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Atmospheric deposition of mercury and cadmium impacts on topsoil in a typical coal mine city, Lianyuan, China



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Jie Liang ^{a, b, *}, Chunting Feng ^{a, b}, Guangming Zeng ^{a, b, **}, Minzhou Zhong ^{a, b}, Xiang Gao ^{a, b}, Xiaodong Li ^{a, b}, Xinyue He ^{a, b}, Xin Li ^{a, b}, Yilong Fang ^{a, b}, Dan Mo ^{a, b}

^a College of Environmental Science and Engineering, Hunan University, Changsha, 410082, PR China

^b Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha, 410082, PR China

HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- Investigating atmospheric deposition fluxes of Hg and Cd in a typical coal mine city.
- Hg and Cd pollution was serious in surface soils.
- Atmospheric Hg and Cd deposition fluxes were positive correlated with soil Hg and Cd concentrations.
- It is predicted that the increment of Hg and Cd contents in topsoil was caused by atmospheric deposition.

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ABSTRACT

Mercury (Hg) and cadmium (Cd) in the atmosphere from coal combustion emissions play an important role in soil pollution. Therefore, the purposes of this study were to quantitatively evaluate the atmospheric Hg and Cd deposition and to determine the influence of atmospheric deposition on Hg and Cd contents in surface soil in a typical coal mine city. Atmospheric deposition samples were collected from May 2015 to May 2016 at 17 sites located in industrial, agricultural and forest areas in the Lianyuan city. Atmospheric Hg and Cd deposition fluxes in the different land use types showed high variability. Curvilinear regression analysis suggested that the atmospheric Hg deposition fluxes were positively related with Hg contents in soils ($R^2 = 0.86359$, P < 0.001). In addition, atmospheric Cd deposition fluxes were also positively correlated with Cd contents in soils when the site LY02, LY04 and LY05 (all belong to agricultural land) were not included in the fitting ($R^2 = 0.82458$, P < 0.001). When they were included, there was no significant relationship between them ($R^2 = 0.2039$, P = 0.05). The accumulation of Hg and Cd concentration in topsoil due to the influence of atmospheric deposition will increase rapidly in the next 30 years, and the mean value of the increment will reach 2.6007 and 33.344 mg kg⁻¹. After 30 years, the Hg and Cd concentration will increase slowly. The present study advocates that much attention should be paid to the potential ecological hazards in soil resulting from the atmospheric Hg and Cd deposition.

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1. Introduction

Hg and Cd are very significant environmental contaminants due

^{*} Corresponding author. College of Environmental Science and Engineering, Hunan University, Changsha, 410082, PR China.

^{**} Corresponding author. College of Environmental Science and Engineering, Hunan University, Changsha, 410082, PR China.

E-mail addresses: liangjie@hnu.edu.cn (J. Liang), zgming@hnu.edu.cn (G. Zeng).

199

to their high toxicity and bioaccumulation (Keeler et al., 2006). Hg can be methylated into highly toxic methylmercury under certain environmental conditions. Methylmercury is easy to accumulate in the body through the food chain, resulting in a series of diseases (Liu et al., 2015). Cd is extremely toxic to animal even at low contents and it can be absorbed into the body through respiration and stored in the liver or kidneys (Liang et al., 2016). At present, 2000–4000 ton year⁻¹ of Hg from human activities is emitted into the atmosphere (Kim and Kim, 1999). Atmospheric dry and wet deposition is commonly regarded as one of the main sources of Hg and Cd in soils (Zhu et al., 2016). Dry deposition of metal occurs by direct collision and gravitational settling on the ground. In wet deposition, aerosols and gases are dissolved or suspended in water droplets or ice crystals and eventually enter into the land (Chance et al., 2015; Soriano et al., 2012). It is reported that sources and loading of heavy metals in the atmosphere have great spatial variability over different sites, which are due to different meteorological conditions and the emission patterns of contaminants (Sakata and Takagi, 2008). Hg and Cd in the atmosphere are related primarily with industrial activities, e.g., coal mining, coal combustion, smelting, chemical manufacturing process (Rossini et al., 2005; Tang et al., 2015). They can be transported for long distances away from their pollution-creating source and their concentrations decrease as transport distance increase. More importantly, Hg and Cd ultimately entered into topsoil via atmospheric deposition (Connan et al., 2013). Heavy metal (Hg and Cd) contamination of soil has always been a global environmental issue. Not only can they change the physical and chemical properties of soils, but also they do harm to plants, animals and human, by altering their metabolism (Nagajyoti et al., 2010). For instance, Pandey and Pandey (2009) reported that long-term atmospheric Hg and Cd deposition will cause a devastating effect on soil fertility and is the major contributor to raise Hg and Cd in edible parts. A study has revealed that exposure of root corns to atmospheric Hg and Cd depositions near industrial district heighten increment of Hg and Cd in the leaves (De et al., 2012). Although plenty of countries make efforts to decrease emissions of atmospheric contaminants, Hg and Cd concentrations in the atmosphere are always considered at risk owing to long-term industrial activities (Bian et al., 2015).

Hg in coal is emitted to the atmosphere during mining and burning and is deposited on the topsoil. China is the most important consumer of coal in the world (Martín and Nanos, 2016). Particularly, there are numerous small-scale coal mines in the Lianyuan city. Cd in the atmosphere mainly originates from fossil fuel burning and manufacturing processes of metals (Wu et al., 2013). In the past few decades, industrialization and urbanization have observably increased atmospheric Hg and Cd concentrations in the Lianyuan city. And once absorbed into soils, Hg and Cd stay a long time and remain an environmental threat especially to Children (Soriano et al., 2012). Consequently, it is very necessary to assess the impact of atmospheric deposition fluxes of Hg and Cd on their contents in soils.

Some previous studies have reported that atmospheric deposition fluxes of Hg and Cd at rural or industrial areas (Azimi et al., 2005; Bi et al., 2006; Kim et al., 2012; Sakata and Takagi, 2008), but few researches have been investigated at agricultural land, forest land and industrial areas together. Furthermore, most studies only considered the relationship between seasonal variation or wind speed and atmospheric deposition fluxes of Hg and Cd (Castillo et al., 2013; Liang et al., 2015, 2017b; Percot et al., 2016; Sharma et al., 2008; Wu et al., 2008), few conclusions have been reported about the relationship between atmospheric Hg and Cd deposition fluxes and their contents in soils. What's more, there are much fewer studies on the prediction of Hg and Cd concentrations increment in surface soils caused by atmospheric deposition. Therefore, the main objectives of this study were: (1) to determine the atmospheric deposition fluxes of Hg and Cd at different sites; (2) to assess the levels of Hg and Cd in surface soils; (3) to elucidate associations between atmospheric Hg and Cd deposition fluxes and their contents in soils; and (4) to predict increment of Hg and Cd contents in topsoil due to the impact of the atmospheric deposition.

2. Materials and methods

2.1. Study areas

The study region is located in Lianyuan (27°27′-28°2′N, 111°33′-112°2′E), in the south-central China. The area has a population of about 1 million and covers 1897 km². The climate type is midsubtropical humid monsoon climate, with an annual mean temperature of 16–17.3 °C and rainfall of 1406 mm. The Lianyuan city is under a comprehensive land uses, including agricultural land, forest area, small reservoir and industrial district. Furthermore, the industrialization of this area is very serious and the main industries are iron and steel smelting, chemical industry, coal mining, machine manufacturing and so on. Moreover, the Lianyuan city has numerous small-scale coal mines.

2.2. Sampling and analysis

Atmospheric deposition samples were collected from May 2015 to May 2016 at 17 sites in the Lianyuan city. The distribution of sampling points was presented in Fig. 1 and the detailed information of the 17 sites was shown in Table S1 (Supplementary material). The sample collector is a dust collecting cylinder, with an inradius and height of 40 cm and 60 cm, respectively. It is composed of glass-ceramic. Before sampling, the dust collecting cylinders were soaked with 10% HCl for 24 h and then rinsed with distilled water. These dust collecting cylinders were generally placed 10 m–15 m above the ground and the sample collection period was set at one year.

After sampling, the samples were placed for 2 d–3 d to clear the upper solution. Then the upper solution was transferred to another container using siphon method. 10 mL of 8 mol L⁻¹ HNO₃ and 5 mL of 5% K₂CrO₇ were added to the upper solution for Cd and Hg measurement, respectively. And these samples were stored in refrigerator for later analysis. The precipitate and suspension samples were filtered through 0.45µm polyester fiber membrane filters and the filters were dried under 65 °C. Then they were weighted at room temperature for further determination of elements.

All solution samples were digested with 1% HNO₃. Hg content was measured by atomic fluorescence spectrometry (AFS) (Reis et al., 2003) and Cd concentration was determined by inductively coupled plasma mass spectrometer (ICP-MS) (Wang et al., 2005). Procedural blanks and standard reference materials (obtained from the Center of National Reference Materials of China) were applied to quality assurance and quality control. Recovery values ranged from 80% to 91%. Blank samples were measured in each batch of samples. The relative standard deviations (RSD) were <10%.

2.3. Calculations and statistical analyses

Atmospheric deposition fluxes of Hg and Cd were calculated by the following formula.

$$Q = Q_d / S + Q_w / S \tag{1}$$

where $Q(\text{mg m}^{-2} \text{ a}^{-1})$ is annual atmospheric deposition flux of Hg or Cd; Q_d (mg a⁻¹) is weight of annual atmospheric dry deposition



Fig. 1. Location of study area and distribution of sampling sites.

of Hg or Cd; Q_w (mg a⁻¹) is weight of annual atmospheric wet deposition of Hg or Cd; and S (m²) is area of the sampling collector.

Prior to geostatistical analyses, the data were logarithmically transformed and the Shapiro-Wilk test was used to verify normal distribution. The spatial distributions of atmospheric Hg and Cd deposition fluxes were established using Kriging interpolation in ArcGIS 9.3.

The sample data for 50 soil Hg and Cd concentrations around each atmospheric deposition sample site were derived from our previous study (Liang et al., 2017a). Curvilinear regression (exponential model) was applied to assess the association between atmospheric Hg and Cd deposition fluxes and soil Hg and Cd concentrations.

According to the technical principles and methods for environmental impact assessment (HJ 2.1–2011), the accumulation of soil Hg and Cd after n years due to the impact of the atmospheric deposition was predicted. The formula was shown as following:

$$C_n = BK^n + EK \frac{1 - K^n}{1 - K} \tag{2}$$

where C_n (mg kg⁻¹) is the accumulation of Hg and Cd in soils after n

Table 1	
The annual atmospheric deposition fluxes (mg $m^{-2} a^{-1}$) of Hg and Cd at different sites worldwide.

Site	Years	Location	No. of sites	Hg	Cd	Reference
The San Jorge river basin, Colombia	2010-2011	Urban	1	0.07	0.43	(Marrugo-Negrete et al., 2015)
Marais Vernier, France	2010-2012	Rural area	1	0.009	0.009	(Connan et al., 2013)
the San Jorge river basin, Colombia	2010-2011	Coal mining area	1	0.77	1.91	(Marrugo-Negrete et al., 2015)
City of Huelva, Spain	2008-2011	Industrial states	3	_	0.60	(Castillo et al., 2013)
The Lianyuan city, China	2015-2016	Urban, rural and industrial area	17	0.17	1.70	This study

years; $B \pmod{\text{kg}^{-1}}$ is soil background value of Hg and Cd; $E \pmod{\text{kg}^{-1}}$ is concentration of annual atmospheric Hg and Cd deposition; and K (%) is the residual rate of Hg and Cd in soils.

Because Hg and Cd are difficult to migrate in soils, the residual rate (K) is usually 90% (Li et al., 2014).

3. Results and discussion

3.1. Atmospheric deposition fluxes of Hg and Cd at different sites

The annual atmospheric deposition fluxes of Hg and Cd at 17 sampling sites were shown in Table S1 as supplementary material. The average values of the atmospheric Hg and Cd deposition fluxes were 0.17 mg m⁻² a⁻¹ and 1.70 mg m⁻² a⁻¹, respectively, with variations ranging from 0.042 to 1.02 mg m⁻² a⁻¹ for Hg and 0.47–14.37 mg m⁻² a⁻¹ for Cd. The atmospheric deposition fluxes of Hg and Cd in this study was compared with data from the literature in different sites such as urban, rural and industrial areas, which were presented in Table 1. Results indicated that atmospheric deposition fluxes of Hg and Cd were highest in a coal mining area and much lower in the rural region.

Fig. 2 presented the spatial variations of atmospheric Hg and Cd

deposition fluxes. The highest Hg deposition fluxes were found at site LY15 and LY16, which may be closely related to numerous small-scale coal mines near the two sampling sites. Feng et al. (2002) reported that Hg deposition flux was 0.0468 mg m⁻² a^{-1} in coal mining regions in Guizhou, China. It was reported that coal mining and combustion were the main sources of atmospheric pollution (Dutt et al., 2009). At site LY01, LY02, LY03, LY05 and LY13, lower deposition fluxes of Hg were observed. These sites were mainly located in farmland and woodland, which were moved away from some coal mine and industrial area. Nóvoa-Muñoz et al. (2008) research has shown that the industrial area had higher atmospheric Hg deposition fluxes than agricultural regions due to coal burning. As can be observed in Fig. 2, atmospheric Hg deposition fluxes increased gradually from the northwest to the southeast, which was consistent with the distribution of small-scale coal mines in the Lianyuan city.

The spatial distribution of atmospheric Cd deposition fluxes was lower in the central part and higher in east-west direction (Fig. 2). Specifically, Cd deposition flux was highest in site LY15, where chemical plants and iron and steel smelting industries were concentrated. In addition, relatively higher Cd deposition fluxes were found in site LY06, LY08, LY10 and LY17, probably as a result of



Fig. 2. Spatial variation of atmospheric Hg and Cd deposition fluxes.

waste gas emitted by non-ferrous metal smelting and chemical industries. This conclusion is in agreement with Maas et al. (2010), who concluded that Cd emission from chemical and smelting industries caused serious pollution to the environment. Marrugo-Negrete et al. (2015) reported that atmospheric Cd deposition flux reached 2.31 mg m⁻² a⁻¹ near the ferronickel factory and 3.23 mg m⁻² a⁻¹ around the gold mining industry in Colombia, which was higher than our investigation in 16 sites except site LY15 in the Lianyuan city. This may be because sampling sites moved closer to the industrial activities in their research area and the ferronickel and gold mining activities can emit higher concentrations of Cd into the atmosphere. Moreover, Marrugo-Negrete et al. (2015) also reported that Cd deposition flux was 1.91 mg m⁻² a⁻¹ near the coal mining region, which was consistent with mean value of Cd deposition flux in this study. It was indicated that coal combustion related to chemical and metallurgical activities was the main source of Cd pollution in the atmosphere in the Lianyuan city. At site LY05, the lowest Cd deposition flux was found and at site LY02, LY03, LY04 and LY12 were also relatively lower, which was caused by not typical industrial activity around these sites. This consequence was similar to the research by Cong et al. (2010), who reported that Cd deposition flux was 0.0018 mg m⁻² a⁻¹ in rural areas far from industrial sources in Nam Co. This tangibly demonstrated that sites with lower atmospheric Cd deposition fluxes stay far away from the industrial activities.

3.2. Levels of Hg and Cd in surface soils

Box-and-whisker plot for Hg and Cd concentrations in surface soils near the 17 sampling sites of atmospheric deposition was shown in Fig. 3. Concentrations of Hg in 17 different sites represent high variability. The highest content was found in site LY11,





followed by site LY15, LY09, LY12 and LY16. The lower concentrations were found in sites LY01, LY02, LY06 and LY08. Fig. S1 (supplementary material) presented the spatial distribution of Hg and Cd concentrations in topsoil. The spatial variation of Hg concentration in soils was almost consistent with the spatial variation of atmospheric Hg deposition flux, indicating atmospheric deposition was the main source of Hg in soils. Pacyna et al. (2006) reported that 45% of Hg in soils came from the atmospheric deposition related to coal combustion.

From Fig. 3, the difference of Cd concentration in the 17 sites was not significant compared with Hg. Relatively high Cd content was observed at site LY02, LY04 and LY05, but the atmospheric Cd deposition fluxes were lower at these sites. Depending on our previous study, Cd in soils originated from agricultural practices, such as use of phosphate fertilizers and pesticides in the Lianyuan city (Liang et al., 2017a). As was shown in Fig. S1, high content of Cd was found around site LY06, LY10 and LY15, which were related to long-term industrial activities, such as chemical manufacturing, metal smelting and coal mining. There were also higher Cd deposition fluxes around the three sites, indicating that atmospheric Cd deposition caused the increase of Cd concentrations in soils. In summary, there was a relationship between atmospheric Hg and Cd deposition and the content of Hg and Cd in soils. So, further research would be needed.

3.3. Relationships between atmospheric Hg and Cd deposition fluxes and their contents in soils

From Fig. 4, the atmospheric Hg deposition fluxes were positively associated with Hg contents in soils ($R^2 = 0.86359$, P < 0.001). This result was consistent with the analyses of previous two sections. Hg from industrial activities (such as coal combustion) was discharged into the atmosphere and ultimately entered into soils through atmospheric deposition, interfering with biochemical processes of plants. In addition, atmospheric Cd deposition fluxes were also positively correlated with Cd contents in soils when the site LY02, LY04 and LY05 (all belong to agricultural land) were not included in the fitting ($R^2 = 0.82458$, P < 0.001). When they were included, there was no significant relationship between them $(R^2 = 0.2039, P = 0.05)$. This was because Cd pollution in farmland soil mainly came from agricultural activities, and soil contamination near the industrial area was caused by atmospheric Cd deposition in the Lianyuan city. As can be seen from Fig. 4, atmospheric Hg and Cd deposition increased faster, soil Hg and Cd concentrations also increased rapidly. Therefore, the increments of Hg and Cd concentrations in soils caused by atmospheric deposition should not be neglected.

3.4. Predicated increments of Hg and Cd content in topsoil due to the impact of the atmospheric deposition

The accumulation of Hg and Cd content in surface soil after 1, 10, 30, 50 and 70 years affected by atmospheric deposition was shown in Table S2 as supplementary material. The increments of Hg and Cd concentration in surface soil were the highest at site LY15, which was consistent with the results of annual atmospheric Hg and Cd deposition fluxes. This indicated that Hg and Cd emissions from industrial activities have caused serious pollution to the surrounding soils. The increase of the distance from the industrial area, the degree of soil contamination was gradually reduced. Zhu et al. (2016) reported that the level of atmospheric heavy metal deposition was considered to be higher in industrial belt than in rural area. For instance, in Huelva industrial belt, City of Huelva and Donana Park, the atmospheric Cd deposition fluxes were 0.2, 0.1 and 0.01 mg m⁻² a⁻¹, respectively (Castillo et al., 2013). According

to reports, Hg emissions from coal-fired power plants affected the spatial distribution of Hg concentrations in surface soils through atmospheric deposition (Keeler et al., 2006). In addition, the increments in Hg concentration in the topsoil were closely related to the distance from the coal-burning power. Some studies have demonstrated that accumulation of Hg in topsoil near the coal-fired power plant increased significantly (Engle et al., 2006; Hua et al., 2015; Nóvoa-Muñoz et al., 2008).

At the 17 sampling sites, we selected 4 sites being the highest, higher, middle and lower atmospheric deposition fluxes, and plotted Fig. 5. Fig. 5 showed a change in the concentration increments of soil Hg and Cd resulted from atmospheric deposition over time. From Fig. 5 and Table S2, the mean values of Hg concentration increments in topsoil after 1, 10, 30, 50 and 70 years will reach 0.3343, 1.7913, 2.6007, 2.6991 and 2.7111 mg kg⁻¹, respectively. And the average values of Cd concentration increments in surface soil after 1, 10, 30, 50 and 70 years will reach 3.545, 22.702, 33.344, 34.638 and 34.796 mg kg⁻¹, respectively. What's more, the concentration of Hg and Cd in topsoil will increase rapidly in the next 30 years, and the mean value of the increment will reach 2.6007 and 33.344 mg kg⁻¹ for Hg and Cd, respectively. After 30 years, the concentration of Hg and Cd will increase slowly. As can be seen from Fig. 5, 30 years later, the concentrations of Hg and Cd in



Fig. 4. Relationship between the atmospheric Hg and Cd deposition and the soil Hg and Cd content.



Fig. 5. Predicted increments of Hg and Cd contents in the topsoil after n years caused by atmospheric deposition.

the surface soil will almost no longer increase. From the above analysis, we can infer that Hg and Cd, which are emitted into the atmospheric by human activities, will almost completely deposit into the surrounding soils within the next 30 years. Additionally, the Hg and Cd will remain in soils for a long time, even for thousands of years (Brookes, 1995). Long-term and increasing atmospheric Hg and Cd deposition have a potentially detrimental effect on soil properties. Hg and Cd interfere with plants metabolism through root and leaf uptake. Previous research has shown that for Cd, the root absorption is an important pathway to enter plants even at low concentrations of the soil environment (De et al., 2015). Moreover, Hg and Cd in plants can be transferred into the human body via the food chain, and eventually pose a threat to the health of residents. Pandey et al. (2012) indicated that the consumption of vegetables polluted with Hg and Cd was the main way for human exposure. Consequently, it is necessary to take effective measure to reduce emissions of Hg and Cd in the atmosphere in a typical coal mine city in order to protect human health and achieve sustainable development.

4. Conclusion

An investigation of atmospheric Hg and Cd deposition fluxes at

17 sites in the Lianyuan city has been conducted with the aim to estimate the effects of atmospheric deposition on the contents of Hg and Cd in soils. The results suggested that average atmospheric deposition fluxes of Hg and Cd were 0.17 mg m⁻² a⁻¹ and 1.70 mg $m^{-2} a^{-1}$, respectively, with variations ranging from 0.042 to 1.02 mg $m^{-2}a^{-1}$ for Hg and 0.47–14.37 mg $m^{-2}a^{-1}$ for Cd. Atmospheric Hg and Cd deposition fluxes in the Lianvuan city exhibited an obvious spatial variability, and the content in the industrial area was significantly higher than that in agricultural and forest area. The highest Hg deposition fluxes were observed in site LY15 and LY16, where high density coal mines and coal-fired industrial activities were distributed. The Cd presented higher content in point LY15 near the chemical plants and iron and steel smelting industries. Curvilinear regression analysis suggested that the atmospheric Hg deposition fluxes were positively related with Hg contents in soils $(R^2 = 0.86359, P < 0.001)$. In addition, atmospheric Cd deposition fluxes were also positively correlated with Cd contents in soils when the site LY02, LY04 and LY05 (all belong to agricultural land) were not included in the fitting ($R^2 = 0.82458$, P < 0.001). When they were included, there was no significant relationship between them ($R^2 = 0.2039$, P = 0.05). The increments of Hg and Cd concentration in surface soil were the highest at site LY15, indicating that the air pollution around this site was very serious. The concentration of Hg and Cd in surface soil will increase rapidly in the next 30 years, and the average value of the increment will reach to 2.6007 and 33.344 mg kg⁻¹ for Hg and Cd, respectively. After 30 years, the concentration of Hg and Cd will increase slowly. It is indicated that Hg and Cd, which are emitted into the atmosphere by human activities, will almost completely deposit into the surrounding soils within the next 30 years and remain in soils for thousands of years. Our study investigated the degree of atmospheric Hg and Cd deposition on soil pollution, providing basic information for further research, especially for typical coal mine cities. And these conclusions are useful to control Hg and Cd emissions from coal combustion by some effective measures in order to protect the natural ecosystem.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.09.046.

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