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Fate of Mercury in a Modern Municipal Solid Waste Landfill in China

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Abstract. Fate of mercury (Hg) in a modern sanitary municipal solid waste (MSW) landfill in Guiyang, China was investigated to assess the potential lost of Hg to ambient environment through different pathways. Mercury content in MSW, leachate, and mercury airborne emission rate through the surface and the vent pipe system were determined. A mass balance calculation showed that, roughly 0.67% of Hg entered into the landfill each year was lost into the surrounding environment, which was dominated by the pathway of working face (98.47%), then soil covers (1.28%), leachate (0.22%) and landfill gas vent pipes (0.03%).

Introduction

Mercury (Hg) enters into the municipal solid waste (MSW) mainly from a variety of Hg-containing products, such as batteries, fluorescent lumps, thermometers, switches, etc. The treating of MSW by incinerators has resulted in a great deal of Hg lost into the atmosphere [1]. While, due to the low cost and low maintenance, the majority of MSW (around 70%) in the world is treated by landfills [2]. As a consequence, quantities of mercury are ended up in landfills. There are 88 and 390 tonnes of Hg been buried in the landfills of 15 European Union countries and USA in 1995 and 2000, respectively [3-4]. In China, there are hundreds of tones Hg discarded into the MSW associated with the Hg-containing products, such as batteries [5] and fluorescent lamps and thermometers [6].

However, the behavior of Hg in the landfill site has rarely systematically studied as that of MSW incinerators. Once Hg enters into the landfill, it could be re-mobilized into the surrounding environment. To provide a detailed insight into Hg cycling in the Chinese landfills, we carried out a series of field campaigns at a modern sanitary MSW landfill (Gaoyan landfill) in Guiyang city, the capital of Guizhou province, SW China, to investigate the Hg fate at the landfill site.

Experimental Methods

The studied landfill, Gaoyan MSW sanitary landfill, started operation since 2001 and is the biggest and highest standard landfill in Guiyang. Gaoyan landfill, treats MSW on a rate of 1300 tonnes per day, is a reprehensive of modern landfills in China, i.e., it covers the MSW by daily soil coverings, collects and treats the leachate by the waste water treatment plant, and discharges the landfill gas (LFG) into the atmosphere directly by vent pipe system.

Hg distribution in MSW, covering soil, leachate and LFG in Gaoyan landfill was investigated, the Hg surface-air exchange was also determined, and finally, a picture of the fate of Hg in this landfill was obtained by a means of mass balance methodology.

Hg in MSW and Covering Soil

Hg content in MSW, as shown in Fig. 1A, ranged from 0.170 to 46.222 mg kg^{-1} (N=40), with an arithmetic mean and geometric mean of 1.870 and 0.603 mg kg^{-1}, respectively. The distribution was highly skewed, with over half the samples having concentrations less than 0.5 mg kg^{-1}, only three had concentrations exceeding 2.0 mg kg^{-1}, these samples maybe contaminated by Hg-containing products. The Hg distribution pattern in MSW at Gaoyan was similar to that of a MSW landfill in Florida, USA, where the range and the geometric mean are 0.033-16.800 mg kg^{-1} (N=106) and 0.178 mg kg^{-1}, respectively [7].

While, Hg levels in covering soil, as shown in Fig. 1B (range: 0.130-0.215 mg kg^{-1}; arithmetic mean: 0.175 mg kg^{-1}; geometric mean: 0.173 mg kg^{-1}, N=16), was more convergent, which reflect a background value for Guiyang.

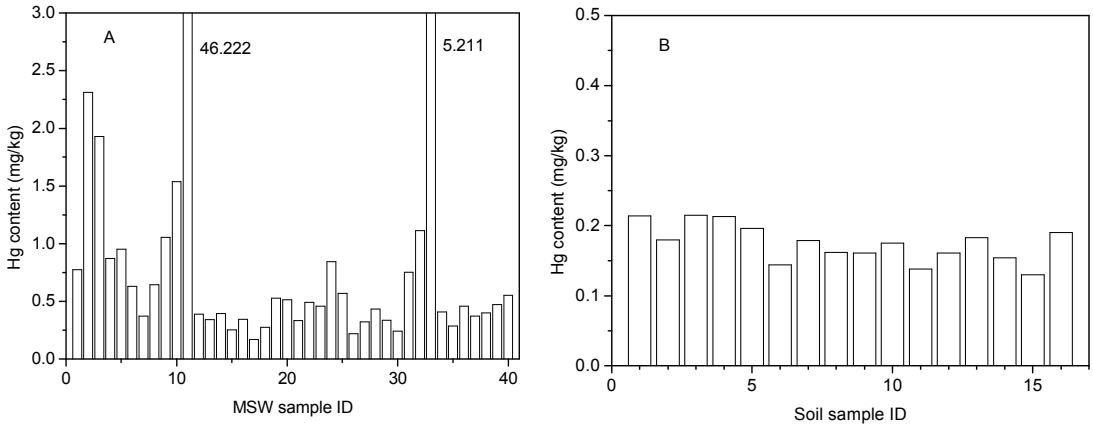

Fig. 1 Hg content in MSW (A) and covering soil (B) at Gaoyan landfill

Hg Speciation in LFG

Three Hg species in LFG emitted from the vent pipe system were determined, i.e., total gaseous mercury (TGM), monomethylmercury (MMHg) and dimethylmercury (DMHg). The range of TGM was 2.0-1406.0 ng m^{-3} and the mean was 89.8 ng m^{-3}. Large variations were observed among different pipes, which reflect the difference in Hg content in MSW. MMHg in LFG of some vent pipes varied between 0.14 and 6.37 ng m^{-3}, with an average of 1.93 ng m^{-3}. The percentage of MMHg to TGM ranged from 0.14 to 1.68%, with an average of 0.51%. 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}. DMHg comprised 0.27 to 3.64% of TGM in the LFG, with an average of 1.79%.

The global background concentrations of MMHg and DMHg in the atmosphere are generally below 10 pg m^{-3} [8], the high concentrations of MMHg DMHg found in LFG hints that landfill is an important methylated Hg source.

Hg in Leachate

The distribution of Hg in the raw leachate and the treated samples at different stages of the on-site treatment plant, are shown in Fig. 2. Total Hg in the raw leachate was 79.4 ng l^{-1}, which was at the lower end of the worldwide landfills (50-160000 ng l^{-1}) [9]. Total Hg and particulate Hg declined significant along the treatment process, while the dissolved Hg remained relative constantly (Fig. 2).

Hg Surface-air Flux

Hg surface-air flux at the non-working area was measured by a dynamic flux chamber (DFC) method, while for the working face area, where the MSW was dumped, spreaded, crushed and covered with the soil covering, a ISCST3 model based on the Gaussion plume model was applied [10]. The results turned out (as see in Table 1) that the Hg surface-air flux was lowest at the soil covering area (generally less than 200 ng m^{-2} h^{-1}), highest at the working face area (nearly 60000 ng m^{-2} h^{-1}), showing the effective of soil covering in the reduction of mercury lost from the surface. The data also revealed that the weather conditions intensively affected Hg emissions from the landfill surface, leading to more Hg lost from the surface at warm and sunny conditions.

Mass Balance of Hg in Gaoyan Landfill

Based on the aforementioned studies, a rough picture can be achieved for Hg cycling in the landfill and its environmental lost (Table 2). There are around 172 kg Hg enters into Gaoyan landfill each year, while 3.36 kg Hg (1.96% of the total) was lost into the surrounding environment at the same time, of which, 97.83% was emitted into the atmosphere, and 2.17% was leached into the surface water. The working face area was the largest pathway for Hg lost from the landfill, accounting for 96.22% of the total loss.

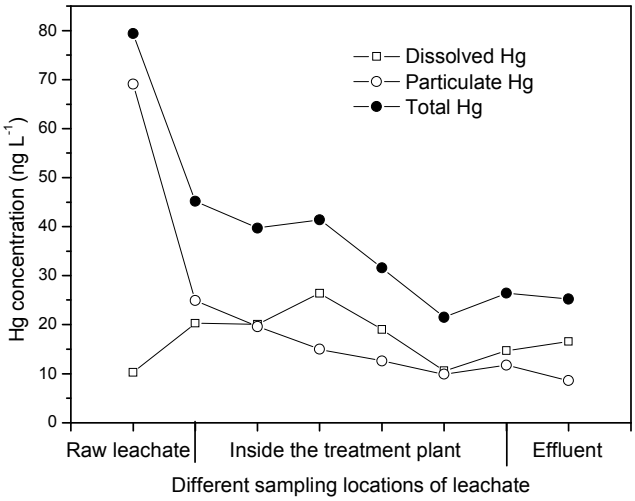

Fig. 2 Mercury concentrations in leachate along different process in the on-site waste water treatment plant

Table 1. Statistical summary of Hg surface-air emissions at different landfill surface sites

Site No.	Site description	Season	Weather condition	Mean±Std (ng m^{-2} h^{-1})	Method
F1	Soil covering area	Warm season	Rainy	78.8±77.9	DFC
F2	Soil covering area	Warm season	Sunny	183.3±191.3	DFC
F3	Soil covering area	Warm season	Sunny	133.3±65.8	DFC
F4	Soil covering area	Warm season	Cloudy	27.8±16.5	DFC
F5	Soil covering area	Cold season	Sunny	29.1±17.5	DFC
F6	Soil covering area	Cold season	Sunny	-1.4±26.2	DFC
F7	Sporadic un-covered MSW	Cold season	Sunny	57.5±83.4	DFC
F8	Sporadic un-covered MSW	Cold season	Sunny	84.5±88.5	DFC
F9	Sporadic un-covered MSW	Warm season	Sunny	664.6±1341.2	DFC
F10	Sporadic un-covered MSW	Warm season	Sunny	537.7±485.1	DFC
F11	Working face area	Warm season	Sunny	57651	ISCST3 Model

Table 2. Annual environmental loss of Hg from different pathways in Gaoyan landfill

Emission pathways	Receptor	Hg emission quantities (g yr^{-1})	Percentage of each pathway (%)
Soil covers	Atmosphere	42.03	1.25
Sporadic un-covered MSW	Atmosphere	10.95	0.33
Working face	Atmosphere	3231.35	96.22
Vent pipes	Atmosphere	0.93	0.03
Leachate	Surface water	73.00	2.17
Total		3358.25	100.00

Conclusions

This study for the first time revealed Hg fate in a real, large and modern landfill in China. Most Hg lose from the landfill was evaporated into atmosphere through the surface, only a minor was discharged into the leachate. The emission patterns of Hg was totally different from other heavy metals (such as Cd, Ni, Pb Cu, Zn) [11], which lost mainly through the leachate, and the lost percentage (0.16%-0.99% for Cd, Ni, Pb Cu, Zn) [11] was obviously lower than that of Hg (1.96%). Due to the high concentrations of methylated mercury in the LFG and their potent toxicology to the creatures, it is highly recommended that the LFG being properly treated before being discharged into the atmosphere. Although, the percentage of Hg lost into the environment each year was relatively small than that of incinerators, however, for a long run, landfill will impact the ecosystem for a longer time. So, to reduce the environmental risk of Hg in landfill, it's better to ban the Hg-containing waste throwing into the MSW, and a recycling and management system for the waste Hg-containing products must be set up in China.

Acknowledgements

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