Spatial distribution of heavy metals in soil around municipal solid waste incinerators at coastal plain and inland mountainous region in China

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Abstract. Heavy metals released from incinerator stacks are considered as the potential sources in soils surrounding the MSWI plants. In this work, heavy metals in soils sampled at different sites around the two MSWI plants with different landforms and weather conditions, namely, windy coastal plain region in Eastern China and calm inland mountainous area in Western China, were investigated. The average concentrations of Cr, Mn, Fe, Cu, Zn and Pb in soils at the coastal plain region were 274.21, 1053.47, 58755.9, 37.38, 90.60 and 40.53 mg/kg, in comparison with those of 224.0, 658.99, 39960.3, 31.72, 33.91 and 70.98 mg/kg in soils at the inland mountain region, respectively. Principal component analysis showed that Mn and Fe were presented as background origin in soils surrounding both the incinerators. Cu, Zn and Pb distributions in the two studied regions were influenced by human activities. The application of pesticides contributed to Cu and Zn contaminations in soils surrounding the coast plain incinerator. Furthermore, the application of pesticides and local traffic had impacts on Cu distributions in soils surrounding the inland mountain incineration plant. The study highlighted that Pb was an important environmental indicator of MSWI air contamination. The source identification and spatial distributions of Pb indicated that the extent of soil contamination by heavy metals in the vicinity of a MSWI was highly ascribed to the stack emission. Furthermore, the terrain and meteorological conditions had a significant effect on heavy metal pollution distributions.

1. Introduction

Incineration is a common technique for treating municipal solid waste (MSW) in many countries. China currently operates about 166 municipal solid waste incinerators (MSWI) with a daily total capacity of approximately 158,488 tons [1]. MSW consists of various compositions such as paper, plastics, glass, metals, textiles, food wastes, and yard wastes. These miscellaneous components contain some quantity of the heavy metals [2]. Although incineration has greatly contributed to reduce the volume of MSW, heavy metals contained in MSW can be released into vicinity environment as
particulate and gas from stack after combustion [3]. Due to their toxicities and potential health hazards, the emission of heavy metals from MSWI has became an issue of great concern [4, 5].

Heavy metals such as lead, cadmium, chromium, zinc, arsenic and mercury are among the pollutants present in the emission from MSWIs. Previous researches indicated that atmospheric deposition of heavy metals emitted from MSWI could be an important source for heavy metal pollutions in soils near the incinerator [3]. Rovira et al. (2010) had reported increased levels of manganese and zinc in soils in the vicinity of a MSWI. It was also found that high emissions of cadmium, lead and mercury from incineration stacks resulted in an increase in the corresponding metal concentrations in soils surrounding the MSWIs [6, 7]. It is well known that compositions of the raw MSW have an impact on chemical and physical properties of incineration ash [8]. In developed countries, separation and sorting of raw MSW before combustion helps to reduce the contents of toxic metals in MSWI, which contributes to the abatement of toxic metal contents emitted from stack. However, in China, the solid wastes have been collected and sent to incineration plant directly. Consequently, higher levels of metal compounds can be found in the incineration stack emissions. Knowing how the influence of waste combustion on the levels and profiles of heavy metals in soils in the vicinity of MSWIs may help in assessing human health hazards and risks caused by operation of MWSIs.

The windblown dust has an important impact on the atmospheric deposition of heavy metals. Therefore, in this study, soil samples in the vicinity of MSWI plants were collected to determine concentrations of the toxic metals in two different representative geographical areas, locating in Jiaxing city in Zhejiang province and Beipei district in Chongqing city. These two selected incinerators provided a number of contrasting features. Jiaxing MSWI is surrounded by flat land and lies not far from the coast, whereas Chongqing MSWI is surrounded by hills and lies more inland.

2. Materials and methods

2.1 Study area

Jiaxing city, located in the northeast of Zhejiang province, is the center of Hang Jia Hu plain in the Yangtze River Delta. It is between latitude 30°21’ N and 30°2’ N, and longitude 120°18’ E and 121°16’ E, facing the sea in the east, leaning on the Qiantang River in the south, bearing on the Tai lake in the north, and connecting with the Tianmu Mountain in the west. The Grand Canal crosses the region. The terrain of the whole city is predominantly flat, with an average elevation of about 3.7 m. Meteorological survey reveals that this area is dominated by southern subtropical monsoon climate, with an annual average temperature of about 16 °C and rainfall of approximately 1200 mm. Monsoon circulation has a significant influence on the seasonal changes of local wind regime. Wind rose diagram displays that wind direction changes frequently in Jiaxing city. The annual average wind speed is 3.5 m·s⁻¹ (Fig. 1a).

Chongqing, namely "the mountain city", located in the eastern Sichuan Basin. which is in the center zone of Yangtze River Three Gorge positioning between latitude 28°10’ N and 32°13’ N, and longitude 105°11’ E and 110°11’ E. The city is surrounded by Daba, Wushan, Wuling Mountain, the mountains Lou in the north, east and south, respectively. The highest point is 950 m on Mt. Jinyun, whereas the low-lying areas along the Yangtze River are <200 m. The city is situated where the Jialing River passes through the Sichuan Basin to join into the main branch of the Yangtze, forming a peninsular. Chongqing’s climate belongs to the humid sub-tropical monsoon climate, with an annual average temperature of around 18 °C and rainfall of about 1000-1450 mm. Wind rose diagram shows that the prevalence of wind blows northeast with the annual average wind speed of 1.12 m·s⁻¹ (Fig. 1b).
Figure 1. Location of sample sites in Jiaxing city (a) and Chongqing city (b), China

2.2 Sampling strategy
The locality of Jiaxing incinerator is generally flat and regarded as agricultural. The area around the incinerator plant was divided into eight sectors (NW, N, NE, E, SE, S and SW). Seventy samples were taken at various sites along the eight defined directions within a radius of 2 km from the stack of the facility (Fig. 1 a). The area surrounding the Chongqing incinerator is located within a network of busy roads, industrial and agricultural activities. A grid with a regular square mesh of 400 m centered on the plant was created. Sixty-one samples were taken at various grids within a radius of 2 km from the facility (Fig. 1 b). Each sampling point was georeferenced using a GPS unit. Soil cores were collected at the same depth (0-20 cm), and each sample consisted of four sub samples obtained in a 3×3 m split in five equal partition. The soil cores were combined to give a composite sample for each site. Gravels and plant materials including roots and leaves in the soil samples were manually removed. Subsequently, the soil samples were air-dried and sieved to <1 mm.

2.3 Chemical analysis
The concentrations of metals (Fe, Mn and Cr) were analyzed using X-ray fluorescence (XRF) spectrometry (Innov AS4000). Samples for XRF analysis were analyzed in triplicate. Accuracy and precision were based on value of coefficient of variation (CV) less than 18%. Microwave digestion was carried out over 30 minutes for the soil samples (0.25 g) with hydrofluoric acid and chloronitric acid. The concentrations of Cu, Pb and Cd in the digest were determined by atomic absorption spectroscopy (AAS, Shimadzu AA-6300). Mercury in the digest was measured using F732-G digital mercury analyzer (Shanghai Huaguang Instrument). Each sample was measured in triplicate with CV of ±7.12%. A reference heavy metal standard solution was used for quality control. The typical recovery percentage is between 90% and 110%.

2.4 Data analysis
Principal Component Analysis (PCA) was conducted by means of SPSS v.22 software (IBM Corporation, Armonk, NY). PCA with a VARIMAX rotation was applied for interpretation of the PCs and factors with eigenvalues greater than unity were retained in the analysis.
3. Results and discussion

3.1 Metal emissions from incinerators

The Jiaxing incinerator was built in 2003 and processes 1900 tons of solid waste per day. The Chongqing incinerator has been operated since 2005, treating 2200 tons of solid waste per day. Both incinerators installed air pollution control systems. Flue gas are purified by means of a semi-dry scrubber with injection of activated carbon, and then released to the atmosphere via a single stack after bag filtration. Volatile heavy metals such as Pb, Cd and Hg are the common pollutants found in the emissions from MSWIs. The stack emissions of these metals are measured regularly by the MSWI's operators. The greater amounts of Pb, Cd and Hg emissions detected in Jiaxing plant than in Chongqing plant are most probably due to differences in pollutant sources (Table 1). Various metal contents in the composition of feed MSW may result in the great difference of stack emissions between Pb and other metals. In comparison with Cd and Hg, Pb can be considered as a major contamination tracer of MSWI stack flue gas.

|            | Hg     | Cd     | Pb    |
|------------|--------|--------|-------|
| Jiaxing    | 8×10⁻⁴ | 4×10⁻² | 0.6   |
| Chongqing  | 5.6×10⁻⁵ | 2.7×10⁻⁴ | 0.2   |

3.2 Descriptive statistical analysis of heavy metals

Table 2 summarized the basic statistics related to heavy metals in soils from coast plain and inland mountainous regions as well as the background values and soil environmental quality used in this study. The enrichment degree of studied metals in soil samples from coast plain and inland mountainous regions based on the mean content values decreased as follow: Fe > Mn > Cr > Zn > Pb > Cu. As comparing the mean values of heavy metals with the background values, the results showed that all heavy metals (Cr, Mn, Cu, Zn and Pb) in soils from both regions were higher than their background values. In particular, Cr in coast plain region and Pb in inland mountainous region were approximately 2.0 and 3.5 times higher than their background values, suggesting that the increase of Cr and Pb in soils is probably due to their emissions from MSWI plants. However, the mean values of the metals except Cr were lower than the related Soil Environmental Quality Standard (SEQS) values for the soils in China [9]. The mean value of Cr is higher than the SEQS value by about 50% and 83% in coast plain and inland mountainous regions, respectively. Cr is the major contamination spread around the investigated areas and expected to cause adverse biological effects in these areas.

| element   | Minimum | Maximum | Mean   | soil background value | Soil Environmental Quality Standard (GB 15618-2008) |
|-----------|---------|---------|--------|-----------------------|--------------------------------------------------|
|           | (n=70)  |         |        |                       |                                                  |
| jiangxing |         |         |        |                       |                                                  |
| Cr        | 146     | 521     | 274.21 | 182.6                 | 150                                              |
| Mn        | 500     | 2278    | 1053.47| 827                   | -                                                |
| Fe        | 33275   | 91210   | 58755.9| -                     | 50                                               |
| Cu        | 22      | 103     | 37.38  | 30.63                 | -                                                |
| Zn        | 51      | 183     | 90.60  | 90.1                  | 200                                              |
| Pb        | 28      | 50      | 40.53  | 19.4                  | 80                                               |
| Cd        | ND      | ND      | -      | -                     | 0.30                                              |
| Hg        | ND      | ND      | -      | -                     | 0.35                                              |
|           | (n=51)  |         |        |                       |                                                  |
| chongqing |         |         |        |                       |                                                  |
| Cr        | 134.00  | 358.00  | 224.00 | 64.45                 | 150                                              |
| Mn        | 86.22   | 1193.78 | 658.99 | 583                   | -                                                |
3.3 Spatial distribution and source identification of heavy metals

3.3.1 Principal component analysis (PCA)
PCA was performed to assist the identification and analysis of sources of heavy metals in surface soils from coast plain and inland mountainous regions. The Bartlett’s sphericity tests ($P<0.001$) indicated that heavy metal contents in surface soils from the studied areas were suitable for PCA. Table 3 showed the results of PCA by applying varimax rotation for heavy metals and Fig. 2 showed the variation diagram in rotated space.

Table 3 Principal component analytical study area mainly heavy metals

| Component matrix | Rotation component matrix |
|------------------|---------------------------|
|                  | 1  | 2  | 3  | 1  | 2  | 3  |
| Ti               | 0.971 | 0.093 | -0.014 | 0.966 | -0.135 | -0.002 |
| Cr               | 0.944 | 0.183 | -0.01 | 0.959 | -0.053 | 0.043 |
| Mn               | 0.855 | 0.157 | -0.1  | 0.874 | -0.015 | -0.045 |
| Fe               | 0.971 | 0.120 | -0.029 | 0.974 | -0.104 | -0.003 |
| Cu               | -0.428 | 0.737 | -0.013 | -0.225 | 0.920 | -0.018 |
| Zn               | -0.164 | 0.911 | -0.157 | 0.051 | 0.894 | 0.28 |
| Pb               | -0.083 | 0.593 | 0.793 | -0.012 | 0.174 | 0.978 |
| eigenvalue       | 3.724 | 1.805 | 0.835 | 3.617 | 1.709 | 1.039 |
| variance contribution rate % | 53.205 | 25.792 | 11.934 | 51.669 | 24.414 | 14.848 |
| Cumulative %     | 53.205 | 78.997 | 90.932 | 51.669 | 76.084 | 90.932 |

The results of metals in coast plain surface soils indicated that PCA reduced the number of variables to three principal components (PCs), which explain 88.58% of the data variance (Table 3 and Fig. 3a). The first PC, loaded heavily by Fe, Cr and Mn, accounted for 46.16% of the total variance. The high loading of Fe (0.944), Cr (0.909) and Mn (0.906) in Fig. 2a indicated the three metals had
similar source. PC2 with a variance loading of 29.25% was dominated by Cu (0.884) and Zn (0.934), suggesting that they may have geochemical affinities. PC 3 with a variance loading of 13.18% was correlated strongly with Pb (0.98), implying that Pb may have different sources in contrast to other metals.

The result of PCA of the metal concentrations in inland mountainous surface soils was shown in Table 3 and Fig. 2b. Four principal components were extracted during PCA, accounting for over 90.42% of the total variance. PC1 with a variance loading of 28.29% was loaded by Zn (0.801) and Pb (0.506). PC2 with a variance loading of 26.70% was loaded by Mn (0.829) and Fe (0.467). PC3 with a variance loading of 17.88% was dominated only by Cr (1.065). PC4 with a variance loading of 17.56% was dominated only by Cu (1.078). The highly different metal loading values may reflect that heavy metals in inland mountainous soils have weak geochemical affinities than in coast plain surface soils.

3.3.2 Spatial distribution of heavy metals in coast plain and inland mountainous surface soils
Using the available measurements for Cr, Mn, Fe, Cu, Zn and Pb concentration as well as semivariograms model, spatial maps of these pollutants were generated using the simple point kriging procedure. Fig. 3 and Fig. 4 showed the spatial distribution patterns of Cr, Mn and Fe gathered from the two study areas. Cr, Mn and Fe, as the first principal component in coast plain surface soils, were mainly controlled by parent material and came from natural source. The spatial distributions of Cr, Mn and Fe had similar patterns and their concentrations were randomly dispersed in the coast plain area (Fig. 3a, b, c). The contents of Cr, Mn and Fe, reflecting the geochemistry of local soil environment, were nearly influenced by the human activities and mainly depended on the soil structural factors such as soil texture and soil erosion. In contrast, the spatial distribution of Cr (PC3) in inland mountainous soils showed some hot spots at northwestern, southern and southeastern parts of the study area (Fig. 4a) The highest concentration of Cr which was about 6 times higher than the background value was found in the southeast of the study area, implying a serious soil pollution caused by Cr. Historic land use indicated that a large chromium salt production plant was located very close to the site where also served as chromium slag accumulation point. Although the plant had been shut down for a quite time, it is most likely to have contributed to the soil Cr load. The spatial distributions of Mn and Fe, second principal component of inland mountainous surface soils, were shown in Fig. 4b and Fig. 4c. Their high contents were sparsely distributed in the study area. The contents of Mn and Fe correspond well to the background value of the study area. This can be attributed to several factors such as the nature of the local geology and soil type.

Figure 3: Spatial distributions of Cr(a), Mn(b) and Fe(c) in soils near Jiaxing incineration plant
Figure 4. Spatial distributions of Cr(a), Mn(b) and Fe(c) in soils near Chongqing incineration plant

The spatial distribution of Cr and Mn (PC3) in coastal plain soils demonstrated similar patterns (Fig. 5a, b). The high contents of the metals were sparsely distributed in the study area. Historic land use showed the majority of the study area was food crop farmland with relatively lower metal concentrations. In contrast, vineyard in the study area had higher metal concentrations. The difference can be attributed to the local pesticide dosage in farmland and vineyard. It is known that dimethoate and zineb are the most commonly used pesticides in local grape cultivation. Thus, it can be assumed that higher Cu and Zn contents in the pesticides resulted in higher Cu and Zn concentrations, with the mean values of 37.38 mg/kg, 90.10 mg/kg, respectively, in vineyard of the study area [10, 11]. The spatial distribution of Cu (PC4) throughout the inland mountainous area exhibited high concentrations occurred at southern and northern part of the study area (Fig. 6a). The southern hot spot was located closely to a cross section of main road. It can be assumed that busy traffic contributes Cu to soils. It is known that lubricant can be oxidized by oxygen in the air to generate organic acids, alcohols, ketones, aldehydes and other organic compounds. These substances cause corrosion of metal parts and oil pump containing Cu in vehicles, subsequently, releasing Cu in soils in the vicinity of the road [12-14]. The northern hot spot was farmland for fruit cultivation. The increase of Cu concentrations was mainly due to the application of pesticides containing Cu.
3.3.3 Spatial distribution of Pb and effect of terrain and wind on Pb distributions

Pb had been considered an important contamination tracer which was highly invested by road traffic. With implementation of the lead free gasoline standard GB17930-1999 in 2000, Pb was no longer considered as major traffic source. In MSWIs, metal contents contained in the solid wastes were discharged from the stack as particulate matters and vapor. However, due to their high volatility, Pb, Cd and Hg could volatilize and escape from the furnace with flue gas, which were then partially removed by APC devices [15]. Therefore, Pb, Cd and Hg were considered important environmental indicators as typical MSWI air contaminants [16, 17] and listed as regularly monitoring substances by Chinese MSWI operators. In Table 1, emission rates of Pb, Cd and Hg from Jiaxing incineration plant were reported. Higher Pb emission (2.3 t/y) was detected, compared with much lower Cd (1.0×10^{-1} t/y) and Hg (3.1×10^{-3} t/y) emissions. Furthermore, the soil background of Cd and Hg concentrations in coast plain regions were low and their concentrations in the studied soils were below detection limit. Therefore, Pb is considered a major contamination tracer of MSW air contaminants. The spatial distribution of Pb in coast plain soils indicated that Pb contaminations in upwind points of the main

Figure 5. Spatial distributions of Cu(a) and Zn(b) in soils near Jiaxing incineration plant

Figure 6. Spatial distributions of Cu(a) and Zn(b) in soils near Chongqin incineration plant
wind directions were not evident, where Pb in soils was equivalent to the background value (Fig. 7a). In contrast, higher Pb concentrations were found along the main wind directions (W, SW, NW). It was observed a positive relationship between the distance from the stack and Pb levels in downwind points of the main wind directions (Fig. 8). The most seriously Pb contaminated sector was that in downwind direction of the incinerator with mean value of 42.22 mg/kg, 2.2 times greater than the background value. In addition, the concentration of Pb was significantly correlated with the distance from the stack (P<0.05). Considering the factors which can affect the Pb emission from Jiaxing incinerator and the subsequent deposition to surrounding soils, the highest Pb concentration will be found at the distance of 1784.83–2648.27 m from stack according to the Gauss model and other air dispersion equations. The experimental data was consistent with the calculated values. The highest Pb concentration was found at 2400 m away from the incineration plant in west section of the studied area. As the even terrain of coast plain nearly affected the Pb distribution, the variability of the Pb concentration with downwind distance determines the spatial pattern of Pb distribution in the vicinity of Jiaxing MSWI. Pb will be considered an important contamination tracer of MSWIs.

Figure 7. Spatial distributions of Pb in soils near Jiaxing(a) and Chongqin (b) incineration plant

Figure 8. The relationship between Pb level in soil and the distance to Jiaxing incinerator stack

The difference between Pb distribution in inland mountainous and coast plain regions was obvious. Pb and Zn, as the first PC in inland mountainous soils, had general similar spatial distributions in the studied region, with the high metal concentrations appearing in the south site of the region (Fig. 6b and Fig. 7b). In addition, High content of Pb was observed in the north site of the region (Fig. 7b). Compared with the high Pb distribution at downwind direction in coast plain region in which the
MSWI is sited, no significant increases in the levels of Pb were detected for prevailing wind directions in inland mountainous region. The high concentrations of Pb and Zn in the surface soils occurred in the areas where anthropogenic activities was intense. Historic land use indicated that some small factories dealing with painting, electroplating and welding businesses were located at north and south sections of the studied area in 1990. The high values of Pb and Zn were probably caused by the industrial activities in north and south sites of the area. The operation report of Chongqing incinerator indicated that emissions of Hg, Cd and Pb were 4.3×10⁻³, 3.2×10⁻³ and 6.2×10⁻² t/a, respectively (Table 1). Pb emission from the inland mountainous incinerator was only 3% of the coastal plain incineration plant. Furthermore, taking into account of the shorter operation time of the Chongqing MSWI (inland mountainous incinerator), there was no evidence that Chongqing incinerators had significantly altered Pb concentrations in surrounding soils.

4. Conclusions
This study investigated the stack emission of heavy metals and their impacts on surrounding soils in two MSWIs located at coastal plain in Eastern China and inland mountainous area in Western China, respectively. The main conclusions include:

a. The results showed that Cr and Mn presented as background origin in soils surrounding the coast plain incinerator. Levels of Cr and Mn laid well within their natural ranges in coast plain soils. Cu, Zn and Pb distributions in both studied regions were influenced by exogenous inputs. Human activity was the dominant source leading to changes in the original distribution of those metals.

b. The results of the PCA and semivariograms model, as well as the values of the soil sample analysis, confirmed that Mn and Fe in surface soils of both studied regions mainly came from natural source. An analysis of the metal speciation indicated that application of pesticides appeared to be a major contributory factor in determining the distributions of Cu and Zn in soils surrounding the coast plain incinerator. In addition, the direction of the prevailing wind influenced the distribution of Pb in the area. Data analysis demonstrated that road traffic and application of pesticides resulted in the changes of distribution of Cu in soils surrounding the inland mountainous incinerator. Meanwhile, Cr, Zn and Pb distributions in the region were likely related to local industrial productions.

c. In comparison with Hg and Cd, stack emission of Pb were relatively high from both plants. The study highlighted that Pb was an important environmental indicator of MSWI air contaminations. The source identification and spatial distribution of Pb in soil in the vicinity of MSWIs confirmed that the extent of soil contamination caused by MSWIs was highly ascribed to the stack emission of heavy metals. The source apportionment and distribution of Pb in soils indicated that total soil pollution level in the areas surrounding MSWI plants mainly depended on pollution emissions. Furthermore, the terrain and meteorological conditions had a significant effect on heavy metal pollution distributions.

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