

LIVESTOCK AIR TREATMENT USING PVA-COATED POWDERED ACTIVATED CARBON BIOFILTER

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ABSTRACT. *This study evaluated the efficacy of polyvinyl alcohol (PVA)-coated powdered activated carbon particles as a biofiltration medium. This material exhibited excellent properties as a biofiltration medium with a water holding capacity of 1.39 g H₂O/g-dry PVA; wet porosity of 0.53; and significantly lower pressure drop than that of commonly reported biofilter media such as compost. Bench-scale biofilters treating off-gas by aerating flushed swine manure samples were used to evaluate ammonia and hydrogen sulfide removal capacities along with greenhouse gas production potentials. Although ammonia adsorption capacity was much lower than granular activated carbon, the PVA biofilter medium retained its ammonia removal capacity because of suspected biological nitrification. The PVA biofilters continued to remove 80% of ammonia in the air for the entire 37 days of operation. While the biofilters produced 0.14 g N₂O-N/L-wet PVA, another greenhouse gas methane production was negligible. Hydrogen sulfide was effectively removed (97%) by the PVA biofilters.*

Keywords. *PVA biofilter, Ammonia, Hydrogen sulfide, Nitrous oxide, Livestock air, Greenhouse gas.*

Reducing ammonia (NH₃) and hydrogen sulfide (H₂S) emissions from livestock facilities is an important issue for many communities and livestock producers. These gases have been regarded as two major odor gases with very low odor thresholds levels: 1.1 ppb for H₂S and 37 ppb for NH₃ (Eikum and Storhaug, 1986; Henry and Gehr, 1980). While the desired reduction of ammonia and hydrogen sulfide emission is facilitated by properly designed animal houses, it can be substantially enhanced by the use of biofiltration technology. This technology employs both natural and synthetic biofiltration media providing surface areas for the attachment of microorganisms that remove air-borne contaminants.

Natural filter media have been more commonly used. They consist of materials such as compost, soil, peat, and woodchips. They have been frequently employed in biofiltration technology to reduce emissions of ammonia, hydrogen sulfide, and other odorous compounds from municipal, industrial, and agricultural facilities (Chung et al.,

2004; Classen et al., 2000; Elmrini et al., 2004; Green et al., 2005; Janni and Nicolai, 2000; Morgan-Sagastume and Noyola, 2006; Nicolai and Janni, 1999; Nicolai et al., 2002; Sheridan et al., 2002; Sun et al., 2000; Wani et al., 1999). Although their costs are low, the natural filter media will decompose, settle, and compact with time (Sun et al., 2000). Alternatively, specially designed synthetic media can overcome some of the structural limitations of the natural filter media. Various synthetic biofilter media have been employed to support microorganisms that remove NH₃ and H₂S such as polyurethane foam cubes (PU), granular activated carbon, synthetic polymer, natural fibers, and proprietary filter media (Ostlie-Dunn et al., 1998; Shareefdeen et al., 2002; Chan and Lu, 2003; Gabriel and Deshusses, 2003; Chan and Lin, 2004; Chung et al., 2004). Recently, powdered activated carbon particles coated with polyvinyl alcohol gel (hereafter PVA particles) were developed as synthetic biofilter media for removing odors from municipal wastewater treatment plants by the Denka Consultant & Engineer Co., LTD in Japan. The PVA particles have high 1) surface areas to support large microbial communities, 2) moisture holding capacities from the hydrophilic PVA polymer, and 3) porosities that reduce pressure drops. These particles can also maintain structural integrity at low pH (Chan and Lin, 2004). Filter material resistance to low pH is an extremely important trait for biofilters treating odorous ammonia- and sulfur-containing compounds which produce large amounts of acidity. The colorless sulfur bacteria from the genus *Thiobacillus* will create acidity via the oxidation of sulfide to sulfate (Kuenen and Robertson, 1992; Chung et al., 1996). The nitrifying bacteria will produce acidity via oxidation of ammonia to nitrate under aerobic conditions of the biofiltration and also lower the pH of the wastewater (Metcalf & Eddy, Inc., 2003).

In addition to acidity, biofiltration may produce greenhouse gases. For instance, N₂O may be formed during either nitrification or denitrification (Bremner and Blackmer, 1978; Tallec et al., 2006; Hunt et al., 2007). Previously, most studies of livestock air biofiltration have focused on odors,

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NH₃, H₂S, or VOCs. To date, greenhouse gas production potential from biofiltration of livestock air has not been reported. In this study, we assessed the effectiveness of the PVA biofilters with respect to greenhouse gases as well as NH₃ and H₂S.

The objective of this study was to evaluate the efficacy of PVA particles as a biofiltration support medium by 1) characterizing the size, pressure drop, and adsorption capacity of the PVA particles; 2) evaluating the performance of bench-scale PVA biofilters in reducing ammonia and H₂S from swine manure aerated air; and 3) examining the greenhouse gas production potential of PVA biofiltration.

MATERIALS AND METHODS

PRESSURE DROP

Resistance to air flow is the major contributing factor for blower power requirement which increases with pressure drop across biofiltration systems. Therefore, accurately predicting pressure drop is an important step in designing of biofilters. The Ergun equation has been frequently used in estimating pressure drop across biofilters and other packed bed systems (Ergun, 1952; Deront et al., 1998; Yang and Allen, 2005; Morgan-Sagastume and Noyola, 2006).

$$\frac{\Delta P}{L} = \frac{150\mu(1-\epsilon)^2 u}{\phi^2 d^2 \epsilon^3} + \frac{1.75\rho(1-\epsilon)u^2}{\phi^2 d \epsilon^3} \quad (1)$$

where

ΔP = pressure drop (Pa),

L = bed height (m),

u = superficial velocity (m s⁻¹)

= Q/A ,

Q = volumetric air flow rate (m³ s⁻¹),

A = cross-sectional area of biofilter (m²),

d = characteristic particle diameter (m),

ϕ = sphericity (surface of sphere/surface of particle of the same volume)

ϵ = porosity,

μ = viscosity of air (kg m⁻¹ s⁻¹), and

ρ = density of air (kg m⁻³).

In order to evaluate the applicability of the Ergun equation in predicting pressure drops across PVA biofilters, pressure drops from passing air (3-13 L min⁻¹) through a 5.1-cm diameter column packed with 1.2 m of clean PVA particles were monitored with a micromanometer (AXD 560, TSI Inc., Shoreview, Minn., accuracy $\pm 1\%$ of reading). The equilibrium pressure drops were achieved rapidly (within 5-10 min). Because the column diameter was about one order of magnitude larger than PVA particles, the wall effects on the pressure drop were assumed to be negligible (Di Felice and Gibilaro, 2004). All pressure drop measurements were conducted in triplicate.

PVA PARTICLE SIZE AND SHAPE CHARACTERIZATION

Ellipsoidal PVA particles of nominal mesh size between 4.8 and 9.5 mm were obtained from the Denka Consultant & Engineer Co., LTD (Chiba, Japan). In order to use the Ergun equation (eq. 1) to predict pressure drops, PVA biofilters were assumed to contain uniform size PVA particles with a characteristic diameter and sphericity. The Sauter-mean diameter or the mean volume-surface diameter has been used

frequently to characterize non-uniform particles (Hinds, 1982; Shieh and Chen, 1984; Ro and Neethling, 1990 and 1994). The Sauter-mean diameter was estimated based on the measurements of 100 randomly selected PVA particles. Major and minor axes of the 100 ellipsoidal PVA particles were measured using a scaled monocular microscope (18 – 36X Zoom, RF Interscience Co, Coram, N.Y.).

$$d_{sm} = \frac{d_v^3}{d_s^2} \quad (2)$$

where

$$d_v = \text{average volume diameter} = \left(\frac{\sum n_i d_{v,i}}{N} \right)^{1/3}$$

$$d_s = \text{average surface area diameter} = \left(\frac{\sum n_i d_{s,i}}{N} \right)^{1/2}$$

$$d_{s,i} = \text{equivalent surface area diameter of particle } i \text{ (m),}$$

$$= \sqrt{2a_i b_i \left(\frac{b_i}{a_i} + \frac{\sin^{-1} \sqrt{1 - (b_i/a_i)^2}}{\sqrt{1 - (b_i/a_i)^2}} \right)},$$

$d_{v,i}$ = equivalent volume diameter of particle i (m),

$$= 2(a_i \cdot b_i^2)^{1/3}$$

a_i, b_i = major and minor axes of an ellipsoidal particle i (radii, m),

n_i = number of i particles, and

N = total number of particles.

The equivalent diameters are the diameters of spheres having the same volume or surface area of non-spherical particles (Narzaroff and Alvarez-Cohen, 2001). The characteristic sphericity of the PVA biofilter particles was then estimated as:

$$\phi = \frac{4\pi d_v^2}{4\pi d_s^2} = \left(\frac{d_v}{d_s} \right)^2 \quad (3)$$

AMMONIA ADSORPTION BY PVA PARTICLES

Ammonia adsorption capacity of the PVA filter was determined with the bed depth service time (BDST) model (Bohart and Adams, 1920; Hutchins, 1973; Aksu and Gonen, 2004; Lin and Wang, 2004). Bohart and Adams (1920) proposed the original BDST model relating filter bed depth to the service time taken for breakthrough to occur. Hutchins (1973) later simplified the original BDST model equation and proposed a linear relationship between service time and filter bed depth (eq. 4).

$$t = \frac{z q_{vo}}{u C_o} - \frac{1}{k C_o} \ln \left(\frac{C_o}{C} - 1 \right) \quad (4)$$

where

C_o = influent solute concentration (mg solute m⁻³),

C = effluent solute concentration (mg solute m⁻³),

k = kinetic coefficient (m³ mg⁻¹ solute-min),

q_{vo} = maximum volumetric adsorption capacity (mg solute m⁻³),

t = service time (min),

u = superficial velocity (m min⁻¹), and

z = filter depth (m).

The PVA adsorption capacity (q_{vo}) was then estimated from the intersection $\left(\frac{zq_{vo}}{uC_o}\right)$ of a regression line (fitted to eq. 4) obtained by plotting t versus $\ln\left(\frac{C_o}{C} - 1\right)$. To assess

ammonia adsorption, a calibrated gas stream with 30.8 ± 2 ppm NH_3 (National Specialty Gases, Research Triangle Park, N.C.) was passed through 5.1-cm diameter columns packed with 0.25 m of PVA particles.

BULK PACKING VOLUME, POROSITY, AND WATER ABSORPTION OF PVA PARTICLES

Due to their hydrophilic properties, PVA particles absorbed water and the particle volume was expanded. Interstitial void fraction, bulk packing volume expansion, and water absorption characteristics were determined gravimetrically using 500-mL graduated cylinders. The water adsorption by PVA particles was estimated by determining the difference of the mass of water using Ohaus analytical balance (readability of 0.1 mg) and a scale (readability of 0.1 g) (Pine Brook, N.J.) that could be drained from the water-PVA mixture (at different adsorption durations up to 125 h) and the initial mass of water added to the graduated cylinder. The interstitial void fraction (hereafter called porosity) was determined by the ratio of the volume of the drained water to the bulk packing volume of the water-PVA mixture. These properties of PVA particles were determined in duplicate.

BENCH-SCALE PVA BIOFILTRATION SYSTEM

The bench-scale biofiltration system consisted of a manure tank, an intake manifold, biofilters, and a blower

(fig. 1). The two biofilters were made of 10.2- × 62.2-cm (4- × 24.5-in.) clear PVC pipe. About 1.3-L bulk volume of wet PVA particles were used for each biofilter. To obtain an inoculation for an active, immobilized microbial culture within the PVA particles, they were initially immersed for a week in an aerated 18.9-L activated sludge mixed liquor. The activated sludge was obtained from a local municipal wastewater treatment plant.

The inflow ammonia and hydrogen sulfide were produced from aerating flushed manure from a 4,400-head finishing swine farm in North Carolina. The aeration was done in a (56.8-L) sealed tank. The off-gas from the aerated manure tank was then introduced at the bottom of the biofilters. Each biofilter receives the off-gas at a rate of $8.4 \times 10^{-5} \pm 2.6 \times 10^{-5} \text{ m}^3 \text{ s}^{-1}$, resulting in an empty bed residence time (EBRT) of 18 ± 8.7 s. Our EBRT was within the range of EBRTs reported for commercial, agricultural, and industrial applications of biofiltration, i.e., 2 s to more than a minute (Leson and Winder, 1991; Deviny et al., 1999; Nicolai and Janni, 1999; Sheridan et al., 2002). The biofilter microbial moisture and nutritional requirements were maintained by recirculating approximately 1.7 L of swine lagoon water (1 h d^{-1}). Replacement of off-gas-generating-manure was required because continuous aeration stabilized the manure in the aerated tank. Fresh manure was used for each experimental run. The duration of each experimental run was mainly determined by availability of fresh manure and operator's work schedule.

ANALYTICAL METHODS

The NH_3 concentration was measured with a photoacoustic multi-gas analyzer (Innova 1412, California Analytical Instruments, Inc., Orange, Calif.). The

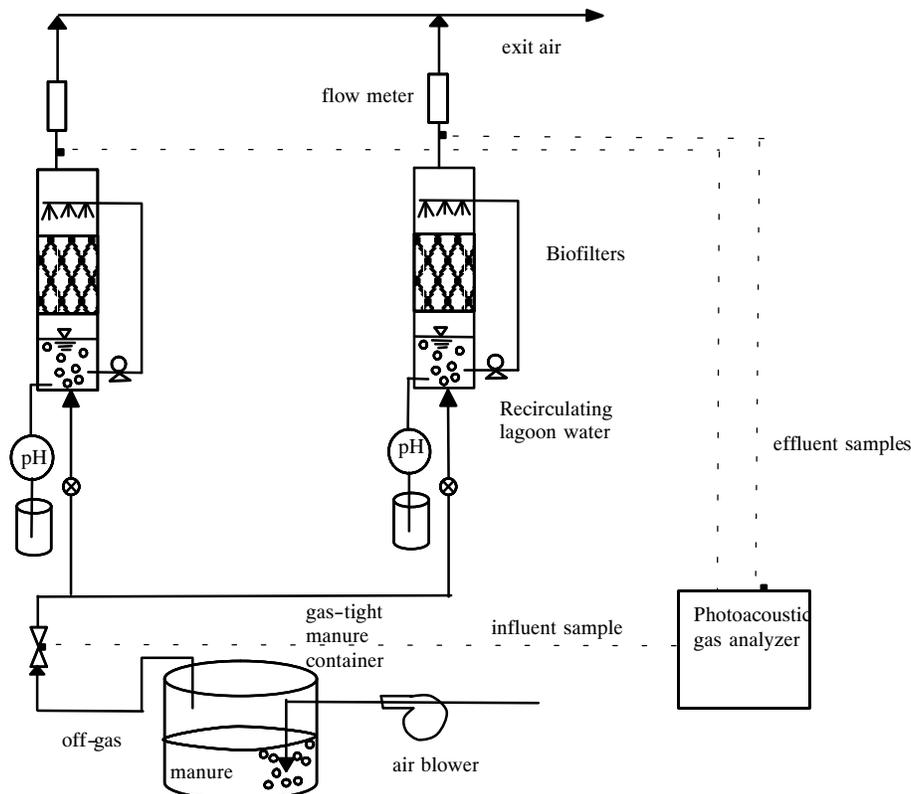


Figure 1. Schematic diagram of bench-scale biofiltration system.

photoacoustic analyzer was factory calibrated and intermittently its accuracy was evaluated with calibrated gases of NH₃, CO₂, CH₄, and N₂O with certified accuracy of less than 5% (National Specialty Gases, Research Triangle Park, N.C.). Hydrogen sulfide concentration was measured daily with a hydrogen sulfide detector utilizing a gold-film sensor (Jerome 631 X, Arizona Instruments, Tempe, Ariz.). The accuracy of the hydrogen sulfide analyzer was ±0.003 ppm at 0.05 ppm (effluent range) and ±0.03 ppm at 0.5 ppm (influent range). Recirculating lagoon water was analyzed for ammonia, nitrate/nitrite, TKN, and pH according to the Standard Method (APHA, 1998).

RESULTS AND DISCUSSION

BULK DENSITY, POROSITY, AND WATER ABSORPTION OF PVA PARTICLES

The PVA particles expanded as they absorbed water (fig. 2). Within 4 h after PVA particles were immersed in water, these particles absorbed water to about 90% of the maximum absorption level of 1.39±0.06 g H₂O/g dry PVA. Concomitantly, the bulk packing volume increased from 2.33±0.09 to 4.68±0.13 cm³g⁻¹ dry PVA (fig. 2). This high water absorption capacity of the PVA particles is important in supporting attached microorganisms (Chan and Lu, 2003). The porosity of the PVA particles changed little with the addition of water. It ranged from a dry porosity of 0.56±0.03 to a fully-expanded state porosity of 0.53±0.02 (fig. 2). The relatively constant porosity indicates that the pressure drop per unit length would not increase appreciably in a PVA filter even after the PVA particles fully absorb water.

PRESSURE DROP

PVA particle size, shape, packing density/porosity, and air flow rate impacted pressure drop. The Sauter-mean diameters for dry and wet PVA particles were 0.72 and 0.93 mm, respectively. The characteristic sphericity of both wet and dry PVA particles was 0.98. Using these PVA particle size and sphericity, the Ergun equation slightly under predicted the observed pressure drops as shown in figure 3. The error bars represent the standard deviation of triplicate tests. The pressure drop of the wet PVA particles was slightly higher than that of dry PVA particles. Nonetheless, the pressure drops of the either wet or dry PVA particles were

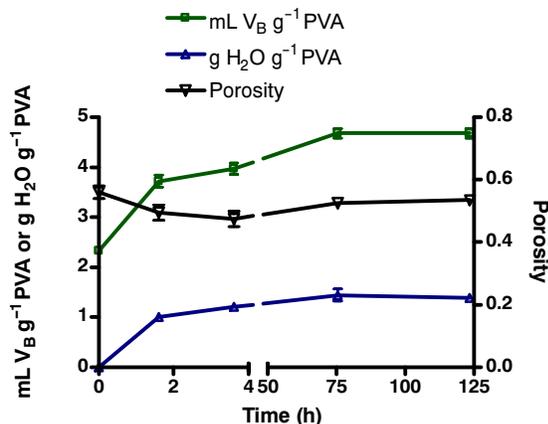


Figure 2. Change in PVA particle characteristics with water absorption (V_B = bulk packing volume).

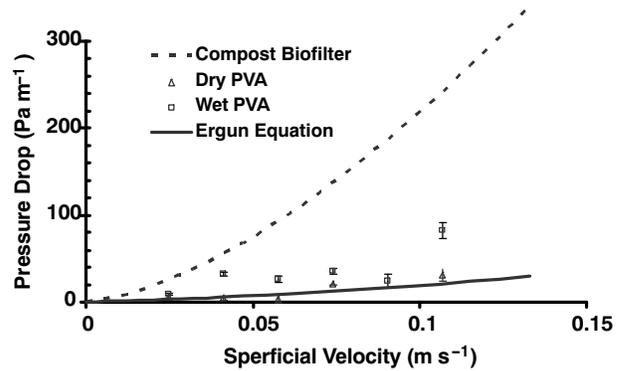


Figure 3. PVA media pressure drops according to equation 4 and estimated pressure for compost (Classen et al., 2000).

very small relative to that of compost biofilter medium as predicted by the empirical equation (eq. 5) as suggested by Classen et al. (2000).

$$\frac{\Delta P}{L} = 7761 \cdot u^{1.55} \quad (5)$$

AMMONIA ADSORPTION BY PVA PARTICLES

Three sets of data with more than 60% ammonia removal were used to estimate the adsorption capacity as shown in figure 4. The ammonia adsorption capacity of the PVA filter was 5.3±0.3 g m⁻³, or 0.012±0.001 mg g⁻¹ dry PVA. This adsorption capacity is far less than 0.6 mg-N g⁻¹ dry granular activated carbon (GAC) as reported by Chung et al. (2005). With this low adsorption capacity, PVA particles would be physically saturated with ammonia on the second day of operation. In other words, this material would not be effective in removing ammonia in the air via physical adsorption alone. Nonetheless, as expected, the PVA biofilters effectively removed ammonia in the air while nitrate concentration was increased in the recirculating lagoon water. The biological component of the PVA biofilters (i.e., nitrification) was able to compensate for the low adsorption capacity of PVA particles and removed most of ammonia in the air. Furthermore, this biological ammonia removal mechanism was not being diminished in time.

AMMONIA REMOVAL EFFICIENCY

For the first 20 days of acclimation period, the influent ammonia concentration ranged from 4 to 96 ppm. The inlet ammonia concentration decreased as the manure was stabilized with the aeration. During this period, the PVA biofilters were not able to remove ammonia from the air and

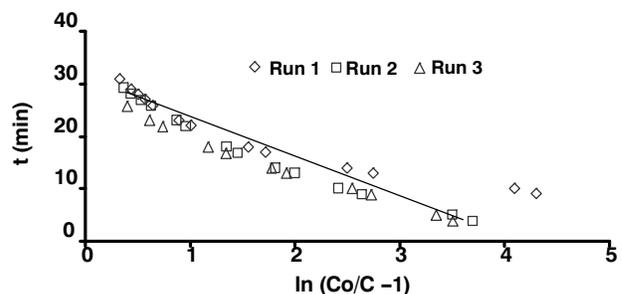


Figure 4. Linear relationships between service time and ln(Co/C-1) of equation 4.

the effluent ammonia concentrations were practically the same as that of influent. However, after the 20 days of acclimation period, the PVA biofilters removed most of ammonia in the influent. The ammonia concentration profiles of the biofilter influent and the average of the two PVA biofilter effluents after 20 days of acclimation period are shown in figure 5. The average coefficient of variation of the two biofilter effluent ammonia concentrations was 10.0%. Influent air ammonia concentration reached as high as 125 ppm; effluent air ammonia concentrations were mostly under 7 ppm. During Run 3, ammonia concentrations in the fresh manure sample were low due to a reduced number of animals in the facility. Although it was not intentionally planned, Run 3 and Run 4 show how well the PVA biofilters sustained its ammonia removal capacity during the period (more than 10 days) of very low ammonia loadings and how quickly the PVA biofilters responded to shock loading in Run 4.

The ammonia removal efficiency was calculated as:

$$\text{removal efficiency (\%)} = \frac{C_o - C}{C_o} \times 100 \quad (6)$$

Ammonia removal efficiency decreased with decrease in the influent ammonia concentration. Less than 80% removal efficiencies were observed when influent ammonia concentrations were less than about 5 ppm. Figure 6 shows the cumulative ammonia mass introduced in the influent and emitted in the biofilter effluents. For the entire 37 days of operations, the PVA biofilters removed 80% of the total ammonia in the influent. The global efficiency of the PVA biofilter was lower than that of compost biofilters which maintained more than 95% efficiency in removing ammonia for 6 days (Pagans et al., 2005). However, the ammonia removal efficiency of the compost biofilter receiving very high ammonia concentration (>2000 mg m⁻³) dropped to 30%. Busca and Pistarino (2003) reported that the ammonia removal efficiencies using natural media such as compost, peat, wood chips, and oyster shells ranged from 70% to 99.7%. The ammonia removal efficiency of the PVA biofilter was comparable to that of biofilters using these natural media.

The ammonia removed from the PVA biofilters was probably nitrified by nitrifying bacteria. The classical biological nitrogen pathway involves a two-step process. Aerobic ammonia oxidizers such as the well-known genera *Nitrosomonas* and *Nitrosospira* oxidize ammonia (not ammonium) to hydroxylamine, which is further oxidized to

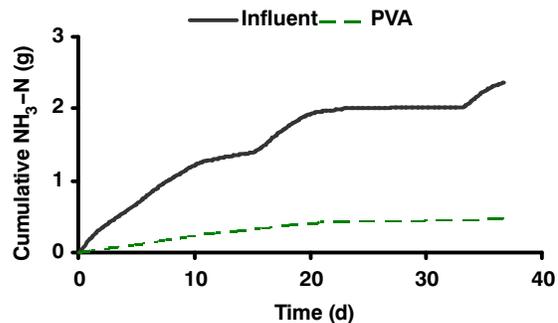
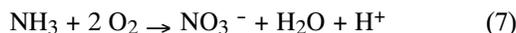


Figure 6. Cumulative mass of NH₃ in influent and PVA-biofiltered air.

nitrite by the enzyme hydroxylamineoxidoreductase (HAO). The nitrite is further oxidized to nitrate by different groups of nitrifying bacteria, *Nitrobacter* and *Nitrosopira* (McCaig et al., 1999; Cebren et al., 2003; Metcalf & Eddy, Inc., 2003; Pukhold et al., 2003). The stoichiometric equations for these nitrification steps are:



Equation 7 suggests that if nitrification occurred in the biofilters, we should have observed a decrease in pH due to production of proton and increase in nitrate. Table 1 clearly shows that these chemical characteristic changes took place in the PVA biofilter recirculating water. After 6 days of operation during Run 1, the ammonia concentration of the PVA biofilter recirculating water was slightly reduced from 356 to 276 mg/L. Simultaneously, NO_x concentrations increased from 0 to 123 mg/L, and the pH decreased from 8.59 to 6.69. These changes indicated that nitrifying bacteria in the PVA biofilter were partly responsible for ammonia removal by oxidizing it to nitrate.

H₂S REMOVAL EFFICIENCY

The PVA biofilters also effectively removed H₂S from the air as shown in Figure 7. The error bars of the effluent concentrations represent the standard deviation of the two biofilter effluent H₂S concentrations. For the entire 37 days of operation, the PVA filters removed 97% of H₂S based on the total mass flow of H₂S in the influent and effluent. This H₂S removal efficiency of the PVA biofilters was slightly lower than 99.8% removal efficiency of a peat biofilter inoculated with *Thiobacillus thioparus* DW44 (Cho et al., 1992). Following the trend of ammonia, the influent H₂S concentration increased abruptly whenever fresh manure was recharged for each run. Although microbial sulfur removal

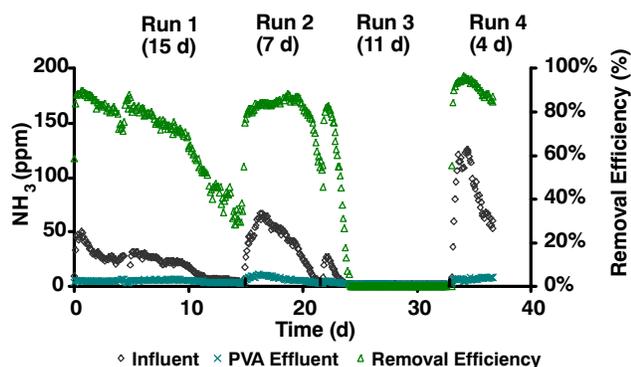


Figure 5. Ammonia concentrations and removal efficiencies of PVA biofilters.

Table 1. Chemical characteristics of PVA biofilter recirculating water.

Chemicals	Initial Concentration	After 6 Days of Operation (Run 1)
pH	8.59	6.69±0.21
Temperature	23.0°C	22.9°C
TKN (mg-N/L)	399	276±27
NH ₃ -N (mg-N/L)	355.8	276±19
NO _x -N (mg-N/L)	Below detection	123±15

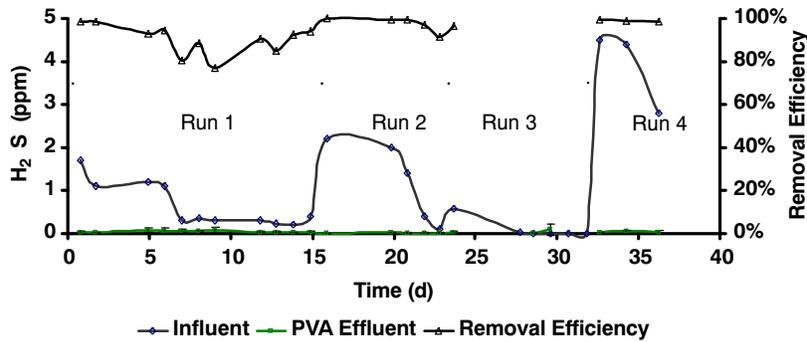


Figure 7. H₂S concentrations and removal efficiencies of PVA biofilters.

mechanism involving *Thiobacillus* species were suggested in the literature (Sun et al., 2000), it is not clear at this time whether the H₂S was removed abiotically such as adsorption, biotically, or both. Further examination of H₂S removal mechanisms is required for effective design of biofilters.

GREENHOUSE GAS EMISSION FROM BIOFILTRATION

Greenhouse gases from influent and effluent from the PVA biofilters were monitored during the 37 days of operation. The greenhouse gas concentrations in figures 8-10 were the average effluent concentration of the two PVA biofilters. The average coefficients of variation of the two biofilter effluent greenhouse gas concentrations were 18,6%, 9,2%, and 6,2% for N₂O, CO₂, and CH₄, respectively. The PVA biofilter emitted slightly more CO₂ than introduced as shown in figure 8. This was likely due to microbial respiration in the PVA medium. The total CO₂ produced from aerating swine manure (i.e., influent to the PVA biofilters) was 0.21 kg; the total CO₂ in the PVA biofilter effluent was 0.23 kg. There was virtually no difference in CH₄ before and after PVA biofilter as shown in figure 9. This indicates that there were little CH₄ reducing microbial activities in the PVA biofilter. However, the PVA biofilter produced significantly more N₂O (0.14 g N₂O-N/L-wet PVA) than was introduced as shown in figure 10. It is likely that N₂O was produced by aerobic nitrifiers in the biofilter. Bremner and Blackmer (1978) also observed N₂O emission from soil during nitrification of ammonium and ammonium producing fertilizers under aerobic conditions. Tallec et al. (2006) reported that autotrophic denitrification, not heterotrophic denitrification, was the major mechanism deriving N₂O emission from activated sludge samples.

CONCLUSIONS

The efficacy of PVA-coated powdered activated carbon biofilters in removing NH₃, H₂S, and greenhouse gases was studied using bench-scale units. The PVA particles expanded as the hydrophilic polymer absorbed water. This provided the necessary moisture for microbial community in the PVA particles. The PVA particles showed excellent pressure drop characteristics, with significantly smaller pressure drops than compost biofilter medium. The ammonia adsorption capacity of the PVA particles was only 0.012 mg NH₃/g-dry PVA, significantly lower than granular activated carbon. Yet, despite the low NH₃ adsorption capacity of the PVA particles,

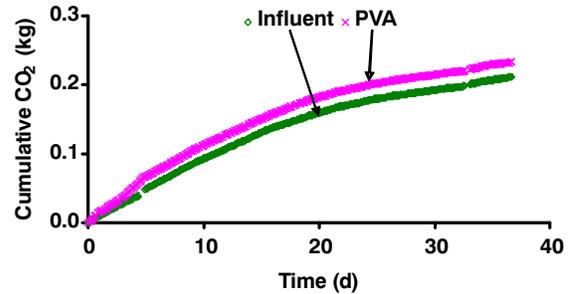


Figure 8. Cumulative CO₂ mass measured in influent and emitted from biofiltration.

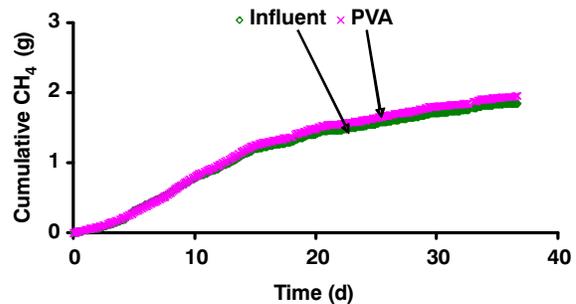


Figure 9. Cumulative CH₄ mass measured in influent and emitted from biofiltration.

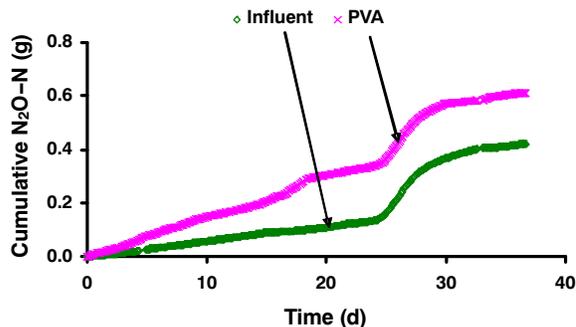


Figure 10. Cumulative N₂O mass measured in influent and emitted from biofiltration.

80% of the total ammonia in the influent was effectively removed by the PVA biofilters during the 37 days of operation. Based on recirculation water chemical characteristics, nitrification of ammonia was suspected as the governing factor in removal of ammonia in the air. The PVA

had no effect on methane. The PVA biofilter produced significantly more N₂O than introduced. Because N₂O is a potent greenhouse gas, its production potential while removing NH₃ and H₂S during biofiltration may pose a serious environmental set back for this technology. It is not clear if other biofilters using different media would also produce greenhouse gases; further study is needed. The removal mechanism for H₂S was not clear; nonetheless, the PVA biofilters removed H₂S effectively. For the entire 37 days of operation, the PVA filters removed 97% of the total H₂S in the influent.

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