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Mercury air-borne emissions from 5 municipal solid waste landfills in Guiyang and Wuhan, China

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Abstract

A detailed study on atmospheric mercury emissions from municipal solid waste (MSW) landfills in China is necessary to understand mercury behavior in this source category, simply because China disposes of bulk MSW by landfilling and a large quantity of mercury enters into landfills. Between 2003 and 2006, mercury airborne emissions through different pathways, as well as mercury speciation in landfill gas (LFG) were measured at 5 MSW landfills in Guiyang and Wuhan, China. The results showed that mercury content in the substrate fundamentally affected the magnitude of mercury emissions, resulting in the highest emission rate (as high as 57 651 ng Hg m^{-2} h^{-1}) at the working face and in un-covered waste areas, and the lowest measured at soil covers and vegetation areas (less than 20 ng Hg m^{-2} h^{-1}). Meteorological parameters, especially solar radiation, influenced the diurnal pattern of mercury surface-air emissions. Total gaseous mercury (TGM) in LFG varied from 2.0 to 1406.0 ng m^{-3}, monomethyl mercury (MMHg) and dimethyl mercury (DMHg) in LFG averaged at 1.93 and 9.21 ng m^{-3}, and accounted for 0.51% and 1.79% of the TGM in the LFG, respectively. Total mercury emitted from the five landfills ranged from 17 to 3285 g yr^{-1}, with the highest from the working face, then soil covering, and finally the vent pipes.

1 Introduction

Mercury (Hg) emissions from municipal solid waste (MSW) incineration are regarded as one of the most important anthropogenic mercury sources to the atmosphere (US EPA, 1997; van Velzen et al., 2002). However, about 70% of the MSW globally are deposited of by landfills, due to their low cost and low maintenance (OECD, 2001). In the past few years, some researches disclosed that landfill also served as a potential atmospheric mercury source (e.g., Lindberg and Price, 1999; Kim et al., 2001), and more importantly, this site is an effective bioreactor which convert inorganic mercury into methylmercury, the most toxic form of mercury (Lindberg et al., 2001, 2005a;

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Hawkins and Prestbo, 2004; Prestbo et al., 2003).

China is one of the largest MSW producers in the world, generating over 150 million tonnes MSW per year, which accounts for 29% of the global total. Landfill is the main disposal method for MSW in China, treating 68.9 million tonnes at 444 landfills in 2004, accounting for 85.4% of the amount which been treated (including landfill, incineration and compost, the rest was not treated, Huang et al., 2006; Liu et al., 2007). The area occupied by MSW landfills in China is about 500 km^2, and the volume of buried MSW has reached up to 664 million m^3 in 2008. Mercury enters the landfill mainly through mercury-containing waste, such as batteries, fluorescent lamps and thermometers (US EPA, 1992). Between 1992 and 1999 in China, 185–802 tonnes of mercury was leached into the environment from discarded batteries (Yang et al., 2003). Although Hg content in batteries was lowered since 2001, 153 tonnes of Hg were still used in batteries in 2004 (Jian et al., 2008). In fluorescent lamps and thermometers, about 200 tonnes of Hg are used each year (Hao and Shen, 2006; Shen and Jian, 2004). Because most (over 90%) of the Hg-containing products were not recycled in China (Yu and Li, 2004), they end up in landfills. Hg emissions from other anthropogenic sources in China, such as coal combustion (Wang et al., 2000; Tang et al., 2007; Wang et al., 2009) and nonferrous metal smelting (Feng et al., 2004a; Li et al., 2009) have been extensively studied, but little information is known about mercury emissions from landfill sites. Unlike other western countries, there are very few facilities that utilize landfill gas (LFG) in China (Huang and He, 2008). As a result, almost all the LFG is emitted into the atmosphere directly, which poses a severe ecological risk. Thus, mercury emissions from landfill sites in China deserve investigation.

In this paper we report the results of mercury emissions from 5 MSW landfills in Guiyang and Wuhan city, China, sampled between 2003 and 2006. Hg emissions were measured from two pathways, i.e., landfill surface and landfill vent pipes, and Hg species in LFG, including total gaseous Hg (TGM), monomethyl Hg (MMHg) and dimethyl Hg (DMHg), were also measured.

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2 Experimental methods

2.1 Landfills studied

Locations of five investigated landfills are given in Fig. 1, basic information is listed in Table 1, and photos showing the sampling sites are shown in Fig. 2. Three landfills are located in Guiyang (capital of Guizhou province), namely Gao-Yan (G-Y), Da-Zhuan-Wan (D-Z-W) and Xian-Ren-Jiao (X-R-J) landfill, and two in Wuhan (capital of Hubei province), namely Jin-Kou (J-K) and Dai-Shan (D-S) landfill. Due to its karstic landscape, landfills in Guiyang are located in valleys, while landfills in Wuhan are located in flat areas. Guiyang has a population of 3.3 million and produces 2100 tonnes of MSW per day, while Wuhan has a population of 7.8 million and produces 6065 tonnes of MSW per day. The climate of both cities is a typical north subtropical monsoon.

Of the five landfills, two were sanitary landfills (G-Y and J-K), and three (D-Z-W, X-R-J and D-S) were simple landfills. During the study period, three landfills (G-Y, J-K and D-S) were in active operation, while the others were closed. A passive LFG vent system was installed at G-Y and J-K landfills, discharging LFG directly into the ambient air (see Fig. 2d), while the other three landfills did not employ the vent pipe system. The landfills studied are representative of Chinese landfills, since they include different types (sanitary or simple), different stages (operational or closed), and different surface types.

2.2 Sampling and analysis methods

The field sampling campaigns were carried out between 2003 and 2006 at G-Y landfill, while the other 4 landfills were measured in 2004. Monitoring focused on two aspects, namely Hg surface-air flux and Hg speciation.

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2.2.1 Hg surface-air flux

Two methods were applied to measure Hg surface-air flux, i.e., a dynamic flux chamber (DFC) method for the non-working face area (Fig. 2c), and a Gaussion plume model for the working face area, where the MSW was dumped, spread, crushed and covered with soil. The principal of DFC measured in conjunction with an automated mercury vapor analyzer (Tekran 2537A) was described elsewhere (Feng et al., 2005; Wang et al., 2005). Briefly, the DFC was a bottom opened, semi-cylinder shaped (Φ20×30 cm) quartz glass chamber, and the turnover time of the flux chamber is about 0.16 min. Hg surface-air flux, with a time resolution of 20 min, was calculated by the difference in TGM concentrations between the outlet and inlet of the chamber, as well as the air flow rate through the chamber and the area covered by it. Tekran 2537A is a sensitive analyzer for TGM with a very low detection limit (about 0.1 ng m^{-3}, e.g., Tekran, 1998), and calibrated by its interior mercury vapour source. The chamber blank was 0.5±1.8 ng m^{-2} h^{-1} (n=77).

The Gaussion plume model (US EPA, 1995) is the ISCST3 model (Industrial Source Complex, Short-Term, Version 3), and is expressed as follows,

$$C(x,y) = \frac{Q}{\pi\sigma_y\sigma_z U}\exp\left[-\frac{y^2}{2\sigma_y^2} - \frac{(z-h)^2}{2\sigma_z^2}\right] \quad (1)$$

where $C(x,y)$ is the pollutant atmospheric concentration at dimensions of (x,y); Q is the pollutant emission rate from the source; U is the horizontal wind speed; h is the effective height of pollution source; σ_y and σ_z are horizontal and vertical dispersion coefficient, respectively; x, y, z are the three dimensions.

The working face was treated as a ground surface source, thus Hg emission rate can be deduced from the meteorological parameters, and atmospheric Hg concentration at downwind and upwind working face sites. The dispersion coefficient was calculated through the onsite meteorological parameters.

2.2.2 Hg speciation in the LFG

Hg speciation in the LFG, including TGM, MMHg and DMHg, was determined at G-Y landfill, while only TGM was measured at J-K landfill. The sampling and analysis methods for mercury speciation were described by Lindberg et al. (2001) and Bloom et al. (2005). TGM was measured by Tekran 2537A onsite at a 5 min interval, and MMHg and DMHg were trapped by diluted HCl (0.5% v/v in double de-ionized water) and Carbotrap^{TM} adsorbent (20/40 mesh, Supelco Inc., Bellefonte, PA), respectively, with sampling time more than 2 h. Then the trapped methylated samples were transported to the laboratory and determined by gas-chromatography separation, and CVAFS detection (Tekran Model 2500, Canada). Excessive moisture in LFG was removed by passing the sample through a water trap (a combination of an empty impinger in ice bath and a soda lime desiccant). The Carbotrap^{TM} was wrapped with aluminum foil during the sampling and storage period to avoid decomposition of the analyte.

MMHg in the LFG was quantified by the MMHg standard solution (supplied by CEBAM Cebam Analytical, Inc., WA, USA), and DMHg was calibrated by purging an aliquot of DMHg standard solution (supplied by Jožef Stefan Institute, Ljubljana, Slovenia) on to the Carbotrap^{TM}, and analyzed similar to that of samples. During each campaign, the field blank samples for MMHg and DMHg were also collected. The method detection limits for MMHg and DMHg were found to be 0.5–0.6 pg (as mercury) absolute or 1.4–1.7 pg m^{-3} (for 2 h sampling) based on 3 times deviation of the blank samples.

2.2.3 Other parameters

In addition, Hg content, pH and organic matter (OM) in the MSW and cover soil of the 5 landfills, TGM in the ambient air above the landfill surface (0.1–2.0 m high), and meteorological parameters, including air/soil temperature, relative humidity, solar radiation, wind speed and wind direction were also monitored to characterize the behavior of Hg emissions from landfill sites. The meteorological parameters were monitored by using

a portable weather station (Global Water IIIB, USA). TGM in the ambient air was determined onsite by Tekran 2537A. Hg content in the MSW and cover soil was analyzed by CVAFS detection (Tekran Model 2500, Canada) after aqua regia ($HCl+HNO_3$ 3:1 v/v) digestion. While, pH and organic matter in the MSW and cover soil were measured by a pH meter in a 2.5:1 (v/m) water/solid suspension and potassium dichromate method, respectively.

3 Results and discussion

3.1 Hg, pH and OM in the MSW and cover soil

Table 2 includes Hg, pH and OM content in the MSWs and cover soils. Hg in MSW was characterized by a significant variability (Guiyang, range: 0.170–46.222 mg kg^{-1}, average: 1.796±7.072 mg kg^{-1}, N=42; Wuhan, range: 0.240–1.271 mg kg^{-1}, average: 0.606±0.349 mg kg^{-1}, N=8). The result for Wuhan was very similar to that reported by Fang and Hong (1988), where Hg concentrations in MSW of Wuhan ranged from 0.13 to1.53 mg kg^{-1} and averaged at 0.52 mg kg^{-1} (N=20). Hg concentrations in cover soils, in general, were more convergent, with averages of 0.310 and 0.058 mg kg^{-1} for Guiyang and Wuhan, respectively. These values are similar to the typical background levels, including 0.222 mg kg^{-1} for Guiyang (Wang, 2004) and 0.056 mg kg^{-1} for Wuhan (Fang and Hong, 1988). A few cover soil samples in D-Z-W landfill contained high Hg (3.124–6.527 mg kg^{-1}), which may be due to unauthorized dumping of MSW that occurred at this landfill after its closure. Hg concentration in MSW was obviously higher compared to the cover soils, which possibly reflected higher mercury-contained substances in the MSW. Organic matter content and pH in MSW were also much elevated than those of cover soils (Table 2), due to the mingling of kitchen waste (such as food remnants and leaves, etc) and coal ash (pH 7.5–12.1, from the domestic cooking and heating), respectively.

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3.2 TGM in the atmosphere above the landfill

Figure 3a shows the ranges of TGM concentration observed in the ambient air over the landfills studied. The range of TGM was from 1.6 to 473.7 ng m^{-3}, with averages (Fig. 3b) at different sites ranging from 8.5 to 155.7 ng m^{-3}. The highest TGM concentrations occurred at the working face and the downwind area for all three landfills (G-Y, J-K and D-S), where TGM was sensitive and proportional to the activities of MSW treating at the working face, as observed at operational landfills in Florida, USA (Lindberg et al., 2005b). Lowest TGM was measured at the closed landfill of X-R-J, where the whole landfill was planted with grass and trees, and this value was close to average TGM concentrations in ambient air of Guiyang (8.4 ng m^{-3}, from Feng et al., 2004b). Since the landfills were located far from other urban Hg emission sources, the elevation of TGM concentrations in the ambient air was predominantly due to the landfill emissions.

3.3 Hg surface-air fluxes

3.3.1 Non-working face areas

Hg surface-air fluxes at the non-working face areas, as determined by the DFC method, are listed in Table 3. The flux indicated large variability from site to site, ranging from −286.2 to 5609.6 ng m^{-2} h^{-1}, with highest averages of about 500–600 ng m^{-2} h^{-1} for the un-covered MSW sites (site F6, F10 in Table 3) and the contaminated soil cover area (site F1), while the lowest was observed at soil covers (site F13, F17) and the grass planted area (F18) with average rates about −1 to 20 ng m^{-2} h^{-1}. Hg flux was clearly higher during the warm season for the same surface type, such as "un-covered MSW" sites at warm season (F6 and F10) versus cold season (F14 and F15), and "temporary soil cover" sites at warm season (F8 and F9) versus cold season (F12 and F13).

The detailed processes of Hg surface-air flux, as well as the concurrent meteoro-

logical parameters at each type of surface are illustrated in Figs. 4–6. Among all parameters, a strong diel cycle with a daytime maximum was observed. Figure 4 shows the contributions of high Hg content (2.313 mg kg^{-1}) in the MSW to the emission rate. Figure 5 shows how vegetation reduced the mercury emission rate, since the mercury surface-air flux at grass planted area of X-R-J was obviously lower than that of two soil covers (with no plantation) at D-Z-W, although under similar weather conditions and with similar mercury contents in the substrate (0.477–0.575 mg kg^{-1}) among three sites. At two similar soil cover sites of J-K landfill, Hg flux was several folds higher under sunny conditions compared to cloudy and rainy conditions (Fig. 6).

Compared with other studies, Hg emissions from soil covers were higher than those from several landfills in Florida, USA, which ranged from 1–20 ng m^{-2} h^{-1} (Lindberg and Price, 1999; Lindberg et al., 2005b), but lower compared to a large closed landfill in Seoul, Korea, which averaged 254±224 ng m^{-2} h^{-1} (Kim et al., 2001). For the uncovered waste, our results obtained in winter 2006 (57.5–84.5 ng m^{-2} h^{-1}) were similar to those measured in an American landfill (70±62 ng m^{-2} h^{-1}, Lindberg et al., 2005b). When compared with the mercury emission rate at local and global background sites (typically less than 30 ng m^{-2} h^{-1}, Wang et al., 2004; Poissant and Casimir, 1998), Hg emitted from the landfill soil cover was the same or several times higher, while the uncovered waste was up to several hundreds times higher.

3.3.2 Working face area

The calculated Hg emission rate by the Gaussion plume model indicated that the emission rate varied from one to two orders of magnitude between landfills (1.9 mg Hg h^{-1} at D-S landfill to 369.0 mg Hg h^{-1} at G-Y landfill, Table 4). This hinted that Hg emissions from working face was correlated to MSW disposal rate (Table 1), Hg content in MSW (Table 2), and the weather conditions at each landfill (Table 4). Hg emission rates at the working face can be deducted when taken into account the MSW disposal rate during the sampling period. The calculated emission factors for D-S, J-K and G-Y landfill were 0.04, 0.63, 6.81 mg Hg $tonnes^{-1}$ MSW disposed, respectively. These results were con-

sistent with those from 7 American landfills (0.7–6.6 mg tonnes^{-1}, Lindberg and Price, 1999; Lindberg et al., 2005b). Hg emission factors indicated that 0.07‰–3.78‰ Hg in MSW was released into ambient air through the working faces, with an average loss rate of 1.63‰. When combined with the emission rate and the actual area of the working face, Hg emissions from the unit area of the working face can been obtained, with a maximum of 57 651 ng m^{-2} h^{-1} at G-Y landfill, which was comparable to a landfill in Florida, USA (70 000 ng m^{-2} h^{-1}, Lindberg and Price, 1999). The results showed that the working face has the highest intensity for Hg emissions among landfill surfaces.

3.3.3 Factors influencing Hg surface-air fluxes

From data in Tables 2 and 3 and Figs. 4–6, it can be easily observed that Hg content in the substrate plays a fundamental role in Hg emission among different sites, as described by other researchers (Gustin et al., 2000; Wang et al., 2005). In this study, Hg in the substrate (MSW or cover soil) showed a Log-Log linear relationship with Hg flux (Fig. 7). Because Hg concentration in MSW was obviously higher than that of the cover soils (Table 2), Hg emissions from uncovered MSW and working face areas were much higher than that of covering soils. The soil cover was found to be an effective barrier preventing mercury emissions.

For the same sampling site, weather conditions affected Hgy fluxes (Figs. 4–6). For example, the flux decreased nearly 50% between sunshine to moderate sunshine conditions at site F6 as shown in Fig. 4. Correlation analysis showed that Hg flux was significantly correlated with solar radiation (r=0.852, p<0.001), followed by soil temperature (r=0.532, p<0.001), air temperature (r=0.347, p<0.001), wind speed (r=0.172, p<0.001), and inversely correlated with relative humidity (r=−0.682, p<0.001).

3.4 Hg species in the landfill gas

3.4.1 TGM

The concentration of TGM in the LFG of 27 vent pipes at G-Y ranged from 2.0 to 1406.0 ng m^{-3}, and 5.0 to 74.0 ng m^{-3} for 6 vent pipes at J-K landfill, respectively. Large variations were observed among different pipes as showed in Fig. 8. Interestingly, for the first time, we found that TGM in the LFG varied under different weather conditions. TGM was much lower and constant on sunny days, but increased sharply during the rainfall (Fig. 9). This phenomenon of increasing TGM during rain events may be due to the following three reasons. First of all, Hg in soil pore air inside the landfill was replaced by rain water. Secondly, the emission pathway of landfill surface was blocked by the rainfall, more LFG was discharged through the vent pipe system. Thirdly, the atmospheric pressure dropped on rainy days, which promoted LFG emission from the passive vent pipes. Many studies reported the flow rate of LFG from passive vent pipes was susceptible to the fluctuation of atmospheric pressure (Gebert and Groengroeft, 2006; Maurice and Lagerkvist, 2003), which generally decreased during the rainfall. We observed an obvious LFG plume from the vent pipes during rainy days as shown in Fig. 2d, whereas imperceptible emissions were found on sunny days. TGM concentrations in LFG increased several fold during the rainfall (Fig. 9), while Hg surface-air flux declined during the rainfall (Fig. 6).

Compared with TGM concentrations in LFG measured at some American landfills, our results were much lower (Lindberg and Price, 1999; Lindberg et al., 2001, 2005a; Hawkins and Prestbo, 2004; Prestbo et al., 2003). In the latter sites, TGM concentrations was approximately at μg m^{-3} level, with the highest of about 12 μg m^{-3}. The results obtained from G-Y and J-K landfill were comparable to landfills in Sweden (Sommar et al., 1999), Germany (Feldmann et al., 1994), South Korea (Kim and Kim, 2002) and Mexico (de la Rosa et al., 2006), ranging from several to a few of thousand of ng m^{-3}. The great discrepancy of TGM in LFG among different landfills may be due to mercury content in MSW and the method for LFG venting (i.e., active or passive

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system).

3.4.2 MMHg

MMHg in LFG of some vent pipes at G-Y landfill varied between 0.14 and 6.37 ng m^{-3}, with an average of 1.93 ng m^{-3} (Fig. 10). The percentage of MMHg to TGM ranged from 0.14 to 1.68%, with an average of 0.51%. The global background concentrations of MMHg in the atmosphere are generally below 10 pg m^{-3} (Munthe et al., 2003), thus the MMHg in LFG was about 3 orders of magnitude higher than the global background values in ambient air, showing landfill was an important MMHg emission source. MMHg concentrations of some American landfills ranged from 1–40 ng m^{-3}, accounting for ca. 0.1% of the TGM (Lindberg et al., 2001, 2005a). These concentrations were much higher than the data observed at G-Y landfill.

3.4.3 DMHg

For the same vent pipes sampled for MMHg, DMHg ranged from 2.54–19.05 ng m^{-3}, with an average of 9.21 ng m^{-3} (Fig. 11). DMHg comprised 0.27 to 3.64% of TGM in the LFG, with an average of 1.79%. DMHg was also detected in LFG in the USA with concentration between 0.2 to 637 ng m^{-3} (Hawkins and Prestbo, 2004; Prestbo et al., 2003; Lindberg et al., 2001, 2005a), which were much higher than that observed at G-Y landfill. DMHg is the most toxic mercury species (Nierenberg et al., 1998), and direct emissions from the landfill site could pose a serious ecological risk. It is highly recommended that LFG in China be utilized, or at least burned before it is discharged into the atmosphere. The latter method will decompose methylated Hg to elemental Hg at high temperatures, reducing their toxicity.

4 Summary and conclusions

Based on field experiments, Hg emission patterns from landfills were estimated (Table 5). Total Hg emissions from the five landfills in 2004 ranged from 17 to 3285 g yr^{-1}, with the highest at G-Y landfill, and the lowest at X-R-J landfill. At G-Y, Hg emissions were dominated by the working face, which accounted for 98.36% of the total, followed by soil cover (1.28%), uncovered MSW (0.33%), and venting pipes (0.03%). A similar pattern was also found at J-K landfill. This confirmed that the working face was the leading source for Hg emissions from landfills, and total emissions from the vent pipes were relatively small. A rough picture for Hg emissions from all landfill sites in China can be obtained by taken into account the total MSW treated by landfill each year, total landfill surface area, total LFG generated and the field data we obtained from this research. The estimated Hg emissions from the Chinese landfill sites ranged from 500–800 kg yr^{-1} under different scenarios. These emission fluxes were relatively low compared to the total emissions of 552–696 tonnes Hg yr^{-1} from Chinese anthropogenic sources between 1995–2003 (Wu et al., 2006). However, based on the limited landfill surface, Hg emission intensity per unit area (up to 57 651 ng m^{-2} h^{-1}), Hg emissions from landfills still cannot be overlooked. In conclusion, Hg emissions from landfill sites depended on the mercury content in the substrate, were maximized at the working face, and were remarkably reduced by applying soil covering or vegetation. Hg emissions from the landfill surface were sensitive to meteorological parameters, especially solar radiation. In comparison to the vent pipe system, Hg emissions from landfill surfaces were the primary pathways. Methylated Hg species produced inside the landfill was especially important, indicated the environmental conditions (such as pH, Eh, T, O_2 level), microbial activity (such as sulfate-reducing bacteria which produce reduced sulfur compounds at landfill, Kim et al., 2005), nutrient levels, and the methyl group (CH_3–) donor, possibly enhanced Hg methylation. However, the exact mechanisms, whether biological or chemical, are still unknown and further research is needed.

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Table 1. Basic situation of the 5 studied MSW landfills.

City	Landfill site	Area (hectares)	MSW disposal rate (tonnes day^{-1})	Disposal method	Surface type	Operation period
Guiyang	G-Y	52	1300	Sanitary	Covered with temporary soil	2001–
	D-Z-W	10	600	Simple	Covered with soil and partially planted with trees	1983–2003
	X-R-J	10	600	Simple	Covered with soil and planted with grass and trees	1994–2001
Wuhan	J-K	40	2200	Sanitary	Covered with soil and partially planted with grass	1989–2005
	D-S	20	1000	Simple	Covered with soil and mostly planted with grass	1989–2007

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Table 2. Statistical summaries of Hg, pH and organic matter in the MSW and cover soil at 5 studied landfills.

Site and sample type	Analyte	Min	Max	Mean	Std
MSW					
Guiyang (N=42)	Hg (mg kg^{-1})	0.170	46.222	1.796	7.072
	pH	7.30	12.30	8.66	1.41
	OM (%)	4.91	17.84	9.82	3.24
Wuhan (N=8)	Hg (mg kg^{-1})	0.240	1.271	0.606	0.349
	pH	7.50	8.30	7.86	0.24
	OM (%)	5.29	12.10	7.70	2.46
Cover soil					
Guiyang-non-contaminated areas (N=23)	Hg (mg kg^{-1})	0.130	1.030	0.310	0.233
	pH	4.90	7.70	6.25	0.85
	OM (%)	0.31	5.58	1.32	1.58
Guiyang-contaminated areas at D-Z-W landfill (N=4)	Hg (mg kg^{-1})	3.124	6.527	5.132	1.473
	pH	8.10	8.30	8.23	0.10
	OM (%)	2.64	4.55	3.80	0.91
Wuhan (N=23)	Hg (mg kg^{-1})	0.037	0.099	0.058	0.014
	pH	7.10	7.90	7.59	0.23
	OM (%)	0.49	1.31	0.79	0.24

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Table 3. The statistical summary of Hg surface-air flux measured by the DFC method at different non-working face sites.

Landfill	Site	Site description	Sampling campaign	Weather condition	Mean±Std ($ng\,m^{-2}\,h^{-1}$)	Min-Max ($ng\,m^{-2}\,h^{-1}$)	Data no.
D-Z-W	F1	Contaminated soil cover	Mar 2004	Sunny	559.1±883.3	47.5–3866.5	85
D-Z-W	F2	Tree planted area	Mar 2004	Cloudy	88.2±136.2	−65.4–550.9	116
D-Z-W	F3	Permanent soil cover	Mar 2004	Rainy	53.6±46.0	−24.0–188.9	91
D-Z-W	F4	Permanent soil cover	Sep 2004	Sunny	50.7±55.5	−27.0–211.5	151
D-Z-W	F5	Permanent soil cover	Sep 2004	Sunny	112.8±108.1	5.7–391.7	181
G-Y	F6	Un-covered MSW	Nov 2003	Sunny	664.6±1341.2	−286.2–5609.6	164
G-Y	F7	Temporary soil cover	Nov 2003	Rainy	78.8±77.9	−72.48–308.7	105
G-Y	F8	Temporary soil cover	Sep 2004	Sunny	183.3±191.3	27.2–1273.3	144
G-Y	F9	Temporary soil cover	Sep 2004	Sunny	133.3±65.8	28.2–260.0	76
G-Y	F10	Un-covered MSW	Sep 2004	Sunny	537.7±485.1	63.2–2877.1	34
G-Y	F11	Temporary soil cover	Sep 2004	Cloudy	27.8±16.5	−16.1–89.0	71
G-Y	F12	Temporary soil cover	Jan 2006	Sunny	29.1±17.5	−47.6–91.0	165
G-Y	F13	Temporary soil cover	Jan 2006	Sunny	−1.4±26.2	−36.1–46.6	43
G-Y	F14	Un-covered MSW	Jan 2006	Sunny	57.5±83.4	−52.3–275.2	98
G-Y	F15	Un-covered MSW	Jan 2006	Sunny	84.5±88.5	−26.4–406.0	133
J-K	F16	Permanent soil cover	Jun 2004	Sunny	192.5±245.3	−40.2–1055.4	178
J-K	F17	Permanent soil cover	Jun 2004	Rainy	19.6±22.2	−47.2–358.8	235
X-R-J	F18	Grass planted area	Sep 2004	Sunny	19.7±27.3	−10.8–103.6	160

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Table 4. Hg emission rate from the working face of 3 operational landfills calculated by ISCST3 model.

Landfill	Sampling campaign	Down wind TGM ($ng\,m^{-3}$)	Distance from WF (m)	Wind speed ($m\,s^{-1}$)	Weather condition	Atmospheric stability*	Emission rate 1 ($mg\,Hg\,h^{-1}$)	Emission rate 2 ($ng\,Hg\,m^{-2}\,h^{-1}$)	Emission factor ($mg\,Hg\,t^{-1}$ MSW)
D-S	Jun 2004	55	50	0.30	Cloudy	D	1.9	291	0.04
J-K	Jun 2004	98	50	0.50	Sunny	A	58.2	9092	0.63
G-Y	Sep 2004	145	90	0.50	Sunny	A	369.0	57 651	6.81

* Determined by the weather conditions. There are six levels (i.e. A, B, C, D, E, F) of atmospheric stability.

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Table 5. Estimation of annual Hg emissions from the studied landfills in 2004.

Landfill	Emission pathways	Hg emission quantities ($g\,yr^{-1}$)	Percentage of each pathway (%)
G-Y	Soil cover	42.03	1.28
	Uncovered waste	10.95	0.33
	Working face	3231.35	98.36
	Vent pipes	0.93	0.03
	Subtotal	3285.25	100.00
J-K	Soil cover	364.34	41.76
	Working face	505.89	57.99
	Vent pipes	2.20	0.25
	Subtotal	872.43	100.00
D-Z-W	Soil cover	175.86	95.58
	Vegetation area	8.14	4.42
	Subtotal	184.00	100.00
D-S	Vegetation area	33.07	69.37
	Working face	14.60	30.63
	Subtotal	47.67	100.00
X-R-J	Vegetation area	17.53	100.00

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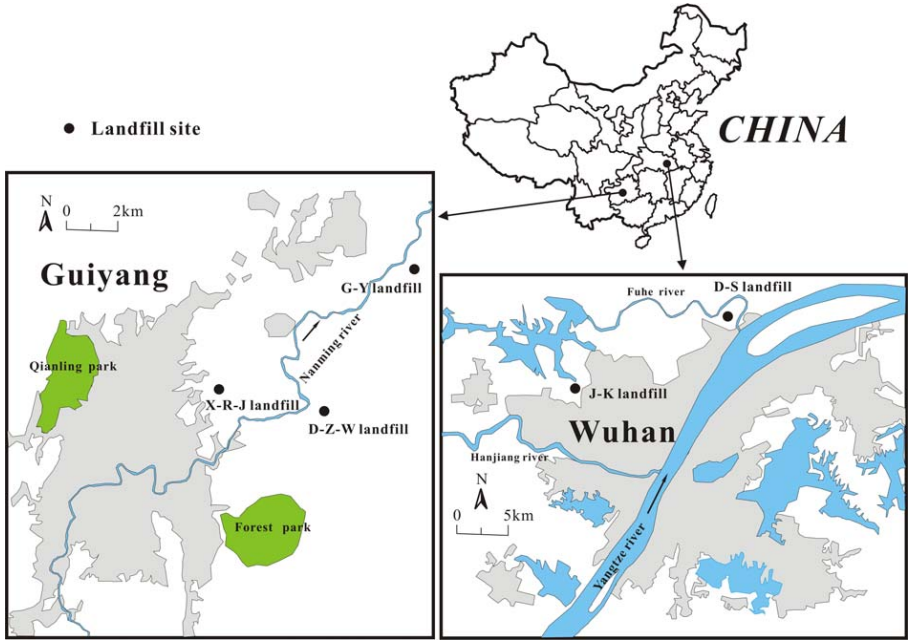

Fig. 1. Sketch map showing the locality of five studied landfills.

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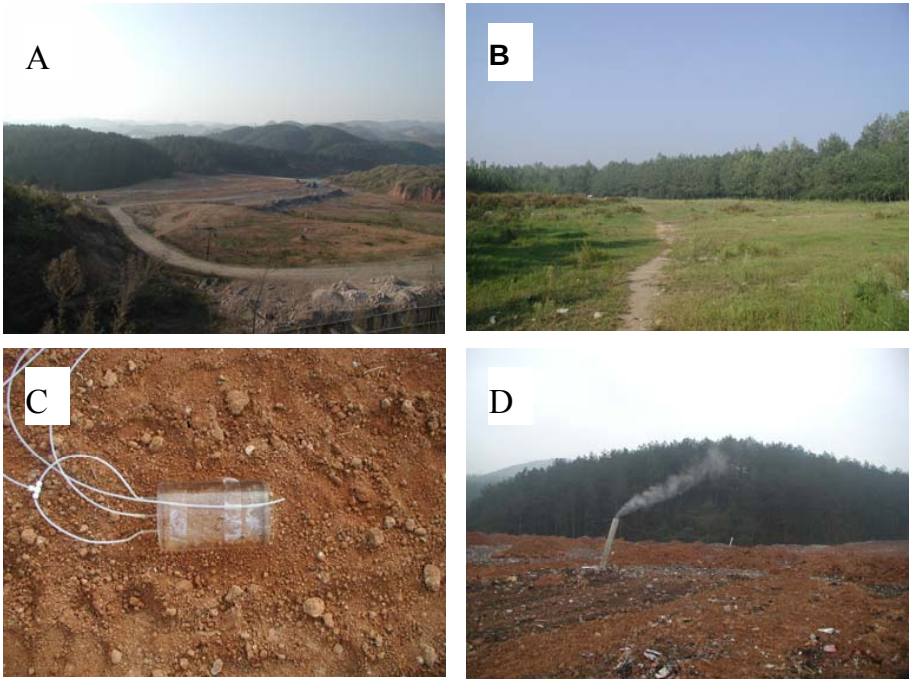

Fig. 2. Photos showing the operation landfill at G-Y **(A)** and closed landfill at X-R-J **(B)**, the DFC method for surface-air flux at the soil covering area **(C)** and the vent pipe system for the LFG at G-Y landfill **(D)**.

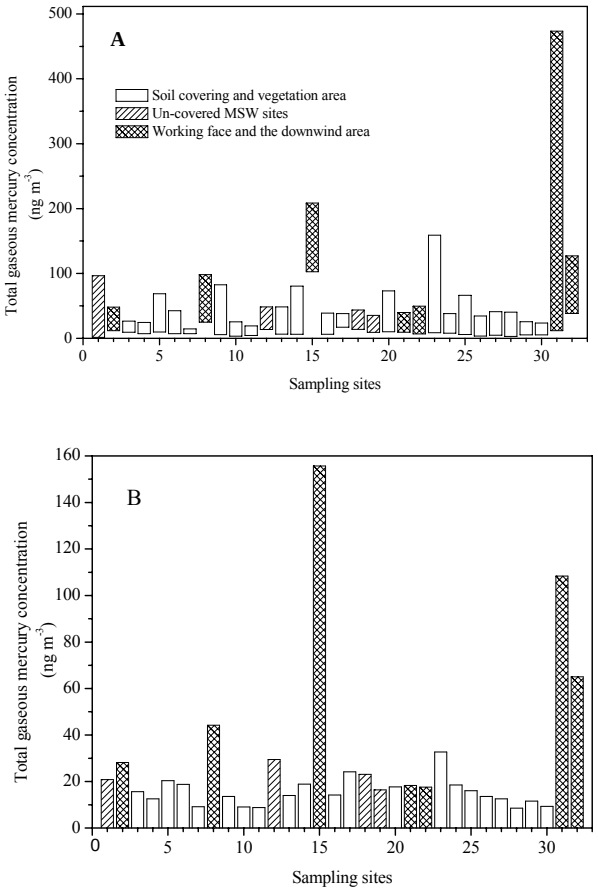

Fig. 3. The range **(A)** and average **(B)** of total gaseous mercury in the atmosphere over different sites of the studied landfills (Sample sites: 1–22 at G-Y landfill; 23–27 at D-Z-W landfill; 28 at X-R-J landfill; 29–31 at J-K landfill and 32 at D-S landfill).

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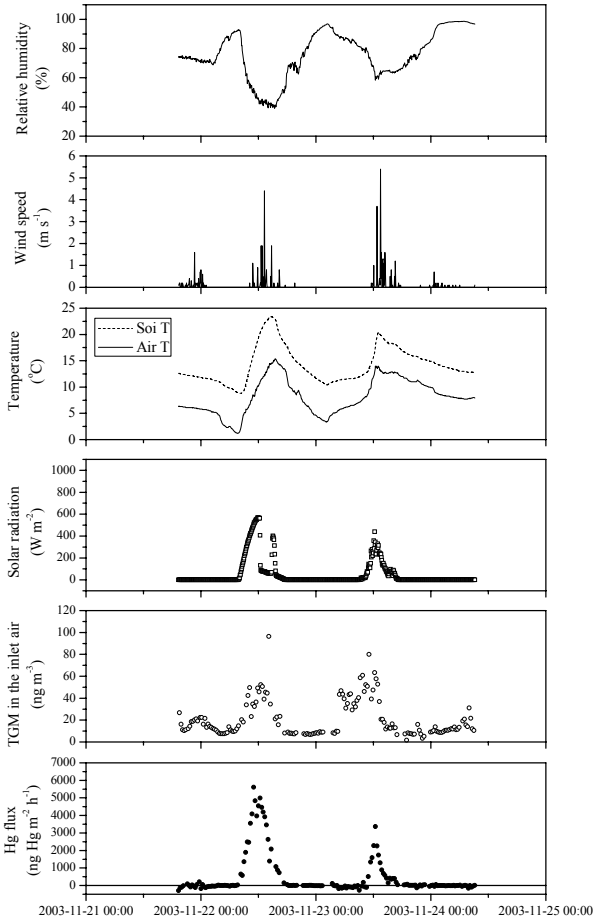

Fig. 4. Mercury surface-air fluxes at un-covered MSW site of G-Y landfill: the contribution of high mercury content in MSW to the mercury emission rate.

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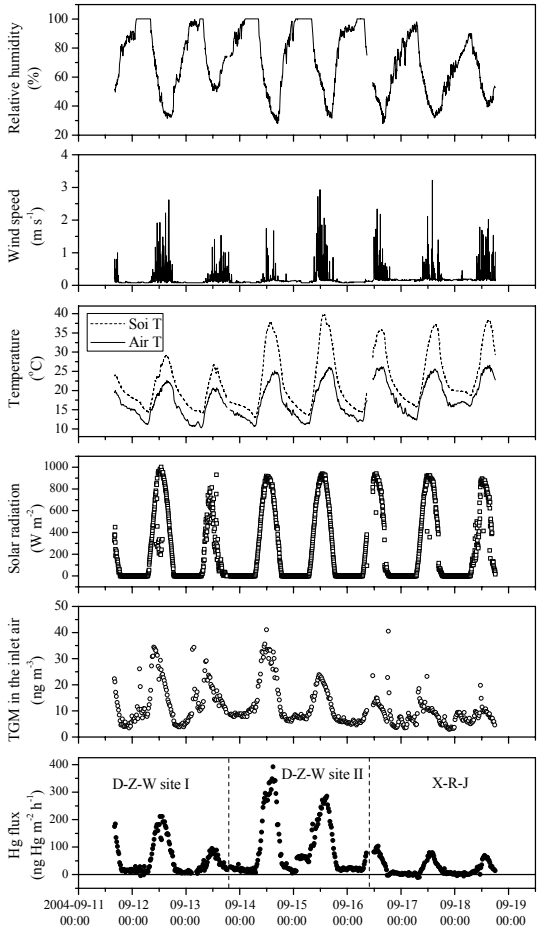

Fig. 5. Mercury surface-air fluxes at soil covering area of D-Z-W and grassed area of X-R-J landfill: the effect of vegetation on the mercury emission reduction.

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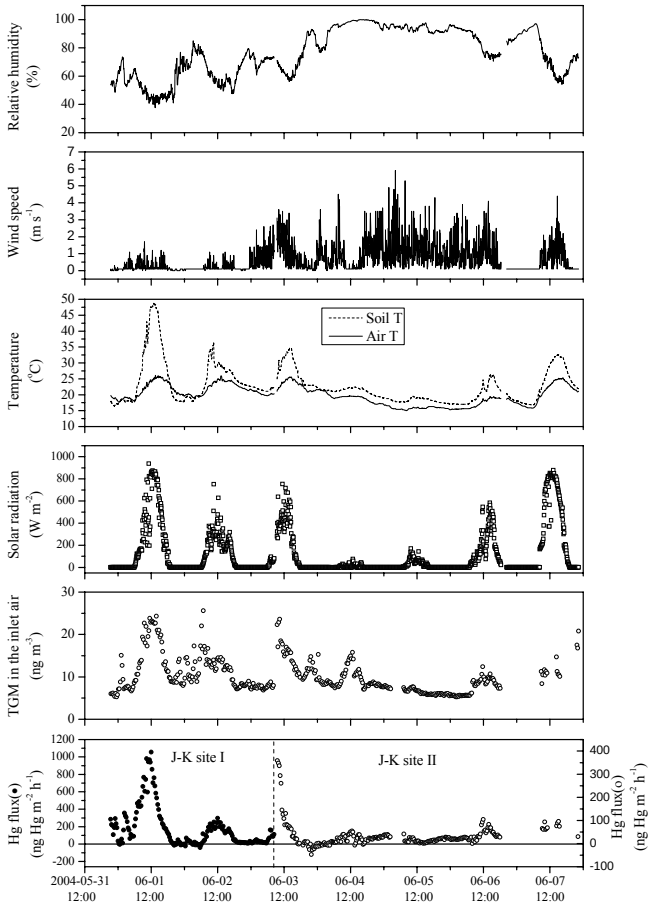

Fig. 6. Mercury surface-air fluxes at soil covering area of J-K landfill: the effect of weather conditions on the mercury emission rate.

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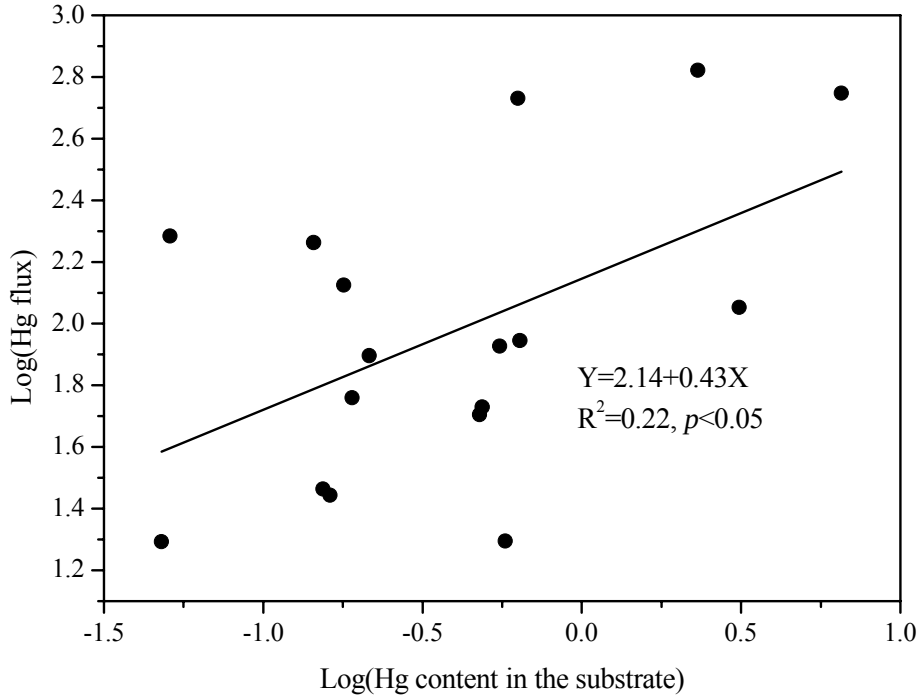

Fig. 7. Correlation between average mercury flux at different sites and mercury content in the corresponding substrates.

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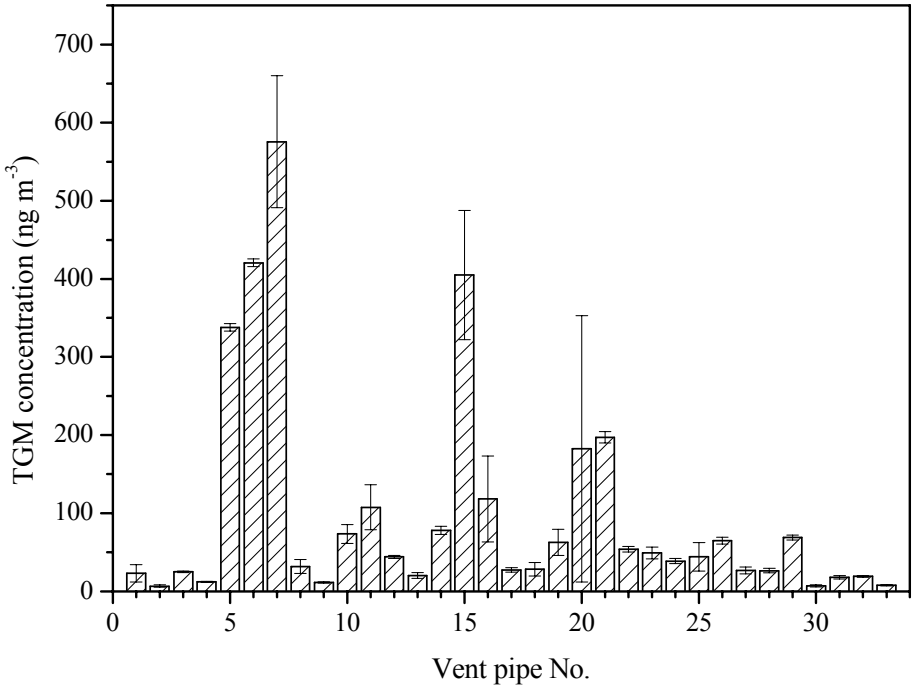

Fig. 8. Average TGM concentrations in the LFG at G-Y (vent pipe No. 1–27) and J-K (vent pipe No. 28–33) landfill.

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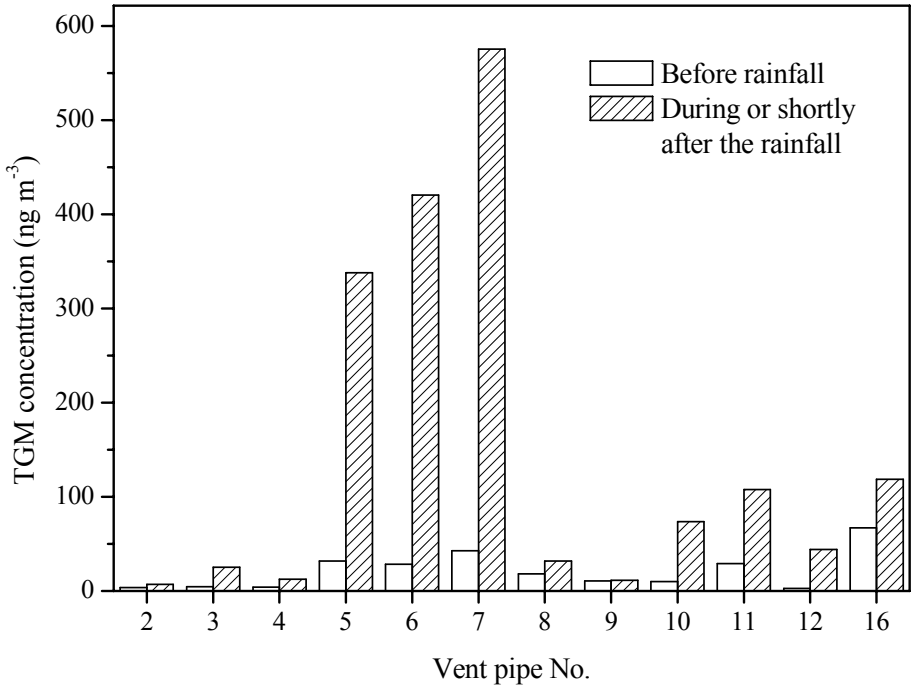

Fig. 9. Comparison of average TGM in the LFG before and after the rainfall event at G-Y landfill.

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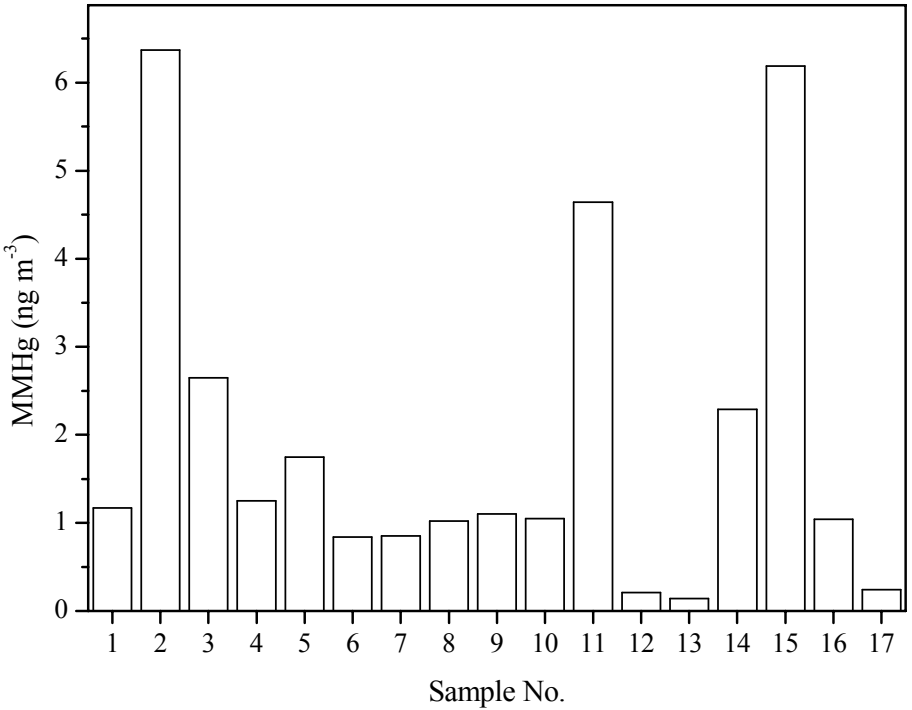

Fig. 10. MMHg in the LFG of selected vent pipes at G-Y landfill.

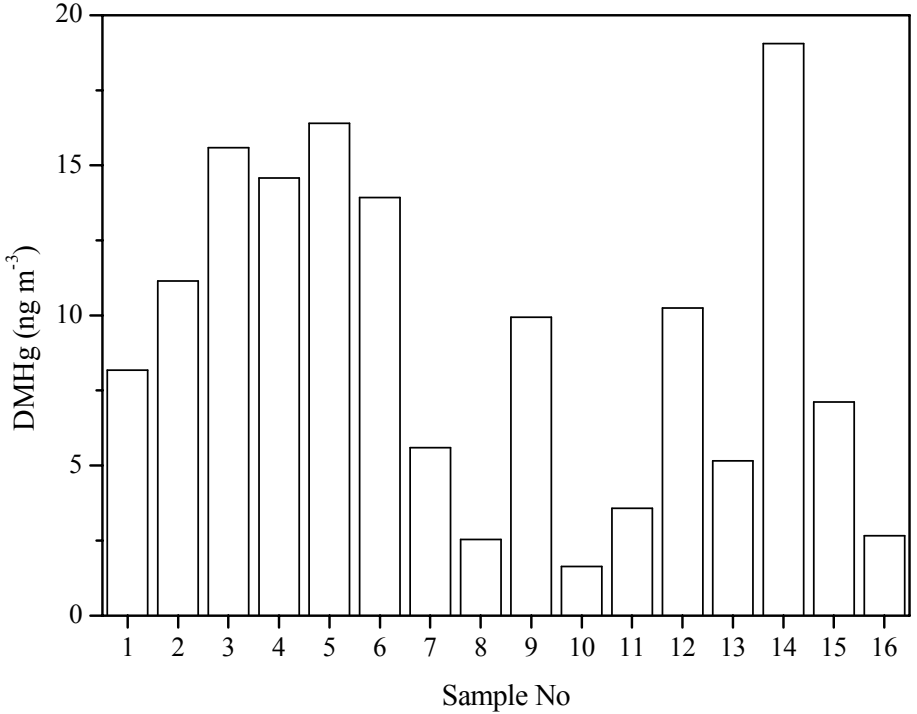

Fig. 11. DMHg in the LFG of selected vent pipes at G-Y landfill.

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