the world's most recognized environmental pollutants and can damage the human nervous, urinary, digestive, and respiratory systems, causing mental and motor functional disorders (Mario et al. 2020, Baragano et al. 2022). Tongren Hg mining area is the largest Hg mining area in China. Long-term mining of Hg has not only caused serious terrain damage and Hg pollution to the surrounding areas but also led to the depletion of the Hg mine's resources. Therefore, the government planted to rehabilitate the mining and surrounding areas, and a large number of vegetable greenhouses were constructed to achieve regional resource transformation. The Hg concentrations of most crops grown in the farmlands of Wanshan Hg mine areas were higher than the governmental reference values (10 or 20 $\mu g \cdot k g^{-1}$) (Xia et al. 2020). As a result, the exposure risk of Hg to residents was high.

Previous studies had found that the light intensity, atmospheric Hg concentration, and land use type were the dominant factors involved in soil Hg release fluxes in Wanshan Hg mining areas (Wang et al. 2021, Zhao et al. 2022, Federico et al. 2023). The greenhouse is in a relatively confined environment where soil Hg release fluxes are generally higher than that of the external bare ground due to the greenhouse effect. The soil Hg release is the main source of interior atmospheric Hg in the greenhouse, which the leafy part of vegetables will assimilate (Liu et al. 2022). Fertilizer application can change the soil physicochemical properties around plant roots, which in turn affects the transport and accumulation of Hg in the soil-plant system (Geng et al. 2023). Humic acid fertilizer (HAF) is a brown or black amorphous polymeric colloidal complex that is the main component of soil organic matter (SOM), and HAF shows great affinity for Hg in soil (Boguta et al. 2019). HAF has a variety of reactive groups, including carboxyl, phenolic hydroxyl, alcohol hydroxyl, amino, and carbonyl groups, which can adsorb and complex with Hg in soil (Zhang et al. 2009).

By changing the amount of HAF in the soil, the adsorption and complexation of Hg with HAF will increase in the soil environment, then the reactivities of mercuric ion (Hg^{2+}) will decrease. As a result, the production of Hg^0 decreases, causing the soil Hg^0 release fluxes to reduce. Ultimately, the contamination of atmospheric Hg caused by soil Hg^0 release may be controlled.

Therefore, the soil Hg⁰ release characteristics from greenhouse soil applied with HAF were identified by the

simulated greenhouse. The adsorption features of Hg²⁺ in soil and HAF system were investigated to elucidate the influencing mechanism of HAF on the soil Hg release process. The purpose of this study is to verify whether the HAF can be used in controlling soil Hg⁰ release from the Hg-contaminated soil in a greenhouse, which will provide a new direction and theoretical basis for remediating Hgcontaminated soil.

MATERIALS AND METHODS

Incubation Experiment

The Hg⁰ release characteristics from the greenhouse soil applied with HAF were identified by using an incubation experiment. The test soil was collected from a vegetable field (106.64°E, 26.40°N) in Guiyang, Guizhou Province, China. After the weeds and debris were removed, the soil was dried at room temperature, crushed, and mixed homogeneously. Then, the soil was spiked with $Hg(NO_3)_2$ solution to make the final soil Hg content to be $\sim 40 \text{ mg.kg}^{-1}$ (Yan et al. 2019). The soil was mixed thoroughly again and left to equilibrium for 6 months. Then 18 kg of Hg-contaminated soil was placed into each greenhouse (length 80 cm, width 60 cm, and height 50 cm) with a height of ~20 cm. The 0 (CK), 180 (1%), 540 (3%), and 900 (5%) g of HAF (Plant Source Ecological Technology, Co. Lid., Tianjin, China) were amended to each greenhouse soil. Both the physical and chemical properties of test soil and HAF are shown in Table 1. The greenhouse film was made of polyethylene, whose transmittance of visible light is 85%. On 0 (just after the HAF was amended to soil), 5, 15, 30, and 60 d, the soil redox potential (Eh), soil temperature, air temperature, light intensity, and soil moisture content were monitored, simultaneously, the greenhouse soils were sampled to detect the total Hg (THg) content, pH, and organic matter (SOM) concentration.

Adsorption Experiment

The adsorption of Hg²⁺ on soil can affect the reactivities of soil Hg, then influencing the generation of soil Hg⁰ via reduction reaction. As a result, the soil Hg⁰ release fluxes were affected by adsorption. The amendment of HAF could change the physical and chemical properties of the soil environment, resulting in the adsorption characteristics

Table 1: The physical and chemical properties of tested soil and HAF.

Parameters	HAF	Soil
Hg concentration [mg·kg ⁻¹]	0.13	0.28
Organic matter [g·kg ⁻¹]	266.45	34.66
Eh [mV]	673.34	569
pH	5.8	6.7

of Hg²⁺ on soil changing simultaneously. Therefore, the adsorption characteristics of Hg²⁺ on the soil amended with HAF need to be identified. According to Table 2, a certain amount of HAF and/or soil was added to a centrifuge tube (50 mL). Then 20 mL Hg solution (50 mg.L⁻¹) was added, and the solution pH was adjusted by using NaOH (1 mol. L^{-1}) and HNO_3 (1 mol.L⁻¹). After that, the centrifuge tube was sealed with a cap and shaken at 120 r.min⁻¹ for a certain time (Table 2) under 25°C. The reaction solution was centrifuged at 4500 r.min⁻¹ for 5 min to collect the supernatant, which was used for Hg content detection. The precipitate was collected to characterize the microstructure. The microstructure and surface morphology of HAF were observed by scanning electron microscopy (SEM) (FEI Versa3D, USA), the surface elements of HAF were analyzed by applying EDS energy spectroscopy (point sweep), and Fourier analyzed the groups of HAF transform infrared spectroscopy (FTIR, Nicolet iS5, Thermo Scientific, USA).

The experimental data was simulated and fitted using pseudo-first-order (eq.(1)) and pseudo-second-order (eq.(2))kinetic model equations.

$$ln(q_e - q_t) = ln q_e - k_1 t \qquad \dots (1)$$

$$\frac{t}{q_t} = \frac{1}{k_2 \times q_e^2} + \frac{t}{q_t} \qquad \dots (2)$$

where, q_e and q_t (mg·g⁻¹) are the adsorption capacities of of Hg²⁺ on HAF at equilibrium and time t (min), respectively, k_1 (min⁻¹) and k^2 (g·mg⁻¹·min⁻¹) are the adsorption rate constants for pseudo-first-order and pseudo-second-order kinetic model equations, respectively.

Measurement Methods

The soil Hg release fluxes were measured using the kinetic fluxes box method, as described in the literature (Liu et al. 2022). The soil THg content was determined with a cold atomic absorption Hg detector (Model III, Brooks Rand, USA) after digestion using aqua regia (50%) (Li et al. 2020).

Table 2: The design of the adsorption experiment.

Treatment	HAF dosages [g]	Soil dosages [g]	рН	t.min ⁻¹	
СК	0	0	3, 4,	1440	
Soil	0	0.1	5, 6, 7 and		
HAF	0.03	0	8		
Soil+HAF	0.03	0.1			
СК	0	0	6	5, 15, 30,	
Soil	0	0.25		60, 120, 240, 480	
HAF	0.075	0		720, and	
Soil+HAF	0.075	0.25		1440	



Fig. 1: The soil Hg⁰ release fluxes in the greenhouse with the application of HAF.

The air-dried soil (particle size 2 mm) was dissolved in deionized water (solid to liquid ratio 1:2.5), stirred for 1 min, rested for 30 min, and determined by a pH meter (PHS-3C, INESA instrument, Shanghai, China). The SOM content was determined by the potassium dichromate oxidation method (Yao et al. 2022). The Soil oxidation-reduction potential (Eh) was determined by an ORP meter (TR-901, Remagnet Shanghai, China). The soil temperature and soil moisture content were determined by a soil temperature and humidity meter (TR-6D, Tianjin, China). The light intensity and air temperature in the greenhouse were measured with an illuminance meter (Digital Lux Mater GM1020, Beijing, China).

RESULTS AND DISCUSSION

Soil Hg⁰ Release Characteristics

Generally, the soil Hg^0 release fluxes decreased with the increasing incubation time and HAF dose (Fig. 1). For all treatments, the soil Hg^0 release fluxes were highest at the initial stage (0 d). On 0 d, for the treatment CK, 1%, 3%, and 5% HAF, the soil Hg^0 release fluxes were measured to be 1633.00, 1734.70, 1748.44, and 1847.65 ng·m⁻²·h⁻¹, respectively. Compared to 0 d, on 5 d, the soil Hg^0 release fluxes respectively decreased by 13.2%, 43.79%, 82.21% and 67.48%. On 15 d, although the soil Hg^0 release fluxes were significantly higher than that on 5 d, 30 d, and 60 d, it still showed a tendency that decrease with increasing HAF dose. The smallest soil Hg^0 release fluxes were found on 60 d. On 60 d, the soil Hg^0 release fluxes respectively decreased

by 33.54%, 56.78%, 79.09%, and 83.59% compared with 0 d. At the initial stage (0 d), the soil needed to be turned over to ensure that the HAF could be mixed uniformly with the soil, causing the originally attached Hg^0 of the soil particles surface to migrate rapidly to the atmosphere (O'Connor et al. 2019). Therefore, the soil Hg^0 release fluxes were high on 0 d. With increasing time and the impact of watering (to maintain soil moisture at ~50%), the soil surface gradually becomes dense, and the exposed area to air decreases; thereby, the soil Hg^0 release fluxes decrease simultaneously (O'Connor et al. 2019). The application of HAF can significantly reduce the soil Hg release fluxes, which can be attributed to the fact that the HAF will adsorb and immobilize Hg ions in the soil, reducing the reactive activity of Hg. The generation of Hg⁰ is inhibited (Yan et al. 2019).

The Influence of Soil Physical and Chemical Properties and Environmental Conditions on Soil Hg⁰ Release Fluxes

Previous studies have found that the soil THg content (Moore & Carpi 2005), pH (Zhang & Lindberg 1999), Eh (Martyniuk & Ckowska 2003), SOM (Wang et al. 2005), light intensity, soil moisture, and temperature (Nascimento & Masini, 2014) are the key factors that can determine the soil Hg⁰ release fluxes. Generally, the soil Hg⁰ release fluxes are elevated when the soil THg concentrations height (Wang et al. 2007). For all treatments, the soil THg concentrations decreased with the increasing incubation time (Fig. 2a). On 60 d, for the treatments CK, 1%, 3%, and 5% HAF, the soil THg concentrations reduced by 6.99, 7.74, 8.17, and 6.84 mg kg, respectively, compared with 0 d. The correlation

analysis found that the coefficient between soil Hg⁰ release fluxes and soil THg concentrations for the HAF addition treatments ($R^2 = 0.31$) was significantly lower than that for the CK treatment ($R^2 = 0.62$, Fig 3e). This indicates that soil Hg concentration is an important factor that can affect the soil Hg release fluxes when HAF is not applied. However, when the soil was amended with HAF, it would hide the influence of soil THg on Hg⁰ release fluxes.

In the soil environment, the rate of reduction of Hg^{2+} to Hg^{0} decreased, and soil Hg^{0} release fluxes decreased under low soil pH conditions (Yang et al. 2007). When the soil pH increases, Hg²⁺ will combine with OH- in the liquid phase of the soil, reducing the surface adsorption of Hg^{2+} on soil minerals and facilitating Hg⁰ release (Xin & Gustin 2007). In the present study, we did not find significant fluctuation of soil pH for the treatment CK. However, for the HAF treatments, the soil pH decreased gradually with the increase of incubation time. Compared to 0 d, the soil pH of 1%, 3%, and 5% HAF treatments decreased by 2.79%, 4.48%, and 9.85% on 60 d (Fig. 2b). This suggests that the application of HAF can reduce soil pH, which the degradation of HAF may cause to generate humic acid (HA), xanthic acid (FA) and humic acid (HU) (Moore & Castro 2012, Peng et al. 2022). The correlation analysis found that the soil Hg^0 is positively related to soil pH ($R^2 = 0.35$). Even so, we think that the soil pH may not be the predominant factor involved in the Hg⁰ release process because the coordination of HAF with Hg may be the key influencing factor.

The soil Eh of CK, 1%, 3%, and 5% HAF treatments were 485, 487, 508, and 495 mV on 0 d, respectively. Then, it was found that the soil Eh showed slightly increasing tendencies with incubation time for the HAF treatments. On 60 d, the respective soil Eh of 1%, 3%, and 5% HAF

treatments increased by 69, 65, and 92 mV, respectively (Fig. 2c). This suggested that the application of HAF could elevate the soil Eh, resulting in that the soil environment would be more oxidative. Therefore, the soil Hg might undergo an oxidation process to generate Hg²⁺ and/or Hg⁺, then the soil Hg⁰ release fluxes decreased (Wang et al. 2005). There is a negative correlation between soil Hg⁰ release fluxes and Eh $(R^2 = 0.46)$, which also suggests that soil Eh is an important factor involved in the Hg⁰ release process.

The SOM can sorb and immobilize Hg ions in the soil, causing the reactivity of soil Hg to decrease, which in turn affects the soil Hg release process (Wang et al. 2005). With the extension of incubation time, the SOM content of CK treatment did not fluctuate significantly. Still, a significant increase of SOM content was found in HAF treatment (Fig. 2d). This could be attributed to the that there was a large amount of organic matter contained in HAF, which was introduced to the soil environment. Correlation analysis revealed that the soil Hg⁰ release fluxes significantly negatively correlated with SOM content after amending with HAF ($R^2 = 0.75$, Fig. 2d). This suggested that the application of HAF could affect the soil's physical and chemical properties, then influencing the Hg⁰ release fluxes.

The correlation analysis showed that the linear regression coefficients between the soil Hg⁰ release fluxes and soil moisture, greenhouse air temperature, light intensity, or soil temperature were all low (Fig. 3). This suggested that the other parameters, except for soil temperature, were not the key factors that can affect soil Hg⁰ release fluxes. The soil temperature was as high as 35.9°C on 15 d, which was significantly higher than the soil temperature at other periods. Then the thermal movement of Hg molecules increased, and the adsorption capacity of Hg^0 on the soil surface decreased,



Fig. 2: The fluctuation of soil THg (a), pH (b), Eh (c), and SOM (d).

promoting the soil Hg^0 entering the pores. As a result, the soil Hg^0 release fluxes elevated with high soil temperature (Moore & Carpi 2005). This may be the key reason for the significant increase in soil Hg release fluxes at 15 d.

It can be seen that soil pH, Eh, SOM, and THg are the key factors that can affect soil Hg⁰ release fluxes in a greenhouse under fertilization conditions, while greenhouse air temperature, light intensity, soil moisture content, and soil temperature have less effect on soil Hg⁰ release fluxes. This result is somewhat different from the conclusion of previous studies that light intensity, temperature, and soil water content are the important factors involved in soil Hg⁰ release (Carpi & Lindberg 1997, Zhang & Lindberg 1999, Hamush et al. 2012, Shen et al. 2019). This is mainly due to the relatively closed environment of the greenhouse, low air mobility, and high temperature of the greenhouse. Besides, the light intensity is weakened by the greenhouse membrane, resulting in temperature, and light intensity is no longer the main influencing factor. In addition, the soil moisture content is maintained at $\sim 20\%$, so it is not a key influencing factor.

Adsorption Characteristics of Hg²⁺ on Soil and HAF Mixed Systems

The main component of HAF is organic matter, which contains functional groups such as -COOH, -SH, -OH, etc. These functional groups have a strong adsorption effect for Hg ions in soil, which affects the reactivity of Hg in soil and then affects the production and release of Hg⁰. Therefore, it is necessary to investigate the effect of HAF on the adsorption of Hg²⁺ in soil. It was found that the soil, HAF, and soil+HAF treatments showed rapid adsorption of Hg²⁺,

basically reaching equilibrium within 5 min (Fig. 4a). The concentrations of Hg²⁺ in the solutions of the soil treatment $(3.58-11.93 \text{ mg} \cdot \text{L}^{-1})$ were higher than those of HAF treatment $(2.130-7.821 \text{ mg} \cdot \text{L}^{-1})$ under different pH conditions. The application of HAF could further reduce the concentrations of Hg in the solutions, indicating that HAF promoted the adsorption of Hg²⁺ by the soil (Fig. 4b). The kinetic model equations found (Table 3) that although the pseudo-firstorder order kinetic model could well describe the adsorption of Hg^{2+} on HAF, soil + HAF, and soil, the fitted theoretical adsorption capacity (q_{e1}) differed significantly from the experimental results. The fitting coefficients (R_2^2) of the pseudo-second-order equation were higher than that of the pseudo-first-order order kinetic model. Additionally, there was no difference between the theoretical adsorption capacity (q_{e2}) and the experimental results. This indicated that the pseudo-second-order kinetic model can better describe the adsorption of Hg²⁺ by HAF. The adsorption process is dominated by chemisorption, which may include surface precipitation as well as specific adsorption (Martyniuk & Ckowska 2003).

FTIR characterization revealed that before the HAF adsorbing Hg^{2+} , the peaks at 3305-3500 cm⁻¹ were the stretching vibration of -OH and -NH₂, the peaks at 1557 cm⁻¹ were C=C and C=O, the peaks at 1378 cm⁻¹, 1005 cm⁻¹, and 912 cm⁻¹ were telescopic vibration peaks of phenolic hydroxyl, C-O and Si-O (Nascimento & Masini 2014, Perla et al. 2023). After the HAF loading with Hg^{2+} , the characteristic peaks of carboxyl, amino, and phenolic groups were shifted to the right, and the intensity of the peaks was significantly weakened, which indicated that carboxyl,



Fig. 3: The relationship between the soil Hg⁰ release fluxes and the light intensity (a), soil moisture (b), soil temperature (c), and greenhouse air temperature (d).



Fig. 4: The adsorption capacity (a) with time and residual Hg concentration (b) with pH for each treatment.

Table 3: The fitting results of pseudo-first-order and pseudo-second-order kinetic model.

Data of this experiment pseudo-first-order order kinetic mo		order kinetic mode	l pseudo-second-order kinetic model				
Туре	$q_e [mg \cdot g^1]$	$q_{e1} [mg \cdot g^{-1}]$	k ₁ [min ⁻¹]	R_1^{2}	$q_{e2} [mg.g^{-1}]$	$K_2[g \cdot mg^{-1} \cdot min^{-1}]$	R_2^{2}
HAF	32.87	1.83	0.0055	0.9673	32.89	0.0111	1
Soil+HAF	7.65	0.14	0.0069	0.9385	7.67	0.1702	1
Siol	9.87	0.69	0.0053	0.9877	9.88	0.0289	1

amino, and phenolic groups could be coordinated with Hg^{2+} to form complexes (Fig. 5).

Before the Hg^{2+} was adsorbed on HAF, whose surface structure was arranged orderly after the HAF adsorbed Hg^{2+} , the pore structure of HAF would collapse. Fragments would exist in the pores (Fig. 6). This could be attributed to the complexation between Hg^{2+} and carboxyl groups, hydrogen bonds, and other functional groups of HAF, then generating cations, which in turn form aggregates (Liu et al. 2016). EDS spectra showed that the main elements in HAF were C, O, Si, S, K, Ca, Al, and Fe (Fig. 7). The Hg elemental was observed in the range of 2-2.5 and 9.1-11.7 keV before HAF adsorbed Hg²⁺. After the adsorption of Hg²⁺, the characteristic intensity of Hg increased slightly, and the weight percentage of elemental Hg increased from 1.3 to 23.91, indicating that a large amount of Hg is attached to the surface of HAF. The characteristic intensity of O and S elements becomes relatively weaker, and their weight



Fig. 5: The FTIR spectra before and after HAF adsorbing Hg²⁺.



Fig. 6: The SEM images of HAF before and after adsorbing Hg²⁺.



Fig. 7: EDS spectrum before (a) and after the HAF adsorbed Hg²⁺.

percentage decreases. The changes in the intensity of characteristic peaks may be due to the complexation of Hg^{2+} with O and S elements to form aggregates.

Based on the results of SEM, FTIR, and EDS elemental analysis, combined with previous research results (Zhang et al. 2020), it was hypothesized that the mechanism of HAF adsorption of Hg^{2+} might be:

$$COOH+Hg^{2+} \rightarrow -COO-Hg^{+}+H^{+}$$
(3)

$$-OH+Hg^{2+} \rightarrow -O-Hg^{+}+H^{+}$$
(4)

$$-SH+Hg^{2+} \rightarrow -SHg^{+}+H^{+} \qquad \dots (6)$$

$$-S-+Hg^{2+} \rightarrow -S(Hg)- \dots(7)$$

The strong adsorption of functional groups contained in HAF may lead to a decrease in the reactivity of Hg in the soil environment, which inhibits the conversion of Hg^{2+} to Hg^{0} and then affects the soil Hg release process in the greenhouse.

CONCLUSION

Temperature, light intensity, soil moisture content, and soil temperature have little effect on soil Hg⁰ release fluxes. This is different from previous studies and may be related to the stable and controllable environmental conditions of the greenhouse versus variable and uncontrollable environmental factors in the natural state. The soil pH, Eh, SOM, and THg have a strong influence on the soil Hg⁰ release fluxes when

the greenhouse soil is amended with HAF. The addition of HAF changed the soil pH, Eh, and SOM, then decreased the soil Hg⁰ release fluxes. The main effects were as follows: (1) Enhancing the soil acidity in the greenhouse caused a decrease in the rate of reduction of Hg²⁺ to Hg⁰, which in turn reduces the soil Hg⁰ release fluxes. (2) Alteration of the soil Eh, which enhanced soil oxidation states and induced a shift of Hg⁰ to a stable higher valence state, thereby reducing the soil Hg⁰ release fluxes. (3) Increased the soil SOM content in the greenhouse, the strong adsorption of functional groups in SOM will adsorb and fix the Hg ions in the soil, reducing the reaction activity of Hg and, thus, the production of Hg⁰ decreased, which in turn reduced the soil Hg⁰ release fluxes. The soil Hg⁰ release fluxes of greenhouse decreased with the increase of incubation time and decreased with the increasing dosage of HAF. The adsorption kinetic characteristics are in accordance with the pseudo-second-order kinetic model, and the adsorption process is dominated by chemisorption, which may include surface precipitation as well as specific adsorption.

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