RESEARCH ARTICLE

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

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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

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 tubes. 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.

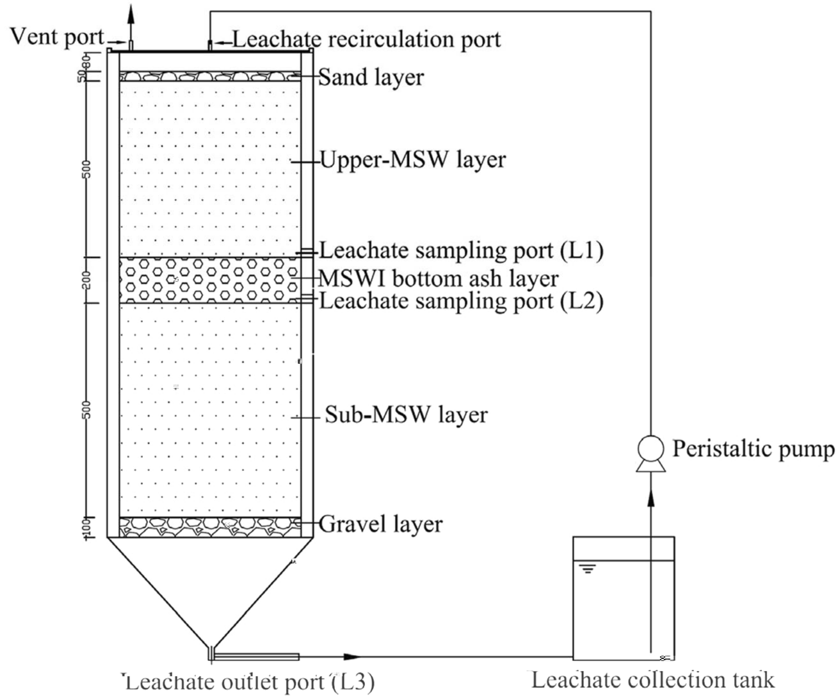


Fig. 1 Schematic of the simulated landfill systems

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 tests 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 tests were carried out in triplicate.

Table 1 Main characteristic of MSWI bottom ash

Moisture content (%)	16.6
Bulk density (kg/m^3)	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)
Al	40,920
Si	223,600
Na	9040
K	15,792
Mg	5997
Ca	69,413
Fe	26,008

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 physicochemical properties of the leachate could be changed after

Table 2 Componen s of he e perimen al MSW

Componen s	Food as e	Plas ic	Paper	Te ile	D s	Ceramic	Me al	Timber	Resid e
w/w (%)	44.3	8.2	7.5	0.3	6.2	5.1	0.1	1.7	27.6

he er ical flo ing hro gh he MSWI bo om ash la er. As sho n in Fig. 2a, he COD le el of L2 as lo er han ha of L1 in he firs 378 da s. I indica ed ha large amo n s of organic ma er ere in ercep ed b he MSWI bo om ash la er. On he one hand, he alkaline minerals in he MSWI bo om ash co ld reac i h he organic acid in he leacha e, hich co ld remo e par s of he acidic organic ma er. On he o her hand, he adsorp ion medi m, s ch as he h dro ide minerals (e.g., al min m (h dr)-o ides, iron (h dr)-o ides) and la ered do ble h dro ides, co ld adsorb he organic ma er (Wei e al. 2011). Af er da 378, he pH of L1 and L3 rose o abo e 7.0 (Fig. 2b), s gges ing ha he as majori of he organic acid had been cons med. Th s, he reac ion of he organic acid i h he alkaline minerals as eakened. In addi ion, he adsorp i e si es migh ha e been mos l occ pied b he ario s poll an s in he leacha e. These reasons res l ed in he rela i el close COD le el be een he L1 and L2 af er da 378. Al ho gh large amo n s of organic ma er ere in ercep ed b he MSWI bo om ash la er, he COD of L3 as reco ered hen he leacha e flo ed hro gh he s b-MSW la er. I s gges ed ha he MSWI bo om ash la er migh no change he final concen ra ion of COD in he leacha e.

The pH of L2 as higher han hose of L1 and L3 all hro gh he s d , indica ing ha he MSWI bo om ash la er co ld increase he pH of he leacha e. The ANC of MSWI bo om ash as p o 1.0 mmol H^+ g^{-1} (Table 1). The high ANC of MSWI bo om ash no abl promo ed he pH of he leacha e. Especiall , he pH of L2 ranged from 8.5 o 11.9 in he firs 44 da s, hich as far higher han ha of L1 (ranged from 4.4 o 6.2). As he dis rib ion pa ern of ammonia (ammonia and ammoni m ion) grea l depended on he pH, he promo ion of he leacha e pH migh change he dis rib ion pa ern of he ammonia in he landfill. Af er da 44, he gap be een pH al es of L1 and L2 s ar ed o narro do n, as he ANC of MSWI bo om ash as grad all cons med. I sho ld be no ed ha no significan difference as obser ed be een he pH al es of L1 and L3. This res l s gges ed ha al ho gh he leacha e pH as increased af er flo ing hro gh he MSWI bo om ash la er, i as con er ed back af er flo ing hro gh he s b-MSW la er.

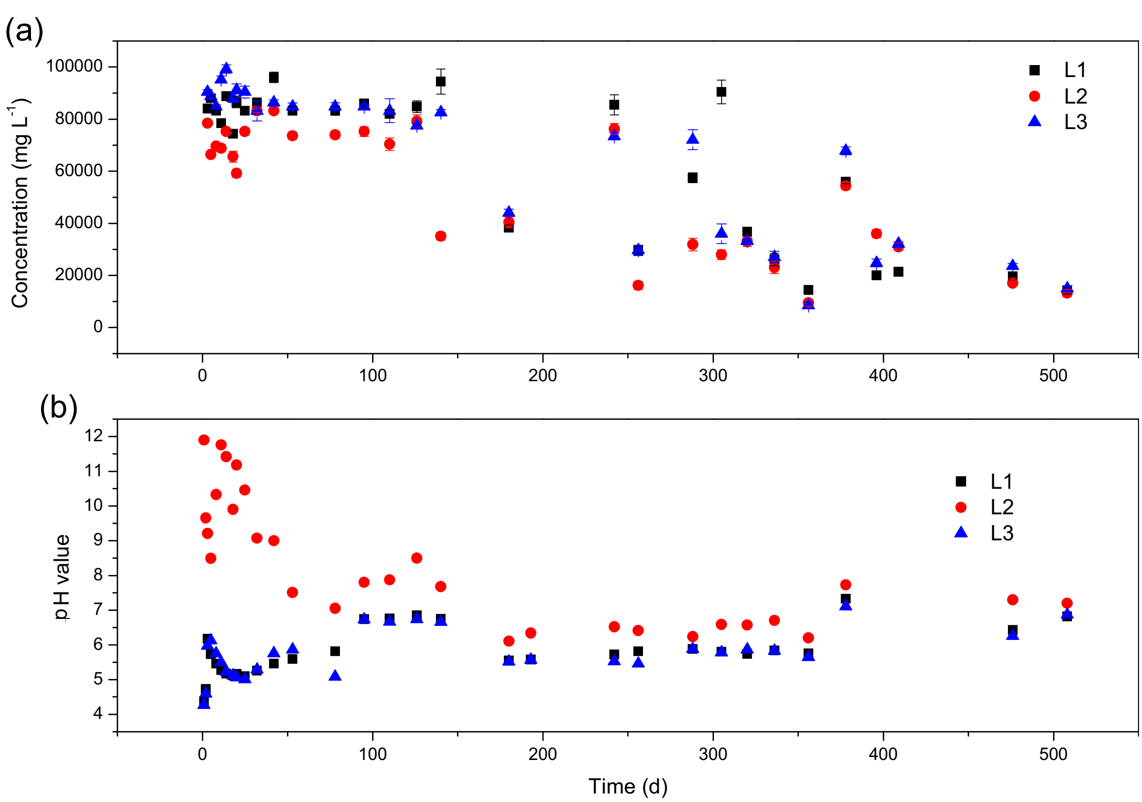


Fig. 2 Varia ion of COD (**a**) and pH (**b**) of L1, L2, and L3

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

Nitrate

Generall , he ni ra e concen ra ions of L1, L2, and L3 e perienced a rise and a decrease d ring he s d . The ini ial concen ra ion of ni ra e as con rib ed b he leaching of he sol ble ni ra e in he MSW. Then, he ref se as degraded b he resid al o gen in he landfill and released cer ain amo n s of ni ra e, res l ing in he increase of he ni ra e concen ra ion. Af er ards, he landfill formed an anaerobic condi ion. The ni ra e as red ced b he deni rifica ion, leading o he decline of he ni ra e le el.

E cep da 9 and da 288, he ni ra e concen ra ion of L2 as significan l lo er han ha of L1 in he firs 396 da s ($P < 0.05$), hich ranged from 86.5 o 253.0 mg L^{-1} and from 94.1 o 287.0 mg L^{-1}, respec i el (Table S1). The lo er ni ra e concen ra ion of L2 compared i h ha of L1 indica ed ha he MSWI bo om ash la er co ld cap re he ni ra e from he leacha e, hich co ld be d e o he adsorp ion of he ni ra e on MSWI bo om ash. According o he leacha e olme and he ni ra e concen ra ion differences be een L1 and L2, i as calc la ed ha abo 1669.5 mg of ni ra e as cap red b he MSWI bo om ash la er in he firs 396 da s. I sho ld be no ed ha he amo n of he re ained ni ra e sho ed a decreasing rend i h he ime e ension in he firs 396 da s (Fig. S1), as he adsorp ion capaci of MSWI bo om ash as grad all cons med. Af er da 396, he ni ra e concen ra ion of L1 became higher han ha of L2, s gges ing ha MSWI bo om ash as nable o f r her adsorb he nira e. Ins ead, some ni ra e as released from he MSWI bo om ash. I migh be d e o he fac ha he adsorp ion of ni ra e on MSWI bo om ash had reached he sa ra ion af er 396 da s' opera ion. Moreo er, he ni ra e concen ra ion in he leacha e decreased i h ime passing b . To keep he eq ilibri m of dis rib ion be een he MSWI bo om ash and he leacha e, he ni ra e as desorbed from he MSWI bo om ash. In he firs 80 da s, he ni ra e concen ra ion of L3 as generall lo er han ha of L1, hich co ld be a rib ed o he adsorp ion of ni ra e on he MSWI bo om ash la er. Ho e er, as he adsorp ion receded, no significan difference as fo nd be een he a erage ni ra e concen ra ions of L1 (123.1 mg L^{-1}) and L3 (125.9 mg L^{-1}) ($P < 0.05$). The preio s res l s indica ed ha he MSWI bo om ash la er co ld affec he release of ni ra e from he landfill a he ini ial s age (Fig. 3).

Nitrite

The aria ion of ni ri e concen ra ions of L1, L2, and L3 is e hibi ed in Fig. 4. The ni ri e concen ra ions of L1 and L3 sho ed a do n ard rend. A he ini ial s age, he sol ble ni ri e as leached from he MSW, res l ing in he rela i el high concen ra ion of ni ri e. Then, he landfill grad all became anaerobic, leading o he deni rifica ion of ni ri e.

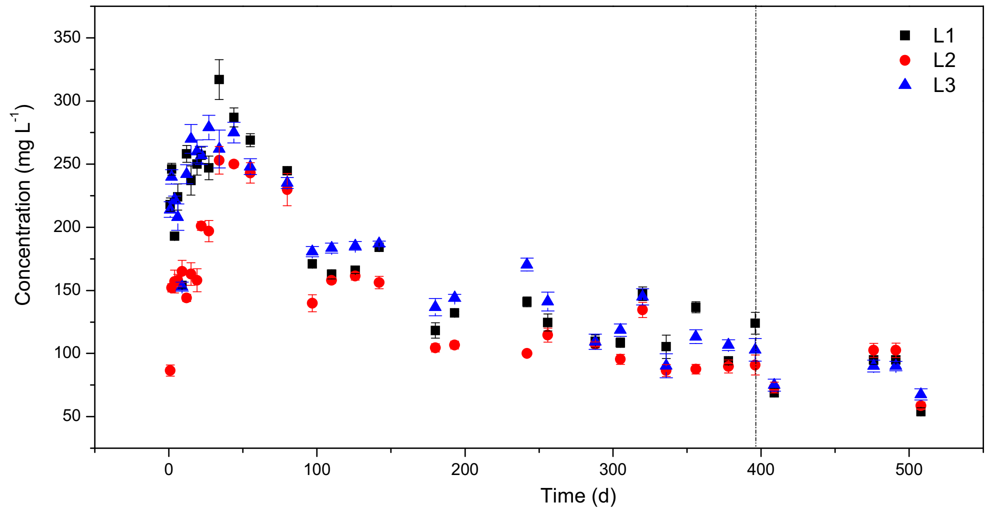
According o he comparison of ni ri e concen ra ions of L1 and L2, he r nning of he landfill co ld be di ided in o hree s ages. A s age I (da 1 o da 34), he ni ri e concen ra ion in L2 as significan l lo er han ha of L1 ($P < 0.05$, Table S2), indica ing ha he ni ri e as cap red b he MSWI bo om ash la er. O r pre io s research sho ed ha MSWI bo om ash had cer ain adsorp ion capaci for ni ri e (Yao e al. 2015a). The adsorp ion of ni ri e on he MSWI bo om ash decreased he ni ri e concen ra ion of he leacha e. I sho ld be no ed ha he difference of ni ri e concen ra ions in L2 and L1 sho ed a declining rend i h he e ension of ime in he firs 34 da s (Fig. S2), s gges ing ha he adsorpion of ni ri e on MSWI bo om ash grad all reached he sa ra ion. A s age II (da 44 o 256), e cep da s 44, 97, and 256, he ni ri e concen ra ion of L2 as significan l higher han ha of L1 ($P < 0.05$), indica ing ha he ni ri e as desorbed from he MSWI bo om ash. A his s age, he ni ri e concen ra ion of L1 decreased o a lo le el. As he adsorp ion as sa ra ed a s age I, he ni ri e as desorbed from he MSWI bo om ash o keep he adsorp ion eq ilibri m be een he leacha e and MSWI bo om ash. A s age III (da s 288 o 508), he a erage ni ri e concen ra ions of L1 and L2 ere close. I mean ha he eq ilibri m be een adsorp ion and desorp ion of ni ri e on MSWI bo om ash had been es ablished. Since he beginning of s age II, he ni ri e concenra ion of L3 as lo er han hose of L1 and L2. I seemed ha al ho gh he desorp ion of ni ri e increased he ni ri e concenra ion of L2, i as ransformed d e o he rela i el rigoro s anaerobic condi ion of he s b-MSW la er. In considera ion of he rela i el lo concen ra ion of ni ri e compared i h hose of ni ra e and ammonia, i is belie ed ha he MSWI bo om ash la er migh no affec he final release of he ni ri e.

Ammonia

As he landfill grad all became anaerobic, he ammonia concen ra ion in he leacha e generall sho ed an increasing rend (Fig. 5), hich as corresponding i h he decreasing of he ni ra e and ni ri e concen ra ions in he leacha e.

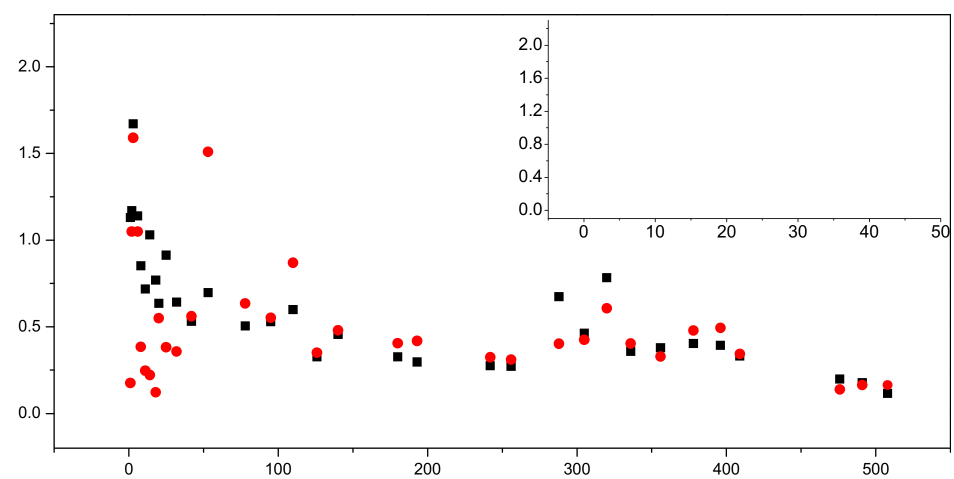
Similar o he si a ion of ni ri e, he landfill co ld also be di ided in o hree s ages according o he comparison of ammonia concen ra ions of L1 and L2. A s age I (from da s 1 o 97), he ammonia concen ra ion of L2 as significan l lo er han ha of L1 ($P < 0.05$, Table S3). Firs l , he MSWI bo om ash had a grea adsorp ion capaci for he ammonia, hich co ld cap re he ammonia from he leacha e. Secondl , he MSWI bo om ash la er significan l increased he pH of leacha e, hich facili a ed he ransforma ion of ammonia ion o ammonia (g). Thirdl , ammonia ion co ld reac i h he Mg in he MSWI bo om ash, forming he immobile

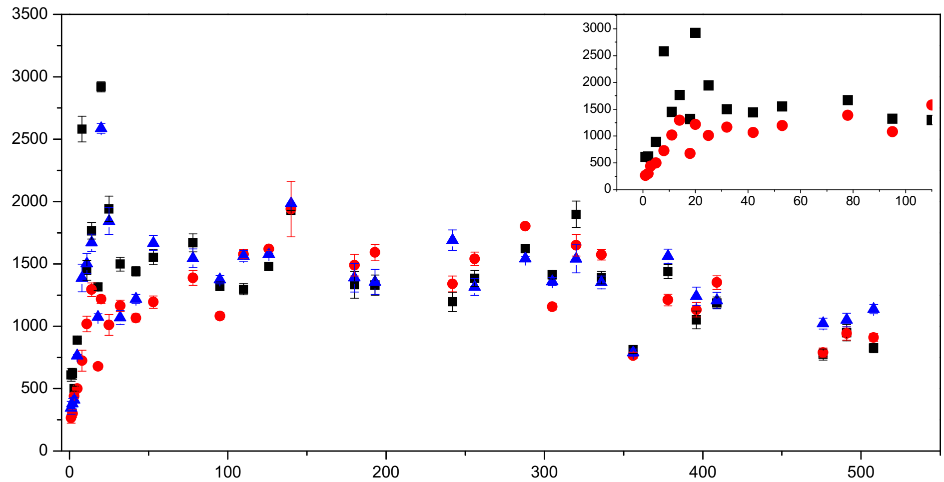
Fig. 3 Varia ion of ni ra e concen ra ion of L1, L2, and L3



precipi a es. Ho e er, he difference of ammonia concen ra ions of L1 and L2 red ced i h he e ension of ime, d e o he sa ra ion of he adsorp ion, reac ion, and he decreasing pH of L2. From da 110 o da 288 (s age II), e cep da 142, he ammonia concen ra ion of L2 as significan l higher han ha of L1 ($P < 0.05$), indica ing ha he ammonia as desorbed from he MSWI bo om ash. Af er da 288 (s age III), he a erage ammonia concen ra ions of L1 and L2 ere close, hich ere 1173.9 and 1148.1 mg L^{-1}, respec i el . I s gges ed ha he eq ilibri m be een adsorp ion and desorp ion of ammonia on MSWI bo om ash had been es ablished. In he firs 78 da s, he ammonia concen ra ion of L3 as generall lo er han ha of L1, hich migh be d e o he adsorp ion and ransforma ion of ammonia on MSWI bo om ash la er. Ho e er, he adsorp ion and ransforma ion receded o er ime, and he anaerobic condi ion of he bo om MSW la er as comple el es ablished. The ammonia concen ra ion of L3 grad all e ceeded ha of L1. These res l s sho ed ha MSWI bo om ash la er co ld affec he release of ammonia from he landfill a he ini ial s age. The ammonia as repor ed o ha e inhibi or effec on he degrada ion of MSW (Poggi-Varaldo e al. 1997). The adsorp ion and ransforma ion of he ammonia in he MSWI bo om ash la er co ld fas en he s abili a ion process of he landfill, hich had been repor ed in he pre io s research (Li e al. 2014).

The pre io s res l s sho ed ha he infl ences of he MSWI bo om ash la er on he ni ra e, ni ri e, and ammonia follo ed he similar pa ern. Firs l , he MSWI bo om ash sho ed cer ain adsorp ion capaci for he ni ra e, ni ri e, and ammonia, hich co ld cap re he ni rogen species in he leacha e. When he adsorp ion as grad all sa ra ed and concen ra ions of ni rogen species in he leacha e decreased, he ni ra e, ni ri e, and ammonia co ld be desorbed from he MSWI bo om ash la er. Finall , he eq ilibri m as es ablished be een he adsorp ion and desorp ion, and he effec of he MSWI bo om ash la er on he ni rogen migra ion became no ob io s. Ho e er, he adsorp ion capaci of MSWI bo om ash for he ni ra e, ni ri e, and ammonia as differen . The concen ra ions of ni ra e, ni ri e, and ammonia in he leacha es ere also aried. Th s, he process of he





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 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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