



# Retention and leaching of nitrite by municipal solid waste incinerator bottom ash under the landfill circumstance



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## HIGHLIGHTS

- Retention and leaching of  $\text{NO}_2^-$  by MSWI bottom ash affect its migration in landfill.
- Retention of  $\text{NO}_2^-$  is affected by the dosage of MSWI bottom ash and pH condition.
- $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and organic matter have disadvantageous effect on the nitrite retention.
- $\text{PO}_4^{3-}$  can enhance the nitrite retention due to the formation of apatite.

## ARTICLE INFO

### Article history:

Received 16 March 2014

Received in revised form 17 June 2014

Accepted 19 June 2014

Handling Editor: J. de Boer

### Keywords:

Nitrite  
MSWI bottom ash  
Landfill  
Retention  
Leaching

## ABSTRACT

The retention and leaching of nitrite by municipal solid waste incinerator (MSWI) bottom ash could affect its migration in the landfill. In this study, the effect of the dosage of MSWI bottom ash as well as the variation of the landfill environmental parameters including pH, anions and organic matter on the nitrite retention and leaching behavior was investigated by batch experiments. The highest removal percentage (73.0%) of nitrite was observed when the dosage of MSWI bottom ash was  $10 \text{ g L}^{-1}$  in  $2 \text{ mg L}^{-1}$  nitrite solution. Further increase of the dosage would retard the retention, as the nitrite leaching from MSWI bottom ash was enhanced. The optimum retention of nitrite was observed when the pH was 5.0, while the leaching of nitrite showed a consistent reduction with the increase of pH. Besides, the presence of  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and acetic acid could enhance the leaching of nitrite and mitigate the retention process. However, the retention of nitrite was enhanced by  $\text{PO}_4^{3-}$ , which was probably due to the formation of the apatite, an active material for the adsorption of the nitrite. These results suggested that MSWI bottom ash could affect the migration of nitrite in the landfill, which was related to the variation of the landfill circumstance.

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## 1. Introduction

The incineration method has many advantages such as the hygienic control, volume and mass reduction as well as energy recovery for the municipal solid waste (MSW) treatment (Wan et al., 2006). Till 2011, there was a total 109 MSW incinerators (MSWI) in China, with a treatment capacity of  $94414 \text{ t d}^{-1}$  (National Bureau of Statistics of China, 2013). Because of the incineration process, a large volume of MSWI bottom ash is produced (Chimenes et al., 1999). Nowadays, MSWI bottom ash is either reused as a secondary construction material or disposed in landfills with MSW (Youcai et al., 2002). All over the world, such as China

and Japan, and Taiwan area, MSWI bottom ash is allowed to be disposed in the MSW landfill site (Inanc et al., 2007; Lo and Liao, 2007). Moreover, large amounts of MSWI bottom ash has been used as the liner, protection layer and leachate drainage layer instead of natural minerals (e.g., gravel, sand, till, and clay) (Lo, 2000; Travar et al., 2009; Su et al., 2013).

MSWI bottom ash is a mineral assemblage (Yao et al., 2010). When it is disposed or reused in the landfill, various minerals will be leached out, which may aggravate the pollution of the landfill. Several researches have been carried out to investigate the environmental impact of the disposal or reuse of MSWI bottom ash in the landfill. For instance, Lo (2005) and Lo and Liao (2007) pointed out that the disposal could increase the metal concentration in the leachate. However, Inanc et al. (2007) indicated that the disposal of MSWI bottom ash would not increase the metal

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leaching notably. Our previous research demonstrated that the effect of the disposal of MSWI bottom ash on the metal leaching greatly depended on the mass proportion ratio of MSWI bottom ash to MSW. These reports are mainly about the metal pollution caused by the disposal of MSWI bottom ash in the landfill. However, few studies focus on the effect of MSWI bottom ash on the nitrogen pollution. Due to the lack of the nitrogen degradation pathway in the anaerobic system, the nitrogen pollution becomes a long term problem in the landfill. It has turned out to be the research focus in the last decade (He et al., 2006, 2007; Shalini and Joseph, 2012; Wang et al., 2013). On the one hand, since the MSWI bottom ash contains considerable amounts of nitrite (Belevi and Moench, 2000), the leaching of nitrite from MSWI bottom ash may further aggravate the nitrogen pollution of the landfill. On the other hand, MSWI bottom ash contained several hydr(oxide) minerals (e.g., aluminum (hydr)-oxides, iron (hydr)-oxides) and layered double hydroxides (e.g. Friedel's salt), which have great ability to adsorb the nitrite (Islam and Patel, 2011; Yang et al., 2014). Thereby, the migration of nitrite in the landfill could be altered by MSWI bottom ash. According to our previous research, the level of nitrite in landfill leachate is from 0.38 to 2.24 mg L<sup>-1</sup> (see Fig. S1). Nitrite is the intermediate product of the transformation of nitrogen, including the nitrification and denitrification process (He et al., 2006). A good knowledge of the retention and leaching characteristic of nitrite by MSWI bottom ash is a prerequisite to understand the impact of MSWI bottom ash on the nitrogen migration of the landfill. Unfortunately, to our knowledge up to date, few studies have been done to investigate the retention and leaching behavior of nitrite by MSWI bottom ash under the landfill circumstance.

In this work, the retention of nitrite by MSWI bottom ash and its leaching behavior from MSWI bottom ash were investigated by batch experiments. The effect of the dosage of MSWI bottom ash as well as the variation of the landfill environmental parameters including pH, anions and organic matter on the nitrite retention and leaching behavior was studied. We aim to provide insights into the effect of MSWI bottom ash on the nitrogen pollution when it was disposed or reused in the landfill.

## 2. Materials and methods

### 2.1. MSWI bottom ash sampling

Bottom ash from MSWI incinerator was taken from the Green Energy MSWI plant in Zhejiang province, East China. The plant consists of three parallel incinerator stokers with an MSW treatment capacity of 650 t d<sup>-1</sup>. The source MSW was collected from several residential areas of Hangzhou without any industrial solid waste. The operating temperature of the incinerators was 850–1100 °C, and the residence time of waste in the incinerator was about 50 min. Bottom ash had been undergone water quenching and magnetic separation before it was sampled. The sampling period lasted for 5 d. Approximately 25 kg fresh bottom ash sample was taken daily from the plant. A total 125 kg bottom ash sample was obtained. Then, the bottom ash sample was mingled and homogenized. Subsequently, an approximate 25 kg of the MSWI bottom ash was oven-dried and grounded into less than 154 μm by the grinder for bulk composition analysis, leaching and adsorption test.

### 2.2. Characterization

The contents of individual elements in the MSWI bottom ash sample were analyzed after it was digested according to the method of Yamasaki (1997). 0.5 g air dried sample was weighed

into a Teflon beaker. 2.5 mL HNO<sub>3</sub> and 2.5 mL HClO<sub>4</sub> were added and heated on the hot plate at 150 °C for 2–3 h. After cooling, 2.5 mL HClO<sub>4</sub> and 5 mL HF were added and heated at 150 °C for 15 min, and then 5 mL HF was added until the residue became almost dry. The residue was dissolved in 5 mL HNO<sub>3</sub> and diluted to 100 mL. The elemental concentrations in the solution were determined by ICP-OES (Thermo Electron Corporation IRIS/AP, USA). The samples were digested and analyzed in triplicate.

The mineralogy of the bottom ash samples, before and after the retention experiment, was carried out by XRD (Rigaku D/max-r B, Japan).

### 2.3. Leaching experiment

The leaching of NO<sub>2</sub><sup>-</sup> from MSWI bottom ash was studied by batch experiments. The solution and MSWI bottom ash were added to the 100 mL Erlenmeyer flasks. The solution was then continually flushed with N<sub>2</sub> to avoid the contact with the atmosphere and was shaken at 150 rpm by an air-temperature-controlled shaker for 2 h to reach the equilibrium (see Fig. S2(a)). Then, the samples were centrifuged and the NO<sub>2</sub><sup>-</sup> concentration in supernatant was determined immediately by ion chromatograph (Dionex ICS-2000, USA). The effects of the dosage, pH, anions and organic matter on the leaching of nitrite were investigated.

#### 2.3.1. Dosage studies

Dosage of MSWI bottom ash ranged from 0.5 to 8.0 g was added to 100 mL distilled water.

#### 2.3.2. pH studies

Generally, the pH of the practical landfill leachate ranged from 4.0 to 10.0 (Long et al., 2008). In this study, the pH of the distilled water was adjusted to 4.0, 5.0, 6.0, 7.0, 8.0, 9.0 and 10.0 by HCl or NaOH solution. 100 mL pH adjusted water and 1.0 g MSWI bottom ash were added to the flasks.

#### 2.3.3. Anions studies

The effects of the presence of Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and PO<sub>4</sub><sup>3-</sup> on the nitrite leaching were investigated. The concentration of the imposed Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and PO<sub>4</sub><sup>3-</sup> ranged from 0 to 5000, 0 to 2000, and 0 to 10 mg L<sup>-1</sup>, respectively, which was according to our previous research (see Figs. S3 and S4) and literature reports (Kjeldsen et al., 2002). 100 mL solution and 1.0 g MSWI bottom ash were added to the flasks.

#### 2.3.4. Organic matter studies

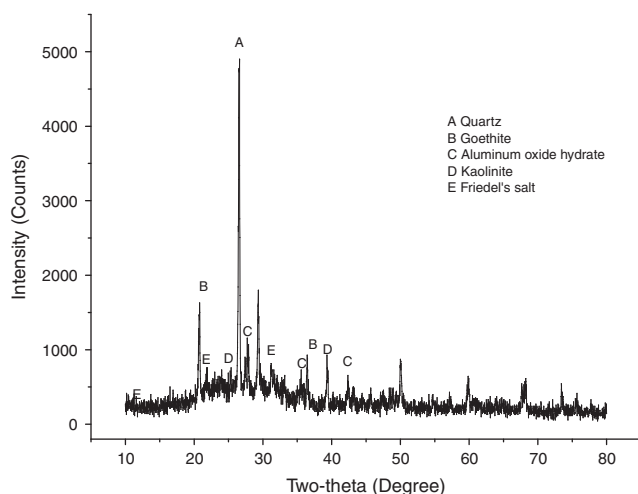
Acetic acid was selected to represent the organic matter in the landfill. The concentration of the imposed acetic acid solution ranged from 10000 to 50000 mg L<sup>-1</sup>, which was corresponding with the variation of COD in the leachate (see Fig. S4, 1.0 mg L<sup>-1</sup> acetic acid equals to 1.06 mg L<sup>-1</sup> COD). 1.0 g MSWI bottom ash and 100 mL acetic acid solution were used in batch experiments.

### 2.4. Retention experiment

The retention of nitrite by MSWI bottom ash was studied by batch experiments. The initial concentration of NO<sub>2</sub><sup>-</sup> was 2.0 mg L<sup>-1</sup>, which was generally in agreement with its presence in the practical landfill leachate (see Fig. S1). MSWI bottom ash and solution were added to the 100 mL Erlenmeyer flasks. The solution was then continually flushed with N<sub>2</sub> to avoid the contact with the atmosphere and was shaken at 150 rpm by an air-temperature-controlled shaker for 2 h to reach the equilibrium (see Fig. S2(b)). Then, the samples were centrifuged and the NO<sub>2</sub><sup>-</sup> concentration in supernatant was determined immediately. The effects of the dosage (0.5–8.0 g), pH (5.0–10.0), ions (chloride,

**Table 1**  
Bulk chemical composition of the MSWI bottom ash sample.

Element	Content (mg kg <sup>-1</sup> dry ash)
Al	40920 ± 1600
Si	223 600 ± 4657
Na	9040 ± 178
K	15 792 ± 167
Mg	5997 ± 115
Ca	69 413 ± 2613
Fe	26 008 ± 28
Mn	1245 ± 231



**Fig. 1.** XRD pattern of the MSWI bottom ash sample.

sulphate, phosphate and carbonate) and organic matter (acetic acid) were investigated, which was in agreement with the leaching test to get a comparable result.

### 3. Result and discussion

#### 3.1. Characterization of MSWI bottom ash

The bulk chemical composition of MSWI bottom ash is exhibited in Table 1. The result showed Si was the major element, which

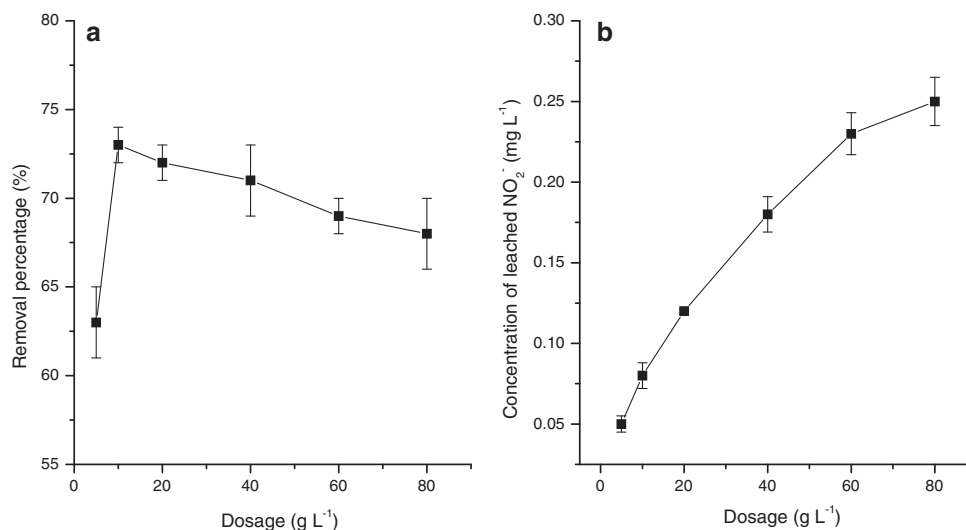
accounted for more than 20%. High levels were found for Ca, Al and Fe, whose contents exceeded 20 000 mg kg<sup>-1</sup>. Besides, the contents of Na, K and Mg were above 5000 mg L<sup>-1</sup>. As MSWI bottom ash was a high temperature product under the oxygen-enriched circumstance, CaO, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> might largely exist in it, which might be transformed into Ca(OH)<sub>2</sub>, Al(OH)<sub>3</sub> and Fe(OH)<sub>3</sub> after the water quench. Moreover, due to the complicate composition of MSWI bottom ash, the presence of some secondary minerals, such as the aluminosilicate and calcium aluminum compounds, was expected.

To further explore the mineral composition, the XRD pattern of the bottom ash was investigated (Fig. 1). The result confirmed the existence of quartz (SiO<sub>2</sub>), goethite (FeO(OH)) and aluminum oxide hydrate ((Al<sub>2</sub>O<sub>3</sub>)<sub>5</sub>H<sub>2</sub>O). Among the minerals, goethite and aluminum oxide hydrate were reported to have a certain adsorption capacity for various pollutants (Wei et al., 2011; Yao et al., 2012). Besides, kaolinite (Al<sub>2</sub>(Si<sub>2</sub>O<sub>5</sub>)(OH)<sub>4</sub>) and Friedel's salt (Ca<sub>2</sub>Al(OH)<sub>6</sub>(H<sub>2</sub>O)<sub>2</sub>) were also observed. Kaolinite is a 1:1 phyllosilicate comprised of neutral layers containing an octahedral sheet of aluminum hydroxide bound to a tetrahedral sheet of silicon oxide through a plane of shared O atoms. It had a great adsorption capacity for anions, such as NO<sub>3</sub><sup>-</sup> (Tribe et al., 2012). Friedel's salt is a layer double hydroxide consisting of cationic brucite layers and interlayer anion (Cl<sup>-</sup>). The interlayer anion (Cl<sup>-</sup>) could exchange with other anions, resulting in the uptake of anions in the solution (Zhang et al., 2011). The presence of the minerals such as the goethite, aluminum oxide hydrate, Kaolinite and Friedel's salt suggested that MSWI bottom ash might have the adsorption capacity for NO<sub>2</sub><sup>-</sup>, which could alter the migration of NO<sub>2</sub><sup>-</sup> in the landfill.

#### 3.2. Leaching and retention behavior of nitrite

##### 3.2.1. Effect of the dosage

The effect of the dosage of MSWI bottom ash on the NO<sub>2</sub><sup>-</sup> retention from aqueous solution is shown in Fig. 2(a). The nitrite removal percentage increased from 63.1% to 73.0% when the dosage increased from 0.5 to 1.0 g. However, the further increase of the dosage did not improve the removal efficiency. The removal percentage decreased to 68.0% though the dosage increased to 8.0 g. This result was distinct from the researches on other adsorbents (Öztürk and Bektaş, 2004; Islam and Patel, 2011), which usually showed a consistent upward tendency with the increase of the dosage. On the one hand, the overlapping of active



**Fig. 2.** Effect of the dosage of MSWI bottom ash on the retention (a) and leaching (b) behavior of nitrite.

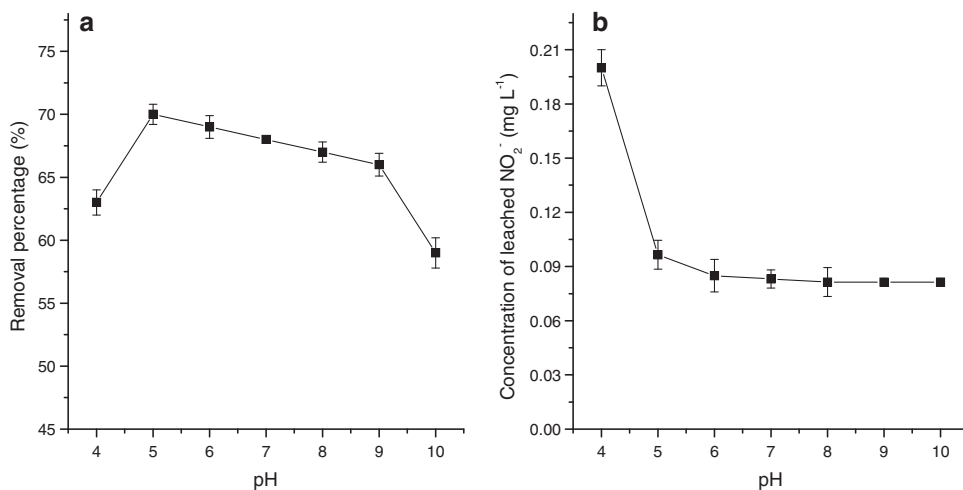


Fig. 3. Effect of pH on the retention (a) and leaching behavior (b) of nitrite.

sites at a higher dosage could limit the removal percentage. On the other hand, and most importantly, the higher dosage could enhance the leaching of NO<sub>2</sub><sup>-</sup> from MSWI bottom ash, as shown in Fig. 2(b). The leaching of NO<sub>2</sub><sup>-</sup> increased consistently with the increase of the dosage. The leached NO<sub>2</sub><sup>-</sup> was 0.05 mg L<sup>-1</sup> when the dosage was 0.5 g, and it came to 0.25 mg L<sup>-1</sup> when the dosage was 8.0 g.

The result indicated that MSWI bottom ash could retain the nitrite when it was disposed or reused in the landfill, which would change the migration of the nitrite. The retention was affected by the mass proportion of the bottom ash. A too high mass proportion of MSWI bottom ash disposed or reused in the landfill might mitigate the retention, owing to the increase of the nitrite leaching from MSWI bottom ash.

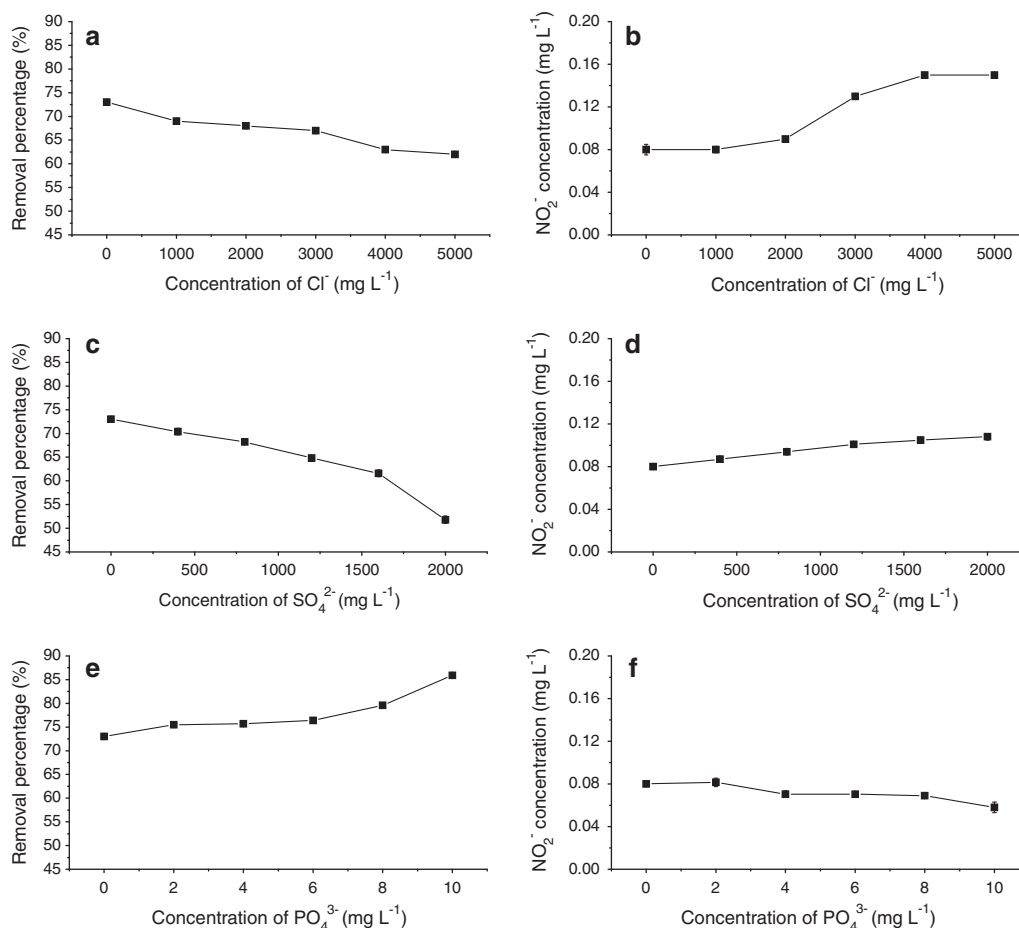


Fig. 4. Effect of Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and PO<sub>4</sub><sup>3-</sup> on the retention and leaching behavior of nitrite.

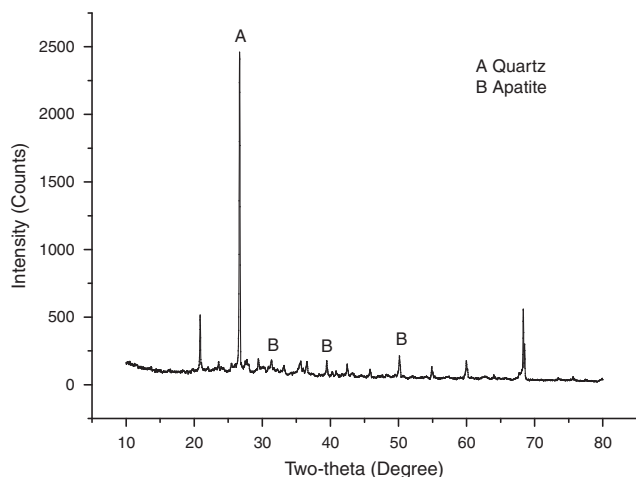


Fig. 5. XRD pattern of the MSWI bottom ash after imposed in  $\text{PO}_4^{3-}$  solution.

### 3.2.2. Effect of pH

The nitrite removal percentage showed first an increase and then a decrease with the increase of the pH. It increased from 63.0% to 70.1% when the pH increased from 4.0 to 5.0 (Fig. 3(a)). Then, the percentage removal consistently declined to 59.1% with the increase of the pH from 5.0 to 10.0. Several researches have pointed out that the low pH was beneficial to the adsorption of  $\text{NO}_2^-$ , as a result of the competition of  $\text{OH}^-$  with  $\text{NO}_2^-$  at the high pH condition (Islam and Patel, 2011; Katal et al., 2012). The result obtained at the pH 5.0–10.0 was basically consistent with the previous findings. However, the removal percentage increased when the pH increased from 4.0 to 5.0, which might result from the leaching of  $\text{NO}_2^-$  from MSWI bottom ash (Fig. 3(b)). The leached  $\text{NO}_2^-$  sharply decreased from 0.201 to 0.097  $\text{mg L}^{-1}$  when the pH increased from 4.0 to 5.0. The descent of the leaching could reduce the  $\text{NO}_2^-$  concentration in the leachate, resulting in the rise of the seeming removal percentage. However, only a slight decrease of  $\text{NO}_2^-$  leaching was observed when the pH further increased to 10.0. As the adsorption was restricted by the increase of the pH, the removal percentage decreased.

The result showed that the effect of pH on the nitrite migration had two sides. The optimum removal was observed when the pH was 5.0. In some cases of landfill studies, the pH of the leachate could be below 5.0 at the acidogenic stage. After the acidogenic

stage, the pH of the leachate would rise, which might reach 9.0 at the mature stage (Long et al., 2010). The retention of nitrite by MSWI bottom ash could be enhanced when the leachate pH increased to 5.0 at the acidogenic stage. However, the further increase of the leachate pH could restrict the retention.

### 3.2.3. Effect of anions

Landfill leachate contained abundant anions which can compete with  $\text{NO}_2^-$  in the adsorption process. In this study,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{PO}_4^{3-}$  were chosen to represent the single valence anion, dianion and trivalent anion, respectively. The nitrite removal percentage showed a downward trend with the increase of  $\text{Cl}^-$  concentration (Fig. 4(a)), which might be partly due to the competitive adsorption of  $\text{Cl}^-$  with  $\text{NO}_2^-$ . On the other hand, the increasing of  $\text{Cl}^-$  could facilitate the leaching of  $\text{NO}_2^-$  from MSWI bottom ash, which might be attributed to the ion-exchange of  $\text{Cl}^-$  with  $\text{NO}_2^-$  in the bottom ash.

Similar results were found for  $\text{SO}_4^{2-}$ . The nitrite removal percentage decreased from 73.0% to 51.8% when the  $\text{SO}_4^{2-}$  concentration increased to 2000  $\text{mg L}^{-1}$ . This result was expected. Firstly, the existence of  $\text{SO}_4^{2-}$  could compete with  $\text{NO}_2^-$  for the adsorption sites. Besides,  $\text{SO}_4^{2-}$  could interact with  $\text{Ca}^{2+}$  to form  $\text{CaSO}_4$  (Eq. (1)), which was beneficial to the releasing of  $\text{NO}_2^-$  as some  $\text{NO}_2^-$  was bound with  $\text{Ca}^{2+}$  in MSWI bottom ash. This was proved by the leaching behavior of  $\text{NO}_2^-$  affected by the presence of  $\text{SO}_4^{2-}$ , which showed a slight rise with the increase of  $\text{SO}_4^{2-}$  (Fig. 4(d)). Furthermore, the formation of  $\text{CaSO}_4$  might occupy the active adsorption sites in MSWI bottom ash, which could further decrease the removal percentage. These might be the possible reasons for the steeper drop observed in  $\text{SO}_4^{2-}$  compared with that of  $\text{Cl}^-$ .



Different from the results of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$ , the removal percentage of nitrite increased with the rise of the  $\text{PO}_4^{3-}$  concentration (Fig. 4(e)). The removal percentage increased consistently from 73.0% to 85.9% when the concentration of  $\text{PO}_4^{3-}$  increased from 0 to 10  $\text{mg L}^{-1}$ . Accordingly, the leaching experiment showed the reduction because of the existence of  $\text{PO}_4^{3-}$  (Fig. 4(f)). These results showed that  $\text{PO}_4^{3-}$  could enhance the retention ability of MSWI bottom ash for the nitrite. It might be due to the formation of the apatite, as shown in Fig. 5. Apatite has been proved to be capable of adsorbing various ions (Gao et al., 2009). The adsorption of nitrite on the apatite resulted in the increase of the removal percentage.

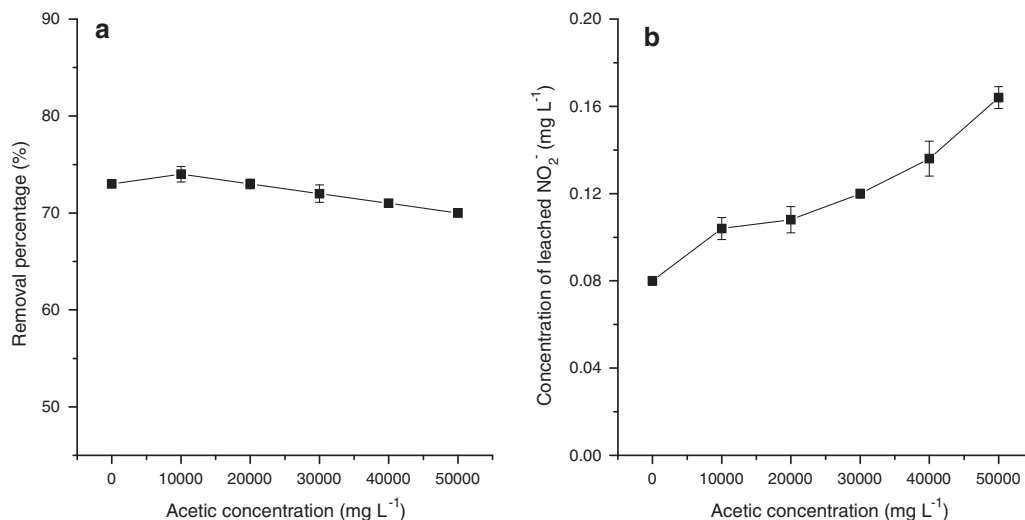


Fig. 6. Effect of acetic acid on the retention (a) and leaching (b) behavior of nitrite.

According to the profile of the anions in the landfill leachate, the highest concentrations of anions were always observed at the acidogenic stage, owing to the low pH condition (Figs. S3 and S4). The high level of anions could affect the retention and leaching behavior of nitrite by MSWI bottom ash. Since different species of anions have different impacts on the retention and leaching behavior of nitrite, further research is needed to figure out the comprehensive results.

### 3.2.4. Effect of organic acid

The presence of organic acid could result in the descent of pH and the increase of the organic anions. The effect of the descent of pH has been shown in Section 3.2.2. It could increase the leaching of nitrite and enhance the retention. The increase of organic anions could compete for the adsorption sites with  $\text{NO}_2^-$ , which was disadvantageous for the retention. The result suggested that the increase of the acetic acid could increase the leaching behavior of the nitrite from MSWI bottom ash (Fig. 6(b)), which was basically in agreement with the result of Section 3.2.2. However, the removal percentage of  $\text{NO}_2^-$  showed a slight decrease due to the increase of the acetic acid (Fig. 6(a)). According to the pH study, the removal percentage of the nitrite was expected to demonstrate an increasing trend, as the pH would fall with the increase of the acetic acid. The decrease of the removal percentage was probably due to the competition of the adsorption sites by the organic anions.

The organic acid in the practical landfill leachate usually experienced a rise and decent all along the running, with the maximum concentration observed at the acidogenic stage. In respect of the organic acid, the retention of nitrite by MSWI bottom ash might be mitigated at the acidogenic stage and promoted at the methanogenesis stage and mature stage.

## 4. Conclusions

The disposal or reuse of MSWI bottom ash in the landfill could affect the migration of nitrite. MSWI bottom ash had the retention capacity for nitrite because of the adsorptive substances, such as kaolinite, Friedel's salt and goethite. However, the leaching of nitrite from MSWI bottom ash was also observed. The retention and leaching of nitrite was influenced by the dosage of MSWI bottom ash, as well as the pH, inorganic anions and organic matter in the leachate. The optimum dosage of MSWI bottom ash for the retention of nitrite was  $10 \text{ mg L}^{-1}$  in  $2 \text{ mg L}^{-1}$  nitrite solution. Further increase of the dosage could enhance the leaching of  $\text{NO}_2^-$ , resulting in the decrease of the seeming removal percentage. The highest removal percentage of nitrite was observed when the pH was 5.0. The decrease of the pH could enhance the leaching of  $\text{NO}_2^-$  while the rise of the pH could increase the competitiveness of  $\text{OH}^-$ . The existence of  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$  and organic matter had disadvantageous effects on nitrite retention. However, the presence of  $\text{PO}_4^{3-}$  could enhance the nitrite retention, due to the formation of apatite. As the leachate's characteristics vary with the running of the landfill, it is assumed that the retention and leaching of nitrite by MSWI bottom ash will be accordingly changed at different stages of the landfill. Further research is required to figure out the effect of the MSWI bottom ash on the migration of the nitrite in practical landfill sites.

## Acknowledgements

This work was financially supported by Natural Science Foundation of Zhejiang province with Grant No. LQ13B070001 and Open Foundation of Zhejiang Provincial Key Laboratory of Solid Waste Treatment and Recycling with Grant No. SWTR-2012-06.

## Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.chemosphere.2014.06.057>.

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