



# The effect of aerobic conditions on the complexation ability between mercury and humic acid from landfill leachate and its implication for the environment



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## HIGHLIGHTS

- ▶ The S content as well as the oxygen-containing ligands of HA played a key role in the complexation with mercury.
- ▶ The anaerobic technique was expected to increase the reaction between Hg and HA.
- ▶ In the long run, the semi-aerobic landfill was more conducive to the transfer of Hg.

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## ABSTRACT

Three-dimensional excitation emission matrix (3DEEM) fluorescence spectroscopy was employed to investigate the structure and characteristics of humic acid (HA) from landfills at different stabilization processes. The results show that the HA in anaerobic landfill leachate stabilized more rapidly than that in semi-aerobic landfill leachate. There were strong interactions between HA and Hg, the S content as well as the oxygen-containing ligands of the HA played a key role in the complexation with mercury. The higher complexation capacity ( $C_L$ ) and stability constant ( $\log K$ ) of HA from anaerobic landfill leachate implies that it is important to strengthen the control of mercury transportation in anaerobic landfills during the early stabilization process. The  $\log K$  and  $C_L$  of HA from semi-aerobic landfill leachate increased with the landfill time indicate that control of leachate Hg contamination in the latter stage is of great significance in semi-aerobic landfills.

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## 1. Introduction

Due to the adopted nonseparated solid waste collection system, a large amount of Hg-bearing solid waste, such as fluorescent lights, batteries, and thermometers, is transported and disposed of in landfills. With the degradation of organic waste, Hg is converted into volatile elemental mercury and highly toxic MeHg (methyl mercury) through biotic and abiotic processes (Lindberg and Price, 1999; Kim et al., 2001; Feng et al., 2004; Li et al., 2010). The conventional anaerobic landfill markedly favors the methylation of Hg and has become an important source of MeHg (Lindberg et al., 2001; Raloff, 2001).

Functional groups of HA, such as carboxyl, hydroxyl and especially thiol, interact very strongly with mercury in the soil, sedi-

ment and aquatic environments (Xia et al., 1999; Haitzer et al., 2002; Ravichandran, 2004). The HA of the landfill differs substantially from that of natural water and soil, due to their different sources and environment conditions (Kang et al., 2002; Sierra et al., 2005; Fu et al., 2007). Furthermore, notable changes were observed in terms of the features, structure and humification degree of HA as time went by (Chai et al., 2007, 2012a; Huo et al., 2008), thus leading to discrepancies in the binding mechanism and binding strength with mercury (Chai et al., 2012b).

A semi-aerobic landfill cannot only accelerate the landfill stabilization process but also reduce the organic substance concentration in the landfill leachate in situ (Borglin et al., 2004; Bilgili et al., 2008; Aziz et al., 2010). Maie et al. (2002) reported that the redox environment exerts a significant influence on the humification process of HSs. These investigations indicated that HA from different landfill technologies were quite distinctive. The complexation between HA and Hg could reduce the methylated mercury production due to the decreased availability of dissolved inorganic Hg to methylating bacteria (Ullrich et al., 2001). Gua et al. (2011)

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found that HA were capable of reducing  $\text{Hg}^{2+}$  to  $\text{Hg}^0$ , leading to lowering the availability of  $\text{Hg}^{2+}$  for methylation. Therefore, The interaction of HA and Hg could influent the transporting and fate of Hg in the landfill and surrounding environment. Although a great deal of literature is now available on the interactions of  $\text{Hg}(\text{II})$  with HA in natural sediments and aquatic environments, the interaction of HA from different aerobic conditions with Hg and its potential impacts on the species and fate of Hg have rarely been examined for different landfill stabilization processes.

In this study, three-dimensional excitation emission matrix (3DEEM) fluorescence spectroscopy was employed to investigate the structure and characteristics of HA from landfill leachate. The complexation capacity ( $C_L$ ) and stability constant ( $\log K$ ) between mercury and HA from different aerobic conditions were determined to examine the role of HS on the release and transfer of Hg from a landfill. Extended knowledge of Hg binding to HA in a landfill will be useful for predicting the transport and fate of Hg and for developing effective measures to prevent Hg from migrating and dispersing from landfills into the surrounding environment.

## 2. Materials and methods

### 2.1. Sample collection and preparation

The simulated semi-aerobic landfill and anaerobic landfill were constructed in situ in the Shanghai Refuse Landfill. The top area and the bottom area were  $33.5 \text{ m} \times 33.5 \text{ m}$  and  $19.5 \text{ m} \times 19.5 \text{ m}$ , respectively. The height of the landfill was approximately 7 m, with a slope of 1:1. To prevent the leachate from polluting the groundwater, a high-density polyethylene (HDPE) membrane was placed on the landfill floor, and leachate collecting pipes with the diameter of 30 cm were installed on the HDPE membrane. For the semi-aerobic landfill, three erect gas venting pipes 30 cm in diameter were 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 air penetration into the land layer. The leachate collection and gas venting pipe make the ambient air naturally flow into the waste body through the leachate collection pipes and subsequently contribute to the aerobic condition. 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.

A total weight of 5000  $\text{m}^3$  of refuse was placed in the simulated semi-aerobic and anaerobic landfills, respectively, from June to July 2009. At last, cover soil was placed on the surface of the landfill.

Leachate samples from different operation times and different landfill technologies were gathered. S1, S2 and S3 were collected from the semi-aerobic landfill unit in April 2010, October 2010 and April 2011, respectively, while A1, A2 and A3 were gathered from the anaerobic landfill in April 2010, October 2010 and April 2011, respectively.

At each sampling point, leachate was collected using a clean glass bottle and promptly transferred to 4-L dark glass flasks with aluminum caps. These leachate samples were filtered to separate the solid and soluble phases of the leachate with a 1- $\mu\text{m}$  polymer membrane filter. The soluble samples were acidified to  $\text{pH} < 2$  using 12 M hydrochloric acid (HCl) and then stored in the dark at 4 °C.

### 2.2. HA extraction process

Isolation and purification procedures were carried out according to the method recommended by the International Humic

Substances Society. Briefly, the leachate sample was initially filtered with 0.45  $\mu\text{m}$  filter paper and then acidified to  $\text{pH} 2.0$  using 36% concentrated HCl. The acidified leachate was passed through a DAX-8 (SUPELCO, USA.) resin column to absorb HSs present in the leachate. The alkaline eluent containing HSs was acidified to  $\text{pH} 1.0$  using 6 M HCl. After precipitation for 12 h, the HA precipitate was separated from the supernatant by centrifugation (6000 rpm, 20 min). Then, the HA precipitate was purified using a mixture of 0.1 M HCl/0.3 M HF and then dialyzed against distilled water until  $\text{Cl}^-$  could not be detected. All the extractions were freeze-dried for further chemical analysis.

### 2.3. Fluorescence quenching titration

The 3DEEM fluorescence spectra were measured with a fluorescence spectrophotometer (F-4500, Hitachi, Japan) with a 150-W Xe arc lamp. The method and data processing had been described in previous report (Chai et al., 2012b).

## 3. Results and discussion

### 3.1. Fluorescence spectra HA

The EEMs of leachate HA landfill under different aerobic conditions at different stabilization process in the absence and presence of Hg (HA:  $50 \text{ mg L}^{-1}$ ,  $\text{pH} 6.0$ ,  $0.01 \text{ M KNO}_3$ ;  $\text{Hg}^{2+}$ :  $40 \mu\text{mol L}^{-1}$ ) was shown in Fig. 1. A prominent peak (peak B) with relatively high fluorescence intensity appeared for all HA samples with an excitation/emission wavelength pair of 340–345/440–450 nm. In addition, peak A, which has a broad shoulder that extends to shorter excitation wavelengths with a less intense peak at 260–270/440–445 nm, was also detected. These two peaks were related to aromatic and aliphatic groups in the DOM fraction and commonly labeled as fulvic-like fluorescence (Coble, 1996). Peak A was ascribed to UV fulvic-like fluorescence, and peak B was ascribed to visible fulvic-like fluorescence (Baker, 2001; Chen et al., 2003; Leenheer and Croue, 2003). The DOM is mainly composed of HA and FA. With the landfill stabilization process extended, the low molecular weight FA will change into high molecular weight and much more stabilized HA. The fluorescence of HA usually have a higher excitation and emission wavelength (Mobed et al., 1996; Chai et al., 2012b), so the fluorescence position of leachate HA showed a red shift compared with that of DOM. The short wavelength appeared in fluorescence peak A, which suggests that there are simple structural components of wide variation of molecular weight and small molecular weight with low degree of aromatic poly-condensation, low level of conjugated chromophores and low humification degree. On the contrary, the long wavelength of the major peak B may be ascribed to the presence of an extended, linearly condensed aromatic ring network and other unsaturated bond systems, which have large molecular weight units and significant degree of humification (Senesi et al., 1991; Chai et al., 2012b).

With the landfill time extended, the fluorescence intensity of peaks A and B in S1–S3 first decreased and then increased. While the fluorescence intensity of peaks A and B in A1–A3 decreased with the landfill time, especially the peak of A3 almost disappeared ultimately. All the results indicate that there are sensible differences between the HA humification processes of landfill leachate under semi-aerobic and anaerobic conditions. The disappearance of peak A and the decreased fluorescence intensity of the fulvic-like peak of HA in the anaerobic landfill suggest that the leachate HA from the anaerobic landfill stabilized more rapidly than that from the semi-aerobic landfill.

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