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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.

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, sediment 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 Hg^{2+} to Hg^0, leading to lowering the availability of 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 Hg(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 m $*$ 33.5 m and 19.5 m $*$ 19.5 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 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-lm polymer membrane filter. The soluble samples were acidified to $pH < 2$ 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 μm filter paper and then acidified to 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 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 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 mg L^{-1}, pH 6.0, 0.01 M KNO_3; Hg^{2+}: 40 μmol 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.

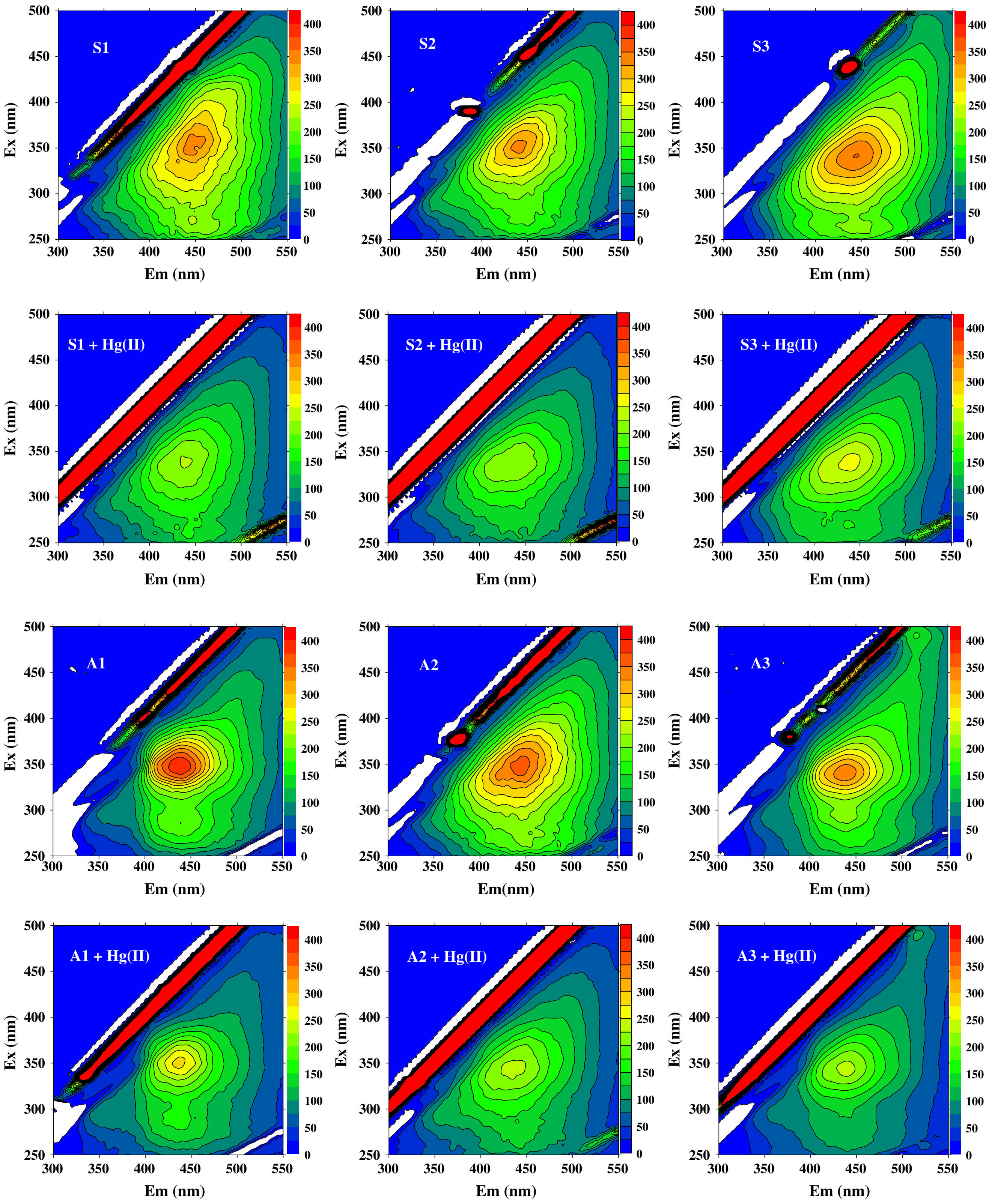

Fig. 1. EEMs of HAs at semi-aerobic and anaerobic conditions (HA: 50 mg L^{-1}, pH 6.0, 0.01 M KNO$_3$; Hg^{2+}: 40 μmol L^{-1}).

Unlike S1, S2, A1 and A2, a less intense fluorescence peak (peak C) at 480–490/515–520 nm was observed in the fluorescence EEM spectrum of S3 and A3, although peak C of S3 was a little obscure. The longer excitation spectrum and the different environment contribute to the appearance of peak C of HA, which was labeled as humic-like fluorescence (Plaza et al., 2006; Huo et al., 2008).

Compared with S1, S2, A1 and A2, the presence of peak C in S3 and A3 can be ascribed to the increased humification degree of HA, which indicates that the humification degree of HA increases as the landfill time was extended. The higher fluorescence intensity of peak C was observed in A3 with respect to S3, suggesting that the anaerobic condition was more favorable than the semi-aerobic condition for achieving high aromatic condensation and a high humification degree. These findings are consistent with the results of elemental and ^{13}C CP/MAS NMR analysis, which indicate that A3 has the highest aromaticity and lowest O content (Tables 1 and 2).

The addition of Hg^{2+} to HA solutions caused relevant changes of the fluorescence EEM spectra, which suggest there are marked modifications of the electronic structure of HA upon interaction with Hg^{2+}. A marked decrease of the fluorescence intensity was observed for all HA samples with the addition of Hg^{2+}. The wavelengths of peak B in S3 and A1–A3 remain almost constant upon Hg^{2+} addition, while a slight blue shift of peak B in S1 and S2 was observed, which indicates that S1 and S2 may undergo greater modification of their electronic structure upon interaction with Hg^{2+}, due to mercury bonds with thiol-type and oxygen-containing functional groups such as carboxyl or phenol groups. After combined with mercury, the structure of humic acid molecular becomes more stable and the fluorescence intensity becomes weak. The complex mode is different when mercury complexes with different functional groups of HA (Skyllberg et al., 2006). For example, mercury is complexed by reduced organic S groups at a distance of 2.33 Å in a linear configuration, while carbonyl-O or amino-N at 2.07 Å and the C atoms at an average distance of 3.14 Å. The different resultant changes of the fluorescence EEM spectra of the HA samples upon interaction with Hg^{2+} demonstrated the different interaction modes and strength of the achieved bonding of Hg^{2+} to HA, which depends on the conformational and functional chemical properties and the humification degree of HA.

3.2. Hg binding parameters of HA

The experimental values of the fluorescence intensity of the main HA fluorophore (peak A) as a function of the Hg^{2+} concentration and the nonlinear regressions generated by best-fitting these data with the model of Ryan and Weber (1982) are shown in Fig. 2. The corresponding best-fit parameters obtained for each HA are listed in Table 3. The high values of the correlation coefficients (r) indicate that the model fits the experimental data sets very well for the HA examined, as was previously demonstrated in other works on similar systems (Plaza et al., 2006; Chai et al., 2012b). The stability constants of mercury ion-HA complexes and the complexing capacities of HAs are described and discussed comparatively as follows.

Under natural conditions (very low Hg/DOM ratios), the binding of Hg to DOM is controlled by a small fraction of DOM molecules containing a reactive thiol functional group (Haitzer et al., 2002), and the Hg-S complexes are quite stable, with a stability constant as high as 10^{20}–10^{47} (Dong et al., 2011). As the experimental Hg/DOM ratios increase, mercury will bind to oxygen functional groups (Yin et al., 1997; Haitzer et al., 2002), which leads to much lower overall stability constants of HA. The much lower stability constants of Hg with HA (log$K$ ranging from 4.3 to 6.0, as shown in Table 3 observed in this study demonstrated that a large portion of Hg forms outer sphere complexes with oxygen-containing functional groups, such as carboxyl or phenol groups. However, it is important to note that the change of complexation capacity (C_L) is generally in accordance with the changes of the S contents in the leachate HA. Therefore, it was the joint function of oxygen-containing ligands and the sulfur-containing ligands that determined the stability constants of Hg–HA complexes and the Hg complexation capacity of HA.

Table 2
Percent distribution of the different C-types of the samples determined by ^{13}C CP/MAS NMR and the aromaticity and aliphaticity degrees.

	S-1	S-2	S-3	A-1	A-2	A-3
0–50 ppm, alkyl-C	33.44	36.72	30.38	30.81	35.32	30.64
50–60 ppm, O—CH_3	8.19	7.06	6.72	7.07	7.52	7.1
60–100 ppm, O-alkyl	9.23	10.93	10.15	10.4	9.3	8.42
100–140 ppm, H-aromatic	26.04	25.16	29.87	28.13	26.55	30.04
140–160 ppm, O,N-aromatic	6.51	6.95	8.78	7.61	8.02	9.56
160–190 ppm, COOH	12.7	9.61	10.79	11.67	9.52	10.98
190–230 ppm, C=O	2.6	3.58	3.31	3.22	2.53	3.27
Aromaticity[a] (%)	39.02	36.98	44.99	42.54	39.87	46.18
Aliphaticity[b] (%)	60.98	63.01	55.01	57.46	60.13	53.82

[a] Aromaticity (%) = [Aromatic C peak area (100–160 ppm)]100/[Total peak area (0–160 ppm)].

[b] Aliphaticity (%) = [Aliphatic C peak area (0–100 ppm)]100/[Total peak area (0–160 ppm)].

Compared with the anaerobic landfill, the lower H/O ratio and higher O/C ratio (Table 1) of HA from the semi-aerobic landfill indicate that the HA has more oxygenic groups, a higher polarity and a lower hydrophobicity, which suggest that it has a high solubility in leachate and has more binding sites for mercury complexation such as phthalic acid-like, salicylic acid-like binding sites, therefore the semi-aerobic condition has a remarkable effect on the migration of Hg through complexation during the stabilization process in the landfill. Two adjacent aromatic carboxyl groups (i.e., a phthalic acid-like binding site) and an aromatic carboxyl group with an adjacent phenolic OH group (i.e., a salicylic acid-like binding site) in the HA structure are known to form highly stable bidentate complexes with metal ions (Elkins and Nelson, 2002; Plaza et al., 2006). Therefore, it can be expected that the leachate HA in a semi-aerobic landfill should possess a relatively higher log$K$ and C_L. However, in this study, the values of log$K$ and C_L were lower in S1 than in A1; this can be attributed to the higher content of S in A1. These results demonstrated that although the content of S was much smaller compared with the content of O and C, S could still exert an influence on the reaction between Hg and HA due to its high binding affinity with Hg and their high stability constant.

With the landfill time extended, the complexation between the Hg and HA was strengthened in the semi-aerobic landfill, while it was weakened in the anaerobic landfill. The increase of log$K$ and C_L in the semi-aerobic landfill was mainly brought about by the S

Table 1
Chemical and structural characteristics of the leachate HA samples.

Samples	Mass (%)					Atomic ratio			
	N	C	S	H	O	C/N	H/C	O/C	H/O
S-1	4.534	40.06	1.747	5.538	48.121	10.31	1.66	0.90	1.84
S-2	3.083	41.79	2.267	5.918	46.942	15.81	1.70	0.84	2.02
S-3	4.41	49.25	2.773	5.692	37.875	13.03	1.39	0.58	2.40
A-1	4.643	43.67	3.487	5.689	42.511	10.97	1.56	0.73	2.14
A-2	3.619	47.2	2.148	6.018	41.015	15.22	1.53	0.65	2.35
A-3	4.889	54.56	2.49	5.769	32.292	13.02	1.27	0.44	2.86

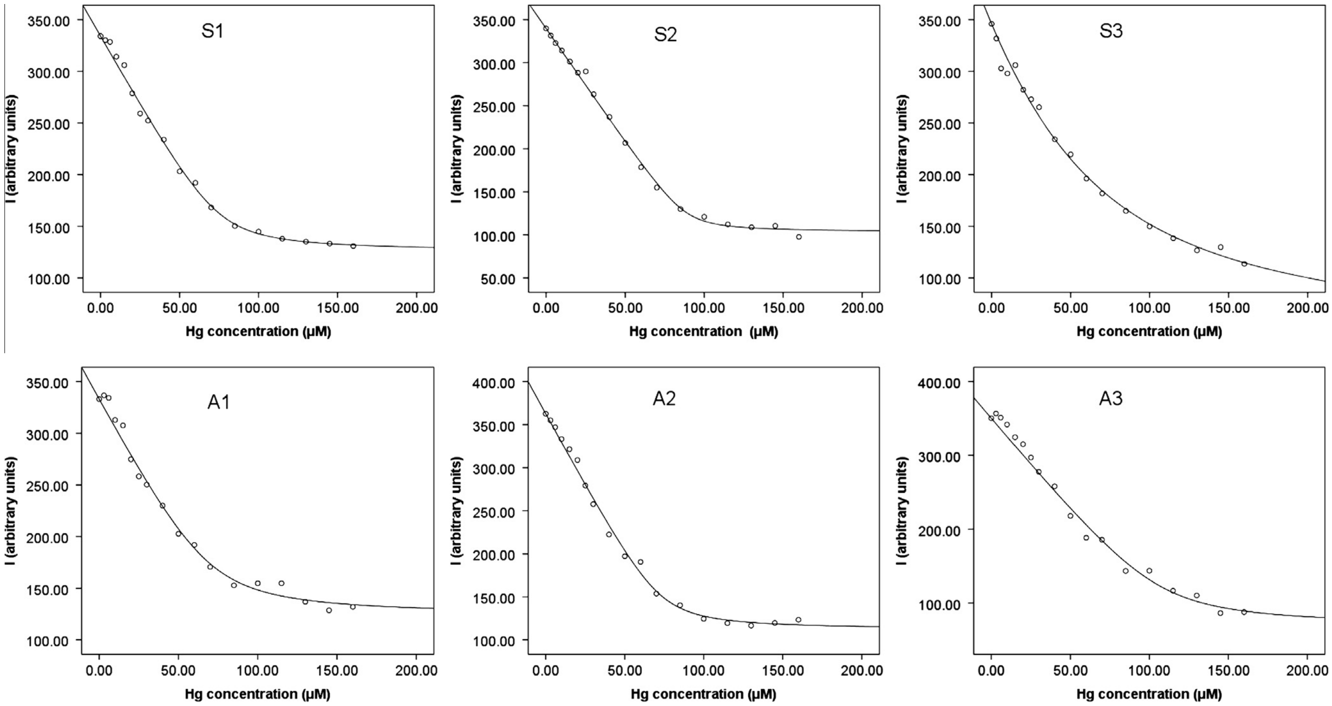

Fig. 2. The fluorescence intensity of the main HA fluorophore (peak A) as a function of the Hg^{2+} concentration (0–160 μmol L^{-1}).

Table 3
Fitting parameters of the nonlinear Ryan-Weber model, i.e., the fluorescence intensity of the Hg(II)–saturated HA complexes (I_{ML}), stability constant (log$K$) of Hg(II)–HA complexes, Hg(II) complexation capacity (C_L) of HA and correlation coefficient for the predicted and measured fluorescence intensities.

Parameter	S1	S2	S3	A1	A2	A3
I_{ML}	123.076	103.037	67.938	125.282	111.434	32.493
log$K$	5.286	6.015	5.307	5.552	5.661	4.335
C_L (μM)	1.415	1.765	2.146	1.512	1.463	0.811
r	0.988	0.997	0.988	0.995	0.995	0.99

content increase. The decrease of logK and C_L in the anaerobic landfill are ascribed to the decrease of the S content and the increase of the aromaticity (Table 2) of HA. The decreased log$K$ and C_L values of A3 can be related to two aspects. On one hand, the carboxylic functional groups decreased with the aromaticity increased, which contributed to the reduction of the number of phthalic acid-like and salicylic acid-like binding sites. On the other hand, the molecular weight and aromatic condensation degree increased with the extended landfill time, which increased the steric hindrance effects for their binding with Hg (Plaza et al., 2006). These results demonstrated that although the S content of the HA plays a key role in the complexation with mercury, the overall stability constant of Hg–HA was also determined by the abundant O-ligands existing in HA.

3.3. Environmental implication

The decrease of the fluorescence intensity of HA with the addition of Hg^{2+} indicates Hg complex with leachate HA, which may inhibit the methylation process in some degree due to the decreased bioavailability of Hg^{2+} to methylated bacteria. The different changes of the fluorescence EEM spectra in different HA samples with the addition of Hg^{2+} demonstrated the interaction between HA and Hg is affected by the components and structure of HA. Different landfill technologies affect the interaction by influencing the physical and chemical properties of the leachate HA. The stability constants and binding capacity determined in this study are beneficial for evaluating the distribution and migration of mercury in landfills, which can provide insight into the control of Hg contamination and landfill management.

The results obtained show that the binding capacities and stable constants of leachate HA in a semi-aerobic landfill are smaller than those in an anaerobic landfill at the early landfill stage, which demonstrated that with respect to the binding capacities and stable constants of leachate HA with Hg^{2+}, the anaerobic technique was expected to increase the complexation between Hg and leachate HA and was more favorable for Hg migration out of the landfill body compared with the semi-aerobic technique. This result suggested that it is important to strengthen the control of mercury transportation and contamination in anaerobic landfills in the early stage.

With the landfill time extended, the complexation between Hg and leachate HA was strengthened in the semi-aerobic landfill, and the log$K$ and C_L values in S3 were obviously higher than those in A3 ultimately, which illustrated that the semi-aerobic landfill technique was more conducive to the transfer of Hg in the long run. Therefore, intensifying the control of leachate Hg contamination in the latter stage is of great significance in semi-aerobic landfills.

4. Conclusion

HA in anaerobic landfill leachate stabilized more rapidly than that in semi-aerobic landfill leachate. The complexation between HA and Hg was strengthened in the semi-aerobic landfill, while it became weakened in the anaerobic landfill with the landfill time extended. Both the S content and the oxygen-containing ligands of HA played a key role in the complexation with mercury. It is important to strengthen the control of mercury transportation and contamination in anaerobic landfills in the early stage. Additionally, intensifying the control of leachate Hg contamination in the latter stage is of great significance in semi-aerobic landfills.

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