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Nutrient addition in biofiltration for air pollution control

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Abstract— A soil biofilter, with internal gas recycle and a suction cell, was used in this study to investigate the influence of nutrient addition on the volumetric removal rates of toluene (as air contaminant) and on the concomitant carbon dioxide production. The use of a suction cell allowed water content control and uniform distribution of nutrients throughout the filter bed medium. Nutrient addition increased the elimination capacity by about five times upon the second addition of nitrate as nitrogen source and the subsequent addition of micronutrients. However, this resulted to the formation of a gelatinous substance in the soil bed. This study gives evidence that calcium accumulation in biofilms plays a significant role in cross-linking extracellular polymeric substances (EPS), components that may affect long-term operation of the biofiltration system. Adding nutrients and maintaining their concentrations at a level that contributes favorably to biofiltration performance continues to be a challenge in vapor-phase biofiltration.

Keywords— biofiltration, nutrient, air pollution control, elimination capacity

I. Introduction

Biofltration is an air pollution control technology where contaminants in a gas stream are metabolized by microorganisms and converted to water, carbon dioxide and biomass [1]. It is often favored over other air pollution control methods since it does not produce secondary pollutants and does not involve expensive operating and maintenance costs. However, reliability of the process is only possible when the biofiltration system is properly designed and operated. Problems such as biomass clogging, nutrient depletion and water content control hinder installation and use of vaporphase biofilters [2,3].

The main factors that play major roles in microbial growth are temperature, water availability, pH, and oxygen [4]. The water in the filter bed medium is essential for microbial growth and for transport of nutrients [5]. Nutrient availability affects biofilm development and it can regulate the structure of a mature biofilm where microorganisms degrade the pollutants [6].

Nitrogen is very important in biofiltration. It (13%) is the second most common element or compound in bacterial cell mass after carbon (50%) and excluding water [7,9].

Biofiltration performance is strongly related to nitrogen availability whether as nitrate or ammonia [8,10,11] Nutrient studies in a biofilter with water content control was done by Beuger and Gostomski [12] using a compost biofilter where the addition of 1.0 gl⁻¹ NaNO₃ solution almost doubled the elimination capacity and the addition of 1.0 gl⁻¹ NH₄Cl increased the elimination capacity almost seven times. Higher elimination capacity was also obtained by Smith et al. [13] and Jorio et al. [14] when using nitrate addition compared to ammonia addition. According to Smith et al. [13], use of nitrate over ammonia for nitrogen addition is preferable because of nitrate's lower biomass yield. In this study, a soil biofilter with internal gas recycle and a suction cell for water content control was used to investigate the influence of nutrient addition on biofiltration performance. The use of a suction cell allowed water content control and uniform distribution of nutrients throughout the filter bed medium. Ideally, the addition of nutrients should maintain the amount of biomass and not promote growth [9]. Surplus biomass can clog the bed and lower the performance [15].

The aim of this study was to investigate the influence of nitrate as nitrogen source and of micronutrients such as calcium, magnesium and iron ions on the volumetric removal rates of toluene as air contaminant. The concomitant carbon dioxide production (PCO₂) was also used as a simple method for assessing the overall activity of the soil microbial community [16] which is crucial in biofiltration. PCO₂ may be used to measure biofiltration performance where a high value confirms effective biodegradation [17].

п. Materials and Methods

A. Biofilter Reactor

A biofilter reactor with water content control developed by Beuger and Gostomski [12] was used in this study (Figure 1) with the following biofilter operating conditions: temperature at 30°C, average flow rate of 26 (\pm 4) ml/min, and average inlet concentration of 223 (\pm 27) ppm of toluene.

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The head space of the reactor was well mixed exposing all of the soil as the medium to a uniform toluene concentration (i.e. CSTR). A suction cell was used to control and manipulate the water content in the soil bed.

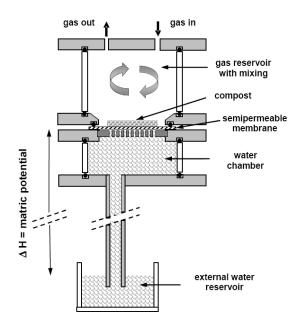


Figure 1 A cut-away of the biofilter

B. Suction Cell

In a suction cell [18], the soil is hydraulically connected to a water reservoir by a semi-permeable membrane (permeable to water but not to air). A vacuum is applied to the water reservoir through the hanging water column between the water chamber and the external reservoir. The matric potential of the soil equilibrates with the vacuum applied to water reservoir causing water to move in and out of the pore space of the soil. The suction is changed by changing the height of the water reservoir (Δ H) in relation to the barrier or membrane. The suction cell not only controls water content. It also allows uniform distribution of nutrients throughout the filter bed medium.

c. Sampling and Analytical Methods

Gas samples (five samples of 0.2 ml) were collected once daily at the inlet and at the outlet sampling ports of the reactor using a 1 ml gas tight syringe (SGE). The concentration of the contaminant in the gas stream was measured using gas chromatography (Varian CP-3800) with a flame ionization detector with a capillary column (Chrompack Cp-Sil 5 CB) and helium as the carrier gas. The temperature of the injector, oven and detector were 220°C, 180°C and 200°C, respectively. Carbon dioxide released at the outlet sampling port was measured daily using a CO₂ analyzer (Vaisala GMP343) which was connected to the sampling port during sampling period for about 30 minutes.

D. Calculation of Parameters

Volumetric removal rate of toluene was measured in terms of elimination capacity (EC) and carbon dioxide production (PCO₂) both in g $m^{-3}h^{-1}$. EC is the mass of contaminant degraded and PCO₂ is the mass of carbon dioxide produced, both per unit volume of filter material per unit time.

E. Nutrient Addition

The concentration of the nutrients such as nitrates, magnesium and ferrous sulfates, and calcium chloride with the duration of their application at each stage in the experiment are shown in Table 1.

TABLE 1. Details of the experiment

Stage	Nutrient	Concentration $(g l^{-1})$	Duration (days)
Ι	H ₂ O		39
II	NaNO ₃	1.0	23
III	H ₂ O		19
IV	$KHPO_4$	0.8	25
	MgSO ₄ ·7H ₂ O	0.4	
	FeSO ₄ ·7H ₂ O	0.0035	
	$CaCl_2 \cdot 2H_2O$	0.02	
V	NaNO ₃	0.5	33

The different nutrient solutions were autoclaved at 120°C for 20 minutes.

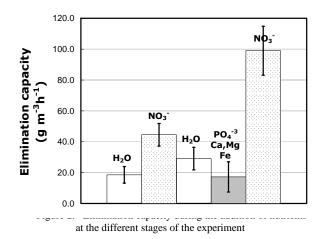
III. Results and Discussion

A. Elimination Capacity

Results of this study (Fig. 2) showed addition of a nitrate solution (1.0 g l^{-1}) increased the elimination capacity (EC) by 140% from 18.5 g m⁻³h⁻¹ in stage I without nutrients to 44.5 g m⁻³h⁻¹ in stage II where the first addition of nitrate was made.

The study of Beuger and Gostomski [12] of toluene removal in a compost biofilter resulted to a more significant increase in EC (7 to 76 g m⁻³h⁻¹) where nitrogen was identified as limiting the biomass growth. Several other studies show the same results of increase in EC upon nitrate addition [8,13,14] and decrease in pollutant removal capacity on nutrient removal or limitation [19].





Furthermore, when the nitrate supply in this study was removed after 23 days, the EC decreased (stage III) by 35%. The study of Song et al. [10] indicated that even under nitrogen-rich conditions, recycling of nitrogen occurred in the biofilm and the fraction of nitrogen demand met by recycling nitrogen increased when the external supply of nitrogen was terminated, and the biofilm became nitrogen limited. When nitrate was supplied again, the biofilm possibly grew thicker and started spreading and when it was taken away it stopped growing (stage I vs II). Non-growth dropped the volumetric consumption rate but left it higher than before growth (stage I vs III).

The increased EC may be explained by the high growth kinetics experienced by the dominant toluene degrading species under high nitrogen concentrations. On the other hand, nutrient limitation reduced microbial growth and resulted to decreased removal rate of the contaminant.Depending on the availability of nutrients, a significant portion of the carbon in the biofilm phase may become inactive and unable to contribute to net growth of active microbial cells [19].

In stage IV, the addition of a solution of phosphate and micronutrients such as calcium, magnesium and iron ions further decreased the EC by 41%. This difference in EC between stage III and IV might not be statistically significant as shown in the error bars (stage I vs. stage IV in Fig. 2). In the study of Beuger and Gostomski [12], there was also no significant change in EC when the same nutrient solution was added.

Interestingly, a second addition of nitrate solution in stage V with half the previous concentration (from 1.0 to 0.5 g 1^{-1}) increased the EC by about five times reaching 100 g m⁻³h⁻¹. More interestingly, as suggested by Delhomenie et al. [7], in line with theoretical considerations, a maximum level of EC at 100 g m⁻³h⁻¹ requires an optimal nitrogen concentration of \approx 2.6 g of N·L⁻¹. The addition of calcium, magnesium and iron salts, though did not increase the EC as they were added, appeared to have influenced the high increase in EC during the

second addition of the nitrate solution. One possible factor may have been the calcium action proposed as a general reset mechanism of cells that allows them prompt and effective response to new situations [20]. Calcium plays important roles in cell cycle and cell division in bacteria, initiation of DNA replication, chromosome partition and cell division [21,22], and in enzyme reactivation [23]. Korstgens *et al.* [24] stated in their study that the observed calcium accumulation in the biofilms plays a significant role in cross- linking/bridging extracellular polymeric substances (EPS) components in the biofilm. In this study, a gelatinous substance was formed in the soil filter bed This gelatinous cover could possibly be due to the presence of calcium.

Iron ions are also essential in biofiltration. Fe limitation and Fe excess both adversely affect biofilm formation in *P. aeruginosa*. In the chemostat study of Dinkla et al. [25], the incomplete removal of toluene observed at low iron/toluene ratios resulted from reduced performance of the cells in terms of degradation kinetics. In this study, the effect of the addition of iron may have contributed to the effects of calcium and magnesium discussed earlier but the addition of these micronutrients still needs to be studied further.

Soil bacterial cells are often embedded in EPS which contributes to a successful adaptation to variations in water content and in nutrient availability as entrapment is enhanced [26]. The amount of EPS synthesis within the biofilm greatly depends on the availability of carbon substrates and on the balance between carbon and other limiting nutrients. In this study, the nutrient addition increased the EC significantly by about five times upon the second addition of nitrate as nitrogen source and the subsequent addition of micronutrients. However, this resulted to the formation of a gelatinous substance in the soil bed. This study possibly gives evidence that calcium accumulation in biofilms plays a significant role in cross-linking EPS), components that may affect long-term operation of the biofiltration system. According to Sutherland [27], the presence of excess available carbon substrate and limitations in other nutrients, such as nitrogen, potassium or phosphate promotes the synthesis of EPS and the slow bacterial growth observed in most biofilms is expected to enhance EPS production.

B. Carbon Dioxide Production

The experimental and the theoretical CO_2 production (PCO₂) rate (in g m⁻³h⁻¹) may be compared easily using Figure 3. The diagonal line marks 100% recovery at which the CO_2 produced experimentally equals the calculated value based on stoichiometry. As the values fall close to the diagonal line, it shows nearly all of the toluene removed has been converted to CO_2 except when the solution containing calcium, magnesium, iron, and phosphates were added and during the second addition of the nitrate solution where there is a discrepancy (about 20 gm⁻³h⁻¹) between carbon from toluene removed and from CO_2 produced. The "lost" carbon may be in dissolved



forms such as carbonates and bicarbonates [28,29] or possibly used in the production of biomass, especially at high toluene concentration [30]. Results in Fig. 3 confirm the low CO_2 recovery during the second addition of nitrate. It also shows the high CO_2 recovery during the addition of the solution containing phosphates and calcium, magnesium and iron ions due to endogenous respiration. The possible influence of magnesium and iron has not been distinctly manifested.

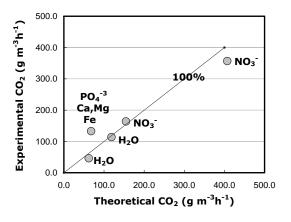


Figure 3. Experimental vs. theoretical carbon dioxide production

In the study of Ben, et. al. 2014 [31], the influence of nutrients on biodegradation was clearly observed in the removal of dimethyl disulfide and ethanethiol but not in the removal of hydrogen sulfide as it was not affected by the lack of nutrients. Similarly, the study of Lee, et. al. 2012 [32] showed the absence of NO_3^- concentration in the perlite media achieved higher ethylene removal efficiencies than the other media and with nitrate. However, a nitrate concentration as high as 2 g L⁻¹ in the original nutrient solution was seen to inhibit the growth or activity of ethylene degraders.

IV. Conclusions

Nutrient addition improved the volumetric removal rate of toluene as it promotes biomass growth. The production of carbon dioxide confirmed effective biodegradation of toluene in the soil biofilter. However, the type of filter media and the concentration of the nutrient affect biofiltration performance. This study appears to give evidence that calcium accumulation in biofilms plays a significant role in cross-linking EPS components and may cause formation of a gelatinous cover in the bed medium which may affect long-term operation of the biofilter as mass transfer of contaminants and nutrients is hindered. Adding nutrients and maintaining their concentrations at a level that does not inhibit biofiltration performance continues to be a challenge in vapor-phase biofiltration.

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References

- J. S. Devinny, M. A. Deshusses and T. S. Webster, "Biofiltration for Air Pollution Control". Boca Raton, FL, USA: Lewis Publishers. CRC Press LLC. 1999.
- [2] C. van Lith, G. Leson and R. Michelsen, "Evaluating design options for biofilters," J. Air Waste Manage. Assoc. vol. 47, pp. 37-48, 1997.
- [3] W. F. Wright, E. D. Schroeder, D. P. Y. Chang and K. Romstad, "Performance of a pilot-scale compost biofilter treating gasoline vapor", J. Environ. Eng. vol. 123, pp. 547-555, 1997.
- [4] . M. Madigan, M. T. Martinko and J. Parker, "Biology of Microorganisms". (8th Ed.) Prentice-Hall Inc. NJ., 1997.
- [5] P. A. Holden and N. Fierer, "Microbial processes in the vadose zone", Vadose Zone J. vol. 4, pp. 1-21, 2005.
- [6] G. C. Goller and T. Romeo, "Environmental influences on biofilm development", Curr. Top. Microbiol. Immunol. vol. 322, pp. 37-66, 2008.
- [7] M. C. L. Delhomenie, S. Bibeau, R. R. Brzezinski and M. Heitz, "Influence of nitrogen on the degradation of toluene in a compost-based biofilter", J. Chem. Technol. Biotechnol, vol. 76, pp. 997-1006, 2001.
- [8] M. C. Delhomenie, L. Bibeau, J. Gendron, R. Brzezinski and M. Heitz, "Toluene removal by biofiltration: Influence of the nitrogen concentration on operational parameters", Ind. Eng. Chem. Res. vol. 40, pp. 5405-5414, 2001.
- [9] E. Morgenroth, E. D. Schroeder, D. P. Y. Chang and K. M. Scow, "Nutrient limitation in a compost biofilter degrading hexane", J. Air Waste Manage. Assoc. vol. 46, pp. 300–308, 1996
- [10] J. Song, J., Ramirez and K. A. Kinney, "Nitrogen utilization in a vaporphase biofilter", Water Res. vol. 37, pp. 4497-4505, 2003.
- [11] R. L. Corsi and L. Seed, "Biofiltration of BTEX: Media, substrate, and loadings effects", Environ. Prog. vol. 14, pp. 151–158, 1995.
- [12] A. L. Beuger and P. A. Gostomski, "Development of a biofilter with water content control for research purposes", Chem. Eng. J. vol. 151, pp. 89-96, 2009.
- [13] F. L. Smith, G. A. Sorial, M. T. Suidan, A. W. Breen and B. Pratim, "Development of Two Biomass Control Strategies for Extended, Stable Operation of Highly Efficient Biofilters with High Toluene Loadings", Environ. Sci. Technol. vol. 30, pp. 1744-1751, 1996.
- [14] H. Jorio, R. Bibeau, G. Viel and M. Hetiz, "Effect of gas flow rate and inlet concentration on xylene vapors biofiltration performance", Chem. Eng. J. vol. 76, pp. 209–221, 2000.
- [15] C. Alonso, M. T. Suidan, G. A. Sorial, F. L. Smith, P. Biswas, P. J. Smith and R. C. Brenner, "Gas treatment in trickle-bed biofilters: Biomass, how much is enough?", Biotechnol. Bioeng. vol. 54, pp. 583–594, 1997.
- [16] R. L. Tate, III, Soil Organic Matter. Biological and Ecological Effects. Krieger Publishing Company, Florida, 1992.
- [17] K. Chang and C. Lu, "Biofiltration of toluene and acetone mixtures by a trickle-bed air biofilter", World J. Microbiol. Biotechnol. vol. 19, pp. 791-798, 2003.
- [18] M A. Ranasinghe and P. A. Gostomski, "A novel reactor for exploring the effect of water content on biofilter degradation rates", Environ. Prog. vol. 22, pp. 103-109, 2003.



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- [19] J. Song and K. A. Kinney, "Effect of vapor-phase bioreactor operation on biomass accumulation, distribution, and activity: linking biofilm properties to bioreactor performance", Biotechnol. Bioeng. vol. 68, pp. 508–516, 2000.
- [20] V. Norris, S. Grant, P. Freestone, J. Canvin, N. Sheikh, I. Toth, M. Trinei, K. Modha and R. I. Norman, "Calcium Signalling in Bacteria", J. Bacteriol. vol. 187, pp. 3677–3682, 1996.
- [21] V. Norris, "A calcium flux at the termination of replication triggers cell division in Escherichia coli", Cell Calcium, vol. 10, pp. 511-517, 1989.
- [22] R. J. Smith, "Calcium and bacteria", Adv. Microbiol. Physiol. vol. 37, pp. 83–103, 1995.
- [23] D. J. Hopper and M. A. Kaderbhai, "The quinohaemoprotein lupanine hydroxylase from Pseudomonas putida", Biochimica et Biophysica Acta, vol. 1647, pp. 110-115, 2003.
- [24] V. Körstgens, H. C. Flemming, J. Wingender and W. Borchard, "Influence of Calcium Ions on the Mechanical Properties of a Model Biofilm of Mucoid Pseudomonas aeruginosa", Water Sci. Technol. vol. 43, pp. 49-57, 2001.
- [25] I. J. T. Dinkla, E. M. Gabor and D. B. Janssen, "Effects of Iron Limitation on the Degradation of Toluene by Pseudomonas Strains Carrying the TOL", (pWWO) Plasmid, Appl. Environ. Microbiol. vol. 67, pp. 3406-3412, 2001.

- [26] D. Or, S. Phutane, and A. Dechesne, "Extracellular Polymeric Substances Affecting Pore-Scale Hydrologic Conditions for Bacterial Activity in Unsaturated Soils", Vadose Zone J. vol. 6, pp. 298–305, 2007.
- [27] I. W. Sutherland, "Biofilm exopolysac- charides: A strong and sticky framework", Microbiol. vol. 147, pp. 3–9, 2001.
- [28] H. Jorio, K. Kiared, R. Brzezinski, A. Leroux, G. Viel and M. Heitz, "Treatment of air polluted with high concentrations of toluene and xylene in a pilot-scale biofilter", J. Chem. Technol. Biotechnol. vol. 73, pp. 183-196, 1998.
- [29] R. S. Singh, B. N. Rai and S. N. Upadhyay, "Performance evaluation of an agrowaste based biofilter treating toluene vapours", Environ. Technol. vol 27, pp. 349-357, 2006.
- [30] T. Sakuma, T. Hattori, and M. A. Deshusses, "The effects of a lower irrigation system on pollutant removal and on the microflora of a biofilter", Environ. Technol. vol. 30, pp. 621-627, 2009.
- [31] M. B. Jaber, B. Anet, A. Amrane, C. Couriol, T. Lendormi, P. L. Cloirec, G. Cogny and R. Fillières, "Impact of nutrients supply and pH changes on the elimination of hydrogen sulfide, dimethyl disulfide and ethanethiol by biofiltration", Chem. Eng. J. vol. 258, pp. 420-426, Dec. 2014.
- [32] Lee, S.H., C. Li and A.J. Heber, "The effect of nitrate on ethylene biofiltration", J. Hazard. Mater. vols. 241-242, pp. 331-339, Nov. 2012.

