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Site-specific diel mercury emission fluxes in landfill: Combined effects of vegetation and meteorological factors

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ABSTRACT

Mercury emission fluxes (MEFs) under different surface coverage conditions in a landfill were investigated in this study. The results show similar diel patterns of Hg emission flux under different coverage conditions, with peak fluxes occurring at midday and decreasing during night. We examined the effects of environmental factors on MEFs, such as the physiological characteristics of vegetation and meteorological conditions. The results suggest that growth of vegetation in the daytime facilitates the release of Hg in the anaerobic unit, while in the semi-aerobic unit, where vegetation had been removed, the higher mercury content of the cover soil prompted the photo-reduction pathway to become the main path of mercury release and increased MEFs. MEFs are positively correlated with solar radiation and air temperature, but negatively correlated with relative humidity. The correlation coefficients for MEFs with different environmental parameters indicate that in the anaerobic unit, solar radiation was the main influence on MEFs in September, while air temperature became the main determining factor in December. These observations suggest that the effects of meteorological conditions on the mercury release mechanism varies depending on the vegetation and soil pathways.

1. Introduction

Mercury, a toxic heavy metal that can be transformed to methylmercury, is known to have serious adverse impacts on environmental and human health (Costa et al., 2016; Mergler et al., 2007; Zhu et al., 2013). It has even been detected in polar regions due to its high volatility, long atmospheric residence time and long-range transmissibility (Assad et al., 2016; Lamborg et al., 2002; Lee et al., 2015; Muir et al., 1999; Xie et al., 2008).

Landfills have been used extensively to dispose of waste in China, due to their low cost and simple technology (Cheng and Hu, 2011). However, collection of unseparated solid waste usually leads to the accumulation of mercury-containing materials in landfills, such as batteries, mercury thermometers and fluorescent lamps (Lindberg et al., 2005). These materials transform into inorganic and organic forms of mercury through chemical and biological processes during the landfill stabilization process. Numerous studies have shown that landfills act as potential atmospheric mercury sources, and can pose a severe ecological risk (Feng et al., 2004; Hang et al., 2008; Kim and Kim, 2002; Kim et al., 2001; Li et al., 2010; Lindberg and Price, 1999; Lindberg et al., 2005). Several studies have reported mercury emission fluxes (MEFs) from landfills during the past decades (Kim and Kim, 2002; Lindberg and Price, 1999; Lindberg et al., 2005). However, all of these studies focused on the mercury flux at the landfill surface, while rarely mentioned were the combined environmental effects of the physiological characteristics of vegetation and the meteorological conditions on MEFs that might provide insight into the mercury emission mechanisms in landfills.

Mercury migration in the soil-plant system is considered to occur through two pathways (Assad et al., 2016; Leonard et al., 1998). The first pathway is more static and involves timeframes of days to months, and involves plant tissues. In the second pathway, mercury is rapidly transported from the soil via the transpiration stream to mesophyll cells inside the leaf, where it volatilizes into the leaf's intercellular spaces as elemental Hg (Hg^0). The time frame for transport in this pathway is measured in minutes to a few hours. The plant-to-atmosphere exchange of mercury occurs when Hg^0 in the intercellular space of the leaf interior diffuses through the stomata into the atmosphere (Leonard et al., 1998). Mercury adsorbed on the leaf's surface can also be emitted to the atmosphere through photo-reduction (Graydon et al., 2012). Vegetation can also shade the underlying soil and weaken the photo-reduction in the soil, thus affecting the migration and release of mercury. Therefore, a variety of environmental factors (e.g., solar

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radiation, temperature and humidity) that affect physiological processes, such as transpiration and photosynthesis, have great influence on mercury emissions by affecting the mercury transport pathways (Leonard et al., 1998).

The entry of mercury into the atmosphere through soil and plant pathways involves physicochemical and biochemical processes, including mercury transformation, migration and release. Dynamic changes in these processes create uncertainty about the extent of mercury emissions. MEFs have become the focus of an increasing number of studies in recent years, but MEFs through the covered soil-vegetation-atmosphere pathway in landfills has rarely been investigated in previous studies. The migration, transformation and release of mercury from landfills are influenced by the combined environmental effects of the physiological characteristics of vegetation and meteorological factors. Thus, greater knowledge of these combined environmental effects on the exchange mechanisms is required to understand the mercury exchange mechanisms in this pathway.

Through the application of a dynamic flux chamber (DFC) system, MEFs under different landfill and coverage conditions were investigated in order to explore the interchange mechanisms of mercury between covered soil, vegetation and the atmosphere. An optimum design for the cover soil and vegetation technology was proposed to inhibit mercury transfer and emission. In addition, meteorological data were monitored simultaneously to document the influences of environmental conditions on MEFs. These results will provide technical guidance for the implementation of effective mercury pollution controls in landfills in order to protect the ecological environment.

2. Materials and methods

2.1. Site descriptions

Flux measurements were carried out in two fields at the Laogang municipal solid waste (MSW) landfill, Shanghai, China, in 2011 and 2012. Descriptions of the simulated anaerobic landfill unit and simulated semi-aerobic landfill unit used in this study have been published in detail (Chai et al., 2015). Briefly, the two landfill units were located in landfill cell #42, with a total volume of 5000 m^3 for each unit. The length and width of each unit were 19.5 m at the top and 33.5 m at the bottom. Fresh refuse was disposed of in each unit until the landfill height reached 7 m in June 2009.

The surfaces of the landfill units were covered after landfilling with cover soil on the anaerobic landfill unit, and with aged refuse on the semi-aerobic landfill unit (Han et al., 2010; Zhao et al., 2002). The semi-aerobic landfill unit was equipped with cowls above the landfill vent pipes, which connected to the leachate collecting pipes. The average Hg content in the fresh refuse that was disposed of in the two landfill units was 0.25 ± 0.092 mg kg^{-1}. In the measurement, the THg concentration of cover soil on the anaerobic landfill was 400 μg kg^{-1}, while the THg concentration of aged refuse on the semi-aerobic landfill was 2000 μg kg^{-1}. The basic characterization of the refuse have been shown in previous studies (Chai et al., 2015). The schematic diagram of landfill units is shown in Fig. 1.

2.2. Sampling and analytical methods

2.2.1. Measurements and estimation of mercury emission fluxes

MEF was measured over the MSW surfaces with different coverage conditions using a dynamic flux chamber made of Teflon and coupled with an automated Tekran 2537A mercury vapor analyzer. Inlet and outlet air of the DFC were measured sequentially at 10-min intervals (two 5-min samples), giving a 20-min temporal resolution for calculated MEFs. The MEFs were calculated according to the following equation: (Xiao et al., 1991)

$$F = \frac{(C_o - C_i) * Q}{A},$$

where F is the MEF (ng m^{-2} h^{-1}), C_o and C_i are the Hg concentrations (ng m^{-3}) of the DFC outlet and inlet airstreams, respectively, A is the surface area enclosed by the DFC (0.05 m^3), and Q is the DFC internal flushing flow rate (m^3 h^{-1}). A relatively high flow rate (30 L min^{-1}) was maintained using a diaphragm vacuum pump (i.e., DAA-V523-ED, Gast Inc., USA) connected to a gas flow meter (Fu et al., 2008) for preventing the possibility of underestimating Hg flux at low-flushing flow rates (Lindberg et al., 2002). The detection limit for the Tekran 2537A mercury analyzer was 0.1 ng m^{-3}. The DFC and all tubing and connections were acid cleaned and rinsed in Milli-Q grade water (i.e., 18.2 MΩ cm) to avoid contamination. The Tekran 2537A mercury vapor analyzer was calibrated in the laboratory before the field experiments, and periodically during the field experiments. In the field, DFC blanks (0.5 ± 0.2 ng m^{-2} h^{-1}) were consistently low, and were not subtracted in the equation above.

MEFs under different vegetation coverage conditions were measured to determine the influence of vegetation on mercury release. A typical landfill plant *Setaria viridis* (*L.*) *Beauv.* was selected for the MEF measurements. *Setaria viridis* (*L.*) *Beauv.* is annual herbaceous plants. Three types of vegetation coverage conditions were defined as follows:

Condition A: The chamber was placed over whole plants that were in normal growth conditions.
Condition B: The whole above-ground plant parts were cut off, and the cuts on stems were sealed with Vaseline to prevent gas diffusion. Then, the parts of the plants that were cut were placed into the chamber to simulate the light-obstructing effect of vegetation on the surface soil as in the condition A.
Condition C: The cut parts of the plant in condition B were removed. The schematic diagram for experiment design is shown in Fig. 2.

Therefore, the difference between MEFs under conditions A and C is the MEF between plant and atmosphere. Each coverage condition was measured over a day, and three days served as a cycle. When the measurement under one coverage condition was finished, the following coverage condition measurement was immediately conducted. The chamber base was buried approximately 1 cm into the soil, and was sealed externally with the same soil, with care taken to ensure that the air inlet was not blocked.

2.2.2. Meteorological data

Meteorological data, including solar radiation, air temperature and relative humidity, were collected on meteorological instruments equipped with a portable weather station logger (i.e., HOBO U-30, Onset Corp., USA). The meteorological data were collected every 5 min on average. The weather station was set-up close to the MEF measurement sites. Sampling was conducted in September and December in order to incorporate a moderate range of meteorological conditions. Meteorological conditions between these two sampling seasons of September and December showed that daily average and peak values of solar radiation, air temperature and relative humidity were higher in September.

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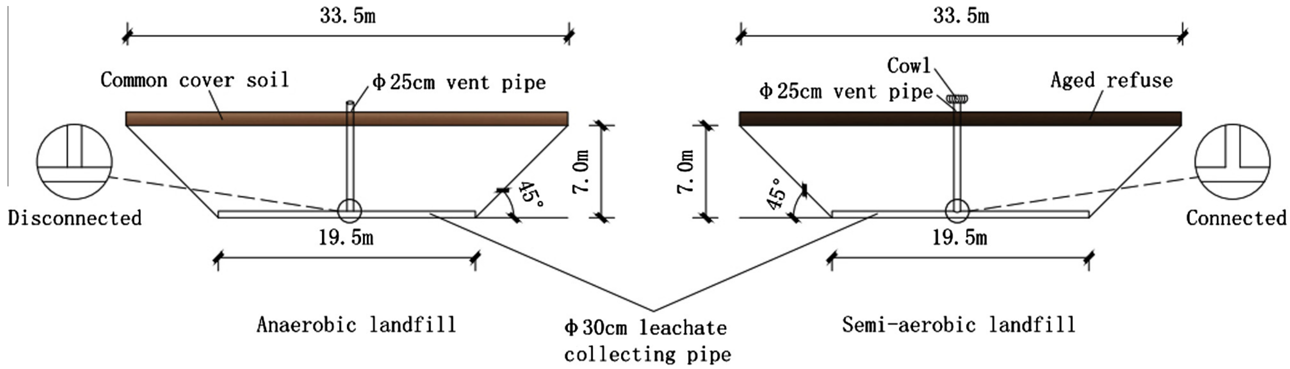

Fig. 1. A schematic diagram of the landfill sites.

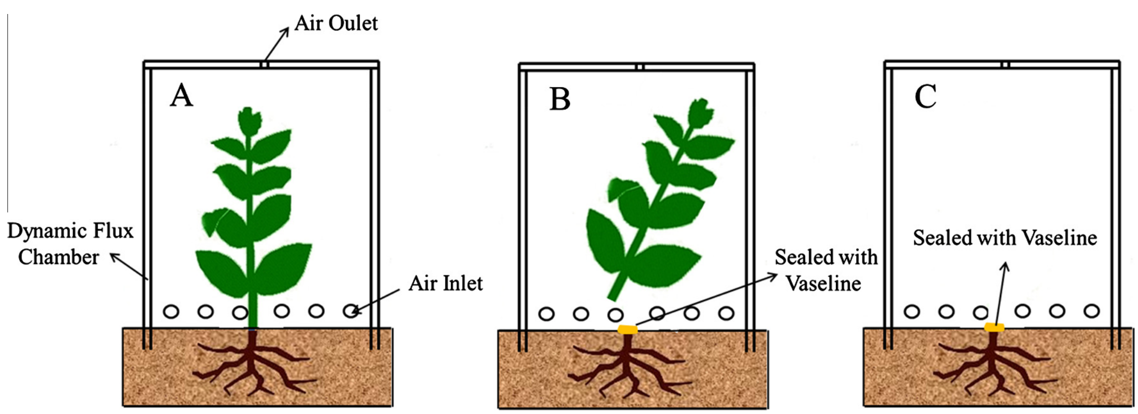

Fig. 2. A schematic diagram for experiment design.

3. Results and discussion

3.1. Characteristics of Hg emission fluxes

3.1.1. Anaerobic unit

The MEFs in the anaerobic unit under different surface coverage and meteorological conditions are shown in Fig. 3. The landfill showed high Hg emission fluxes (57.80 ng m^2 h^{-1}, Fig. 3, II). The diel pattern in MEFs shows a similar trend under different coverage conditions (i.e., A, B and C in Fig. 3), with peak fluxes occurring at midday and decreasing during the night (Li et al., 2010; Zhu et al., 2013). The measured MEFs were in the same range as those reported for landfills in Florida, which range from 1 to 20 ng m^{-2} h^{-1} (Lindberg and Price, 1999; Lindberg et al., 2005), but much lower than those reported for landfills in Seoul (Kim and Kim, 2002). The Hg emission fluxes in landfill were significantly above the soil background value in United States (Ericksen et al., 2006), which indicated that landfills are an important source of gaseous mercury to the atmospheric environment.

Refuse components, landfill stabilization processes, coverage conditions, climate and environmental conditions exert significant influence on MEFs (Li et al., 2010). In the anaerobic unit, the peak value of MEFs occurred near 12:00 a.m., except for under condition C in September (i.e., 2:25 p.m.). The highest MEFs during the daytime were found under condition A, and this was especially evident in September, which indicated that the growth of vegetation facilitated the release of Hg in the daytime. The MEFs varied with different landfill conditions, because the mercury in intercellular spaces of plant leaves can be released to the atmosphere by biochemical processes (Carrasco-Gil et al., 2013). Alternatively, the increased air temperature in September could raise the mercury vapor pressure within the leaf and accelerate the volatilization of mercury. Similarly, the more intense solar radiation also increases leaf temperature and facilitates mercury release (Leonard et al., 1998). Furthermore, increased photosynthesis and transpiration of vegetation in September accelerates the transport and release of Hg (Hussein et al., 2007; Moreno et al., 2008; Stamenkovic and Gustin, 2009).

The mean, standard deviation and range (i.e., minimum and maximum) of the measured MEFs are shown in Table 1. The seasonal difference in MEFs varies for different coverage conditions. Differences in MEFs were observed in different seasons under conditions B and C, with MEFs for condition C in December being higher than for condition B, with the pattern reversed in September. The seasonal variation of MEFs under different conditions may be attributed to different mechanisms. In December, the physiological activities of vegetation are much lower, and surface covered soil becomes the only pathway for Hg release after the vegetation was cut and removed. Due to the coverage of vegetation in condition B, with lower air temperature and less solar radiation being absorbed by soil-covered surface inside the chamber, the MEFs decreased. However, in September, the daily mean solar radiation for conditions B and C were 105.70 W m^{-2} and 34.26 W m^{-2}, respectively. The lower solar radiation resulted in lower MEFs for condition C, which indicated that solar radiation is the more dominant influence of MEFs in this situation.

It has been reported that solar radiation can significantly enhance Hg emissions from soil (Gustin et al., 2002). Because Hg^{2+} in soil can be reduced to Hg^0 by photo-reduction, stronger solar radiation leads to higher Hg^0 concentrations in soil air, which in turn increases the mercury emission rate from soil to the

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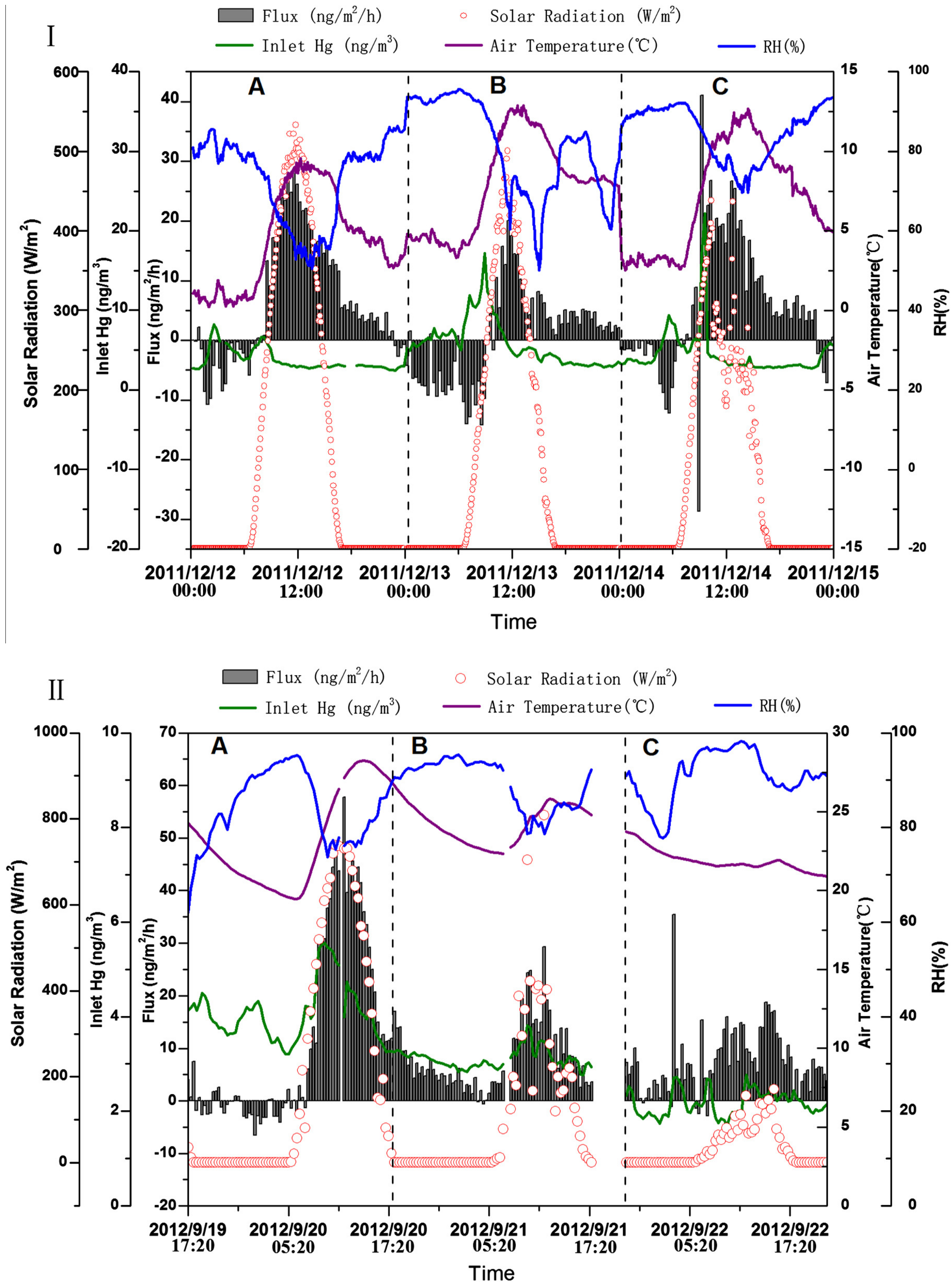

Fig. 3. Meteorological conditions and MEFs in the anaerobic unit: (I) December, (II) September (The dashed lines used to distinguish condition A, B and C).

atmosphere. The increased activity of Hg^{2+} with rising soil temperature accelerates the speed of this photo-reduction of Hg^{2+} to Hg^0. Meanwhile, the air driven from soil pores by elevated soil temperature may increase the Hg^0 diffusion into the atmosphere. The MEFs for conditions A and B measured in September were significantly higher than those measured in December, which are probably due to the effects of solar radiation and temperature on MEFs. The rapid degradation of refuse and strong microbial activity induced by relatively higher temperatures result in a massive release of Hg. In addition, intense solar radiation and higher temperatures accelerate the photo-reduction of Hg^{2+} and volatilization of Hg^0 (Gustin et al., 2002; Leonard et al., 1998; Li et al., 2010).

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Table 1
The mercury emission fluxes under different coverage conditions in the anaerobic unit (ng m^2 h^{-1}).

	Daytime		Night	
	Mean ± std	Range	Mean ± std	Range
December				
Condition A	14.45 ± 9.28	−5.85 ~ 28.76	−0.20 ± 4.48	−10.77 ~ 6.54
Condition B	3.03 ± 9.64	−14.19 ~ 20.06	−1.00 ± 5.32	−9.40 ~ 5.15
Condition C	12.48 ± 12.11	−28.70 ~ 41.01	−0.36 ± 5.00	−12.21 ~ 7.44
September				
Condition A	24.12 ± 17.45	−2.13 ~ 57.80	−1.13 ± 2.53	−6.57 ~ 7.55
Condition B	11.07 ± 6.72	2.54 ~ 29.31	5.28 ± 3.88	−0.65 ~ 17.03
Condition C	9.74 ± 5.57	−2.85 ~ 18.74	5.25 ± 5.84	−1.09 ~ 35.40

(Daytime is defined as the time immediately after sunrise and before sunset)

In the anaerobic unit, MEFs during the daytime were higher than those at night, whilst atmospheric Hg deposition events occurred. The phenomenon of higher MEFs in daytime resulting from the effects of solar radiation and temperature are discussed above. Moreover, the enhanced photosynthesis and transpiration of vegetation stimulated by intense solar radiation may influence the stomatal conductance of vegetation, which would facilitate the release of Hg (Lindberg et al., 1979; Stamenkovic and Gustin, 2009). The atmospheric Hg deposition event that occurred under condition A during the nights in September is mainly attributed to the presence of vegetation.

3.1.2. Semi-aerobic unit

The MEFs in the semi-aerobic unit under different surface coverage and meteorological conditions are shown in Fig. 4 and Table 2. The diel pattern in Hg emission flux shows a similar trend under different coverage conditions (i.e., A, B and C in Fig. 4), with peak fluxes occurring at midday and decreasing during the night. The MEFs exhibit higher concentrations during the daytime, and in September.

The MEFs from the semi-aerobic unit exhibited opposite trends under different conditions, with the highest value under condition C, and the lowest value under condition A occurring in daytime, and opposite results at night with the highest value under the condition A and the lowest value under the condition C. The high MEFs observed under the condition C in daytime indicate that the surface cover soil is the main pathway for Hg emissions. The covered mineralized refuse, which has a THg concentration of approximately 2000 μg kg^{-1}, is the principal source of MEFs due to diffusion of Hg stimulated by photo-reduction in the soil. The vegetation shading that obstructed solar radiation from the cover soil decreased the Hg emissions under condition A. The MEFs under condition B were slightly higher than those under condition A, which may have resulted from the weaker shading from the cut vegetation with its lower physiological activity. However, the lowest MEFs observed under condition C were at night, which confirms that solar radiation is the principal factor influencing the MEFs.

The MEFs at night under condition A were at least twice as high as those under condition C, indicating that mercury could be released to the atmosphere through non-stomatal pathways at night. Non-stomatal processes might be important for cycling Hg between foliar surfaces and the atmosphere (Stamenkovic and Gustin, 2009).

3.2. Factors affecting mercury emission flux

3.2.1. Anaerobic unit

The measurement of MEFs under different coverage and meteorological conditions allows the relationships between MEFs and meteorological conditions to be investigated. Table 3 gives the correlation coefficients for MEF against meteorological parameters under specific conditions in the anaerobic unit, and these shows strong correlation between MEF and three meteorological parameters under different coverage conditions. Positive correlations were obtained between MEF and solar radiation and between MEF and air temperature, whereas a negative correlation was obtained between MEF and relative humidity.

Compared with the correlations under different coverage conditions, the highest positive correlation coefficients between MEFs and solar radiation and between MEFs and air temperature were found under condition A, which indicates that solar radiation and air temperature are important factors affecting MEFs under a vegetation-covered condition. The temperature dependence of mercury flux is attributed to changes in the contaminant's vapor pressure in the leaf interior. Increased temperature accelerates the biotic or abiotic transformation of Hg^0 and enhances the mercury vapor pressure to facilitate the volatilization of mercury. Furthermore, intense solar radiation accelerates the photo-reduction of Hg^{2+} and the volatilization of Hg^0 (Leonard et al., 1998; Rutter et al., 2011; Lindberg et al., 1979; Stamenkovic and Gustin, 2009). In addition, the thermal energy absorbed by plants from solar radiation can increase the mercury vapor pressure and transport mercury from plants into the atmosphere by convection (Gustin et al., 2002).

Higher correlation coefficients were obtained between MEF and solar radiation and between MEF and air temperature under condition B than under condition C, except for MEF and solar radiation in December. This indicates that meteorological conditions had a weaker effect on MEFs under the soil-covered condition for which the THg concentration was 400 μg kg^{-1}. The lower correlation coefficient between MEF and solar radiation under condition B in December is due to weaker physiological activity of plants and decreased solar radiation obstructed by covered cut vegetation, which reduces the MEFs from covered soil.

Notably, MEF and relative humidity are negatively correlated. Enhanced mercury atmospheric deposition results from increased relative humidity, which can combine vapor and mercury (Briggs and Gustin, 2013; Lodenius, 2013). Meanwhile, vapor can occupy air gaps in the soil and block the release of landfill gas. The lowest correlation coefficient was between relative humidity and MEF under condition C in the anaerobic unit.

The correlation coefficient between solar radiation and MEF was higher than that between air temperature and MEF in September, while the pattern was the opposite in December. In September, photo-reduction driven by solar radiation, rather than temperature, was the main factor affecting MEFs. The increase in solar radiation can not only accelerate the photo-reduction, but also increase air temperature, thereby promoting MEFs (Gustin et al., 2002). The above observations suggest that solar radiation was the restrictive factor on mercury release in September. However, in December, lower temperatures caused weaker physiological activity of plants and microorganisms and decreased the leaf area index of plants,

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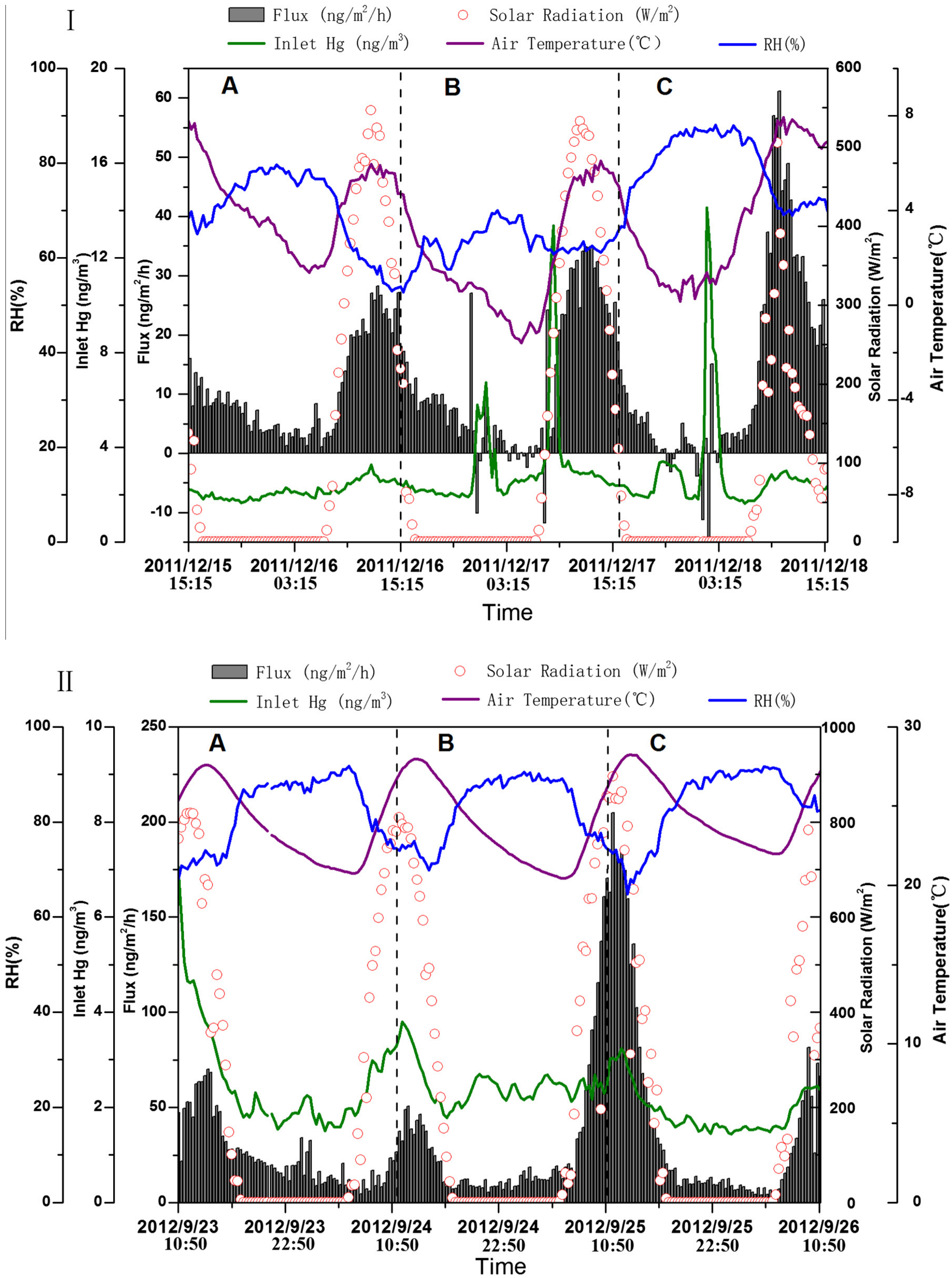

Fig. 4. Meteorological conditions and MEFs in the semi-aerobic unit: (I) December, (II) September (The dashed lines used to distinguish condition A, B and C).

which weakened the MEFs (Li et al., 2010). The highest correlation coefficient between MEF and air temperature was found in December. These results indicate that in anaerobic landfills, solar radiation is the main factor influencing mercury release in September, whilst air temperature is probably the more important driver in December.

3.2.2. Semi-aerobic unit

A similar pattern was observed in the semi-aerobic unit, with positive correlations between MEF and solar radiation and between MEF and air temperature, while a negative correlation was observed between MEF and relative humidity (Table 4). However, there were differences in the values of the correlation

Table 2
The mercury emission fluxes under different coverage conditions in the semi-aerobic unit (ng m^2 h^{-1}).

	Daytime		Night	
	Mean ± std	Range	Mean ± std	Range
December				
Condition A	16.67 ± 8.01	2.61 ~ 28.19	5.59 ± 2.78	1.10 ~ 10.89
Condition B	20.23 ± 12.23	−11.79 ~ 35.08	4.38 ± 5.80	−10.09 ~ 27.06
Condition C	27.57 ± 15.61	3.05 ~ 61.15	1.80 ± 4.98	−16.03 ~ 15.10
September				
Condition A	31.50 ± 21.10	4.50 ~ 70.18	18.15 ± 6.43	9.02 ~ 33.91
Condition B	47.23 ± 41.53	7.19 ~ 170.49	11.17 ± 3.42	5.47 ~ 19.19
Condition C	74.49 ± 60.01	6.65 ~ 204.92	9.51 ± 3.51	4.01 ~ 18.11

Table 3
Correlation of meteorological conditions and MEFs in the anaerobic unit.

	Coverage conditions	Solar radiation	Air temperature	Relative humidity
December	A condition	0.891**	0.920**	−0.883**
	B condition	0.581**	0.853**	−0.704**
	C condition	0.695**	0.717**	−0.704**
September	A condition	0.960**	0.784**	−0.504**
	B condition	0.812**	0.513**	−0.786**
	C condition	0.572**	−0.148	0.270*

* $0.01 < p < 0.05$.
** $p < 0.01$.

Table 4
Correlation of meteorological conditions and MEFs in the semi-aerobic unit.

	Coverage conditions	Solar radiation	Air temperature	Relative humidity
December	A condition	0.905**	0.671**	−0.949**
	B condition	0.851**	0.878**	−0.357**
	C condition	0.925**	0.872**	−0.737**
September	A condition	0.561**	0.802**	−0.725**
	B condition	0.688**	0.327**	−0.522**
	C condition	0.897**	0.746**	−0.814**

*$0.01 < p < 0.05$.
** $p < 0.01$.

coefficients for the two landfill units, especially under condition C, which suggests that meteorological conditions play different roles in the mechanism of MEFs in the two types of landfill technology.

The highest correlation coefficient between MEF and solar radiation was found under condition C, indicating that solar radiation was the most important factor affecting mercury release from the covered soil. Due to the higher mercury content of the covered soil in the semi-aerobic unit, mercury volatilization caused by photo-reduction is the main path of mercury release. In surface soils, increasing temperature enhances not only the rate of biotic or abiotic processes leading to Hg^0 production, but also the rate of contaminant volatilization through a direct effect on the vapor pressure (Leonard et al., 1998). In September, extensive vegetation shaded the covered soil and weakened the effect of photo-reduction, which resulted in the lower correlation under conditions A and B. The effect of vegetation shading decreased the emission rates, and this effect was most pronounced at the highest temperature studied (Lindberg et al., 1979). The correlation coefficient between MEF and solar radiation under different coverage conditions varied within a smaller range in December than in September. In December, the plants withered leading to the inevitable decrease of leaf area index, which diminished the effect of shading on soil, and reduced the differences between correlation coefficients for different coverage conditions.

4. Conclusions

MEFs changed greatly with diel and seasonal pattern. MEFs were higher in daytime and in September. Difference of MEFs varies between different landfill unit and different coverage conditions. MEFs in the landfill were closely related to the mercury in the cover soil, vegetation conditions and meteorological conditions. In anaerobic unit, the growth of vegetation facilitated the release of mercury in the daytime, while mercury deposition phenomenon was occurred at night. In semi-aerobic unit, higher mercury content of covered soil increased MEFs in the vegetation removed condition and prompted photo-reduction pathway to become the main path of mercury release. The existence of vegetation weakened the effect of photo-reduction and decreased the mercury release. And also it could facilitate mercury release through non-stomatal pathway at night.

Meteorological conditions like solar radiation, air temperature and relative humidity influenced the mercury emission fluxes and the correlation coefficients were different under different coverage conditions. In anaerobic unit, solar radiation was the main influence on MEFs in September, while air temperature became the controlling factor in December. All of these suggested that the effects of meteorological conditions on mercury release process mechanism varied under vegetation pathway and soil pathway.

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