Waste Management xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

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

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ARTICLE INFO

Article history:
Received 6 June 2016
Revised 6 September 2016
Accepted 7 September 2016
Available online xxxx

Keywords:
Mercury emission fluxes (MEFs)
Landfill
Vegetation
Meteorology parameters
Solar radiation

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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http://dx.doi.org/10.1016/j.wasman.2016.09.006
0956-053X/© 2016 Published by Elsevier Ltd.

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.