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The dependence of the methylation of mercury on the landfill stabilization process and implications for the landfill management

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HIGHLIGHTS

- The semi-aerobic landfill was more favorable for the release of Hg.
- The anaerobic landfill was more conductive to the methylation of Hg.
- The ORP condition was the controlling factor of the methylation process.

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ABSTRACT

Mercury species and other chemical characteristics of the leachate from anaerobic and semi-aerobic landfills were analyzed to investigate the factors that control mercury methylation during the landfill stabilization process. At the early landfill stage, the total mercury (THg) and the monomethyl mercury (MMHg) released rapidly and significantly, the THg concentration of the semi-aerobic landfill leachate was obviously higher than that of the anaerobic landfill leachate, while compared with the semi-aerobic landfill, the MMHg concentration in the anaerobic landfill was higher. As the landfill time increased, both of THg and MMHg concentration decreased quickly, the THg concentration in the anaerobic landfill was much higher than that in semi-aerobic landfill, while the MMHg concentration in the anaerobic landfill was lower than that in the semi-aerobic landfill. Generally, the concentrations of dimethyl mercury (DMHg) in the anaerobic landfill leachate were slightly higher than in the semi-aerobic landfill leachate during the stabilization process. A significant positive correlation was found between the DMHg concentrations and the pH value in anaerobic landfill leachate, but this correlation was opposite in the semi-aerobic landfill. The oxidative–reductive potential (ORP) condition was found to be the controlling factor of the methylation process during the early stage. However, the chemical characteristics, especially the TOC concentration, appeared to be the dominant factor affecting the methylation process as the landfill time increased.

1. Introduction

Mercury (Hg) is well known to bioaccumulate and biomagnify as neurotoxic methylmercury (CH_3Hg^+) in organisms, particularly fish, and the consumption of these fish and other organisms represents the primary exposure route of mercury to humans (Lawson and Mason, 1998; Clarkson and Magos, 2006). The species of Hg will convert between each other under certain conditions (Fig. 1).

Due to the unseparated solid waste collection system widely adopted in China, a large amount of Hg-bearing solid waste, such as fluorescent lights, batteries, and thermometers, is collected and disposed of in landfills. As this refuse degrades, the mercury is discharged via landfill gas or leachate, resulting in secondary pollution to the environment. Numerous studies have indicated that landfills have become a potential mercury emission source, especially of methylated mercury (Lindberg and Price, 1999; Lindberg et al., 2001; Kim et al., 2001; Feng et al., 2004; Lindberg et al., 2005; Southworth et al., 2005; Li et al., 2010). However, all of these studies just focused on Hg species in landfill gas (LFG) and the Hg flux of the landfill surface, while the Hg species in the leachate were rarely mentioned, which can assistant in making clear of the methylation process in the landfill.

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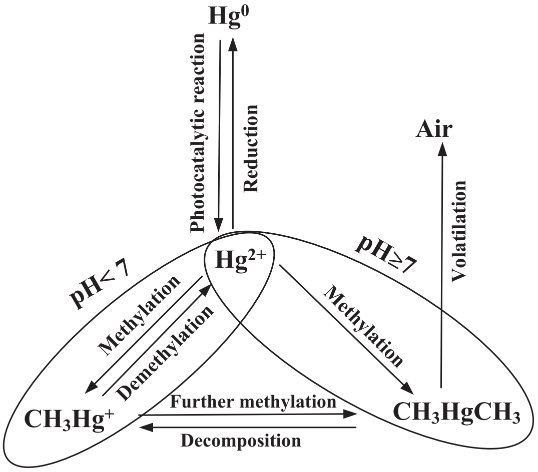

Fig. 1. The conversion between Hg species.

Previous studies have reported that the highly toxic compound methyl mercury is readily formed under the reducing conditions of anaerobic landfills via the activity of sulfate- and iron-reducing bacteria (SRB and IRB, respectively) (Acha et al., 2012; Yu et al., 2012). The formation of monomethyl mercury (MMHg) is a complex biochemical process driven by a wide variety of environmental factors (Ullrich et al., 2001), such as microbial activity and the concentration of the bioavailable Hg (rather than the total Hg pool), which, in turn, are associated with parameters such as temperature, pH, organic material, ORP, sulfide concentration, and salinity (Celo et al., 2006; Sunderland et al., 2006; Hammerschmidt et al., 2008; Kwaansa-Ansah et al., 2012).

A large body of literature is now available on the distribution of Hg and the factors affecting methylation in aquatic and sediment environments (Ullrich et al., 2001; Louis et al., 2007; Han et al., 2010). However, due to the more complicated environmental conditions in landfills, the distribution and methylation mechanisms of mercury are expected to be markedly different and more complex than in natural environments and have rarely been examined. In particular, the environmental factors that control the distribution and transformation of mercury species in both semi-aerobic (Aziz et al., 2010) and anaerobic landfills remain poorly understood.

In the present study, field experiments were used to investigate the distribution of total mercury (THg), MMHg, dimethyl mercury (DMHg) and other chemical characteristics of the leachate at different time during the stabilization process of both anaerobic and semi-aerobic landfills. The aim of this study was to find out factors that influence Hg methylation in landfills and provide guidance and recommendations for landfill management.

2. Materials and methods

2.1. Landfill site studied

The studies were carried out on a simulated semi-aerobic landfill and a simulated anaerobic landfill, both of which were constructed in situ in the Shanghai Lao Gang Refuse Landfill. A total volume of 5000 m^3 of fresh refuse was placed in each of the simulated semi-aerobic and anaerobic landfills from on June to July in 2009. The schematic diagram is shown in Fig. 2.

For the semi-aerobic landfill, the erect gas venting pipe, 25 cm in diameter, was fixed on a portion of the leachate collecting pipe. The erect gas venting pipes were perforated and protected by gravel cages (40 cm in diameter) to allow air flow and penetration into the landfill body. The leachate collection pipe (30 cm in diameter) and the gas venting pipe were connected, making it possible for the ambient air naturally flow into the landfill body through the leachate collection pipes and subsequently contribute to the aerobic condition. In order to further accelerate the air flow into the landfill body, a cowl was installed above the vent pipe. A leachate collection well was installed at lower ground level than the landfill site and was connected to the main leachate collection pipes in order to collect the leachate. In comparison, a standard anaerobic landfill with the same capacity was constructed according to the China standard. It is worth noting that the leachate collection pipe and the gas vent pipe were not connected, and no cowl was installed in anaerobic landfill.

2.2. Waste material

The refuse used in this experiment was fresh domestic garbage in Shanghai, the basic characterization of the refuse was listed in Table 1. The average content of Hg in the refuse was 0.25 ± 0.092 mg kg^{-1}.

2.3. Sample collection and preparation

The leachate samples were taken from the outlet of the leachate collecting pipe at 4 time points (282, 452, 654, 806 d), labeled A1, A2, A3, and A4, respectively, for the samples from the anaerobic landfill and S1, S2, S3, and S4, respectively, for the samples from the semi-aerobic landfill.

At each sampling point, the leachate was collected in borosilicate glass bottles, which were acid-cleaned and then pre-cleaned by heating for 1 h in a muffle furnace at 500 °C. All of the samples were stored in the dark at 4 °C. The mercury species were analyzed within 24 h after the samples were taken back to the laboratory.

2.4. Analytical method

Prior to the anion analysis, all of the samples were centrifuged and filtered with a 0.45 μm microfiltration membrane. The pH of the leachate was measured with a pH meter (PHS-3C, Shanghai precision & scientific instrument, China), and the TOC and TN (nitrogen) were measured with a TOC analyzer (Shimadzu Inc., Japan). The chlorine and sulfate levels were measured with an ion chromatography system (ICS-1000, Dionex, USA) that was equipped with an AG11-HC column with potassium hydroxide used as the eluent.

For the THg analysis, 25 mL of leachate was transferred to 50 mL pre-cleaned centrifuge tubes and digested with 5 mL of a

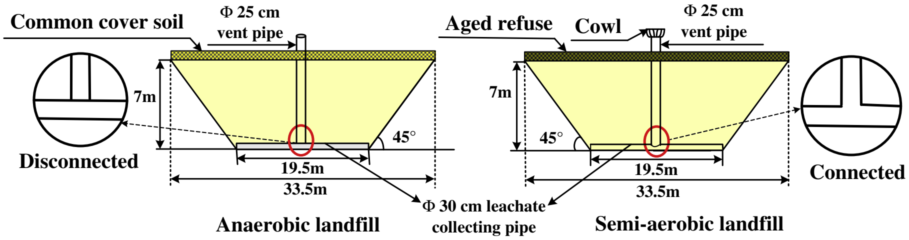

Fig. 2. A schematic diagram of the landfill site.

Table 1
The mass compositions of the refuse (%) (wet base).

Average	Content (%)
Paper	9.43
Plastic	26.03
Wood bamboo	0.91
Cloth	3.76
Kitchen waste	53.31
Fruit peel	2.30
Metal	0.37
Glass	1.86
Ceramics and stone	1.97
Coal ash	0.04
Others	0.01

These data was provided by the Monitoring Center of Shanghai Environmental Sanitation Engineering Design Institute.

fresh strong-acid solution ($HNO_3 + H_2SO_4$, v/v 3:1) in a water bath at 95 °C for 2 h. After 1.5 h, 0.5 mL of BrCl was added, followed by reduction with NH_2OH HCl and $SnCl_2$. Finally, the reduced samples were detected by cold vapor atomic absorption spectrometry (CVAAS).

The MMHg concentrations in the leachate were determined using solvent extraction (CH_2Cl_2), Sodium Tetraethylborate (NaBEt4 ethylation), and isothermal gas chromatography cold vapor atomic fluorescence spectrometry (GC-CVAFS) detection according to USEPA Method 1630 (USEPA, United States Environmental Protection Agency, 2001). 40 mL of leachate was transferred to 50 mL pre-cleaned centrifuge tubes before the addition of 5 mL of CH_2Cl_2, 0.5 mL of HCl, and 1 g of Na_2SO_4. The subsequent procedure was the same as for the measurement of MMHg in sediments (Liang et al., 2004).

For the DMHg analysis, the samples were purged with high-purity nitrogen gas onto Carbotrap^{TM} adsorbent (20/40 mesh, Supelco Inc., Bellefonte, PA). The DMHg on the Carbotraps was determined using GC-CVAFS (Horvat et al., 1993; Conaway et al., 2009).

Quality control for the THg, MMHg and DMMHg analyses was implemented with the use of method blanks, blank spikes, matrix spikes, and blind duplicates. The detection limit for THg was 0.1 ng L^{-1} at a blank level of 0.3 ng L^{-1}. The detection limit for MMHg was 0.1 ng L^{-1} at a blank level of 0.1 ng L^{-1} in water samples. The detection limits for DMHg was 0.1 pg L^{-1} (as mercury) based on 3 times deviation of the blank samples. All the samples were determined by 3 blind duplicates. The recoveries on the matrix spikes of THg, MMHg in water samples were 81.5%, 117.5%, respectively.

3. Results and discussion

3.1. THg, MMHg, and DMHg concentrations

A pronounced decrease in the THg concentrations was observed from 18914 ± 2011 to 294 ± 64 ng L^{-1} in the semi-aerobic landfill and from 14103 ± 1365 to 621 ± 48 ng L^{-1} in the anaerobic landfill (Fig. 3(a)), which was much higher than that in rivers (<10 ng L^{-1}) (Hurley et al., 1995). After day 452, the concentrations of THg in both landfills exhibited a slight declining trend. This process indicated that the release of Hg from the refuse was much more obvious during the early stage, especially in the first year after landfill. As shown in Fig. 4 (Han, 2011), the pH value of the leachate was acid at the early stage, which potentially facilitated the dissolution and leaching of Hg from refuse. After one year, the pH value of the leachate in the two landfills was similar and maintained around 8, reduced the releasing rate of Hg.

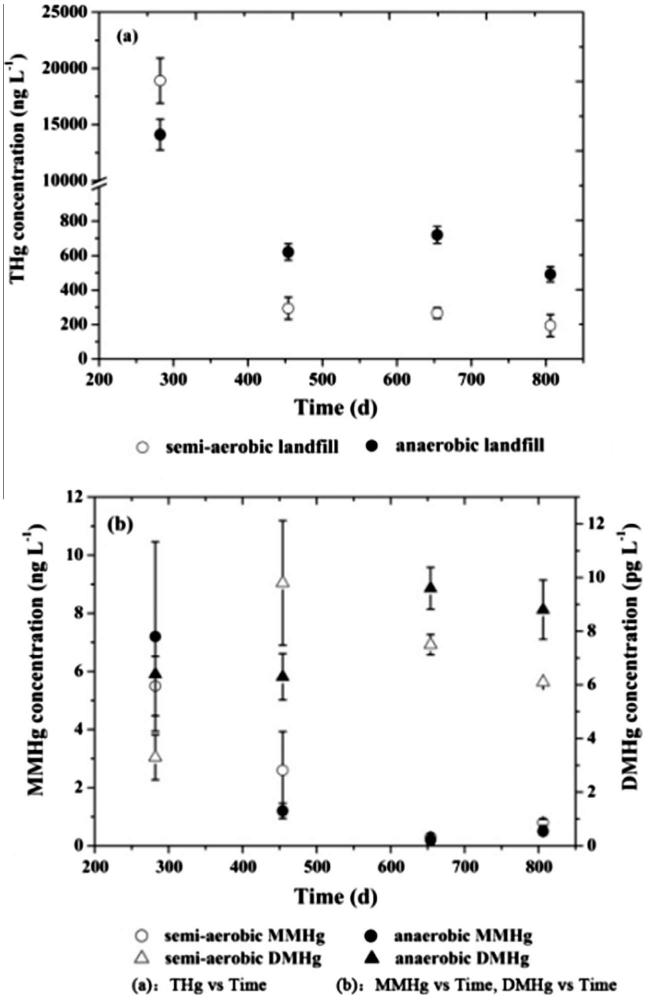

Fig. 3. Effect of time on leachate THg, MMHg and DMHg concentrations in semi-aerobic and anaerobic landfills.

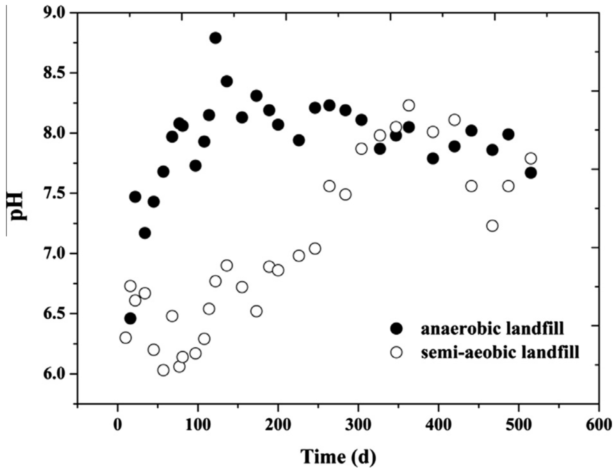

Fig. 4. Effect of time on leachate pH value from semi-aerobic and anaerobic landfills.

When the operation time was less than one year, the THg concentration at S1 was obviously higher than that at A1. However, the THg average concentrations of the anaerobic landfill leachate were twice higher than that of the semi-aerobic landfill leachate

(Fig. 3(a)) as the landfill time increased. The pH value of the leachate in semi-aerobic landfill was obviously lower than that in anaerobic landfill during the first year after landfill, which was more favorable for the release of Hg. In addition, the structure of semi-aerobic landfill facilitated the forming of aerobic environment, and thus was more conductive to the degradation of refuse and the release of Hg compared with anaerobic landfill (Borglin et al., 2004; Giannis et al., 2008; Erses et al., 2008). Therefore, the Hg in anaerobic landfill experienced a relatively slower releasing process compared with that in semi-aerobic landfill at the early stage, thus at the later stage more Hg will be released with the further degradation of refuse in anaerobic landfill.

The MMHg was mainly produced during the early stage of the semi-aerobic and anaerobic landfills, with concentrations of 5.5 ± 1.02 and 7.2 ± 3.26 ng L^{-1}, respectively. The MMHg concentration of the anaerobic landfill leachate first decreased quickly and then fluctuated at 0.8 ± 0.11 ng L^{-1} during the later stage. Although the concentrations of MMHg in the semi-aerobic landfill leachate exhibited the same trend as those in the anaerobic leachate, the average MMHg concentration at A1 was clearly higher than that at S1, whereas the concentrations of MMHg at A2, A3, and A4 were lower than those at S2, S3, and S4, respectively (Fig. 3(b)).

Compared with the concentrations of THg and MMHg, the concentration of DMHg showed a different trend as the landfill time increased. DMHg formation is favored under neutral and basic (pH > 7) conditions, the varied basic pH value was considered to contribute to the variation concentration of DMHg (Jensen and Jernelöv, 1969; Fagerström and Jernelöv, 1972). The concentration of DMHg ranged from 3.3 to 9.8 pg L^{-1}, which is approximately at the same level as seawater (Mason et al., 1998; Louis et al., 2007; Conaway et al., 2009). The low stability, rapid degradation rate of DMHg contributed to the low DMHg levels (Black et al., 2009). In addition, since DMHg is highly volatile, there is possibility that DMHg produced inside the landfill and majority was emitted through the vent pipes, only a small portion was retained in the leachate. The concentrations of DMHg in the anaerobic landfill leachate were higher than those in semi-aerobic landfill leachate except at the second time point, which can be attributed to the higher methylation and lower demethylation rates under anaerobic conditions than under aerobic conditions (Olson and Cooper, 1976; Compeau and Bartha, 1985). Therefore, the anaerobic condition was relatively more favorable for the formation of DMHg.

At A1 and S1, the proportion of MMHg to THg was 0.05% and 0.03%, respectively. This low proportion can be linked to the extremely high concentrations of THg released during the early stage. The percentages of the methylated Hg were 0.88%, 0.11%, and 0.41% at S2–S4, respectively, and 0.19%, 0.03%, and 0.10% at A2–A4, respectively. The higher MMHg concentrations and the MMHg ratio at A1 compared with that of S1 can be ascribed to the low ORP in the anaerobic landfill. Under aerobic conditions, the activity of SRB was reduced and the demethylation of MMHg was accelerated, resulting the lower production and stability of MMHg (Olson and Cooper, 1976; Delaune et al., 2004; Bridou et al., 2011).

The lower pH value in semi-aerobic landfill leachate appears to increase the amount of available Hg for methylation bacteria, which would result in favoring the methylation of Hg. This increase may be related to H^+ compete with Hg^{2+} for combination with organic compounds (Barkay et al., 1997). In addition, under acidic conditions H^+ stimulates methylmercury production by accelerating the adsorption of methylation bacteria for Hg^{2+} (Kelly et al., 2003). Therefore, the lower pH value in semi-aerobic landfill made it more favorable for Hg methylation. However, at the early stage, the MMHg concentration in semi-aerobic landfill was lower than that in anaerobic landfill, implying that the ORP was the dominate factor of Hg methylation process at the early stabilization stage rather than the pH value.

With the landfill time increased, the concentration and percentage of MMHg at S2–S4 became higher than those at A2–A4, implying that the factor that dominated methylation process changed and that the ORP was no longer the controlling factor. In order to make clear which factor dominated the methylation process in the later stage, the correlations between leachate characteristics and MMHg concentration were explored.

3.2. The correlation between leachate characteristics and MMHg concentration

3.2.1. pH

No correlation was found between the MMHg concentration and the pH value in this study (Fig. 5(a)). However, a significant correlation between the DMHg concentrations and the pH value was observed (Fig. 5(b)). This was because that low pH values favor the production of MMHg (Miskimmin et al., 1992; Kelly et al., 2003) and high pH values favored the formation of volatile DMHg (Jensen and Jernelöv, 1969; Fagerström and Jernelöv, 1972), the basic pH values observed in this study are not favorable for the production of MMHg but stimulate the formation of DMHg.

Interestingly, the correlations between the DMHg concentrations and the pH of the semi-aerobic and anaerobic landfill leachate exhibited opposing trends, which was found to be positive in anaerobic landfill and negative in semi-aerobic landfill. The apparently conflicting effect of pH on the mercury methylation under aerobic and anaerobic conditions can be ascribed to the different ORP conditions. The high pH and anoxic condition of semi-aerobic landfill favor the dimethylmercury formation, while the lower pH and lower ORP of anaerobic landfill stimulate methylmercury production and accumulation (Michael et al., 1990).

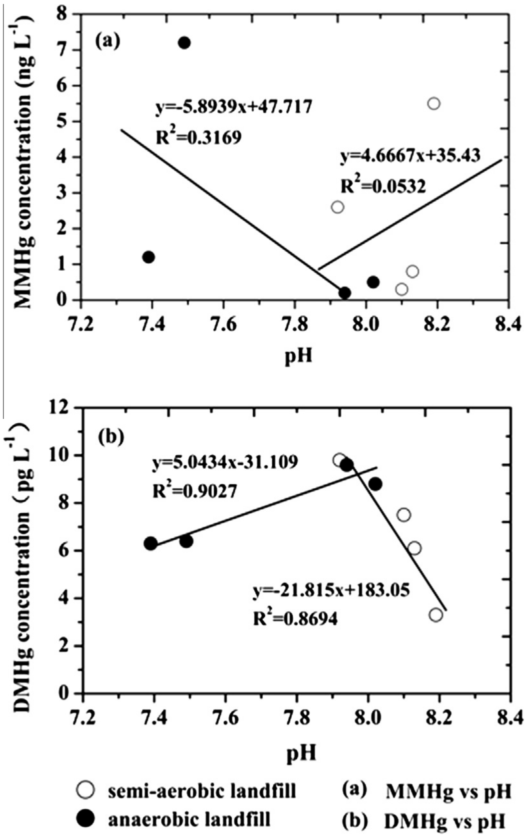

Fig. 5. The correlation between the MMHg, DMHg concentration and pH.

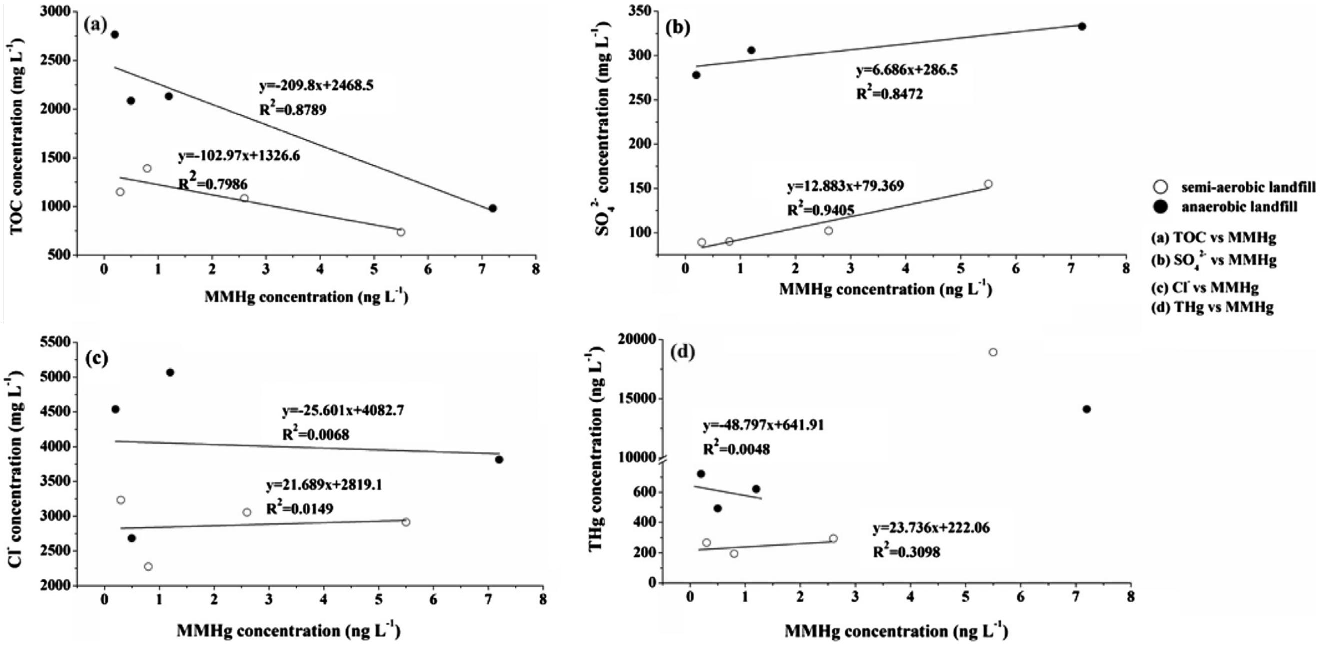

Fig. 6. The correlation between leachate characteristics and MMHg concentration.

3.2.2. TOC

The amount of organic matter is a major controlling factor on the availability of dissolved inorganic Hg to methylating bacteria and may affect methylation and the MMHg concentrations through several mechanisms. A negative relationship between the MMHg concentrations and the TOC was found, especially in the anaerobic landfill leachate (Fig. 6(a)). This relationship exists because high TOC values inhibit Hg methylation by complexation with Hg, thus reducing the availability of the dissolved inorganic Hg to the methylating bacteria, particularly in the neutral pH range (Ullrich et al., 2001). Therefore, the high TOC levels have a mitigating effect on both the production and bioaccumulation of MMHg. Furthermore, humic acid (HA), as a component of the TOC, was an important factor in controlling the solubility and mobility of Hg. On one hand, HA was capable of reducing Hg^{2+} to Hg^0, leading to the reduced availability of Hg^{2+} for methylation (Gua et al., 2011). On the other hand, the degree of humification of HA and the amount of organic matter increased as the landfill time increased (Chai et al., 2013), thus decreasing the availability of effective carbon, such as the electron-donating group like O–CH_3, alkal-C, which is one of the major variables affecting SRB populations and activities (Lambertsson and Nilsson, 2006). The methylation of Hg is therefore limited by the increasing TOC of the landfill leachate.

3.2.3. Sulfate

Sulfate-reducing bacteria are important methylators of Hg in anaerobic sediments, and sulfate is a prerequisite for SRB, which can stimulate the formation of MMHg. In this study, all of the concentrations of sulfate in the leachate were much higher than 50 mg L^{-1}, which exceed the optimal sulfate concentration ranges (20 to 50 mg L^{-1}) for Hg methylation by SRB as reported by Gilmour et al. (1992). Previous findings suggested that low sulfate concentrations stimulate the reduction of SO_4^{2-} to sulfide by SRB, and this SO_4^{2-} reduction stimulates Hg methylation (King et al., 1999; Han et al., 2010; Bergman et al., 2012), whereas high sulfate concentrations that exceed the optimal concentration ranges inhibit methylation (Gilmour et al., 1992; Shao et al., 2012). With the oxygen in the refuse consumed up, the ORP in the landfill decreased and became anaerobic, which was favorable for the reduction of sulfate. In the presence of hydrogen sulfide, Hg^{2+} precipitates as HgS, thus reduced the bioavailability of Hg^{2+}. Therefore, the high sulfate concentration in the leachate had a negative effect on the methylation of mercury. However, the positive correlation between the sulfate and MMHg concentrations indicate that other characteristics of the landfill leachate, such as TOC, exerted stronger influences on the Hg methylation than sulfate (Fig. 6(b)).

3.2.4. Cl^- and THg

In this study, no correlation was observed between Cl^- and MMHg (Fig. 6(c)), indicating that Cl^- may have a minor effect on the Hg methylation. Similarly, there was also no correlation between the THg and MMHg concentrations (Fig. 6(d)), which was in close agreement with the reports that THg concentrations are generally not useful in predicting MMHg concentrations (Kelly et al., 1995; Lambertsson and Nilsson, 2006).

In summary, TOC of the leachate played an important role in Hg methylation, which exhibited a negative effect on Hg methylation, while the pH value, SO_4^{2-}, Cl^- and THg did not show important effects on methylation as they were expected. Compared with the semi-aerobic landfill, the anaerobic landfill possess higher concentrations of TOC, implying that the inhibition effect of TOC on methylation process was much bigger in the anaerobic landfill and thus resulted the lower MMHg concentration in the anaerobic landfill leachate. These results demonstrated that with the landfill time increased, leachate characteristics, especially TOC, became the dominating factor that controlled Hg methylation process at the later stage.

3.3. Implications for the landfill management

The high concentrations of THg and MMHg in the leachate indicated that the leachate was a potential emission source of THg and MMHg, especially at the early landfill stage. The majority of Hg was released during the early landfill stage of the semi-aerobic landfill, which is convenient for the management of Hg pollution. Although the semi-aerobic landfill was more conducive to the formation of MMHg compared with the anaerobic landfill in the later stage, both the levels of MMHg in the anaerobic and semi-aerobic landfill leachate were very low. In addition, the release of Hg in the anaerobic landfill involved an extended span of time, which would require much more attention on the part of landfill managers to prevent Hg contamination in the anaerobic landfill throughout the long stabilization process. Therefore, easy control of Hg pollution is a further advantage of semi-aerobic landfill compared with anaerobic landfill.

4. Conclusions

At the early landfill stage, THg and MMHg released significantly and rapidly, the semi-aerobic landfill was more favorable for the release of THg, ORP was found to control the methylation process. With the landfill time increased, the release amount of THg and MMHg decreased fast, the concentration of THg in the anaerobic landfill leachate was much higher than that in semi-aerobic landfill, the semi-aerobic landfill seemed to be more favorable for the formation of MMHg, and the chemical characteristics especially TOC concentration appeared to be the dominant factor affecting the methylation process. Compared with the semi-aerobic landfill, the anaerobic landfill suffered a longer Hg releasing extension time. Therefore, considering the controlling of Hg pollution, the semi-aerobic landfill was a sound choice.

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