



## Effect of organic matter and pH on mercury release from soils

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### Abstract

An investigation was conducted on the effect of organic matter (OM) and pH on mercury (Hg) release from soils. Hg release flux was measured using the dynamic flux chamber (DFC) combined with the Lumex<sup>®</sup> multifunctional mercury analyzer in both laboratory experiment and field monitoring. The results showed that Hg emission from the OM-added soils was apparently low because of the high affinity of OM to Hg, resulting in the reverse order as the amount of OM addition. Meanwhile, Hg release flux from different pH value soils exhibited the same trend for both Hg<sup>2+</sup> and Hg<sub>2</sub><sup>2+</sup> treatment, increasing the Hg flux with pH value of soils increasing. The trend of Hg release in the pH dependence experiment has been well in agreement with that from the field test. In addition, Hg release seemed to be related to its species in the soil, the flux from Hg<sup>2+</sup>-added soil was obviously higher than that of Hg<sub>2</sub><sup>2+</sup>-added soil by the laboratory experiment.

**Key words:** mercury; organic matter; pH; release flux

### Introduction

In recent years, a great deal of research has been devoted to improving the understanding of atmospheric mercury on local, regional and global scales (Schroeder and Munthe, 1998; Kim and Kim, 2001; Nadim *et al.*, 2001; Pirrone *et al.*, 2001; Ebinghaus *et al.*, 2002; Kellerhals *et al.*, 2003; Kock *et al.*, 2005; Wang *et al.*, 2006b; Hall *et al.*, 2006). As a consequence of natural and anthropogenic sources, Hg emission from terrestrial systems has been gradually elevated after the Industrial Revolution (Mason *et al.*, 1994; Slemr and Langer, 1992). Thereinto, Hg emission from soils is considered to be the major contributor of global atmospheric Hg inventory (Schroeder *et al.*, 1989; Kim *et al.*, 1995; Ericksen *et al.*, 2006). According to available literature, evasion of Hg from soils dominantly acts as a function of many multiple and interacting factors, mainly including the climate parameters (containing air/soil temperature, solar radiation, relative humidity, wind speed and so on), soil properties (including organic matter (OM), pH cation exchange capacity (CEC), etc.), Hg content and speciation in the substrate (Carpi and Lindberg, 1997; Poissant *et al.*, 1999; Boudala *et al.*, 2000; Gustin *et al.*, 2002; Scholtz *et al.*, 2003; Wang *et al.*, 2006a). In contrast, the OM concentration and soil pH values have been demonstrated to be two of the most important factors which may significantly influence the Hg geochemistry under

various conditions, including soil transport, transformation processes, and also exchange/release flux via the interface (Semu and Singh, 1987; Meili, 1991; Wallschläger *et al.*, 1998a). Previous studies have shown that Hg<sup>2+</sup> was the predominant form in the soil which was mainly associated with OM because of its high affinity to Hg through the different functional groups such as hydroxy carboxylic, aromatic acids and S-containing ligands. Additionally, there were studies reporting that the effect of the soil pH values on the processes of Hg in the soil was dependent on a series of influence on soil natures owing to the change of soil pH values (Semu and Singh, 1987; Wallschläger *et al.*, 1998b).

Despite many investigations on the subject, suggesting that the OM content and pH value are two of the most important factors which affect Hg behaviors in the soil or through the soil/air interface, the embedded Hg reaction in the soil is still not clear. Furthermore, to our knowledge, few studies are available on the relation between the soil properties (especially for OM and pH value) and Hg release flux at present. In order to develop a more detailed knowledge about these processes, it is essential to make a thorough study on the mechanism of mercury release from soils.

## 1 Materials and methods

### 1.1 Experimental design

Three types of purple soils, Dystric Purpli-Udic Cambosols, Typic Purpli-Udic Cambosols and Calcaric Purpli-

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Udic Cambosols (including two kinds of Dystric Purpl-Udic Cambosols, denoted as DPUC1, DPUC2, TPUC, CPUC, respectively, throughout the text), mainly cultivated soils in Chongqing, China, were collected to investigate the impact of the OM and pH value of soils on the Hg release from soils. One type of the soil, the TPUC, was chosen to investigate the effect of OM on the Hg flux from or to the soil. Ten kilogram of the soil was weighed, in 10 replicates for two parallel groups, each of which added a certain amount of OM from commercialized organic manure, which OM content is 56%, in order that its concentration may be increased by 0, 5, 10, 15, and 20 g/kg (given the initial OM content of 16.05 g/kg). Half a month later, certain  $\text{Hg}_2(\text{NO}_3)_2$  and  $\text{Hg}(\text{NO}_3)_2$  solutions were added to the soil, respectively, until the Hg content of the soil was increased by 1 mg/kg.

The three types of soils (DPUC1, DPUC2 and TPUC) with similar soil properties but different pH values were chosen to observe the influence of pH on the Hg release flux. Ten kilogram of the soil was weighed, in three replicates for each type of soils, to two of which were added certain  $\text{Hg}_2(\text{NO}_3)_2$  and  $\text{Hg}(\text{NO}_3)_2$  solution, respectively, so that the Hg concentration in the soil was increased by 1 mg/kg, the same as mentioned earlier, and the third one made no treatment as control check (CK). All the treatments of soils for simulated test were placed in a plastic box with dimensions of 0.6 m × 0.4 m × 0.2 m. The Hg exchange through the soil-air surface was measured everyday along with the environmental parameters (including solar radiation, air pressure, air-soil temperature, relative humidity, wind direction and speed) half a month later after adding the Hg solution. The work was ended until the Hg flux was almost unchanged for a few days. The measurement of field test at the four sites was conducted during the laboratory experiment campaign.

## 1.2 Measurement methods

Hg release flux from and to the soils was measured using a rectangular dynamic flux chamber (DFC) (Fig.1) with dimensions of 0.4 m × 0.2 m × 0.2 m, resulting in a covered surface area of 0.08 m<sup>2</sup> and a volume of 16 L. The DFC method was described in detail and was widely used in earlier studies (Xiao *et al.*, 1991; Ferrara and Mazzolai, 1998; Carpi and Lindberg, 1998; Engle *et al.*, 2001; Zhang *et al.*, 2002; Gabriel *et al.*, 2006). The chamber for the current study comprised of plexiglas, which was chosen as the construction material because it was cheap, inert and clear, and because of its hard characteristics which permitted the transmission of visible radiation and easily achieved low blank of the chamber. The chamber

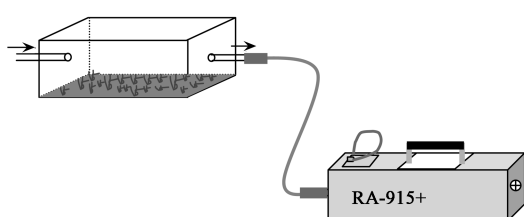


Fig. 1 Schematic diagram of the DFC operation system.

was linked through the outlet with an automatic mercury analyzer by semi-transparent Teflon tube. The total gaseous mercury (TGM) concentrations were determined by cold vapor atomic absorption spectrometry (CVAAS) using the Lumex<sup>®</sup> multifunctional mercury analyzer RA-915+ (Lumex Ltd., Russia). Both inlet and outlet TGM concentrations were measured alternatively to get a 10-min average over 5 min intervals. Twenty minute average flux values were obtained by the following equation:

$$F = \frac{(C_o - C_i)Q}{A} \quad (1)$$

where,  $F$  is the Hg release flux (ng/(m<sup>2</sup>·h)),  $C_i$  and  $C_o$  are the TGM concentrations of the inlet and outlet (ng/m<sup>3</sup>), respectively,  $Q$  is the flow rate into chamber (1.2 m<sup>3</sup>/h in the present study) and  $A$  is the enclosed emission area (m<sup>2</sup>). Positive flux values indicate Hg emission from the soil to air, whereas, negative one represents Hg deposition to the soil from air. Fig.1 schematically illustrates a typical setup for the DFC operation. To determine the potential for contamination/accumulation of the system, the chamber blanks were routinely determined before and after collecting the soil flux measurements by sealing the chamber bottom to a clean plexiglas sheet, resulting in an average of 1.3±0.2 ng/(m<sup>2</sup>·h) during the whole campaign. The soil temperature in surface layer (2–3 cm) was measured with a soil temperature meter and solar radiation determined with a TES-1332A Digital Lux meter (TES, Taiwan, China). Other environmental parameters (including air temperature, air humidity, air pressure, wind speed and direction) were also monitored with Kestrel<sup>®</sup> 4000 Pocket Weather tracker (Nielsen-Kellerman, USA).

## 2 Results and discussion

### 2.1 Effect of organic matter on Hg release from soils

For both the treatments with the addition of  $\text{Hg}_2^{2+}$  and  $\text{Hg}^{2+}$  solution, there was a significant reduction in release flux of Hg through the soil-air interface with increasing OM content added into the soil based on our experiment, as indicated in Fig.2. Additionally, it was evidently shown that the flux from the CK soil was higher than that observed from the other OM-added soils by comparing with the

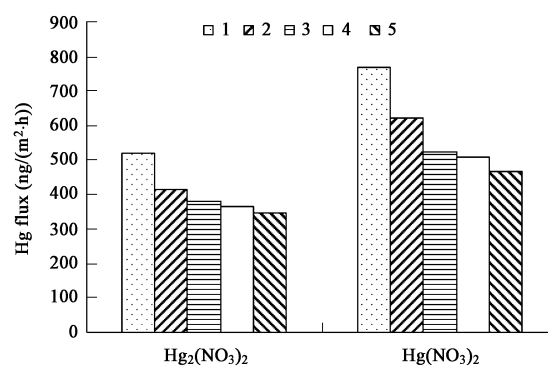


Fig. 2 Mean Hg flux of the whole campaign from the OM-added soils. 1, 2, 3, 4, and 5 represent the added amounts of OM 0, 5, 10, 15, and 20 g/kg, respectively.

mean flux. It can be partially explained that the OM in the soil absorbed a large of Hg with extremely high affinity, suggesting low availability/mobility (Semu and Singh, 1987; Skyllberg *et al.*, 2000). As added into the soil, the OM could also, to a certain extent, change the soil characteristics such as the amount of absorption sites and surface electricity which play an important role in the absorption of heavy metals in the soil. As a result of the strong binding of Hg to OM, the reduction processes of oxidized Hg were restrained significantly resulting in the decrease in Hg flux from the soils.

The time series of Hg flux from the OM-added soils is illustrated in Fig.3. Hg flux for all the treatment soils fluctuated drastically during the measurement period, as the variance coefficients were fairly high, nearly surpassing 100%. The great variation day by day can be mainly attributed to the reason that the climatic condition varied greatly during the whole campaign, which played an extraordinarily important role in the Hg evasion from the soil by impacting the formation and the reduction of oxidized Hg. This was in agreement with the results of several previous studies, suggesting that evasion of elemental Hg from the soils was primarily influenced by the environmental parameter, such as solar radiation, air temperature, relative humidity and certain other synergetic factors (Gillis and Miller, 2000). Therefore, because of a sharp decrease in the amount of elemental Hg forming on a cloudy or rainy day, the Hg release from the soils deeply declined. As can be seen in Fig.3, the Hg release evidently decreased on the fourth or fifth day for all treatments, resulting in a low site because of the rainy weather. At the earlier experiment period, the Hg release flux differentiated extremely and fluctuated greatly as is evident in Fig.3. However, there was a general trend of the Hg evasion flux decreasing as time went by. Fifty days later, the negative Hg flux intermittently took place, in other words, the Hg deposition from the air to soil happened. After 73 d, all the flux from different treated soils became less variable and finally reached a stable state.

## 2.2 Effect of pH on Hg release from the soils

It is well established that pH value is important in impacting the mobility and availability of the heavy metal (including Hg) in the soil, not only by the effect on the metal speciation in the soil solution but also the changing of soil characteristics (Barrow and Cox, 1992; Yin *et al.*,

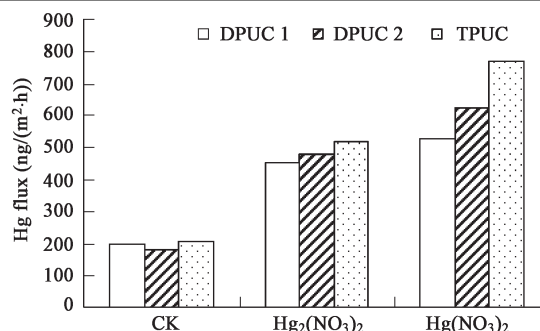


Fig. 4 Mean Hg flux from the soils with different pH values. DPUC1, DPUC2, and TPUC are three types of soils, the pH values of which are 4.82, 6.20, and 7.11, respectively.

1996). Fig.4 presents the mean Hg flux from the soils with different pH values. For the CK treatment there was slight difference between each other and the flux can be shown in the order of TPUC > DPUC 1 > DPUC 2. After the addition of certain amounts of Hg<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> and Hg(NO<sub>3</sub>)<sub>2</sub> solution, there was an evident increase in Hg flux. And both the Hg solution addition treatments exhibited the same order of mean Hg exchange flux: TPUC > DPUC 2 > DPUC 1. In other words, the flux emission performed the same sequence as the pH value of soils. However, it must be pointed out here that because of the existence of a slight difference in the other aspects of soil properties, the no-significant variety in Hg flux cannot be interpreted just by the difference between the pH values. Therefore, more detailed research is needed to comprehensively understand the Hg flux as a function of the soil pH values.

Large variability of Hg flux was observed as shown in Fig.5. The variance coefficients in all the treatments were quite large, nearly the same as the results of OM experiment. The above-mentioned mechanism of multiple interacting factors of environmental parameters on the release flux, therefore, could be also the primary explanation to the identical fact.

## 2.3 Effect of Hg species on release from soils

The results from both the soils with OM added and various pH had the same phenomenon. The significant difference in soil Hg flux was observed between the two kinds of addition of Hg<sup>2+</sup> and Hg<sub>2</sub><sup>2+</sup>, and the former was much higher. The percentage increased (Fig.6) shows the results of pH dependence experiment. It is well known that Hg<sub>2</sub><sup>2+</sup> hardly change based on the fact for  $E^0(\text{Hg}^{2+}/\text{Hg}_2^{2+})$

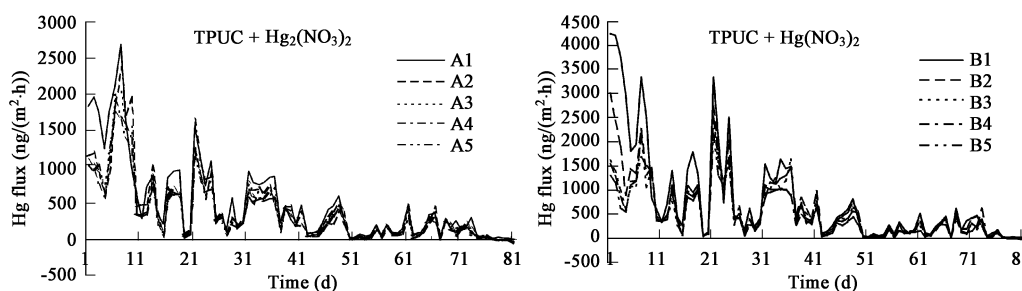


Fig. 3 Time series of Hg flux from the OM-added soil. A and B represent Hg<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> and Hg(NO<sub>3</sub>)<sub>2</sub> added soil and 1, 2, 3, 4, and 5 represent the added amount of OM 0, 5, 10, 15, and 20 g/kg, respectively. TPUC is one type of soils.

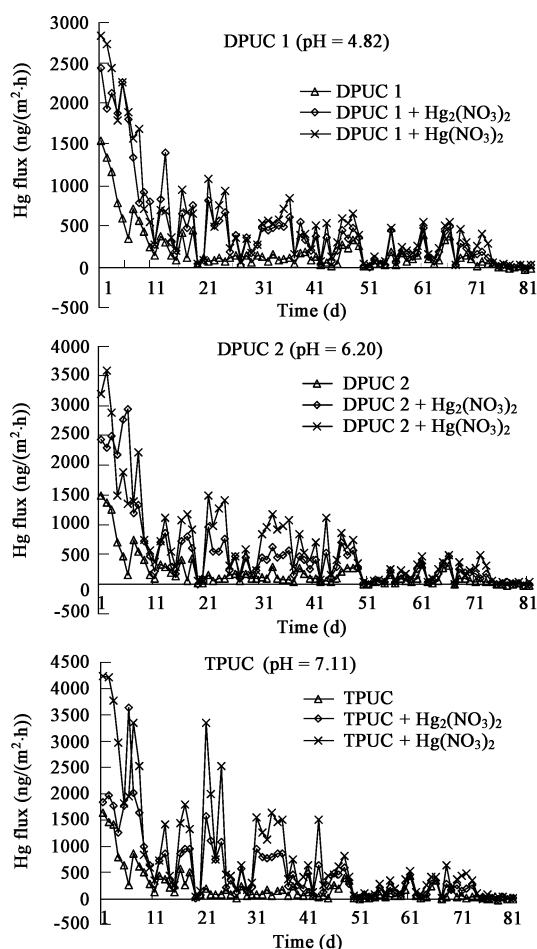


Fig. 5 Time series of Hg flux from the soils with different pH values.

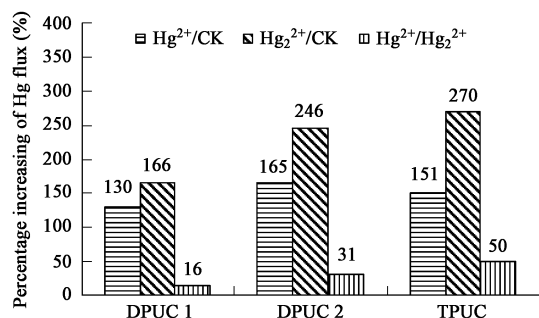


Fig. 6 Percentage increasing of Hg flux in addition of  $\text{Hg}^{2+}$  and  $\text{Hg}_2^{2+}$ .  $\text{Hg}^{2+}/\text{Hg}_2^{2+}$  denotes the increasing flux ration from the  $\text{Hg}^{2+}$ -added soil and  $\text{Hg}_2^{2+}$ -added soil, the same as the other two outlines.

$> E^0(\text{Hg}_2^{2+}/\text{Hg})$ . On the other hand, most of the  $\text{Hg}_2^{2+}$  was changed into the insoluble state combining with other substances once added to the soils, because most of

$\text{Hg}_2^{2+}$  compounds were of low solubility. Therefore, the reduction process of oxidized Hg was seriously suppressed and thus decreased the formation of elemental Hg in the soil. As a result, there was an obvious decrease in the Hg flux obtained in the experiment.

## 2.4 Results of field monitoring

The flux results from field test were well in agreement with that of the pH laboratory experiment. Hg flux from different monitoring sites increased with the increasing pH values of soils. Four sites, three from the monitoring base and one as the polluted site, were chosen to obtain flux measurement from a range of pH values with similar soil properties. Long time of measurement was implemented and the related data is summarized in Table 1. Fig. 7 further illustrates the mean flux during the field campaign. It can be observed that the difference among the measurement sites was not significant as we expected, except that the contaminated soil site quite elevated and proximate 2 or 3 times higher than those found in the other sites. The greater flux of the polluted site was mainly because of the high concentration of Hg in the soil (approximately 32 times as great as that of the base sites), indicating that the factor of Hg content was quite important in mercury release from the soils (Gustin *et al.*, 2000). These high values should be considered as normal to such an intense and prolonged laying of the mercury trash.

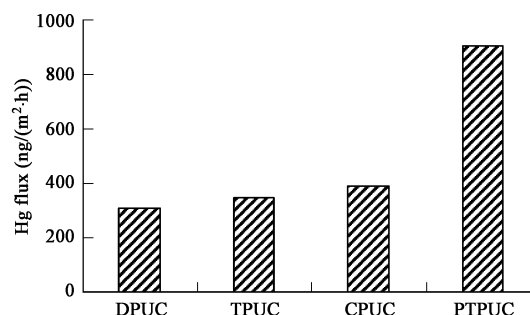


Fig. 7 Mean Hg flux of the measurement sites in the field test.

## 3 Conclusions

On the basis of the large quantity of Hg flux measurement from a variety of soils in both the laboratory experiment and field test campaign, we determined that the OM and pH value were two of the most important soil factors affecting the Hg emission from soils. Hg emission from the OM-added soils was apparently suppressed,

Table 1 Results of Hg flux measurement in field test (ng/(m<sup>2</sup>·h))

Date (m-d)	DPUC (pH=5.11)		TPUC (pH=6.87)		CPUC (pH=8.15)		PTPUC (pH=6.91)	
	Range	Mean	Range	Mean	Range	Mean	Range	Mean
04-23	69–864	407.3	99–834	434.0	159–894	495.7	534–2484	1437.3
10-10	144–534	294.0	114–714	380.3	159–804	425.3	144–489	286.5
11-05	99–504	285.0	84–714	372.0	114–834	429.0	339–2184	1407.0
11-25	309–714	410.3	279–654	432.8	219–624	421.5	234–1029	635.3
12-16	–6–249	72.0	–6–219	81.0	24–219	105.0	99–234	165.0

PTPUC represents the monitoring site 4 for the field test—the soil of polluted TPUC.

mainly as a consequence of the Hg binding to OM with high affinity, thus decreasing its availability and mobility which were quite important for the formation of elemental Hg from the reduction of oxidized mercury. Finally, there was the obvious decrease in the Hg flux as compared to the CK treatment. On the other hand, Hg flux from different pH value soils with similar properties exhibited the same increase trend in the Hg flux with increasing pH value of soils. The trend of Hg release flux in the laboratory experiment has been well in agreement with that from field test. Because of the high content of Hg in the soil, emission flux from polluted measurement site was approximately 2 or 3 times higher than those found in the state-monitoring base sites. In addition, Hg release seemed to be related to its species in soil, the flux from the Hg<sup>2+</sup>-added soils was obviously higher than that from Hg<sub>2</sub><sup>2+</sup>-added soils.

Although our experiment is relatively systemic and precise, it cannot comprehensively account for the impact mechanism of soil properties of OM and pH on Hg release. Thus, it is important to keep in mind that more detailed research is needed to further elucidate the Hg reaction processes under the soil condition, to find out the detailed impacting mechanism of soil properties on Hg emission then lessen the negative impact on atmospheric mercury.

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