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RESEARCH ARTICLE

Migration of nitrate, nitrite, and ammonia through the municipal solid waste incinerator bottom ash layer in the simulated landfill

Jun Yao[1,2] · Luxi Chen[1] · Huayue Zhu[1] · Dongsheng Shen[3] · Zhanhong Qiu[1]

Abstract Simulated landfill was operated for 508 days to investigate the effect of municipal solid waste incinerator (MSWI) bottom ash layer on the migration of nitrate, nitrite, and ammonia when it was used as the intermediate layer in the landfill. The result suggested that the MSWI bottom ash layer could capture the nitrate, nitrite, and ammonia from the leachate. The adsorption of the nitrate, nitrite, and ammonia on the MSWI bottom ash layer was saturated at the days 396, 34, and 97, respectively. Afterwards, the nitrogen species were desorbed from the MSWI bottom ash layer. Finally, the adsorption and desorption could reach the equilibrium. The amounts of adsorbed nitrate and nitrite on the MSWI bottom ash layer were 1685.09 and 7.48 mg, respectively, and the amount of the adsorbed and transformed ammonia was 13,773.19 mg, which was much higher than the desorbed. The water leaching test and synthetic precipitation leaching procedure (SPLP) results showed that the leachable nitrate, nitrite, and ammonia in the MSWI bottom ash were greatly increased after the landfill operation, suggesting that the adsorbed nitrogen could be finally leached out. Besides, the results also showed that MSWI bottom ash layer could affect the release of nitrate and ammonia at the initial stage of the landfill. However, it had little effect on the release of nitrite.

Keywords MSWI bottom ash layer · Nitrate · Nitrite · Ammonia · Migration · Release

✉ Zhanhong Qiu
13958559929@163.com

1   Center of Marine and Ecological Environment Protection, Taizhou University, Linhai 317000, China

2   Department of Environmental Engineering, Zhejiang University, Hangzhou 310029, China

3   Zhejiang Provincial Key Laboratory of Solid Waste Treatment and Recycling, Zhejiang Gongshang University, Hangzhou 310018, China

Introduction

Incineration has become more and more important for the treatment of municipal solid waste (MSW), due to the shortage of the urban land (Wan et al. 2006; Yao et al. 2010a). For instance, in the past 10 years, the number of municipal solid waste incinerator (MSWI) facility increased sharply from 47 to 166 in China, with the treatment capacity increased from 15,000 to 158,488 t day^{-1} (National Bureau of Statistics of China 2015). However, incineration is not a final waste treatment. Large volumes of solid residues, such as the bottom ash, fly ash, and air pollution control residue, are generated during the incineration process. Among the residues, MSWI bottom ash is the main waste stream, which accounts for approximately 80% (Chimenos et al. 1999). MSWI bottom ash is allowed to be disposed in the MSW landfill sites in several countries and areas, such as China, Japan, and Taiwan (Inanc et al. 2007; Lo and Liao 2007; Zhao et al. 2002). It is often used as the liner, intermediate layer, and leachate drainage layer instead of natural minerals in the landfill (Lo 2000; Travar et al. 2009; Su et al. 2013).

MSWI bottom ash is a mineral assemblage, which contains various adsorption media, such as aluminum (hydr)-oxides, iron (hydr)-oxides, and layered double hydroxides (Yao et al. 2010b, 2012, 2014b). Our previous study showed that MSWI bottom ash had considerable adsorption capacity for

the nitrate, nitrite, and ammonia (Yao et al. 2015a, b). The adsorption capacity of MSWI bottom ash for the nitrate, nitrite, and ammonia could be up to 7.5, 0.15, and 156.2 mg/g, respectively. Therefore, it is assumed that the nitrate, nitrite, and ammonia can be adsorbed by the MSWI bottom ash when it is used as liner or protection layer in the landfill, which may affect the migration and release of nitrogen pollutants. Due to the lack of degradation pathway in the anaerobic system, the nitrogen pollution turns out to be a long-term problem in the landfill, which has been the research focus in the last decade (He et al. 2006, 2007; Shalini and Joseph 2012; Wang et al. 2013). Several researches have been done to reveal the environmental impact of the MSWI bottom ash layer on the landfill. For instance, Lo (2005) and Lo and Liao (2007) suggested that MSWI bottom ash could enhance the metal release from the landfill. Inanc et al. (2007) and Su et al. (2013) argued that the MSWI bottom ash could not increase the metal leaching notably. Our previous research showed that MSWI bottom ash could increase the metal content of the refuse (Yao et al. 2014a). These results mainly concerned about the metal pollution, while few studies have been done concerning the effect of the MSWI bottom ash layer on the nitrogen pollution in the landfill. To figure out the comprehensive effect of MSWI bottom ash layer on the nitrogen pollution and to verify our assumption, it is necessary to investigate the effect of MSWI bottom ash layer on the migration of nitrate, nitrite, and ammonia in the landfill. Unfortunately, to our knowledge up to the date, few studies have been done on this scheme.

In this work, a leachate recirculated landfill bioreactor was established and operated for 508 days. MSWI bottom ash was disposed as the interlayer of the landfill. The variation of the nitrate, nitrite, and ammonia concentrations in the leachate, which was sampled above the MSWI bottom ash layer (L1), beneath the MSWI bottom ash layer (L2), and at the bottom of the landfill (L3), was monitored. Besides, the water leaching test and synthetic precipitation leaching procedure (SPLP) were carried out on the MSWI bottom ash samples to reveal the long-term behavior of the nitrogen species adsorbed on the MSWI bottom ash. We aimed to provide insights into the effect of MSWI bottom ash layer on the release of inorganic nitrogen pollutants when it was used as the intermediate layer in the landfill.

Materials and methods

Experimental setup

A simulated landfill bioreactor with a leachate recirculation system was set up. The schematic diagram of the experimental setup is shown in Fig. 1. The reactor was 287 mm in diameter and 1430 mm in height, with a total working volume of 92 L. Five ports were equipped for the reactor: the two ports at the top were used for gas exporting and leachate recirculation; the two ports at the side were used for sampling the leachate above (L1) and beneath (L2) the MSWI bottom ash layer; the port at the bottom was used for leachate drainage and sampling (L3). A 100-mm-thick layer of gravel was placed at the bottom of the landfill to simulate a leachate collection system and to prevent clogging of the leachate withdrawal outlets. The MSWI bottom ash layer was placed between the MSW layers. Another 50-mm-thick layer of sand was placed at the top of each landfill to simulate intermediate cover and top drainage layer.
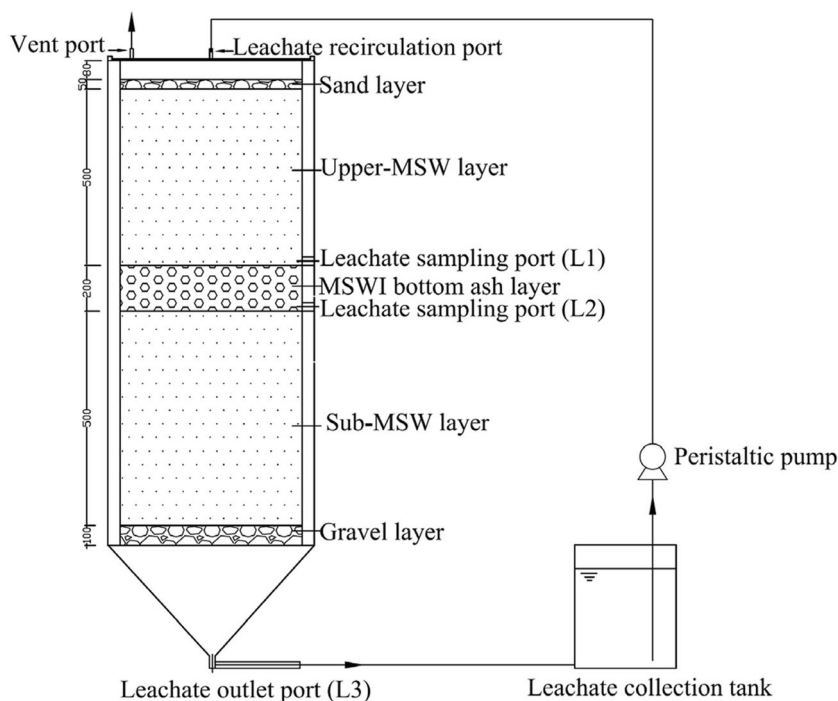
MSWI bottom ash and MSW

MSWI bottom ash was sampled from the Green Energy MSWI Plant in Zhejiang Province, East China. The plant consists of two parallel stoker incinerators with an MSW treatment capacity of 650 t day^{-1}. The MSWI bottom ash samples had undergone water quenching and magnetic separation before being sampled. The main characteristics of the MSWI bottom ash are shown in Table 1.

The MSW used in this work was collected from the Jiazhi Transport Station of Taizhou, Zhejiang, East China. MSW was sampled in the morning, afternoon, and night on the same day. Then, the large particles of the refuse were shredded into approximately 20 mm in diameter. The shredded refuse was homogenized by a shovel as thoroughly as possible before it was loaded to the landfill reactor. The moisture content of the refuse was 59.6%. The components of the MSW are exhibited in Table 2.

Operation of the reactor

Firstly, about 25 kg MSW was loaded and compacted using the shovel and sledgehammer. Then, 16 kg MSWI bottom ash was loaded and compacted. Finally, another 25 kg MSW was loaded and compacted. The mass proportion of MSWI bottom ash to MSW chose in this study was close to the production ratio of MSWI bottom ash to MSW in Zhejiang Province, China. The densities of MSW and MSWI bottom ash in the landfill were 0.78 and 1.28 t (m^3)$^{-1}$, respectively. The moisture content of the MSW was adjusted to 75% by adding tap water, which was reported to be an initial rapid decomposition threshold for the anaerobic organic refuse mineralization in bioreactor landfill (Benson et al. 2007; Lay et al. 1998). After the loading, the reactor was sealed with a gasket and silicone sealant. The leachate generated from the landfill reactor was collected in the leachate collection tank and continuously recirculated using pumps with the flow rates ranging from 2.2 to 2.9 mL min^{-1}, which was adjusted according to the leachate volume. To keep the volume equilibrium of leachate, the consumed leachate during the analytical process was replenished by the same volume of tap water (~20 mL) every time.

Fig. 1 Schematic of the simulated landfill systems

SPLP and water leaching test

MSWI bottom ash sample was sampled from the MSWI bottom ash layer before and after the landfill operation. SPLP (US Environmental Protection Agency 1996) and water leaching test were carried out on the MSWI bottom ash samples. In the SPLP, 5 g sample was added into 100 mL acid solution with a pH of 4.20 (adjusted with HNO_3 and H_2SO_4 with a volume ratio of 4:6). In the water leaching test, 5 g sample was added into 100 mL deionized water. Solutions were shaken for 20 h at 25 °C. After being shaken, the solutions were filtered through a 0.45 μm membrane filter. The resulting solutions were analyzed for nitrate, nitrite, and

ammonia concentrations. The SPLP and water leaching test were carried out in triplicate.

Analytical procedure

The corrected leachate samples (L1, L2, and L3) were analyzed for pH; chemical oxygen demand (COD); and concentrations of nitrate, nitrite, and ammonia. All these analyses were performed in accordance with standard methods. The pH was determined by GB 6920-86 (Ministry of Environmental Protection of the People's Republic of China 1986). The COD was determined by GB 11914-89 (Ministry of Environmental Protection of the People's Republic of China 1989). The nitrate was determined by GB 7480-87 (Ministry of Environmental Protection of the People's Republic of China 1987). The nitrite was determined by GB 7493-87 (Ministry of Environmental Protection of the People's Republic of China 1987). The ammonia was determined by GB 7479-87 (Ministry of Environmental Protection of the People's Republic of China 1987). All the analyses were performed in triplicate.

Result and discussion

Effect of MSWI bottom ash layer on the variation of COD and pH in the leachate

MSWI bottom ash layer has high acid neutralization capacity (ANC) and abundant adsorption medium. The physico-chemical properties of the leachate could be changed after

Table 1 Main characteristic of MSWI bottom ash

Moisture content (%)	16.6
Bulk density (kg/m³)	1278
LOI (%)	2.23
pH	11.2
Acid neutralization capacity at titration point of 7.5 (ANC$_{7.5}$)	1.0 mmol H$^+$ g^{-1}
Element	Content (mg kg^{-1} dry wt)
Al	40,920
Si	223,600
Na	9040
K	15,792
Mg	5997
Ca	69,413
Fe	26,008

Table 2 Components of the experimental MSW

Components	Food waste	Plastic	Paper	Textile	Dust	Ceramic	Metal	Timber	Residue
w/w (%)	44.3	8.2	7.5	0.3	6.2	5.1	0.1	1.7	27.6

the vertical flowing through the MSWI bottom ash layer. As shown in Fig. 2a, the COD level of L2 was lower than that of L1 in the first 378 days. It indicated that large amounts of organic matter were intercepted by the MSWI bottom ash layer. On the one hand, the alkaline minerals in the MSWI bottom ash could react with the organic acid in the leachate, which could remove parts of the acidic organic matter. On the other hand, the adsorption medium, such as the hydroxide minerals (e.g., aluminum (hydr)-oxides, iron (hydr)-oxides) and layered double hydroxides, could adsorb the organic matter (Wei et al. 2011). After day 378, the pH of L1 and L3 rose to above 7.0 (Fig. 2b), suggesting that the vast majority of the organic acid had been consumed. Thus, the reaction of the organic acid with the alkaline minerals was weakened. In addition, the adsorptive sites might have been mostly occupied by the various pollutants in the leachate. These reasons resulted in the relatively close COD level between the L1 and L2 after day 378. Although large amounts of organic matter were intercepted by the MSWI bottom ash layer, the COD of L3 was recovered when the leachate flowed through the sub-MSW layer. It suggested

that the MSWI bottom ash layer might not change the final concentration of COD in the leachate.

The pH of L2 was higher than those of L1 and L3 all through the study, indicating that the MSWI bottom ash layer could increase the pH of the leachate. The ANC of MSWI bottom ash was up to 1.0 mmol H^+ g^{-1} (Table 1). The high ANC of MSWI bottom ash notably promoted the pH of the leachate. Especially, the pH of L2 ranged from 8.5 to 11.9 in the first 44 days, which was far higher than that of L1 (ranged from 4.4 to 6.2). As the distribution pattern of ammonia (ammonia and ammonium ion) greatly depended on the pH, the promotion of the leachate pH might change the distribution pattern of the ammonia in the landfill. After day 44, the gap between pH values of L1 and L2 started to narrow down, as the ANC of MSWI bottom ash was gradually consumed. It should be noted that no significant difference was observed between the pH values of L1 and L3. This result suggested that although the leachate pH was increased after flowing through the MSWI bottom ash layer, it was converted back after flowing through the sub-MSW layer.
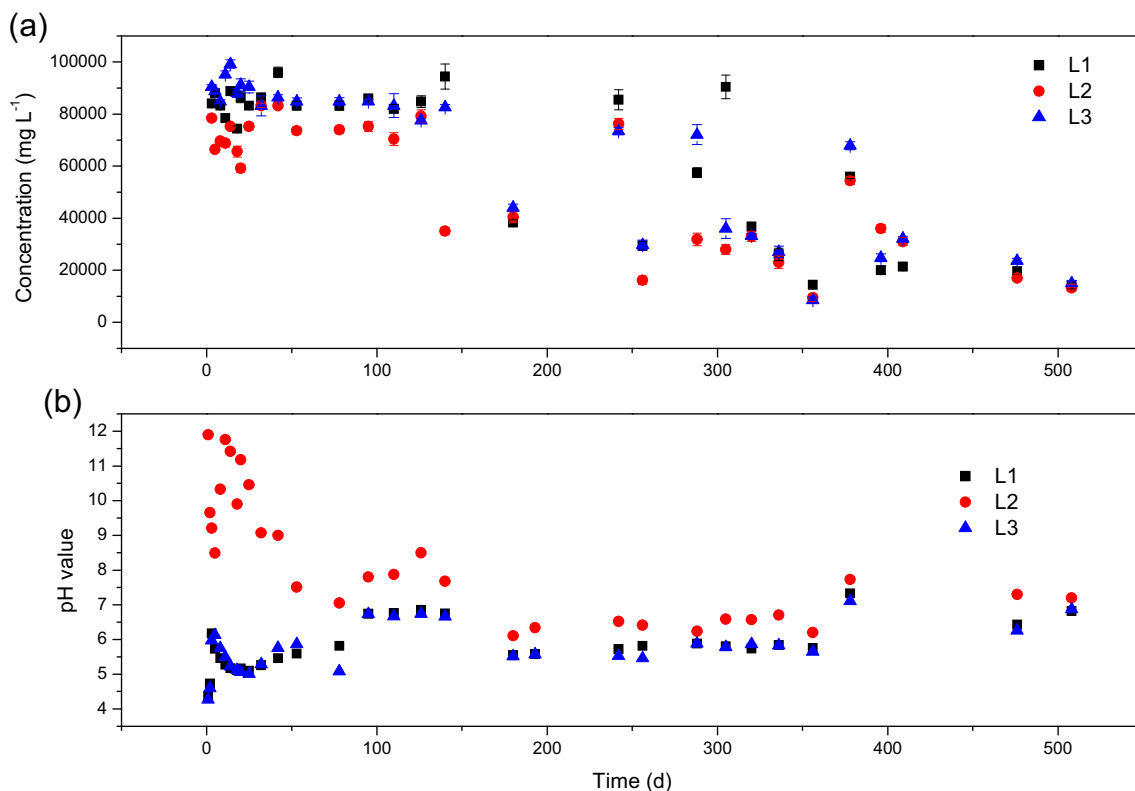
Fig. 2 Variation of COD (**a**) and pH (**b**) of L1, L2, and L3

Effect of MSWI bottom ash layer on the migration of nitrate, nitrite, and ammonia

Nitrate

Generally, the nitrate concentrations of L1, L2, and L3 experienced a rise and a decrease during the study. The initial concentration of nitrate was contributed by the leaching of the soluble nitrate in the MSW. Then, the refuse was degraded by the residual oxygen in the landfill and released certain amounts of nitrate, resulting in the increase of the nitrate concentration. Afterwards, the landfill formed an anaerobic condition. The nitrate was reduced by the denitrification, leading to the decline of the nitrate level.

Except day 9 and day 288, the nitrate concentration of L2 was significantly lower than that of L1 in the first 396 days ($P < 0.05$), which ranged from 86.5 to 253.0 mg L^{-1} and from 94.1 to 287.0 mg L^{-1}, respectively (Table S1). The lower nitrate concentration of L2 compared with that of L1 indicated that the MSWI bottom ash layer could capture the nitrate from the leachate, which could be due to the adsorption of the nitrate on MSWI bottom ash. According to the leachate volume and the nitrate concentration differences between L1 and L2, it was calculated that about 1669.5 mg of nitrate was captured by the MSWI bottom ash layer in the first 396 days. It should be noted that the amount of the retained nitrate showed a decreasing trend with the time extension in the first 396 days (Fig. S1), as the adsorption capacity of MSWI bottom ash was gradually consumed. After day 396, the nitrate concentration of L1 became higher than that of L2, suggesting that MSWI bottom ash was unable to further adsorb the nitrate. Instead, some nitrate was released from the MSWI bottom ash. It might be due to the fact that the adsorption of nitrate on MSWI bottom ash had reached the saturation after 396 days' operation. Moreover, the nitrate concentration in the leachate decreased with time passing by. To keep the equilibrium of distribution between the MSWI bottom ash and the leachate, the nitrate was desorbed from the MSWI bottom ash. In the first 80 days, the nitrate concentration of L3 was generally lower than that of L1, which could be attributed to the adsorption of nitrate on the MSWI bottom ash layer. However, as the adsorption receded, no significant difference was found between the average nitrate concentrations of L1 (123.1 mg L^{-1}) and L3 (125.9 mg L^{-1}) ($P < 0.05$). The previous results indicated that the MSWI bottom ash layer could affect the release of nitrate from the landfill at the initial stage (Fig. 3).

Nitrite

The variation of nitrite concentrations of L1, L2, and L3 is exhibited in Fig. 4. The nitrite concentrations of L1 and L3 showed a downward trend. At the initial stage, the soluble nitrite was leached from the MSW, resulting in the relatively high concentration of nitrite. Then, the landfill gradually became anaerobic, leading to the denitrification of nitrite.

According to the comparison of nitrite concentrations of L1 and L2, the running of the landfill could be divided into three stages. At stage I (day 1 to day 34), the nitrite concentration in L2 was significantly lower than that of L1 ($P < 0.05$, Table S2), indicating that the nitrite was captured by the MSWI bottom ash layer. Our previous research showed that MSWI bottom ash had certain adsorption capacity for nitrite (Yao et al. 2015a). The adsorption of nitrite on the MSWI bottom ash decreased the nitrite concentration of the leachate. It should be noted that the difference of nitrite concentrations in L2 and L1 showed a declining trend with the extension of time in the first 34 days (Fig. S2), suggesting that the adsorption of nitrite on MSWI bottom ash gradually reached the saturation. At stage II (day 44 to 256), except days 44, 97, and 256, the nitrite concentration of L2 was significantly higher than that of L1 ($P < 0.05$), indicating that the nitrite was desorbed from the MSWI bottom ash. At this stage, the nitrite concentration of L1 decreased to a low level. As the adsorption was saturated at stage I, the nitrite was desorbed from the MSWI bottom ash to keep the adsorption equilibrium between the leachate and MSWI bottom ash. At stage III (days 288 to 508), the average nitrite concentrations of L1 and L2 were close. It meant that the equilibrium between adsorption and desorption of nitrite on MSWI bottom ash had been established. Since the beginning of stage II, the nitrite concentration of L3 was lower than those of L1 and L2. It seemed that although the desorption of nitrite increased the nitrite concentration of L2, it was transformed due to the relatively rigorous anaerobic condition of the sub-MSW layer. In consideration of the relatively low concentration of nitrite compared with those of nitrate and ammonia, it is believed that the MSWI bottom ash layer might not affect the final release of the nitrite.

Ammonia

As the landfill gradually became anaerobic, the ammonia concentration in the leachate generally showed an increasing trend (Fig. 5), which was corresponding with the decreasing of the nitrate and nitrite concentrations in the leachate.

Similar to the situation of nitrite, the landfill could also be divided into three stages according to the comparison of ammonia concentrations of L1 and L2. At stage I (from days 1 to 97), the ammonia concentration of L2 was significantly lower than that of L1 ($P < 0.05$, Table S3). Firstly, the MSWI bottom ash had a great adsorption capacity for the ammonia, which could capture the ammonia from the leachate. Secondly, the MSWI bottom ash layer significantly increased the pH of leachate, which facilitated the transformation of ammonia ion to ammonia (g). Thirdly, ammonia ion could react with the Mg in the MSWI bottom ash, forming the immobile

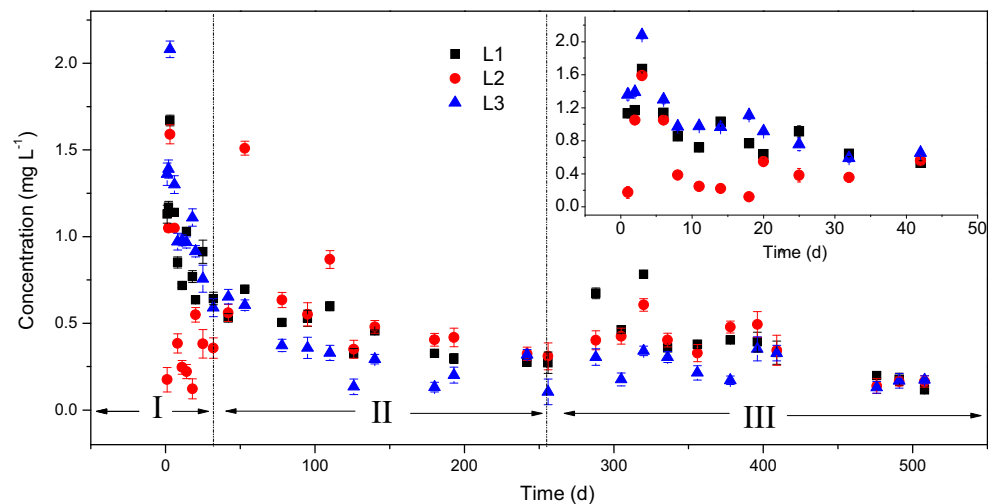**Fig. 3** Variation of nitrate concentration of L1, L2, and L3
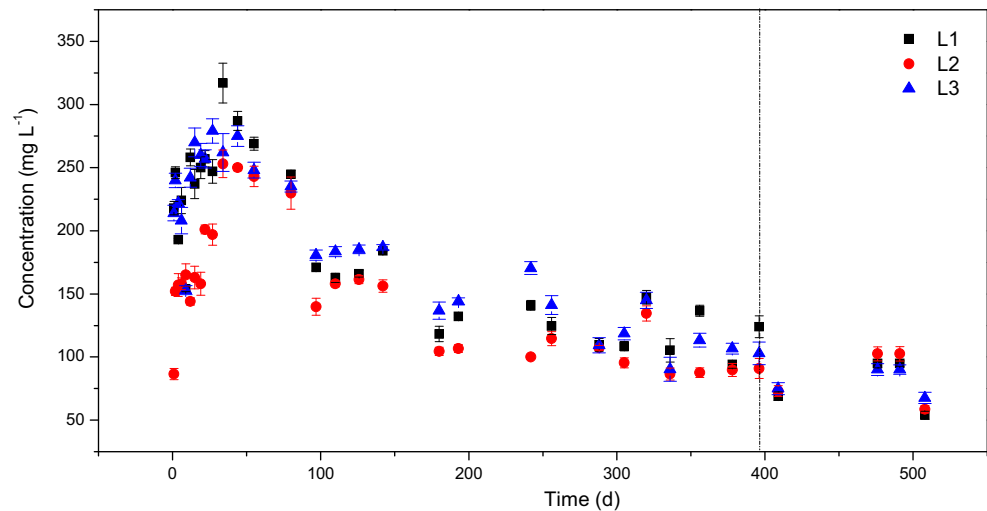
precipitates. However, the difference of ammonia concentrations of L1 and L2 reduced with the extension of time, due to the saturation of the adsorption, reaction, and the decreasing pH of L2. From day 110 to day 288 (stage II), except day 142, the ammonia concentration of L2 was significantly higher than that of L1 ($P < 0.05$), indicating that the ammonia was desorbed from the MSWI bottom ash. After day 288 (stage III), the average ammonia concentrations of L1 and L2 were close, which were 1173.9 and 1148.1 mg L^{-1}, respectively. It suggested that the equilibrium between adsorption and desorption of ammonia on MSWI bottom ash had been established. In the first 78 days, the ammonia concentration of L3 was generally lower than that of L1, which might be due to the adsorption and transformation of ammonia on MSWI bottom ash layer. However, the adsorption and transformation receded over time, and the anaerobic condition of the bottom MSW layer was completely established. The ammonia concentration of L3 gradually exceeded that of L1. These results showed that MSWI bottom ash layer could affect the release of ammonia from the landfill at the initial stage. The ammonia

was reported to have inhibitory effect on the degradation of MSW (Poggi-Varaldo et al. 1997). The adsorption and transformation of the ammonia in the MSWI bottom ash layer could fasten the stabilization process of the landfill, which had been reported in the previous research (Li et al. 2014).

The previous results showed that the influences of the MSWI bottom ash layer on the nitrate, nitrite, and ammonia followed the similar pattern. Firstly, the MSWI bottom ash showed certain adsorption capacity for the nitrate, nitrite, and ammonia, which could capture the nitrogen species in the leachate. When the adsorption was gradually saturated and concentrations of nitrogen species in the leachate decreased, the nitrate, nitrite, and ammonia could be desorbed from the MSWI bottom ash layer. Finally, the equilibrium was established between the adsorption and desorption, and the effect of the MSWI bottom ash layer on the nitrogen migration became not obvious. However, the adsorption capacity of MSWI bottom ash for the nitrate, nitrite, and ammonia was different. The concentrations of nitrate, nitrite, and ammonia in the leachates were also varied. Thus, the process of the

Fig. 4 Variation of nitrite concentration of L1, L2, and L3

**Fig. 5** Variation of ammonia concentration of L1, L2, and L3
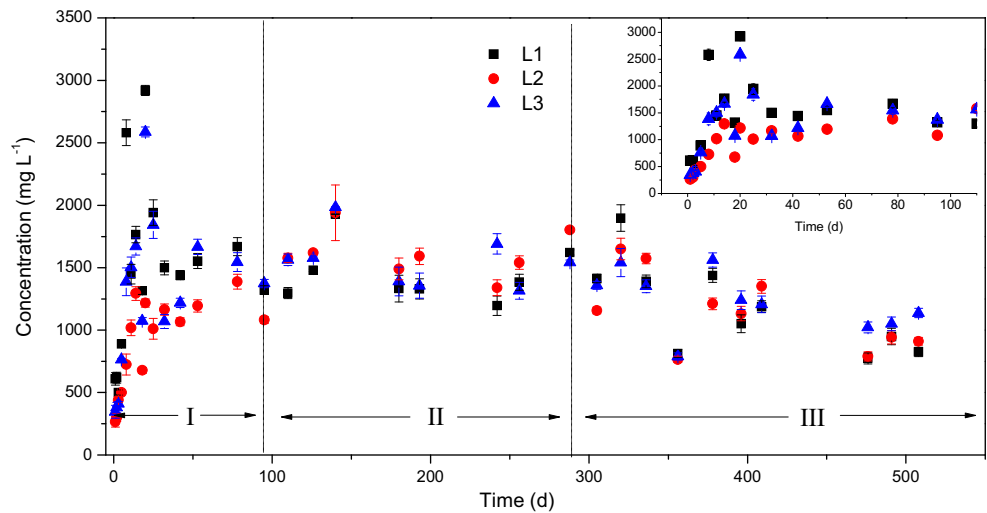
adsorption, desorption, and establishment of the equilibrium was different among the three nitrogen species. For example, the saturation adsorption time was 396 days for nitrate, while it was 34 days for nitrite and 97 days for ammonia. The similar pattern was also observed when the total inorganic nitrogen was concerned (Fig. S3). In the first 142 days, the total inorganic nitrogen concentration of L1 is higher than that of L2, suggesting that the nitrogen was captured by the MSWI bottom ash layer. From day 142 to day 288, the total inorganic nitrogen concentration of L1 is lower than that of L2, indicating that the nitrogen was released from the MSWI bottom ash layer. After day 288, the average concentration of the inorganic nitrogen became close. It should be noted that although the

migration of the nitrogen species was affected, the distribution pattern of the inorganic nitrogen in the leachate was not influenced by the MSWI bottom ash layer (Fig. S4). It seemed that the MSWI bottom ash layer had not changed the redox state of the landfill.

Leaching behavior of nitrate, nitrite, and ammonia from MSWI bottom ash samples

The water leaching test and SPLP were carried out on the MSWI bottom ash samples before and after the landfill operation. After the study, the MSWI bottom ash lost 12.8% of the total mass, which was probably due to the

Fig. 6 Leaching of nitrate, nitrite, and ammonia from MSWI bottom ash samples in water leaching test (**a**) and SPLP (**b**)
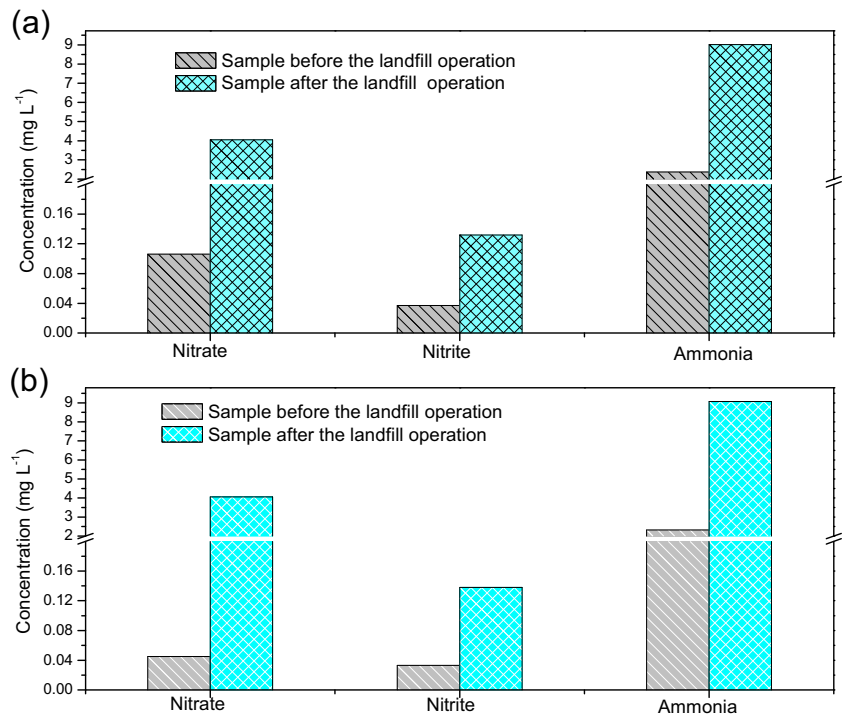
release of minerals during the landfill operation. The leaching of the nitrogen species was at the same level in the water leaching test (Fig. 6a) and SPLP (Fig. 6b). It suggested the acid addition in SPLP could not increase the nitrogen leaching from the MSWI bottom ash sample, although it was beneficial for the dissolution of the minerals. This result meant that the leached nitrogen was released from the surface of the MSWI bottom ash sample, which was probably adsorbed during the landfill operation. The leaching of nitrate, nitrite, and ammonia was greatly enhanced after the landfill operation, both in the water leaching test and in the SPLP. This result was consistent with our expectation. According to the calculation, about 1685.09 mg of nitrate and 7.48 mg of nitrite were adsorbed on the MSWI bottom ash layer. About 13,773.19 mg of ammonia was adsorbed or transformed in the MSWI bottom ash layer. However, only 55.89 mg of nitrate, 2.97 mg of nitrite, and 3013.45 mg of ammonia were desorbed from the MSWI bottom ash layer (Tables S4, S5, and S6). According to the previous results, it is believed that the captured nitrate, nitrite, and ammonia by the MSWI bottom ash layer could be finally leached out if the rain fell on the landfill sites.

Conclusion

MSWI bottom ash layer could affect the migration of nitrate, nitrite, and ammonia in the landfill. The effects followed the similar pattern for the nitrogen species. The nitrate, nitrite, and ammonia in the leachate were firstly adsorbed by the MSWI bottom ash layer due to its great adsorption capacity, which could retard the migration of the nitrogen species with the leachate. After the adsorption of nitrate, nitrite, and ammonia was saturated at the days 396, 34, and 97, respectively, the desorption was observed. Finally, the equilibrium between the adsorption and desorption could be established. The amounts of nitrate, nitrite, and ammonia adsorbed were much higher than the desorbed, resulting in the accumulation of the nitrogen species in the MSWI bottom ash layer. These nitrogen species could be finally leached out if the rain fell on the landfill. Besides, the MSWI bottom ash layer could affect the release of nitrate and ammonia from the landfill at the initial stage. However, the release of nitrite might not be affected due to the low concentration and the rigorous anaerobic condition of the sub-MSW layer.

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