Mercury in soils of three agricultural experimental stations with long-term fertilization in China

Yuan-Ming Zheng^a, Yu-Rong Liu^a, Hong-Qing Hu^b, Ji-Zheng He^{a,*}

^a State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

^b Key Laboratory of Subtropical Agricultural Resources and Environment, Huazhong Agricultural University, Wuhan 430070, China

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ABSTRACT

Mercury (Hg) in the agricultural ecosystem is a global concern because of its high potential toxicity. The objectives of this study were to determine the concentration and distribution of Hg in soils from three long-term experimental stations, i.e., Taoyuan (TY) and Qiyang (QY) in Hunan Province and Fengqiu (FQ) in Henan Province of China, and thus to assess the possible food and health risk of long-term applications of fertilizers. Soil samples at each site were collected from different fertilization plots and also from soil profiles with depths 0–100 cm. There were significant differences in soil Hg concentrations in 0–20 cm (A) or 20–40 cm (B) horizon among the three experimental stations. QY station showed significantly higher Hg concentrations than TY and FQ stations. However, there were no significant differences in soil Hg concentrations between A and B horizons at each station. It was concluded that the soil Hg concentrations at the three sites were mainly controlled by the parent materials. Moreover, chemical fertilizer, especially phosphorous fertilizers, could influence the soil Hg concentrations to some extent at the station with lower soil Hg concentrations, for example, at TY station. There were minimal amounts of Hg resulting from applications of the other chemical fertilizers and organic manure, and thus the fertilization had very low risk to the food security of the agro-ecosystems in the terms of Hg inputs and contamination.

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1. Introduction

Mercury (Hg) is a nonessential element and concerned as it is a potential neurotoxin (Zagury et al., 2006). Because of its persistence in the environment and possible toxicity to organisms, Hg has been listed as one of the priority pollutants by many international agencies (Jiang et al., 2006), such as United Nations Environment Programme, World Health Organization and Food and Agriculture Organization of the United Nations. Many previous studies have shown the contributions of mining and smelting activities to the concentration of Hg in ecosystems (Fernandez-Martinez et al., 2005; Kotnik et al., 2005; Feng et al., 2006; Qiu et al., 2006). Other anthropogenic activities, such as petroleum refining and fossil fuel combustion (in particular coal), industrial production processes, waste incineration, application of fungicides and land spreading of sewage sludge, are also important sources of Hg to the soil environment (Steinnes, 1995).

In China, the amount of Hg utilization has increased significantly due to the rapid development of industry and agriculture during the past two decades, for example, the use in the chlor-alkali production, batteries, fluorescent lamps, and fungicides

(Streets et al., 2005). The anthropogenic Hg emissions in China were estimated at 536 (± 236) tons in 1999 (Streets et al., 2005), and total consumption of Hg was over 900 tons in 2000, accounting for 50% of the world production (Jiang et al., 2006). High Hg accumulation has been found in many ecosystems. Serious Hg pollution caused by wastes discharged from chemical plants occurred in some regions, such as the Songhua and Jiyun Rivers (Jiang et al., 2006). Guiyang, capital of Guizhou Province, southwest of China, has serious Hg contamination in the atmosphere (Feng et al., 2003). And Wanshan, located in eastern Guizhou Province, has the largest Hg deposition in China, where the concentrations of soil Hg range from 24.3 to 34.8 mg kg⁻¹ (Ding et al., 2004; Jiang et al., 2006).

It has been found that many other factors, such as parent material (Rodrigues et al., 2006), organic matter (Lee et al., 1994), soil microbes (Barkay and Wagner-Dobler, 2005), played important roles in influencing the concentration and distribution of Hg in the soil. Clay minerals and oxides, particularly iron (Fe) and aluminum (hydr) oxides, controlled the transfer of Hg along soil profiles (Roulet et al., 1998). Therefore, high variability of soil Hg concentrations was closely correlated to the soil characteristics (Fernandez-Martinez et al., 2005; Rodrigues et al., 2006). Some of the crucial changes in soil Hg concentration could also be induced by the changes of land use and soil fertility (Lacerda et al., 2004; Mainville

* Corresponding author. Tel.: +86 10 62849788; fax: +86 10 62923563.

E-mail address: jzhe@rcees.ac.cn (J.-Z. He).

et al., 2006). In addition, plants could function as conduits for the interfacial transport of Hg from the soil to the atmosphere, and this capacity of transportation often varied according to the plant species (Leonard et al., 1998; Moreno-Jimenez et al., 2006).

However, most of the data generated from these studies were only concerned about the ecosystems polluted seriously or farmlands with no regular agricultural management practices. There is little information about the concentration and distribution of Hg from different agricultural soils with long-term consistent fertilizer applications. Therefore, it is necessary to reveal the status of Hg in agricultural soils through investigating the effects of agricultural management practices, which are vitally correlated to the security of agricultural products. The objectives of this study were to investigate the concentration and distribution of Hg in soils from three long-term fertilization experiments. Based on the investigation, some sources of soil Hg would be discussed and the possible health risk of soil Hg would also be assessed in agricultural ecosystems.

2. Materials and methods

2.1. Study area

Three long-term agricultural experimental stations, i.e., Taoyuan station (TY, 111°27'E, 28°55'N), Qiyang station (QY, 111°53'E, 26°45'N) and Fengqiu station (FQ, 114°24'E, 35°00'N), were selected for the comparison of soil Hg concentrations. The three long-term experiments, located at Taoyuan County, Qiyang County, and Fengqiu County, respectively, represented three different agroecosystems. TY and QY are both in Hunan Province. The annual rainfall is about 1255–1448 mm and the mean temperature is 16–18 °C in this region. The soil type is red soil (clay 45.0%, silt 46.3% and sand 8.7%) in QY, and paddy soil (about 40% clay) in TY. Both of them were derived from quaternary red clay earth. The long-term fertilization experiments were commenced in 1990 at these two stations. FQ is located in Henan Province and belongs to the Huang-Huai-Hai Plain, which is an important agricultural area of China with dry farming agriculture. Fengqiu County has an annual rainfall of 650 mm and an annual mean temperature of 13.9 °C, showing the temperate monsoon climate. The soil type is fluvo-aquic soil derived from the sediments of Yellow River. It is a typical soil in the region with a texture of sandy loam in the plough layer and loam in the subsoil. The long-term fertilization experiments were commenced in 1990.

2.2. Experimental treatments, sampling and analytical methods

The fertilization treatments and some basic soil chemical characteristics in the three experimental stations are listed in Table 1. There were three replicates for each treatment in TY and four replicates in QY and FQ stations. All treatments were arranged randomly and managed with long-term continuous fertilization practices for about 16 years. The soil samples were collected from all treatments at 0–20 cm (A) in depth and some of the treatments also at 20–40 cm (B) in depth.

Meanwhile, at these three sites, one soil profile was set in the cropland of the experimental station without fertilization (labeled as P1) and the nearby vegetable field (labeled as P2) which usually received more intensive agricultural management (e.g., more fertilizers and pesticides) than the cropland, to examine the variation of Hg concentration along the soil profiles. In this way, six soil profiles, i.e., TYP1, TYP2, QYP1, QYP2, FQP1 and FQP2, were obtained. At each profile, soil samples were taken every 5 cm between 0 and 40 cm depths and every 10 cm between 40 and 100 cm depths.

Soil samples were air-dried and ground to a size finer than 0.149 mm. Half a gram of each sample was digested with HNO₃ + HCl (10 ml, 1:1 v/v) in a teflon tube with cover at 100 °C for 2 h (Lacerda et al., 2004; Feng et al., 2006). The total Hg concentration was then determined using an atomic fluorescence spectrometer (AFS, AFS-2202E Haiguang Analytical Instrument Co., Beijing, China). Two standard reference materials, GBW-07401 (GSS-1) and GBW-07405 (GSS-5), obtained from Center for National Standard Reference Materials of China, were inserted to the analytical process for quality assurance/quality control. The measured Hg concentrations of the two standard soil samples GSS-1 and GSS-5 were $0.033 \pm 0.003 \text{ mg kg}^{-1}$ ($n = 8$), and $0.30 \pm 0.02 \text{ mg kg}^{-1}$ ($n = 8$), respectively, which were comparable with their certified value of $0.032 \pm 0.004 \text{ mg kg}^{-1}$ and $0.29 \pm 0.04 \text{ mg kg}^{-1}$, respectively.

2.3. Data analysis

Analysis of variance was performed using ANOVA with SPSS for Windows. Data were presented as mean and standard deviation for the same treatment at the same experimental site. The soil Hg concentrations of A horizons were divided by the corresponding values of B horizons and thus the ratios of Hg concentrations for these two horizons (RCH) were obtained for further comparison.

Table 1

Fertilization treatments and some chemical properties of agricultural soil samples from Taoyuan (TY) and Qiyang (QY) in Hunan Province and Fengqiu (FQ) in Henan Province, China, where the chemical fertilizers and manure applications have lasted for 16 years

TY	Treatment	CK ^a	NK ^b	NP ^c	NP+C ^d	N+C ^e	NPK ^f	NPK+C ^g
	pH H ₂ O	4.05 ± 0.05	4.10 ± 0.15	4.11 ± 0.03	3.98 ± 0.03	4.01 ± 0.05	4.03 ± 0.01	4.03 ± 0.02
	Organic matter (g kg ⁻¹)	28.89 ± 1.71	30.37 ± 6.15	30.67 ± 7.21	48.60 ± 1.43	41.62 ± 4.48	35.09 ± 3.12	46.27 ± 0.77
QY	Treatment	CK	NK	NP	OM ^h	PK ⁱ	NPK	NPK + OM ^j
	pH H ₂ O	5.47 ± 0.18	3.76 ± 0.12	3.95 ± 0.11	5.31 ± 0.19	5.03 ± 0.14	4.04 ± 0.06	5.81 ± 0.15
	Organic matter (g kg ⁻¹)	13.60 ± 0.56	14.32 ± 0.52	16.36 ± 0.87	21.74 ± 0.07	14.60 ± 0.72	16.88 ± 0.81	21.25 ± 0.75
FQ	Treatment	CK	NK	NP	OM	PK	NPK	NPK + OM
	pH H ₂ O	8.77 ± 0.04	8.47 ± 0.04	8.45 ± 0.04	8.62 ± 0.04	8.67 ± 0.04	8.40 ± 0.04	8.54 ± 0.03
	Organic matter (g kg ⁻¹)	7.29 ± 0.26	7.45 ± 0.23	9.56 ± 0.17	16.77 ± 0.67	8.42 ± 0.46	9.89 ± 0.04	12.82 ± 0.03

^a CK = Control without fertilizers.

^b NK = Nitrogen and potassium fertilizers.

^c NP = Nitrogen and phosphorus fertilizers.

^d NP + C = Nitrogen, phosphorus fertilizers and incorporation with rice straw.

^e N + C = Nitrogen fertilizer and incorporation with rice straw.

^f NPK = Nitrogen, phosphorous and potassium fertilizers.

^g NPK + C = Nitrogen, phosphorous, potassium fertilizers and incorporation with rice straw.

^h OM = Organic manure.

ⁱ PK = Phosphorus and potassium fertilizers.

^j NPK + OM = Nitrogen, phosphorous, potassium fertilizers and organic manure.

3. Results

3.1. Mercury concentrations in soils of the three experimental sites

The descriptive statistics of the Hg concentrations in two soil horizons from different stations are shown in Table 2. The highest mean Hg concentration was observed in samples from QY, ranging from 0.470 mg kg⁻¹ to 0.777 mg kg⁻¹. Based on the ANOVA statistical analysis, the Hg concentrations of QY were significantly higher than those of TY and FQ, and TY significantly higher than FQ.

The mean soil Hg concentration in A horizon was higher than in B horizon in TY station while contrary trend was observed in QY station. It was found that all of RCH values in QY were smaller than 1 with mean value of 0.870, while all of the RCH in TY were greater than 1 with the highest mean value of 1.380 among the three stations. The RCH of FQ showed complex variation ranging from 0.381 to 1.826. The highest standard deviation of RCH was observed in FQ which was much higher than those in TY and QY.

3.2. Mercury concentrations in the soils under different fertilization treatments

Soil Hg concentrations of A horizon under different fertilization treatments of the three experimental stations are shown in Figs. 1–3. For TY (Fig. 1), the treatments of nitrogen and phosphorus fertilizers (NP) and nitrogen, phosphorus and incorporation with rice straw (NP + C) had significantly higher levels of Hg than the others except the treatment of nitrogen, phosphorus, potassium fertilizers and incorporation with rice straw (NPK + C), and the treatments fertilized with phosphorus except the treatment of nitrogen, phosphorus, and potassium fertilizers (NPK) had significantly higher Hg concentration than the treatments without phosphorous fertilizers, especially than control without fertilization (CK). For QY (Fig. 2), Hg concentrations in CK were higher than those in the others. However, there were no significant differences among the all treatments. For FQ (Fig. 3), Hg concentrations in the soils of the NPK treatment were significantly higher than the concentrations of the others. Higher concentrations of Hg were found in the treatments using phosphorus fertilizers compared with CK.

3.3. Distributions of mercury along soil profiles in the three agricultural experimental stations

The variation trend of Hg distribution along the soil profiles was different at the three sites (Fig. 4). In TYP1 and TYP2 the concentra-

Table 2
Total Hg concentrations and RCH values in soils of the three agricultural stations

Site	Horizon	Sample number	Minimum	Maximum	Median	Mean	SD
Concentration: mg kg ⁻¹							
TY ^a	A ^b	21	0.160	0.246	0.188	0.196	0.036
	B ^c	15	0.126	0.159	0.140	0.140	0.012
QY ^d	A	28	0.470	0.644	0.530	0.539	0.055
	B	28	0.547	0.777	0.611	0.622	0.039
FQ ^e	A	28	0.020	0.058	0.038	0.037	0.014
	B	28	0.025	0.0520	0.032	0.035	0.008
RCH ^f							
TY		15	1.146	1.740	1.342	1.380	0.027
QY		28	0.787	0.969	0.887	0.870	0.063
FQ		28	0.381	1.826	1.201	1.138	0.516

^a TY = Taoyuan.

^b Samples from 0 to 20 cm depth.

^c Samples from 20 to 40 cm depth.

^d QY = Qiyang.

^e FQ = Fengqiu.

^f RCH = ratios of soil Hg concentrations in A to B horizons.

Fig. 1. Soil Hg concentrations in depth 0–20 cm under different fertilization treatments in TY station (the meanings of the treatments are present in Table 1). Different letters labeled in the figure showed the significant differences between treatments.

Fig. 2. Soil Hg concentrations in depth 0–20 cm under different fertilization treatments in QY station. Different letters labeled in the figure showed the significant differences between treatments.

Fig. 3. Soil Hg concentrations in depth 0–20 cm under different fertilization treatments in FQ station. Different letters labeled in the figure showed the significant differences between treatments.

tions of Hg did not differ a lot along the soil profiles, while in FQP1 and FQP2 soil Hg concentrations tended to decrease with the profile depth. There were no significant differences between P1 and P2 in these two sites. However, soil Hg concentrations of QYP2 were significantly lower than those of QYP1.

Fig. 4. Mercury distribution along soil profiles from the three experimental sites. The TYP1, QYP1 and FQP1 were the soil profiles collected from croplands without fertilization in the three experimental stations, and the TYP2, QYP2 and FQP2 were collected from vegetable lands with normal fertilization nearby the each experimental station.

4. Discussion

4.1. Mercury concentration in soils under different agricultural management practices

To some extent agricultural management practices could affect the characteristics of soil (Tomiyasu et al., 2003; Xiang et al., 2006; Manna et al., 2007). The results showed that human activities were influencing the soil Hg concentrations to the different extents.

Plant roots and shoots could be a reservoir of heavy metals and the biomass of plant would be related to the uptake amount of heavy metals by the plant (Moreno-Jimenez et al., 2006). In QY, mean Hg concentrations in maize (main crop of QY) followed the order: leaf (81.1 $\mu\text{g/kg}$) > root (43.1 $\mu\text{g/kg}$) > stem (29.4 $\mu\text{g/kg}$), which were 3–10 times higher than those in grains showing that biomass of plant contribute greatly to the remove of soil Hg from field. When collecting the samples on the site, it was observed that there was the smallest biomass of plant in CK treatment compared with the others in QY. Therefore, soil Hg concentration in CK treatment of QY was slightly higher than those in the others although all of these could hardly affect the soil Hg significantly. This phenomenon was only found in QY because of the higher soil Hg compared with other sites.

Moreover, fertilizers might be important sources of heavy metals in agricultural soils (Van Loon, 1974; Hutton and Symon, 1986). In this study, the analysis revealed high level of Hg (0.85 mg kg^{-1}) in the phosphorous fertilizers, which was coincided with the higher concentrations of Hg in the treatments fertilized with phosphorus compared with CK treatment in TY and FQ ($P < 0.05$). Nevertheless, there was no similar trend in the treatments of QY, which might be due to its high background level of Hg which covered up the effect of phosphorus fertilizer on the increase of Hg

concentration, i.e., the disturbances of soil Hg from fertilization could only be found given the low concentrations of soil Hg. This effect would be further discussed in the following section.

4.2. Effects of parent materials on mercury concentration in the soils

Soil parent material is an important factor affecting the concentration of trace metals in the soil (Kauranne, 1992) and in areas with heterogeneous lithology heavy metal contents in soils can exhibit a high variability controlled by the parent materials (Facchinelli et al., 2001), while climate factors related to the weathered process of parent rock, such as rainfall and temperature, could also contribute to the effects of parent materials on the metal level in the soil, such as Hg (Rodrigues et al., 2006). In this study, significant differences of Hg concentrations existed not only among the A horizon from the three stations, but also among the B horizon (Table 2). In general, the concentrations of trace metals in B horizon were considered to be closer to the parent materials. Thus the background concentrations of Hg in the soil were much different in the three stations because of the big differences of the physical geographical conditions, which also meant that agricultural practices from human could not change the soil Hg to such a great extent. All of the effects mentioned in the previous section were based on the “natural” concentrations of soil Hg. Taken into account the high level of soil Hg in the P1 of QY, the minor variation of Hg concentration along the P1 also embodied the key role of parent materials clearly.

4.3. Potential risk of the agricultural practices

Based on the results and discussion above, it could be concluded that the concentrations of Hg in the soils could be controlled by several factors. Parent materials gave the original concentrations of Hg while human management practices disturbed the natural background. The mean RCH of TY and of FQ were greater than 1 which showed a limited trend of Hg accumulation in the soil, while the RCH of QY was smaller than 1 which indicated the reduction trend of Hg in the soil. Thus, the disturbance of human activities varied with the different agro-ecosystems. However, the effects of agricultural management practices were still much less important than the effects of soil parent materials since no significant differences were observed between the soil Hg concentrations of A and B horizons in the three stations.

Considering the effects of all these factors, the application of phosphorous fertilizer might influence the soil Hg concentrations to some extent. However, this phenomenon only emerged in the soils with lower Hg background. The application of other chemical fertilizers and organic manure would not significantly change the soil Hg concentrations in the current study. It could be concluded that there was little Hg accumulation risk in the soil from the long-term fertilization practices.

Table 3
Mercury concentrations in soils reported in the literature

Soil type/land use	Location	Median/mean (mg kg^{-1})	Range (mg kg^{-1})	Reference
Suburb soil	Shanghai, China	0.086	0.031–0.169	Lu et al., 2006
Suburb soil	Nanjing, China	0.176	0.122–0.233	Fang et al., 2002
Suburb soil	Wuhan, China	0.181	0.006–0.447	Wang et al., 2005
Suburb soil	Taiyuan, China	0.11	0.061–0.297	Wang et al., 2003
Farmland soil	Guiyang, China	0.237	0.13–0.42	Deng et al., 2006
Agricultural soil	Chongqing, China	0.096	0.045–0.116	Guo et al., 2000
Agricultural soil	Changjiang Delta, China	0.587	0.2–1.04	Zhang et al., 2006
Agricultural soil	Zhujiang Delta, China	0.498	0.16–0.84	Zhang et al., 2006
Agricultural soil	Canada	0.04	0.005–0.13	Reimann and Caritat, 1998
Agricultural soil	Belgium	0.12/0.24	0.03–4.19	Tack et al., 2005
Background	China	0.038	0.006–0.272	Fang et al., 2002

4.4. Comparison of soil mercury concentrations in different studies

There was no uniform standard to assess the potential risk of soil Hg contamination. We attempted to show the characteristics of Hg distribution in soils of the three agricultural ecosystems by comparing with other agricultural soils in the world. Mercury concentrations in soil from QY station, exceeding mean Hg background of China a lot (CNEMC, 1990), were comparable to the concentrations in Changjiang and Zhujiang Delta (Zhang et al., 2006) and much higher than those in other cities or regions of China (Table 3). The concentrations of Hg in TY soils were found to be close to the concentrations of soils from Nanjing and Wuhan (Fang et al., 2002; Wang et al., 2005), which were near the Hunan Province, and had similar natural conditions. TY also had the similar soil Hg concentrations with Belgium in the agricultural soils (Tack et al., 2005). The mean concentrations in FQ were nearly equal to the mean Hg background concentration of China and the concentrations in agricultural soil of Canada (Reimann and Caritat, 1998). The results of comparison confirmed that total Hg concentrations were characterized with high variability in different locations.

5. Conclusions

The concentrations of soil Hg differed a lot among the three agricultural experimental stations with long-term fertilization practices. The order of the Hg concentration of A horizon in the three stations was: QY (0.539 ± 0.055) > TY (0.196 ± 0.036) > FQ (0.037 ± 0.014). The effects on the concentration and distribution of Hg in the soils mainly came from the parent materials. The application of phosphorous fertilizers would influence, to some extent, the concentration of Hg in the surface soils with lower Hg background level. The application of the other chemical fertilizers and organic manure could not result in significant changes of soil Hg concentrations in the three experimental stations with long-term fertilization practice.

Since the data sets were obtained from only three experimental sites, further investigation would be necessary to confirm the factors affecting Hg concentrations in the soils from different agro-ecosystems. Moreover, it is essential to address the speciation and bioavailability of Hg in the soils for assessing its food and health risks.

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