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Reducing Nitrous Oxide Emission Using Goat Manure-Compost Based Biofilter

Tania Surya Utami, Lila Adriaty, Heri Hermansyah and M. Nasikin

Chemical Engineering Department, Faculty of Engineering,
University of Indonesia Campus, UI Depok, Indonesia

Abstract: Biofiltration is the latest technology of pollution control for removing N_2O with compost as medium filter. This technology has many advantages, such as low installation and operation cost, secure operation, low energy consumption, good stability and able to remove pollutant with high efficiency. Effects of N_2O flow rate, water content and use of natural and synthetic nutrient supplement in compost which is added by *Nitrobacter, sp.* will be investigated towards N_2O gas removal efficiency for 9 hours in batch system. Decreasing concentration of N_2O was analyzed with Gas Chromatography (GC) and increasing quantity of microorganism in compost as filter medium was analyzed with Total Plate Count (TPC). The result indicates that the highest N_2O gas removal efficiency is obtained under biofilter length of 50 cm, gas flow rate of 72.02 cc/min and 60% water content. The result shows that N_2O gas removal efficiency could be optimized by adding synthetic nutrient supplement to compost which has been mixed with *Nitrobacter, sp.*, with 75.9% of removal efficiency. Results of TPC show increasing bacteria population after biofiltration process.

Key words: Biofilter · Compost · N_2O, *Nitrobacter* · *sp.* · Removal efficiency

INTRODUCTION

Nitrous oxide (N_2O) is a by-product of fixed nitrogen application in agriculture. N_2O can be emitted from agricultural soils or can be deposited on non-farm land and biochemically converted and emitted into the atmosphere. N_2O has a global warming potential (GWP) 310 times that of CO_2 (US Dept. of Energy, 2007), which means 1 kg of N_2O has the same global warming impact as 310 kg of CO_2. Agriculture is responsible for over 50% of global N_2O emissions. N_2O is an important greenhouse gas (GHG) because it has a global warming potential 310 times than that of CO_2 and contributes to the destruction of stratospheric ozone (O_3). N_2O is a powerful and long-lived greenhouse gas, it occurs naturally in the environment, but the concentrations are now rising, mainly as a result of fertilizer used and composting process becomes an alternative for organic waste treatment.

Biofiltration and biotrickling filtration is biological technology which very often used in air pollution control [1, 2]. Biofiltration is a process of gas pollutant treatment in a medium bed where pollutant will be degraded by microorganism, while in biotrickling filtration there is liquid dripped by above medium bed to produce optimum condition at biofilm [3]. In biofilter, pollutant will be transferred directly from biofilm, while in biotrickling filter transfer of pollutant conducted through the liquid dripped by above filter medium. Technology of biological treatment of gas emission, in this case biofiltration, is preeminent in some aspects: safe operating condition that does not need high cost in the provision of microorganism involved, high removal efficiency of pollutant, low energy consumption, simple equipments design, practically no undesirable byproducts and oxidized organic and inorganic compounds are converted to nonhazardous compounds.

In some researches, biofilter systems proven to be effective in eliminating odors, ammonia and Volatile Organic Compound (VOC) are: benzene biofiltration with compost medium [4], styrene biofiltration [5], phenol biofiltration utilizing microorganism *Pseudomonas putida* [6], hexane biofiltration with compost medium [7], odors biofiltrations in piggery [8] and toluene and styrene biofiltration from the air [9].

Nowadays, researches in biofilter focus in the reduction of NH_3 and H_2S. Only a few researchers interested to apply it in the reduction of NOx. To support further application, studies in laboratory and pilot scale are needed to optimize the operating parameters, especially the influence of type of medium co-material, input gas flow rate, medium humidity, nutrition, etc. The

Correspoding Author: Tania Surya Utami, Chemical Engineering Department, Faculty of Engineering, University of Indonesia, Campus UI Depok, 16424, Tel: +6221-7863516, Fax: +6221-7863515, E-mail: nana@che.ui.ac.id.

objectives of this research are to develop biofilter system in reducing N_2O emission simultaneously by utilizing compost as filter medium, to determine biofilter operating parameters, to evaluate the ability of biofilter in reducing N_2O and to develop microorganisms in medium.

MATERIALS AND METHODS

The first phase of this research is designing biofilter system according to the research requirement and based on the information obtained from literature. Flow regulation of biofilter system in this research is batch system using flow meter to regulate the flow rate at a certain speed of flow. Input flow rate of N_2O is regulated as minimum as possible so that N_2O gas residence time in the filter medium is longer and N_2O gas concentration which is adsorbed by the filter medium becomes maximum. Schematic diagram of biofilter used in this research is shown in Figure 1 below. Design and selection of materials are conducted by considering preventive aspect of leakage, so that it is achieved the most possible effective design [4]. Biofilter column is designed using acrylic material and pipe line made of stainless steel with minimal extensions. Two (2) sampling ports are positioned on the top and the bottom of the column for gas sampling before and after biofiltration process.

The next phase is compost preparation as filter medium. Filter medium used in this process is compost. The compost used is from Green-Lab., Sekolah Alam Indonesia. The compost consists of goat manure mixed with domestic waste, peat and coconut peat. Before used as filter medium, the compost is going through a drying and screening process. Drying performed at room temperature (27°C) and 70% humidity and continues with screening. Diameter of sieve used is approximately 1-1.5 mm (100 mesh) to get uniform particle size of compost in order to prevent pressure drop.

This research also investigates the effect of natural and synthetic nutrient in the medium of biofilter on the reduction of air contamination. Nutrient supplement is provided to the biofilter as the source of carbon (glucose), inorganic nutrients and moisture. 40 mL of synthetic nutrient is added to the medium. The nutrient solution (pH 8.0) contains the following components (in 1 L of H_2O): K_2HPO_4 (0.4 g), KH_2PO_4 (0.15 g), NH_4Cl (0.3 g), $MgSO_4 \cdot 7H_2O$ (0.4 g), sodium acetate 2.93 g and 2 mL of trace element solution consisting of (in 1 L of H_2O): EDTA (50.0 g), $ZnSO_4 \cdot 7H_2O$ (2.2 g), $CaCl_2 \cdot 2H_2O$ (5.5 g), $MnCl_2 \cdot 4H_2O$ (5.06 g), $FeSO_4 \cdot 7H_2O$ (5.0 g), $(NH_4)6Mo_7O_{24} \cdot 2H_2O$ (1.1 g), $CuSO_4 \cdot 5H_2O$ (1.57 g) and

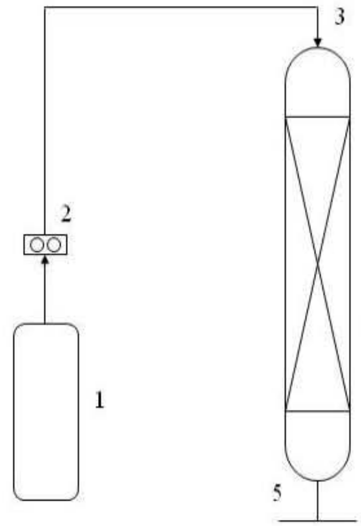

Fig. 1: Schematic diagram of laboratory scale biofilter. (1: N_2O gas supply, 2: Flow meter, 3-5: Sampling port, 4: Biofilter column)

$CoCl_2 \cdot H_2O$ (1.61 g). These inorganic materials are selected because they have already been used to grow aerobic nitrifying bacteria [4]. Natural nutrient supplement is liquid waste from cow husbandry in Kukusan, Depok, Indonesia.

The third phase is biofiltration process optimization through evaluation of the ability of biofilter system in reducing N_2O concentration. Optimization process conducted in batch system, focusing on some operating parameters, such as N_2O flow rate, influence of water content in filter medium and use of nutrition against biofilter adsorption. The change of microorganism number in compost medium before and after biofiltration process is also evaluated. Data is then collected by paying attention to 2 statistical important principles, i.e. randomization and repetition.

N_2O concentration measured using Gas Chromatography (GC, Shimadzu, Poropak Q column, detector type TCD), with injector temperature 60°C and detector temperature 100°C and carrier gas He. The effluent is analyzed every hour for 9 hours. The results of biofiltration experiment is summarized and reported. Data are obtained from N_2O peak area which will be plotted towards time. The N_2O removal efficiency can be obtained using the formula below:

$$N_2O\ removal\ efficiency\ at\ t_n = 100\% \times \frac{A_{t=0} - A_{t=tn}}{A_{t=0}} \quad (1)$$

Where: $A_{t=0}$ = N_2O peak area at t = 0 hours ; $A_{t=tn}$ = N_2O peak area at t = n hours

Analysis is also conducted to compost medium in order to know the change of microorganism colony number in the medium with TPC (Total Plate Count) method before and after biofiltration process.

RESULTS AND DISCUSSION

The Effects of Gas Flow Rate: The graph in Figure 2 shows that longer retention time between the compost and N_2O gas will affect the concentration of N_2O taken from the lower biofilter column. Figure 2 also shows that lower gas flow rate gives higher N_2O removal efficiency. Figure 2 shows that at t = 1-2 hours the gas flowing through the medium is still not stable. It is also shown in Figure 2 that the removal efficiency profile for the flow rate of 127.22; 185.74; 232.89 cc/minute have fluctuation at t = 1 hour and t = 2 hours. Since the flow rate in this biofiltration is very high, the residence time on the filter media will be short. This can also occur due to the dry compost used, which will facilitate the flow of N_2O. Dry compost may have less moisture content, which will cause channeling [5].

Figure 3 showed that the highest N_2O removal efficiency occurred when N_2O gas flow rate is 72 cc/min with removal efficiency 56.7%. N_2O removal efficiency tends to increase when the flow rate is lower because N_2O gas residence time in the filter medium is longer and the contact time between N_2O gas and biofilter medium is also longer. Consequently, the intensity of N_2O gas undergoing adsorption and degradation processes is greater when the N_2O gas flow rate is lower. In line with the above explanation, N_2O gas removal efficiency is also greater when the contact time between the compost and N_2O gas is longer. Furthermore, if it is associated with the adsorption curve in general, the concentration of adsorbate will decrease because the adsorbate is adsorbed by the adsorbent, so that the removal efficiency of N_2O gas as a contaminant (adsorbate) will increase when the flow rate is lower. Accordingly, it can be concluded based on the bar graph in Figure 3 that the smaller flow rate, the greater N_2O removal efficiency.

The Effects of Water Content: This experiment aims to find out the effects of water content in the compost medium on the reduction of N_2O. Addition of water is intended to increase the humidity of filter medium, which will affect the microbial growth in it. Water content is varied from 30%, 40%, 50%, 60% and 70% (w/w). Variation in the water content 30-70% is based on previous studies investigating the effects of reduction of moisture content on reduction of other pollutants [6]. The height of the filter medium used is 50 cm and uses the lowest flow rate (72 cc/min).

Figure 4 presents the concentration profiles of N_2O with the variation of water added to the filter. The Figure above also illustrates the trend of the profile with three segments: up, down and up again slowly. When the experiment is carried out and started (t = 0), it is assumed that N_2O gas concentrations in the reactor is close to zero so that the N_2O gas output is also considered zero. This is why N_2O removal efficiency equals to zero. However, at t = 1 hour, N_2O has not been detected, whereas at t = 2 hours there is a gas containing N_2O with very small concentration, which then increases until a certain time (6-7 hours). N_2O gas residence time in filter medium is longer because humidity of biofilms increases, which makes the medium porosity smaller.

It can be seen in Figure 5 that the highest removal efficiency is 70.13% with moisture content 60% (w/w). In this process, humid biofilms have bigger effects on N_2O removal efficiency if compared to dry compost. Biofilms on the particles of filter medium is aerobic area and contains water and microbes for degradation of pollutants. Moreover, biofilms provide essential nutrients for biological activity and maintain moisture for bacterial growth. Thus, optimum humidity in this area will improve the performance of biofilms in degrading microbes. N_2O which diffuses into the pores of the particles will be dissolved into the compost through layers of biofilms and degraded by the microbes contained therein.

N_2O gas removal efficiency tends to increase at the optimum water content for each particular filter medium. As shown in Figure 5 the most effective moisture content 60% (w/w), gives optimum N_2O removal efficiency. When the water content is more than 0.6 g water /g medium dry weight, the biofilter efficiency is reduced. The removal efficiency in this experiment increases than that of previous experiment due to the optimum moisture of compost and decrease in porosity, which result in longer residence time of N_2O in the compost. This will make more N_2O undergoing the adsorption and degradation processes, so that N_2O removal efficiency is also growing at every hour of contact time between the compost and N_2O gas. Consequently based on Figure 5, it is concluded that at 60% moisture content of compost will give higher N_2O removal efficiency.

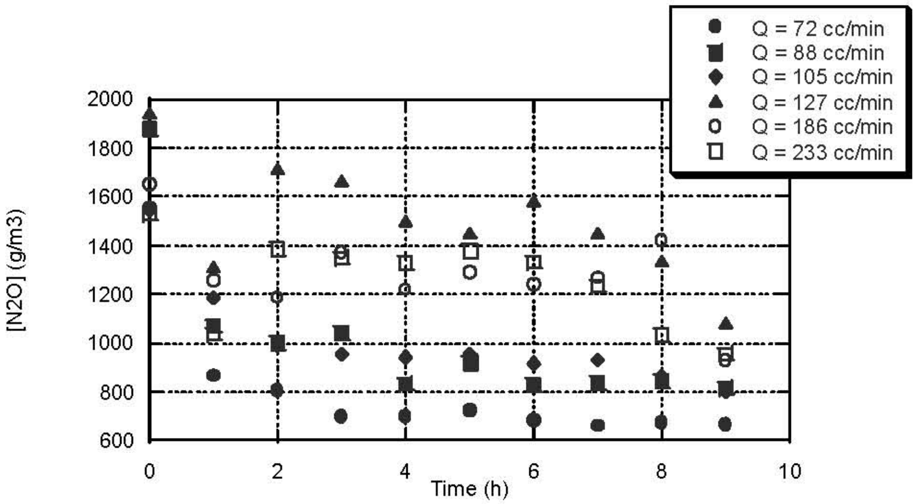

Fig. 2: Effects of gas flow rate on N_2O removal efficiency.

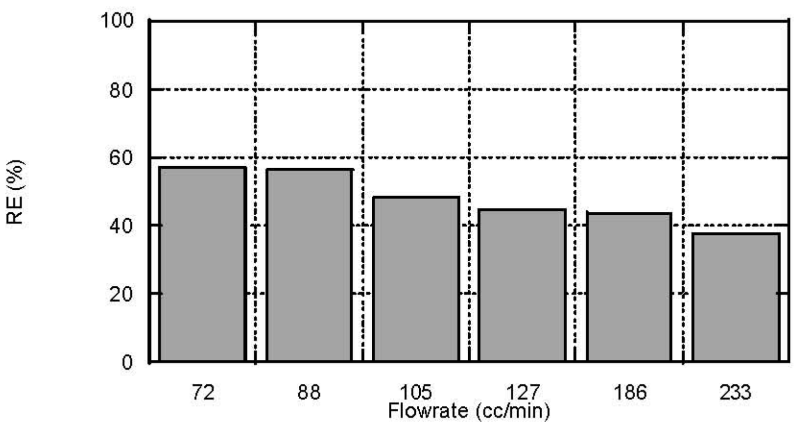

Fig. 3: N_2O removal efficiency in various N_2O flow rate.
(h = 50 cm, dried medium, t = 9 hours)

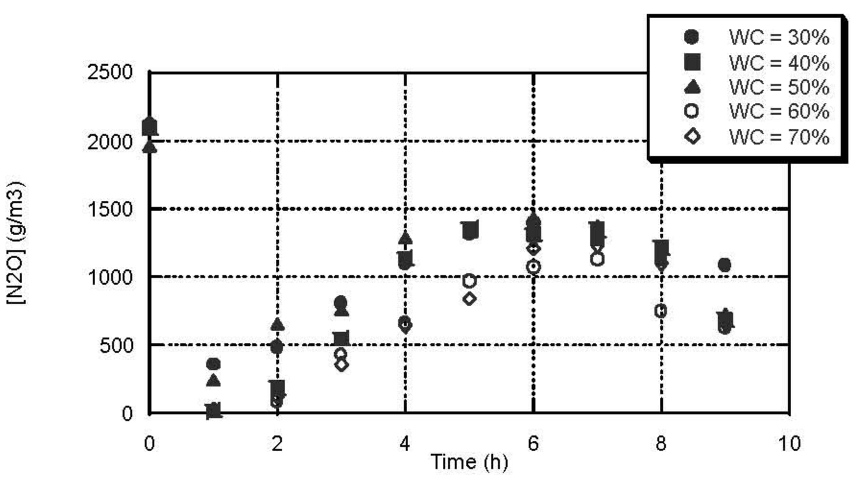

Fig. 4: Effects of water content on N_2O removal efficiency.

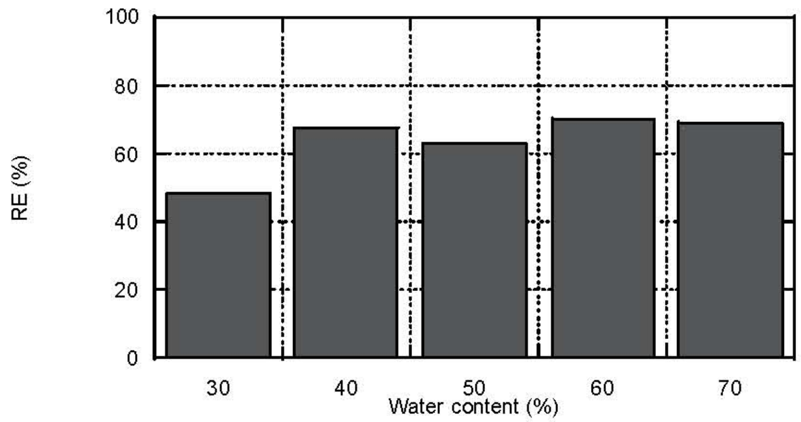

Fig. 5: N_2O removal efficiency in water content variation.
(h = 50cm, f =72 cc/minutes, dried medium, t = 9 hours)

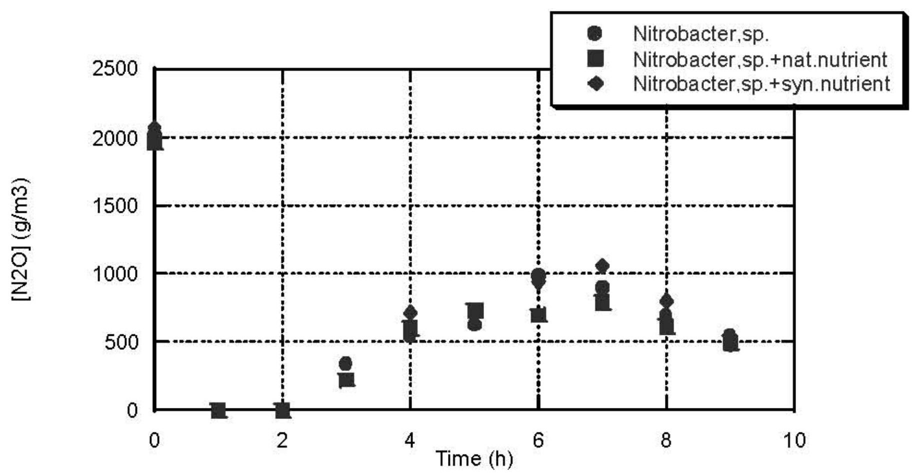

Fig. 6: Effect of nutrition supplementation on N_2O removal efficiency.

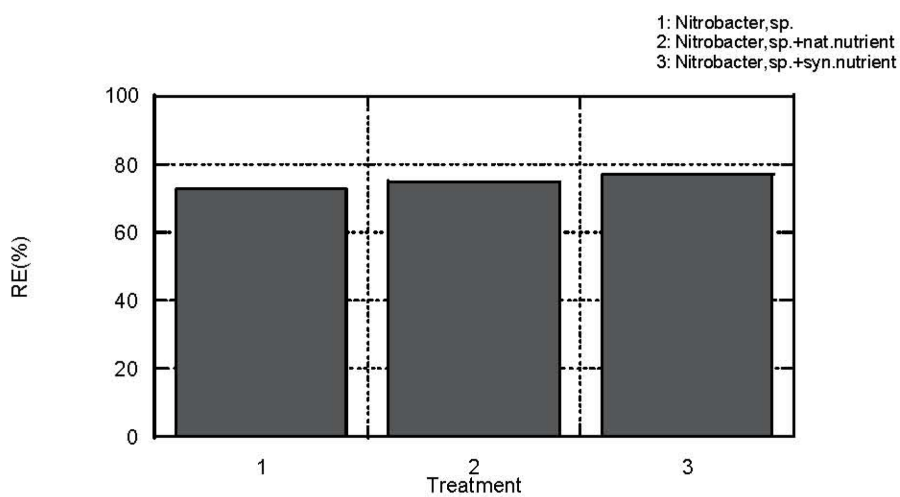

Fig. 7: The effect of nutrients addition on N_2O removal efficiency.

Comparison of Synthetic and Natural Nutrition: This experiment has the objective to find out the effects of addition of natural and synthetic nutrition on N_2O removal efficiency. In this experiment, compost as the filter medium is provided with *Nitrobacter, sp.* as nitrification bacteria. Then, this filter medium is added with nutrition which could increase the activity of microbes to reduce N_2O. The microbes needed the nutrition to survive and increase their activities.

The experiment is conducted with a filter medium height 50 cm, flow rate 72 cc/min and water content of 60% (w/w). Nutrients are dissolved in an amount of water reported as the best volume according to the previous experiments. The natural nutrient added is liquid waste from dairy farms, while the synthetic nutrient is added in the form of solution consisting of nutrient and trace elements explained above. Figure 6 shows the profile of the addition of natural and synthetic nutrients in the compost enriched with *Nitrobacter, sp.* to increase N_2O removal efficiency.

Figure 6 shows that the graphic comprises of three segments like the result from previous experiment. This is due to the condition of water content of the same filter medium making N_2O distribution in the compost is not different in unsteady phase of 6-7 hours. The difference is only in the nutrients addition which increases the moisture content of biofilter and degrading microbes' performance in N_2O reduction. Nutrients addition and water addition is different. The difference between the two experiments lies on the adsorption and degradation process in biofilter. Compost added with water and natural or synthetic nutrition will have N_2O removal efficiency higher than added with water only.

The purpose of nutrients addition is to support the growth of nitrifying bacteria. In order to maximize the growth of the bacteria, therefore, it is added natural or synthetic nutrients to the compost, wherein the addition is conducted after the addition of *Nitrobacter, sp.* To have a comparison, for initial parameters, experiment is also conducted to compost medium added with *Nitrobacter, sp.* without nutrients, natural or synthetic, addition. From these parameters, the effect of nutrients addition can then be compared between the synthetic and natural nutrients addition to the increase in N_2O removal efficiency. From Figure 7, it can be seen the difference among the three treatments.

Figure 7 shows that the highest removal efficiency is in the addition of synthetic nutrients with N_2O removal efficiencies 76.9%. When compared to experiment without nutrients addition, the removal efficiency is 4.2% higher. When compared to the experiment with addition of natural nutrients, N_2O removal efficiency is 2.2% higher. The increase in N_2O removal efficiency is the result from the minerals contained in the synthetic nutrients needed for particular microbes directly related to the degradation of N_2O. Meanwhile, natural nutrients do not have mineral content as complete as synthetic nutrients. However, natural nutrients also contain many additional microbes originated from cow manure, in which the microbes are also helpful in the biofilter performance.

Nutritional composition and trace elements added contain the elements of N, S, P, Ca, K, Na, Mg, Fe, Co and Zn. According to Shuler and Kargi (1992) minerals needed by the microbes are S, P, Ca, K, Na, Mg, Fe, Co and Zn. It is these elements contained in the synthetic nutrients causes better N_2O degradation than the natural nutrients.

Microbes Testing in Compost Medium: Test for microbial growth in compost before and after the biofiltration is conducted by Total Plate Count (TPC). TPC Total Plate Count (TPC) is one of analysis methods to determine colonies number of microbes in sample. TPC is one of the calculation techniques using Nutrient Agar (NA) as a medium for bacteria to be counted. The results of TPC calculation will be presented as Colony Forming Units (CFU) per gram of compost tested. In the results of TPC test, it can be seen different numbers of bacteria colony before and after biofiltration, visually or by calculation and it is seen that the number of colonies after biofiltration is higher.

Based on Table 1, it is seen that after biofiltration, the number of microbes calculated by TPC method is increasing. This is seen from the increase of microbes from 5.32×10^9 to 1.08×10^{10} CFU/g. This increase may occur as described in previous reports, one of which is the energy (ATP) derived from the transformation of air pollutants that flow to the biofilter [7]. It is shown that the number of bacteria contained in the compost after biofiltration is higher than that of dry compost (before biofiltration). Water content can create optimum

Table 1: TPC Test results before and after biofiltration

	Σ Bacteria (CFU/g)
Compost before Biofiltration	$5.32.10^9$
Compost after Biofiltration	$1.08.10^{10}$

environmental conditions for microbial growth, thus increasing the performance of these bacteria in degrading and get more nutrients from the results of such degradation.

The phenomenon of increasing number of bacteria in the compost after biofiltration strengthens the hypothesis that degradation processes occur in biofilter to reduce pollutants. It can also be concluded that the compost that has been used in biofiltration process will have better quality, because the increase in the number of bacteria as fertilizer in the compost is also increasing.

CONCLUSION

Performance of biofilter in this study in the N_2O removal efficiency achieves 75.9% with a medium height of 50 cm, N_2O flow rate 72 cc/min, 60% water content and the addition of synthetic nutrients and *Nitrobacter*, sp. to compost as a filter medium. It is showed that after biofiltration the number of microbes calculated by the TPC method is increasing and visual test from SEM confirmed this result.

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