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Trimethylamine (TMA) biofiltration and transformation in biofilters

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Abstract

Bioremoval of trimethylamine (TMA) in two three-stage biofilters packed with compost (A) and sludge (B), respectively, was investigated. Both biofilters were operated with an influent TMA concentration of 19.2–57.2 mg m^{-3} for 67 days. Results showed that all of the inlet TMA could be removed by both biofilters. However, removal efficiency and transformation of TMA in each section of both biofilters was different. In the Introduction section, TMA removal efficiency and maximum elimination capacity of the compost medium were greater than those of sludge medium under higher inlet TMA concentration. In comparison with biofilter A, considerably higher NH_3 concentrations in effluent of all three sections in biofilter B were observed after day 19. Although, NO_2^--N concentration in each section of biofilter A was relatively lower, NO_3^--N content in each section of biofilter A increased after day 26, especially in the Materials and method section which increased remarkably due to a lesser amount of TMA and higher ammonia oxidation and nitrification in compost medium. In contrast, neither NO_2^--N nor NO_3^--N were detected in either section of biofilter B at any time throughout the course of the experiment. The cumulative results indicated that compost is more favorable for the growth of TMA-degrading and nitrifying bacteria as compared to the sludge and could be a highly suitable packing material for biodegradation and transformation of TMA.

Keywords: Biofiltration; Trimethylamine; Three-stage biofilter; Transformation

1. Introduction

Biofiltration is believed to be one of the most efficient technologies in terms of its technical and economical qualities, especially for the treatment of low-concentration polluted air streams. Under the proper conditions, high removal efficiencies can be achieved through biofiltration and the process is environmentally sound [1,2].

Trimethylamine (TMA) is a volatile organic compound (VOC) responsible for strong odor emission. It is often released from fish-meal manufacturing processes [3–5], wastewater treatment, waste disposal landfills, livestock farming, and hog manure [6–9]. Due to its potentially toxic and likely carcinogenic properties, TMA is considered as a strong environmental pollutant [10]. Moreover, TMA has also been reported to inhibit the synthesis of macromolecules such as DNA, RNA and proteins and to have a teratogenic effect on animal embryos [11]. Thus, microbial degradation would be useful for the purpose of eliminating TMA from contaminated environments [5].

Over the past decade, attention has been given to the treatment of TMA for the purpose of bioremediation, as this method was determined to be more economically feasible and environmentally friendly in comparison to other conventional means [9]. To the authors' knowledge, there is only one published study using an aerobic biofiltration system containing entrapped mixed microbial cells for removal of TMA-containing waste gases [9]. No report has been published on the bioconversion of TMA as a sole target in biofilters. Further, research concerning variations in physical and chemical properties of the packing materials in biofilters with respect to time or height is scarce, especially for the studies on treating TMA-containing waste gases.

The main objectives of this research were to (1) determine the TMA removal efficiency of two different packing materials, (2) investigate the elimination capacity of TMA in laboratory-scale biofilters, (3) detect ammonia emissions in outlets of both biofilters and (4) study the physical and chemical changes occurring at different heights of the reactor over time. Results from this research will be useful for understanding the mechanisms concerning the bioconversion of the target contaminant

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Nomenclature

C_0	influent concentration (mg m^{-3})
EC	elimination capacity (g m^{-3} h^{-1})
FID	flame ionization detector
L_m	mass loading rate (g m^{-3} h^{-1})
Q	gas flow rate (m^3 h^{-1})
RE	removal efficiency (%)
TKN	total Kjeldahl nitrogen (g kg^{-1})
VOC	volatile organic compound

Greek letter

τ	empty bed residence time (s)

and providing economical and effective alternatives for odor control.

2. Materials and methods

2.1. Biofilters and packing materials

The biofilter system consisted of a gas source (air pump and syringe pump), a humidifying apparatus, a mixing cell, a gas flow control unit (flow meters, valves, and pipes), an air treatment unit (parallel biofilters), and an effluent absorption apparatus (Fig. 1). Two identical bench-scale biofilters made of transparent rigid plexiglass with an inner diameter of 0.15 m and a height of 1.0 m were used for this experiment. The height of the filter bed was 0.45 m in each reactor and it was divided into three 0.15 m equal sections, leaving a 20 mm space between two sections for representative gas sampling. Sections were flanged and each section could be dismantled to replace and sample the packing material. TMA gas was made by injecting TMA-containing solution into the mixing bottle using a syringe pump and a gas-tight syringe. Inlet air from the air pump was humidified to prevent the biofilter matrix from drying and then flowed into the biofilters mixed with TMA-containing gas. The gas inlet flow was controlled by a rotameter. TMA concentration of the gas flow was kept constant by adjusting the injection rate of the solution and the flow rate of the air stream. The exhaust was absorbed by solution in gas washing bottles.

Two kinds of biofilter packing materials, compost and sludge, were used in this study due to their easy availability and inherently diverse microbial communities. The compost used was commercially produced from pig feces and rice straw under aerobic composting conditions. It was amended with 30% perlite (v/v) to increase the porosity of filter medium and decrease the pressure drop across the filter bed. In addition, digested dehydrated granular sludge with moisture content of approximately 30% obtained from the municipal wastewater treatment plant at Cixi (Zhejiang Province, China) was used in this study.

2.2. Inoculation

The packing materials were, respectively, pre-inoculated with 4 L of acclimated activated sludge suspension which was enriched with 40% activated sludge (Sibao wastewater treatment plant, Hangzhou, China), 60% nutrient medium with TMA as main carbon source. The suspension was prepared when TMA concentration and mixed liquid suspended solids increased to 0.1 and 4–5 g L^{-1}, respectively. The nutrient medium (g L^{-1}) contained soluble starch 0.268, beef extract 0.068, peptone 0.132, urea 0.008, $NaHCO_3$ 0.08, $MgSO_4$ 0.066, $CaCl_2$ 0.006, KH_2PO_4 0.0488, $(NH_4)_2SO_4$ 0.112, $FeSO_4$ 0.0003 and $MnSO_4$ 0.006.

2.3. Operation conditions

The biofilters were filled with inoculated compost and sludge, respectively. Both reactors were first acclimatized for 14 days under the influent TMA concentration (C_0) of 19.2 mg m^{-3}.

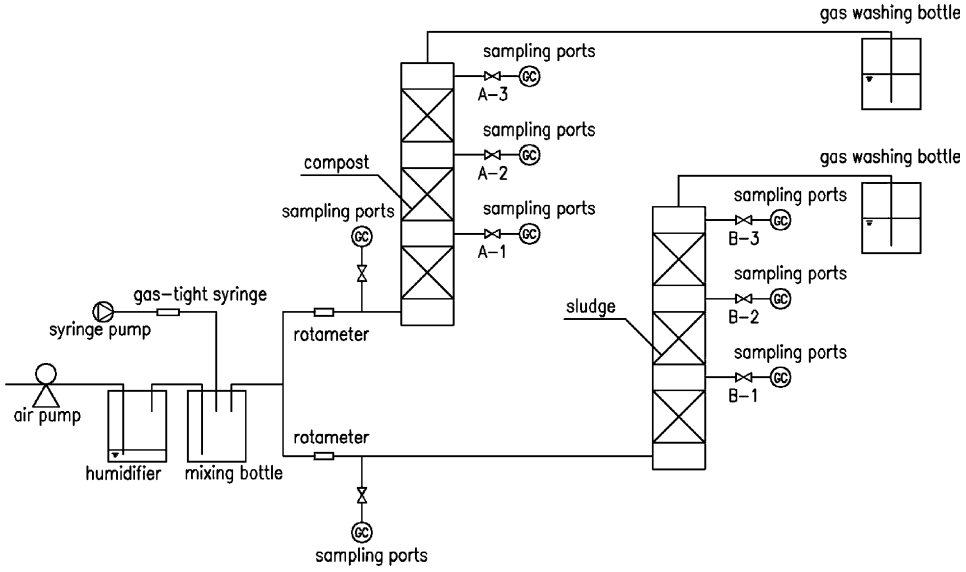

Fig. 1. Schematic experimental system.

Then, C_0 was gradually increased from 19.2 to 57.2 mg m^{-3}. According to the range of C_0, the mass loading rate of TMA (L_m) of Section 1 were 3.48–10.35 g TMA m^{-3} h^{-1}. The biofiltration of TMA was carried out over a period of 67 days at a gas flow rate (Q) of 0.48 m^3 h^{-1} and an empty bed residence time (τ) of 60 s. All reactors were operated at 25 ± 2 °C.

2.4. Sampling and analytical methods

After inoculation, the properties of the media such as porosity and bulk density were analyzed as described by Hirai et al. [12]. Average particle size was measured by sieving [13]. Volumetric specific area was analyzed by AUTOSORB-1-C gas adsorption apparatus (Quantachrome, USA) based on the physical adsorption of nitrogen on the solid surface, using the BET method [14]. About 0.1 mL TMA gas sample was periodically taken using a gas-tight syringe at the outlet of every section and analyzed by gas chromatography on a glass column with GDX-401 at 130 °C, coupled to a flame ionization detector (FID) with N_2 as carrier gas. Ammonia (NH_3) from the outlet of every section was absorbed in 25 mL of 0.005 mol L^{-1} H_2SO_4 solution by an atmosphere sampling instrument at 0.5 mL min^{-1} for 10 min. Then 10 mL of the absorbed solution was analyzed with ammonium-Nessler's reagent colorimetric method at 420 nm. Packing materials in each section of the biofilters were sampled five times during the entire experimental period with days 0, 13, 25, 40, 60 for biofilter A and days 0, 12, 24, 39, 59 for biofilter B, for the measurement of pH, moisture content, organic matter, total Kjeldahl nitrogen (TKN), ammonia-N (NH_4^+-N), nitrite-N (NO_2^--N) and nitrate-N (NO_3^--N). These physical and chemical characteristics of compost and sludge filter bed materials were analyzed as described by Chen et al. [13]. Five grams of sample (wet basis) was drawn from each section of the reactors and filtered after shaking the samples with 2 M KCl for 30 min to analyze NH_4^+-N and NO_2^--N. Another 10 g was drawn and extracted with $CuSO_4$–Ag_2SO_4 in a rotary shaker for 10 min to analyze NO_3^--N. All analyses were carried out in duplicate.

3. Results and discussion

3.1. Properties of medium

Initial characteristics of filter medium in both biofilters A and B are shown in Table 1. As described by Chang et al. [9], optimal pH for TMA treatment should be between 6.0 and 8.0, with a neutral pH being most favorable for maximum enzyme activity for TMA degradation. Initial pH of compost medium and sludge medium was 7.22 and 6.82, respectively, which was in the optimal range. Williams and Miller [15] point to bed moisture content as the single most important parameter for biofilter viability. Optimal moisture contents varied from 20% to 60% in their review of operational biofilters. Due to different absorbability, after inoculation the moisture of sludge medium is 35.5%, while compost medium is 88.9%. The former is in optimal range of moisture content but the latter is too high. Actually, 35.5% is the highest moisture content in such kind of sludge medium.

Table 1
Characteristics of the media in biofilters A and B

Characteristics	Biofilter A (compost)	Biofilter B (sludge)
pH	7.22	6.82
Moisture (w/w, %)	88.9	35.5
Bulk density (kg m^{-3})	121	457
Porosity (%)	39	53
Average particle size[a] (wt.%)		
>4 mm	37.6	62.9
2–4 mm	36.3	34.8
<2 mm	26.1	2.3
Specific surface area (m^2 g^{-1})	4.175	7.081
Organic matter (w/w, %)	15	36
TKN (g kg^{-1})	18.52	20.52
NH_4^+-N (g N kg^{-1} medium)	0.021	0.344
NO_2^--N (g N kg^{-1} medium)	0.0004	0.0004
NO_3^--N (g N kg^{-1} medium)	0.447	0.0034
Seed sludge (l)	4	4
Total volume (l)	8	8

[a] Obtained by sieving.

As shown in Table 1, compost medium and sludge medium had bulk densities of 121 and 457 kg m^{-3}, respectively, indicating that compost was more volumetric on a weight basis than sludge. Porosity of compost medium was lower than sludge medium probably because it was subject to bed compaction and had higher moisture content. In addition, the relationship between pressure drop and superficial gas velocity is a very important parameter in determining the operational cost. Data on the pressure drop for gas streams through both media are shown in Fig. 2. Under room temperature (25 °C), pressure drop of both biofilters increased with the increase of superficial gas velocity from 27.2 to 90.6 m h^{-1}. There is a good correlation between pressure drop and gas velocity with the coefficient determination of R^2 values larger than 0.97 in both biofilters. The pressure drop of biofilter A was always higher than that of biofilter B. It is mainly because compost had higher moisture content and lower porosity resulting in a higher gas resistance.

As shown in Table 1, sludge medium has a higher volumetric specific area than compost medium. Bardtke et al. [16] reported that media with a higher volumetric specific area were more

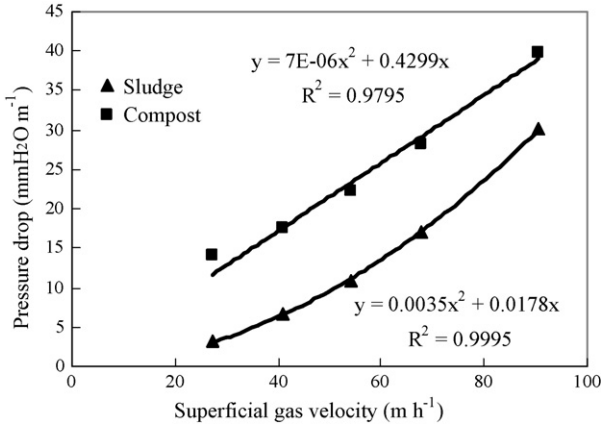

Fig. 2. Pressure drop of the experimental filter media.

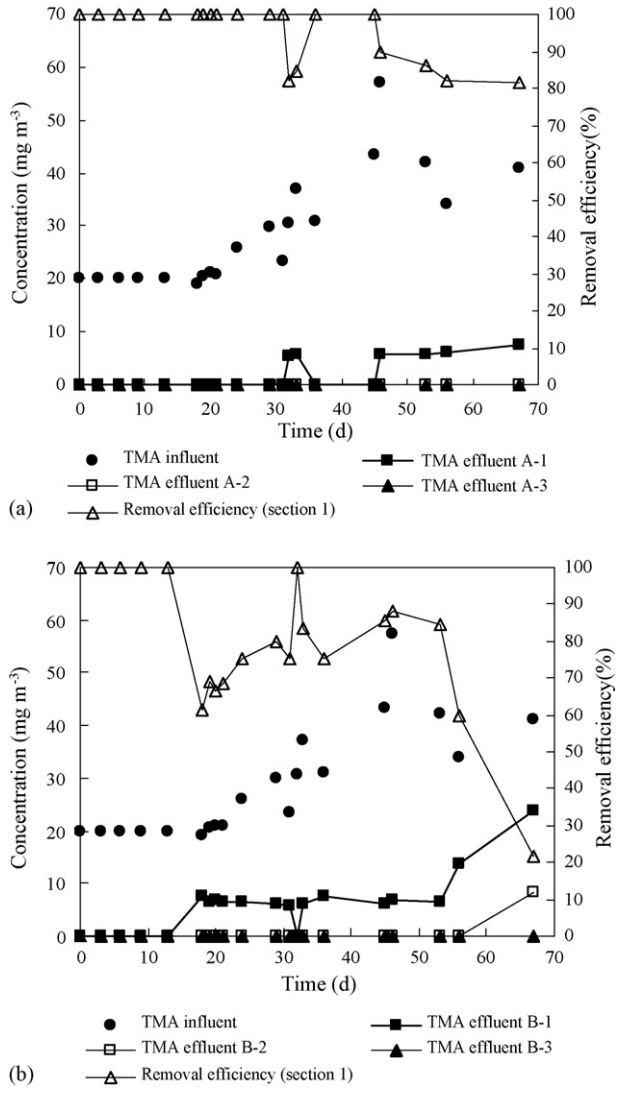

Fig. 3. TMA removal efficiency of Section 1, influent concentration and effluent concentrations at sampling ports of biofilters A (a) and B (b).

efficient than those with lower specific area in removing VOCs from gas streams. From this point of view, sludge in biofilter B was expected to achieve greater effectiveness than compost in biofilter A in terms of TMA removal. However, the result was not accordant as described in the following parts.

3.2. Temporal changes of TMA

TMA removal in biofilters A and B is shown in Fig. 3. C_0 were fluctuating in order to test the adaptability and stability of the systems. As shown in Fig. 3a, when C_0 was about 19.2 mg m^{-3} in the first 14 days, TMA concentrations in effluent A-1, A-2 and A-3 were below detection levels and the TMA removal efficiency (RE) was 100%. When C_0 increased gradually from 19.2 to 43.3 mg m^{-3}, TMA concentration in effluent A-1 was steady and RE in Section 1 maintained 100% except on days 32 and 33, indicating that TMA-degrading bacteria inoculated in the compost medium exhibited high activity. When C_0 increased from 43.3 to 57.2 mg m^{-3}, RE in Section 1 dropped again and never increased even if C_0 returned to a relatively lower level. It suggested that the high TMA concentration might pose a detriment to the TMA-degrading bacteria and the systemic performance of Section 1 in biofilter A. Nevertheless, it was noticed that a majority of TMA was removed in the first two sections and the RE in Section 1 remained above 80%. In contrast, a different performance of TMA removal was observed in biofilter B (Fig. 3b). Although, RE of TMA in Section 1 maintained 100% over the first 14 days, it decreased to 61% once the influent TMA concentration was increased. However, with the gradual increase of TMA concentration from 19.2 to 57.2 mg m^{-3}, RE of TMA in Section 1 increased to over 60% while TMA concentration in effluent B-1 remained at 7 mg m^{-3} until day 53. Afterwards, TMA concentration in effluent B-1 increased and RE in Section 1 of biofilter B decreased dramatically even though influent TMA concentration was lower than 57.2 mg m^{-3}. This indicates that Section 1 of biofilter B cannot treat TMA further at this level of C_0 and the sludge medium was not as favorable as compost medium for the growth of inoculated TMA-degrading bacteria under this condition probably due to the poor sorption of TMA and the reduced microbial activity as a consequence of decreasing moisture content in sludge medium (Fig. 4). TMA elimination capacities in Section 1 of biofilters A and B under different mass loading rates were determined and are shown in Fig. 5a and b, respectively. For biofilter A, TMA removal efficiency was over 80% with a mass loading rate less than 10.4 g TMA m^{-3} h^{-1} and ranged from 61% to 100% in biofilter B under the same loading rate up to 53 days. The maximum elimination capacities of Section 1 of biofilters A and B were 9.31 and 9.13 g TMA m^{-3} h^{-1}, respectively.

3.3. Ammonia emissions in biofilters

TMA is oxidized to dimethylamine by the action of the enzyme TMA dehydrogenase, then to monomethylamine by

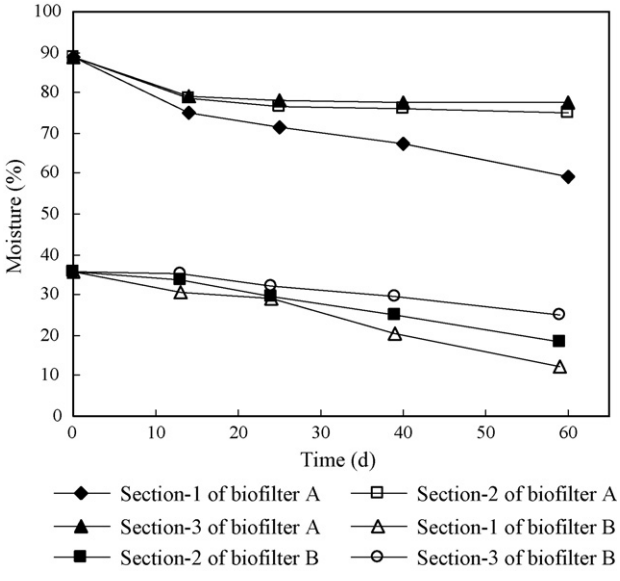

Fig. 4. Variations of moisture content in each section of biofilters A and B over time.

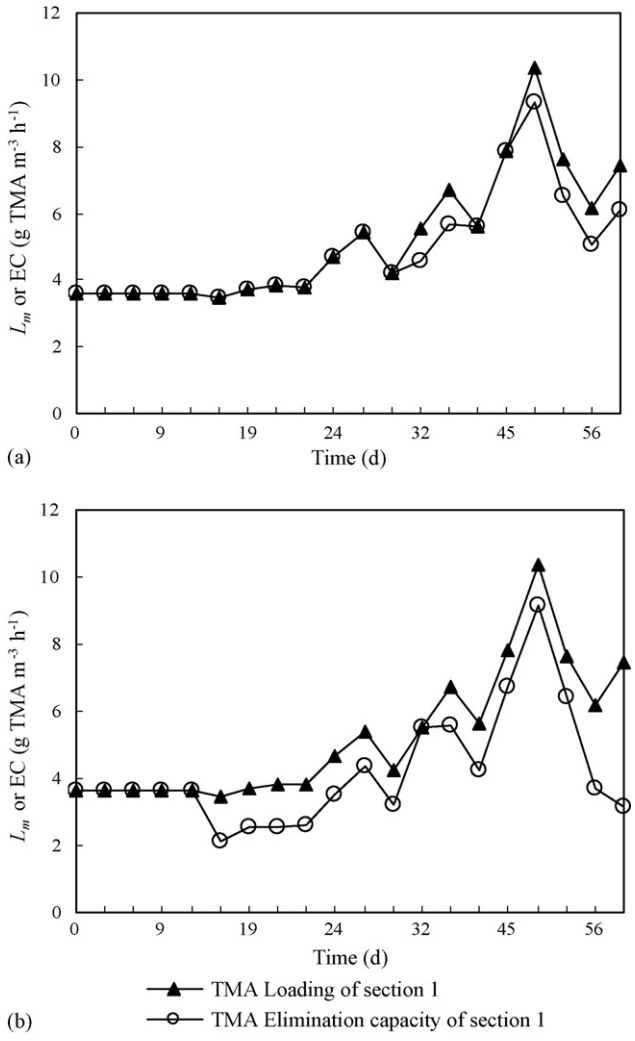

Fig. 5. TMA loading and elimination capacity of Section 1 of biofilters A (a) and B (b) during the experiment.

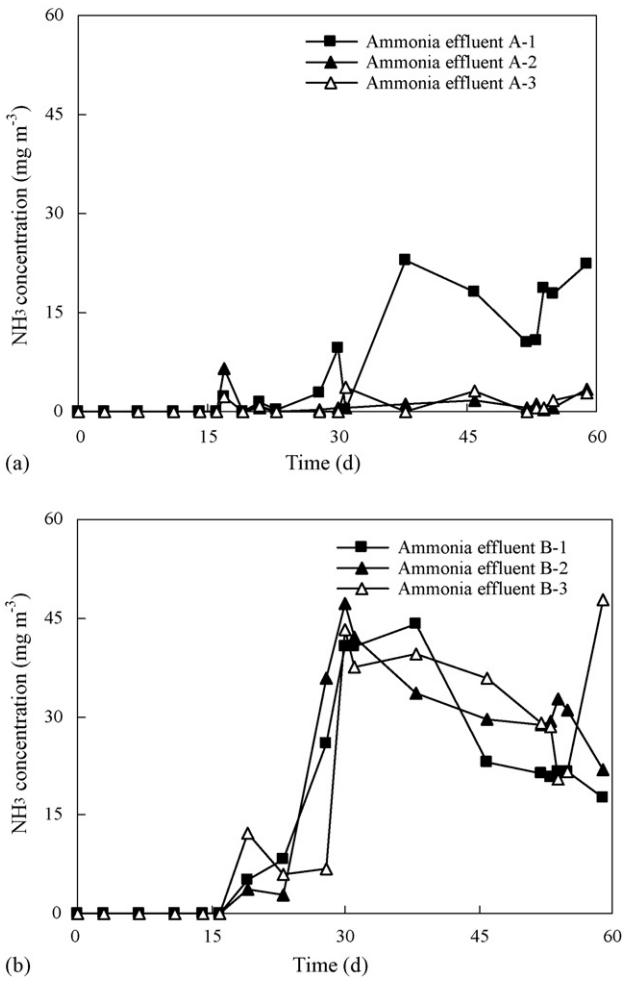

Fig. 6. Variation of ammonia concentration in effluent gas streams of biofilters A (a) and B (b) over time.

the activity of dimethylamine monoxygenase and finally to formaldehyde and ammonia by the action of the enzyme monomethylamine dehydrogenase before carbon uptake by the cell and incorporation into the serine pathway [10]. As one main product of biodegradation of TMA, ammonia (NH_3) in effluent gas streams of biofilters A and B is shown in Fig. 6a and b, respectively. No NH_3 was detected at any outlet of either biofilters during the initial period, i.e. before day 15, indicating complete elimination of TMA through sorption and absorption. However, on day 28 in biofilter A and on day 19 in biofilter B, an increasing concentration of NH_3 was observed in both outlets A-1 and B-1, which indicated that the absorbed TMA had been hydrolyzed into NH_3 by the microorganisms in the packing materials. Higher concentration of NH_3 was detected in the effluent A-1, but less in A-2 and A-3 (Fig. 6a). It can be estimated that biodegradation of TMA in biofilter A has occurred mainly in the first section. Ammonia generated in the first section of biofilter A was absorbed and partially nitrified in Section 2. Due to the relatively low NH_3 loading, nearly all NH_3 reaching Section 3 was eliminated. Consequently, 100% removal of TMA was observed in biofilter A over the duration of the operation period. In contrast, biofilter B behaved differently. Higher concentrations of NH_3 were detected at the outlet of biofilter B after day 19. These results showed that the packing material in biofilter B was quickly saturated by NH_3 and the biodegradation of NH_3 was significantly lower than that in biofilter A. Furthermore, TMA degradation might be inhibited by elevated NH_3 concentrations. This would explain both the poor TMA degradation in biofilter B from the beginning of the experiment on (Fig. 3b) and the decrease in efficiency of Section 1 of biofilter A in the second half of the experiment (Fig. 3a).

3.4. Variations in the properties of medium over time

Variations of pH, ammonia-N, nitrite-N, and nitrate-N contents in both biofilters are shown in Figs. 7 and 8. As discussed in the previous section, TMA in the medium was biodegraded into NH_3. Due to the alkaline nature of TMA and NH_3, it can be expected that the sorption of TMA and NH_3 might raise the pH of the medium. However, pH might be reverted or drop further by subsequent nitrification of ammonia to nitrite or nitrate. Therefore, the pH of the medium is determined by the rate of accumulation of ammonia and the rate of ammonia oxidation and nitrification of the reduced forms of nitrogen [17]. As

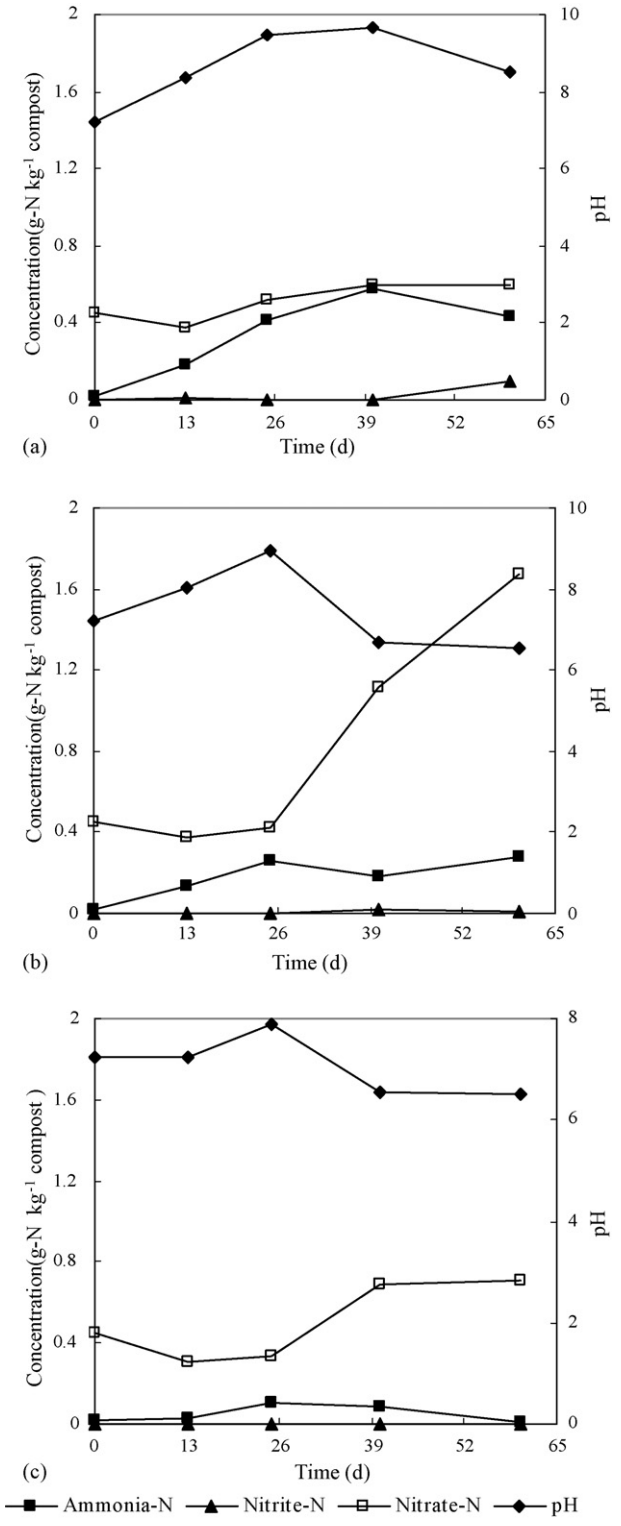

Fig. 7. Variations of pH, ammonia-N, nitrite-N, nitrate-N contents at different sections (a: Section 1; b: Section 2; c: Section 3) in biofilter A over the experimental period.

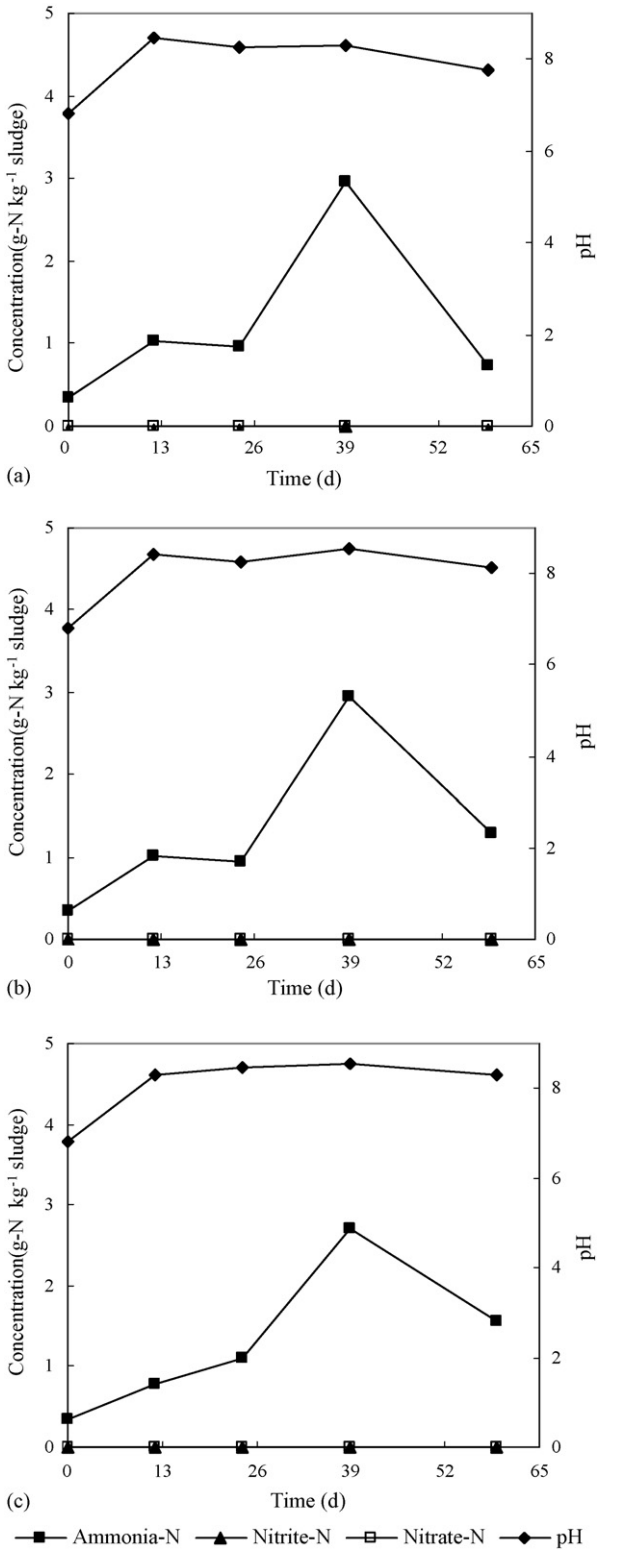

Fig. 8. Variations of pH, ammonia-N, nitrite-N, nitrate-N contents in different sections (a: Section 1; b: Section 2; c: Section 3) in biofilter B over the experimental period.

shown in Fig. 7, variations of pH and NH_4^+-N concentration in each section of biofilter A exhibited a similar trend with a gradual increase followed by a slight decline. Shifts of nitrate-N concentration in each section of biofilter A are also similar. From day 26, content of NO_3^--N in each section of biofilter A increased remarkably, indicating that nitrification had taken place. Ammonia-N was continuously transformed into NO_3^--N by nitrobacteria, which accumulated in each section resulting in pH decline. In comparison to Sections 1 and 3, NO_3^--N concentration in Section 2 was much higher after day 26. As described by Chou and Shiu [17], given different initial pH values, media differed significantly in their ability to biodegrade. Nitrification of ammonia is active only in a slightly alkaline environment—for example, in the pH range of 7.5–8.7. Due to the highest nitrogen loading, the highest initial pH (>8.7) in Section 1 of biofilter A inhibited nitrifying bacteria while the initial pH in Section 2 of biofilter A was very suitable for nitrifying bacteria. It was noted that NH_4^+-N and NO_3^--N in the packing material of Section 1 were gradually accumulated. A slight decline of the NH_4^+-N content in Section 1 was observed after day 39. Apparently, Section 1 of biofilter A with the highest nitrogen loading experienced a higher alkaline condition resulting in lowest capability for nitrification, while Section 2 exhibited better nitrification activity giving rise to the remarkable accumulation of NO_3^--N after day 26 (Fig. 7b). In Section 3, a small quantity of NH_4^+-N absorbed was almost completely converted into nitrate (Fig. 7c) which gradually acidified the medium. The pH of the medium remained around 7.0 and the concentration of NO_3^--N increased slightly suggesting that Section 3 of biofilter A still has great potential for nitrification. During the whole period of operation, NO_2^--N concentration in each section of biofilter A was notably low, indicating rapid transformation of NO_2^--N to NO_3^--N.

In contrast, neither NO_2^--N nor NO_3^--N was detected in each section of biofilter B over the whole study period confirming the absence of nitrifying bacteria in sludge packing material. It is also the reason why the variations of ammonia in effluent gas streams of each section in biofilter B were unapparent (Fig. 6b). Since nitrification is an autotrophic process, the availability of carbon sources is an important factor. Since sludge medium contains double the amount of organic matter (Table 1), nitrification may be inhibited by available carbon sources in sludge. Moreover, ammonium-N concentration in each section of biofilter B showed a similar trend with a gradual increase up to day 39 and a sudden drop thereafter. This might result from the saturation of NH_3 (Fig. 6b) and the lower moisture content (<20%) of sludge packing material (Fig. 4).

It is reasonable for the compost packing material with low bulk density, sufficient nutrients, and large water holding capacity to be able to enhance the growth of a variety of bacteria including the ammonia-oxidizer. Our result was in agreement with that of Bardtke et al. [16]. In addition, our previous study indicated that ammonia-oxidizing bacteria had already existed in compost [13]. Moreover, it is necessary for the sludge packing material to be inoculated with nitrifying bacteria in order to initiate ammonia oxidation in a bioreactor [13,18,19] and thus a continuous operation of biofilter B would be feasible.

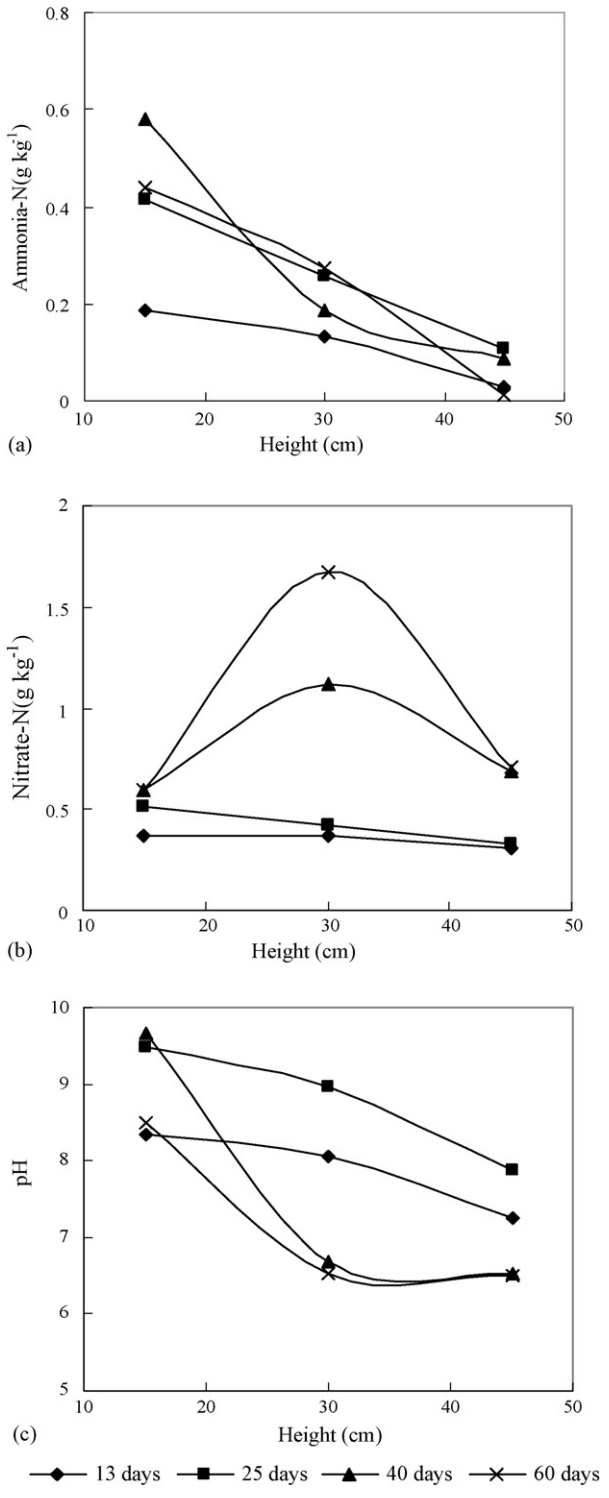

Fig. 9. Variation of ammonia-N (a), nitrate-N (b), and pH (c) with packing height for biofilter A.

3.5. Variations in the properties of the compost packing material with height

Fig. 9 showed variations of NH_4^+-N, NO_3^--N, and pH levels at different heights of biofilter A, respectively. As discussed in the previous section, within Section 1 above 80% influent

TMA was eliminated. In general, pH and NH_4^+-N levels of the packing material decreased with the increase of biofilter height. Due to a high level of the biodegradation in Section 1, a high amount of NH_4^+-N was generated and absorbed by the packing material resulting in the increase of pH. As a result of the high pH, nitrification in Section 1 was somewhat inhibited and the concentration of NO_3^--N in Section 1 was relatively low. With the increase of influent TMA, the highest level of NO_3^--N concentration was observed on days 40 and 60 since most of the NH_3 generated from the biodegradation of TMA in Section 1 (0–15 cm) was transformed into NO_3^--N within Section 2 (16–30 cm) under the favorable nitrification pH.

4. Conclusions

(1) The packing materials: compost and sludge, have different properties, which determine the different patterns of operation. TMA was successfully removed by both biofilters with 100% efficiency, with most removal occurring in the first two sections. The efficiency of Section 1 of biofilter A is more than 80% and that of biofilter B is also more than 60%. The maximum elimination capacities in Section 1 of biofilters A and B were 9.31 and 9.13 g TMA m^{-3} h^{-1}, respectively.

(2) NH_3 produced by biodegradation of TMA could be removed and converted to NO_3^--N in biofilter A, while in biofilter B the ammonium accumulates due to lack of nitrification. Compost medium are more favorable for the growth of TMA-degrading and nitrifying bacteria as compared to the sludge, which could be extraordinary packing material for biodegradation of TMA and nitrification of NH_3. It is necessary for the sludge packing material to be inoculated with nitrifying bacteria in order to initiate ammonia oxidation in bioreactor and thus a continuous operation would be feasible.

(3) Moreover, for the up-flow biofilters, the bottom (Section 1) was subjected to high TMA or ammonia loading, and its pH varied greatly due to the rate of accumulation of ammonia and the rate of ammonia oxidation and nitrification. The middle parts (Section 2) were subjected to moderate TMA and ammonia loading suitable for nitrification, and their environmental conditions lay on composite effect of biodegradation, nitrification and sorption.

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