



Distribution and ecological effect of mercury in Laogang landfill, Shanghai, China

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Abstract

Laogang landfill near Shanghai is the largest landfill in China, and receives about 10000 t of daily garbage per day. Samples of topsoil and plants were analyzed to evaluate mercury pollution from the landfill. For topsoil samples, there were significant correlations among total mercury (Hg_T), combinative mercury (Hg_C) and gaseous mercury (Hg_G), and content of total organic carbon (TOC), but, no significantly relationship was found between Hg content and filling time. Hg content changes in vertical profiles with time showed that the average Hg_T of profiles 1992, 1996, and 2000 was similar, but their average Hg_G was quite different. Hg_T was significantly correlated with Hg_C in profile 1992 and 2000, and Hg_T was significantly correlated with Hg_G in profile 1996. Hg_G/Hg_T ratio in profile samples decreased in the order of $(Hg_G/Hg_T)_{1992} > (Hg_G/Hg_T)_{1996} \gg (Hg_G/Hg_T)_{2000}$. A simple outline of Hg release in landfill could be drawn: with increasing of filling time, degradation undergoes different biodegradation, accordingly, gaseous mercury goes through small, more, and small proportion to total mercury. Distribution of Hg in plants was inhomogeneous, following the order of leaf>root>stem. The highest value of leaf may be associated with higher atmospheric Hg from landfill. Ligneous plants (e.g. *Phyllostachys glanca*, *Prunus salicina* and *Ligustrum lucidum*) are capable of enriching more Hg than herbaceous plants.

Key words: mercury; landfill; distribution; plant; topsoil

Introduction

Mercury (Hg) is the pollutant controlled by all countries and international organizations such as UNEP, WHO, FAO etc. with top-priority. With a low vapour pressure, Hg is the only liquid metal at normal temperature. Point Hg pollution could be spread to the globe through air cycle. Scientists have been paying attention to mercury for tens of years for its jeopardy to environment and health of human beings (Lin and Pehkonen, 1999). Recently, release of Hg to the air, soil and underground water from large-scale landfill catches sight of scientific researchers (Lindberg *et al.*, 1999, 2001, 2005; Raloff, 2001; Kim *et al.*, 2001, 2002; Feng *et al.*, 2004; Li *et al.*, 2006). Along with Chinese rapid economy development, more and more Hg-bearing garbage produces in daily life i.e. fluorescent lights, battery, thermometer, barometer, and abandoned electric apparatus and so on. Meanwhile, garbage classification system is not widely adopted in China, a large amount of Hg-bearing garbage is treated as normal daily

refuse. Therefore, most Hg in the garbage pollutes the environment.

Shanghai is the biggest city of China, and faces the serious Hg pollution day by day (Ding *et al.*, 2005). Laogang landfill near Shanghai (31°00'N, 120°52'E), the largest landfill in China, is just located in north of Hangzhou Gulf and south of mouth of Yangtze River. The landfill is separated into many small pits (400 m×125 m), it started to receive garbage about 10000 t/a from 1989. Banks, waterway, dock, road, bridges and separating bands are equipped in the landfill. Reaching 4 m in height, garbage is covered with 30 cm thick of soil. Sewage is collected to ponds from every pit by ditch. After anaerobic digestion and oxidation in the ponds, sewage is directly released to the East China Sea, 1 km away from the landfill.

Environmental and ecological effects of Hg from landfill can not be overlooked. In this paper, Hg status was determined by analysis of topsoil and plants, and then, influencing factors, release process, and environment and ecological effects on local ecosystem were discussed accordingly.

1 Materials and methods

Topsoil is simply composed of quartz and few clay min-

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erals. Topsoil samples (0–15 cm) were collected in different pits filled during 1989 to 2001, and three profile samples were respectively collected in pits filled in 1992, 1996, and 2000.

Solidago canadensis, which was an exotic plant and grows everywhere around Shanghai, was selected as representative plant species to compare the Hg effect released from landfill pits with different filling time. Different plant species growing in pit 1994 were taken to compare their assimilating ability of mercury. These plants include *Solidago canadensis*, *Nerium indicum*, *Cana indica*, *Ilex purpurea*, *Chenopodium album*, *Setaria viridis*, *Sesbania cannabina*, *Phyllostachys glanca*, *Trachycarpus fortunei*, *Ligustrum lucidum*, and *Prunus salicina*. Root, stem, leaf component were collected.

Profile samples were cut into sequential sections every 3 cm. After dried in room temperature, those profile samples and topsoil samples were ground to pass a 100-mesh sieve. Moisture, pH, and total organic carbon (TOC) were analyzed according to SCACAPC (1983). Rinsed with tap water and DI water in turns, plant samples were analyzed in a fresh basis.

Total mercury contents (Hg_T) of all samples were analyzed in duplicate on an AMA 254-Automatic solid/liquid Hg Analyzer (Milestone, Italy), with an absolute detection limit of 0.01 ng.

Gaseous mercury (Hg_G): 1.0 g soil sample weighted to put in a vassal of 50 ml, and then dried in an oven at 180°C for 48 h, cooled down and determined the total mercury (Hg_H). The difference between Hg_T and Hg_H is gaseous mercury (Hg_G) (Lechler *et al.*, 1997); the difference between Hg_T and Hg_G is combined mercury (Hg_C). Usually, gaseous mercury is more chemical reactive and released from soil easily (Lechler *et al.*, 1997). The European reference material (BCR, CMR143) is served as the quality control. All measurements were replicated three times and analyzed using SPSS 11.0 for Windows.

2 Results and discussion

2.1 Distribution of Hg in topsoil samples from different pits

Contents of moisture, pH, TOC, Hg_T , Hg_G ; Hg_C of topsoil samples from different pits are listed in Table 1. Moisture content of topsoil is below 4.0%. The topsoil is weak alkali with the averaged pH of 8.58 (range 8.14–9.66). TOC ranges 2.99–29.48 g/kg, with a average of 11.74 g/kg. The Hg_T mean value of topsoil samples is the same as the national soil mean value (71 $\mu\text{g/kg}$) (Xia *et al.*, 1984), but most are lower, except for two exceptionally high values (Table 1). We repeated these two samples and got the same results.

There was a strongly negative correlation between moisture content and pH value ($r=-0.7173$). High correlative coefficients were found among TOC, Hg_T , Hg_G and Hg_C (Table 2), but low between Hg and moisture content. Hg_T and Hg_C decreased with filling time during 1998–2001, but no significant relationship was found between Hg content and filling time (Table 1).

Hg level in topsoil was influenced by its own properties (such as TOC, mineral composition), and degradation degree (filling time) as well. TOC content affects strongly enrichment and transportation of Hg (Wang *et al.*, 1997). More organic matters produced during the degradation tend to combine with Hg, and store in soil.

Garbage degradation has a great effect on Hg releasing. Complicated degradation occurs after garbage is buried. William (1986) divided garbage degradation into three periods: aerobic biodegradation, facultative biodegradation, and anaerobic biodegradation. Garbage degradation was divided into five periods: adjusting period; transitional period; acid-forming period; methyl-forming period; and maturity period (Yuan *et al.*; Raloff, 2001). Hg-bearing garbage degrades in different rate, for examples, Hg in crushed fluorescent lights can be discharged into environment quickly, but that in battery and thermometers is hard to release (Clear and Berman, 1994; Tetra Tech Inc. and Frontier Geosciences Inc., 1994).

2.2 Distribution of Hg in profile samples

Three profile samples were selected to study the transportation of Hg in topsoil of pits with different filling time (1992, 1996, 2000, respectively). The contents of moisture, TOC, and different Hg species in profile samples are shown

Table 1 Properties of topsoil samples

Filled time	Moisture (%)	pH	TOC (g/kg)	Hg_C ($\mu\text{g/kg}$)	Hg_T ($\mu\text{g/kg}$)	Hg_G ($\mu\text{g/kg}$)	Hg_G/Hg_T (%)
1989	3.05	8.14	21.13	55.70	71.97	16.27	22.61
1990	1.87	8.22	12.18	31.46	47.18	15.72	33.32
1991	1.69	8.26	13.05	36.74	62.70	25.96	41.40
1992	1.09	8.76	5.05	15.72	21.89	6.17	28.19
1993	0.82	9.66	2.99	20.84	24.32	3.48	14.31
1994	3.13	8.15	29.48	199.89	250.80	50.91	20.30
1995	1.19	9.28	6.25	22.93	28.47	5.54	19.46
1996	1.67	8.24	12.57	21.93	30.93	9.00	29.10
1997	2.69	8.49	19.86	206.58	259.80	53.22	20.48
1998	2.55	8.34	3.84	13.08	18.21	5.13	28.17
1999	1.82	8.75	6.45	24.53	25.20	0.67	2.66
2000	1.14	8.81	12.90	32.34	37.63	5.29	14.06
2001	1.63	8.38	6.84	35.22	46.92	11.70	24.94
Mean	1.87	8.58	11.74	55.15	71.23	16.08	22.57
SD	0.76	0.47	7.85	66.63	83.26	17.32	20.80
CV	42%	6%	67%	121%	117%	108%	92.31%

Table 2 Correlations among TOC, Hg_T, Hg_G and Hg_C in topsoil samples (*n*=13)

	TOC	Hg _C	Hg _T	Hg _G
TOC	1			
Hg _C	0.8045**	1		
Hg _T	0.8147**	0.9979**	1	
Hg _G	0.8213**	0.9497**	0.9681**	1

**Correlation is significant at the 0.01 level (2-tailed).

in Fig.1, and their mean value, standard deviation (*SD*), coefficient of variation (*CV*) are listed in Table 3.

The Laogang landfill was put into use in 1989, but it took 22–23 years for the garbage to degrade completely (Zhao *et al.*, 2000). Hg content in the three profiles changed irregularly (Fig.1). (1) In profile 2000, Hg_T and Hg_C were higher in the top and bottom than those in the middle, and Hg_G was higher in surface (0–6 cm) of the profile and lower in the deep section (>6 cm) of the profile. TOC decreased with depth and Hg_T was significant correlated with Hg_C. With a *CV* of 41.28%, Hg_G was small proportion to Hg_T (11.35%), which showed that only small gaseous Hg formed in the young pit, and garbage degradation just started. (2) In profile 1996, Hg_T and Hg_C decreased irregularly with depth, two exceptionally high values appeared in depth of 9 cm and 21 cm. There was no apparent trend with Hg_C and TOC. Hg_T was significant correlated with Hg_G, meanwhile, with a highest *CV* of 56.95%, Hg_G was about 1/3 of Hg_T. It might show that garbage degradation was actively. (3) In profile 1992, Hg_T and Hg_C increased with depth, and Hg_G was higher in the middle than those in the top and bottom. TOC was not apparent regularity, but Hg_T was significant correlated with Hg_C. With a lowest *CV* of 24.76%, Hg_G was 34.15% of total Hg, and it was the highest in the three profiles. It demonstrated that pit 1992 still was in active degradation period after 10 years, but it was different with pit 1996. Large quantity of gaseous Hg forming in degradation became more stable, and therefore, total Hg was dominated by combinative Hg.

In the three profiles, average Hg_T was closely, but average Hg_G was discrepant widely. Hg_G decreased in the order of pit 1996 (12.87 µg/kg)>pit 1992 (9.34 µg/kg)>pit 2000 (3.67 µg/kg). *CV* of Hg_T (20.68%–29.10%) decreased with increasing of filling time. *CV* of Hg_G changed greatly from 24.76%–56.95%. Hg_T was significant with Hg_C in profile 1992 and profile 2000, and Hg_T was significant with Hg_G in profile 1996 (Table 4). Ratio of Hg_G/Hg_T in profile samples decreased in the order of (Hg_G/Hg_T)₁₉₉₂>(Hg_G/Hg_T)₁₉₉₆>(Hg_G/Hg_T)₂₀₀₀. It is

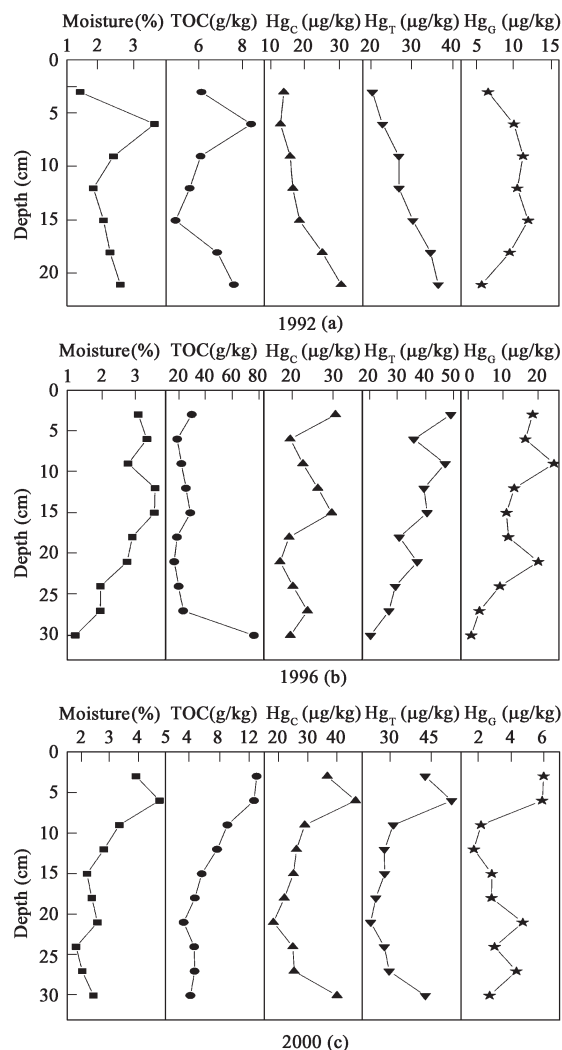


Fig. 1 Contents of TOC, Hg_T, Hg_G and Hg_C in different profiles. (a) profile 1992, (b) profile 1996, (c) profile 2000.

interesting to find that there was not significant relationship between TOC and Hg (Hg_T, Hg_G and Hg_C) in profile 1992 and 1996. The reason still remained unknown.

There is scant information on old pits, outline of Hg release in landfill could be drawn from above properties: gaseous mercury is formed and released from landfill after garbage is buried, at the beginning of degradation (such as pit 2000), gaseous mercury is only small proportion to total mercury, and total mercury is depend on combinative mercury; with increasing of filling time, degradation becomes more active, and reaches a peak period, in which

Table 3 Mean, *SD*, *CV* of Hg_C, Hg_T, Hg_G, Hg_G/Hg_T, moisture, and TOC in profiles (the year 1992, 1996, 2000)

Year		Moisture (%)	TOC (g/kg)	Hg _C (µg/kg)	Hg _T (µg/kg)	Hg _G (µg/kg)	Hg _G /Hg _T (%)
1992	Mean	2.37	6.52	19.01	28.35	9.34	34.15
	<i>SD</i>	0.63	1.19	6.55	5.86	2.31	9.86
	<i>CV</i>	26.64%	18.27%	34.43%	20.68%	24.76%	28.86%
1996	Mean	2.70	27.75	22.73	35.60	12.87	33.67
	<i>SD</i>	0.76	17.38	4.64	8.94	7.33	15.90
	<i>CV</i>	28.04%	62.63%	20.42%	25.12%	56.95%	47.21%
2000	Mean	2.84	7.06	29.46	33.13	3.67	11.35
	<i>SD</i>	0.930	3.50	8.91	9.64	1.52	4.42
	<i>CV</i>	32.81%	49.60%	30.25%	29.10%	41.28%	38.98%

Table 4 Correlations among moisture, TOC, Hg_C, Hg_T, and Hg_G in profiles (the year 1992, 1996, and 2000)

Year		Moisture (%)	TOC (g/kg)	Hg _C (μg/kg)	Hg _T (μg/kg)	Hg _G (μg/kg)
1992	Moisture (%)	1				
	TOC (g/kg)	0.7797*	1			
	Hg _C (μg/kg)	0.0418	0.2133	1		
	Hg _T (μg/kg)	0.1013	0.0605	0.9364**	1	
	Hg _G (μg/kg)	0.1383	-0.4502	-0.4566	-0.1152	1
1996	Moisture (%)	1				
	TOC (g/kg)	-0.5876	1			
	Hg _C (μg/kg)	0.4578	-0.0022	1		
	Hg _T (μg/kg)	0.7441*	-0.4701	0.5759	1	
	Hg _G (μg/kg)	0.6182	-0.5723	0.0696	0.8556**	1
2000	Moisture (%)	1				
	TOC (g/kg)	0.9089	1			
	Hg _C (μg/kg)	0.7231*	0.6781*	1		
	Hg _T (μg/kg)	0.7599*	0.7049*	0.9897**	1	
	Hg _G (μg/kg)	0.5815	0.4962	0.4141	0.5401	1

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

gaseous mercury becomes more and more and dominates total mercury (such as pit 1996); and then, degradation enters a relative active period, in which quantity of gaseous mercury becomes stable, and total mercury is depend on combinative mercury again (such as pit 1992); finally, degradation is over, and garbage enters maturity period, in which gaseous mercury will become less and less.

2.3 Distribution of Hg in plants

Hg levels in root, stem, leaf of a representative plant, *Solidago canadensis*, from different pits are shown in Table 5, and those of other species from pit 1994 are shown in Table 6. Among these plants, *Solidago canadensis*, *Nerium indicum*, *Cania indica*, and *Ilex purpurea* are herbaceous plant, *Chenopodium album*, *Setaria viridis*, *Sesbania cannabina*, *Phyllostachys glanca*, *Trachycarpus fortunei*, *Ligustrum lucidum*, and *Prunus salicina*, are ligneous plants.

Average of mercury contents in root, stem, and leaf of *Solidago canadensis* respectively are 2.67, 1.55 and 23.05 μg/kg. Hg content of plant components decreases in the order of leaf>root>stem. Although Hg levels of stem and leaf increased with filling time for *Solidago canadensis* growing in old pits (1989–1993), no obvious relationship

Table 5 Mercury content in (*Solidago canadensis*) plants from different landfill pits (μg/kg, fresh weight)

Year	Root	Stem	Leaf
1989	3.45	5.84	27.90
1990	3.86	1.35	24.08
1991	3.89	1.72	26.32
1992	3.86	1.63	25.31
1993	3.81	1.14	23.48
1994	2.85	1.03	24.43
1995	1.69	1.62	14.52
1996	2.51	0.69	31.58
1997	2.50	1.02	23.50
1998	2.09	1.05	16.90
1999	1.22	1.02	26.16
2000	1.36	1.30	20.93
2001	1.58	0.79	14.57
Mean	2.67	1.55	23.05
SD	1.03	1.33	5.10
CV	38.41%	85.58%	22.12%

*Collected from pit 1994 (n=15).

Table 6 Mercury levels of different plants from the same pit of 1994 (μg/kg, fresh weight)

Sample	Root	Stem	Leaf
<i>Solidago canadensis</i>	2.85	1.03	24.43
<i>Chenopodium album</i>	0.82	0.25	17.10
<i>Setaria viridis</i>	2.88	1.04	7.87
<i>Sesbania cannabina</i>	4.69	0.60	6.94
<i>Ilex purpurea</i>	6.86	0.79	12.47
<i>Nerium indicum</i>	2.85	0.52	7.04
<i>Phyllostachys glanca</i>	10.02	2.07	20.13
<i>Cania indica</i>	/	0.85	13.74
<i>Trachycarpus fortunei</i>	/	2.17	20.47
<i>Ligustrum lucidum</i>	/	2.14	34.00
<i>Prunus salicina</i>	/	1.88	13.80

“/” no detected.

was found between Hg content of plant components and soil properties (moisture, TOC, Hg_T, Hg_G and Hg_C).

Distribution patterns of other plants are consistent with that of *Solidago canadensis*. For the same species, the pattern is following the order: leaf>root>stem. Hg content of leaf is as 23.7 times high as that of stem for *Solidago canadensis*, and it can reach as high as 68.4 times for *Chenopodium album*. Plants may assimilate and enrich heavy metals massively from air (Pan *et al.*, 1987; Chen *et al.*, 2002). Wang and Mou (1999) found that Hg content in soil and plant increased with the rising atmospheric Hg content, and Hg accumulation in soil-plant system was strongly influenced by atmospheric mercury; Hg content of plant aerial parts mainly comes from the atmosphere, and root mercury comes from soil. The higher the atmospheric Hg content, the higher the plant aerial parts relative to corresponding underground parts (Wang and Mou, 1998). Leaf is the main organ to absorb and enrich Hg from the atmosphere.

Previous studies indicated that distribution of Hg in plants was inhomogeneous (Xu *et al.*, 1995; Wang *et al.*, 1997). Total gaseous mercury occurs at concentrations in the μg/m³ range and is released into the atmosphere from landfill (Lindberg *et al.*, 1999, 2005). Considering pipes are equipped for degassing from underground during garbage degradation, atmospheric mercury was higher than normal. The fact that mercury content of plant's aerial parts is higher than that of underground parts shows

exceptionally atmospheric mercury.

Stem mercury of ligneous plants such as *Phyllostachys glanca*, *Ligustrum lucidum*, and *Trachycarpus fortunei*, was 2–8 times higher than that of herbaceous plants. And, root mercury of *Phyllostachys glanca* is much higher than that of herbaceous plants such as *Solidago canadensis*, *Chenopodium album*, and *Setaria viridis*. By comparison, woody plants have stronger capacity of enriching Hg than herbaceous plants.

3 Conclusions

For topsoil samples, there were significant correlations among total mercury (Hg_T), combinative mercury (Hg_C) and gaseous mercury (Hg_G), and content of total organic carbon (TOC), whereas, no significantly relationship was found between Hg content and filling time.

With similar Hg_T , the average Hg_G was quite different in different profile. Hg_G/Hg_T ratio in profile samples decreased in the order of $(Hg_G/Hg_T)_{1992} > (Hg_G/Hg_T)_{1996} \gg (Hg_G/Hg_T)_{2000}$.

Distribution of Hg in plants followed the order of leaf>root>stem. Ligneous plants are capable of enriching more Hg than herbaceous plants. *Phyllostachys glanca*, *Prunus salicina* and *Ligustrum lucidum* are good example plants.

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