

Journal of Hazardous Materials B80 (2000) 259-269



www.elsevier.nl/locate/jhazmat

Long-term results of ammonia removal and transformation by biofiltration

Yongkun Liang^a, Xie Quan^a, Jingwen Chen^{a,*}, Jong Shik Chung^b, Joon Y. Sung^c, Shuo Chen^a, Daming Xue^a, Yazhi Zhao^a

> ^a School of Environmental Science and Technology, Dalian University of Technology, Zhongshan Road 158-129, Dalian 116012, PR China

> ^b School of Environmental Engineering, Pohang University of Science and Technology, San 31, Hyoja-Dong, Pohang 790-784, South Korea

> ^c LG Institute of E.S. & H., Yonsei Engineering Research Center, 134 Shinchon-Dong, Seodaemun, Seoul 120-749, South Korea

Received 26 February 2000; received in revised form 18 August 2000; accepted 21 August 2000

Abstract

In this paper, long-term (>8 month) results of ammonia removal in biofilters was studied. Compost was used as the biofilter medium and activated carbon as an added material. The ammonia removal was normally >95% at influent ammonia concentrations of 20–500 ppmv. According to the test results, the influent ammonia concentration should be <200 ppmv (0.1570 g-ammonia/kg-media per day) so that the effluent concentration of ammonia is <1.0 mg/m³ (the emission standard of China), and the biofiltration system can achieve good long-term performance. In the biofiltration system utilized in this study, the shortest retention time that the system could attain was 0.532 min. However, the retention time can be decreased further without decreasing the ammonia removal efficiency. Countercurrent flow is favorable, as it enhances the moisture retention ability of the media. In the bioreactors, ammonia can be converted into the nitrate. The bioreactors have a stratification phenomenon for ammonia removal over the biofiltration depth, which implies that different parts of the bioreactor play different roles in the ammonia removing process. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Ammonia removal; Biofiltration; Air pollution control

* Corresponding author. Tel.: +86-411-3685113; fax: +86-411-3685113. *E-mail address:* destqc@dlut.edu.cn (J. Chen).

0304-3894/00/\$ – see front matter © 2000 Elsevier Science B.V. All rights reserved. PII: S0304-3894(00)00314-9

1. Introduction

Biofiltration has gained much interest in recent years for controlling the emission of odors air, toxics and VOCs [1–4]. Compared with traditional methods of air pollution control, biofiltration can be an inexpensive and effective gaseous waste treatment method without secondary-stream issues [5,6]. A biofilter consists of one or more beds of solid biologically active materials, such as peat, compost, soil, leaves, or woodbark [2,3,5,7]. The packing may be mixed with inert support materials such as porous clay, polystyrene spheres, granular activated carbon (GAC), diatomaceous earth, perlite, or vermiculite [4–6,8] to increase its reactive surface and durability, to reduce back pressure, and to prevent compaction [5,8].

The media hold and provide nutrients for the microbial consortium, which forms a biofilm on the media surface. The contaminant in the air dissolves in the water of the biofilm. The microbial consortium utilizes the contaminants to grow and metabolize, ultimately converting the contaminants to biomass and metabolites [3,8].

Ammonia is an air pollutant which is colorless and odorous. Gaseous ammonia is generated mainly by livestock, farming, petrochemical refining, metal manufacturing, and food, pulp and paper, and textile production [9]. The traditional treatment of ammonia emissions is based on physical and/or chemical processes, such as activated carbon adsorption and/or wet scrubbing, both of which are expensive and produce secondary pollutants.

There have been a few studies on biofiltration utilized to remove ammonia. Togashi et al. investigated NH₃ removal using peat as the packing medium. They also examined the feasibility of NH₃ removal by peat seeded with nitrifying bacteria [7]. Chung and Huang utilized the chemoautotrophic microorganism, *Nitrosomonas europaea*, to remove gaseous ammonia [10]. Martin et al. attempted to illustrate the control factors of a peat biofilter that allowed an effective operation of the gaseous nitrogen pollutant [11]. As far as we know, there are no reports on long-term results of ammonia removal and transformation by biofiltration, which was the purpose of this study.

2. Materials and methods

2.1. Biofilter system

The biofilter system utilized in this work consisted of a gas source, a gas flow control unit (flow meters and valves), mixing cells, and two parallel bioreactors (Bioreactors 1 and 2). The biofilter system is shown in Fig. 1. Ammonia from a gas cylinder was diluted in the mixing cell by air supplied by an air compressor. Contaminants in air were analyzed using a GC (Model 1102, Shanghai Analytical Equipment Plant); no chromatographic peaks of VOCs were observed. The bioreactors, 110 mm inner diameter and 1000 mm height were made of polymethyl methacrylate. There was a water distributor in the top of each reactor to maintain moisture in the media. Water was added manually to the media in the reactors once each day to maintain a moisture content of approximately 55–65%. Relative to the water flow, the contaminant gas could be introduced into the bioreactors by one of two modes, that is, in concurrent or countercurrent flow. For concurrent flow, the gas inlet was



Fig. 1. Scheme of the bioreactor system.

located in the top of the reactor and the gas outlet was located in the bottom of the reactor. The countercurrent flow was the inverse of the concurrent flow. There were three sampling outlets located on the wall of each reactor. The total empty volume of each reactor was 9.50 L.

The media composition of the bioreactors is shown in Table 1. Compost not only has a rich bacterial population that can transform ammonia, but also has an excellent capability to maintain moisture and nutrients [2,6]. Thus, compost was selected as the main packing medium. GAC was also added to the compost to reduce compaction and channeling, as well as increasing the reactive surface and durability of the filter materials. GAC also augmented the adsorptive ability of a biofilter with a goal of improving performance under shock waste gas loading [5,8].

Bacteria that can transform ammonia were collected from activated sludge from a wastewater treatment plant that treated domestic and industrial wastewater. The bacteria were acclimated to ammonia in the reactors. The acclimation time was approximately 2 weeks.

Table 1

Media composition of the bioreactors				
Volume (l) Weight (kg)		Composition of the media (weight ratio)		
Bioreactor 1 6.65	6.5	Compost:activated carbon = 4:1		
Bioreactor 2 7.03 5.8		Compost: activated sludge: granular activated carbon $= 4:4:3^{a}$		

^a Activated sludge was added to accelerate the acclimation of Bioreactor 2.

Table 2 The experimental arrangements
Bioreactor 1
From 12 October 1998 to 25 January 1

From 12 October 1998 to 25 January 1999	
1. Influent ammonia concentration: ca. 20 ppmv	
2. Gas flow direction: counter current	
From 12 October 1998 to 17 November 1998	
Influent gas flow rate = $0.30 \text{ m}^3/\text{h}$, EBRT = 1.330 min	
From 17 November 1998 to 23 December 1998	
Influent gas flow rate = $0.45 \text{ m}^3/\text{h}$, EBRT = 0.887 min	
From 24 December 1998 to 25 January 1999	
Influent gas flow rate = $0.75 \text{ m}^3/\text{h}$, EBRT = 0.532 min	
From 18 March to 30 August 1999	
1. Influent ammonia concentration: ca. 20 ppmv	
2. Gas flow direction: concurrent	
3. Influent gas flow rate = $0.75 \text{ m}^3/\text{h}$, EBRT = 0.532 min	
4. Analysis of nitrogen species in media	
5. Observation of the pH changes of biofilter	
preactor 2	
From 17 March to 25 November 1999	
1. Influent gas flow rate = $0.50 \text{ m}^3/\text{h}$, EBRT = 0.844 min	
2. Gas flow direction: counter current	
3. Analysis of nitrogen species in media	
From 17 March to 19 May 1999	
Influent emmonie concentration: as 100 ppmy	

Bio

From 17 March to 25 November 1999				
1. Influent gas flow rate = $0.50 \text{ m}^3/\text{h}$, EBRT = 0.844 min				
2. Gas flow direction: counter current				
3. Analysis of nitrogen species in media				
From 17 March to 19 May 1999				
Influent ammonia concentration: ca. 100 ppmv				
From 21 May to 30 August 1999				
Influent ammonia concentration: ca. 200-250 ppmv				
From 7 October to 25 November 1999				
Influent ammonia concentration: ca. 300-500 ppmv				



Fig. 2. Ammonia removal efficiency at different EBRT (Bioreactor 1).

262

Table 3	
Ammonia loading and elimination	rate

Influent ammonia concentration (ppmv)	Ammonia loading (g-ammonia/kg- media per day)	Effluent ammonia concentration (ppmv)	Removal efficiency (%)	Ammonia elimination rate (g-ammonia/kg- media per day)
20 ^a	0.0210	0.2-1.0	95.0-99.0	0.0200-0.0208
100 ^b	0.0785	0.4-1.1	98.9–99.6	0.0776-0.0782
200 ^b	0.1570	0.5-1.5	99.3-99.8	0.1558-0.1566
300 ^b	0.2355	0.8-2.2	99.3–99.7	0.2338-0.2349
500 ^b	0.3925	2.0-40.0	92.0–99.6	0.3611-0.3910

^a Represent for Bioreactor 1; influent gas flow rate = $0.75 \text{ m}^3/\text{h}$.

^b Represent for Bioreactor 2; influent gas flow rate = $0.50 \text{ m}^3/\text{h}$.



Fig. 3. Ammonia removal efficiency at an influent ammonia concentrations of 20 ppmv (Bioreactor 1).

B. Concurrent flow



Fig. 4. Ammonia removal efficiency at different influent ammonia concentrations (Bioreactor 2).

The experiments for Bioreactors 1 and 2 lasted for approximately 11 and 8 months, respectively. Details of these experiments are described in Table 2.

2.2. Analytical method

The diluted ammonia gas, sampled by an impulse sampler at the inlet and outlet of the reactor, was analyzed using the sodium salicylate/sodium hypochlorite spectrophotometric method that is a standard analysis method of PR China [12].

To investigate the transformation of ammonia in the reactors, 5 g solid samples were taken from the three sampling outlets of the reactors, impregnated in distilled water and separated from the water phase. In the effluent water, the concentrations of ammonium nitrogen (NH_4^+ -N), nitrite nitrogen (NO_2^- -N), nitrate nitrogen (NO_3^- -N), kjedahl nitrogen (K-N) and total nitrogen (T-N) were determined using standard Chinese analytical methods [12–14].

To determine the pH values of the media, some distilled water was added to a 5 g sample and then the mixture was blended [14] and used for measurement. Moisture was measured by weight loss after the medium sample was dried at $103-105^{\circ}$ C to constant weight [14].

3. Results and discussion

3.1. Empty bed retention time (EBRT)

Empty bed retention time (EBRT) is an important biofilter operating parameter. Assuming a constant concentration of NH_3 , in principle, the shorter the EBRT, the higher the influent ammonia loading. As shown in Fig. 2, at EBRTs of 1.330, 0.887 and 0.532 min, the ammonia removal efficiencies were in the range of 91.4–99.0%. These results indicate that the three different EBRTs had no significant effects on NH_3 removal, which can be attributed to the



Fig. 5. Concentration changes of NO_3^- -N, NO_2^- -N, NH_4^+ -N, organic-N, K-N and T-N in the media of Bioreactor 1.

fact that the influent NH_3 loading in Bioreactor 1 was relatively low. As shown in Table 3, the ammonia loading in Bioreactor 1 was 0.0200-0.0208 g-ammonia/kg-media per day. It appears that the retention time could be reduced further without decreasing the ammonia removal efficiency. Nevertheless, 0.532 min was the shortest retention time that we could attain in Bioreactor 1 being limited by the maximum flow rate of air that could be supplied by the air compressor.

3.2. Gas introduction modes

As shown in Fig. 3, ammonia removal in Bioreactor 1 employing countercurrent flow and concurrent flow modes was 93.0–99.9 and 91.4–99.3%, respectively. For both of the gas introduction modes, Bioreactor 1 achieved a high ammonia removal efficiency. However, for concurrent flow, we found that the media in the upper and middle portions of the biofilter has a poor ability to retain moisture, resulting in deteriorated performance.

3.3. Influence of initial ammonia concentration on ammonia removal

Fig. 4 illustrates the ammonia removal efficiency in Bioreactor 2 when the influent ammonia concentration was 100–500 ppmv. The ammonia loading and ammonia elimination rate in Bioreactor 2 are shown in Table 3. At influent ammonia concentrations of 100 and 200 ppmv, the mean ammonia elimination rate was equivalent to 0.0779 and 0.1562 g-ammonia/kg-media per day, respectively, and the effluent ammonia concentration was below 1.5 ppmv. At influent ammonia concentrations of 300 and 500 ppmv, the mean ammonia elimination rate was equivalent to 0.2344 and 0.3761 g-ammonia/kg-media per day, respectively; the effluent ammonia concentration was in excess of 1.5 ppmv. It can be concluded that Bioreactor 2 has a relatively high capability for ammonia removal. However, with the increase of influent ammonia concentration and operational time, the effluent ammonia concentration increased slowly and ultimately exceeded the emission standards for odoriferous pollutants set by the EPA of China [12], namely, 1.0 mg/m³ for ammonia



Time of Analysis (year-month-day)

Fig. 6. Change in pH of Bioreactor 1.



Fig. 7. Concentration variations of NO_3^--N , NO_2^--N , NH_4^+-N , organic-N, K-N and T-N in the media of Bioreactor 2.

nia. These results imply that the influent ammonia concentration should be <200 ppmv (0.1570 g-ammonia/kg-media per day for ammonia loading) for Bioreactor 2 in order to achieve good long-term performance.

3.4. Analysis of various forms of nitrogen in the media

3.4.1. Bioreactor 1

Bioreactor 1 was operated in a concurrent flow mode. The initial concentrations of various N forms are shown in the results of the first sample in Fig. 5. Due to the utilization of concurrent flow, ammonia loading in the upper part of the bioreactor was higher than in the middle and lower parts. From Fig. 5, it can be seen that nitrate was the main inorganic nitrogen form. Thus, it can be concluded that the ammonia absorbed by the media was ultimately converted into nitrate. As shown in Fig. 6, the pH value in the biofilter media decreased slowly. This result may be caused by nitrate accumulation in the media.

In Fig. 5, it can be seen that Bioreactor 1 exhibited a stratification phenomenon for ammonia removal over the biofiltration depth. The upper section of the biofilter was the main part that absorbed and transformed ammonia; the mid section was the next efficient location and the last section was the least efficient in removing ammonia.

3.4.2. Bioreactor 2

Bioreactor 2 was operated in a countercurrent flow mode. The influent ammonia concentrations ranged from 100 to 500 ppmv. It can be seen in Fig. 7 that Bioreactor 2 also showed a stratification phenomenon in ammonia removal over the biofiltration depth. The lowest part of the biofilter was the main part to absorb and transform ammonia. The initial concentrations of various N forms are shown in the first data points in Fig. 7. It can be seen from Fig. 7 that nitrate and ammonium were the main inorganic nitrogen forms in the media. Thus, it can be concluded that ammonia absorbed by the media was partly converted into nitrate. In further studies, the volatilization of absorbed NH₃ should be monitored after turning off or decreasing the ammonia in the influent stream.

4. Conclusions

In this study, long-term (>8 month) results of ammonia removal in biofilters were measured. The ammonia removal efficiencies of the biofilter systems were high (>95%). According to these results, the influent ammonia concentration should be <200 ppmv (0.1570 gammonia/kg-media per day), so that the effluent concentration of ammonia is <1.0 mg/m³ (the emission standard of China) and the biofiltration system can achieve good long-term performance. In the biofiltration system used in this study, the shortest retention time that the system could attain was 0.532 min. The retention time may be decreased further without decreasing the ammonia removal efficiency. Countercurrent flow is the optimal gas introduction mode, since the moisture retention ability of the media can be enhanced. In the bioreactors, ammonia can be converted into the nitrate. Finally, in our opinion, biofiltration is the best method for treatment of ammonia of low emission concentration (<200 ppmv) due to its low equipment cost, low operating cost and low reclamation values of ammonia.

Acknowledgements

This study was supported by the Science and Technology Fund of the Liaoning Province of China (Grant No. 9910600202), the Teaching and Research Award Program for Outstanding Young Teachers in Higher Education Institutions of MOE, China, the Research and Development Fund of Dalian Municipal Government of China, and the Scientific Research and Development Fund of the LG Group, Korea. The financial supports are gratefully acknowledged.

References

- [1] R.D. Pomeroy, J. WPCF 54 (1982) 1541.
- [2] H.L. Bohn, J. Air Pollut. Control Assoc. 25 (1975) 953.
- [3] C. Quinlan, K. Strevett, M. Ketcham, J. Grego, J. Air Waste Manage. Assoc. 49 (1999) 544.
- [4] T.S. Webster, J.S. Devinny, E.M. Torres, S.S. Basrai, Environ. Prog. 15 (1996) 141.
- [5] G. Leson, A.M. Winer, J. Air Waste Manage. Assoc. 41 (1991) 1045.
- [6] R.L. Corsi, L. Seed, Environ. Prog. 14 (1995) 151.
- [7] I. Togashi, M. Suzuki, M. Hirai, H. Kubota, J. Ferment. Technol. 64 (1986) 425.
- [8] W.J. Swanson, R.C. Loehr, J. Environ. Eng. 123 (1997) 538.
- [9] J.E. Ryer-Power, Plant Operations Prog. 10 (1991) 228.
- [10] Y.C. Chung, C.P. Huang, Environ. Prog. 17 (1998) 70.
- [11] G. Martin, M. Lemasle, S. Taha, J. Biotechnol. 46 (1996) 15.
- [12] National Standards Compilation of Environmental Protection, 1st Edition, Standards Press of China, Beijing, 1995.
- [13] National Standards Compilation of PR China (86), 1st Edition, Standards Press of China, Beijing, 1992.
- [14] Methods for Monitoring and Analyzing of Water and Wastewater, 3rd Edition, Environmental Science Press of China, Beijing, 1989.