

Article

Landfill GHG Reduction through Different Microbial Methane Oxidation Biocovers

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Abstract: Emissions from daily and final covers of municipal solid waste (MSW) landfills can produce significant impacts on local and global environments. Simplifying, landfills can cause local impacts with odor emissions and global impacts with GHGs. This work focuses on hydrogen sulfide (H₂S) and methane (CH₄) emissions, with the aim of studying how it is possible to reduce their impacts by means of biofiltration systems. Both field and laboratory investigations have been carried out in Casa Rota Landfill (Tuscany, Italy). In the field trials, four pilot-scale biocovers made of compost from a source-selected organic fraction (SS compost), compost from a mechanical biological treatment plant—the residual fractions of the MSW, a mixed compost (SS-MSW compost) and sand were monitored in the daily cover area of the landfill, where high emissions were detected. Results showed that high CH₄ and H₂S emissions reductions occurred in the mixed SS-MSW compost plot, given a maximum methane oxidation efficiency of greater than 98% and an average oxidation efficiency of about 75%. To assess the specific oxidation rate, laboratory tests using SS-MSW compost sampled from the biocovers were done.

Keywords: microbial methane oxidation; landfill odor mitigation; biocovers; biofiltration systems; greenhouse gases emissions mitigation

1. Introduction

Landfills are considered important anthropogenic sources of odors and greenhouse gases, mainly due to uncontrolled biogas emissions. The odor pollution impact from landfill sites is often within 2.0 km, but under bad conditions, such temperature inversion, it can go beyond 6.0 km [1]. Methane is a large potential contributor to climate change with a global warming potential of 21 [2].

Landfilling is still the most widely used form of disposal of municipal solid waste (MSW) in Europe. Eurostat shows that in 2018, on average, in the 28 member states, 23% of MSW produced was disposed of in landfills [3,4]. With reference to 2018, in Italy the MSW disposed in landfill amounted to the 22% of the produced waste [5].

Biogas collection systems and daily and final covers are the used methods for emissions control [6], and several studies demonstrated that these two systems are highly dependent on each other: the rate of efficiency of the gas extraction system varies from 50% to 95% depending on the type of cover system (daily, intermediate or final) [7]. The conventional possibilities of reducing the greenhouse effect from waste landfilling consist of landfill gas flaring or combustion with energy recovery [8,9]. Furthermore, over time, with the natural decrease in biogas production and the reduction in the methane content [10], conventional treatment technologies become technically and economically unsuitable [11–13]. Biofiltration systems are considered effective alternatives in the management of landfill gas for both methane oxidation of fugitive emissions with low calorific value [14,15] and for the mitigation of odorous compounds [12,16,17].

Researchers have found that, among the processes that naturally occur in the landfill covers, the presence of CH₄ and O₂ counter gradients provides the necessary conditions for the development of methanotrophic bacteria, able to degrade methane in landfill gas. As the investigation of microbial CH₄ oxidation in landfill progressed, the potential to exploit the process in engineered systems was quickly recognized [18–21].

In particular, the environmental factors that could affect the process have been found to be: temperature [14,22–24], moisture content [25,26], micronutrient availability [18,27], material characteristics of the filter media [14,28,29] and climatic and inhibition conditions [30–32].

Biofiltration is one of the biological methods which is used for odor elimination [16,33,34]. Three important general factors determine compost biofilter performance: (a) the type of the filter media (including void fraction, particle size, moisture content, microbial diversity and nutrients), (b) the prevailing conditions of gas flow inside the biofiltration unit (including superficial velocity, gas distribution, temperature and inlet pressure) and (c) the substrate concentration, solubility and biodegradability [7,18,33]. Similarly to biological methane oxidation studies, also in this case, numerous researchers have studied the optimal process parameters and the performances of different filtering materials [35,36].

Since many factors influence the efficiency of the microbial oxidation processes of the landfill gas and it is difficult to define the process conditions [37], an innovative combined approach between the field pilot scale and the laboratory scale has been chosen here.

The aim of this study was to compare different types of biocover filter media produced by plants by treating the organic fraction of municipal solid waste, compost, for instance, in order to identify which performs best in terms of reducing methane emissions. This paper describes the results of field and laboratory investigations that have been carried out in Casa Rota Landfill (Tuscany, Italy). A pilot biocovers system was trialed to mitigate the uncontrolled emissions from the daily cover area. Compost from a source-selected organic fraction, MSW compost from a mechanical biological treatment plant, mixed compost and sand were compared in the experimental biocover. Furthermore, some laboratory column tests were carried out in order to assess the performances of the materials used in the field test with a known inlet biogas flow.

2. Materials and Methods

2.1. Description of the Site

Casa Rota Landfill is in the Province of Arezzo (Tuscany, Italy). In the landfill, non-hazardous municipal and industrial solid waste has been accepted from 2006 until now, with an average rate of 262,530 t yr⁻³. The total waste disposal capacity is 5.2 million m³ in its 11 cells, and it has a total surface area of about 190,000 m². The active gas management system is composed of 126 vertical perforated wells, 52 horizontal drainage pipes and 33 dual leachate/gas extraction wells. The experimental biocover was constructed in the daily cover area of the landfill, where high emissions were detected. In general, the construction and management of the landfill studied complies with the European Landfill Directive [38].

2.2. Biocovers System Design

In order to construct the biocover system, the temporary cover (about 35 cm of compacted clay) was removed to enhance the gas flow up to the biocover.

The biocover system consisted of four plots of 25 m² delimited by a clay bank. Each of the experimental plots included a 70 cm thick layer of substrate underlined by a 20 cm drainage layer of gravel. Plots were directly fed by the biogas produced by 1-year-old unsorted waste.

The materials used in the biocover were 1-year-old compost from source-selected organic fraction (SS compost), MSW compost from a mechanical biological treatment plant (MSW compost), a mixture of the previous compost (SS-MSW compost) and sand. The composts were mixed with sand with

a volume ratio of 5:1, as suggested by Jugnia et al. (2008) [39]. All the materials were available in large amounts at the landfill, since a mechanical biological treatment and composting plant is near the landfill.

The organic matter content of the SS compost (loss of ignition at 550 °C) was 27.4% w/dw, the pH 7.1 and the dynamic respiration index (DRI) was 373 mgO₂h⁻¹kg⁻¹ of volatile solids (VS) [40,41]. An organic matter content of 23.3% w/dw, a pH of 7.6 and a DRI of 427 mgO₂h⁻¹kg⁻¹VS were measured for the MSW compost. Respirometry tests were performed according to the UNI/TS 11184, and, as suggested by the standard, the obtained results are typical for biologically stable substrates [40,41].

Twenty-seven probes were installed into each plot to measure the soil gas concentration and the temperature at 15, 25, 35, 45, 55, 65 and 75 cm depths. Probes were assessed in triplicate for each plot and consisted of high-density polyethylene (HDPE) pipes each with an internal diameter of 8 mm. In order to measure the gas composition outside the experimental area, 4 probes were externally installed at 25 and 35 cm depths next to each plot.

2.3. Column Tests

To assess the performances of the materials used in the field test with a known inlet biogas flow, some laboratory trials were done.

A column was constructed with an HDPE pipe with an external diameter of 250 mm, a length of 1700 mm and a wall thickness of 16 mm. The column was filled with a gas distribution layer of coarse gravel of 35 cm in thickness covered by about 105 cm of tested sample. From the bottom, the column was continuously fed with moisturized synthetic gas controlled by a flow meter, while on the top in the headspace, a fan was used for air-mixing. Vertically, 11 probes, consisting of HDPE pipes each with an internal diameter of 8 mm and a length of 10 cm, were set at intervals of 10 cm for the gas pore composition analysis in the filtering media.

This work shows the results obtained with a sample of the SS-MSW compost cover. The sample used had a pore volume of 36.5% *v/v*, a pH of 8.1 and a water content of 30.4% *w/w*. At a minimum of two weeks duration each, two case studies were set up: non-aerated column tests with pure methane (99% *v/v* CH₄), and an aerated test, using a gas mixture of 12% *v/v* CH₄ and 88% *v/v* of air. The tested flows were high to simulate biofilter applications: 41 and 206 g m⁻²d⁻¹ in the non-aerated tests; 82 and 312 g m⁻²d⁻¹ in the aerated biofilter simulation.

2.4. Measurement and Monitoring Program

In the monitoring program of the experimental biocover, implemented from August until November 2015, the conditions of the substrates used in the biocover, the soil gas concentrations and the gas emissions were detected.

Furthermore, during the monitoring period, meteorological data, including precipitation, air temperature, air humidity and atmospheric pressure, were continuously recorded by the weather station installed near the experimental biocover.

In each day of monitoring, samples of materials from each plot were taken at 15 cm depth. Total solids (TS) and the water content were analyzed by weighting the samples before and after drying at 105 °C for 24 h until a constant weight; VS were measured as loss of ignition at 550 °C and pH of the water suspension of the sampled substrates was measured too, according to ANPA (2001).

Soil gas concentration profiles were studied by the multilevel probe system previously described for both biocover and column tests. The concentrations of methane, carbon dioxide, oxygen and hydrogen sulfide were measured by a portable infrared gas analyzer (Ecoprobe 5, RS Dynamics, CZ) integrated with a H₂S-AH Hydrogen Sulfide Sensor. For the analysis of soil gas emissions below 500 ppm, the gas was sampled by means of a foil bag and analyzed within 4 h with the micro-gas chromatograph (INFICON, Switzerland) according to the procedure described by Baldi et al. (2019) [42,43]. In order to get stable concentration measurements, the gas sampling was done with a pump speed of 0.25 l min⁻¹ and different measurement periods. Measurements were done starting from the internal probe and

moving toward the external ones in a clockwise direction. In the column tests the pore gas composition was analyzed starting from the bottom. All readings were made in triplicate to ensure data repeatability.

Gas emissions were measured by the static accumulation chamber method using an infrared gas analyzer and a sample cell of 6 l with an internal diameter of 200 mm [44]. In the field tests a regular grid was used for the flux measurements, and two measurements for each experimental plot and eight measurements in the external area in a clockwise direction were done [45]. All readings are made in triplicate to ensure data repeatability. The monitoring model applied here has been defined by other studies and then validated by the repeated field application [12,19,27,45].

2.5. CH₄ Oxidation and Odor Mitigation Efficiencies

Scheutz et al. (2009) [18] suggest that the use of mass ratios and mass balances for the CH₄ and CO₂ to derive CH₄ oxidation is problematical because, in addition to production of CO₂ via methanogenesis and CH₄ oxidation, CO₂ is also produced and consumed by multiple subsurface and near-subsurface processes (soil respiration, organic matter oxidation, photosynthesis). As such, in the present research the measured biogas emissions were used to qualitatively assess any effect of the biofiltration.

Nevertheless, to give an indication of the oxidation efficiency, the method proposed by Gebert et al. (2011) [14] was used in field and laboratory trials. According to the assumptions, in a biofilter with high CH₄ loading and in temporary landfill covers, the volume of oxidized CH₄ at a certain depth can be derived from the Equation (1):

$$\frac{[\text{CO}_2]_{LFG} + x}{[\text{CH}_4]_{LFG} - x} = \frac{[\text{CO}_2]_i}{[\text{CH}_4]_i} \quad (1)$$

where x is the share of oxidized methane (% v/v); $[\text{CO}_2]_{LFG}$ and $[\text{CH}_4]_{LFG}$ are the concentrations of methane and carbon dioxide in the landfill gas (% v/v), which correspond to the external probe measurements; $[\text{CO}_2]_i$ and $[\text{CH}_4]_i$ are methane and carbon dioxide concentrations at a i depth (% v/v).

To assess the odor mitigation effects of the experimental biocovers, concentrations of the hydrogen sulfide measured in the internal and external probes will be compared.

3. Results and Discussion

3.1. Climatic Conditions and Characteristics of the Filter Material

Figure 1 shows the daily atmospheric temperatures and the atmospheric pressure logged during the monitoring period, and the average atmospheric temperature in each monitoring day. Data showed the seasonal decreasing of the atmospheric temperature during the trial (from 22 to 9 °C) and the fluctuation of the atmospheric pressure.

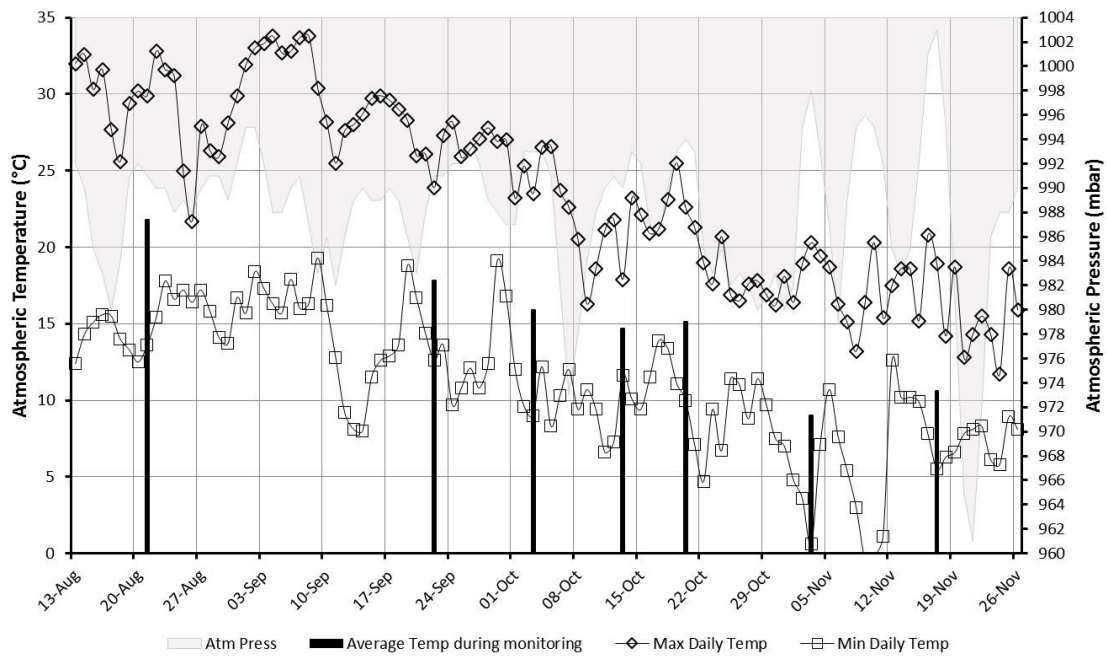


Figure 1. Atmospheric temperature and pressure at Casa Rota Landfill during the monitoring.

Figure 2 shows the relative humidity and the daily rainfall logged. As expected, precipitation was frequent and intensified in autumn months with the overall increasing of the relative humidity.

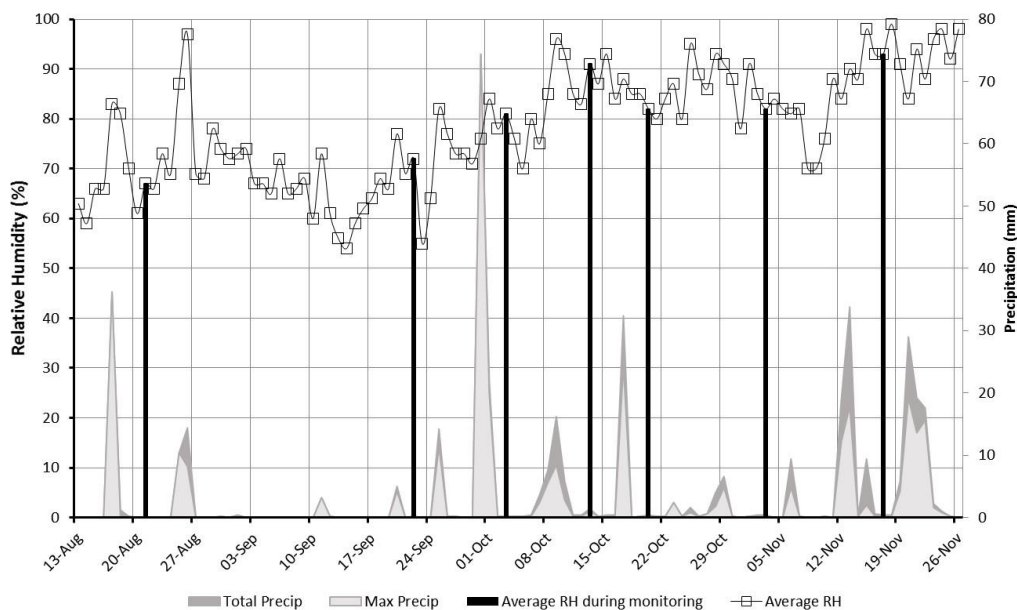


Figure 2. Relative humidity and precipitation at Casa Rota Landfill during the monitoring period.

In SS compost, MSW compost and SS-MSW compost biocovers, the pH ranged between 7.4 and 8.1, whereas the sand plot was lightly acidic, between 6 and 6.5. The water content values were always lower than 30% *w/w*, except in the compost cover in the last monitoring day probably because of the heavy rainfall of the previous days. Overall, the SS-MSW compost cover maintained the highest water content vs. reduction among the three covers made of compost measured during the monitoring period; the values measured for the sand cover were constant and about 2% *w/d*—typical for inert materials.

3.2. Gas Composition, Temperature and CH₄/CO₂ Ratio Profiles

The gas profiles measured in each cover (Figure 3) showed that the methane oxidation mainly occurred between 45 and 85 cm of depth, depending on the porosity and density of the material.

In the sand cover, oxygen was present up to high depth (95 cm), while the concentrations of methane and carbon dioxide, with very similar profiles, decreased rapidly between 75 and 45 cm of depth and then remained constant.

Oxygen concentrations in the mixed cover SS-MSW compost was close to zero up to about 65 cm of depth, and above this depth, the concentrations of CH₄ and CO₂ rapidly decreased. In this cover, methanotrophic bacteria seemed to be active and concentrated in a narrow zone between 85 and 55 cm of depth. The gas concentrations measured in the superficial probes were close to the detection limit.

A gradual decrease of gas concentration was measured in the SS compost cover in which oxygen could diffuse to greater depths. Oxygen, a limiting factor for the development of bacterial communities, was present at all the depths investigated. CH₄ and CO₂ concentrations showed a rapid decrease between 55 and 95 cm depth in this cover too.

In the MSW compost plot, oxygen concentrations decreased up to 75 cm and reductions of methane concentration were measured between 35 and 85 cm of depth.

These trends were similar to those observed by other authors both in the case of biocovers and biowindows (in practice, in those cases where oxygen was present, it was because it infiltrated from the surface atmospheric air without a prior dilution of the incoming gas, such as in the biofilters) [19,46,47].

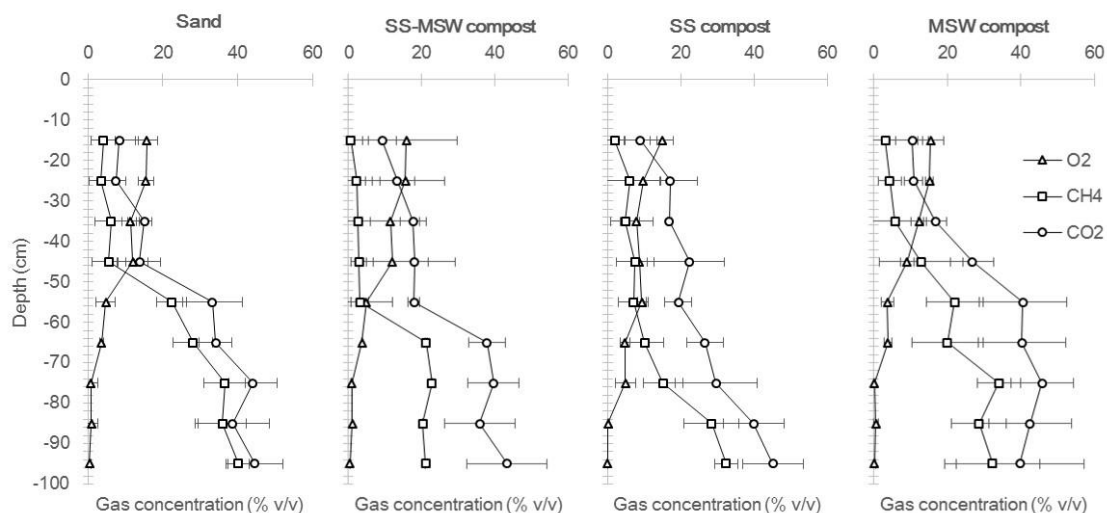


Figure 3. Average gas composition profiles.

Figure 4 compares the average profiles of temperature, CH₄/CO₂ ratio and H₂S concentration measured in each plot with reference to the average values calculated in the external probes (dashed line).

Results showed that the average temperatures followed a parabolic trend within all the experimental plots. Temperatures started to increase at 75 cm of depth with the vertex at 35 cm of depth and ranged between 19 and 36 °C; that could be considered an optimum range for methane oxidation [19,23,25,48].

All the experimental covers showed a progressive decrease of the ratio as a function of depth. The decrease of this ratio is a strong indicator of the consumption of natural gas [48,49]. The measured ratio for the external probes was similar to those recorded at depths of 95 cm in each cover. The values of the ratios found in the surface probes were 0.1 in the mixed SS-MSW compost cover; values were between 0.2 and 0.3 in the SS compost and MSW compost covers; and the highest value was recorded in the sand cover (0.54). The decrease of the CH₄/CO₂ ratio in the MSW compost and SS-MSW compost covers occurred mainly at 55 and 75 cm of depth. In the sand cover, characterized by higher ratios,

a decrease was recorded at 45 cm of depth; in the SS compost plot, the ratio decreased almost linearly with the depth.

Regarding the concentration of hydrogen sulfide, measurements showed that for all the covers, the concentrations in the higher probes were one order of magnitude lower than those recorded in the external probes. In fact, at 15 cm of depth, an average H_2S concentration of about 1 ppm was present in the sand, in the SS compost and in the mixed MSW-SS compost covers. In the MSW compost cover, there was a concentration of 3 ppm; 15 ppm of H_2S was found in the external probes.

It is important to notice that the concentrations of hydrogen sulfide measured at 95 cm depth were found to be different among the covers; in particular, an average concentration of 19 ppm was measured in the sand cover, 48 ppm in the mixed SS-MSW compost cover, 57 ppm in SS compost and 64 ppm in the MSW compost covers. This allows one to observe the effect of H_2S mitigation in biocovers, as other authors have found [17,19,34].

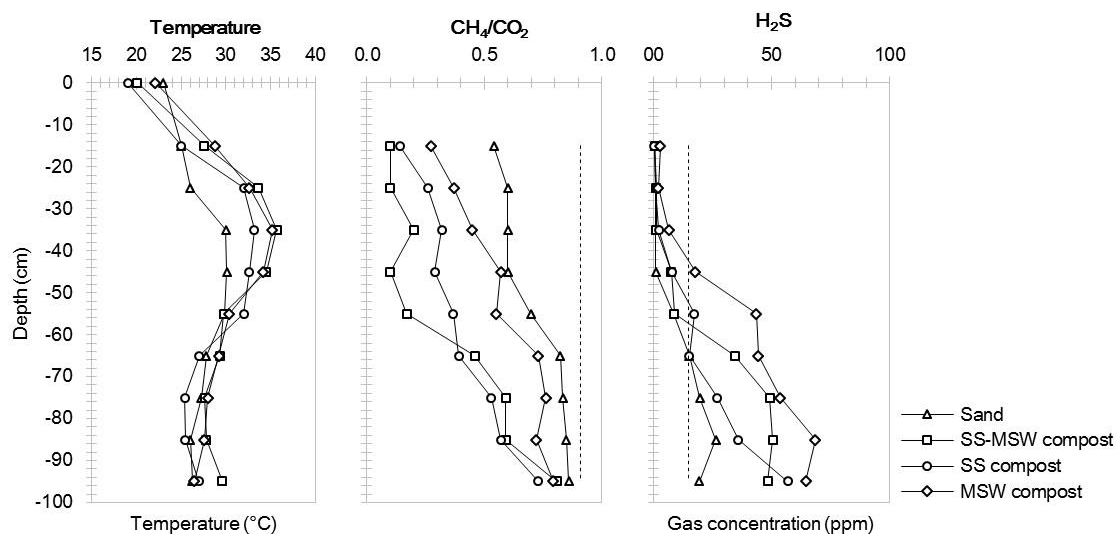


Figure 4. Average temperature, CH_4/CO_2 ratio and H_2S concentration profiles. Dotted lines represent the expected limits.

3.3. CH_4 Oxidation Efficiencies and Methane Emission Measurements

Starting from every over gas concentration profiles, the oxidation efficiencies were calculated for each cover according to Gebert et al. (2011) [14] (Figure 5). The results underlined that in each plot, the efficiency decreased with depth, indicating that the presence of oxygen in the cover allowed for the development of methanotrophic bacteria. Data showed that high efficiency was achieved in the last monitoring days, probably due to a better acclimatization of methanotrophs. However, the worst performances were observed on November 17, when high values of water content were measured in the cover as a result of heavy rainfall. The average total efficiency (corresponding to 15 cm depth) of the mixed SS-MSW compost cover was registered to be the highest (75%), followed by the efficiencies of SS and MSW compost plots (65% and 56% respectively). The maximum oxidation efficiencies were 100% in the SS-MSW compost plot, 96% in the SS compost and 81% in the MSW compost.

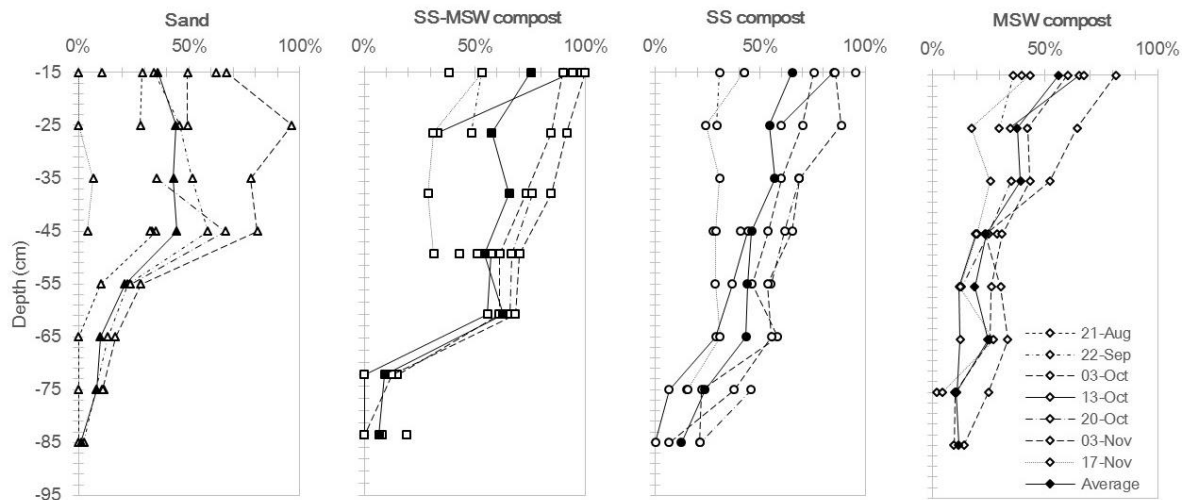


Figure 5. Methane oxidation efficiencies.

Figure 6 compares the average methane emissions measured inside and outside the trial biocover and the calculated average methane oxidation efficiency. Following a regular grid, about six flux measures inside and five measurements in the external surrounding area were done for each plot. Methane emissions in the SS compost cover were higher than the emissions measured in the other experimental plots, but overall the external methane emissions were the highest. Furthermore, as expected, higher methane emissions correspond to lower oxidation efficiencies.

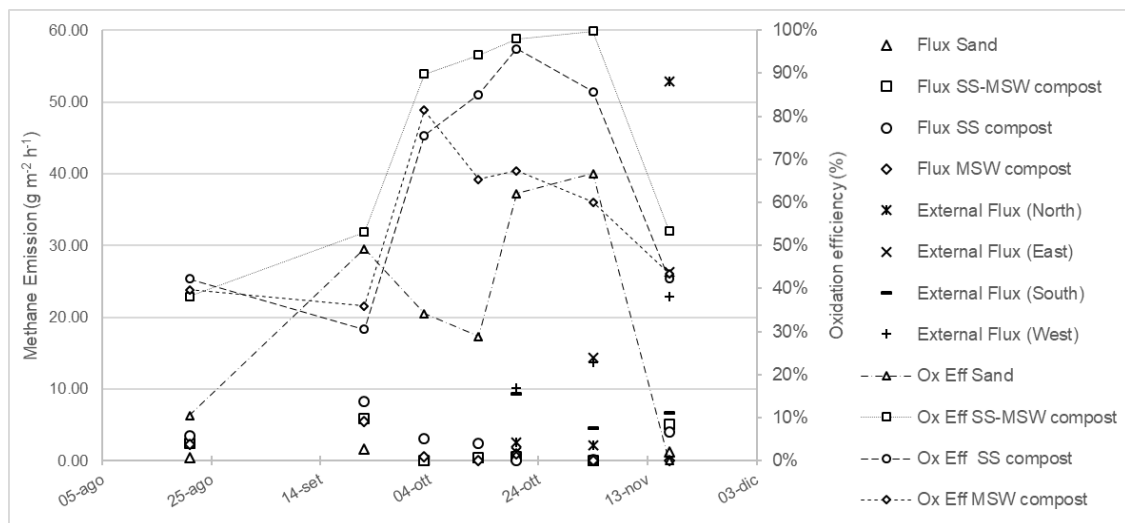


Figure 6. Methane emission measurements.

In Figure 6, it is possible to observe for the four different average filter media, different oxidation efficiencies of CH₄ over the whole monitoring period, and in particular: 36% ± 25% for sand, 75% ± 26% for SS-MSW compost, 65% ± 26% for SS compost and 56% ± 17% for MSW compost. The best result of the compost with respect to sand was indeed expected, since the compost contains the nutrients for the organic substances and an excellent porosity that allows for the passage of oxygen into the pores, even at a depth of 65–75 cm [25,49].

3.4. Column Tests

In order to determine the specific yields of the filter materials used in the experimental plots, some laboratory tests on samples of SS-MSW compost extracted from the biocover were performed. The objective of the tests was also to estimate parameters useful to implementing other systems of

biofiltration as active bio-filters. The tested flows were: 41 and 206 g m⁻² d⁻¹ in tests 1 and 2; and 82 and 312 g m⁻² d⁻¹ in test 3 and 4.

Observing the gas composition profiles of test 1 and test 2 (Figure 7), it can be noticed that the oxygen diffuses into the filter material only up to 30 cm of depth. Furthermore, in test 1 with a lower methane inlet load, the methanotrophic bacteria seemed to be active and concentrated in a larger zone; the methane concentrations measured at 10 cm of depth were in fact lower than those detected in test 2.

Gas composition profiles of the aerated tests, in which oxygen was supplied to inlet methane, show that the methane concentrations decrease with a linear trend with depth, probably because of the presence of active methanotrophic bacteria along the entire length of the column. The performances of the SS-MSW compost in test 3 and test 4, were similar, despite the increase of the inlet methane load.

The higher oxidation efficiencies were reached during the aerated tests as result of higher oxygen concentrations that allow for the activation of the entire column. In the aerated tests the higher efficiencies (94% and 87% in the tests 3 and 4 respectively) were reached between 30 and 20 cm depth; in the tests 1 and 2, the maximum efficiencies were more superficial. Furthermore, in tests 1 and 2, the oxidation efficiency decreases with increase of inlet load, and the aerated tests had comparable results.

The study in column tests shows that working with a gas diluted with incoming oxygen makes it possible to activate microbiological oxidation over the entire length of the column and thus increase oxidation efficiencies.

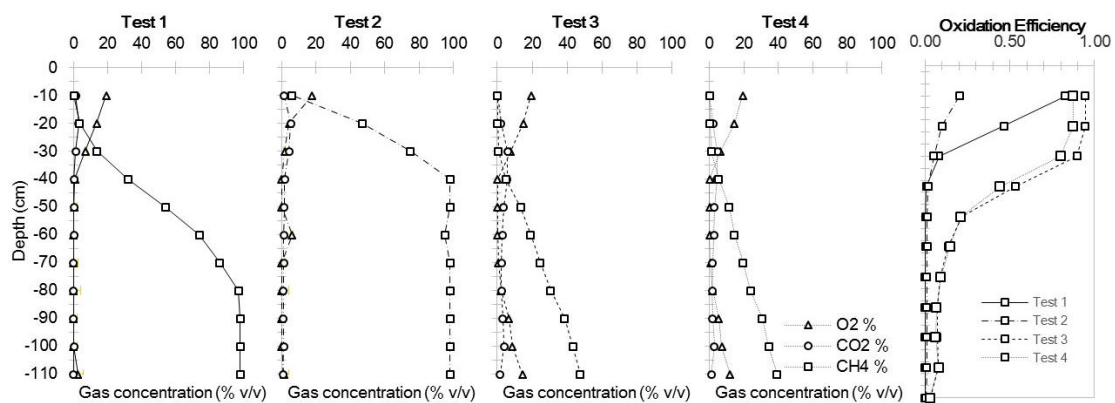


Figure 7. Average gas composition profiles and methane oxidation efficiencies in the column tests.

4. Conclusions

Field and laboratory investigations were carried out to study the possibility of mitigating biogas emissions' impacts by means of biofiltration systems. In particular, four pilot-scale biocovers made of compost from a source-selected organic fraction (SS compost), MSW compost from a mechanical biological treatment plant, mixed compost (SS-MSW compost) and sand were constructed and monitored in the daily cover area of the Casa Rota Landfill.

Results showed that the biocovers were an effective system to reduce landfill gas emission, especially for the mixed SS-MSW compost cover. Anyway, the worst performances were observed with no good atmospheric conditions and with low oxygen concentrations, proving that further investigations are necessary for the real applicability of biofiltration systems.

From the results it is also possible to conclude that compost is an excellent medium filter for measuring the effects of microbiological oxidation of methane emitted from landfill covers.

In laboratory trials, the methane oxidation efficiencies of the tested SS-MSW compost samples were higher when air was added to the inlet gas. Results hence suggested that, in real active biofilter systems, gas suction could be properly increased.

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Conflicts of Interest: The authors declare no conflict of interest.

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