

TOWARDS NEW MANAGEMENT TOOLS FOR THE MONITORING AND MANAGEMENT OF LANDFILL GAS IN SMALL SITES

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SUMMARY: In this study, a monitoring campaign on the biofilter used to treat the biogas produced by the landfill located in Venosa (Basilicata, Southern Italy) was carried out to measure CH₄ and CO₂ concentrations in the gas entering in the biofilter, as well as in the treated air, at different operational conditions (i.e. biogas flow rate and moisture content). Mass balances between the influent and off-gas flow rates allowed us to estimate the CH₄ removal efficiency of the biofilter. Moreover, biological analyses on the filter medium were performed to evaluate the influence of ammonia nitrogen and temperature on the methanotrophic growth. The findings show that the CH₄ removal efficiency improves when the biogas flow rate decreases, because the lower flow rates minimize possible air contaminations from pipe losses. Regarding the factors influencing microorganisms growth, high incubation temperature cause an inhibition of biological processes, whereas the possible inhibition effect of ammonia depends on the dilution conditions in which the soil sample are prepared.

1. INTRODUCTION

Waste landfilling is nowadays the most common technique used in Italy for disposing municipal solid waste (MSW). The landfill is considered as a biological reactor working under anaerobic conditions in which the biodegradable organic portion of disposed waste is biodegraded. The biodegradable organic compounds are hydrolysed by microorganisms and dissolved in the liquid phase contributing to leachate production. Moreover, the anaerobic biological processes affect the production of landfill gas (LFG), which is mainly composed by methane (CH₄) and carbon dioxide (CO₂), as well as of nitrous oxide (N₂O). CH₄ is the second powerful gas contributing to climate change, because of its global warming potential (GWP) 25 times higher than CO₂ (IPCC, 2006). Even though CH₄ has been considered as the major contributor to climate change from landfill emissions, recent studies have paid attention also on N₂O to assess the environmental impact of MSW management (Nag et al., 2016). Particularly, N₂O causes concern because of its GWP, which is 289 times higher than CO₂ (IPCC, 2006), as well as its strong weight on ozone layer depletion.

The solid waste management cycle can be considered as an anthropogenic source of

greenhouse gas (GHG), requiring investigation on biogas and N₂O production/emission towards a sustainable waste management. Solid waste disposal can be considered the most important source of CH₄, contributing to 18% of the total anthropogenic CH₄ emissions (Scheutz et al., 2009). Therefore, in the last decades, researchers have paid attention on the main processes controlling CH₄ emissions and oxidation during waste treatment, focusing on innovative off-gas measurements and environmentally friendly techniques for biogas treatment. During the MSW disposal, CH₄ produced in the dump as biogas is collected by means of a collection system and then utilized to save energy or treated by means of on-site technologies (Hettiarachchi et al., 2011). However, the recovery of biogas as a source of renewable energy is economically sustainable within landfills of large size, since the high flow rate of produced biogas and the high concentration of CH₄ are the required elements for an efficient combustion. Otherwise, other techniques are preferred when CH₄ content is lower than 25%, especially in small sites with a volume smaller than 10000 m³ (Amodeo et al., 2015).

Biofiltration can be considered an alternative method to combustion in order to reduce the methane concentration in the flow leaving the dump surface, particularly in case of low flows and methane concentrations less than 20-30% (Amodeo et al., 2015, Haubrichs et al., 2006). Therefore, the development of biofilters as technology to treat biogas produced in landfills can be considered a reliable solution to control methane emissions from MSW (Menard et al., 2009), when the produced amount is not enough for a sustainable energy recovery (Hettiarachchi et al., 2011). The biofilter operating is based on biological oxidation of CH₄ into CO₂ and water by means of methanotrophic bacteria which use the methane content in the biogas as the only carbon and energy source for their activities.

In order to ensure the growing of methanotrophic bacteria, the biofilter is filled with a granular medium, such as wood chips added to compost, kept wet to favour the nutrient supply needed to the microorganisms activities (Park et al., 2002). The water content plays a fundamental role in CH₄ consumption that is inhibited at low water content (17% of water-holding capacity) at all temperatures (Einola et al., 2007). Moreover, water is also produced by microbiological reactions inside the biofilter, avoiding moisture losses, as well as contributing to maintain wet the deeper filter layers (Hettiarachchi et al., 2011).

Since methanotrophs are aerobic bacteria, a system that provide oxygen to the filter medium is necessary to ensure optimum CH₄ removal, as well as to improve the biofilter performance. Therefore, a landfill equipped with a suitable aerated biofilter is a reasonable solution for the biogas treatment (Haubrichs et al., 2006).

In this paper, biogas samples treated in a biofiltration unit were analyzed by means of gas chromatograph equipped with a Barrier Ionization Discharge (BID) detector to evaluate the better conditions for methanotrophs growth. Even though the commonly detectors used to analyze CH₄ and CO₂ concentrations are the flame ionization detector (FID) and the thermal conductivity detector (TCD), respectively (Pascale et al., 2017, Di Bella et al., 2011), BID allowed us to detect both the compounds simultaneously, saving time and money (Pascale et al., 2017).

The experimental activity focuses on the variation of the operational parameters of the biofilter, such as oxygen content, moisture rate, biogas flow, and methane concentration. In fact, these parameters, as well as their variations, are always taken into account in the literature to investigate methane oxidation (Stein et al., 2001; Abichou et al., 2011). Moreover, different environmental conditions were tested on the biological activities, varying the temperature and ammonia nitrogen content, because the rate of the biological methane degradation is temperature dependent (Streese et al., 2003). Moreover, methanotrophs can be classified into two different groups based on

operating conditions: the type I bacteria seem to be favoured by limiting methane combined with high concentration of nitrogen compounds, whereas type II bacteria prefer environments with high levels of methane and low concentrations of oxygen and nitrogen (Hanson et al., 1996).

The results can contribute to amplify the knowledge about the performance of aerated biofiltration systems, as well as on the methanotrophic activities. Moreover, strategies and policies can be suggested to improve the waste management towards the reduction on LFG emissions from landfills.

2. MATERIALS AND METHODS

2.1 Site under study

The case study landfill is located in Venosa in which the MSW of the north county of Potenza (Southern Italy) are disposed (Figure 1). The cell n.1 is equipped with a biogas capture system consisting in 21 wells linked to a collection system. The applied depression of few ten centimeters of water column avoids the air infiltration in the dump. Before the testing period, a biogas flow of $150 \text{ Nm}^3 \text{ h}^{-1}$ was conveyed to the flaring system.

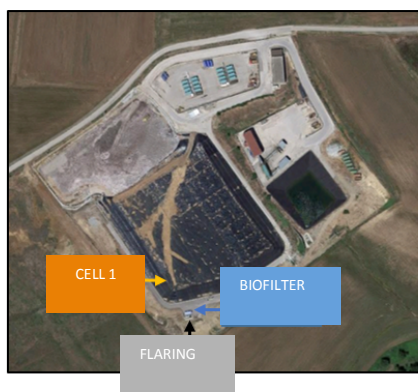


Figure 1. The landfill of Venosa (PZ): aerial view.

Subsequently, the landfill was then equipped with a biofilter (geCO₂ - Entsorga Italia SRL) with a volume of 22 m^3 (LxWxH, 6.5m x 2.5m x 2.65m) to treat the biogas produced in the landfill cell n. 1 (Figure 2), discharging the treated air through a chimney located on the biofilter cover. The biofilter was transported in the Venosa landfill in 2014 and, afterwards all the installation works, it was put in operation in 2015.

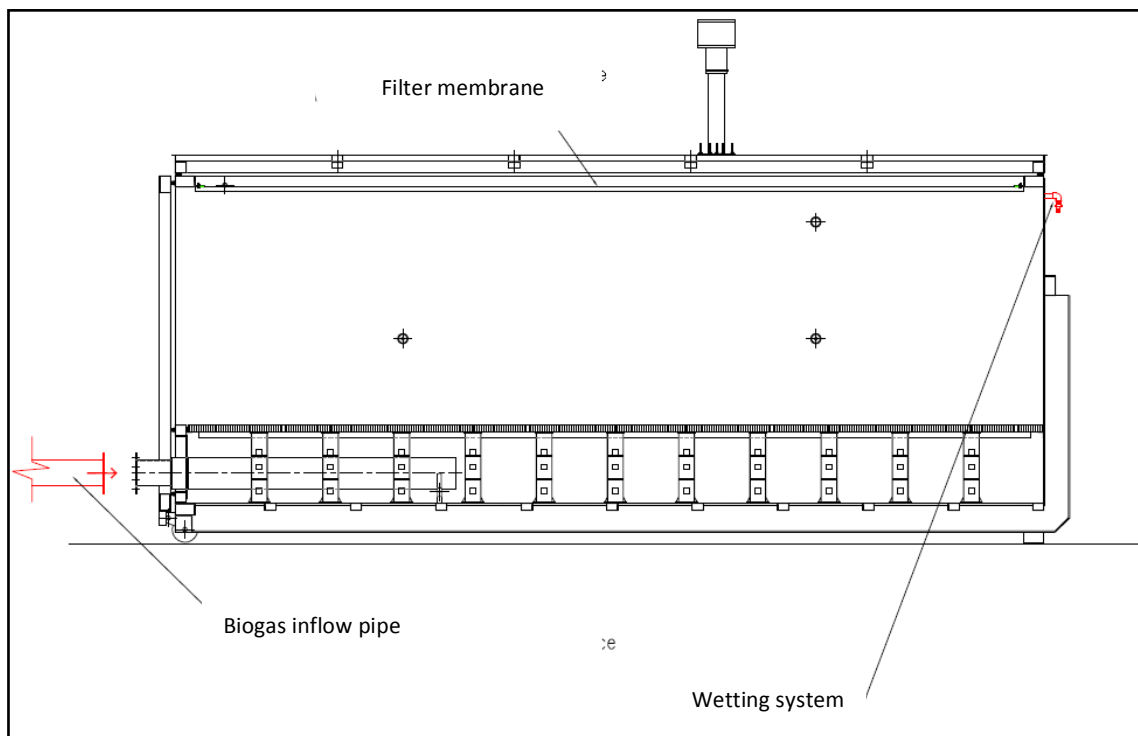


Figure 2. Biofilter (geCO₂ - Entsorga Italia SRL) layout.

An aeration system was positioned on the bottom of the biofilter, blowing the biogas on the entire filtering volume, in order to ensure a homogenous distribution of the collected biogas inside the filling material. A water storage was provided to ensure the necessary moisture content to the filling material. The filling material was of a mixture of biological activated matters, i.e. a mix of wood chips and compost. The compost has an important role during biological processes, ensuring the organic matter necessary for the microorganism’s activity (Haubrichs et al., 2006).

About 19 m³ of filling material were introduced in the container. About 20 cm of empty space was left on top, between the filling material and a geomembrane located under the closing system. The geomembrane has the task of retaining the moisture in the system, letting pass the treated LFG. A discharge valve was also installed at the bottom to unload the leachate.

The biofilter worked in two phases, the first was a biological process involving the filling material, whereas the later was a refining treatment involving the semi-permeable membrane used to demolish methane molecules.

2.2 Sampling plan

Two monitoring campaigns were planned to sample gas bags from the biofilter and the cell n.1, respectively. The first monitoring campaign was performed on the biofilter from February to June 2016. CH₄ and CO₂ concentrations in the gas entering in the biofilter, as well as in the treated air, were measured in different operational conditions, considering the variation of the influent biogas flow rate, as shown in Table 1:

Table 1. Variation of the influent biogas flow rate during the testing periods.

tasting date	biogas flow rate (Nm ³ h ⁻¹)
February 8th	24
May 13th	12.6

May 27th	10.2
June 15th	10.2
June 23rd	10.2

In order to avoid air infiltration, the biogas flow rate was decreased from 24 Nm³h⁻¹ to 10.2 Nm³h⁻¹. Therefore, mass balances between the influent and off-gas flow rates allowed us to estimate the CH₄ removal efficiency of the biofilter, as suggested in Equation 1:

$$CH_4 \text{ removal } [\%] = \frac{C_{in} - C_{out}}{C_{in}} \cdot 100$$

where C_{in} and C_{out} are the inlet and outlet concentrations, respectively.

Furthermore, gas samples were collected in both dry and wet working conditions, wetting the filter medium for 2 minutes per day for seven days.

Tedlar sampling bags (Recom Industriale s.r.l., Italy), equipped with vacuum pump (Gilian Air Plus), were used to collect the biogas from the biogas flow pipe, and the off-gases from the biofilter top. The Tedlar sampling bags for gases collection provided good sample storage properties and secure transportation means for reliable gas-chromatographic analyses (Caivano et al., 2016).

2.3 Analytical method

Measurements of CH₄ and CO₂ concentration were carried out by using a Shimadzu 2010 Plus Tracer gas chromatograph (Kyoto, Japan) equipped with a 2m×1mm (i.d.) ShinCarbon ST micropacked column (Restek, Bellefonte, USA) and a Barrier Ionization Discharge detector (BID). The injection temperature was set to 150 °C. The oven temperature program was 30 °C (hold time for 5 min) and increased to 120 °C with a 10°C min⁻¹ rate, as suggested in Pascale et al. (2017). All injections were made in the direct mode. Helium was used as carried gas at a flow rate of 15.0 ml min⁻¹. The BID detector was operated at 250°C; 80 ml min⁻¹ discharge gas flow was used for analysis. A 250 µl gastight syringe was employed to inject the gas sample into GC-BID system. The CH₄ and CO₂ concentrations were quantified by comparing the peak areas of samples against the standard curves over the range of desired concentrations (50-1000 ppm_v, i.e. 0.005-0.1%). The detection limits under these GC conditions were 60.80 ppm_v and 3.50 ppm_v for CO₂ and CH₄, respectively. The limits of quantification were 184.23 ppm_v for CO₂ and 10.61 ppm_v for CH₄. All biogas samples were analysed by diluting 1:500 with ambient air.

2.4 Biological tests on the filter medium

In order to assess the better conditions for methanotrophs growth, that is the better working conditions for the biofilter, biological analyses on the filling material were carried out in collaboration with the School of Agricultural, Forestry, Food and Environmental Science (SAFE) at the University of Basilicata, according to the guidelines reported in "Methods of soils analysis" (Page et al., Second Edition)

As suggested by guidelines, 1L of simple culture medium and 1L of culture medium with ammonia nitrogen (NH₃) was prepared to evaluate the influence of nitrogen on microorganisms growth. In fact, to our knowledge, how the ammonia nitrogen influences the abatement of methane, that is the methanotrophs growth rate, during biological processes is still unclear in the scientific literature. However, as found in Wang et al. (2011) and Gómez-Cuervo et al. (2015), nitrogen compounds, i.e. ammonia, nitrite, and nitrate, may have an inhibitory effect on CH₄ consumption by methanotrophs, even though the mechanisms are still unknown.

In order to prepare the culture medium, 0.5 g of K_2HPO_4 , 0.5 g of KH_2PO_4 , 0.2 g of $MgSO_4 \cdot 7H_2O$, 0.015 g of $CaCl_2$, 0.001 g of $FeSO_4 \cdot 2H_2O$, 0.001 g of $Na_2MoO_4 \cdot 2H_2O$, 10 ml of filter medium, and 12.5 g of Agar, were added to 1L of distilled water. By means of a pH probe, the pH of the culture medium was controlled and eventually corrected with nitric acid (HNO_3) to be in the range 6.8-7.

Then, a Ringer solution (25%) was prepared adding 2.25 g of sodium chloride (NaCl), 0.105 g of potassium chloride (KCl), 0.045 g of calcium chloride ($CaCl_2$), 0.05 g of sodium bicarbonate ($NaHCO_3$), and 0.034 g of citric acid to 1L of distilled water. Moreover, a sodium pyrophosphate solution (1.8%) was prepared adding 1.8 g of $Na_4P_2O_7 \cdot 10H_2O$ to 100 ml of distilled water.

Then, 100 ml of soil sample was produced from 10g of filter medium from the biofilter previously sifted to 2 mm and added with 90 ml of Ringer solution (25%) and 10 ml of sodium pyrophosphate solution (1.8%), sonicated for 2 minutes, and left at 4 °C for 15 minutes. The prepared sample diluted at 10^{-1} was further diluted by means of the Ringer solution (25%) until 10^{-3} and 10^{-5} .

48 Petri plates were filled with 20 ml of simple culture medium and 100 μ l of soil sample at different dilutions, whereas other 48 Petri plates were filled with 20 ml of culture medium with NH_3 and 100 μ l of soil sample at different dilutions.

Therefore, the 96 Petri plates were tightly closed in 4 sampling bags (30 L in volume) with 24 plates each, used to collect two different mixtures of air and biogas and investigate on the growth rate of methanotrophs. The first mixture (MIX 1) was composed by 50% of laboratory air and 50% of biogas from the landfill sited in Montegrosso-Pallareta (Potenza), that is 27% of methane and 11% of oxygen, obtaining a CH_4/O_2 volumetric ratio of 1:0.4. The second mixture (MIX 2) was composed by 28% of laboratory air and 72% of biogas from the landfill sited in Montegrosso-Pallareta (Potenza), that is 15% of methane and 15% of oxygen, obtaining a CH_4/O_2 volumetric ratio of 1:1. The variation of the CH_4/O_2 volumetric ratio allowed us to investigate on CH_4 removal efficiency under O_2 limiting conditions. In fact, as suggested by Park et al. (2009), the CH_4 abatement is hindered by lack of oxygen, avoiding the CH_4 oxidation into CO_2 .

The 4 sampling bags were filled with MIX 1 and MIX 2 at different levels of biogas and air, as shown in Table 2, and left to rest at air temperature or in oven for 2 weeks:

Table 2. Operational conditions in which the sampling bags

sampling bag	MIX	exercise temperature	biogas (L)	air (L)
1	1	35°C in oven	6	6
2	2	35°C in oven	3.4	8.6
3	1	air temperature	15	15
4	2	air temperature	8.4	21.6

3. RESULTS AND DISCUSSIONS

3.1 CH_4 and CO_2 emissions

Table 3 summarizes the data obtained from gas analyses for each testing day. Particularly, after a week of wetting (June 23rd) an aerobic conversion of the landfill was observed as the biogas entering in the biofilter is composed by 6.8% of CO_2 and 0.32% of CH_4 . However, comparing the values of CH_4 in the inlet and outlet, a scarce abatement of methane is found after the treatment inside the biofilter, demonstrating no conversion of CH_4 into CO_2 .

Table 3. CH₄ and CO₂ concentrations in the biogas entering in the biofilter (in) and in the off-gas leaving the biofilter surface (out). All values are in %.

testing date	CH ₄ in	CH ₄ out	CO ₂ in	CO ₂ out
February 8th	1.15	0.65	0.62	0.53
May 13th	3.4	1.9	4.4	2.2
May 27th	8.3	4.04	15.1	13.8
June 15th	0.48	0.25	11.5	12.5
June 23rd	0.32	0.24	6.8	7.4

During the second and the third testing date (i.e. May 13th and 27th), the data about CO₂ concentrations in the inlet and outlet show that the operating conditions of the biofilter could favour the growing of chemoautotrophic bacteria which use CO₂ as carbon source. In fact, the decrease of CH₄ is not coupled with an increase of CO₂, demonstrating that the amount of CO₂ produced from CH₄ conversion is consumed by chemoautotrophs.

Furthermore, the findings of the fourth day of tests (i.e. June 15th) show an abrupt decrease of CH₄ in the biogas leaving the landfill and entering the biofilter, coupled with a further increase of CO₂, probably due to air infiltration inside the dump causing the aerobic digestion of the waste. Therefore, thanks to a comparison with literature data, it can be concluded that the landfill of Venosa works in semi-aerobic conditions instead of anaerobic ones. The CH₄/CO₂ ratio can be used as index to verify the aerobic, semi-aerobic, or anaerobic behaviour of the landfill (Jeong et al., 2015). Indeed, the CH₄/CO₂ ratio represents the ration between of anaerobic and aerobic decomposition, assuming values in the range 1.08 – 1.46 (Jeong et al., 2015).

Table 4. CH₄/CO₂ ratio

testing date	Biogas flow rate (Nm ³ h ⁻¹)	Biogas CH ₄ content (%)	Biogas CH ₄ /CO ₂ (%)
February 8th	24	65	1.85
May 13th	12.6	43.5	0.77
May 27th	10.2	35.5	0.55
June 15th	0.48	11.5	0.04
June 23rd	0.32	6.8	0.05

Particularly, Yang et al. (2007) suggest a CH₄/CO₂ value of 1.9 if the landfill works under anaerobic conditions and a value of 0.8 if the landfill works in semi-aerobic ones. Therefore, looking at the data in Table 4, the landfill of Venosa is under semi-aerobic conditions.

The results in Table 5 show the performance of the biofilter in terms of CH₄ removal at three main biogas flow rates and in wetting conditions.

Table 5. Biofilter efficiency

testing date	Biogas flow rate (Nm ³ h ⁻¹)	Biogas CH ₄ content (%)	CH ₄ removal efficiency (%)
February 8th	24	65	43.5
May 13th	12.6	43.5	44.1
May 27th	10.2	35.5	51.3

The removal efficiency improves when the biogas flow rate decreases. Indeed, lower influent flow rates minimize possible air contaminations from pipe losses.

3.2 The influence of ammonia nitrogen on methanotrophic growth

The results regarding the methanotrophic growth are reported in Table 6. Regarding the plates incubated at air temperature (bag 3 and bag 4), after two weeks of incubation, colonies of microorganisms were found on the Petri plates in which was added simple culture medium (i.e. without NH₃). More colonies (i.e. 35) were counted on the plates filled with soil sample at higher concentration (10⁻¹) and incubated with the highest concentration of methane in the sampling bags (MIX 1).

Table 6. Microbial counts of methanotrophic bacteria. Values represent means (n = 3) ± standard deviation. Values with different letters in the same column are statistically different at P ≤ 0.05, according to Duncan's multiple comparison test.

Medium	Temp. (°C)	CH ₄ :O ₂ ratio	Days of incubation				
			5	10	13	21	30
M1 (- N)	20	1:1	0 ± 0 a	120 ± 25 b	3000 ± 80 b	2100 ± 160 b	2300 ± 350 b
		1:0.4	0 ± 0 a	250 ± 25 a	3500 ± 95 a	2700 ± 250 a	3000 ± 220 a
	35	1:1	0 ± 0 a	10 ± 5 d	1000 ± 120 d	800 ± 180 d	1000 ± 150 c
		1:0.4	0 ± 0 a	30 ± 10 c	1600 ± 235 c	1200 ± 350 c	1200 ± 190 c
M2 (+ N)	20	1:1	0 ± 0 a	0 ± 0 e	140 ± 20 f	150 ± 25 f	300 ± 50 e
		1:0.4	0 ± 0 a	0 ± 0 e	300 ± 30 e	400 ± 30 e	600 ± 50 d
	35	1:1	0 ± 0 a	0 ± 0 e	0 ± 0 g	0 ± 0 g	0 ± 0 f
		1:0.4	0 ± 0 a	0 ± 0 e	0 ± 0 g	0 ± 0 g	0 ± 0 f

The presence of NH₃ caused the inhibition of microorganisms growth also on the Petri plates filled with soil sample at 10⁻³ and 10⁻⁵, however in the first case the number of colonies is greater than in others, but the size is lower, whereas in the second case the number of colonies is lower than in others, but the size is greater.

Therefore, a positive correlation was found between the number of colonies and the concentration of methane inside the sampling bag.

Moreover, a low number of colonies were counted on the plates incubated at 35 °C in oven (bag 1 and bag 2), demonstrating the inhibition effect of high temperatures on microorganisms growth.

In fact, as suggested by Wang et al. (2011), the CH₄ oxidation rate increases exponentially in the range 10 – 30 °C and then decreases until to a completely inhibition at 60°C.

Furthermore, after a week of measurements, because of the methane concentration decrease inside the sampling bags due to biological processes, the mixture inside the 4 sampling bags was renewed with a new one. In fact, since the methane was the only source of carbon, the decreasing of its concentration caused a decrease of methanotrophic rate, as well as an inhibition of microorganisms growth. Particularly, because the bag 4 and bag 2 were filled with MIX 2, the effects of low methane concentration were greater, that is the inhibition of growth occurred before than in bag 3, especially on the plates filled with soil sample at 10⁻¹.

Therefore, the bag 4 and the bag 2 were filled with a new mixture (MIX 3) composed by 30% of laboratory air and 70% of biogas from the landfill sited in Montegrosso-Pallareta (Potenza), that is 38% of methane and 6% of oxygen, whereas the bag 3 and bag 1 were filled with MIX 1 again. Then, bags 1 and 2 were incubated at 30 °C in oven, whereas bags 3 and 4 were incubated at air temperature. Small-sized colonies grew on Petri plates filled with soil sample at 10⁻¹ and with the culture medium added with NH₃, whereas no more colonies grew on the plates filled with simple culture medium. Furthermore, the influence of NH₃ on the microorganisms growth on the plates filled with soil sample at 10⁻⁵ can be considered negligible.

Particularly, a comparison between the plates incubated in the bag 1 and the ones incubated in the bag 2 showed how the air rich in methane (MIX 3 in bag 2) favoured the microorganisms growth, confirming that methane was the only carbon source for methanotrophs. However, the colonies size on the plates incubated at 30 °C was lower than the size of ones incubated at air temperature, confirming the negative effects of high temperatures on biological processes.

Regarding the dilution rate, the effect of NH₃ on the plates filled with soil sample at 10⁻⁵ was different from that of previous measurement, showing no more inhibition of microorganisms growth. Therefore, the effect of ammonia nitrogen on the methanotrophic growth is still unclear, even though the findings on the plates left to rest at air temperature are more reliable, suggesting the possibility to add nitrogen-based fertilizer to the filter medium inside the biofilter.

4. CONCLUSIONS

The experimental results found in this work show that the biofilter is able to reach a significant decrease of methane in the influent biogas, even working under different conditions of oxygen percentage, moisture content, and temperature. However, a significant improving of decrease of CH₄/CO₂ ratio is recorded at low biogas flow rate because of reduction of air contaminations.

Since the landfill of Venosa worked under stabilization conditions, that is low concentration of methane in the biogas, the findings of this work can be useful to test the measurement of the CH₄/CO₂ ratio and the biological growth on fresh dump in which the methane production is relevant.

The biological analyses on the methanotrophs activities allowed us to individuate the operational conditions to improve the biofilter performance. Particularly, the methane concentration in the biogas entering the biofilter, the ammonia content in the filter medium, the exercise temperature, and the oxygen content need to be controlled to obtain a significant abatement in methane. Since a standard protocol for methanotrophs processes evaluation is not still available in literature, these findings can help to develop suitable analytical protocols to conduct standard tests on microorganisms activities inside a biofilter.

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