

# Influence of Ammonium Concentration on the Performance of an Inorganic Biofilter Treating Methane

Marc Veillette, Antonio Avalos Ramirez, and Michèle Heitz

**Abstract**—Among the technologies available to reduce methane emitted from the pig industry, biofiltration seems to be an effective and inexpensive solution. In methane ( $\text{CH}_4$ ) biofiltration, nitrogen is an important macronutrient for the microorganisms growth. The objective of this research project was to study the effect of ammonium ( $\text{NH}_4^+$ ) on the performance, the biomass production and the nitrogen conversion of a biofilter treating methane. For  $\text{NH}_4^+$  concentrations ranging from 0.05 to 0.5  $\text{gN-NH}_4^+/\text{L}$ , the  $\text{CH}_4$  removal efficiency and the dioxide carbon production rate decreased linearly from 68 to 11.8 % and from 7.1 to 0.5  $\text{g}/(\text{m}^3\text{-h})$ , respectively. The dry biomass content varied from 4.1 to 5.8  $\text{kg}/(\text{m}^3 \text{ filter bed})$ . For the same range of concentrations, the ammonium conversion decreased while the specific nitrate production rate increased. The specific nitrate production rate presented negative values indicating denitrification in the biofilter.

**Keywords**—Methane, biofiltration, pig, ammonium, nitrification, denitrification.

## I. INTRODUCTION

WITH a total production of 109 million tons of pig meat in 2010 [1], the world pork industry was also responsible for water, air and soil pollution [2]. Among the compounds responsible for air pollution, this agricultural sector released volatile fatty acids, ammonia ( $\text{NH}_3$ ), hydrogen sulphide ( $\text{H}_2\text{S}$ ) and greenhouse gases (GHG) such as carbon dioxide ( $\text{CO}_2$ ) and methane ( $\text{CH}_4$ ) [3]. In Canada (2009),  $\text{CH}_4$  emissions represented 13% of the total GHG emissions (690 Mt eq.  $\text{CO}_2$ ) which corresponds to 90 Mton eq.  $\text{CO}_2$ [4]. In 2004, the world  $\text{CH}_4$  anthropogenic emissions represented around 6.9 Gton eq.  $\text{CO}_2$ [5]. With a heat of combustion of 890  $\text{kJ}/\text{mol}$  (25 °C, 1 atm) [6],  $\text{CH}_4$  is an interesting compound produced by anaerobic digestion of organic matter [3]. However,  $\text{CH}_4$  emissions, even if they are lower than  $\text{CO}_2$  emissions, are not neglectable in terms of global warming because  $\text{CH}_4$  has a global warming potential 25 times higher than  $\text{CO}_2$  over a period of 100 years [7].

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Even if  $\text{CH}_4$  can theoretically be thermally oxidized, the latter requires a minimal  $\text{CH}_4$  concentration in air ranging from 5 to 15% (v/v) [8]. In case of  $\text{CH}_4$  emitted from slurry storage, the concentrations are generally lower than 3% (v/v), which is not enough to use thermal oxidation [9]. On the other hand, several studies have shown that low  $\text{CH}_4$  concentrations can be treated effectively and relatively non-expensively by biofiltration [9]. In order to increase the biofilter performance, some parameters must be controlled such as moisture, temperature and nutrients [10]. Among the nutrients, microorganisms require nitrogen because it represents up to 14% of dry cell weight [11]. Usually, nitrogen is supplied to inorganic bed biofilters as a form of nitrate ( $\text{NO}_3^-$ ) [12] because ammonium ( $\text{NH}_4^+$ ) had a negative effect (inhibiting potential on  $\text{CH}_4$  oxidation) on methanotrophic bacteria in soil studies [13, 14], but also had a positive effect (stimulation of  $\text{CH}_4$  oxidation) in other soils studies [15].

The objective of this study was to test the effect of  $\text{NH}_4^+$  concentration in the nutrient solution of the performance of an inorganic packed bed biofilter treating  $\text{CH}_4$ . The performance of the biofilter was determined by analyzing the carbon and the nitrogen balance.

## II. MATERIALS AND METHODS

Fig. 1 presents the inorganic packed bed biofilter used for the experiments. The biofilter was a Plexiglas cylinder with an inlet diameter of 15 cm, divided into 3 sections. The biofilter was packed with an inorganic material for a total bed height of 1 m (volume of 18 L). The exact nature of the filter bed cannot be revealed for confidential reasons.

A mixture of pure  $\text{CH}_4$  (Praxair) and compressed air containing oxygen ( $\text{O}_2$ ) was fed at the bottom of the biofilter and the treated air was released at the top. In order to avoid filter bed desiccation, the air mixture was previously saturated with water by passing through a humidification column. A nitrate salts medium (NMS) was used to supply nutrients and moisture to the filter bed [17]. At the top of the biofilter, the nutrient solution was fed (1.5 L; once a day) while the leachate was collected at the bottom of the biofilter. Concurrently,  $\text{NO}_3^-$  (as sodium nitrate) concentration was decreased by 0.05  $\text{gN-NO}_3^-/\text{L}$  increasing steps and  $\text{NH}_4^+$  (as ammonium carbonate) concentration was increased in order to keep the total nitrogen concentration in the nutrient solution at 0.5  $\text{gN}/\text{L}$ . The  $\text{NO}_3^-$  concentration was decreased from 0.45 to 0  $\text{gN-NO}_3^-/\text{L}$  while the  $\text{NH}_4^+$  concentration was increased from 0.05 to 0.5  $\text{gN-NH}_4^+/\text{L}$ .

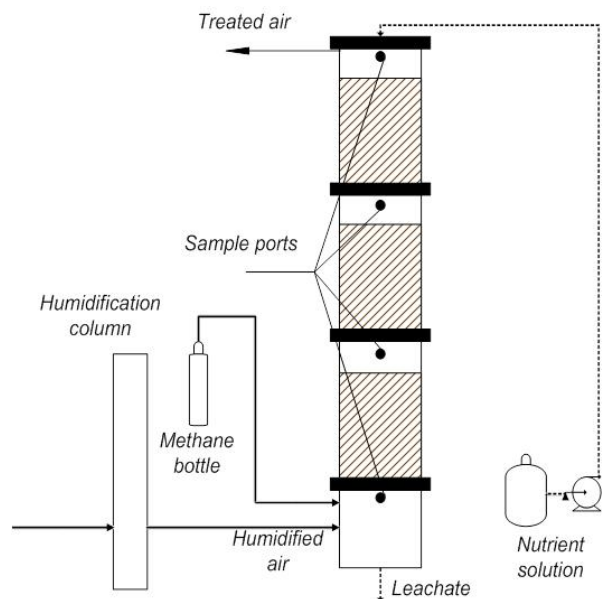


Fig. 1 Biofilter set up for the biofiltration of CH<sub>4</sub>

Suspended biomass contained in leachate samples was removed using filter paper. Ionic chromatography (Dionex ICS-1000, Canada) was employed to determinate the concentrations of NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> in leachate and nutrient solution [18]. The weight difference of the packing material sample dried at 105 °C and calcined at 500 °C was used to determinate the dry biomass concentration in the packed bed [19].

At the bottom of the biofilter, the CH<sub>4</sub> concentration and air flow rate were respectively set at 1500 ppmv (0.15% v/v) (inlet load of 10 g/(m<sup>3</sup>-h)) and 3 L/min. At each sample port, a total hydrocarbon analyser equipped with a continuous flame ionisation detector (Horiba model FIA-510, USA) was utilized to measure the CH<sub>4</sub> concentration in the gas phase. A gas analyser detector (Ultramat 22P, Simens, Germany) was also employed to measure the CO<sub>2</sub> concentration in the gas phase. Table I summarizes the main parameters considered to

evaluate the performance of the biofilter. The theoretical dry biomass production rate (DBR), listed in Table I, was used to evaluate the theoretical dry biomass production. This parameter is evaluated by means of a molar balance of CH<sub>4</sub> and CO<sub>2</sub>.

### III. RESULTS AND DISCUSSION

#### A. Biofilter Performance

Fig. 2 presents the methane removal efficiency (CH<sub>4</sub>-RE) and the P<sub>CO<sub>2</sub></sub> as a function of the NH<sub>4</sub><sup>+</sup> concentration in the nutrient solution. For NH<sub>4</sub><sup>+</sup> concentrations from 0.05 to 0.5 gN-NH<sub>4</sub><sup>+</sup>/L, the CH<sub>4</sub>-RE decreased linearly from 68 to 12%. For NH<sub>4</sub><sup>+</sup> concentrations ranging from 0.2-0.25 gN-NH<sub>4</sub><sup>+</sup>/L, the CH<sub>4</sub>-RE decreased quickly from 50 to 24%. For NH<sub>4</sub><sup>+</sup> concentrations ranging from 0.05 to 0.15 gN-NH<sub>4</sub><sup>+</sup>/L, the P<sub>CO<sub>2</sub></sub> increased from 7.1 to 12.4 g/(m<sup>3</sup>-h) and from 0.15 to 0.5 gN-NH<sub>4</sub><sup>+</sup>/L, the P<sub>CO<sub>2</sub></sub> decreased from 12.4 to 0.5g/(m<sup>3</sup>-h).

The fact that CH<sub>4</sub>-RE decreased with the NH<sub>4</sub><sup>+</sup> concentration shows the effect of NH<sub>4</sub><sup>+</sup> on the populations of methanotrophic bacteria present in the biofilter. Many studies have shown that NH<sub>4</sub><sup>+</sup> reduces the CH<sub>4</sub> oxidation rate in soil [20, 21, 22, 23], compost [24] and biofilters [18, 25, 26]. For example, in paddy soil, for CH<sub>4</sub> inlet concentrations of 1500 ppmv, Cai and Mosier [20] found that for an increase of NH<sub>4</sub><sup>+</sup> concentration from 0 to 0.05 mgN-NH<sub>4</sub>/kg soil, the CH<sub>4</sub> oxidation rate decreased from 338 to 166 ngC-CH<sub>4</sub>/(g soil-h). In the present study, the decrease of CH<sub>4</sub>-RE from 68 to 12% (-83%) was more important because, in soil, the nitrifying bacteria are already present, which reduced the NH<sub>4</sub><sup>+</sup> concentration in the filter bed. This could also mean that a NH<sub>4</sub><sup>+</sup> concentration of 0.5 gN-NH<sub>4</sub><sup>+</sup>/L has more negative effect on CH<sub>4</sub> oxidation than the NH<sub>4</sub><sup>+</sup> concentration used by Cai and Mosier [20](0.05 mgN-NH<sub>4</sub>/kg soil).

Between 0.05 and 0.15 gN-NH<sub>4</sub><sup>+</sup>/L, the P<sub>CO<sub>2</sub></sub> increased from 7.1 to 12.4 g/(m<sup>3</sup>-h) (+76%) even if CH<sub>4</sub>-RE decreased from 68 to 57% (-16%). This may mean that less carbon was used to produce biomass inducing a lower methanotrophic

TABLE I  
MAIN PARAMETERS USED TO EVALUATE THE BIOFILTER PERFORMANCE

Parameters	Equations	Units
Methane removal efficiency (CH <sub>4</sub> -RE)	$CH_4 - RE = \frac{C_{gin} - C_{gout}}{C_{gi}}$	Dimensionless
Carbon dioxide production rate (P <sub>CO<sub>2</sub></sub> )	$P_{CO_2} = \frac{Q \cdot (C_{dout} - C_{din})}{V}$	g/(m <sup>3</sup> -h)
Theoretical dry biomass production rate (DBR)	$DBR = \left[ \frac{(C_{gin} - C_{gout})}{W_{CH_4}} - \frac{(C_{dout} - C_{din})}{W_{CO_2}} \right] \left( \frac{Q \cdot W_B}{V} \right)$	g biomass/(m <sup>3</sup> -h)
Nitrate production rate (P <sub>NO<sub>3</sub></sub> )	$P_{NO_3} = \frac{(NO_3^-_{in} - NO_3^-_{out}) \cdot Q_{NS}}{V \cdot DB}$	gN/(g biomass-h)

a: C<sub>gin</sub> and C<sub>gout</sub> are the inlet and outlet concentrations of CH<sub>4</sub> (g/m<sup>3</sup>); Q is the air flow; V is the volume of the biofilter (0.018 m<sup>3</sup>); C<sub>din</sub> and C<sub>dout</sub> are the inlet and outlet concentrations of carbon dioxide (g/m<sup>3</sup>); W<sub>CH<sub>4</sub></sub>, W<sub>CO<sub>2</sub></sub> and W<sub>B</sub> are the molecular weights of CH<sub>4</sub>, CO<sub>2</sub> and biomass produced (g/mol), assuming an empirical formula of C<sub>5</sub>H<sub>7</sub>NO<sub>2</sub> for biomass with an average value of 113 g/mol [16]; NO<sub>3</sub><sup>-</sup><sub>in</sub> and NO<sub>3</sub><sup>-</sup><sub>out</sub> are the concentration of NO<sub>3</sub><sup>-</sup> in the nutrient solution and the leachate, respectively (gN/L); Q<sub>NS</sub> is the flow of nutrient solution (L/h); DB is the dry biomass in the filter bed (g biomass/(m<sup>3</sup> filter bed)).

activity, explaining the CH<sub>4</sub>-RE decrease. For NH<sub>4</sub><sup>+</sup> concentrations higher than 0.15 gN-NH<sub>4</sub><sup>+</sup>/L, the P<sub>CO<sub>2</sub></sub> followed the same tendency than the CH<sub>4</sub>-RE. Between 0.20 and 0.25 gN-NH<sub>4</sub><sup>+</sup>/L, the quickly decrease of P<sub>CO<sub>2</sub></sub> from 9.0 to 4.7 (-48%) confirms the assumption that for this concentration range, a major change in bacteria population occurred in the biofilter as observed in a previous study [19].

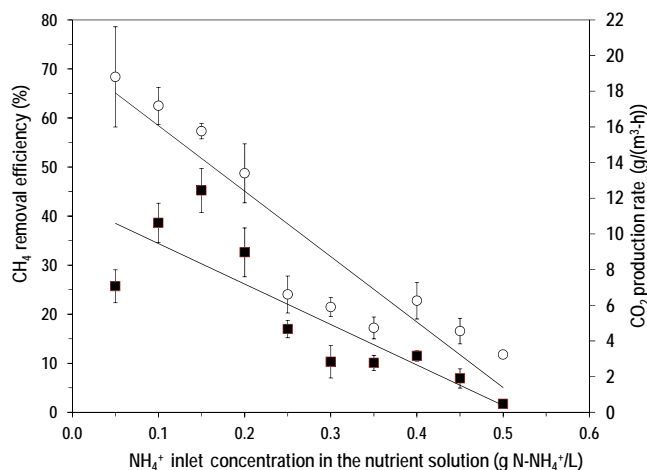


Fig. 2 CO<sub>2</sub> production rate (■) and CH<sub>4</sub> removal efficiency (○) as a function of the NH<sub>4</sub><sup>+</sup> inlet concentration in the nutrient solution

### B. Biomass

Fig. 3 presents the average dry biomass content (DB) and the theoretical dry biomass production rate (DBR) as a function of the inlet NH<sub>4</sub><sup>+</sup> concentration in the nutrient solution. For NH<sub>4</sub><sup>+</sup> concentrations ranging from 0.05 to 0.5 gN-NH<sub>4</sub><sup>+</sup>/L, the DB decreased with the NH<sub>4</sub><sup>+</sup> concentration and varied from 5.8 to 2.5 kg/m<sup>3</sup> filter bed. The DBR decreased also with the NH<sub>4</sub><sup>+</sup> concentration and followed a logarithmic tendency with values ranging from 30 to 5 g/(m<sup>3</sup>-h).

The decrease of DBR with the NH<sub>4</sub><sup>+</sup> concentration was also observed by Wilshusen et al. [24]. In order to explain this phenomena, the authors hypothesized that the exopolymeric substances could serve "as a carbon cycling mechanism for type I" methanotrophic bacteria. The fact that DBR decreased could also indicate that more carbon was transformed into CO<sub>2</sub>, which explains the decrease of CH<sub>4</sub>-RE observed in Fig. 2 as less new biomass was formed.

The fact that the DB (linear) followed a different tendency than DBR (logarithmic) indicates that some microorganisms other than methanotrophic bacteria (like denitrifying and nitrifying bacteria) can generate biomass. However, a visual inspection of the biofilter shows a decrease of the biomass in the filter bed which may mean that the biomass produced by other microorganisms may be more soluble in water. The dry biomass content is also influenced by the amount of biomass washed out of the filter bed at each daily watering. The decrease of DB observed in the filter bed (-57%) was lower than the DBR (-83%). As a consequence, for the NH<sub>4</sub><sup>+</sup> concentrations tested, less biomass would be lost in the leachate as the NH<sub>4</sub><sup>+</sup> concentration increased.

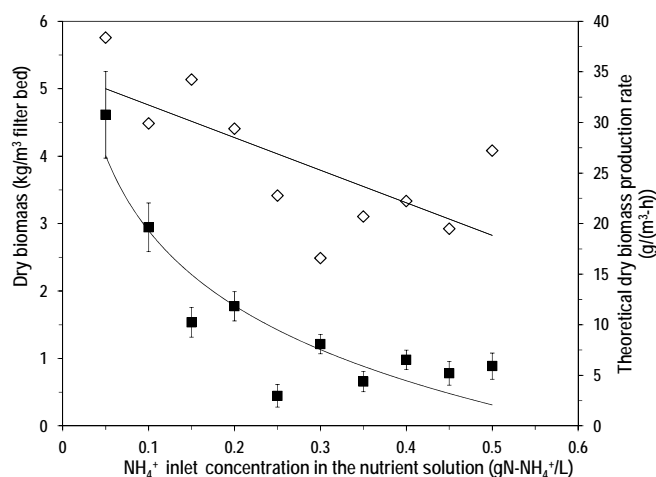


Fig. 3 Dry biomass content (◇) and theoretical dry biomass production rate (■) as a function of the NH<sub>4</sub><sup>+</sup> inlet concentration in the nutrient solution

### C. Nitrogen Conversion

Fig. 4 presents the NH<sub>4</sub><sup>+</sup> conversion and the specific NO<sub>3</sub><sup>-</sup> production rate (P<sub>NO<sub>3</sub></sub>) as a function of the NH<sub>4</sub><sup>+</sup> inlet concentration in the nutrient solution. For NH<sub>4</sub><sup>+</sup> concentrations in the nutrient solution ranging from 0.05 to 0.5 gN-NH<sub>4</sub><sup>+</sup>/L, the NH<sub>4</sub><sup>+</sup> conversion decreased linearly from 48 to 26 % while the P<sub>NO<sub>3</sub></sub> increased from -0.01 to 0.16 gN/(m<sup>3</sup>-h).

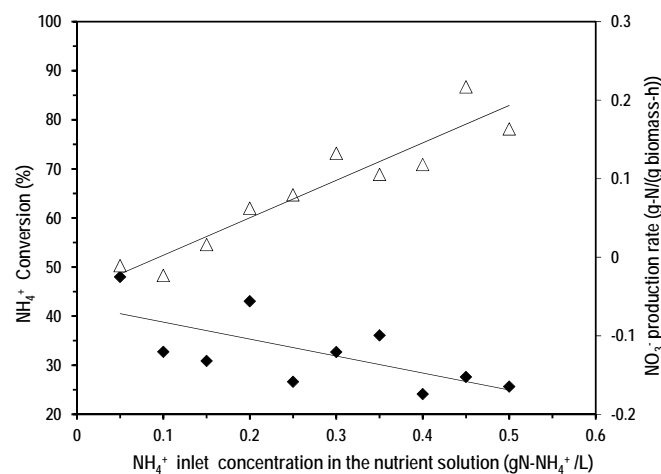


Fig. 4 Ammonium conversion (◇) and specific nitrate production rate (△) as a function of the NH<sub>4</sub><sup>+</sup> inlet concentration in the nutrient solution

The fact that the NH<sub>4</sub><sup>+</sup> conversion decreased with the NH<sub>4</sub><sup>+</sup> concentration could be due to the decrease of CH<sub>4</sub>-RE (Fig. 2). In fact, the increase of CH<sub>4</sub> concentration could lead to the decrease of NH<sub>4</sub><sup>+</sup> conversion as CH<sub>4</sub> is an inhibitor of nitrifying bacteria [27]. Moreover, the increase of CH<sub>4</sub> concentration in the biofilter could also lead to changes of number and kind of microorganisms specific to NH<sub>4</sub><sup>+</sup> conversion which could lead to the NH<sub>4</sub><sup>+</sup> conversion decrease. The P<sub>NO<sub>3</sub></sub> presented some negative values at 0.05 and 0.10 gN-NH<sub>4</sub><sup>+</sup>/L of -0.01 and -0.02 gN-NO<sub>3</sub><sup>-</sup>/(g biomass-h).

h), respectively. This indicated that there was a consumption of  $\text{NO}_3^-$  by methanotrophic bacteria or a denitrification.

#### IV. CONCLUSION

Increasing the  $\text{NH}_4^+$  concentration in the nutrient solution reduced the performance of an inorganic biofilter treating  $\text{CH}_4$  at an inlet concentration of 1500 ppmv, as follows: the  $\text{CH}_4$ -RE, the  $\text{P}_{\text{NO}_3}$  and the dry biomass content decreased respectively from 68 to 12 %, from 7.1 to 0.5  $\text{g}/(\text{m}^3\text{-h})$  and from 5.8 to 4.1  $\text{kg}/\text{m}^3$  filter bed. For the same range of concentrations, the  $\text{NH}_4^+$  conversion also decreased from 48 to 26% whereas the  $\text{P}_{\text{NO}_3}$  increased from -0.01 to 0.16  $\text{gN}/(\text{m}^3\text{-h})$  which suggests that denitrification occurred. This study shows that the nature and the concentration of the macronutrients (nitrogen) present in the nutrient solution are important for the performance of an inorganic biofilter treating  $\text{CH}_4$ .

#### ACKNOWLEDGMENT

This work was supported by a Strategic grant from the Natural Sciences and Engineering Council of Canada to Michèle Heitz in partnership with le Centre de Recherche Industrielle du Québec, Viaporc inc. the Civil Engineering Department in Université Laval (Professeur Paul Lessard). The authors also want to express their gratitude to the laboratories staff of Chemical and Biochemical engineering in Université de Sherbrooke. Thanks are also provided to Professor Ryszard Brzezinski and MSc Student Pascal Viens from the Department of Biology at Université de Sherbrooke for their support with the microbiological aspects throughout the project.

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