A Preliminary study on mercury speciation in municipal waste landfill gas from Guizhou, China

SHUNLIN TANG[1, 2], XINBIN FENG[1,*], ZHONGGEN LI[1, 2], SHAOFENG WANG[1, 2], LIAN LIANG[3]

[1]State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, PR China.
[2]The graduate school of Chinese Academy of Sciences, Beijing 100039, PR China.
[3]Cebam Analytical Laboratories, 3927 Aurora Ave, Seattle, WA 98103, USA.
*Corresponding author; E-mail: xinbin.Feng@mail.gyig.ac.cn

Abstract: The concentrations of gaseous mercury species emitted from the vents of a municipal waste sanitary landfill in Guizhou province, China, were measured. The average concentration of TGM in 6-month vent gas, 12-month vent gas and 24-month vent gas, was 665.52 ± 291.25 ng/m^3 (n=305), 25.6 ± 3.2 ng /m^3 (n=13), 14.5 ± 1.8 ng/m^3 (n=28), respectively. The average concentration of MMHg in 6-month vent gas and 24-month vent gas was 2.06 ± 1.82 ng/m^3 (n=11), 0.18 ± 0.06 ng/m^3 (n=2), respectively. The average concentration of DMHg in vent gas was 9.45 ± 5.18 ng/m^3 (n=12). The preliminary results illuminated that the landfill was, not only a significant TGM source to the atmosphere, but also toxic MMHg and DMHg source to the atmosphere.

Key words: Landfill gas, TGM, MMHg, DMHg

INTRODUCTION

Many types of anthropogenic mercury emission sources including incinerators, coal-fired power plants, sewage sludge amended fields, mine waste, have been widely investigated. However, large uncertainties still remained with respect to the potential of certain sources like landfill, especially with mercury species released from landfill gas. More than 50 % of the total Hg content in garbage is attributable to used batteries, while other industrial products such as fungicides, paints, thermometers, electronic components and fluorescent tubes, are also responsible for some Hg pollution (REIMANN, 1986). The mercury compounds could be transformed into more toxic monomethyl (MMHg) or dimethyl Hg (DMHg) under anaerobic conditions (COMPEAU AND BARTHA, 1985; LINDBERG, 2001), which are common in landfills.

LINDBERG ET AL. (1999; 2001) conducted measurements of Hg emissions and methylated mercury species from several municipal landfill sites in South Florida, USA. KIM (2001) using micrometeorological techniques investigated the vertical exchange of Hg across the boundary of air and surface soils and the concentration from vents at the Nan-Ji-Do (NJD) landfill. In this work, we carried out a measurement program to measure the concentrations of gaseous mercury species emitted from the vents of Gaoyan municipal waste sanitary landfill in Guiyang, the capital of Guizhou province, southwest China, from November 20 to 29, 2003.

RESULTS AND DISCUSSION

In contrast to industrialized countries, the landfill is a primary waste treatment way in China, which account for 70 % of all waste treatment. Gaoyan municipal waste sanitary landfill located in 14km northeast of the center of Guiyang. It received an average of 800 tones per day of municipal waste and its operation began in Dec. 2001. The municipal wastes firstly were sanitized with HClO solution every day and then were covered up with clean soils when the wastes were accumulated up to 4 – 5 m high in depth. Finally a vent pipe was installed (15 cm i.d. and height of about 2m above soil surface) across every 40 to 60m. Almost all the vent pipes are found to release gases with exceptionally high CH_4 levels. The landfill is divided in three sectors that can be named as 6-month waste disposed area, 12-month waste disposed area and 24-month waste disposed area in terms of the disposed time.

Total gaseous mercury (TGM) was collected and analyzed using Mercury Vapor Analyzer Tekran® 2537A. The Tekran® has two channels which trap vapor-phase mercury onto gold adsorbents. The two adsorbent cartridges thermally desorbs alternately, after which Hg is detected using cold vapor atomic fluorescence spectroscopy, enabling a time resolution down to 5 min. MMHg was collected at flow rates of 400 ml/min in a series of 2-bubbler which contained 45 ml of 0.5 % HCl solution and analyzed by thermal desorption–gas chromatography–atomic fluorescence spectrometry (TD–GC–AFS) after isolating MMHg from the matrix by distillation and conversion to methylethylmercury by aqueous phase ethylation (Bloom and Von der Geest, 1995).

DMHg was collected using Carbotrap adsorbers™ (20/40 mesh) and analyzed by TD–GC–AFS (LIANG L ET AL., 1994). The method used to determine DMM is very sensitive and highly selective (Lindberg, 2001), which found to have high adsorption capacity for DMM, while allowing the bulk of mercury, present as Hg^0, to pass through. In order to stabilize the DMM on the Carbotrap™ sorbent, the traps were immediately purged after sampling with dry nitrogen to remove residual water.

Fig. 1 depicted the results of TGM concentration for a 6-month vent gas, which indicated

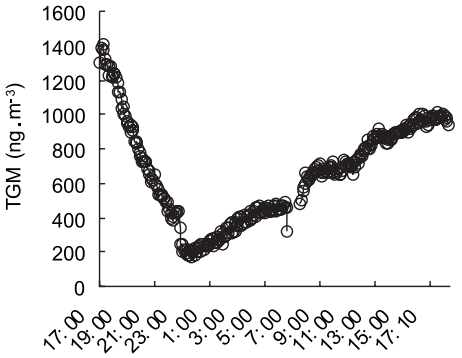

Fig.1 The concentration and trend of TGM in a 6-months landfill vent gas

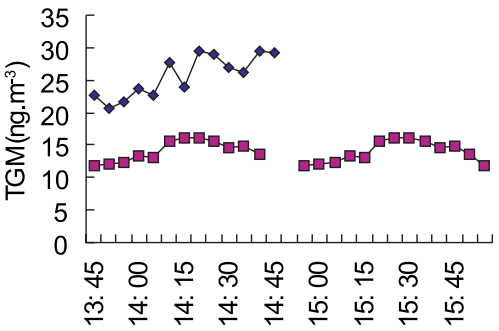

Fig 2. The TGM concentration in one year and two years landfill vent gas

a clear trend of higher TGM concentrations in daytime and lower TGM at night. The average concentration of TGM in 6-month vent gas was 665.52 ± 291.25 ng/m^3 (n=305). TGM data obtained from a 12-month and 24-month vent gas (Fig. 2), with the average concentration of 25.6 ± 3.2 ng /m^3 (n=13) and 14.5 ± 1.8 ng/m^3 (n=28) respectively, were lower than that of 6-month vent gas. TGM concentration of this landfill vent gas is comparable to Nan-ji-du landfill in Seoul (3.45- 2952 ng/m^3), Palm Beach landfill (630 ± 920 ng/m^3) and Martin county (560 ± 560 ng/m^3) in Florida (KIM, 2002; LINDBERG, 1999). Whereas Brevard county landfill in Florida showed a highly consistent level of TGM (7200 ± 300 ng/m^3).

LINDBERG (2001) used Henry's Law to estimate the MMHg concentration in Brevard county landfill to be about 6 ng/m^3. In this work, MMHg was detected with the average concentration of MMHg in 6-month vent gas and 24-month vent gas was 2.06 ±1.82ng/m^3 (n=11) and 0.18 ± 0.06 ng/m^3(n=2), respectively (Table 1).

DMHg in landfill gas are the only measured anthropogenic emission sources of methylated mercury compounds (LINGDBERG, 2001). We also measured DMHg concentration with the average value of 9.45 ± 5.18 ng/m^3, which represented nearly 1.5 % of TGM in landfill gas.

CONCLUSIONS

The positive higher-level concentration of TGM, MMHg and DMHg in this landfill vent gas indicated the landfill was not only a TGM source, but also a type of toxic MMHg and DMHg source to the atmosphere.

Acknowledgements

This work was supported financially by the National Natural Science Foundation of China (No.40203009). We would like to thank the staff of Gaoyan landfill for their heartily support in the sampling.

REFERENCES

BLOOM, N. S., VON DER GEEST, E. J. (1995): Matrix modi.cation to improve the recovery of MMHg from clear water using distillation; *Water, Air and Soil Pollution* 80, pp. 1319-1323.

COMPEAU, G. C., BARTHA, R. (1985): Sulfate-reducing bacteria: principal methylators of mercury in anoxic estuarine sediment; *Applied Environmental Microbiology* 50, pp. 498-502.

KIM, K.H., KIM, M.Y. (2002): Mercury emissions as land.ll gas from a large-scale abandoned land.ll site in Seoul; *Atmospheric Environment* 36, pp. 4918-4928.

LIANG, L., HORVAT, M., BLOOM, N. S. (1994): An improved speciation method for mercury by GC/CVAFS after aqueous phase ethylation and room temperature precollection; *Talanta* 41, pp. 371-379.

LINDBERG, S. E., PRICE, J. (1999): Measurements of the airborne emission of mercury from municipal land.ll operations: a short-term study in Florida; *Journal of Air and Waste Management Association* 49, pp. 174-185.

LINDBERG, S. E., WALLSCHLAGER, D., PRESTBO, E. M., BLOOM, N. S., PRICE, J., REINHART, D. (2001): Methylated mercury species in municipal waste landfill gas sampled in Florida, USA; *Atmospheric Environment* 35, pp. 4011-4015.

REIMANN, D. (1986): Mercury output from garbage incineration; *Waste Manage. Res.* 4, pp. 45-56.