Field-Scale Performance of Biochar-Amended Soil Covers for Landfill Methane Oxidation

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Revised manuscript submission to

Journal of Biomass Conversion and Biorefinery

Special Issue on "From Feedstock to Production of Biochar and its Application"

March 28, 2021

ABSTRACT

A field validation of three biochar-amended soil covers (2%, 10% and 100% biochar amended soil) along with a soil control cover was conducted within the intermediate cover of an active municipal solid waste (MSW) landfill in conjunction with laboratory studies evaluating the effect of biochar in enhancing methane (CH_4) oxidation in cover soils. Baseline CH_4 emissions and pre-existing site conditions were characterized prior to installation of test plots simulating three cover designs evaluated in related laboratory studies. Static chamber measurements of surface CH_4 fluxes and sampling of soil pore gas at different depths was conducted across the 8-month monitoring period to assess cover performance. Surface fluxes from the test plots exhibited wide spatial variability, with one location emitting fluxes > 1100 g CH_4 m^{-2} d^{-1} in one survey. Potential rates of CH_4 oxidation were determined in batch assays of exhumed soil core subsamples following termination of the field trial and ranged from ~1 to 350 µg CH_4 g^{-1} d^{-1}. The heterogeneity of the waste led to nonuniform CH_4 loads in the test plots. The soil control test plot was exposed to higher CH_4 loads and biochar amended test plots were exposed to significantly lower CH_4 loads. As a result, the soil control test plot showed higher CH_4 oxidation rates (257-289 µg CH_4 g^{-1} d^{-1}) than the biochar amended test plots. Similarly, the soil control plot also showed higher relative abundance of methanotrophs which was positively correlated with the CH_4 oxidation rates. The test plot with 10% biochar amended soil experienced CH_4 loads nearly 25% of that in soil control and still showed CH_4 oxidation rates (260 µg CH_4 g^{-1} d^{-1}) comparable to that of soil control which showed the efficacy of biochar amendment in enhancing CH_4 oxidation rates. The environmental and CH_4 exposure conditions affected the microbial community composition in the test plots and showed the dominance of Type I methanotrophic genus such as *Methylomonas* and *Crenothrix* spp. Overall, the waste heterogeneity led to

nonuniform CH_4 exposure conditions at each test plot making it hard to distinctly quantify the effect of biochar amendment on CH_4 oxidation rates.

Keywords: Biochar; biocover; landfill cover; field test; CH_4 oxidation

1 Introduction

Landfill gas (LFG) is generated during the anaerobic decomposition of waste after disposal in engineered landfills and is composed primarily of methane (CH_4) and carbon dioxide (CO_2) which are major greenhouse gases (GHGs). Municipal solid waste (MSW) landfills have emerged as a major contributor of GHG emissions over the past few decades [1-2]. CH_4 is of particular concern with respect to global climate change due to its relatively high potential for atmospheric warming (~25 times greater than CO_2) [3]. In newly constructed U.S. landfills, LFG must be captured before it reaches the atmosphere via active gas extraction systems. However, there remain many old and abandoned landfills for which installation of active gas extraction system is neither economical nor practical. At these sites, several organic amendments (e.g., compost, sewage sludge and biochar) to landfill cover soil have been investigated as a passive means of promoting microbial CH_4 oxidation and removal [4-7]. Biochar, a solid byproduct generated during anaerobic biomass pyrolysis, has recently garnered interest for both agricultural and geoenvironmental applications owing to its unique physical and chemical properties, such as its high surface area and porosity, and ability to adsorb a variety of compounds, including CH_4 found in LFG [8-10]. Biochar amendments to cover soils are also an economic solution to reduce CH_4 emissions from landfills as they are composed of a more stable form of carbon than other organic amendments (a longer effective lifetime) and implementation is relatively inexpensive if feedstock materials are locally available.

An important aspect of this work is the assessment of the feasibility and effectiveness of biochar-amended landfill covers under field conditions. While large column incubation tests can be used to recreate field conditions in the laboratory, the impacts of seasonality and precipitation events, as well as waste heterogeneity on CH_4 oxidation are difficult to simulate in the laboratory

and must be evaluated in field trials. Moreover, performance metrics obtained from laboratory studies often do not accurately reflect the performance of the same cover in the field due to the additional external factors impacting CH_4 oxidation in field situations [11]. Prior field studies on compost-based biocovers have found substantial variability between potential CH_4 oxidation rates in various cover substrates in laboratory settings as compared to those observed in the field [12]. As such, field-scale testing of a proposed alternate cover design is essential to understanding site-specific considerations for optimal CH_4 removal.

Field validation of three design configurations of biochar-amended soil covers, evaluated in laboratory column tests [13-14], was conducted at an active landfill in northeastern Illinois (USA) in order to evaluate the actual performance of these covers under dynamic field conditions throughout the year. In doing so, the effects of important parameters such as average temperature, precipitation, and natural variability on the overall performance of the biocover was examined by correlating the variation in temperature, precipitation, and natural variability with the CH_4 emissions rates, CH_4 oxidation rates and microbial community distribution.

2 Materials and methods

2.1 Site selection and background

An active landfill site in northeastern Illinois (USA) was selected for field-scale implementation of the biochar-amended soil covers designed and evaluated in Phase I and Phase II testing [13]. The landfill was actively accepting both MSW and some construction and demolition (C&D) waste from the Chicago metropolitan region throughout the testing period. The total landfill footprint was estimated to be ~20 to 25 hectares (~50 to 60 acres), making it one of the largest active MSW landfills in the area. Prior to implementation, relevant background information for

the site was obtained, including the approximate age of the landfill and average age of waste below the test site; details regarding landfill configuration and geometry; and information on the gas extraction system currently operating at the site (i.e., locations of gas extraction wells and average negative pressures of the extraction wells). At the test plot location, the waste was last received at approximately 1.5 years prior to the start of the field test. Nominal vacuum pressure applied for the gas extraction wells according to the landfill operator was 85 inches of water column. Meteorological data was obtained from a nearby airport (Waukegan Regional Airport) and recorded during each site visit. The soil used for the intermediate and final covers comprised of glacial till mainly silty clay. The landfill cover soil at some locations of the site had a high proportion of large, gravel-sized particles mixed with clay.

A location within the intermediate cover of the landfill upwind of the active filling zone was selected for the installation of the test plots. Prior to installation, the soil properties of the existing intermediate cover were characterized and the depth to the underlying waste was estimated by manual excavation of the existing cover until the underlying waste layer was reached. The existing intermediate cover soil thickness in the test area ranged from 0.9 to 1.5 m (3 to 5 ft) at locations closer to sloped areas. Existing cover soil generally contained high proportions of stiff clay as well as gravel and pebble-sized particles.

2.2 Baseline emissions survey

After finalizing the test plot locations, a baseline survey of the surface emissions from the existing intermediate cover was performed in order to establish the pre-existing conditions and surface fluxes at the site prior to installation of the biochar-amended cover plots. The static chamber flux method was employed to measure CH_4 emissions out of the landfill cover before

and after installation of the test pad. The static flux chamber has been considered an appropriate and simple tool to assess landfill CH_4 emissions [12, 15-18]. A total of three baseline survey campaigns were conducted prior to installation of the test plots which are summarized below:

- Survey 1: During survey 1, three transects (designated as transect A, B and C) were drawn across the study area and surface fluxes of CH_4 were measured at 18 distinct locations using the static chamber technique as shown in **Fig. 1**.
- Survey 2: A second survey was conducted to assess the physical characteristics of the existing intermediate cover soil at 3 locations along the transect A which are represented by trenches A, B and C (**Fig. 1**). The soil pore gas was sampled at depths of 0.30, 0.60, 0.90 and 1.20 m (1, 2, 3 and 4 ft) by installing gas probes clusters (GP 1-3) at three different location between the transects A, B and C (**Fig. 1**).
- Survey 3: A third and final baseline emission was conducted within the test area that included surface flux measurements at 6 random locations as well as from the 3 gas probe clusters (GP 1-3) installed in the second survey (**Fig. 1**). For this survey, random locations were selected for surface flux measurements over the test area. The gas samples were withdrawn from gas probes using 10 mL syringe fitted with stopper and a filter.

All gas samples were stored in evacuated 10 mL glass vials sealed with butyl rubber septa prior to analysis on a HP 6890 gas chromatograph equipped with a flame ionization detector operated at 40 °C. Additional details regarding gas analysis procedures are given in Yargicoglu [11].

2.3 Static chamber flux measurements

In chamber flux measurement method, the surface mass flux of CH_4 (J_{CH_4}) is determined from the change in headspace CH_4 concentration over time in the sealed headspace of the chamber using the following equation:

$$J_{CH_4} = \frac{dC}{dt} \times \frac{V}{A} \quad (1)$$

where dC/dt is the rate of CH_4 loss in the headspace in ppmv/min; V is the chamber volume in m^3; and A is the chamber base area in m^2. The change in CH_4 concentration over time (dC/dt) was determined from the linear slope of CH_4 concentration (ppmv) vs. time (minutes) plot for each trial. The ideal gas law ($PV = nRT$) was used to convert ppmv CH_4/min to g CH_4/min at the average temperature recorded during sampling. This was then converted into mass fluxes per unit area for units of g CH_4 m^{-2} day^{-1}.

The following procedure was followed for all static chamber flux measurements:

1. At each designated location (**Fig 1**), the bottom anchor of the static chamber was pounded firmly into the ground using a sledgehammer. The chamber top was placed and sealed with three large clamps. Deionized water was poured around the lip of the chamber tops to check the leakage at the chamber joint.
2. Samples were withdrawn using a syringe equipped with a luer-lock 3-way stopcock and needle from the sampling port at the top of the static chamber. For each trial, a sample was withdrawn immediately after closing the chamber (t = 0) and every 4 minutes for a total of 5 to 6 points over 16 to 20 minutes. Chamber tops were removed between trials for a minimum of 10 minutes to allow sufficient mixing with atmospheric air prior to the successive trial.
3. Once sampling was completed for a given location, the static chambers were disassembled, and samples were returned to the laboratory for analysis via gas

chromatography (GC). Samples were stored in evacuated vials no longer than 48 hours prior to analysis. The dC/dt was obtained from linear regression of CH_4 concentration versus time plot and then the CH_4 flux was calculated according to **Eq. 1**. Slopes with R^2 values above 0.8 were considered acceptable and fluxes with R^2 below 0.7 were not included when determining average flux values.

Measurements of biocover emissions were compared to the baseline flux values in order to assess any emission reductions attained during biocover operation.

2.4 Measurement of field gas profiles

In addition to the surface flux measurement, gas profiles at different depths in the cover were evaluated by collecting samples through gas probes and analyzed via GC. The gas probes were made of hollow stainless steel tubes of internal diameter (ID) 6.25 mm (0.25 in.) of varying lengths. The gas probes had perforations at the bottom to collect gases from the targeted depth. The probes had gas sampling ports at the top, made of Ultra-torr fittings with a butyl rubber septum. Gas was withdrawn through these ports during sampling campaigns after flushing the air space within the probes several times with LFG to obtain a fresh gas sample. Data on gas profiles was used to aid interpretation of measured fluxes and allow inferences on the zones of greatest CH_4 removal within the experimental covers.

2.5 Biocover test pad design and installation

A schematic of each test plot profile is shown in **Fig. 2**. The cover profiles were tested in duplicate making a total of eight test plots which are summarized below:

- P1/P5: Soil control

- P2/P6: 2% biochar-amended soil at 0.15 to 0.30 m (0.5 to 1 ft) depth below ground surface (bgs)
- P3/P7: 100% biochar layer of 0.025 m (1 inch) thick at 0.15 m (0.5 ft) depth (bgs)
- P4/P8: 10% biochar-amended soil at 0.15 to 0.30 m (0.5 to 1 ft) depth (bgs)

Biochar-amended soil cover test plots were installed following the completion of baseline survey, and the surface CH_4 flux and gas profiles at the test plots were monitored for ~8 months. A total of eight test plots each covering an area of 1.5 m^2 (16 ft^2) were installed within the existing intermediate cover to a depth of 0.90 m (3 ft) (total biocover area = 12 m^2). At the location of each test plot, existing interim cover soil was excavated until the waste layer was reached. The waste was visually observed, and it was noted that the waste conditions varied at each test plot location, which did not provide the same gas emissions from the waste. Then, the excavation was backfilled so that all test plots had 0.30 m (1 ft) of intermediate cover soil overlying the waste. This layer of soil was placed due to the landfill operator's concern of unintended CH_4 releases during field trials; hence a 0.30 m thick soil layer was placed above the waste to minimize the risk of any unwanted CH_4 releases. This step also helped to homogenize the thickness of soil below each test plot. A 0.30 m thick layer of pea gravel was placed above the existing intermediate cover soil layer to serve as the gas distribution layer. In the test plots with biochar amended soil layers, a 0.15 m thick topsoil layer was provided as an erosion protection layer.

2.6 Initial sampling and characterization of biocover substrates

Physical and chemical properties were characterized for the initial (before installation of the test plots) and terminal samples (after exhumation) of biocover substrates. For the initial samples, bulk samples of each layer were taken while placing them in the test plots. For the terminal

sampling, core samples were retrieved from the center of each test plot by a stainless-steel Shelby tube. Additional bulk samples were taken from the upper (0 to 0.30 m) and lower depths (0.30 to 0.60 m) of each test plot. Physical and chemical properties of the samples were determined according to ASTM standards as follows: moisture content (ASTM D2216); specific gravity (ASTM D854); particle size distribution (ASTM D422); organic matter content via loss on ignition (ASTM D2974); pH, oxidation-reduction potential, and electrical conductivity (ASTM D4972). Because sampling at depth during the experimental period would introduce disturbances in the cover physical structure with potential impacts on microbial community activity, detailed physical, chemical, and microbiological characterization was limited to initial and terminal sampling.

2.7 Long-term monitoring of test plots

Long-term monitoring of biocover performance was undertaken over eight field campaigns conducted throughout the eight-month test duration. Emissions were measured at the center of each test plot for three consecutive trials using the static flux chamber as described previously. In addition to fluxes of CH_4 and CO_2, micrometeorological conditions were also monitored over the experimental period. Monitored climatic parameters include barometric pressure, precipitation events, and average daily air and ground surface temperature. Surface measurements of ground temperature and volumetric water content (VWC) were taken during flux measurements using a soil thermocouple and a Delta-T soil moisture sensor using time-domain reflectometry.

2.8 Batch incubation testing

Soil samples were refrigerated at 4 °C until subjected to batch testing in the laboratory to quantify potential CH_4 oxidation rates of soils sampled from the test plots after eight months of

exposure to the LFG. CH_4 oxidation rates were determined via batch incubation testing in clear 125 mL serum vials (Wheaton Glass, Milville, NJ). All vials and sampling equipment were sterilized in an autoclave (Napco 8000 Model DSE) at 121°C and > 1.5 bar for one hour to prevent cross-contamination among samples or equipment. All batch tests were run in duplicate or triplicate. Rates of gas consumption or production were determined from linear regression analysis of the change in volumetric gas concentration over time (dC/dt) based on zero-order kinetics observed during batch testing. Volumetric gas concentration gradients were converted to mass gradients using the ideal gas law ($PV = nRT$). Statistical analysis of batch test results (t-tests for equivalency of sample means; $\alpha = 0.05$) was performed using OriginPro™ (version 9.1) software.

2.9 DNA extraction and genetic analyses

Additional subsamples were frozen at -20 °C in sterilized 5 mL vials for DNA-based assays aimed at characterizing the microbial community that had developed after addition of biochar to the existing intermediate cover soil. DNA was isolated from ~0.3 g of soil collected from exhumed field samples using the MoBio Power Soil DNA Isolation Kit (Cat. 12888-100) according to manufacturer's instructions. DNA samples were then amplified using the 515F/806R primer set targeting the 16S rRNA genetic region and subjected to next-generation sequencing on an Illumina BioSystems platform for taxonomic classification and analysis. The total abundance of bacteria and methanotrophic bacteria can be qualitatively compared among samples by comparing the total number of sequences matches in terms of operational taxonomic units (OTUs) to known taxonomic groups within each sample.

3 Results and discussion

3.1 Baseline survey of surface emissions and gas profiles

The baseline surface CH_4 flux at different locations during surveys 1 and 3 are summarized in **Table 1**. CH_4 concentrations measured along the depth of existing intermediate cover soil during survey 3 are shown in **Table 2**. In general, the CH_4 emissions at the test site prior to any cover modifications were low, with average flux ranging from negative values in survey 1 (-0.013 g CH_4 m^{-2} d^{-1}) to relatively low positive values (~0.28 g CH_4 m^{-2} d^{-1}) in survey 3. Overall average surface emissions during surveys 1 and 3 was ~0.114 g CH_4 m^{-2} d^{-1} (n = 17). If only positive flux data are considered, the average surface flux increase slightly to 0.14 ± 0.5 g CH_4 m^{-2} d^{-1} (n = 14). These data were obtained during the summer months when oxidation rates are expected to be relatively high in cover soils and reduced CH_4 emissions are often observed [11, 19-20]. The other reasons for lower surface flux could be the presence of a thick interim soil cover and an active gas extraction system in operation at the landfill. In addition, a relatively higher moisture contents in the intermediate soil (~14-20 % (*w/w*)) could also have led to lower surface emissions.

The CH_4 concentrations varied significantly at the three locations. The CH_4 concentration at a depth closer to the waste varied from 0.007% to 56% (*v/v*) at three different locations which could be attributed to the heterogeneity of the waste (as observed during the excavation during the test plot construction). The CH_4 concentrations were consistently lower near the ground surface at all three locations (0.004% to 0.047% at 0.30 m depth below ground surface) which could be partly due to CH_4 oxidation within the intermediate cover soil and partly due to dilution from atmospheric air ingress.

3.2 Physical and chemical properties of test plots and existing intermediate cover

In general, a high variability in physical soil characteristics was observed within the existing intermediate cover soil (**Table 3**), leading to significant spatial variability within the test area. USCS classification for the existing soil cover ranged from clayey gravel (GC) to low plasticity clay (CL). In general, all plots had a high proportion of gravel (P1 35.3%, P3 42.4%, P4 27.5%, P5 32%, P7 24.7% and P8 33.8%), except for soils sampled from P2 and P6 which had lower gravel content (P2 13.2% and P6 4.7%) and higher proportion of sand and fines (silt and clay). The initial physico-chemical properties within each relevant soil layer of each test plot are given in **Table 3.**

The organic matter content (OC) of the existing cover soil ranged from ~3.8 to ~5% in the upper soil layers without any biochar amendment (0 to 0.15 m depth) (**Table 3**). Due to the high heterogeneity of the existing cover soil, the effect of biochar on the overall OC of the soil was not uniform across all test plots, though a clear increase in OC was observed in the 2% and 10% biochar-amended soil layers in test plots P2/P6 and P4/P8, respectively (**Table 3**). Initial average OC in the 2% biochar-amended soil layers in P2 and P6 and 10% biochar-amended soil layers in P4 and P8 were ~7.63% and ~11.44%, respectively, much higher than the unamended soil layers. Thus, the addition of biochar to the cover soil led to an increase in OC that ranged from approximately 3 to 7% overall. The average initial pH values of the cover soil did not vary significantly among the treatments and was within the optimal range previously reported for methanotrophic bacteria [21]. The pH ranged from 7.29 to 7.39 in the various test plots across various depths.

3.3 Meteorological monitoring data

Meteorological data obtained during each emission survey are presented in **Table 4** which includes measurements during late summer through early spring. The temperature varied from

~0.5 to 27 °C and VWC from ~3 to 20% (*v/v*) during this period. As will be shown in the following sections, climatic factors especially soil and air temperature have a large influence on rates of LFG generation as well as CH_4 oxidation in cover soils.

3.4 Surface CH_4 emissions

Surface CH_4 emissions from each test pad were monitored over the course of 8 monitoring campaigns (or surveys) during 8-month period, which are summarized in **Table 5**. Both negative and positive CH_4 fluxes were observed, with high spatial variability. Although the test plots were constructed in duplicate, the surface emissions could not be replicated due to the existing heterogeneity in the waste and intermediate cover soil. Negative fluxes were observed which ranged from -15.9 to -0.06 g CH_4 m^{-2} d^{-1} in test plots P2, P3, P4 and P5, suggesting net CH_4 uptake by the soil. Plot 1 (soil control) mostly showed higher CH_4 fluxes (max. flux 199.4 g CH_4 m^{-2} d^{-1}), however Plot 5 which was the duplicate of Plot 1 showed significantly lower flux rates (max. flux 10.4 g CH_4 m^{-2} d^{-1}) as shown in **Table 5**. This clearly shows the heterogeneity in the waste which was also observed during the test plot construction. Similarly, lower fluxes were observed in the test plots P2, P3 and P4 which had biochar amended soil layers. Again, their duplicate test plots (P6, P7 and P8) showed relatively higher CH_4 fluxes with P7 showing highest flux (max. flux ~1133 g CH_4 m^{-2} d^{-1}) (**Table 5**). It is hard to distinguish the effects of biochar amendment on the CH_4 oxidation potential of the landfill cover soil due to the inherent heterogeneity of the waste and variable CH_4 loading on each test plot. As stated earlier, to avoid the risk of CH_4 release during installation of the test plots, a 0.30 m (1 ft) thick intermediate cover soil layer was placed just above the waste at each test plot (**Fig. 2**). This layer may have restricted inflow of the LFG from waste upward into the test plots and it may also have

contributed to partial CH_4 oxidation. Since, the existing intermediate cover soil was highly heterogeneous, it could also have contributed to the variable CH_4 fluxes through the different test plots.

Overall highest average surface CH_4 emissions were observed at P7 (thin (0.025 m) biochar layer at 0.15 m depth). Average surface emissions at P7 ranged from 4.7 to 1132.8 g CH_4 m^{-2} d^{-1}, with the highest individual flux measurement (~1289 g CH_4 m^{-2} d^{-1}) recorded in late summer (survey 1-Sept 2^{nd}) under relatively warm and dry weather conditions. The CH_4 emissions gradually reduced with time which could be due to the onset of CH_4 oxidation in the soil. The initial high CH_4 emission could be due to the low CH_4 oxidation because of lag phase experienced by the microbes to acclimate to the test plot environment. Although, reduction in CH_4 emissions was observed after placement of the test plots, the CH_4 emissions did not fall to zero which could be due to the persistent lower temperatures during the testing period (avg. 11.2 ℃) as shown in **Table 4**. Temperature is found to have profound effect on CH_4 oxidation rates and optimum temperature for CH_4 oxidation ranges from 25-35 ℃ [11, 21]. The VWC in the initial four surveys conducted between September to December were lower (3.03 to 6.96% *v/v*) whereas it was higher during the last four surveys conducted during March and April (7.36 to 19.55% *v/v*) (**Table 4**) which was due to the high precipitation events which occurred during that period. The high soil moisture content could also be a reason for lower CH_4 fluxes during later phases of the survey impeding the diffusive flux of LFG through the cover [22].

Overall, CH_4 fluxes observed at the site fall within the wide range of previously reported CH_4 emissions from landfill covers, ranging from < 0.0004 to > 4000 g m^{-2} d^{-1} [23].

3.5 Average gas profiles

Gas profiles were determined at 5 of 8 test plots (P1 to P4 and P8) during each monitoring campaign by sampling soil pore gas from gas probes clusters installed to 4 depths (0.30, 0.60, 0.90 and 1.20 m bgs) at each location. In some instances, gas samples could not be withdrawn from the probes due to the presence of water in the cover soil at depth of measurement (due to the precipitation event), and in some cases low gas volume within the probe was also encountered, preventing retrieval of soil gas at that depth.

Average gas profiles along the depth of each test plot across the monitoring period are illustrated in **Figure 3**. A clear difference between the test plots can be seen however, the effect of biochar amendment cannot be delineated due to the heterogeneity of the waste leading to variable gas production rates. Soil control plots (P1) generally had elevated CH_4 and CO_2 concentrations throughout the cover depth across the monitoring period. A particular trend was not detected in the CH_4 concentration profile across the soil depth in P1 which may be due to the variability in the soil properties within the cover. The test plots with biochar amended soil layers (P2 to P4) except for P8 consistently showed lower CH_4 concentrations across the depth of the cover (**Figure 3**). However, the difference between gas profiles of duplicate plots (P4 and P8) clearly shows difference in gas generation rates owing to the waste heterogeneity at the test plot locations and hence makes it hard to derive a strong correlation between biochar amendment and CH_4 oxidation.

3.6 Terminal properties and potential CH_4 oxidation rates of test cover materials

At the end of the monitoring period, soil cores and bulk soil samples were taken from each test plot and characterized for field moisture content, bulk density, organic matter content, and CH_4 oxidation potential in batch assays. Physical properties of the samples from soil cores and bulk

samples obtained from various depths of test plots are summarized in **Table 6.** Field moisture contents were relatively high during terminal sampling due to a recent rain event, ranging from ~14 – 20% (*w/w*) within the soil cores (**Table 6**). Samples were taken from soil above the biochar-amended layers as well as within the biochar-amended soil layers to compare the effect of treatment type on the CH_4 oxidation capacity of the microbial community at field moisture contents. CH_4 oxidation rates for each upper unamended soil layer (0 to 0.15 m) and the underlying treatment zone (0.15 to 0.30 m) for each test plot are shown in **Fig. 4**.

The soil control (P1 and P5) showed higher CH_4 oxidation rates (257-289 µg CH_4 g^{-1} d^{-1}) than the treated soils which is attributed to the exposure to higher CH_4 concentrations in the soil control (**Fig. 3**) likely leading to higher abundance of CH_4-oxidizing bacteria [24]. The other tests plots showed lower CH_4 oxidation rates which can be attributed to the lower CH_4 exposure in the field. Röwer et al. [25] also observed lower CH_4 oxidation capacities (< 8.64 g CH_4 m^{-2} d^{-1}) in soils with lower exposure to CH_4 in the field. The 10% biochar amended soil in P8 showed higher CH_4 oxidation (265 µg CH_4 g^{-1} d^{-1}) (**Fig. 4**) which again shows strong correlation between CH_4 availability and abundance of methane-oxidizing bacteria (MOB) as P8 had higher CH_4 concentrations across the cover depth than its duplicate test plot P4 (**Fig. 5**). Very short lag periods (on the order of 1 to 4 hours) were observed for all field samples during batch incubation, even after storage for > 1 week at 4 °C (**Fig. 6**). This indicates that the CH_4-oxidizing bacterial communities were well-developed and capable of withstanding extreme shifts in soil temperature and moisture.

CH_4 oxidation rates observed in this study were within the range of previously reported rates for landfill cover soils (~1 to 644 µg CH_4 g^{-1} d^{-1}). CH_4 availability was found to have profound effect on CH_4 oxidation rates which is consistent with other studies [26]. A t-test

assuming equal variances among P1 and P5 batch test units was performed to test equality of means, and the results indicate the two populations were not significantly different among the control plots (p = 0.23; p = 0.153 if unequal variances are assumed). Thus, the oxidation rates achieved in the unamended soil plots were not statistically significantly different, despite the much higher surface emissions measured throughout testing at P1 relative to P5.

Evaluating the effect of biochar treatments on the potential oxidation rates observed in batch incubations was limited by the strong spatial variability observed at the site. Indeed, several other studies have also noted that extreme spatial and temporal variability with respect to LFG concentrations and surface fluxes commonly observed in landfill settings can complicate assessment of cover effectiveness and the relationships among soil properties and CH_4 emissions [20, 25, 27].

3.7 Microbiological characterization of select field soils

Select field samples taken from terminal soil cores from test plots P1, P2, P3 and P4 were analyzed for microbial community characteristics. The microbial as well as methanotrophic bacterial abundance on the sequenced samples are summarized in **Table 7**.

Soil samples from all four test plots (P1 to P4) were inhabited by MOB resulting in significant relative abundance of methanotrophs in the samples. Among the four test plots, the soil control plot (P1) had the highest methanotrophic abundance ranging from 34.5% to 75. 29% (**Table 7**). In P1, the methanotrophic abundance increased with depth, however, the same trend was not noticed in other test plots (P2 to P4). In test plot P2, which had a lower CH_4 oxidation rate in batch assays relative to P1 soils, the average relative abundance of methanotrophic genera was only ~14.36%. At P3, percentages of MOB were even lower than at P2, ranging from ~3.6 to ~5.5%. P4 had relatively higher relative abundance of MOB than P3. P2 and P4 which had

biochar mixed soil layers showed comparable MOB relative abundance. The lower MOB abundance in test plots P2-P4 could be due to the lower CH_4 availability at those locations as shown in **Fig. 3**.

Major fraction of MOB belonged to the class *Gammaproteobacteria*, with a high number of species belonging to the genus *Methylomonas* (Type I MOB) observed especially in P1. In addition, the genera *Methylomicrobium* and *Methylobacter* were also observed, though in lower abundance as compared to the *Methylomonas* in P1, which appeared to be a dominant methanotroph in this soil. *Methylomonas* was found in the samples from P2, P3 and P4 as well, although the relative abundances were lower. Another dominant genus observed in most samples was *Crenothrix*. It is interesting that the relative abundance of genera *Methylobacter* was relatively low which otherwise is generally found to dominate the methanotrophic populations in the landfill cover soils at a temperature range of 23-30 °C [28]. The persistent low temperatures at the landfill test site could have favored the growth of *Methylomonas* over *Methylobacter.*

Overall, the trends among species present within each sample appeared consistent among the field samples sequenced, with the genus *Methylomonas* being most predominant, followed by *Crenothrix*. Both these genera belong to the same order *Methylococcaceae* within the class *Gammaproteobacteria* and have been previously detected in landfill cover soils [29]. Moreover, species within both genera are known to be capable of cyst formation, which is especially useful for maintaining survival in dynamic environments and across seasonal changes in soil moisture and temperature [21].

Methanotrophic abundance is affected by CH_4 availability and exposure duration in the field [30] which is consistent with the observations made in this study. A positive correlation among CH_4 oxidation rates in batch assays and total percentage of methanotrophic genera

detected in the DNA isolates based on 16S rRNA taxonomy was observed as shown in **Fig. 5**. It provides further support for the hypothesis that the differences in CH_4 oxidation rates observed in the field soils are a direct result of differences in methanotrophic abundance, likely due to differing levels of CH_4 exposure during the field trial which was the outcome of waste heterogeneity in the landfill. Similar results have been observed in other studies, most recently by Gebert and Perner [31], who observed an increased abundance of MOB along preferential flow pathways in landfill covers due to greater cumulative CH_4 exposure at those locations. Moreover, a number of other studies have reported higher oxidation rates in materials due to higher prior CH_4 exposure and resultant increases in methanotrophic populations [18, 32-33].

3.8 Effect of field conditions on biocover performance

Surface emissions from the test plots exhibited high degrees of spatial and temporal variability, hindering the evaluation of the effect of the different cover treatments on the overall cover performance in the field. Results of surface emission measurements over time showed elevated emissions occurring in early fall and early spring (**Table 5).** As shown in **Fig. 7**, air temperature was found to be positively correlated with both the maximum and average measured CH_4 flux during each monitoring survey (R^2 = 0.87). It should be noted that the correlation between temperature (either air or ground surface) varied significantly among individual plots and the correlations noted above correspond to either maximum or average fluxes across all plots for a given survey. This likely reflects the fact that higher temperatures result in higher LFG generation rates within the landfill, as well as drying of the soil near the surface giving rise to formation of cracks and thus preferential flow paths. The generally drier soils in the summer months may also limit the microbial oxidation of CH_4 during the summer months [27]. In general, lower fluxes are observed at higher temperatures due to increased CH_4 oxidation in soils

[27]. However, in this study, the test plots were set up during warmer climate (early September) when the LFG generation rates are normally high but simultaneous higher rate of CH_4 oxidation could not be obtained due to the potential lag phase of the microbial activity thereby resulting in higher CH_4 fluxes. The reduced flux with temperature could be due to reduced CH_4 generation as well as increased CH_4 oxidation with the acclimatization of MOB in the soil environment.

CH_4 exposure in the field was also found to have a direct impact on the capacity for CH_4 oxidation during terminal batch assays. Higher surface emissions associated with higher rates of CH_4 oxidation in terminal batch assays (especially in P1) and higher abundance of MOB relative to the total bacterial population. Exposure to seasonal variation may have led to the development of a more robust soil microbial community as compared to that cultivated in laboratory soil columns, as activity in terminal batch assays from field soil cores resumed much more quickly upon re-exposure to CH_4 than laboratory soils evaluated in Yargicoglu [13]. Similar observations were made by Henneberger et al. [33] and Krause et al. [34]. They observed that in environments with high variability in physical and chemical conditions, a "microbial seed bank" within the soil of dormant microorganisms that become active when favorable conditions return is critical to maintaining optimal functioning over seasonal and temporal changes. The presence of a viable microbial "seed bank" within the field cover soil may be responsible for the much shorter lag phases observed in the field batch assays as compared to laboratory incubations of samples taken from soil columns subjected to controlled, relatively static conditions [13].

4 Conclusions

Four different cover profiles, three with biochar amendment at different proportions and one soil control, were tested at a landfill by exposing the cover profiles to LFG generated from the waste

for 8 months (early September to late April). The existing waste heterogeneity in the landfill led to variable CH_4 fluxes through the test plots and thus affected the performance of the tested biocover profiles. Although biochar amendment was strongly correlated with elevated CH_4 oxidation rates in laboratory column experiments, the same could not be replicated in the field conditions due to the low CH_4 availability at the biochar amended test plots leading to lower methanotrophic abundance and thus lower CH_4 oxidation rates. However, despite being exposed to low fluxes of CH_4, 10% biochar amended soil showed significant CH_4 oxidation rates in terminal batch incubation confirming the positive effect of biochar amendment. In contrast, the soil control plot which was exposed to higher CH_4 load from the waste mass showed higher CH_4 oxidation rates. This shows that the CH_4 oxidation rates are dependent on the CH_4 exposure conditions and CH_4 loading rates. The microbial surveys also confirmed that exposure to higher CH_4 load gives rise to the development of higher microbial abundance as shown by significantly higher relative abundance of methanotrophs in soil control. The study showed dominance of *Methylomonas* (Type I methanotroph) in all the samples instead of *Methylobacter* which is generally found as a dominant Type I methanotroph in laboratory incubation studies at optimal temperature (23-30 °C) and wide range of pH conditions [35-36]. This shows that the seasonal variation in temperature and moisture affects the microbial community composition in the field. The relative abundance of MOB varied significantly across the treatments and was positively correlated with potential oxidation rates observed in terminal batch assays.

Additional controlled field pilot study is warranted with homogeneous waste and cover soil conditions to accurately quantify the impacts of the biochar-amendment on the microbial community composition and CH_4 oxidation under long-term conditions. From this study, it can be shown that the activity of methanotrophic bacteria in landfill cover soils is strongly influenced

by environmental factors, such as soil moisture, average air and ground temperature, and characteristics of the waste. The extent of prior CH_4 exposure may be one of the most critical factors to affecting the potential oxidation rate of cover soils, though control on this parameter is difficult to achieve in passive cover systems.

Author contributions: Krishna R Reddy: Research conceptualization, funding, experimental planning, supervision, reviewing, and editing.

Erin Yargicoglu: Literature searching, experimental planning, performing experiments, data analysis, and writing

Jyoti K Chetri: Literature searching, writing, data plotting, reviewing, and editing

Funding information: This research project was funded by the U.S. National Science Foundation (Grants CMMI #1200799 and CMMI #1724773).

Compliance with ethical standards

Conflict of interest: The authors declare that they have no conflicts of interest

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Table 1. Summary of baseline surface methane emissions measured prior to removal of 30 cm of existing intermediate cover and installation of test plots.

	Survey 1	Survey 3	Surveys 1+3
No. of Locations	18	6	24
Total No. of Trials	18	16	34
No. of Samples Accepted	10	7	17
No. of Positive Fluxes	8	6	14
No. of Negative Fluxes	2	1	3
Avg. Surface CH_4 Flux (g m^{-2} d^{-1})	-0.0024 ± 0.008	0.279 ± 0.683	0.114 ± 0.46
Positive CH_4 Fluxes (g m^{-2} d^{-1})			
Min.	2.95×10^{-6}	2.56×10^{-6}	2.56×10^{-6}
Max.	2.55×10^{-3}	1.95	1.95
Mean ± SD	0.00034 ± 0.0008	0.326 ± 0.73	0.14 ± 0.50
Negative CH_4 Fluxes (g m^{-2} d^{-1})			
Min.	-2.63×10^{-2}	-	-2.63×10^{-2}
Max.	-1.12×10^{-5}	-	-3.06×10^{-6}
Mean ± SD	-0.01316 ± 0.0132	-3.06×10^{-6}	-8.78×10^{-3}

Table 2. Average methane concentrations in gas probes installed during baseline Survey 3.

Depth (cm)	Avg. CH_4 (% v/v) Profiles		
	Cluster 1	Cluster 2	Cluster 3
30	0.002	0.047	0.004
60	1.211	0.485	10.535
90	0.007	23.986	55.916
120	1.351	5.534	ND

Table 3. Summary of initial physico-chemical properties of soils sampled from each depth layer for each test plot during installation**.**

	Layer	Depth (cm)	MC (% w/w)	OC (%)	pH	ORP (mV)	EC (mS/cm)
P1	Soil, unamended (control)	0 to 60	12.6	3.9	7.3	-35.7	0.32
	Regraded waste below test pad	120+	15.0	2.7	7.6	-48.8	0.23
P2	2% biochar -amended soil	15 to 30	11.0	7.2	7.3	-36.6	0.32
	Soil layer below treated layer	30 to 60	12.5	5.0	7.4	-43.9	0.26
	Regraded waste below test pad	120+	20.9	4.3	7.3	-31.0	0.36
P3	Soil, unamended above biochar layer	17 to 60	9.3	4.1	7.4	-38.3	0.3
	IC soil below GDL	90 to 120	20.0	3.7	7.3	-36.5	0.32
	Regraded waste below test pad	120+	14.9	4.2	7.5	-42.8	0.27
P4	10% biochar-amended soil	15 to 30	10.1	10.9	7.2	-30.8	0.37
	IC soil below GDL	90 to 120	12.5	4.8	7.4	-39.2	0.30
	Regraded waste below test pad	120+	21.9	6.2	7.4	-39.7	0.29
P5	Soil, unamended (control)	0 to 60	11.4	4.6	7.4	-43.1	0.27
	Regraded waste below test pad	120+	14.20	4.3	7.4	-39.8	0.29
P6	Soil, unamended above treated layer	0 to 15	13.5	3.8	7.4	-43.0	0.27
	2% biochar amended soil	15 to 30	11.7	8.1	7.4	-41.1	0.28
	Soil, unamended below treated layer	30 to 60	11.6	5.1	7.4	-42.8	0.27
	IC soil below GDL	90 to 120	15.8	4.6	7.3	-37.8	0.31
	Regraded waste below test pad	120+	10.1	2.3	7.9	-73.9	0.12
P7	Soil, unamended above biochar layer	0 to 15	12.3	4.9	7.4	-43.1	0.27
	Soil layer, below biochar layer	15 to 60	10.9	6.9	7.5	-47.3	0.24
	Regraded waste below test pad	120+	6.9	1.7	7.7	-60.9	0.17
P8	Soil, unamended above treated layer	0 to 15	8.9	6.0	7.5	-44.8	0.26
	10% biochar-amended soil	15 to 30	9.7	12.0	7.4	-39.7	0.29
	Soil, unamended below treated layer	30 to 60	11.9	3.4	7.5	-47.3	0.24
	IC soil below GDL	90 to 120	10.1	4.0	7.4	-41.3	0.28
	Regraded waste below test pad	120+	14.0	3.8	7.4	-44.1	0.26

MC = Moisture Content; OC = Organic Matter Content; EC = Electrical Conductivity; IC = Intermediate Cover; GDL = Gas Distribution Layer

Table 4. Meteorological conditions during each sampling campaign conducted.

Event No.	Day of Year	Weather Condition[a]	Ground Temp.[a] (°F)	Soil Moisture[a] (% *v/v*)	Daytime Air Temp[a] (°C)	Pressure[a] (mb)	Rel. Humidity[a] (%)
1	245	Clear	26.1	3.65 ± 1.4	24.9	1011.26	53%
2	269	Mostly clear, some fog	12.4	3.03 ± 0.7	10.6	1023.63	96%
3	294	Overcast	24.9	6.02 ± 1.5	8.8	1022.15	73%
4	346	Overcast, cold	0.9	6.95 ± 1.8	1.1	1026.27	85%
5	76	Overcast	8.1	7.36 ± 2.9	3.9	1025.14	81%
6	84	Clear	4.5	19.55 ± 2.2	3.6	1014.3	34%
7	91	Clear	8.1	7.90 ± 1.9	10.0	1017.63	92%
8	94	Mostly cloudy	4.9	9.71 ± 2.3	3.2	1024.53	47%

[a] Values shown represent average values recorded during each sampling event. Pressure reported refers to barometric pressure.

Table 5. Average and standard deviation of surface methane fluxes (g CH_4 m^{-2} d^{-1}) measured from the center of each test plot for each survey.

Survey No.	P1	P2	P3	P4	P5	P6	P7	P8
1	199.4 ± 68.2	-0.06[c]	-8.48 ± 7.36	-7.26[c]	0.8 ± 0.7	39.9 ± 36.9	1132.8 ± 159.6	4.5 ± 0.7
2	53.8 ± 5.4	-0.21[c]	-0.61[c]	-0.02[c]	10.4 ± 14.8	114.0 ± 8.2	321.6 ± 111.7	0.8 ± 0.2
3	0.25 ± 0.1	0.0[a]	-0.14[c]	-0.33 ± 0.7	0.18[c]	4.4 ± 0.7	515.8 ± 86.2	1.2 ± 0.3
4	119.4 ± 42.2	-0.82 ± 1.2	4.16 ± 3.9	2.41[c]	ND[b]	ND[b]	ND[b]	ND[b]
5	2.5 ± 1.8	0.04 ± 0.1	0.06 ± 0.1	0.11 ± 0.2	-0.96 ± 1.0	19.2 ± 9.4	307.8 ± 19.0	7.9 ± 1.2
6	12.3 ± 4.5	0.02[c]	-0.01[c]	0.01[c]	0.01[c]	3.7 ± 0.7	4.7 ± 0.3	110.2 ± 53.8
7	69.6 ± 21.1	ND[b]	ND[b]	0.0[a]	ND[b]	31.1 ± 5.4	225.4 ± 14.5	17.8 ± 5.9
8	140.0 ± 29.8	ND[b]	ND[b]	ND[b]	0.6 ± 0.3	7.1 ± 1.8	170.1 ± 43.2	17.3 ± 2.0

[a] No net positive or net negative flux observed.

[b] ND: No data obtained.

[c] Only 1 of 3 trials performed yielded data with acceptable R^2 values.

Table 6. Physical properties of the soil cores obtained from the center of each test plot during terminal sampling.

Plot No.	Bulk Density (g/cm^3)	Moisture Content [a] (% *w/w*)	Dry Density (g/cm^3)	Organic Matter[a] (%)	Porosity (%)
1	2.095	19.5	1.75	4.137	0.28
2	2.144	17.6	1.82	6.489	0.26
3	1.826	14.5	1.60	8.478	0.35
4	1.900	16.6	1.63	10.310	0.33
5	1.500	13.7	1.32	5.538	0.46
6	1.975	14.3	1.73	5.692	0.29
7	1.877	18.7	1.58	7.653	0.35
8	1.963	18.0	1.66	6.102	0.32

[a] Reported moisture and organic matter contents are averages for the whole core

Table 7. Total number of sequences matches to operational taxonomic units detected in samples from each test plot and the percentage of those corresponding to methanotrophic genera (%MOB) for select field samples characterized.

	DNA Conc. (ng/ul)	Moisture Content (% *w/w*)	Organic Content (%)	Depth targeted (cm)	Absolute abundance	Total MOB	% MOB
P1	11.3	16.94	5.59	~10	45967	15847	34.47%
	17.6	16.94	5.591	~10	67458	38922	57.70%
	8.6	17.58	4.37	~15	68413	38335	56.03%
	34.2	17.58	4.37	~15	64518	48578	75.29%
P2	3	14.28	4.87	~10	40129	11441	28.51%
	bdl	14.28	4.87	~10	104311	16020	15.36%
	3.1	12.49	7.11	~20	51378	6587	12.82%
	1.3	12.49	7.11	~20	100832	11636	11.54%
P3	2.8	11.98	4.14	~10	119748	4299	3.59%
	bdl	11.98	4.14	~10	110787	6791	6.13%
	0.3	12.30	12.60	~20	100556	4386	4.36%
	2.7	12.30	12.60	~20	93987	5144	5.47%
P4	8.5	13.22	3.76	~11	75736	15002	19.81%
	5.2	13.22	3.76	~11	61185	6004	9.81%
	4.4	14.47	9.43	~15	61153	4637	7.58%
	3.6	14.47	9.43	~15	45612	3693	8.10%

MOB = Methane Oxidizing Bacteria; bdl = below detection limit

Figure Captions

Figure 1. Schematic of transects taken during baseline surveys (not to scale) with respect to test plot locations.

Figure 2. Profiles of test plot designs installed at the field site. Profiles for the replicate plots P5 to P8 are identical to P1 to P4, however no gas probes were installed at P5, P6 and P7.

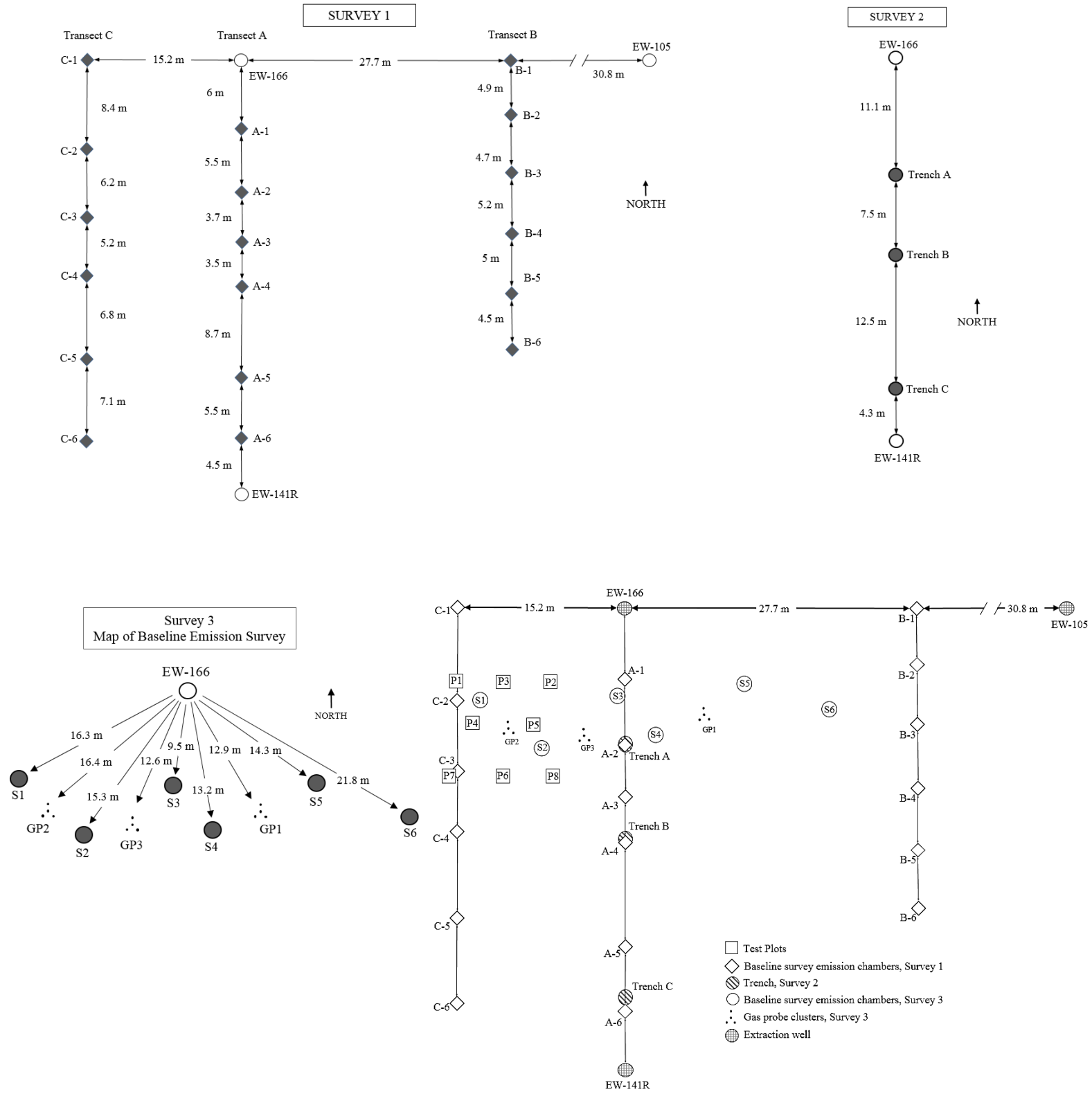
Figure 3. Average gas profiles across all monitoring event for test plot with gas probes installed (P1, P2, P3, P4 and P8).

Figure 4. Average rates of methane oxidation, CO_2 production, and O_2 consumption observed in batch assays of field soils taken from soil cores exhumed during terminal sampling; upper (above treatment layer); lower (within treatment layer).

Figure 5. Percentage of operational taxonomic units detected in next-generation sequencing of the 16SrRNA gene in field soil samples versus potential methane oxidation rates observed in those samples during terminal batch assays.

Figure 6. Plots of CH_4 and CO_2 versus time (hours) for terminal batch

Figure 7. Maximum and average methane flux observed during each survey versus average air temperature.



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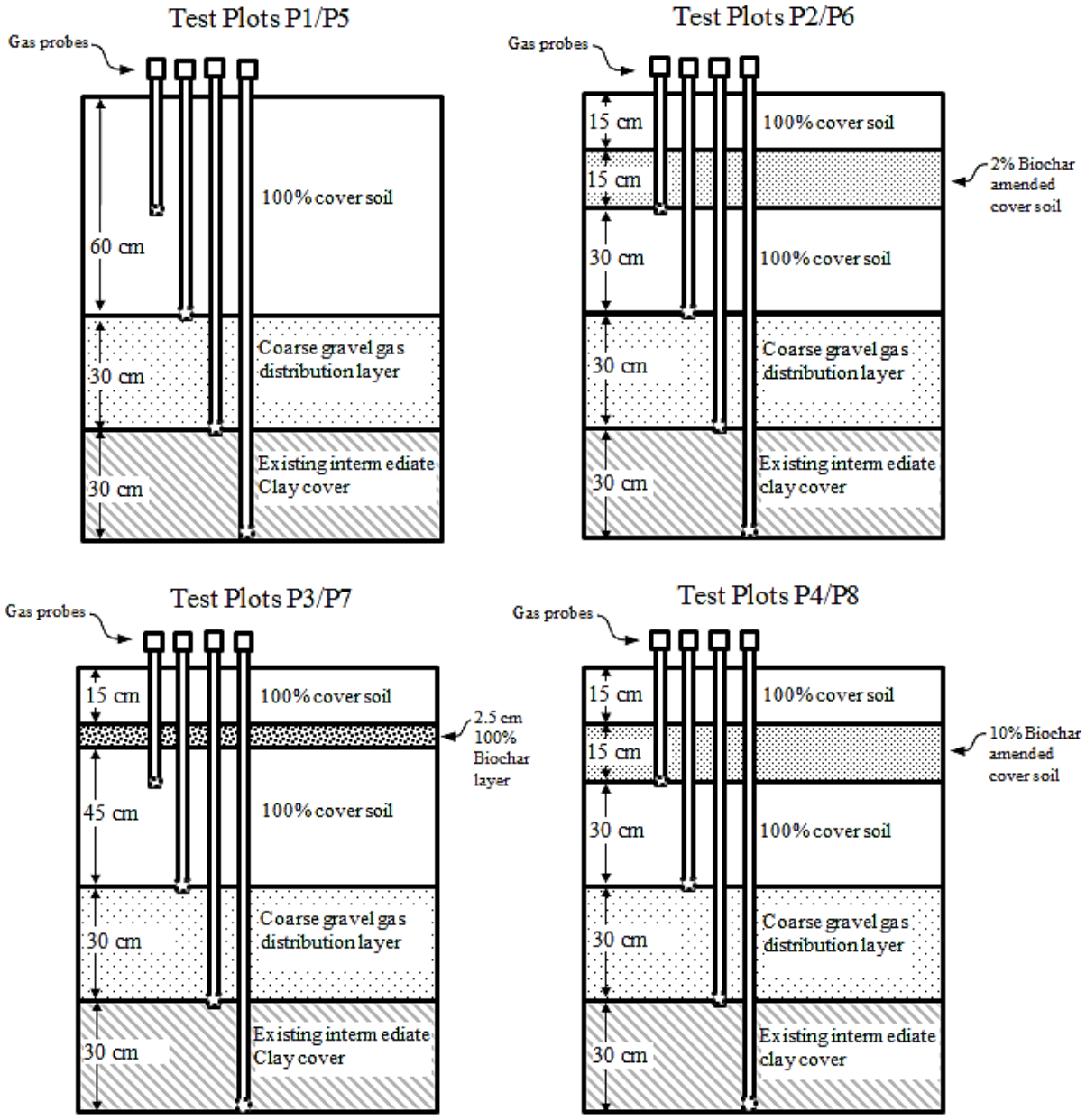

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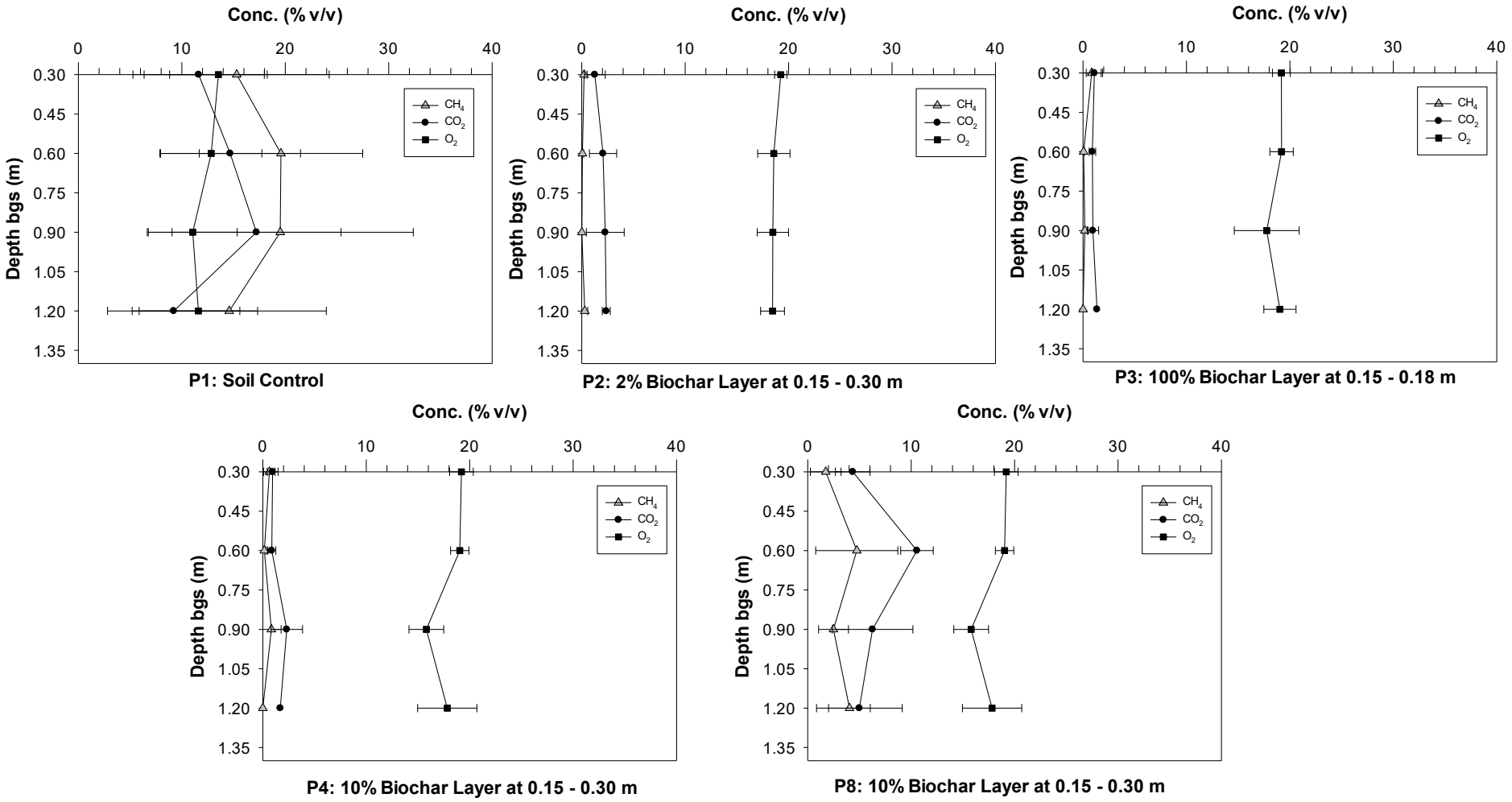
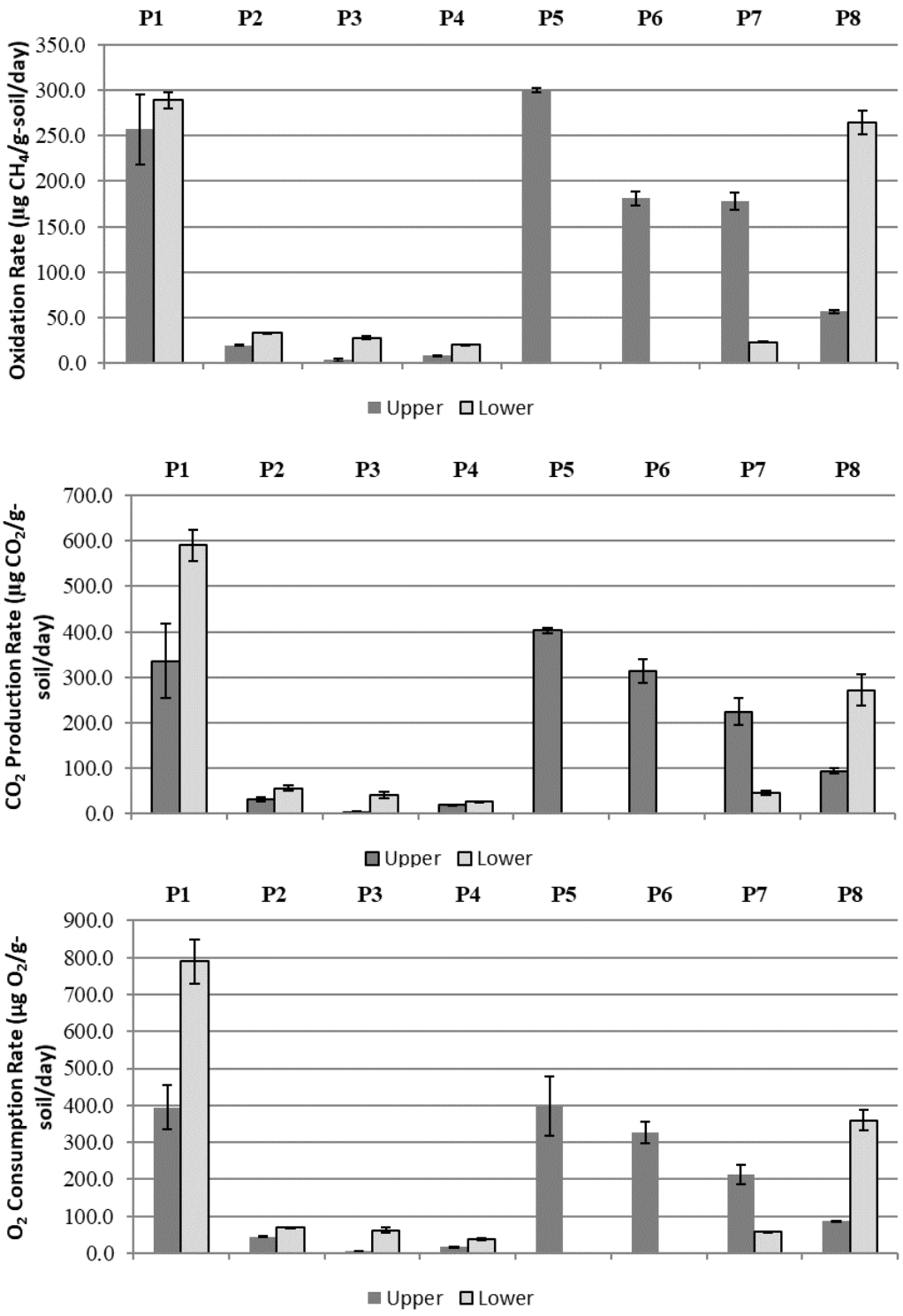


Figure 3. Average gas profiles across all monitoring event for test plot with gas probes installed (P1, P2, P3, P4 and P8).



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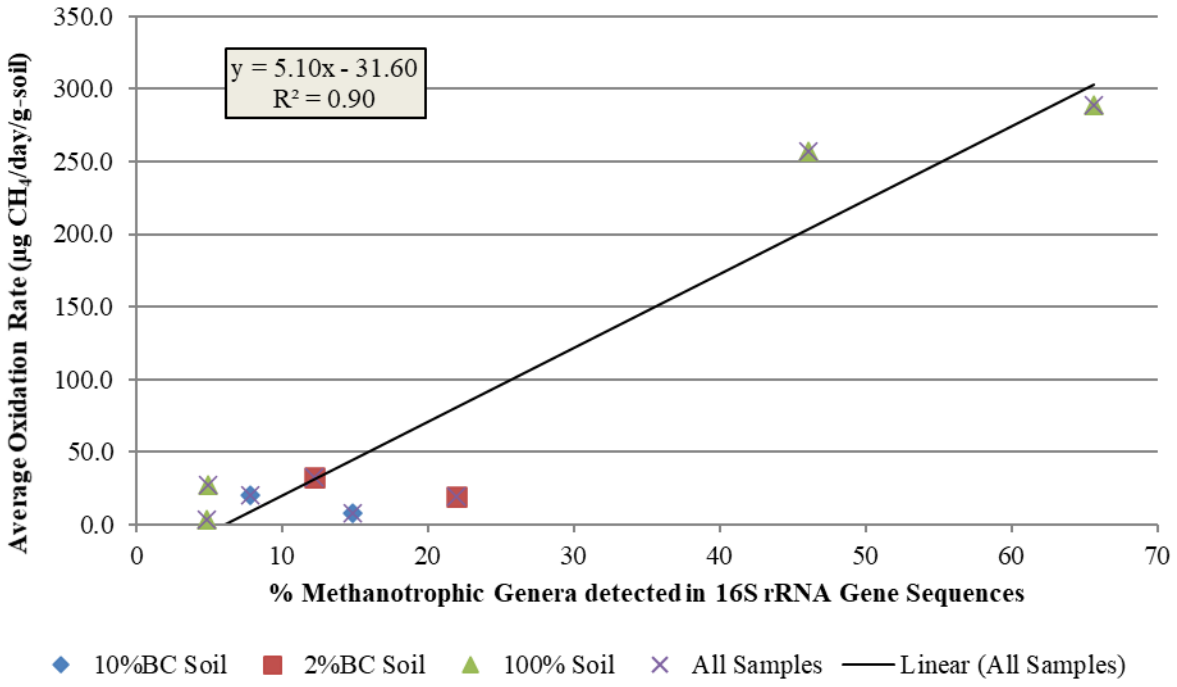

Figure 5. Percentage of operational taxonomic units detected in next-generation sequencing of the 16SrRNA gene in field soil samples versus potential methane oxidation rates observed in those samples during terminal batch assays.

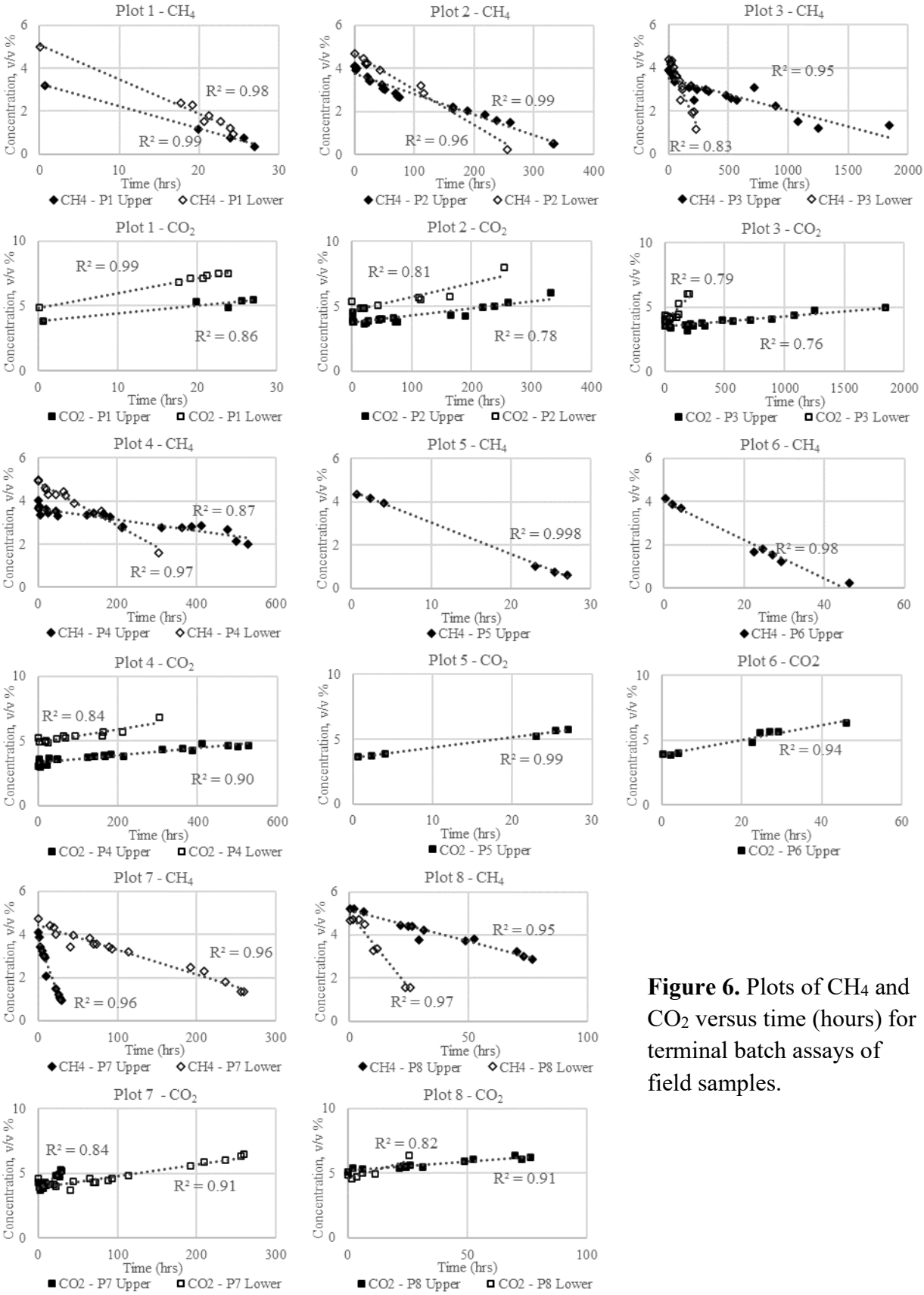

Figure 6. Plots of CH_4 and CO_2 versus time (hours) for terminal batch assays of field samples.

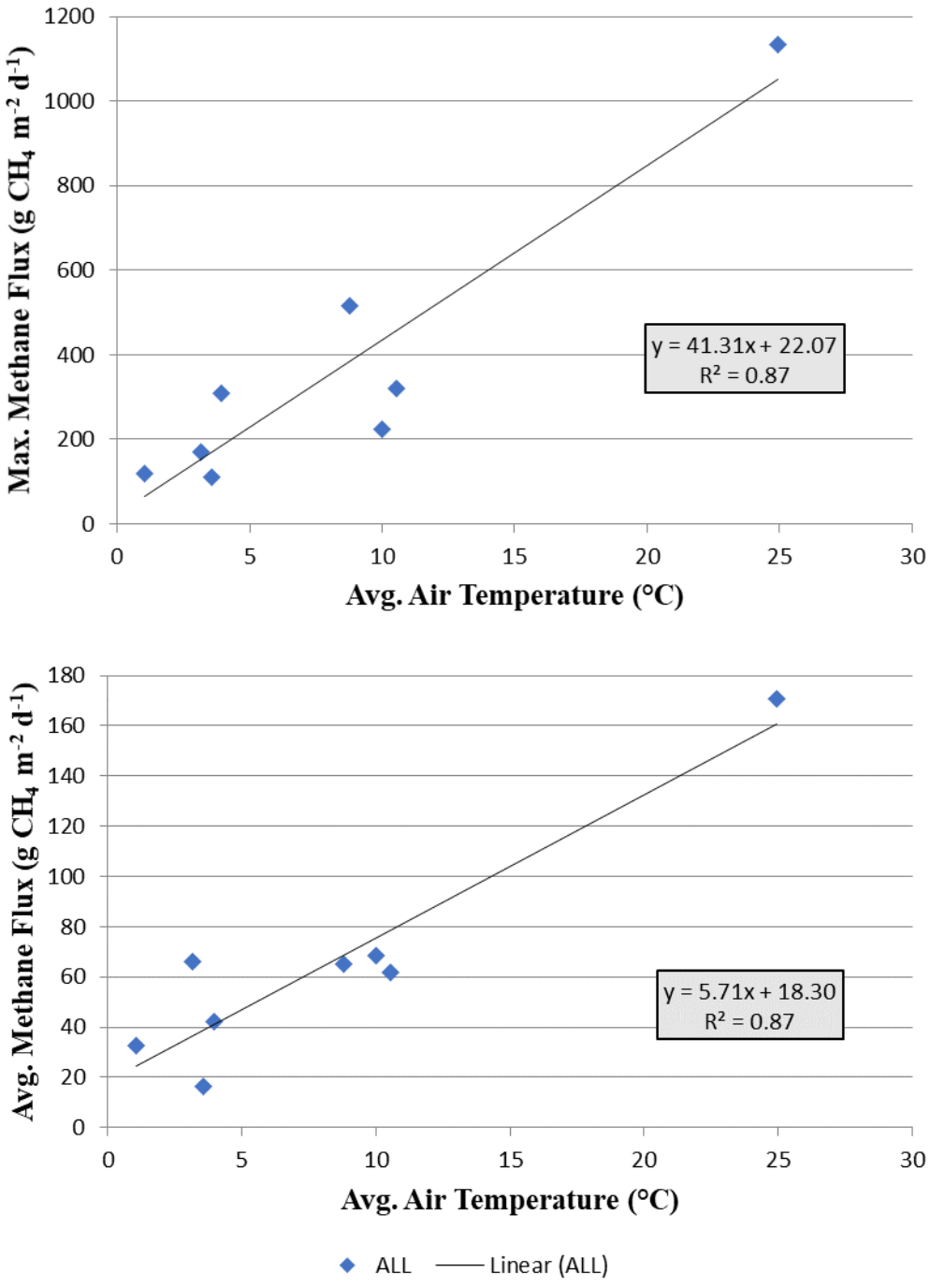

Figure 7. Maximum and average methane flux observed during each survey versus average air temperature.