

DESIGN AND DEVELOPMENT OF A NOVEL BIOFILTER

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ABSTRACT

The design and development of a novel biofilter is described in this paper. Moisture control is a key aspect of the design. Moisture from the gas stream leaving the biofilter is condensed and returned to the top of biofilter as reflux of pure water. The refluxed water maintains a moist environment for the bacteria throughout the total biofilter column. It also washes down microbial product/s (nitrite and nitrate in the case of ammonia oxidation) that may be toxic to microorganism at a high concentration. This allows microbial activity to be maintained at its most favorable level in the top section of the biofilter. Optimization of moisture balance between evaporation and condensation can result in a system whereby water level on the media surface is sufficient for microbes to thrive while no leachate is produced.

In a proof-of-concept study, a laboratory-scale computer controlled biofilter system employing the reflux process was operated to remove ammonia from an air stream ($2.05 \mu\text{mol} / \text{L} = 50 \text{ ppmv}$) at a volume load of 1 L/L/min . Inoculated zeolite (clinoptilolite) was used as filter media. Depth profile analysis of the filter bed showed the development of a steep gradient of ammonia, nitrite and nitrate from the top layer to the bottom layer of filter media in the biofilter. Aside from the well-known ammonium adsorption on zeolite, consistent microbial nitrification continued to take place. Nitrite and nitrate level at the bottom of the biofilter continued to increase over 180 day period of experiment. As a result, ammonia removal efficiency remained close to 100% in the leachate-free biofilter.

Keywords: biofilter, ammonia removal, zeolite, clinoptilolite, moisture control, reflux

INTRODUCTION

All microorganisms need water, e.g. for nutrient transfer and mobility, which require moist conditions in a biofilter (Jun and Wenfeng 2009). Commonly used biofilter systems have been constructed with a water supply on top of the biofilter (Baquerizo et al. 2009). Differences occur depending whether fresh water or nutrient solution is introduced (Pandey et al. 2009) or whether the water is recycled (Sakuma et al. 2009). For most systems, the accumulated water (leachate) from the bottom is reintroduced to the top (recycling) of the biofilter and causes a uniform filter bed (Sakuma et al. 2009) giving no room for the development of different microenvironments with unique capabilities. Furthermore, reintroducing the water-dissolved pollutants on top of the biofilter, leads to an increased risk of incomplete treatment and discharge. Studies reveal that leachate production is often a common problem of conventional biofilters (Baquerizo et al. 2009, Pandey et al. 2009, Kim et al. 2008). Leachate contains accumulated water dissolvable pollutants (e.g. NH_3) and their degraded compounds (e.g. NO_2^- , NO_3^-) and needs further treatment prior to disposal, which leads to questioning the efficiency of biofiltration (Baquerizo et al. 2009, Nisola et al. 2009).

This paper describes a novel design concept and implementation of a biofilter that achieved moisture controlled environments with zero leachate production leading to a gradient of concentration distribution favouring microbial activities within the biofilter.

METHODS

Design concept

We propose the application of the reflux mechanism to biofiltration. The reflux mechanism is based on temperature differences between biofilter inlet and outlet, causing water to evaporate at the biofilter bottom and to condense at the top. Due to this condensation cycle, minimal amount of water is required for the process. This mechanism is well known from applications to crystallogenetic adsorbents (Lamb and Ohl 1935), COD- analysis (APHA 2005) and extraction chemistry (Eiserbeck et al. 2011).

Concept of the reflux process

The process of condensation of vapours and the return of this condensate to the system from which it originated is widely accepted as a reflux technique. Evaporation of water at the biofilter inlet (biofilter bottom) occurs by controlling the moisture content and temperature of the gas stream entering the biofilter to below the conditions at which the biofilter is operated. An installed cooling device on top of the filter system causes water condensation and reintroduction of pure water to the system. The water percolation through the biofilter prevents the filter from drying out, and a wash-down of accumulated pollutants and metabolites is achieved. Temperatures and corresponding water content can be calculated as described below.

Application of the reflux technique to the biofiltration concept would reduce the concentration of pollutant in the top section and increase concentration in the bottom section of the biofilter system, thereby creating a concentration gradient of the pollutant throughout the filter. This gradient allows biological activity in the top and concentrates the pollutants and microbial products that are generally toxic to microbes at a high concentration in the bottom section.

Concept of moisture control

A moisture controlled biofilter system was developed on the basis of gas stream humidification and condensation (Figure 1). It consists of an inlet dryer before the biofilter and a condenser above the biofilter.

The vapour pressure of air over water is saturated when the number of water molecules condensing equals the number evaporating from the surface of water. The saturation vapour pressure at a certain temperature can be calculated according to Tetens (1930) to determine the water content for a saturated gas in [Pa] (Equation 1). Equation 1 shows the saturation vapour pressure (610.78 [Pa]) at the temperature of 273.16 [K] multiplied by the exponential temperature (T) constant term and the vapour pressure of water at this temperature as 17.2694 mm Hg.

$$ps = 610.78 + \exp(T/T + 238.3) \times 17.2694 \quad \text{Equation 1}$$

Equation 2 allows the calculation of the water content (wc [kg m^{-3}]) at a certain saturation vapor pressure (Wexler 1976). The molar mass of water ($M = 0.018$ [kg mol^{-1}]) divided by the gas constant ($R = 8.314$ [J (K mol)^{-1}]) multiplied by the ratio of saturation vapor pressure and temperature (T [$^{\circ}\text{C}$]).

$$wc = M/R + ps/T + 273.16 \quad \text{Equation 2}$$

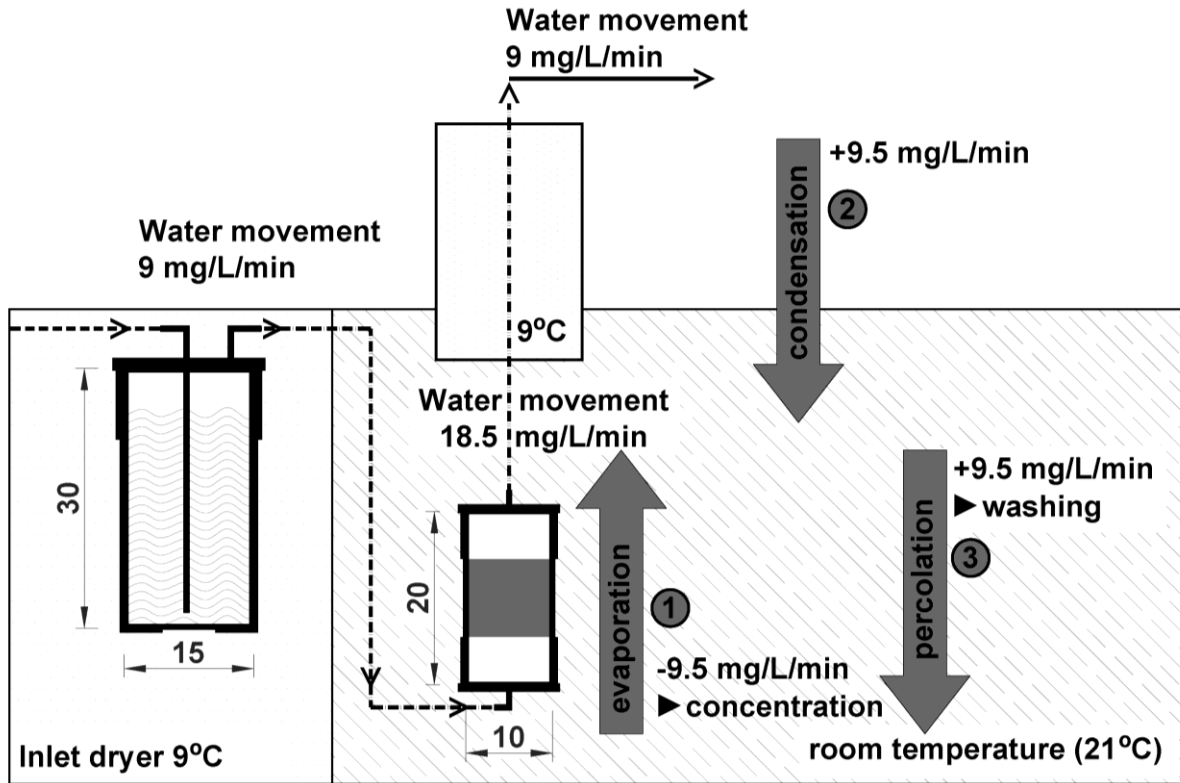


Figure 1: Schematic diagram of gas flow consisting of the three temperature zones. (1) Evaporation of water from the biofilter due to introducing gas containing lower water content. Causing the gas to gain moisture of $9.5 \text{ mg L}^{-1} \text{ min}^{-1}$. (2) Condensation of moisture in the gas by cooling to lower temperature with the result that $9.5 \text{ mg L}^{-1} \text{ min}^{-1}$ condenses and reintroduced into the biofilter. (3) $9.5 \text{ mg L}^{-1} \text{ min}^{-1}$ water percolates and thereby washing accumulated compounds to the bottom where the process begins again.

Employing Equation 3, the evaporated or condensed water content $\Delta_{w,c} [\text{kg m}^{-3}]$ between two temperatures (T_1 and T_2 [$^{\circ}\text{C}$]) can be calculated. A positive $\Delta_{w,c}$ indicates condensation; a negative $\Delta_{w,c}$ implies evaporation.

$$\Delta_{w,c} = WC_{T_1} - WC_{T_2}$$

Equation 3

The above reported mechanisms lead to the design and development of an inlet dryer (Figure 1) which adjusts the moisture content of the gas stream entering the biofilter. An installed outlet dryer (Figure 1) maintained the water content within the biofilter and prevented the escape of water. The outlet dryer operated on the same principle as the inlet dryer.

Proof-of concept experimental set up

The computer-controlled biofilter with reflux system was set up (Figure 2) based on the above design concept. The biofilter control system consisted of an inlet dryer, a biofilter column rig, an outlet dryer and an online gas analyser. The in-house compressed air supply was used as a carrier gas for the dilution of a 1% NH_3 in air (gas cylinder) to 50 ppm_v ($2.05 \text{ } \mu\text{mol L}^{-1} \text{ min}^{-1}$). Required parts were mounted to a metal frame to hold the biofilter columns, condenser, manifolds, solenoid valves, analogue flow meter (150 mm long, equipped with 16-turn high precision needle valves (4x $0\text{--}18.7 \text{ mL min}^{-1}$ and 4x $16737 \text{ mL min}^{-1}$)) and other electronic equipment (such as pH signal amplifier, thermocouples and wire). The system was controlled and monitored using an ADAM 4520 interface module (Advantech's ADAM) and Labview (8.5.1) software by National Instruments.

① Analogue flow meter with needle valves

② Reactor column

① Thermocouple*

② pH probes*

③ Digital flow meter*

*Computer controlled and connected

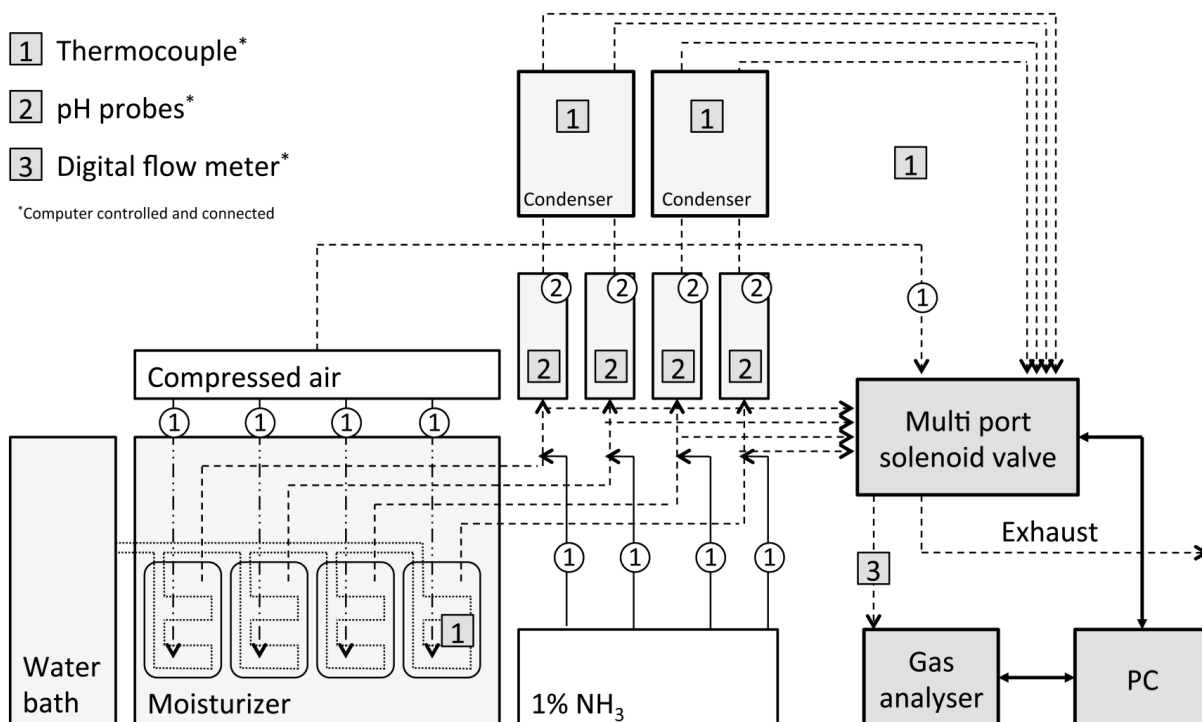


Figure 2: Flow chart of the biofilter system (operating four individual biofilter columns)

Biofilter column

An up-flow biofilter column was constructed as shown in Figure 3. A stainless steel mesh installed within the biofilter provided support to hold up to 1 L filter material and simultaneously provided 300 mL homogenization (mixing) space at the bottom. A pH probe was installed approximately 2.5 cm above the stainless steel mesh and connected to a data logging module for automated pH data acquisition. The biofilter column was operated at room temperature (21 ± 1 °C). Clinoptilolite (Zeolite Australia Pty. Ltd.) was used as a non-degradable filter media sieved to 2.4 – 4.0 mm. Prior to use the clinoptilolite was incubated at a local composting facility for 6 months. After that period, the clinoptilolite was harvested and carefully washed to clean the filter material from accumulated dust and organic material. One litre drained but moist clinoptilolite was placed within the biofilter column.

Inlet dryer

The inlet air (1 L min^{-1}), in-house compressed air supply, was sparged into the moisturizer vessel at the bottom of a chilled (9 °C) water reservoir Figure 1. The moisturizer vessel contained a maximum of 4.5 litre of deionised water (plus approximately 1 litre headspace). The moisturised air left the vessel at the top of the vessel before being mixed with known amount of 1 % NH₃ in air (gas cylinder) to give final concentration in air stream of 50 ppm_v NH₃. Due to the temperature difference between the introduced gas stream from the moisturiser vessel (chilled and saturated) and the biofilter column (see Figure 3) (operated at room temperature of 21 ± 1 °C), evaporation in the bottom layer occurred.

The saturation vapour pressure at a certain temperature can be calculated according to Tetens (1930). At 9 °C, a water content of 9 mg L^{-1} can be calculated under the assumption that the gas leaving the moisturizer is fully saturated. When this gas stream enters a water phase at a higher temperature, the water content per litre of gas will increase due to evaporation.

Under the assumption that the gas reaches saturation during passage through the moist biofilter (operated at 21 °C) an increase in water content of 9.5 mg L⁻¹ to a total 18.5 mg L⁻¹ is calculated. At a flow rate of 1 L min⁻¹ a total discharge of 13.75 mL d⁻¹ can be expected. Additional measurements of the biofilter moisture content showed total water content of 250 mL L⁻¹ biofilter material. Further calculations reveal that the biofilter would theoretically be dried out after 18 days unless the water is replenished.

Outlet dryer

To prevent water loss, a condenser was installed on top of the biofilter column and aligned with the outlet of each biofilter. A Tygon tube was vertically installed within the condenser which was operated at the same temperature (9 °C) as the inlet dryer (Figure 1). By cooling the gas in the PVC tube the moisture condensed on the tubing wall and dripped back into the biofilter. The water inventory of the biofilter remained. The percolation of the clean condensate through the biofilter allowed wash-down of soluble compounds accumulated over the depth of the biofilter. These compounds accumulated at the bottom where due to continuous introduction of dry gas (saturated at a lower temperature) the reflux process starts again and causes a cumulative increase in accumulated compound concentration.

Analysis

A HORIBA VA/ VS 3000 multigas analyser (HORIBA Ltd. 2004) equipped with nitric oxide (NO_x), nitrogen dioxide (N₂O) and NH₃ sensors was used for gas analysis. For analysis of the liquid surrounding clinoptilolite the 1 L biofilter media was manually separated into 200 mL lots (layers) every fortnight. Subsamples of 10 g of each homogenized layer were washed for 1 min in 10 mL deionised water. Eluents were syringe filtered (0.45 μm), frozen (below -20 °C) and stored until analysis. The liquid was analysed for Ammonium⁺ by using an AGILENT 1200 series HPLC with a “Universal Cation HR, 3 μm, 7.0 x 53 mm” column coupled to a conductivity detector (Alltech Model: 350). A mobile phase of 3 mM methanesulfonic acid was used at a flow rate of 2.5 mL min⁻¹. Nitrate and nitrite analysis were determined based on Standard Methods for the Examination of Water and Wastewater (APHA 2005).

RESULTS AND DISCUSSION

The biofilter with reflux system treating ammonia-contaminated air was operated for over 180 days. During that time no additional water was added and no leachate was produced. In general, ammonia removal mechanism in a biofilter starts with the dissolution of ammonia gas (NH₃) to NH₄⁺ in the water phase of the wet filter bed. Nitrifying bacteria (*Nitrosomonas* and *Nitrobacter*) then convert NH₄⁺ to nitrite (NO₂⁻) and nitrate (NO₃⁻). The metabolites will accumulate until the biofilter activity is inhibited due to high concentration. Flushing the metabolites out (by water irrigation) or replacing the filter material is required to prevent the inhibition.

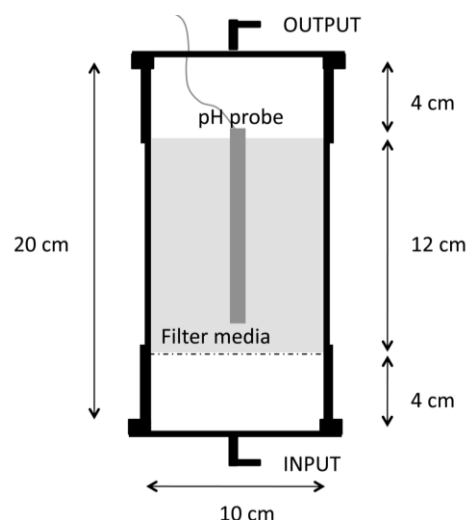


Figure 3: Schematic of the biofilter column design

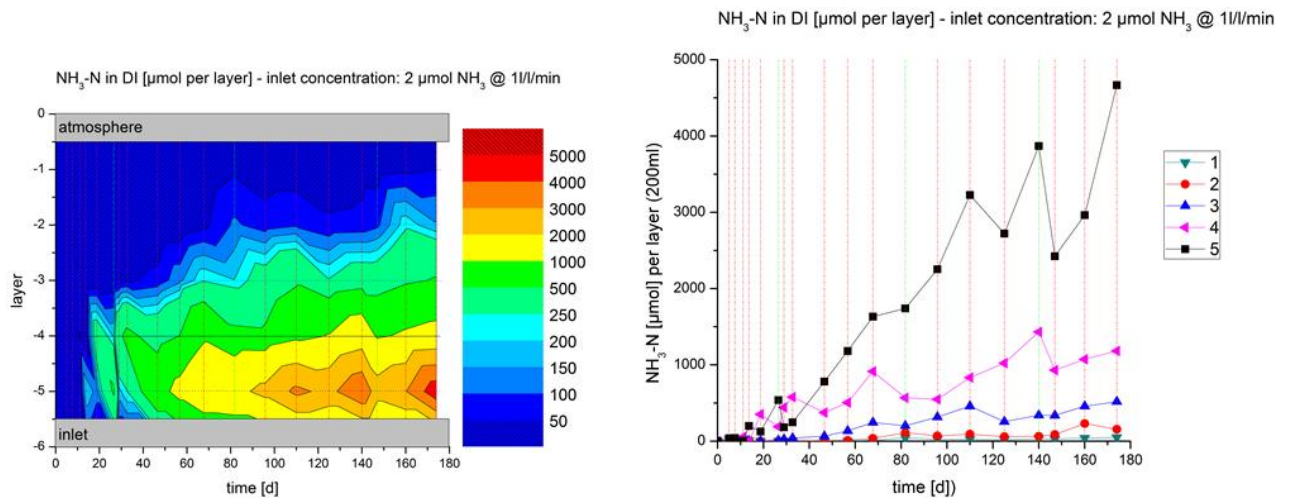


Figure 4: Biofilter depth profile for NH₄⁺ in 5 layers over time. Left: interpolated over depth and time presented as a contour plot and Right: a conventional line plot.

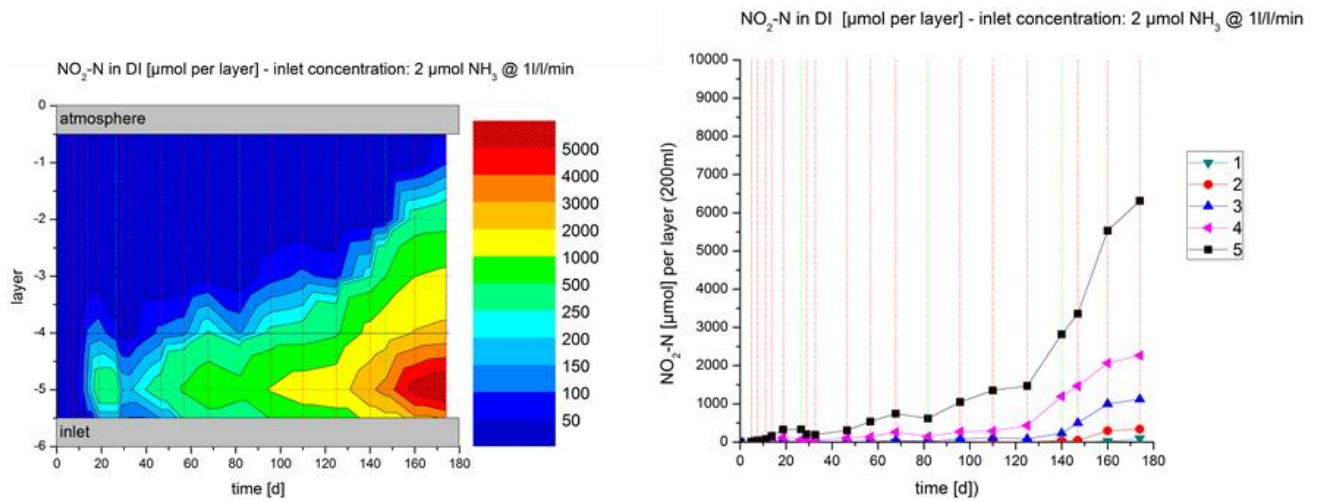


Figure 5: Biofilter depth profile for NO₂⁻ in 5 layers over time. Left: interpolated over depth and time presented as a contour plot and Right: a conventional line plot

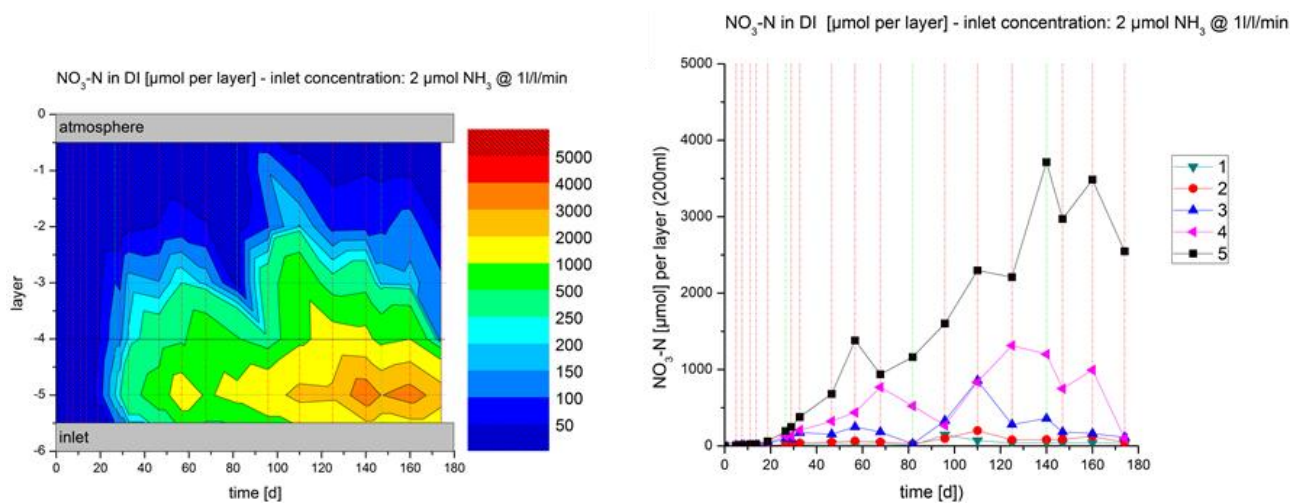


Figure 6: Biofilter depth profile for NO₃⁻ in 5 layers over time Left: interpolated over depth and time presented as a contour plot and Right: a conventional line plot

By operating the biofilter using reflux system, depth profile analysis of filter bed in Figure 4 to Figure 6 show clear vertical gradient of NH_4^+ and its metabolites (NO_2^- and NO_3^-) throughout the column. Over the period of 180-day experiment, close to 100% ammonia removal was achieved. NO_2^- and NO_3^- level continued to increase over the entire period of experiment and indicated that microbial conversion of NH_4^+ to oxidized metabolites had not been inhibited. The build up of NH_4^+ , NO_2^- , and NO_3^- was apparent at the bottom layer of biofilter and were above the inhibitory concentration (Anthonisen et al. 1976, Fdz-Polanco et al. 1994, Kim et al. 2003, Vadivelu et al. 2007), allowing no biological ammonium oxidation. In contrast, the concentration of NH_4^+ , NO_2^- , and NO_3^- was below detection limit in the top layer of the reactor column allowing microbial activity to be maintained at its most favourable level.

From the results, it can be postulated that the biological oxidation of NH_4^+ occurred in the top section, and the metabolites were washed down to the bottom of the biofilter with the introduction of the pure water to the top of the reactor. In this way, ammonia in air stream can be continuously treated at high efficiency through out the experimental period of 180 days without leachate production. Long-term operation is required to determine appropriate time and measure to remove concentrated pollutants at the bottom section of the biofilter.

This may be done by building the biofilter in modular section and simply removing the bottom section when full and replacing new section on the top, or by flushing the biofilter to remove the pollutant as high strength wastewater. A major operational cost is the energy to cool air. Theoretical energy requirement to cool 1 m³ of dry air from 21°C to 9°C is 0.0043 kWh. Actual cost involving heat exchange may be estimated at 0.017 kWh (at four times the theoretical figure). However, compared to conventional biofilter whereby leachate is continuously produced and requires treatment or disposal, the cost (inlet humidifier, water circulation pump, treatment and disposal of large quantity leachate, etc.) will be significantly reduced.

CONCLUSIONS

Based on the proof-of concept experiment, the following conclusions can be drawn;

1. By using reflux system, the cycle of vaporising and condensing the water in the biofilter caused (a) a stable biofilter moisture inventory and (b) a gravitational washing of compounds to the bottom of the biofilter
2. The addition of the reintroduced condensate caused a concentration gradient across the biofilter with (a) lower concentrations on top of the biofilter and (b) accumulation of the compounds at the bottom
3. Low concentrations in the upper layer preclude biological inhibition and favour biological reaction such as the nitrification of ammonium to nitrite and nitrate in this case
4. Accumulated pollutants in the bottom part of the biofilter can be removed as high concentrated low volume liquid waste which is generally easier to treat or dispose off.
5. Minimal or no leachate is produced.

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REFERENCES

- Anthonisen, A., Loehr, R., Prakasam, T., Srinath, E. (1976). Inhibition of nitrification by ammonia and nitrous acid. *Journal Water Pollution Control Federation*, 48(5), 835-852.
- American Public Health Association. (APHA). (2005). *Standard Methods for the Examination of Water and Wastewater*. Washington, D.C.
- Baquerizo, G., Maestre, J. and Machado, V. (2009). Long-term ammonia removal in a coconut fiber-packed biofilter: Analysis of N fractionation and reactor performance under steady-state and transient conditions. *Water Research* 43, 2293-2301.
- Eiserbeck, C., Nelson, R.K., Grice, K., Curiale, J. and Reddy, C.M. (2012). Comparison of GC-MS, GC-MRM-MS, and GC × GC to characterise higher plant biomarkers in Tertiary oils and rock extracts. *Geochimica et Cosmochimica Acta*, 87, 299-322.
- Fdz-Polanco, F., Villaverde, S., Garcia, P. (1994). Temperature effect on nitrifying bacteria activity in biofilters: activation and free ammonia inhibition. *Water Science and Technology*, 30(11), 121-130.
- HORIBA Ltd. (2004) HORIBA VA/VS - 3000 manual, pp. 1-61.
- Jun, Y, and Wenfeng, X. (2009). Ammonia biofiltration and community analysis of ammonia-oxidizing bacteria in biofilters. *Bioresource Technology* 100(17), 3869-76.
- Kim, J.H., Rene, E.R. and Park, H.S. (2008). Biological oxidation of hydrogen sulfide under steady and transient state conditions in an immobilized cell biofilter. *Bioresource Technology* 99(3), 583-88.
- Kim, D., Chang, J., Lee, D., Han, D., Yoo, I., Cha, G. (2003). Nitrification of high strength ammonia wastewater and nitrite accumulation characteristics. *Water Science and Technology*, 47(11), 45-51.
- Lamb, A., Ohl, E. (1935). Heats of Adsorption of Gases and Vapors upon Crystallogenic Adsorbents. *Journal of the American Chemical Society*. 57(11), 2154-2161.
- Nisola, G.M., Cho, E., Orata, J.D., Redillas, M.C.F.R., Farnazo, D.M.C., Tuuguu, E., Chung, W.J. (2009). NH₃ gas absorption and bio-oxidation in a single bioscrubber system. *Process Biochem*, 44(2), 161-167.
- Pandey, R., Mudliar, S., Borgaokar, S. (2009). Treatment of waste gas containing diethyldisulphide (DEDS) in a bench scale biofilter. *Bioresource Technology*, 100(1), 131-135.
- Sakuma, T., Hattori, T., Deshusses, M.A. (2009). The effects of a lower irrigation system on pollutant removal and on the microflora of a biofilter. *Environmental Technology*, 30(6), 621-627.
- Tetens, O. (1930). Über einige meteorologische Begriffe. *Zeitschrift Geophysik*, 6, 297-309.
- Vadivelu, V., Keller, J. and Yuan, Z. (2007). Effect of free ammonia on the respiration and growth processes of an enriched Nitrobacter culture. *Water Research*, 41(4), 826-834.
- Wexler, A. 1976. Vapor pressure formulation for water in the range 0° to 100°C. *Journal of Research of the National Bureau of Standards*, 80A, 775-780.