MICROBIAL OXIDATION TECHNOLOGIES FOR LANDFILL GAS EMISSIONS MITIGATION IN LANDFILL AFTERCARE: ABATEMENT OF METHANE AND TRACE COMPONENTS IN BIOFILTRATION PROTOTYPES IN MEDITERRANEAN REGIONS

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SUMMARY: Landfills are listed as the second antropogenic source of atmospheric methane (CH_4) and contributes significantly to climate change. To reduce surface emissions from landfills have been developed biocover or biofilter to enhance biologically methane oxidation by methanotrophic bacteria. The RE Mida project is a demonstrative project in the frame of Climate Change Mitigation co-finance by LIFE-Programme. Within the project have been designed and constructed two biofiltration systems: a biofilter, located at Podere il Pero landfill in province of Arezzo, and 7 passive biowindows, located at Le Fornaci landfill in province of Siena. The aim of this paper is to show the result of the monitoring campaigns made to evaluate the performance and the CH_4 oxidation efficiency of the prototypes. For the biofilter the results show an average oxidation efficiency among 60-70%. Despite the biowindows show an oxidation efficiency depending on the inlet methane concentration, but has been achieved oxidation efficiencies of 100%. The results of the monitoring NMVOC and odorous emissions show a significant reduction of the pollutant inlet the biofiltration prototypes.

1. INTRODUCTION

One of the main environmental challenges associated with landfills is the generation of LFG, gas produced by the anaerobic decomposition of the organic waste fraction. LFG is mainly composed by methane (CH_4) and carbon dioxide (CO_2), and its production lasts until most of the organic material in waste is degraded, which can take several decades. Suboptimal cover design and improper landfills management can lead to undesired LFG emissions. Infact, landfills are significant anthropogenic sources of odours and greenhouse gases. In particular, landfills are the second antropogenic sources of CH_4 emissions (IPCC 2013).

LFG emissions reduction is a central issue of the EU Landfill Directive (Directive 99/31/EC).

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In order to reduce the CH_4 emissions from the waste management sector the Directive acts through two main measures: (i) the mandatory use of LFG extraction and utilization systems or LFG flaring in all new disposal sites; (ii) the progressive reduction of the amount of biodegradable waste going to landfill. Despite these measures, some critical issues related to LFG management are still present, in regard to residual LFG gas with low calorific value. Indeed, when the concentration of CH_4 in LFG collected is too low for combustion, the gas collected is often vented without thermal treatment. LIFE RE Mida aims to provide other direct environmental benefits by the mitigation of uncontrolled NMVOCs emissions. LFG contains a wide variety of trace NMVOCs including other climate-relevant gases, organosulfur compounds and H_2S (El-Fadel et al., 1997). These trace components are either the sub-products of the biological and chemical process occurring in the waste (alcohols, ketones, esters, organic acids, furans and sulphur compounds) or they may have anthropogenic origin (such as halogenated hydrocarbons) and have been deposited together with the waste. It has been demonstrated that NMVOCs emissions from landfill may pose risk to human health (odour nuisance), cause photochemical smog, and contribute to global warming and to ozone depletion (Bacchi, 2016).

Within the project, two demonstrative biofiltration systems were designed and constructed in two landfills with different features, chosen as application cases. The prototype constructed in Podere Il Pero Landfill (Arezzo, Italy) is a biofilter fed by using a part of the LFG collected by the existing active gas extraction system, that currently goes to a flare. In Le Fornaci di Monticiano Landfill (Siena, Italy), a passive biofiltration system (biowindow), was preferred since in this plant the active LFG extraction system has never been installed.

Beside the prototypes construction, the project involves monitoring of the biological process in order to assess the performance of the demonstrative systems in terms of CH_4 oxidation and NMVOCs abatement.

Concerning long term results, LIFE RE Mida aims to encourage the development of biofiltration technologies in the field of LFG management in both managed and unmanaged landfills. Indeed, the target sites of the proposed technologies are all the landfills in which the concentration of CH_4 in LFG collected is too low for combustion without the use of additional fuel, such as natural gas. Whatever the landfill site, this occur every time that the CH_4 concentration is lower than 20% v/v. According to our experience, this could be caused both because of landfill site characteristics and because of specific needs in LFG management.

The aim of this paper is to show the results of the monitoring campaign made to asses the performance of the prototype installed at Podere il Pero e Le Fornaci landfills and to describe the methods used to evaluate the oxidation efficiency of CH_4. Besides, the design and construction phases are described.

2. SITE CHARACTERIZATION

Podere il Pero Landfill is a landfill for non-hazardous waste located in the Province of Arezzo (Tuscany). The landfill is divided into four sections. The landfill was active from 1994 until March 2014. The final volume achieved is amount 631,000 m^3. The operations of closure (installation of final capping) started in the summer 2015 and are currently ongoing. A total of approximately 660,000 tons of waste had been disposed at Podere il Pero. The dominating waste types are soil fill, household refuse and sludge from wastewater treatment plants.

An active extraction system is use for LFG control. The system consists in 44 vertical wells and 8 leachate wells and a network of pipes connected to 6 points for suction control (Figure 1).

Since September 2004 an internal combustion engine, with an electrical power of 625 kW, has been used to recover energy, but from 2012 the energy production was no more technically possible due to LFG quantity and quality. For this reason, currently a LFG flare is used.

The current data, measured on continuous basis by CSAI (industrial partner of the project), shows in 2015 an average LFG flow rate of 128 m^3 h^{-1} with an average CH_4 concentration of 33.9% v/v, ranging between 26.5 and 40.5% v/v. Furthermore, by comparing the data measured at each main header, resulted that the LFG collected from header 5 is characterized by the lower concentration of CH_4 and the higher concentration of O_2. To evaluate the LFG emissions from the capping was performed a monitoring campaign by using the static flux chamber method. From the survey resulted that a total amount of 54 Nm^3 h^{-1} of LFG is emitted from the capping (corresponding to a specific flow of 1.1 Nl $h^{-1}m^{-2}$) of which 28.2 Nm^3 h^{-1} of CH_4.

Le Fornaci di Monticiano Landfill is an old landfill of MSW in the Province of Siena. This plant has been closed before the entry in force in Italy of the Landfill Directive. The landfill consists into two sections. One is an old landfill managed directly by the municipality of Monticiano between the 80s and the 90s, the other has been constructed by Sienambiente (SA, industrial partner of the project). About the older part there is not reliable information. The more recent section was active between the 1996 and the 2001. In this section, about 29,300 tons of waste had been disposed. The LFG is managed through a passive system (no extraction blower is present). The system installed provides a control method of allowing migrating LFG to escape from the soil without active mechanical system. The passive venting involves 15 vertical wells equipped with riser pipes surrounded by gravel pack. Each well supports little flares with manual ignition (Figure 1). The LFG generation over the time has been estimated through a LFG model by considering the quantities and the typology of waste disposed of in the landfill. The model shows in 2016 negligible LFG generation from the landfill site.

Despite this result, a field survey, using the static accumulation method, has been done in order to detect possible hot spots (high emissions points) on the landfill cover. The higher emissions were detected in the surrounding of the vertical wells: points of discontinuity of the landfill cover from which the residual LFG can be easily emitted. Beside the flux measurements, the LFG composition within the wells was also measured. Significant CH_4 concentrations were observed in all the wells. In some wells high O_2 concentrations were measured, probably due to air contamination within the wells. The highest CH_4 emission was detected at the well 11, but also at wells 12, 9 and 1 significant CH_4 were measured.

Figure 1. LFG management system - Podere il Pero (on the left), Le Fornaci (on the right)

3. PROTOTYPE DESIGN AND CONSTRUCTION.

3.1 Biofilter

Based on the outcomes of preliminary site characterization action, the biofilter has been designed to treat a LFG flow ranging between 20 and 50 Nm^3/h. An average LFG composition of 20% CH_4, 35% CO_2, 10% O_2 and 35% N_2 has been considered.

The prototype was built near the blower station of the existing LFG extraction system. This was the most suitable place because of the presence of all the needed utilities (LFG, water, electricity etc.). The biofilter has a total area of 270 m^2, 18x15 meters. The containment walls, made of concrete, are 2 m high. A pitched roof consisting in a metal carpentry structure with a PVC cover lining was designed and set up.

The LFG is supplied to the prototype by header 5 that collects the LFG from the oldest part of the landfill. By means of a system of valves, the LFG quality is controlled to maintain the requested composition (maximum CH_4 concentration of 20% v/v).

The LFG supply system includes a dedicated blower, an inverter, a flowmeter, a manometer, a temperature probes, a gas analyser and a PLC. From the blower, the LFG enters into the biofilter through a system of pipes. A bottom distribution layer of coarse gravel (grain size 15-30 mm) of 40 cm high layer was made (Figure 2). A geogrid was used to separate this layer and the top filtering media (compost). The biofilter floor has a slight inclination in order to eventually remove water from the bottom. In this case, the leachate is collected by gravity from the biofilter and moved to a tank through a drain trap system.

In both prototypes compost mixed to sand in a volume ratio of 5:1 as structural material (Jugnia et al., 2008) was used as filtering media; indeed, from previous literature results, compost has proved to be a suitable substrate for biocovers constructions (Mor et al., 2006; Huber-Humer et al., 2009; Scheutz et al., 2009).

The compost used in the prototypes fulfilled most of the criteria indicated by the authors Huber-Humer et al. (2009) showing optimal characteristics in terms of physical, chemical and maturity properties.

Figure 2. Construction of the pilot biofilter, LFG distribution system, filtering bed

3.2 Biowindows

Based on the data observed, in the case of Le Fornaci di Monticiano Landfill a passive biofiltration system was considered the most effective method to treat emissions from hot spots.

Based on the outcomes of site characterization 7 hot spots on the landfill cover have been identified. On each point, the existing LFG passive control system has been substituted by a biowindow. Each biowindow was designed to treat the highest LFG emission measured during the preliminary survey.

Each biowindow has a volume of about 5 m^3 with a filtering section of 4 m^2 (2m x 2m). The biowindows consists of a gravel layer for the proper LFG distribution and an overlying compost layer, where CH_4 oxidation occurs. The thicknesses of the layers were approximately 120 cm (compost), and 20 cm (gravel layer, grain size 15-30 mm). Sand in a volume ratio 5:1 has been added to improve compost structure. A geogrid was used to separate the two layers. Metal formworks were used to contain the filtering bed and reinforce the whole module.

The LFG migrates through the landfilled waste (below the landfill cover) to the biowindows due to pressure differences and diffusion (Gebert and Groengroeft, 2006). The biowindow media has a higher permeability compared to the landfill cover layer, and thus LFG moves through this towards the atmosphere. To limits rainwater entry, each biowindow has been protected with clay levees (Figure 3).

Figure 3. Design and construction of the pilot biowindows, filling of the formworks and final works

4. MATERIALS AND METHODS

4.1 Monitoring and control plan

A detailed review of the state-of-the-science regarding microbial CH_4 oxidation has been done to identify the appropriate methods and operations to control the factors affecting the biological process and the oxidation rate. Based on the results of the review a monitoring control plan has been established (Table 1).

Basically, the assessment of the performance of the biofiltration systems is done by measuring the gas concentrations and the temperature at different depth (gas concentrations and temperature profiles) and the LFG fluxes by the surface, defined as the routine monitoring. See the following paragraphs for the methodology applied.

Table 1. Monitoring and control plan

Activity	*Timing*
Filter media characterization: pH, organic matter (Total Volatile Solid - TVS), TOC, TKN, Ptot, NH4-N, NO3-N, SO4	Fourth-Monthly
Leachate (eventually collected) characterization: BOD5, TOC, COD, SO4, N-NH4, sulphates, ammonia nitrogen, nitrous and nitric acid, metals and total solid contents.	Monthly
Visual inspection of the surface	Monthly
Control and analysis of the LFG inlet flow (CH4, CO2, O2, H2S)	Continuous biofilter
Measurement of the temperature profile of the biofilter media	Monthly
Continuous acquisition of the environmental climate conditions, data analysis	Continuous
Detection of anaerobic conditions (O_2 concentration)	Monthly
Study and analysis of the gas concentrations profiles (CH4, CO2, O2, H2S)	Monthly
Survey of NMVOCs and odorous emissions	Six-Monthly

4.2 Gas concentration profiles and temperature profile

Gas concentration profiles are studied by placing multilevel probes and drawing gas samples to be analyzed for main landfill gas components: CH_4, CO_2, O_2 and H_2S. The gas concentration probes have been made of PVC pipes of 16 mm ID and closed at both ends. The sampling probes have 12 different lengths and are placed at 10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 110 e 130 cm below the surface level. Each pipe, at the lower end, is provided of six 2 mm slots to drawing gas samples at desired depths. The instrument uses to measure pollutant concentrations (CH_4, CO_2, O_2 and H_2S) is Ecoprobe 5 (IR analyser; Rs Dynamics). For each sampling probe, the measurement integration time was determined based on the probe volume.

For the passive biofiltration systems (biowindows) the gas concentration profiles have been evaluated in duplicate. Despite, for the biofilter have been installed 6 sampling lines (Figure 4). Besides, are measured the temperatures of the biofiltering bed at different depth (10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 110 cm) with a penetration probe (Hanna Instruments, HI 935005).

4.3 Surface flux measurements

Surface emission of CH_4 and CO_2 were determined using static flux chambers. According to the method, the meausure of a surface flow of a pollutant is based on the increase of the concentration of the pollutant within the chamber. The emissive flow of a pollutant can be estimated by evaluating the α derivative in the initial trajectory of the dependency function between the pollutant concentration in the chamber and the time within the system conditions (or control volume) are respected.

The equipment used in the field is made up of a flux chamber and a gas analyzer (Ecoprobe 5, IR analyser, Rs Dynamics). In the case of study, two flux chambers have been used, both of which have circular shape, developed during Ph. D thesis by Pecorini (2010). The largest has a radius of 10 cm and a height of 20 cm, for a total volume of 0.006 m^3, while the smallest has a radius of 9.95 cm and a height of 10.5 cm. The flux chambers are made of HDPE with a thickness of 1 cm. The upper base is enclosed with a 27 cm square plexiglas panel and a thickness of 1 cm, on which three valves are inserted, two of which are connected to PVC pipes of 0.6 cm diameter forming a closed loop between the flux chamber and the analyzer. Inside the flux chamber is installed a fan to achieved a homogenization of the collected gas. See Figure (4) for the sampling point scheme of both biofiltration systems.

Instead, the dynamic flux chamber method has been used to control NMVOC's emissions. According to this method, the chamber is located on the area that usually represent an emissive hot-spot. The chamber is made of PTFE, with an inner diameter of 50 cm and an outside diameter of 20 cm, inside the chamber there is a multifire spiral gas distribution system, made of PTFE and a tubular shape windshield with diameter of 25 cm and height of 40 cm.

The methodologies UNI EN 13725:2004, U.S. EPA TO 11A 1999, NIOSH 6013:1994 were applied to evaluate respectively the odorous emissions, the H_2S concentration and the NMVOC concentration. For the biofilter was decided to perform three surface sampling and to evaluate the NMVOCs and odorous compound also on the inlet biogas. Besides, were choosen 3 biowindows on which perform a surface and biogas sampling.

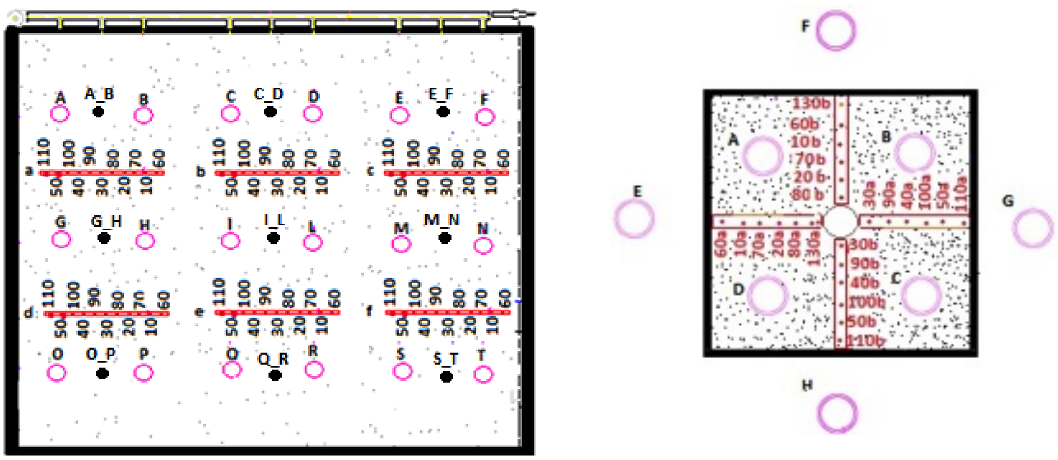

Figure 4. Monitoring and control plan, sampling point scheme

4.4 Evaluation of protoype performance

For the assessement of the oxidation efficiency of the biofiltrations systems has been decided to use the methodology proposed by the authors Gebert et al., (2011).

The authors Gebert et al. (2011) proposed a method to calculate the oxidation efficiency of biofilters, biowindows and biocovers, based on the gas concentration profile of CH_4 and CO_2.

In both the biofiltration systems studied are installed gas sample probes and the proposed method can then be applied. The main assumption on which the method is based is that the

CO_2 concentration is mostly due to the oxidation of CH_4 by methanotrophic bacteria and therefore, the biomass respiration phenomena are negligible. Under this hypothesis, the oxidation efficiency of CH_4 can be reconstructed along the gas concentration profile by the CH_4/CO_2, as follows:

$$\frac{CO_{2\_LFG} + x}{CH_{4\_LFG} - x} = \frac{CO_{2\_i}}{CH_{4\_i}}$$

Where CO_{2_LFG}= CO_2 concentration of the landfill gas (%v/v), CH_{4_LFG}= CH_4 concentration of the landfill gas (%v/v), CO_{2_i}= CO_2 concentration at depth *i* (%v/v), CH_{4_i}=CH_4 concentration at depth *i* (%v/v) and x= share of oxidized CH_4 (%v/v).

By dividing the share of oxidized CH_4 up to depth *i* by the concentration of CH_4 in the landfill gas (CH_{4_LFG}), one obtains the cumulative percentage of CH4 oxidized, Eff_{ox} (%) (Gebert et al., 2011):

$$Eff_{ox} = \frac{x}{CH_{4\_LFG}}$$

For the biofilter CH_4 oxidation efficiency is also calculated using a mass balance method, from the fluxes into and out the system, as follows (Gebert et al., 2011):

$$Efficienza\ globale\ [\%] = \frac{CH_{4in} - CH_{4out}}{CH_{4in}}$$

Where CH_{4in}= specific flux of methane inlet the biofilter calculated as the product of the biogas flow in the biofilter and the average concentration of CH_4 of the landfill gas (%v/v) divided by the section of the biofilter ($Nm^3m^{-2}h^{-1}$).

4.5 Environmental climate condition

With regard to the determination of the environmental climate conditions at the Podere il Pero landfill, the average, minimum and maximum temperature data, wind direction, and wind speed, humidity and precipitation are available every 15 minutes from weather station in the plant.

For the site of Le Fornaci, weather data are available on the site of the Functional Center of the Tuscany Region (CFR), where the data from the meteorological unit installed at Monticiano are part of the monitoring network of the Tuscany Region.

5 RESULTS AND DISCUSSION

According to the timeline in Table 2, have been carried out 6 complete monitoring campaigns for the biofilter. The first two were made to validate the routine monitoring protocol. In addition to material and methods validation, the results of these preliminary monitoring campaigns have been used to assess the preliminary performance of the prototypes.

Table 2. Operation, control and monitoring of the biofilter, monitoring campaign performed

Monitoring campaign	*Date*	*Activity*
I campaign	15/12/2017/ 31/01/2017	Routine monitoring
II campaign	31/01/2017	Routine monitoring
III campaign	14/02/2017	Routine monitoring
IV campaign	02/03/2017	Routine monitoring + NMVOC and odorous compounds emissions
V campaign	15/03/2017	Routine monitoring
VI campaign	19/04/2017	Routine monitoring

The oxidation efficiencies were estimated through the gas concentrations measured within the biofiltering bed (Gebert et al, 2011). Concerning the biofilter in most of the sampling lines a significant oxidation efficiency has been observed, ranging from 60 to 70% (Figure 5). Furthermore, the high temperature measured in the biofilter bed indicates the presence of active biological process (results not show). However, comparing the results among the different sampling lines, it was possible to assess that the biological process is not yet uniformly developed within the prototype (Figure 5). This could be due to a non-uniform distribution of the inlet LFG. The inlet average methane load is among 8.33 $g_{CH4}m^2h^{-1}$.

Observing the CH_4 oxidation performance estimated during the complete monitoring period (Figure 5), it was observed a slightly increase of the performance. In this first semester, the inlet load has been kept constant in terms of both LFG flow (about 20 Nm^3h^{-1}) and CH_4 concentration (between 16 and 19% v/v) since the goal of the activities was to grow methanotrophs bacteria. The highest average CH_4 oxidation efficiencies were measured at 10 cm of depth and ranged between 62% and 70.5%.

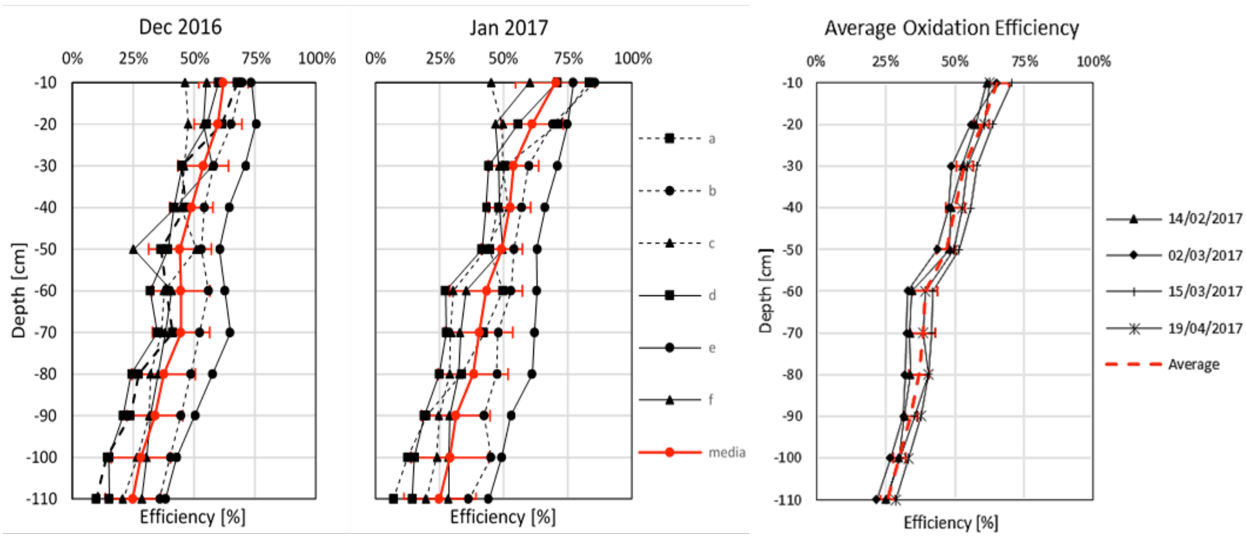

Figure 5. Biofilter oxidation efficiencies for sampling lines and average oxidation efficiencies

The results of the NMVOCs on the inlet biogas, show the predominance of hydrocarbon compounds (isobutane, n-butane, n-pentane, n-hexane, n-heptane and propylene), which are among the VOC species most present in the analyzed samples and are produced by degradation processes usually associated with older sections of the landfill (Parker et al., 2002).

The oxygenated and halogenated compounds were found at appreciable but not very high concentrations. Overall, the concentrations of sulfuric and terpene compounds are lower than those normally found in the landfill gas. In the surface biogas emission, there is a significant reduction of pollutants in particular of halogenated and aromatic sulfur compounds whose concentration is not appreciable in the gas emitted due to the limits of low detection that can be achieved with the methods used.

The odor concentrations, of the superficial gas emitted by the device were quite low at all sampling points. The higher odor concentration detected was 166 [UO/Nm^3], below 300 UO/Nm^3, the concentration limit normally prescribed for exhaust air treatment devices (biofilters waste treatment plants).

Despite, for the passive biofiltration systems, according to the timeline in Table 3, have been carried out 7 complete monitoring campaigns.

Table 3. Operation, control and monitoring of the biowindow, monitoring campaign performed

Monitoring campaign	*Date*	*Activity*
I campaign	26/10/2016	Routine monitoring
	16/11/2016	Routine monitoring
II campaign	30/11/2016	Routine monitoring
	19/12/2016	Routine monitoring
III campaign	26/01/2017	Routine monitoring
IV campaign	22/02/2017	Routine monitoring
V campaign	01/03/2017	NMVOC and odorous compounds emissions
VI campaign	23/03/2017	Routine monitoring
VII campaign	26/04/2017	Routine monitoring

Comparing the performance of the biowindow, three main behavior of functioning depending on the inlet methane concentration has been observed: biowindows 4 and 6 (CH_{4in}<10% v/v), biowindows 3 and 14 (CH_{4in}=10-25% v/v) and biowindows 11, 12 and 8 (CH_{4in}> 25% v/v). The oxidation efficiencies indeed vary accordingly and the higher efficiencies were achieved by biowindows 4 e 6 (100%), average efficiencies were estimated for biowindows 3 e 14 and lower efficiencies were measured for biowindows 8, 11 e 12, where high methane load are present. Although lower efficiencies were measured for these prototypes, the efficiencies achieved were still quite high (75%).

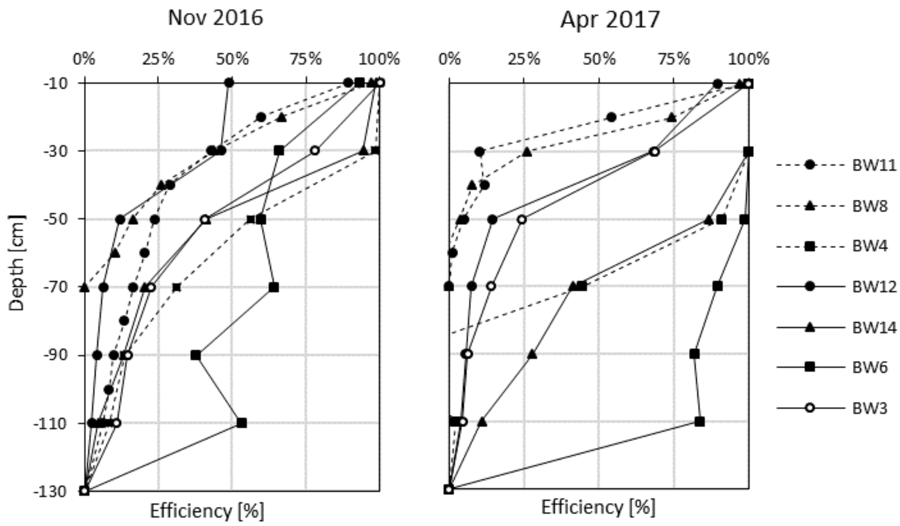

Figure 6. Average efficiency of the biowindows

As a result of the monitoring campaigns, to control NMVOCs and odorous compounds was decided to sample the most emissive biowindows (numbers 8, 11 and 12). The oxygenated and halogenated compounds were detected at appreciable but not very high concentrations except the gas entering the device 12 where concentrations of these VOC families were detected at maximum values (8 mg/m^3). The concentrations of sulfuric and terpene compounds are lower than those normally found in the landfill gas. For device number 11 an inlet concentration of 36.6 mg / Nm^3 of H_2S was found, negligible concentrations were detected for the other modules analyzed. About the odorous emission were found very low concentratios: BW8 18 UO/Nm^3, 32 UO/Nm^3, 23 UO/Nm^3.

6. CONCLUSIONS

The monitoring and control plan implemented is a viable approch to evaluate the performance and the CH_4 oxidation efficiency of the prototypes installed at Podere il Pero e Le Fornaci landfills. During the monitoring campaigns for both biofiltration systems was detected metanotrophic activity. In addition, during the first period of bacteria acclimatation phase, high oxidation efficiency (more than 50% biofilter and over 70% for biowindows) was calculated. Also, a high level of abatment of NMVOCs and even odorigen compounds was observed. However, to assess the long-term performance of both biofiltration systems, the monitoring campaign are going on till December 2018. During this period will be assess the influence of environmental climate condition on passive biofiltration system. Besides, will be evaluated the performance of the active biofilter setting various inlet methane load.

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