INDUCTION OF ENHANCED METHANE OXIDATION IN COMPOST: TEMPERATURE AND MOISTURE RESPONSE

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Abstract

Landfilling is one of the most common ways of municipal solid waste disposal. Degradation of organic waste produces CH_4 and other landfill gases that significantly contribute to global warming. However, before entering the atmosphere, part of the produced CH_4 can be oxidised while passing through the landfill cover. In the present study the oxidation rate of CH_4 was studied with various types of compost as possible landfill cover. The influence of incubation time, moisture content and temperature on the CH_4 oxidation capacity of different types of compost was examined. It was observed that the influence of moisture content and temperature on methane oxidation is time-dependent. Maximum oxidation rates were observed at moisture contents ranging from 45% to 110% (dry weight basis), while the optimum temperature ranged from 15 to $30^{o}C$.

Key words: CH_4 oxidation, greenhouse gas, compost, landfill, landfill gas.

1. Introduction

Methane (CH_4) accounts for approximately 22% of the enhanced global warming (Lelieveld et al., 1998). Relative to CO_2, the global warming potential of CH_4 is estimated at 23 using a 100-year time horizon (IPCC, 2001). The atmospheric concentration of CH_4 has increased from a concentration of 700 ppb to 1745 ppb between the years 1750 and 1998 (IPCC, 2001).

Landfilling is considered to be an important global source of this greenhouse gas. Emissions of CH_4 from waste landfills are ranked third among anthropogenic CH_4 sources and range between 19-40 Tg/yr (Doorn and Barlaz, 1995; Bogner and Matthews, 2003). These emissions are mainly caused by inadequate gas collection systems, uncontrolled emissions from old dumps and from unauthorized open dumping. Furthermore, because of the increase in population in developing countries, CH_4 emissions are estimated to increase by up to 60% within the next two decades (Meadows et al., 1997). Landfill gas extraction from smaller and older landfills with low CH_4 generation rates is economically not beneficial.

A low-cost approach such as increasing microbial activity to increase CH_4 oxidation using a suitable cover layer can help reducing CH_4 emissions from landfills. Approximately 60% of the CH_4 produced from anthropogenic and natural sources is oxidised microbially adjacent to the zone of production or in soils functioning as atmospheric sinks (Reeburgh et al., 1993; Bender and Conrad, 1995).

CH_4 oxidation in conventional soil covers, typically of older landfills, has been reported to be effective in reducing the amount of CH_4 emitted (Bogner et al., 1997; Chanton and Liptay, 2000; Börjesson et al., 2001). The oxidation rate depends on different environmental factors including moisture and temperature (King and Adamsen, 1992; Adamsen and King 1993; Boeckx and Van Cleemput, 1996; Whalen

and Reeburgh 1996; Visvanathan et al., 1999; De Visscher et al., 2001). The effect of inorganic nitrogen on CH_4 oxidation is very complex and can be both stimulatory and inhibitive (Bodelier and Laanbroek, 2004). De Visscher and Van Cleemput (2003) found that the influence of ammonium and chloride on CH_4 oxidation by soils is time dependent. Time dependence of the influence of temperature and moisture on CH_4 oxidation has never been studied systematically. However, Gebert et al. (2003) and Börjesson et al. (2004) found that methanotrophic enrichments cultured at 10°C lead to a shift towards more type I methanotrophs with a lower optimum temperature, indicating that temperature response in media like soil or compost might be time-dependent.

CH$_4$ oxidation rates in conventional soils have been studied extensively, but there are fewer studies on environmental factors influencing the rate of CH_4 oxidation in compost. Streese and Stegmann (2003) studied compost as biofilter material for microbial CH_4 degradation and reported high degradation rates of up to 63 g CH_4 m^{-3} h^{-1}. Wilshusen et al. (2004a,b) studied CH_4 oxidation and formation of exopolymeric substances in compost for the performance of CH_4 biofilters and the effect of oxygen concentration. Humer and Lechner (1999, 2001) studied municipal solid waste compost and sewage sludge compost as cover soil to increase oxidation of CH_4 and found that complete CH_4 oxidation is possible. Barlaz et al. (2004) reported that compost covers oxidized more CH_4 than conventional clay covers in field trials, but warned that compost covers can also produce CH_4 if the moisture content is too high.

In the present study experiments were performed at laboratory scale to examine how CH_4 oxidation depends on temperature and moisture for different types of compost and to assess the suitability of compost as landfill cover. Particular attention was given to the time dependence of these influences, an aspect that has

received little or no attention to date. The CH_4 oxidation capacity of compost will help to determine its potential usefulness to landfill operators as a tool to curb CH_4 emissions in order to meet regulatory requirements.

2. Materials and Methods

2.1. Compost characteristics and analysis

Table 1 shows the characteristics of the composts used in this study. They were obtained from VLACO, Mechelen, Belgium. Two mixed kitchen and garden waste composts (labelled GFT1 and GFT2) were tested, together with three garden waste composts (labelled G1, G2 and G3). GFT1 was produced by intensive aeration in a compost hall; GFT2 by intensive aeration in a compost tunnel. G1 was produced by intensive aeration for 3 months, whereas G2 and G3 were produced by passive aeration for 6 months. All compost samples were analysed by VLACO. Compost moisture contents were determined gravimetrically, by drying 10-g samples for 48 h at 105 °C. pH was measured by shaking 20 ml of compost in 100 ml of water for 24 hours and analysis by pH meter. Electrical conductivity was determined in similar extracts. Organic matter was determined gravimetrically by heating to 550°C for 4 hours. Organic carbon content was assumed to be organic matter content divided by 1.8. Total nitrogen was determined by Kjeldahl destruction followed by steam distillation and capturing the resulting ammonia in boric acid, and back titration of the resulting ammonium borate with HCl. For inorganic N analysis, 30-g samples were extracted with 60 ml 2 M KCl by shaking for 1 h and then the compost slurry was filtered. The NH_4^+, nitrate and nitrite contents were analysed by auto analyser (Bran + Luebbe Auto Analyser 3). Additional compost samples were used for kinetic experiments of methane oxidation as described in the next section.

2.2. Kinetic experiments

The batch experiments were carried out at room temperature ($22\pm1°C$) in glass bottles. All kinetic experiments were performed in duplicate. About 30 g of the compost samples was transferred into 215-ml bottles that can be closed with a rubber septum. Each test bottle was injected with 10 ml of 100% CH_4, leading to an initial CH_4 concentration of about 5%. The bottles were sampled periodically with an interval of 30-60 minutes to determine the CH_4 consumption. Typically 5 samples (100μl) were taken. The CH_4 uptake rate was determined by linear regression of the measured CH_4 concentration versus time.

To determine the respiration activity of the compost about 30 g of fresh compost samples not previously exposed to CH_4 were transferred into 215-ml bottles, closed with rubber septa and the concentration of CO_2 was determined at different times at 22°C. The CO_2 production rate was determined by linear regression of the CO_2 concentration versus time.

2.3. Compost incubations

2.3.1. Influence of incubation time

Incubations were carried out in batch mode because this technique enables accurate moisture control by gravimetry. It has been shown that column experiments can lead to moisture loss, accumulation and redistribution, both in forced-aeration mode (De Visscher and Van Cleemput, 2003) as in passive-aeration (counter-gradient) mode (Stein and Hettiaratchi, 2001).

The compost taken to measure the effect of incubation time on CH_4 oxidation rate was fresh and not previously exposed to CH_4. All five compost samples were

incubated in 215-ml bottles at $22\pm1°C$ to test their potential to oxidise CH_4. When CH_4 or O_2 was depleted the bottles were opened for about 15 minutes for aeration, closed again and 10 ml CH_4 was added, leading to a headspace mixing ratio of approximately 5%. This procedure was repeated until the end of the incubation.

2.3.2. Influence of moisture content

To test the influence of moisture content, fresh samples of the most active composts (G1 and G3) were pre-incubated in jars with CH_4 at room temperature of $22\pm1°C$ by periodically adding 10% CH_4 after aeration. The incubation time in the jars was two weeks for compost G1 and five weeks for G3. The moisture response of CH_4 oxidation was determined by adjusting samples to different moisture contents, transferring them to bottles and measuring the CH_4 oxidation rate. The moisture contents tested were 29, 45, 65, 85 and 110% for G1 and 34, 52, 72, 92 and 110% (w/w) for G3. All moisture percentages are expressed on a dry weight basis. Moisture contents were increased by adding demineralised water and decreased by evaporating the water content to the desired level. 30 g of sample were brought into the bottles and acclimatised overnight with changed moisture content at a temperature of $22\pm1°C$.

2.3.3. Influence of temperature

To test the influence of temperature, fresh samples of the most active composts (G1 and G3) were pre-incubated in jars with CH_4 at a temperature of $22\pm1°C$ by periodically adding 10% CH_4 after aeration. The incubation time in the jars was two weeks for compost G1 while for G3 it was five weeks. 30 g of sample was brought into the bottles and acclimatised overnight at a temperature of 7, 15, 22, 30

and 40°C. The CH_4 oxidation rate was measured by determining the decrease of CH_4 versus time after addition of 10 ml of CH_4.

2.4. Gas analysis

The headspace concentration of CH_4 was determined with a Chrompack CP 9000 chromatograph using a flame ionization detector (FID). Helium was used as carrier gas. The analysis was carried out under the following conditions: injector temperature 65°C, oven temperature 35°C, and detector temperature 150°C. A stainless steel column with NaI-inactivated alumina (100-120 mesh) stationary phase was used for separation. The chromatograms were registered and analyzed using the Thermo Separation System "WOW" software package. CO_2 was determined with a Shimadzu 14 B GC using an electron capture detector (ECD). Nitrogen was used as carrier gas. The analysis was carried out under the following conditions: injector temperature 100°C, oven temperature 35°C, and detector temperature 250°C. Gases were separated on a 1-m Porapak-Q pre-column (80-100 mesh, outer diameter 0.3 cm), followed by a 2-m column with the same characteristics. A backflush valve between the pre-column and the main column was used for protection of the dectctor. The chromatograms were registered and analyzed using the "SHIMADZU CLASS VP" software package.

3. Results and Discussion

Table 2 shows the respiration rate of the different types of compost. A high CO_2 production rate shows high levels of microbial activity, indicating that the compost is still decomposing. In theory, CO_2 production can also result from inorganic sources (carbonates), but since no major changes in the compost properties

(like a pH decrease leading to carbonate dissolution and CO_2 release) were imposed prior to the experiment, such effects will be negligible. It was expected that the CH_4 oxidation potential would be low when the compost is not yet stable and respiration rate is high. Therefore, we tested the relationship between respiration rate and CH_4 oxidation. However, no relation between the oxidation potential and respiration rate was observed. Only two garden waste composts (G1 and G3) showed a significant CH_4 oxidation activity even after one month of incubation and hence these composts were used for further study of CH_4 uptake rates and of the effect of temperature and moisture content on CH_4 oxidation. Both composts had a low inorganic N and salt content in comparison with the other composts tested.

3.1. Effect of incubation time

Figure 1 shows the CH_4 oxidation rate as a function of time at 45% moisture content for G1 and 52% moisture content for G3 at $22 \pm 1°C$. After an initial lag phase at low activity, the compost evolved to a more active phase and showed a peak activity after 20-25 days of incubation. Thereafter the activity started to decline. A similar phenomenon was observed by Kightley et al. (1995), Hilger et al. (2000), and De Visscher and Van Cleemput (2003) in soils, and by Wilshusen et al. (2004b) in compost. In some studies the peak occurred after a few days (De Visscher et al. (1999, 2001), De Visscher and Van Cleemput (2003) for soils; Wilshusen et al. (2004b) for leaf compost and municipal solid waste compost), whereas in some studies CH_4 oxidation developed gradually over several weeks (Hilger et al. (2000) for soils; Wilshusen et al. (2004b) for wood chip compost). The peak activity of G1 was 1.81 μmol kg_{dw}^{-1} s^{-1}, while for G3 it was 0.82 μmol kg_{dw}^{-1} s^{-1}. In compost G1, the peak activity was observed for about 5 days and was followed by a gradual decrease of the

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activity. The peak activity in compost G3 lasted only 2 days and was followed by a rapid decline. Average activity after the decline was 0.61 μmol kg_{dw}^{-1} s^{-1} and 0.18 μmol kg_{dw}^{-1} s^{-1} for G1 and G3, respectively.

The observed decline can be due to exopolymer production (Hilger et al., 1999; 2000; Wilshusen et al., 2004a,b), or due to depletion of nutrients (De Visscher et al., 1999, 2003). As there was no visible slime production in the samples, we assume the latter to be more probable in the present experiment. De Visscher et al. (1999) observed that methane oxidation rates stay stable for several months after the activity decline. Sometimes a resurge of the activity is observed after 2-3 months (De Visscher and Van Cleemput, 2003).

3.2. Moisture response experiments

Figure 2 and 3 show the influence of moisture content on the CH_4 oxidation rate by compost G1 and G3, respectively. At 110% moisture content compost G1 had a maximum oxidation rate of 2.05 μmol kg_{dw}^{-1} s^{-1} as observed on day 5 (Figure 2). Thereafter, the rate decreased to below 1 μmol kg_{dw}^{-1} s^{-1} on day 15 and increased slightly to 1.16 μmol kg_{dw}^{-1} s^{-1} towards the end of the experiment. At 85% moisture content pattern was similar but a maximum activity of about 2.35 μmol kg_{dw}^{-1} s^{-1} was observed on day 5. The pattern was similar at 65% moisture content, but it did not show a recovery of the activity after 2 weeks. At 45% moisture content a maximum activity of about 1.51 μmol kg_{dw}^{-1} s^{-1} was reached on day 9 and showed only slight decrease afterwards. At this moisture content the samples showed more steady behaviour. The activity was low throughout the experiment at 29% moisture content. The optimum moisture content was found to be 45-85%, and was time-dependent. The observed trends tended to slow down with decreasing moisture content.

Unlike sample G1, sample G3 showed highest CH_4 oxidising activity at 110% moisture content throughout the experiment (Figure 3). Except at low moisture contents, the CH_4 oxidation rate increased steadily during the entire experiment. Oxidation rates for different moisture contents of G3 were in the following order, 110>85>65>45>29%. Apparently, the range of moisture contents examined was too narrow to determine the optimum.

The optimum moisture content of compost G1 (31% organic matter) and G3 (52% organic matter) for CH_4 oxidation were 45–85% and ≥110% moisture on a dry weight basis, respectively. These values are much higher than the optimum for mineral soils with 1 to a few percents of organic matter, which is typically around 15% (Whalen and Reeburgh, 1996; Boeckx and Van Cleemput, 1996). For a bog soil containing about 90% organic matter, on the other hand, a value on the order of 350% has been reported (Whalen and Reeburgh, 1996). Organic matter appears to be the key property determining the optimum moisture content for CH_4 oxidation.

These results indicate that the CH_4 oxidation potential of compost G3 was not really lower than the CH_4 oxidation potential of compost G1, as could be expected from the results in the previous section. Rather, G1 was close to optimal moisture content during the first experiment, whereas G3 was not. The three inactive composts (GFT1, GFT2 and G2) had substantially lower moisture contents than the active composts and higher organic matter content than G1. This may well explain their inactivity, as their optimum moisture content was probably around 100%, 2.5–3.5 times the moisture content at the time of the experiment. Another reason for the lack of activity might be the high inorganic nitrogen content of these composts. It is concluded that tests for CH_4 oxidation potential in composts should not be limited to the natural moisture content obtained by the composting process.

3.3. Temperature response experiments

The effect of temperature on the evolution of the CH_4 oxidation rate is shown in Figure 4 and 5. In the case of G1 at 22°C, maximum oxidation rate of 1.68 µmol kg_{dw}^{-1} s^{-1} was observed on day 9 while at 30°C, maximum oxidation rate observed was 1.65 µmol kg_{dw}^{-1} s^{-1} on day 6 after the start of the experiment. A much lower activity was observed at 7°C. Maximum oxidation observed at this temperature was 0.15 µmol kg_{dw}^{-1} s^{-1} on day 9. The general shape of the curve on day 6 is very similar to results obtained with soils (De Visscher et al., 2001; Whalen and Reeburgh, 1996) with an optimum temperature of 30°C. However, on day 14 the temperature response was very different, with no significant influence between 15 and 30°C. A similar observation was also made on day 26. On the long term almost no influence of temperature between 15 to 30°C was observed.

Compost G3 showed a much slower and more erratic response (Fig. 5). This is probably due to the sub-optimal moisture content of these samples.

The changing patterns observed in G1 can be explained in terms of shifts in the methanotrophic population present in the compost. Gebert et al. (2003) and Börjesson et al. (2004) found that enriching methanotrophic mixed cultures at low temperature (10°C) lead to a pronounced predominance of type I methanotrophic strains with lower optimum temperature (22°C instead of 38°C). It is assumed that the composts incubated at 15°C developed a more psychrophilic population than the composts incubated at higher temperatures.

3.4. Compost as landfill cover material

In our study with compost, the maximum activity observed was 1.812 μmol kg_{dw}^{-1} s^{-1} on day 23 after the start of the experiment. This value refers to CH_4 mixing ratios of several percents, which is substantially higher than the Michaelis-Menten half-saturation constant (K_m) typically encountered with CH_4 oxidation (usually <1%). Also, the CH_4 oxidation was zero-order. Therefore, it is assumed that the values reported here approach V_{max}. De Visscher et al. (2001) have reported a V_{max} value of 0.820 μmol kg_{dw}^{-1} s^{-1} in soil on day 34 after the start of an incubation experiment, while Whalen et al. (1990) have reported a V_{max} value of 0.039 μmol kg_{dw}^{-1} s^{-1} in soil samples that were taken from a landfill.

Streese and Stegmann (2003) observed a V_{max} value of 2.43 μmol kg_{dw}^{-1} s^{-1} in compost, which was a mixture of an equal volume of yard waste, peat and squeezed spruce wood fibre. Wilshusen et al. (2004a) observed a V_{max} value of 3.77 μmol kg_{dw}^{-1} s^{-1} after one month and 2.77 μmol kg_{dw}^{-1} s^{-1} after two months in compost, which was prepared from municipal green waste such as leaves mixed with manure. Values of V_{max} are not easy to interpret mechanistically because they are the result of both the number of micro-organisms in the medium and their activity (Bender and Conrad, 1992; Dunfield and Conrad, 2000). The lower values found in our study in comparison with the results of Wilshusen et al. (2004a) are probably due to the fact that the incubation of the samples of Wilshusen et al. (2004a) were at higher CH_4 concentrations (>30%). Also, CH_4 concentrations were rapidly depleted in our batch experiments when the activity was high, leading to daily periods of CH_4 limitation in the compost samples. Another reason could be the difference in composition of the compost starting material.

In spite of these limitations, the activities found in our study were substantially higher in compost than the activity normally found in soils. Furthermore, compost has

more air-filled pore space than soils (on the order of 50% compared to 20-30% for soils). Therefore it will allow more oxygen to penetrate for the oxidation of CH_4 and create a more extended aerobic zone in the cover. Also, Barlaz et al. (2004) argued that the use of compost avoids the occurrence of cracks in the landfill cover, reducing the risk of emission hot spots. Hence compost is expected to be a better material for biological CH_4 oxidation than soils. Nevertheless, suitability of compost as a cover material should be evaluated on a site-by-site basis, as compost has a lower density than soil, leading to lower potential contact between CH_4 molecules and the micro-organisms, and because high-porosity substrates might dry out more quickly. Furthermore, Barlaz et al. (2004) reported that compost covers can also produce CH_4 if the moisture content is too high. Berger et al. (2005) observed that a clear borderline between cover layers with different properties can lead to water accumulation. The use of soil-compost mixtures might be a solution here.

3.5. Application of compost as low cost approach

The applicability of compost as cover material to reduce greenhouse gas emissions was theoretically assessed for the Gazipur landfill site in Delhi, India. It is an unplanned landfill site and has no collection system for CH_4 recovery. Mor et al. (submitted manuscript) have estimated that $36–71$ m^3 CH_4 m^{-2} yr^{-1} corresponding to $51–101$ μmol CH_4 m^{-2} s^{-1} is produced from this site. Many approaches can be applied to reduce such emissions but they require a proper design and plans for landfills and hence may be costly. The application of compost as cover material offers a low cost approach to reduce such emissions and hence is suitable for the developing countries.

Based on the emission estimates of Mor et al. (submitted manuscript) and a steady state activity of 0.61 μmol kg$_{dw}$$^{-1}$ s^{-1}, it is estimated that a compost dry weight

of 83–165 kg is needed to oxidise all CH_4 emitted by 1 m^2 of landfill. Assuming a compost bulk density of 600 kg fresh weight per m^3 and a moisture content of 100% (dry weight basis), a compost layer of 28–55 cm can theoretically oxidise all CH_4 emitted by the Gazipur landfill site. Assuming that the steady-state activity is representative to long-term application of compost cover soils, it can be an adequate option for mitigating CH_4 emission from this landfill.

Besides, compost can be prepared from organic waste at the landfill site. Therefore, there will be no extra cost in transporting soil from another place. Hence, promoting microbial CH_4 oxidation within a cover soil can be a good management practice for designing and managing the landfill cover. This is a cheap and effective option for reducing emissions from smaller and older landfills with lower amounts of CH_4 generation making gas extraction economically less beneficial.

Conclusion

The temporal patterns of CH_4 oxidation rates observed in compost were similar to the patterns observed previously. The influence of temperature and moisture content on CH_4 oxidation was time-dependent. The temperature response tended to be flatter at longer incubation times. It is hypothesized that this is the result of a shift towards more psychrophilic methanotrophs during incubation at a temperature of 15°C in comparison with higher temperatures. The moisture response of CH_4 oxidation was also time-dependent in one of the composts tested. When comparing composts for their suitability as a landfill biocap, care should be taken to optimize moisture conditions for CH_4 oxidation. Compost has a significant potential to oxidize CH_4 and hence it can be used as cover material to reduce the emission of this greenhouse gas from landfills. Calculations show that complete oxidation of the

methane produced in the Gazipur landfill in Delhi, India, could be feasible if the activities observed can be sustained for a sufficiently long time.

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Table 1

Characteristics of different types of compost

Parameter	GFT1	GFT2	G1	G2	G3	Unit
Moisture content	29.2	41.4	49.5	36.8	58.5	wt%[a]
E.C.(1/5)	2410	2010	971	1750	688	µS/cm
pH(water)	8.8	8.8	8.3	7.4	6.7	-
Chloride	1767	1575	n.d.[c]	1650	700	mg/l
Organic matter content	39.1	42.9	31.1	38.2	52.1	wt%
Total nitrogen	1.81	1.53	1.54	1.46	1.08	wt%
NH_4-N	131.0	343.0	14.0	<25	86.0	mg/l
NO_3-N	118.0	<25	130.0	350.0	<25	mg/l
C/N	12.0	15.6	11.2	14.5	26.9	-
Total nutrient content						
Total P_2O_5	0.88	0.58	n.d.	0.43	0.25	wt%
Total K_2O	1.36	1.15	n.d.	1.06	0.58	wt%
Total CaO	2.91	2.25	n.d.	1.93	1.41	wt%
Total MgO	0.65	0.47	n.d.	0.37	0.20	wt%
Extractable content						
Extractable P	752	794	n.d.	698	173	mg/l
Extractable K	3587	4947	n.d.	5381	2447	mg/l
Extractable Ca	2946	4652	n.d.	5674	1985	mg/l
Extractable Mg	639	510	n.d.	773	466	mg/l

[a] All weight fractions are on a dry weight basis

[b] Volumes used for expressing concentrations in compost are bulk volumes. For approximate mass fractions in mg kg^{-1}, multiply by 2.

[c] n.d.: not determined

Table 2

Respiration rate of the compost

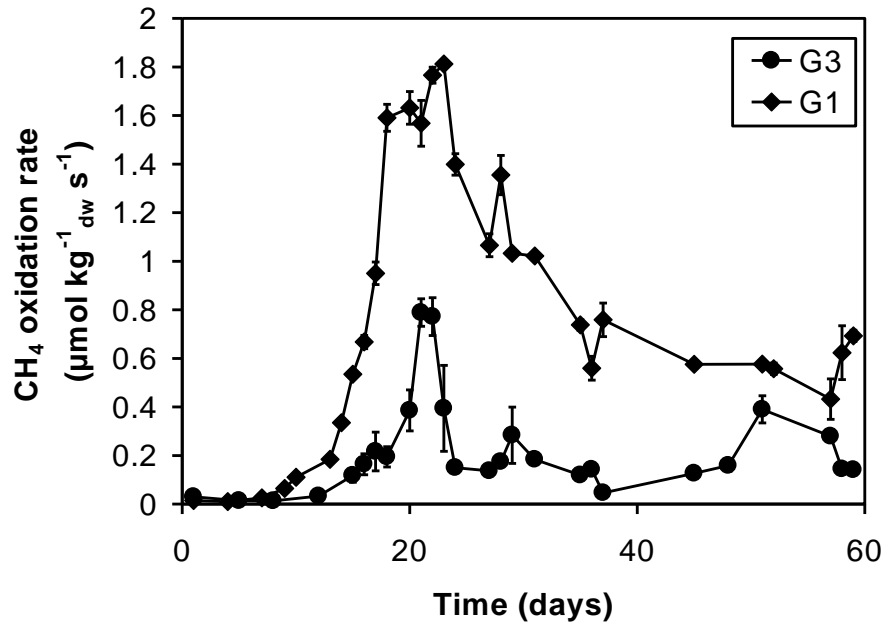| Compost	mmol $CO_2/kg_{dw}/day$
GFT1	4.10
GFT2	15.12
G1	6.28
G2	2.96
G3	8.75

Fig. 1. CH$_4$ oxidation rate observed in two composts over a period of 60 days at 22°C.
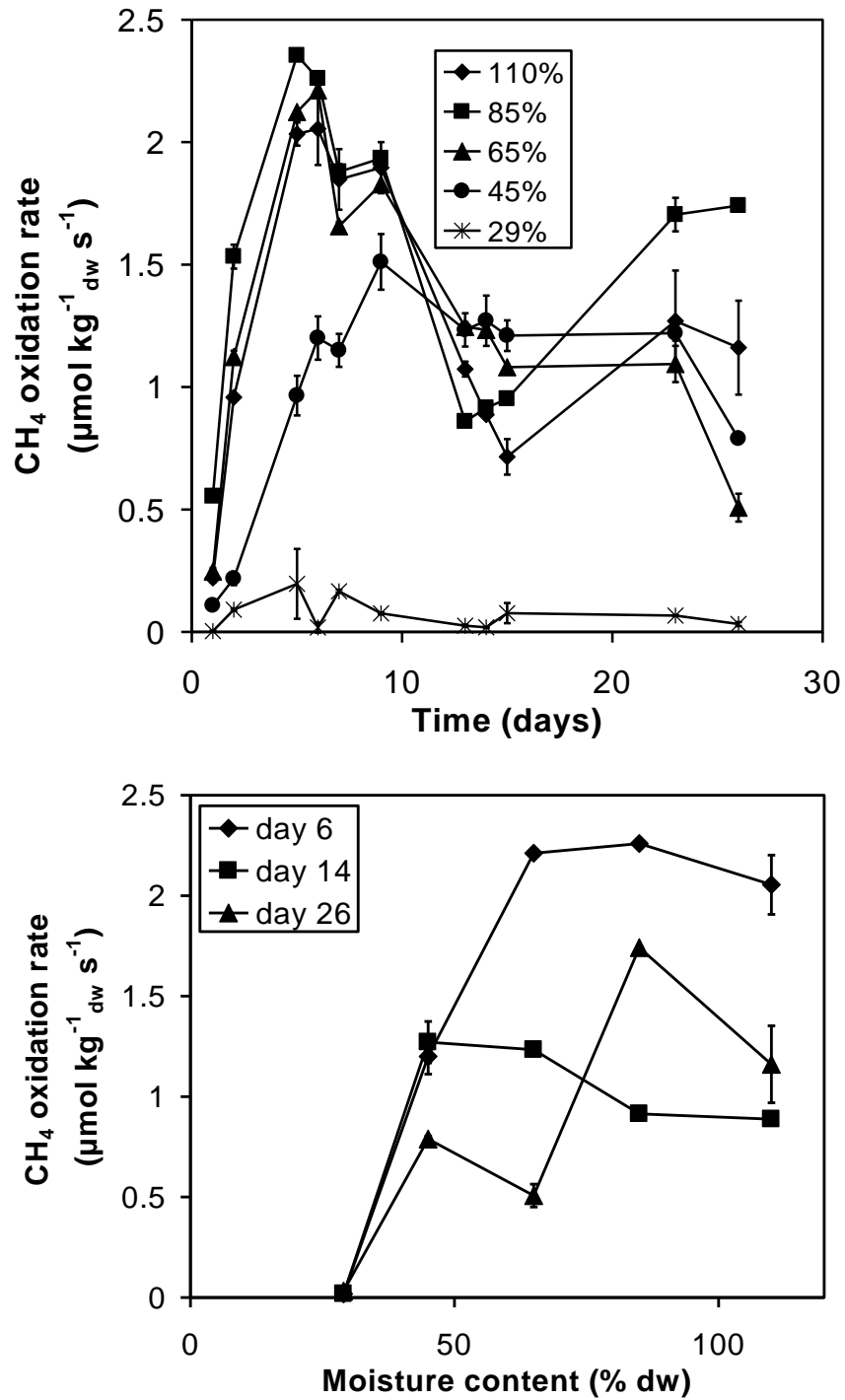
Error bars are standard deviation.

Fig. 2. Effect of moisture content on CH$_4$ oxidation rate by compost G1 versus time. Error bars are standard deviation.
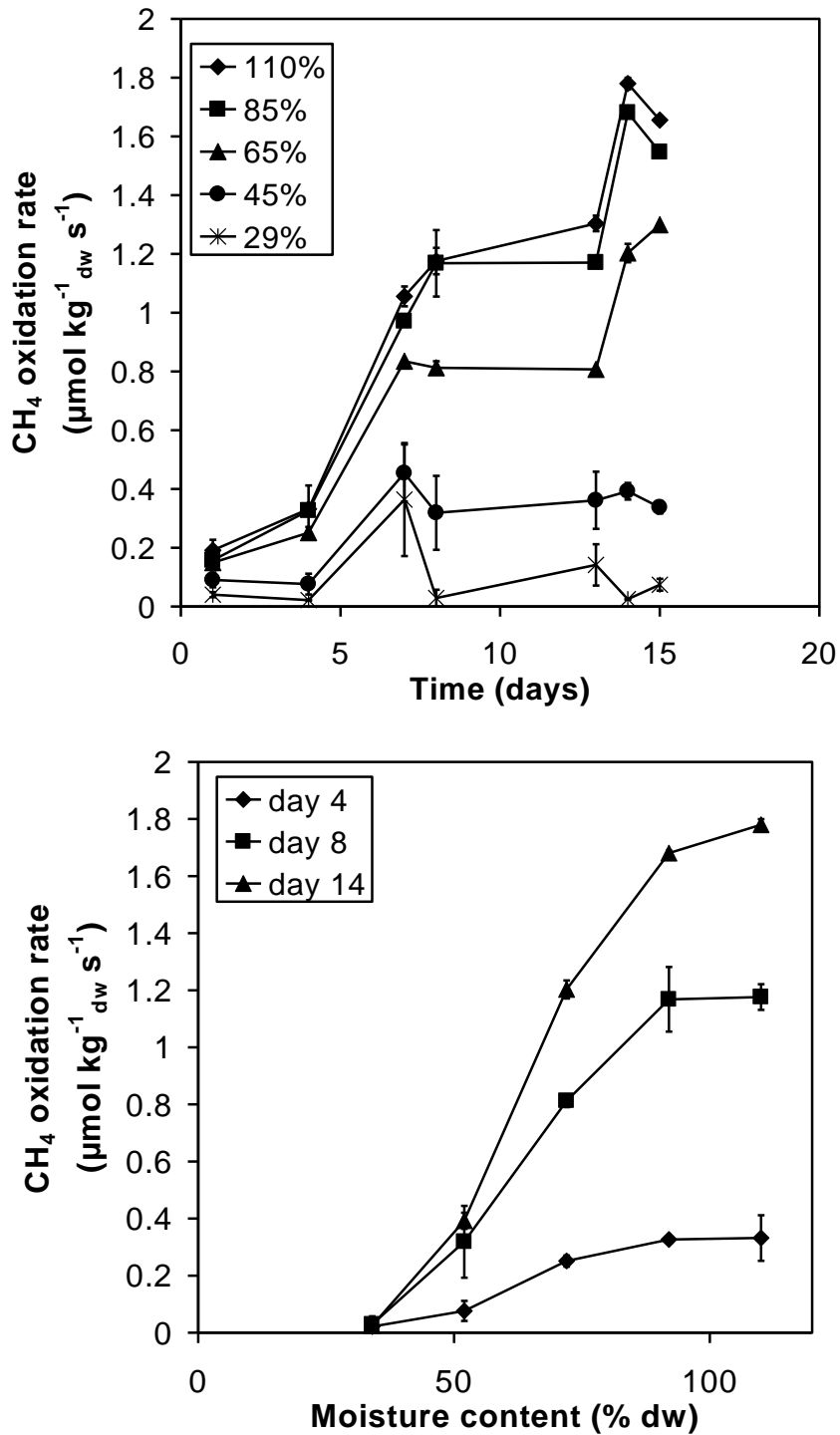
Fig. 3. Effect of moisture content on the CH_4 oxidation rate by compost G3 versus time. Error bars are standard deviation.
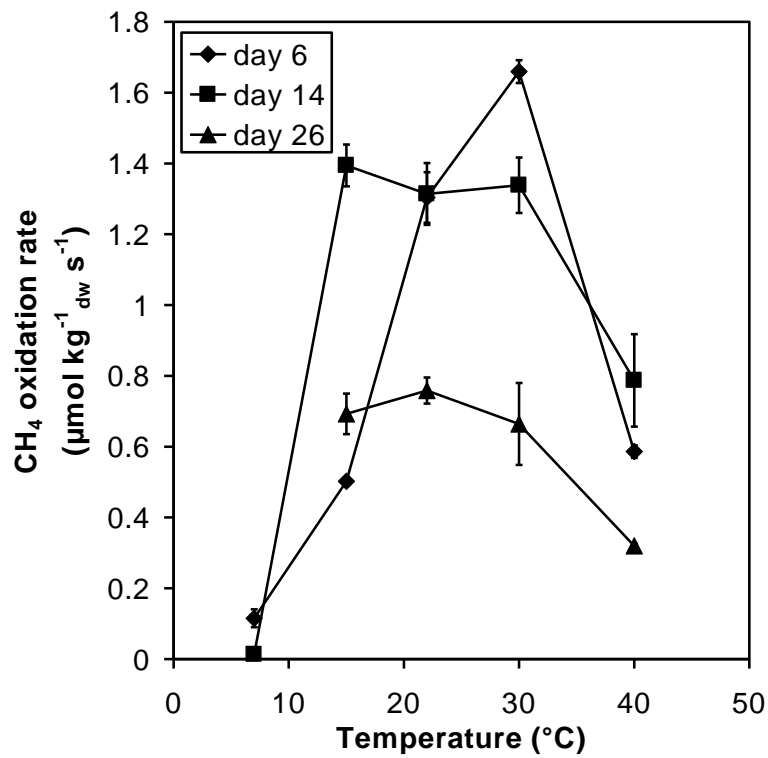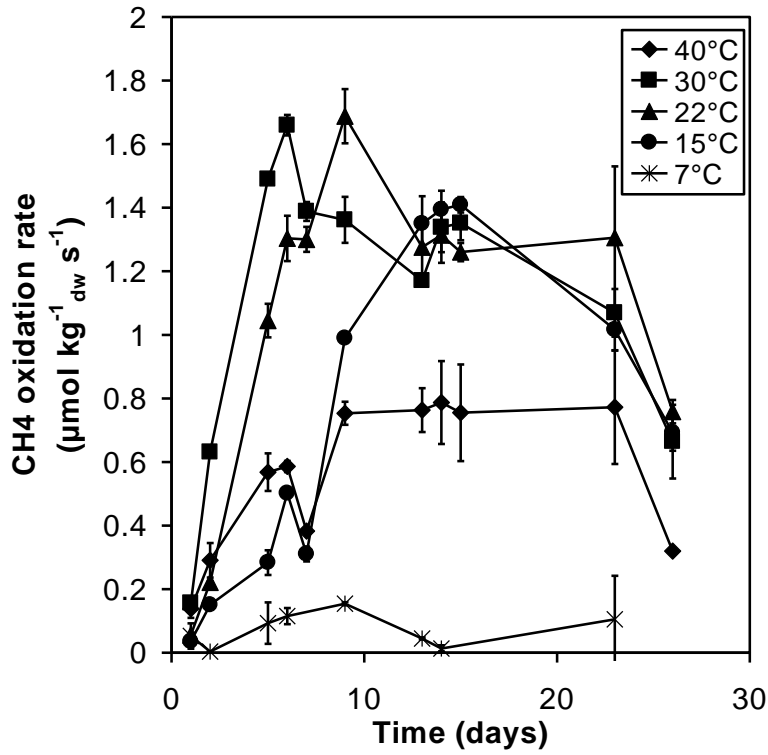
Fig. 4. Effect of temperature on the CH₄ oxidation rate by compost G1 versus time.
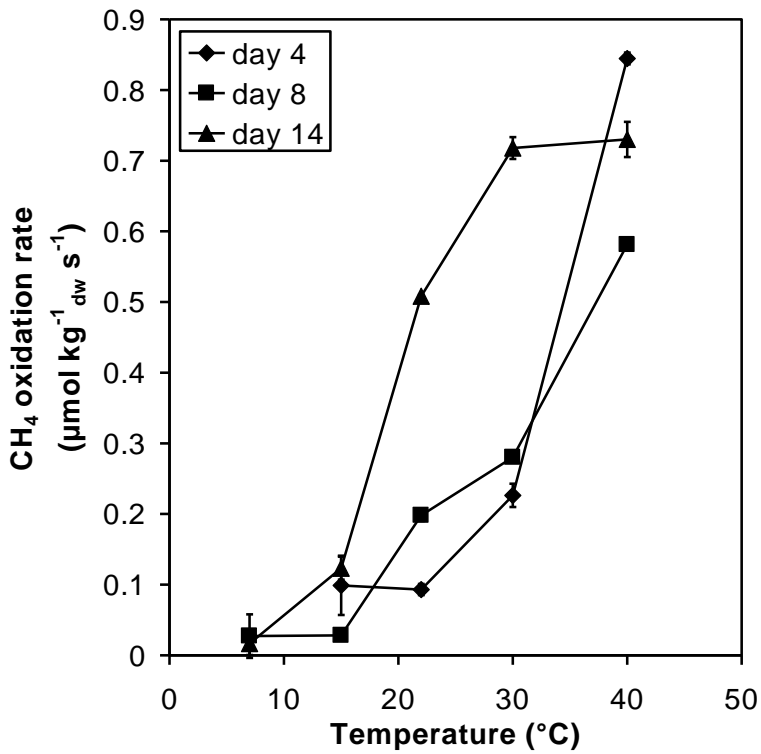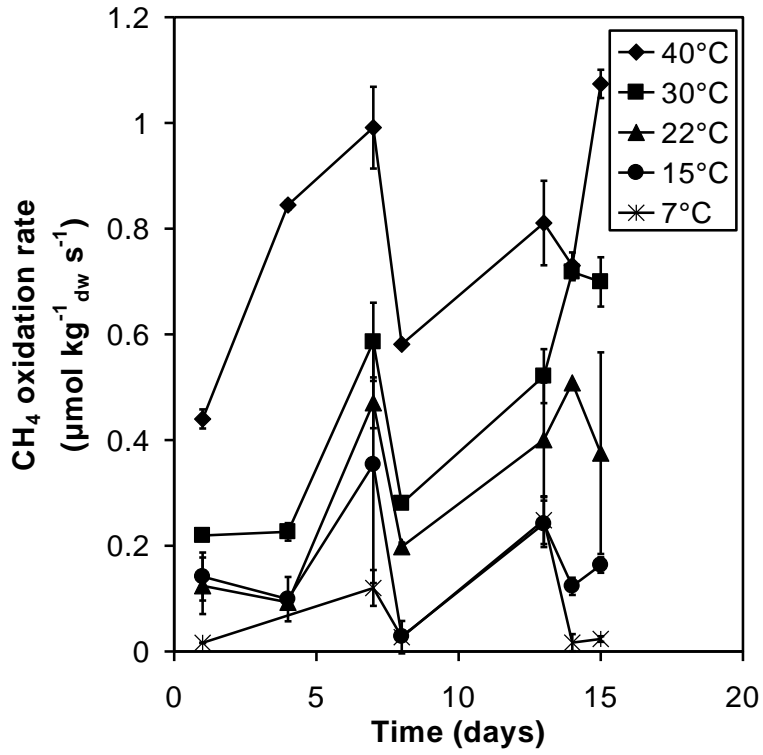
Error bars are standard deviation.

Fig. 5. Effect of temperature on the CH$_4$ oxidation rate by compost G3 versus time. Error bars are standard deviation.