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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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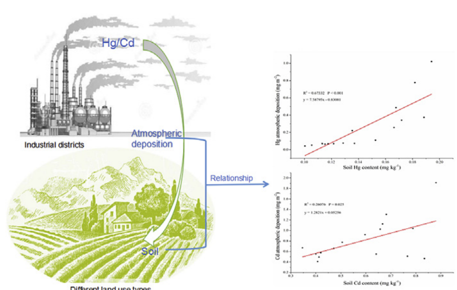
HIGHLIGHTS

- 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.

GRAPHICAL ABSTRACT



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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.

1. Introduction

Hg and Cd are very significant environmental contaminants due

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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 km^2. The climate type is mid-subtropical 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} HNO_3 and 5 mL of 5% K_2CrO_7 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% HNO_3. 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 (mg m^{-2} a^{-1}) is annual atmospheric deposition flux of Hg or Cd; Q_d (mg a^{-1}) is weight of annual atmospheric dry deposition