Distribution and sources of mercury in soils from former industrialized urban areas of Beijing, China

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Abstract Fifty-seven typical surface soils and 108 deeper soils were collected from five former industrial sites in Beijing and concentrations of

total Hg (ΣHg) as well as pH, total carbon (TC), total nitrogen (TN), total sulfur, and dissolved organic carbon concentrations determined. The mean concentration of ΣHg in surface soils was significantly greater than background concentrations in the vicinity of Beijing. Forty-eight percent of the samples exceeded the “critical” concentration of 1.0 mg Hg/kg, dry weight in soils, which has been established by the Chinese government. At depths of 0–80 cm in the soil, profile concentrations of ΣHg also exceeded the background value. There were significant correlations between concentrations of ΣHg , TC, and TN in the industrial soils. The greater concentration of ΣHg in most soils could have been due in part to combustion of coal and leakage from industrial processes.

Keywords Hg · Metal · Pollution · Contamination

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Introduction

Mercury (Hg) is one of the most toxic elements and can cause renal and neurotoxicity to humans and wildlife. It has been identified as a priority toxic substance in many countries (Gaudet et al. 1995). The amount of Hg mobilized and released into the environment has increased since the beginning of the industrial age (Mason et al. 1994).

However, Hg contamination and its effects upon the environment and human beings were only recognized as a significant environmental issue after the tragic accident that occurred in the Bay of Miuamata, Japan in 1956. Because of Hg's volatility and toxicity, it is recognized as a global environmental problem by scientists and the general public (Agamuthu and Mahalingam 2005).

Hg pollution is primarily due to human activities. Emissions of Hg from human activities accounted for 60% to 90% of the total Hg (ΣHg) emitted in some industrialized regions (Nriagu 1989). Between 1990 and 1995, emissions of Hg due to human activities in Asia increased from approximately 30% of the total global emissions to 56% (1,074/1,912 t). Within that 5-year period, due to rapid industrialization, emissions in China alone increased by 250 t (Pacyna and Pacyna 2001). Most of the world's industries are located in urban settings, and a large proportion of Hg emissions from industries are deposited locally (Landis et al. 2004). The greatest concentrations of ΣHg in soils of industrialized areas were at least 20-fold greater than background concentrations (Linde et al. 2001).

The industrialization of the Beijing began in the 1950s. The main industries included electronics, manufacturing, including what is classified as heavy industry, and chemical production. The number of industrial facilities eventually totaled about 20,000. With Beijing's successful application to host the Olympic Games in 2008 and the rapid extension of urban areas in recent years, most industrial plants in the city are being closed or moved from downtown to suburban areas. These former industrial sites have been converted into commercial and residential properties. It is increasingly being recognized that these industrial sites might have become significantly contaminated by these industrial activities to levels that might be significant risks to human health. Mercury pollution of soil is one potential environmental problem because Hg was contained in airborne fallout, waste dumping, liquid leakage and incidental emissions from the workshops, waste treatment plants, and storage locations of old equipment in the former industrial plants. It has been reported that there are relatively great concentrations of Hg not only in the air, snow

(Liu et al. 2002; Wang et al. 2004), and drinking water in the Beijing urban areas (Gao et al. 2004) but also in soils on the outskirts of Beijing (Tan et al. 2005). Soils of Beijing exposed to industrial sewage and sludge have been found to be contaminated with Hg (Zhu 2001). An investigation indicated that the concentration of Hg in people's hair was greater in industrial areas of Beijing (Wu et al. 1991). Communities built on such polluted sites can be at increased risk of environmental and human health problems, even long after the source of Hg has been discontinued. Therefore, it became important to determine potential sources of Hg, including the distribution, and in soils at these former industrial sites and the potential exposure of urban inhabitants, especially children.

It is, however, rare to find information on Hg in soils from former industrialized areas of Beijing in the open literature. In order to ascertain the sources and distribution of Hg, as well as the location and extent of contamination with Hg, and to provide policymakers with remediation measures for the affected soils, a detailed study of surface soils and complete soil profiles was conducted in industrialized urban areas of Beijing. Concentrations of ΣHg were measured in soils, and the profiles of ΣHg concentrations were interpreted as a function of depth and their relationship with soil properties and possible sources in the former industrial sites discussed. The results obtained can be of use for the municipal land planners and managers to identify pollution sources, locate polluted areas for remediation measures, and to choose the proper cleanup strategies. Moreover, the data may be of interest to environmental agencies for their activities of soil monitoring.

Materials and methods

Site description

The industrialized urban area was located approximately 1–8 km southeast of the center of Beijing. Industrial activity in the area started in the 1950s and ended around 2006. Five representative industrial sites (denoted as A, B, C, D, and E) in the industrialized area were studied.

Plant A was established in 1958 and its main products were chemical agents, fluorescent, and photosensitive materials. Plant B was established in 1956 and produced pesticides, polyvinyl chloride plastic, and rubber products. Plant C was built in 1959 and its main products were coke, coal gas, ammonium sulfate, benzene, naphthalene, and hydroxybenzene. Plants D and E were established, respectively, in 1954 and 1956. Plant D mainly produced pesticides, lacquer, and building and industrial paint, while Plant E mainly produced dyestuffs, paint products, and vitriol. All of the plants have now been closed or moved, and some residential communities have been, or will be, established on the land. There are also other facilities in the area, such as hospitals, schools, residential communities, restaurants, and guesthouses. The soils in the investigated area developed on a sandy-textured alluvial deposit and are dominated by silt and sandy loams; the predominant soil types are typically fluvial in origin. Sampling locations taking up most of the site were selected based on locations of production and waste treatment sectors, which were potentially important sources of contamination.

Sample collection

Fifty-seven representative surface soils and 108 deep layer soil samples from 27 profile pits were collected from the five sites (Fig. 1). Four subsamples were collected at each sampling point and mixed thoroughly to get a representative soil sample. The deeper samples of soil were taken from each profile pit at depths of 20, 80, 180, or 400 cm.

Soil analysis

Prior to analysis, soil samples were air-dried, crushed in an agate mortar, and passed through a nylon sieve of 0.149 mm. For total Hg analysis, 0.2–0.3 g dry soil samples were digested at 95°C in a water bath with an acid mixture of HCl/HNO₃. Mercury concentrations were measured in digested sample solution by use of cold-vapor atomic absorption spectrometry, with the instrument detection limit of 0.01 µg Hg/kg, wet weight. A standard soil sample GBW-07405 (GSS-5)

0.29 ± 0.04 mg Hg/kg dry weight (dw), from the Center of National Standard Reference Material of China, was used to evaluate the analytical accuracy within 5%. The concentration measured was 0.30 ± 0.01 mg Hg/kg, dw, with the average recovery rate of 103% in triplicate analysis. Precision was estimated by use of analysis of duplicate. The precision was greater than 95%.

Fifteen grams of sample was dissolved in 15 ml of deionized water, and after mixing for 24 h, the pH was measured. For dissolved organic carbon (DOC) measurements, 7 g of soil was shaken with 28 ml of distilled water for 1 h, followed by centrifugation and filtration through 0.45-µm filters (Wright et al. 2005). Extracts were analyzed for DOC by persulfate oxidation using a total organic carbon analyzer (model 7000, OI Corporation, TX, USA). Concentrations of total carbon (TC), total nitrogen (TN), and total sulfur (TS) were determined by use of a Universal CHNOS elemental analyzer (Elementar Vario EL III, Germany; Mai et al. 2003).

Statistical treatment

Statistical analyses were carried out using Microsoft Excel and SPSS 10.01 software (SPSS Inc., USA). The normal distribution of Hg concentrations was tested by the Kolmogorov–Smirnov method. Analyses of significant differences of Hg concentrations between two sites were performed by the analysis of variance (data with normal distribution) or by the Kruskal–Wallis *H* test (non-parametric data). A significance level of *P* < 0.05 was used throughout the study. Relationships between parameters were investigated by use of Spearman rank-order correlation (non-parametric data) or Pearson product-moment correlation (normal distribution).

Results and discussion

Total concentrations of mercury in surface soils

Concentrations of Hg in soils were plotted as histograms and normal probability plots either arithmetically (Fig. 2) or after logarithmic transformation (Fig. 3). Skewness or kurtosis indicates

Fig. 1 Map of sampling locations in the industrialized urban areas of Beijing, China

departures from normality. The normal probability plot provides a visual estimate of the nearness of the data to a normal distribution. If the data are normally distributed, their points will fall on a straight line in such a plot, and curvature in the points indicates a deviation from a normal

distribution. Thus, the distribution of ΣHg in the surface soil samples was lognormal (Figs. 2 and 3).

Bare soil on the ground surface may serve as local “hot spots” for Hg emissions because of the large percentage of ΣHg lying in the upper few centimeters of soil. The mean concentration

Fig. 2 Histogram and normal probability graph of ΣHg concentrations in surface soil samples

of ΣHg (4.85 mg Hg/kg, dw) in the Beijing industrial soils (Table 1) was significantly greater than the background concentration of ΣHg in other Beijing soils (0.06 mg Hg/kg, dw; CNEMC 1990), as well as the Beijing urban soil background concentration (0.28 mg Hg/kg, dw; Zhang et al. 2006). According to the guidelines for Hg (Wu and Zhou 1991), 100% of the soil samples from Beijing industrial sites exceeded the “ideal” con-

centration (0.1 mg Hg/kg, dw) and “acceptable” concentration or “threshold” value (0.2 mg Hg/kg, dw), which indicates that very careful environmental management is necessary. Seventy-seven percent of the soil samples exceeded the “tolerable” concentration, which also corresponds to the “warning” value (0.5 mg Hg/kg, dw). This means that 77% of the area is only mildly ecologically impacted, and the foodstuff grown on this area

Fig. 3 Histogram and normal probability graph of ΣHg concentrations in surface soil samples after $\log_{10}$ transformation of the data

Table 1 Descriptive statistics of ΣHg concentrations and mean values of general parameters of the surface soils

Site	No. of samples	pH	TN (%)	TC (%)	TS (%)	DOC (mg/kg)	Hg (mg/kg)		SD	Min	Max	Skew	Kurt
A	6	8.2	0.02	1.40	0.02	374.98	Mean	9.65a	17.30	1.06	44.70	2.37	5.68
B	13	8.1	0.06	2.18	0.07	627.66	Mean	13.22a	20.46	1.37	76.27	2.80	8.61
C	14	8.3	0.05	1.89	0.08	200.99	Mean	1.24b	1.75	0.26	6.77	3.05	9.89
D	10	8.5	0.04	2.11	0.06	483.46	Mean	0.61b	0.44	0.23	1.65	1.92	4.28
E	14	8.2	0.05	1.89	0.13	149.13	Mean	1.08b	1.63	0.22	6.60	3.43	12.29
Total	57	8.3	0.05	2.02	0.08	261.03	Mean	4.85	12.30	0.22	76.27	4.51	22.84

For the indicated ΣHg , those values not showing a common superscript (a, b) are significantly different ($P < 0.05$)

had elevated concentrations of Hg. Forty-eight percent of the soil samples exceeded the “critical” concentration (1.0 mg Hg/kg, dw), which means that 48% of the area is affected by anthropogenic input of ΣHg . Due to the chemical and physical nature of surfaces of the plants in the area, their large spatial variability, the time series characteristics of industrial Hg emissions (i.e., industrial burning, processing, waste incineration; Liu et al. 2002; Feng et al. 2003), the ambient Hg concentrations, and the urban meteorology, the ΣHg concentrations in the soils of the industrialized urban land was significantly greater than those of farmland (Table 2). This result is in agreement with the results of another study (Hollis 1991). Furthermore, based on the soil cleanup standards (SCS) for Hg in soils used for residential or industrial purposes in some countries (Table 3), only two samples contained concentrations of ΣHg greater than the USA residential SCS, four samples were greater than the German SCS, and five were greater than the SCS from Belgium. However, based on the standards of other countries, 50 samples (89%) contained concentrations that were less than 8.0 mg Hg/kg, dw, which implies that 89% of the industrialized urban land can be used for residential purposes. According to the soil cleanup standards for Hg for industrial land use in most countries, most industrialized urban land (except one or two places) in Beijing can continue to be used for industrial purposes.

Because none of the distributions of Hg concentrations in soils was normally distributed based on skewness and kurtosis (Table 1), the Kruskal–Wallis test, a method used for non-normal data distributions, was used to test for differences between the means of the Hg concentrations in the surface soil samples of different sites. Mean Hg

Table 2 Comparison of ΣHg concentrations (mg ΣHg /kg, dw) in industrial soils with those in farmland soils in Beijing

	N	Mean	SD	Min	Max
Farm land ^a	45	0.04	0.03	0.01	0.16
Industrial land	57	4.85	12.30	0.22	76.27
Background value ^b	38	0.06	0.08	0.00	45.90

^aLi et al. (2006)

^bCNEMC (1990)

Table 3 Overview of the soil cleanup standards for ΣHg (mg/kg, dw) for residential and industrial land uses in various countries (Provoost et al. 2006)

Country	Residential land	Industrial land
Belgium	15	30
Germany	20	80
France	7	600
Sweden	1	7
Great Britain	8	480
Canada	6.6	50
USA	23	310

concentrations of soils were the following decreasing order: $B > A > C > E > D$. Mean Hg concentrations in soils of sites B (13.22 mg Hg/kg, dw) and A (9.7 mg Hg/kg, dw) were significantly greater than any other site ($P < 0.05$). This indicates that the surface soils in areas of industries like those at Plants B and A may have the greatest Hg pollution potential.

Total mercury concentrations in soil profiles

There was no statistically significant difference in mean ΣHg concentrations between samples taken at 20- or 80-cm depths as a whole (Table 4). However, the mean concentrations of ΣHg at these depths were significantly greater than those at depths of 180 and 400 cm. At depths of 20 or 80 cm, there were six soils (three at 20 cm and three at 80 cm) whose Hg concentrations were greater than the maximum soil cleanup standard for Hg for residential areas (Table 3). No soil sample from depths of 180 and 400 cm had a ΣHg concentration greater than 6.6 mg Hg/kg, dw (Fig. 4). Although these five plants had almost the

Table 4 Descriptive statistics of mercury concentrations at different soil depths in the industrial area (mg ΣHg / kg, dw)

Depth (cm)	Number of samples	Mean ^a	SD	Min	Max
20	27	3.64a	6.27	0.22	22.72
80	27	3.65a	7.00	0.01	29.66
180	27	0.98b	1.52	0.05	6.31
400	27	1.11b	1.36	0.08	4.25

^aFor the indicated elements, those values not showing a common superscript (a, b) are significantly different at the level ($P < 0.05$)

Fig. 4 Mercury (ΣHg) concentration profiles with depth at different industrial sites

same periods of production, they had different distributions of ΣHg concentration in the deep soils. Soils from site B contained the greatest mean concentration of ΣHg in the profile. For profiles in which the maximum ΣHg content was found to lie between 20 and 80 cm, three possible causes are possible. First, Hg could have leached downward by colloidal transport (Jacobsen et al. 1998). Second, Hg could have evaporated from the surface soil (Schlüter 2000) as either methyl mercury or elemental mercury Hg^0 , but the degree to which Hg^0 evaporates is small. Nater and Grigal (1992) indicated that soils retain about 90% of the ΣHg deposited on them. Similar findings were observed in other studies and it was found that in spite of the evaporation of Hg^0 , there is a net accumulation of Hg in soils (Mason et al. 1994). Finally, plants may have absorbed Hg from the soil. All three of these possibilities most likely have contributed to decreasing the concentrations of ΣHg in the upper soils. Of these possibilities, colloidal transport appears to be the most likely mechanism for decreasing the ΣHg concentration in the upper soils. Since maximum mean ΣHg concentrations were observed at a depth of 80 cm, some remedial actions should be taken to remove the soils in the depth range of 0–80 cm. The economic aspects of remediation are usually not easily accessible, although they generally determine whether a remediation project will be executed or not. Based on the study by Hylander and Goodsite (2006), remediation costs range between US \$2,500 and

US \$1.1 million per kilogram Hg isolated from the biosphere depending on local circumstances such as quantities to secure, nature of pollution, media, geography, technology chosen, etc. The remediation technologies are expensive and complex processes, mostly effective at very high mercury concentrations. Remediation technologies appropriate for mercury removal include liquid extraction, thermal treatment, electrolytic methods, mercury flotation, or immobilization. Among them, phytoremediation may provide an efficient and practical cost-effective solution to mercury contamination (Sas-Nowosielska et al. 2008).

Soil properties and mercury concentrations

To investigate the common characteristics (behavior, origin, etc.) of ΣHg in soils in the urban areas of Beijing, correlation coefficients (Pearson product-moment coefficients) between mercury and the soil properties were calculated. Organic matter in soil is known to play a role in adsorption of Hg (Semu et al. 1987), although its quantitative contribution is not clearly understood. In this study, there was a positive correlation between TC and ΣHg concentration (Table 5). This result is in agreement with the results of Semu and Singh (1987). Surface soils, rich in organic matter, adsorb significantly greater amounts of Hg (Trost and Bisque 1972) and create strong organic complexes of great stability (Shuster 1991). Due to the affinity of Hg for organic matter, soil Hg leaching rates should be small because the industrialized urban soils in Beijing have great concentrations of organic matter (2.02%; Table 1). Furthermore, the results of Landa (1978) sug-

gested that in addition to quantity, the nature of a soils' organic matter also influences adsorption of Hg. It is important to pay close attention to DOC because of its large influence on sorption of Hg (Reddy and Aiken 2001). However, there was no significant correlation between ΣHg concentration and dissolved organic carbon in our study (Table 5), which is not in agreement with results reported in other studies (Shuster 1991; Linde et al. 2007). Further study is needed to explain this. Changes in pH can also have a major effect on a soil's ability to retain Hg. Mercury adsorption generally decreases with decreasing pH due to H^+ ions removing and replacing Hg ions. However, in our study, all samples were slightly alkaline, with a mean pH value of 8.2 (Table 1), and there was no significant correlation between soil pH and ΣHg concentration (Table 5). This means that adsorption of Hg by soils would not change when the soil is in alkaline. Furthermore, concentrations of Hg were significantly correlated with concentrations of TN, but not C/N ratio or TS (Table 5). C/N ratio is often related to humification (Gobat et al. 1998). In this study, high organic C/N ratios (>36) is characteristic of a low humification state of organic matter and of poor bacterial activity (Semu et al. 1987; Gobat et al. 1998). Little humification of organic matter and of poor bacterial activity may have resulted in relatively little complexation of Hg. Therefore, the C/N ratio was poorly correlated with Hg. In relatively more-reducing environments, Hg appears to be converted, in large part, to relatively insoluble HgS (Lechler et al. 1997). However, in this study, due to the small concentration of TC (about 2%) in soils of industrial sites and great oxygenation and oxidant-to-reducing environmental conditions at the surface soils, HgS may have become soluble (Moore and Ramamoorthy 1984). This may be the reason for the poor correlation between Hg and TS in the soils. Significant relationships between ΣHg concentrations and TC and TN suggest that organic matter and nitrogen in the soils control ΣHg concentrations of industrial urban soils and that adsorption and retention of ΣHg in these industrial soils is influenced by organic matter and nitrogen. Thus, organic matter and nitrogen may be important sinks for ΣHg in the soils studied.

Table 5 Rank correlation coefficients for ΣHg and chemical properties in the soils ($n = 57$) from the Beijing industrialized urban area

	Hg	pH	TN	TC	TS	C/N	DOC
Hg	1.00						
pH	-0.12	1.00					
TN	0.35**	-0.11	1.00				
TC	0.35**	0.05	0.68**	1.00			
TS	0.17	-0.12	0.67**	0.53**	1.00		
C/N	0.06	0.15	-0.44**	0.25	-0.27**	1.00	
DOC	0.23	0.12	0.44**	0.50**	0.52**	-0.27	1.00

**Correlation is significant at the 0.01 level (two-tailed)

Source of mercury

Industrialized urban areas are complex systems in which human activities have disturbed the natural development of soils. The possible inputs of Hg from industrial activities to urban lands include emissions from coal-burning power plants, residential or industrial boilers, waste incinerators, cement and lime kilns, petroleum refineries, landfills, fly ash from coal-burning locomotives, sewage sludge, and other sources. Mercury is also released during the life cycle of industrial products such as fluorescent lamps, electrical switches (electronics industry), thermostats, pressure sensing devices, products of chlorine-alkaline industry, paints, products of the paper industry, and products of the pharmaceutical industry (Sznopce and Goonan 2000). Based on our investigation, Plants A and B produced mercuric salts and phenylmercury acetate, respectively, but the other plants did not produce any Hg-containing products. Therefore, the soil at sites A and B had relatively great concentrations of ΣHg (Table 1 and Fig. 3). We concluded that the processes of producing Hg-containing products likely resulted in contamination of soils with Hg at these Beijing industrial sites. However, since industrial boilers, combustors, or incinerators relied heavily on coal for energy in industrial processes, the relatively great concentrations of ΣHg in soils from former industrialized urban areas of Beijing likely came primarily from industrial combustion of coal. The range of concentrations of Hg in coal has been reported to be 0.1 to 0.56 mg Hg/kg (Li 1992). Furthermore, soils with relatively great concentrations of ΣHg also contained relatively great concentrations of SO_2 from burning high-sulfur coal (Gao 1987). This indirectly indicates that Hg pollution in the soils studied may have come from coal combustion. Concentrations of ΣHg in surface soils in the vicinity of plants A and C were 1.7 and 0.51 mg Hg/kg, dw, respectively. This was most likely due solely as a result of burning coal (Liu et al. 1997). Beyond emissions of Hg from industries, residential coal burning in winter could be another factor contributing to Hg pollution in the industrial soils. The average Hg concentration in coal used by Beijing residents is 0.3 mg ΣHg /kg, which is greater than the average concentra-

tion in coal of China (0.2 mg ΣHg /kg; Jiang et al. 2005). Although ΣHg concentrations in surface soils decreased gradually from the boundary of Beijing industrial plants to residential areas, the values in residential areas were still as great as 0.2–1.0 mg Hg/kg, dw. Atmospheric Hg can be deposited into the terrestrial ecosystem through dry and wet precipitation, which increases the concentrations of mercury in soils. Atmospheric mercury concentrations in urban Beijing were reported to be 10–13 ng $\Sigma\text{Hg}/\text{m}^3$ in summer and 8.3–25 ng $\Sigma\text{Hg}/\text{m}^3$ in winter, respectively (Liu et al. 2002), which are several times greater than the global background range (1–3 ng $\Sigma\text{Hg}/\text{m}^3$; Tayban and Preston 2005). In addition, mercury concentrations in two snow samples from industrial areas of Beijing were 1.6 and 1.5 ng $\Sigma\text{Hg}/\text{l}$, respectively, which were greater than the background concentration of ΣHg in wet precipitation (0.2 ng Hg/ml; Liu et al. 1997, 1998). Another source of Hg pollution is industrial leakage and runoff accidents. For example, the annual runoff of Hg from Plant A in 1977 was about 583 kg based on an investigation. Therefore, it was reported that there was a very great concentration of mercury in the soil profile from 0- to 100-cm depth in industrial areas as a result of leaching (Liu et al. 1998). Thus, the amount of Hg from leakage and runoff was one of the important factors that caused contamination of soils at Beijing industrial sites with Hg.

Conclusions

The accumulation of ΣHg in the surface soils is particularly obvious in industrialized urban lands in Beijing compared with the farmland. Concentrations of ΣHg in 48% of surface soils in the industrial sites exceeded the “critical” concentration and were only mildly ecologically impacted as a result of the anthropogenic input. However, 89% of the industrialized urban land can be used for residential purposes. All the industrial surface soils need careful environmental management. Since Hg contamination was primarily confined to the top 0–80 cm, some engineering measures should be taken to remove these soils. Organic matter and nitrogen may be important sinks for

Hg in the industrial soils. The making processes of Hg-containing products have caused heavy soil pollution of Hg at the industrial sites. Atmospheric Hg emitted by industrial and residential coal burning might have been the main cause of soil mercury contamination at most industrial sites in Beijing. Another source of Hg in industrial soils was industrial leakage and runoff.

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