Biological removal of air loaded with a hydrogen sulfide and ammonia mixture

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Abstract: The nuisance impact of air pollutant emissions from wastewater pumping stations is a major issue of concern to China. Hydrogen sulfide and ammonia are commonly the primary odor and are important targets for removal. An alternative control technology, biofiltration, was studied. The aim of this study is to investigate the potential of unit systems packed with compost in terms of ammonia and hydrogen sulfide emissions treatment, and to establish optimal operating conditions for a full-scale conceptual design. The laboratory scale biofilter packed with compost was continuously supplied with hydrogen sulfide and ammonia gas mixtures. A volumetric load of less than $150 \, gH_2S/(m^3 \cdot d)$ and $230 \, gNH_3/(m^3 \cdot d)$ was applied for about fifteen weeks. Hydrogen sulfide and ammonia elimination occurred in the biofilter simultaneously. The removal efficiency, removal capacity and removal kinetics in the biofilter were studied. The hydrogen sulfide removal efficiency reached was very high above 99%, and ammonia removal efficiency was about 80%. Hydrogen sulfide was oxidized into sulphate. The ammonia oxidation products were nitrite and nitrate. Ammonia in the biofilter was mainly removed by adsorption onto the carrier material and by absorption into the water fraction of the carrier material. High percentages of hydrogen sulfide or ammonia were oxidized in the first section of the column. Through kinetics analysis, the presence of ammonia did not hinder the hydrogen sulfide removal. According to the relationship between pressure drop and gas velocity for the biofilter and Reynolds number, non-Darcy flow can be assumed to represent the flow in the medium.

Keywords: biological removal; hydrogen sulfide; ammonia mixture

Introduction

Waste gases containing reduced sulphur and nitrogen compounds such as hydrogen sulfide (H_2S) and ammonia (NH_3) have an unpleasant odor even at extremely low concentrations. They are toxic air pollutants, and control of their emission is important for public health and welfare. The hazardous caused by high concentrations of H_2S and NH_3 are known, but there is little information on human exposure to very low concentrations, although recent studies have shown that the adverse health effects of malodorous sulphur compounds occur even at lower concentrations (Partti-Pellinen, 1996).

The nuisance impact of air pollutant emissions from wastewater pumping stations is a major issue of concern to China. Wastewater treatment plants can be constructed well away from populated areas, but the sewage must still be conduced through pumping stations located in populated areas, so complaints from neighbors are common. Kangas *et al*. (Kangas, 1986) analyzed ammonia, methane, hydrogen sulphide and methyl mercaptides in the atmospheres of 16 Finnish municipal wastewater treatment plants and in 18 pumping stations. Under normal operating conditions, sulphides varied from < 0.07 to 53 mg/m^3, with the highest concentrations found at the sludge presses. In pumping stations sulphide vapours varied from 0.07 to 0.5 mg/m^3. Ammonia concentrations were 0.007—3.5 mg/m^3. According to the results of our investigation to several pumping stations in summer 2001, hydrogen sulfide and ammonia are the primary odor, and their concentrations in the investigated pumping stations were 0.38—8.75 mg/m^3 and 1.31—33.1 mg/m^3, respectively. Existing methods for controlling odorous off-gas include activated carbon adsorption, caustic and acid scrubbers, thermal oxidation, and so on. Although these methods provide high odor removal efficiency, they are also costly and may lead to the generation of secondary pollutants. Biofiltration, on the other hand, is thought to be one of the best technologies in terms of its technical and economical aspects.

Compost used as packing material in biofilter for hydrogen sulfide and ammonia removal has high liquid retention and develops a high mass transfer area without inducing high pressure drops. Compost is thus favorable to bacterial colonization and nutrient solutions containing sources of carbon, nitrogen and phosphorus do not have to be supplied to maintain the growth and activity of sulfur oxidizing bacteria and ammonia-oxidizing bacteria. Hence, it is interesting to develop the biofiltration process using compost and more accurately to study the potential of this support for olfactory nuisance removal. Moreover, in order to better control and optimize biofiltration for a full scale implementation, it is important to study the performance of laboratory pilot biofilters whose inlet gaseous effluent is a malodorant pollutants mixture.

In this study, biofilter was employed to treat odorous hydrogen sulfide and ammonia gases in a laboratory-scale. The scope of this work was to evaluate the elimination capacity of a compost biofilter to purify waste gases containing

H_2S and NH_3. Based on the results, it was also aimed to develop the excellent biofilter which can be practically applied.

1   Materials and methods

1.1   Laboratory-scale biofilter

The biofilter consisted of a Plexiglas column with an internal diameter of 0.15 m and a height of 1.1 m (Fig.1). The column was packed with compost on top of a perforated sieve plate fitted at the bottom of the column to ensure the uniform distribution of the inlet gas. The column was provided with four outlets, located at intervals of 20 cm. Hydrogen sulfide and ammonia levels were measured and samples taken at these outlets. The flow of airstreams was 20 L/min, and empty bed retention time is 20 s. H_2S was generated by chemical method and NH_3 was supplied from gas cylinder. They were first diluted with compressed air and flowed upwards through the bottom of the biofilter.
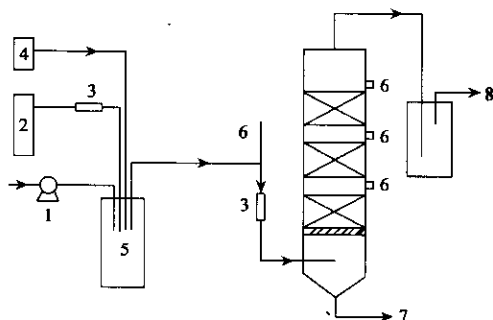

Fig.1   Schematic of the lab scale biofilter

1. air pump; 2. NH_3 gas cylinder; 3. flow meter; 4. H_2S gas; 5. mixing bottle; 6. sampling port; 7. percolate waters; 8. effluent air

The compost produced from yard waste was used as a carrier material. Twenty percent (w/w) of perlite (sieve fraction between 1.2 and 4.0 mm) were mixed with the compost to increase voidage of the biofilter. The wet bulk density of the carrier material was 0.692 g/cm^3. The moisture content of the filter material was 37%. The compost was inoculated with activated sludge from the Sibao Wastewater Treatment Plant. Water was splashed manually on the compost the column every three days to maintain moisture content of the compost at about 40%.

1.2   Analysis of the pollutants

Ammonia and hydrogen sulfide were transferred in aqueous solution by bubbling the gaseous influent in a solution of sulfuric acid and zinc acetate, respectively. The ammonia concentration was the evaluated by a spectrophotometric method using Nessler reagent. The pH of the carrier material was measured after mixing 10 g carrier material with 100 ml distilled water during 15 min. The moisture content was calculated by the weight difference before and after drying at 105℃ to constant weight. The wet bulk density was determined by weighing a column with known volume before and after filling with compost material. The concentrations of ammonium nitrogen, nitrite nitrogen and nitrate nitrogen of the carrier material were determined using standard Chinese analytical method.

1.3   Surface and mass loading

The surface loading rate (L_s) is a measure of the volumetric gas loading to a biofilter. Mass loading rate(L_m), a combination of the waste airflow rate and the contaminant concentration in the waste gas stream, is defined as the mass of pollutant introduced into a biofilter per unit volume of filter material per unit time. Removal efficiency (RE) is the operating parameter used to judge the success of a biofilter in terms of bioconversion of a contaminant.

Empty bed residence time(s), waste air surface loading rate(m/h), contaminant mass loading rate($gNH_3/(m^3 \cdot h)$), and removal efficiency (%) were determined using the relationships between the influent and effluent gas phase concentrations, waste airflow rate, and the volume of the biofilter material as follows:

$$\tau = V/Q, \tag{1}$$
$$L_s = Q/A, \tag{2}$$
$$L_m = Q/V \times C_{in}, \tag{3}$$
$$RE = [(C_{in} - C_{out})/C_{in}] \times 100. \tag{4}$$

Where, Q is the waste airflow rate(m^3/h); A is the area of cross-section of biofilter column (m^2); V is the volume of filter material (m^3); M is the molecular weight of the contaminant(17 for NH_3 and 34 for H_2S); and C_{in} and C_{out} are the contaminant concentrations in the inlet and outlet waste gas streams(mg/m^3).

1.4   Biomass counting

The biomass measurements were obtained as follows: most probable number (MPN)-Griess counting for nitrifying bacteria: The protocol used for MPN-Griess counting was based on that of Schmidt et al. (Schmidt, 1994). The dispersed packing suspension was subsampled before sedimentation for a series of dilution(1 : 10) and inoculations of MPN tubes with a five-fold Nitrosomonas autotrophic medium with (NH_4)$_2$$SO_4$ 0.50 g/L or a five-fold Nitrobacter autotrophic medium with $NaNO_2$ 1 g/L. Samples were incubated for three months at 28℃ in the dark, the Griess reagent was added, and the number of Nitrosomonas or Nitrobacter cells per gram of compost was determined with MPN tables(Lorch, 1995).

MPN counting for thiobacilli: The dispersed packing suspension was subsampled before sedimentation for a series of dilution (1 : 10) and inoculations of MPN plates with Thiobacillus thioparus autotrophic medium. Samples were incubated for 15 d at 28℃ in the dark. The cell numbers were expressed in colony forming units(cfu).

1.5   Kinetic analysis

Two processes, mass transport and microbial utilization of contaminants, simultaneously occur in biofiltration. The

particles in the filter bed are surrounded by a wet, biologically active layer, called a biofilm. Convection and dispersion of contaminants take place in the air phase, while the biodegradation occurs within the biofilm. In general, by assuming that the oxygen concentration required for the aerobic respiration of the microorganisms in the biofilm is not limiting, under selected conditions, the substrate utilization rate of a compound by the microbial flora as well as enzymatic reaction can be expressed by a Michaelis-Menten type relationship(Hirai, 1990; Wani, 1999). At steady state the growth rate of the microorganisms due to biodegradation is balanced by its own decay, resulting in no net growth and eventually biological equilibrium is achieved, so that kinetic constant over the time period considered. Under such conditions, i. e. when the microbial population does not change, the half saturation constant(K_m) and the substrate concentration(C) may have comparable value. The modified Michaelis-Menten equation:

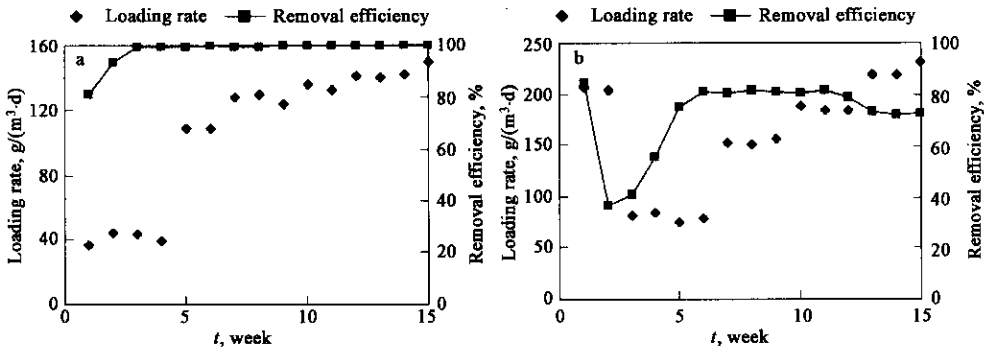$$\frac{1}{R} = \frac{K_m}{V_m}\frac{1}{C_{ln}} + \frac{1}{V_m},\qquad(5)$$

where $R(g/(m^3 \cdot d))$ is apparent removal rate; $C_{ln} = [(C_0 - C_e)/\ln(C_0/C_e)]$, logarithmic means concentration of pollutant in the biofilm; $C_0(mg/m^3)$ is the concentration of pollutant in the biofilm at the inlet; $C_e(mg/m^3)$ is the concentration of pollutant in the biofilm at the outlet; $V_m(g/(m^3 \cdot d))$ is the maximum apparent removal rate, and K_m (mg/m^3) apparent half-saturation constant. From the linear relationship between $1/C_{ln}$ and $1/R$, V_m and K_m were calculated from the intercept and slope, respectively.

2 Results and discussion

2.1 Reactor efficiency

In the process of experiment, the inlet concentrations of H_2S and NH_3 were 10—50 mg/m^3 and 25—80 mg/m^3, respectively. The loading rates of H_2S and NH_3 were 40—150 $g/(m^3 \cdot d)$ and 80—230 $g/(m^3 \cdot d)$, respectively. The removal efficiency and loading rate of H_2S and NH_3 as a function of time is shown in Fig.2 for the compost biofilter.

Fig.2 Compost biofilter removal efficiency and loading rate evolution as a function of time
(a: H_2S; b: NH_3)

For biological removal of H_2S, there was a short start-up period(about a week), however, complete H_2S-removal was obtained from week 2. As a steady state is reached, removal efficiency remained above 99%. No critical or limiting load for hydrogen sulfide was found from data (Fig. 3a). The highest load during this study was at 150 $g/(m^3 \cdot d)$. These results showed that low concentrations of NH_3 have no effect on H_2S efficiency.

NH_3 removal reached low values during the first four weeks($\leqslant 55\%$). As shown in Fig.2b, due to the physico-chemical interaction of NH_3 with the carrier material, high removal efficiencies were obtained during the first week. From week 2, however, a strong reduction in EC was obtained. Subsequently, although NH_3 loading rate was deceased, the removal efficiency did not increase. These results are explained by the adaptation phase of nitrifying bacteria community during this delay. The biofilter environmental conditions and a long generation time of 10 h (Bock, 1989) could be also unfavourable to the nitrifying

community growth. So the elimination capacity of NH_3 decreased when nonbiological NH_3 removal reached saturation. As a steady state was reached, the NH_3 oxidation efficiency varied between 70% and 80%, and the maximum elimination capacity was about 160 $g/(m^3 \cdot d)$ (Fig.3b). The possible reason for low NH_3 efficiency is that in a biofilter distinct microbial populations frequently interact with each other and the sulfide oxidizing community could have a negative effect on the nitrifying community growth.

The hydrogen sulfide oxidation was then carried out more easily than the ammonia oxidation. Kowal (Kowal, 1993) showed that hydrogen sulfide oxidation also happens using a chemical pathway which is observed during adaptation phase of sulfide-oxidizing bacteria. Because of the sensitivity of nitrifying microorganisms, however, biofiltration of waste gases containing high NH_3 concentrations(above 50 ppmv) has been reported to be questionable. In an inoculated activated carbon biofilter, Yani *et al*. found a complete NH_3 removal up to loading rate 95 $gNH_3/(m^3 \cdot d)$, whereas the
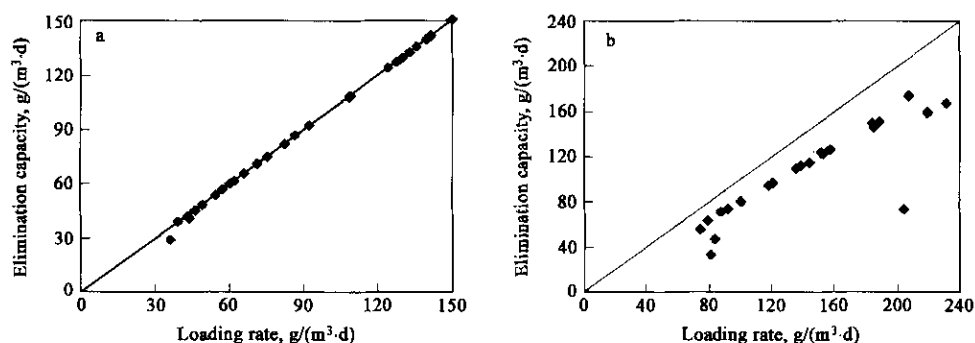
Fig.3 H_2S and NH_3 elimination capacity of the compost biofilter as a function of load

elimination efficiency decreased at higher loads (Yani, 1998). The highest EC observed was 220 $gNH_3/(m^3 \cdot d)$ at LR 250 $gNH_3/(m^3 \cdot d)$, and at an empty bed residence time of 52 s(Yani, 1998). Smet(Smet, 2000) obtained EC up to 350 $gNH_3(m^3 \cdot d)$ in a compost biofilter($t = 131$ s).

2.2 Concentration profiles

Fig. 4 presents the experimental concentration profiles obtained for the compost biofilter. The concentration profile shows that H_2S is completely eliminated in the first section of the biofilter, 71.4% of NH_3 inlet load is degraded in the first section of the column and 10% in the other sections. High percentages of NH_3 or H_2S are oxidized in the first section of the column. H_2S oxidation activity and nitrification are greater in the first section and in the other sections is due to the utilization of countercurrent flow. These results showed that the NH_3 oxidation has no negative effect on H_2S removal.
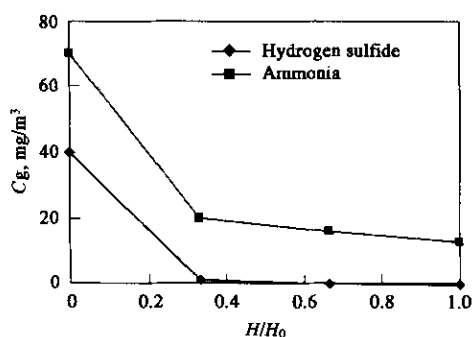
Fig.4   Concentration profiles in the compost biofilter as a function of height

2.3 Effect of pH on NH_3 removal

The effect of pH(3.0—7.5) on H_2S removal was not significant(data not shown). However, NH_3 removal effection was highly dependent on pH. Due to nitrification, pH will decrease during biological removal of NH_3. So it is necessary to consume alkalinity to balance H^+ produced in the reaction to maintain the neutral or alkalescent environment where nitrifying bacteria can grow well. Therefore, the NH_3 removal efficiency was highly dependent on pH. When pH is below 5.70, nitrification process in the solution will almost not occur. The compost samples used for acidity tests were treated with dilute HCl or NaOH solutions to bring the initial

pH of the compost to the desired range. Following a few days of operation, the inlet and outlet gas sample concentrations were measured when the bed operation become stable(Fig. 5). NH_3 inlet concentration was about 50 mg/m^3, and superficial gas velocity in the experiment was 20 L/min. In addition, the removal efficiencies of NH_3 were average values.
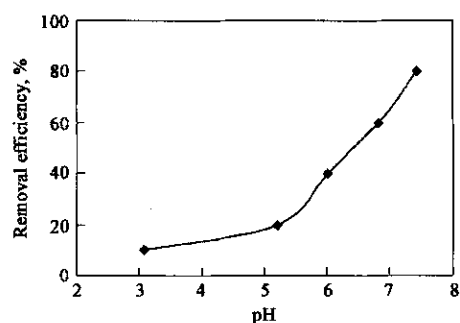
Fig.5   Effect of compost pH on NH_3 removal efficiency

The NH_3 removal efficiency was highly dependent on pH above 7.0 but was almost independent of pH at lower values. The maximum NH_3 removal occurred at a compost pH value of 7.8 (83.1%). Nitrifying bacteria can live in environments having a narrow pH range(7.7—8.1). For the higher pH range, chemical reaction between nitrate and the compost material can significantly enhance its removal.

2.4 Product analysis

Due to its rather low Henry Constant $H_{20℃}$ of 5.6×10^{-4} (Perry, 1984) and its pH-dependent protonation, ammonia in biofilters is partly retained by physico-chemical processes(such as absorption or adsorption). Some authors (Togashi, 1986; Cho, 1992) reported a chemical NH_3-removal in a biofilter loaded with NH_3 and sulfur compounds due to the neutralization reaction of NH_3 with the metabolite sulfuric acid.

Therefore at the different time of the experiment, NH_4^+-N, NO_2^--N and NO_3^--N contents of the compost were analyzed. The NH_3 removal in the biofilter resulted in a strong increase of NH_4^+-N content, while NO_2^--N and NO_3^--N contents of the compost material slightly increased

(Table 1).

Table 1 NH$_4$-N, (NO$_2$ + NO$_3$)-N and SO$_4^{2-}$-S of the carrier material during the biofilter experiment

	Time, week			
	0	5	10	15
NH$_4$-N, g/kg	0.05	0.60	1.47	2.36
(NO$_2$ + NO$_3$)-N, g/kg	0.006	0.015	0.045	0.34
SO$_4^{2-}$-S, g/kg	0	0.55	1.85	3.31

As Table 1 shown, the increase in ammonia content of the compost is the highest, nitrite and nitrate content are very low. It shows that ammonia in biofilter is mainly removed by adsorption process and absorption process. The nitrification process is not significant. These results seem to be accordant with Don(Don, 1985). In addition, this assumption could be supported by the counts realized at the end of the experiment (Table 2): the number of ammonia oxidizing bacteria(10^3—10^4 bacteria/g packing material) is 10 superior to that of nitrite oxidizing bacteria (10^2—10^3 bacteria/g packing material) in the biofilter.

Table 2 Number of nitrifying and sulfide oxidizing bacteria

	H/H_0		
Number of bacteria, bacteria/g compost	0.3	0.7	1
Ammonia oxidizing bacteria	1.7×10^4	2.7×10^3	4.7×10^3
Nitrite oxidizing bacteria	5.4×10^3	3.4×10^2	2.1×10^2
Sulfide oxidizing bacteria	9×10^5	5×10^5	2×10^5

2.5 Kinetic analysis

Biodegradation rate parameters, V_m and K_m, were estimated from modified Michaelis-Menten model(Eq.(5)). Step changes in inlet concentrations were made in the biofilter columns where H$_2$S was biodegraded as a substrate and the elimination capacities over the first section of the filter bed were estimated. After adjusting the inlet concentration the system was allowed to stabilize for 24 h prior to analyzing the outlet concentration and changing to new inlet concentration.

The reciprocal of H$_2$S elimination capacities of the biofilter plotted against the reciprocal of log-mean concentrations of H$_2$S are summarized in Fig.6. The apparent kinetic parameters of the maximum removal rate and half-saturation constant to degrade H$_2$S under the presence of NH$_3$ are calculated by the Lineweaver-Burk method. Fig. 6 indicates that a low NH$_3$ concentration has a little effect on the metabolism of H$_2$S. The K_m and V_m values were calculated to be 12.8 mg/m^3 and 213 g(H$_2$S)/(m$^3 \cdot$ d), respectively without ammonia. While the K_m and K_m values were calculated to be 14.4 mg/m^3 and 256 g(H$_2$S)/(m$^3 \cdot$ d), respectively, at the existence of NH$_3$.

Generally, if we inferred a physical meaning for K_m analogous to enzymatic kinetics, a higher of K_m value indicated a lower enzymatic affinity for H$_2$S(Chung, 2001). Thus, the existence of NH$_3$ has a little effect on H$_2$S removal by the biofilter.

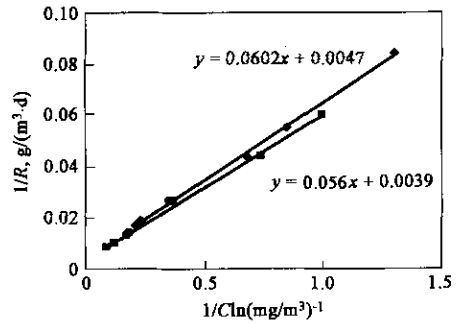

Fig.6 Effect of NH$_3$ concentrations on H$_2$S removal by compost biofilter

2.6 Pressure drop

The relationship between pressure drop and superficial gas velocity is an important parameter in determining the operational cost. The influence of gas velocity on pressure drop is shown in Fig. 7. Pressure drop of the biofilter increased with increasing gas velocity. The relationship between gas velocity and pressure drop was nonlinear. There is a good agreement between pressure drop and square of gas velocity with a R^2 value of more than 0.97. The interstitial Reynolds number (R_e) can be found by the following equation:

$$R_e = \frac{D_P \nu_\infty \rho}{(1 - \theta)\mu}. \tag{6}$$

Where D_P = average particle diameter, ν_∞ = approach velocity, ρ = density of air, θ = medium porosity, and μ = viscosity of air(25℃ assumed).

At 20 L/min, the approach velocity is 18.9 mm/s. The average particle size was assumed to be 4 mm, and the porosity was experimentally determined to be 71.6%. Therefore, the Reynolds number was 14. Darcy flow is usually applicable in medium where the interstitial Reynolds number is less than 1. So non-Darcy flow can be assumed to represent the flow in the medium in this study.
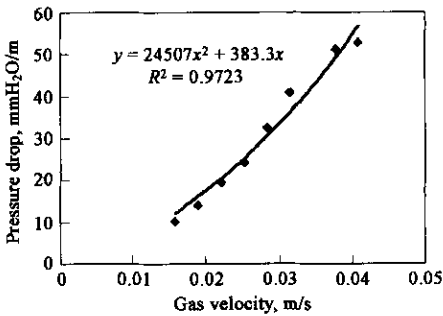

Fig.7 Profile of pressure drop vs. gas velocity for the biofilter

3 Conclusions

Even though odor control in pumping stations which transport wastewater has become a major concern, the use of biofilters has limited application and does not have high a success record in China. The performance of laboratory-scale biofilter using compost was investigated in this study. During

the operation period of 15 weeks, the compost biofilter showed good removal efficiency of more than 99% to low concentration of H_2S. However, NH_3 removal efficiency was only 70%—80%. The maximum elimination capacities of H_2S and NH_3 were 150 $g/(m^3 \cdot d)$ and 160 $g/(m^3 \cdot d)$, respectively. While no toxicity effect of low NH_3 concentrations on the biofiltration of H_2S was observed. From macrokinetic analysis, the existence of NH_3 has a little effect on H_2S removal by the biofilter. The NH_3 removal efficiency was highly dependent on pH above 7.0 but was almost independent of pH at lower values. The maximum NH_3 removal occurred at a compost pH value of 7.8(83.1%). Biofiltration of H_2S and NH_3 resulted in the accumulation of NH_4^+ -N, $NO_2 + NO_3$-N and SO_4^{2-} -S in the compost material. Ammonia in the biofilter is mainly removed by adsorption onto the carrier material and by absorption into the water fraction of the carrier material. High percentages of hydrogen sulfide or ammonia are oxidized in the first section of the column. There is a good agreement between pressure drop and square of gas velocity with a R^2 value of more than 0.97. According to the relationship between pressure drop and gas velocity for the biofilter and Reynolds number, non-Darcy flow can be assumed to represent the flow in the medium.

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