THE EFFECT OF LANDFILL LEACHATE IRRIGATION ON SOIL GAS COMPOSITION: METHANE OXIDATION AND NITROUS OXIDE FORMATION

A. WATZINGER[1,*], T. G. REICHENAUER[2], W. E. H. BLUM[1], M. H. GERZABEK[1] and S. ZECHMEISTER-BOLTENSTERN[3]

[1]*Institute of Soil Research, Department of Forest and Soil Sciences, University of Natural Resources and Applied Life Sciences, Peter JordanStrasse 82, 1190 Vienna, Austria;* [2]*Department of Environmental Research, ARC seibersdorf research GmbH, 2444 Seibersdorf, Austria;* [3]*Department of Forest Ecology, Federal Office and Research Centre for Forests, Seckendorff-GudentWeg 8, 1131 Vienna, Austria*

([*]author for correspondence, e-mail: andrea.watzinger@boku.ac.at; Tel.: +43-1-47654-3109, Fax: +43-1-47654-3130)

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Abstract. The treatment of landfill leachate by irrigation of the recultivation layer of landfills sites might interfere with greenhouse gas cycling in the soil through alteration of the microbial methane oxidation capacity and promotion of nitrous oxide formation. The interaction of landfill leachate irrigation, methane oxidation and nitrous oxide formation was investigated in a compost – gravel recultivation substrate with and without landfill gas fumigation during a two year lysimeter experiment. Microbial methane oxidation started 3 days after landfill gas addition, and it was promoted by less than 150 mm of landfill leachate application. While long term landfill leachate irrigation negatively affected methane oxidation corresponding to the increasing soil moisture content. In respect to nitrous oxide, formation was low under landfill gas fumigation, while landfill leachate application triggered nitrous oxide production. Only low amounts (<200 mm) might avoid increasing greenhouse gas concentrations in landfill leachate irrigated soil.

Keywords: methane oxidation, nitrous oxide, landfill leachate, irrigation, landfill cover, recultivation

1. Introduction

Carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are important greenhouse gases. Although present in lower concentrations, CH_4 and N_2O have respectively a 23 and 296-fold global warming potential (GWP) refering to the 100 year horizon values compared to CO_2. The atmospheric concentration of CH_4 has increased by a factor of 2.5 since the preindustrial era. 13% of the global anthropogenic emissions derive from landfills (IPCC, 2001), while in Austria, the main source of CH_4 (42%) are landfills (Federal Environment Agency Ltd., 2002). 5% of the released CH_4 is estimated to be reduced by the soil (IPCC, 2001). N_2O emissions from soils, which are believed to be caused by increasing soil N availability driven by increasing fertiliser use and N deposition, can explain the increase in the atmospheric

N_2O abundance. Soils contribute 65% to the natural and anthropogenic N_2O emissions (IPCC, 2001). In Austria, 49% of the total anthropogenic N_2O emission originated from agricultural soil in 2001 (Federal Environment Agency Ltd., 2002).

Methane generation from landfill sites is likely to increase in future, especially in developing countries (Meadows *et al.*, 1997). Microbial methane oxidation (MMO) in the top cover of landfill sites seems to be a preferable option for reduction of rate of CH_4 emissions because: (1) CH_4 emissions cannot be reduced completely, even if a landfill gas collection system is installed (Humer and Lechner, 1999; Börjesson and Svensson, 1997); (2) for low CH_4 production, such as in old and small landfill sites, collection and use is technically difficult and in most cases not feasible (Damman *et al.*, 1999); and (3) low cost options for methane reduction are required especially in developing countries. MMO is controlled by physical parameters such as temperature, moisture content, soil structure and texture, and by chemical parameters such as availability of nutrients and toxic substances and the microbial community. High water content and low soil porosity limit the CH_4 and O_2 supply to microorganisms, while low moisture content induces physiological stress to the methanotrophic population (Boeckx and Van Cleemput, 2000). Low temperature becomes MMO rate limiting at low moisture contents (Boeckx and Van Cleemput, 2000). CH_4 oxidation influences the N turnover by altering the redox potential of the soil and consequently influencing nitrification and denitrification processes. N_2O production is increased under high moisture content and high CH_4 concentration in the soil owing to O_2 limitations (Börjesson *et al.*, 1998; Bogner *et al.*, 1999).

Landfill gas is not the sole emission from waste sites. Often more of a concern is landfill leachate. Landfill leachate irrigation has been described as a cost-effective and relatively simple treatment option which can improve leachate quality and reduce its quantity (Cureton *et al.*, 1991; Hasselgren, 1992; Ettala, 1992). Irrigation has been proposed on landfill cover soil, mainly to save land and to use it efficiently for biomass production as a carbon neutral energy source.

Methane oxidation has been mainly investigated in laboratory scale experiments (Börjesson *et al.*, 1998; Hilger *et al.*, 2000a; Kightley *et al.*, 1995). Field experiments focused on actual gas emissions (Einola *et al.*, 2003; Giani *et al.*, 2002; Maurice and Lagerkvist, 2003) rather than on the process of gas production in the soil profile. To quantify the effect on greenhouse gas emissions, all relevant gases (N_2O, CH_4, CO_2), their behaviour and interaction have to be monitored. Only a few authors have measured all of these gases (Börjesson *et al.*, 1998, Einola *et al.*, 2003). Processes of N_2O production and elimination have largely been investigated in natural soil (Smith *et al.*, 2003). However, data are scarce for N_2O gas profiles in artificial landfill cover soil, except for results published by Bogner *et al.* (1999). Little is known about the effect of vegetation on the gas composition, but effects are expected via the influence vegetation has on soil microorganisms, e.g. in the rhizosphere. Hilger *et al.* (2000a) found higher peak uptake of CH_4 and an upward shift in the location of biomass accumulation in vegetated soil columns. Also, the effect of landfill leachate irrigation on greenhouse gas emissions under CH_4 addition is unclear. Maurice *et al.* (1999) reported a supportive effect of landfill leachate irrigation on MMO if plant growth was stimulated as well. N_2O production under landfill leachate irrigation increased, but the irrigated substrate was not used for CH_4 oxidation (Lee *et al.*, 2002; Hui *et al.*, 2003).

This study presents the findings of an investigation into the impact of landfill leachate irrigation on the distribution profile of greenhouse gases (CH_4, N_2O, CO_2) in an artificial revegetated landfill cover soil under outdoor conditions.

2. Materials and Methods

At the beginning of February 2002 a lysimeter experiment was set up at the ARC research seibersdorf GmbH in Austria (16°30′24″E, 47°58′34″N). The precipitation during the experimental period (April 2002 – September 2003) amounted to 557 mm in 2002 and 357 mm in 2003 (Figure 1a). Sixteen lysimeters, each with a volume of 200 l and a surface area of 0.29 m^2, were filled with a landfill cover substrate consisting of a mixture of 50 vol% biowaste compost ($\leq$10 mm) (Table I), 33 vol% quartz gravel and 17 vol% carbonate gravel and a 0.20 m thick drainage layer of quartz gravel at the bottom (Figure 2). The texture of the quartz gravel was a clayey sand having a sand/silt/clay distribution of 76/1/23% with 46% gravel ($\leq$7 mm). The carbonate gravel was a sandy clay (72/1/27%) with 51% gravel ($\leq$8 mm). Each lysimeter was vegetated with two cuttings of *Salix viminalis* L. and *Populus nigra* L. at the beginning of March 2002 and irrigated with 152 mm tap water to allow rooting until the irrigation regime was implemented in June (Figure 1b). A perforated ring was placed within the drainage

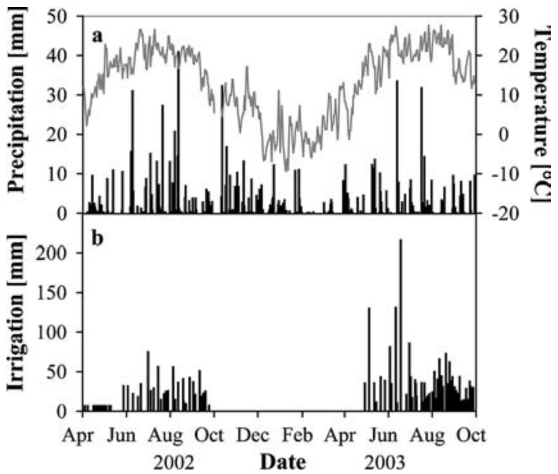

Figure 1. (a) Daily temperature (grey) and precipitation (black) and (b) irrigation during the experimental period.

TABLE I
Characterisation of the compost (≤10 mm)

Parameter	Unit	Value
Water capacity	%	174
Dry density	g cm^{-3}	0.49
pH		7.12
EC	mS cm^{-1}	1.98
C_{org}	%	17.9
N_{total}	g kg^{-1}	14.7
NO_3-N	mg kg^{-1}	16
NH_4-N	mg kg^{-1}	177
P_{total}	g kg^{-1}	1.3
P_{CAL}[a]	g kg^{-1}	0.37
K_{total}	g kg^{-1}	3.4
K_{CAL}	g kg^{-1}	2.2
Ca_{total}	g kg^{-1}	56.7
Mg_{total}	g kg^{-1}	14.9

[a]Extracted with calcium-acetat-lactat.

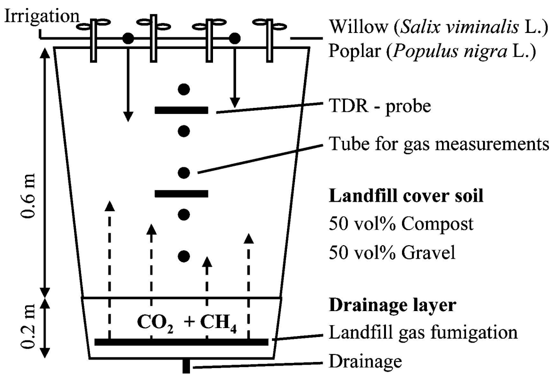

Figure 2. Experimental design of the lysimeters.

material for fumigation purposes. The recultivation substrate was filled in 0.01 m layers, which were compacted to an average density of 1.2 g cm^{-3}. Perforated tubes were installed at depths of 0.1–0.5 m for soil gas collection.

Four different treatments, tap water irrigation (W), landfill leachate irrigation (LL), tap water irrigation and fumigation with landfill gas (W + LG) and landfill leachate irrigation and fumigation with landfill gas (LL + LG), were set up in four replicates. Lysimeters were fumigated with 166.6 l m^{-2} d^{-1} of artificial landfill gas consisting of 60 vol% CH_4 and 40 vol% CO_2 from May 2002 onwards.

During the growing season (June–September 2002 and April–September 2003) the lysimeters were irrigated either with landfill leachate or tap water as a control if the soil moisture content at 0.15 m in the control treatment (W) dropped below 45 vol% and 40 vol% in the irrigation years 2002 and 2003, respectively. In total, 749 mm of irrigation water were applied in 2002, and 1738 mm tap water and 1763 mm landfill leachate in 2003 (Figure 1b). The landfill leachate was obtained from an operating municipal solid waste landfill site in Vienna (Rautenweg) (Table II). The soil moisture content was measured every two hours at 0.15 and 0.35 m by time domain reflectometry (TDR) probes and water potential was measured by tensiometers (Soilmoisture Equipment Corp., USA). Plant performance was monitored throughout the experiment. On the 1st and 2nd of October 2003, the lysimeters were dismantled and the soil profile was described.

CH_4, CO_2, O_2 and H_2S concentrations were measured by a portable gas analyser (LMSx, Gas Data Ltd, UK) weekly in 2002 and fortnightly in 2003. N_2O concentrations were measured by gas chromatography (GC). Samples were collected in September 2002 and in June, July and September 2003. The gas sampling tube in the lysimeter and the syringe were flushed with 50 ml before sampling. Samples collected in 2002 were analysed by GC (Varian 3400, column: RCX, 30 m length,

TABLE II

Chemical characterisation of the landfill leachate in 2002 and 2003

Parameter	Unit	2002	2003
pH		7.14	6.98
EC[a]	mS cm^{-1}	7.00	8.50
COD[b]	mg l^{-1}	250	59
TOC[c]	mg l^{-1}	62	79
NO_3-N	mg l^{-1}	6.8	21
NH_4-N	mg l^{-1}	136	130
PO_4-P	mg l^{-1}	n.d.[d]	n.d.
SO_4	mg l^{-1}	270	150
Cl	mg l^{-1}	1400	1900
Na	mg l^{-1}	800	1000
K	mg l^{-1}	240	310
Ca	mg l^{-1}	130	140
Mg	mg l^{-1}	190	210
B	mg l^{-1}	3.1	4.1
Cu	μg l^{-1}	<5.0	<5.0

[a]Electric conductivity.
[b]Chemical oxygen demand – $KMnO_4$ demand.
[c]Total organic carbon.
[d]Below detection limit.

0.32 mm inner diameter, 3 μm film thickness) and mass spectrometry (ITS 40, Finnigan) and injected by using a headspace sampler (HS 40, Perkin Elmer). All other samples were analysed with a GC (Hewlett Packard 5890 II series) equipped with a ^{63}Ni electron capture detector (ECD) connected to a headspace sampler (DANI HSS 86.50). All statistical analyses were done by SPSS for Windows 11.0. All differences presented were confirmed by *t*-test or ANOVA and a post hoc test (Scheffe test) at a 0.05 confidence level.

3. Results

3.1. Vegetation performance and description of the soil profile

In the first month, plant growth was greater under landfill leachate irrigation in comparison with the water treatment, but then deteriorated. In spring 2003, before irrigation was commenced, plants had recovered. However, after the onset of landfill leachate irrigation, plant growth declined again and most of the plants died before the end of the experiment. Landfill gas fumigation influenced vegetation little, but positively.

In non landfill gas fumigated soil, the profile was homogenous and roots extended into the drainage layer. However, roots have died under landfill leachate irrigation. Rooting depth was restricted under landfill gas fumigation and distinct horizons developed in W + LG (Figure 3). A reddish brown band of 0.05 m was identified below 0.10–0.15 m, followed by an approximately 0.025 m thick layer containing white filaments. A dense root mat developed above these layers. Some dead roots still occurred at lower depth. The reddish brown colour primarily occurred along the roots. Apart from the reddish and white discolouring, the soil was darker and the substrate was aggregated. Distinct horizons were not noted in LL + LG. However a darker, 0.05 m thick layer was found within the first 0.20 m. A strong smell of H_2S, high water content and low stability of the soil aggregates was noted when LL + LG lysimeters were dismantled.

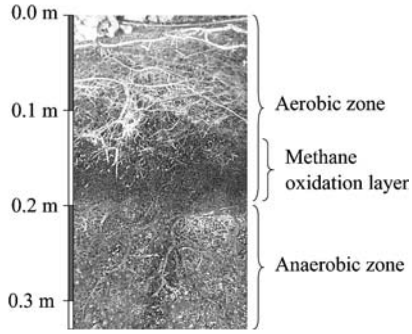

Figure 3. Soil profile of W+LG.

3.2. Soil temperature and soil moisture

Soil temperature significantly increased under landfill gas fumigation in comparison with non fumigated lysimeters at the end of April 2003, especially in LL + LG. After irrigation was commenced, soil temperature dropped in LL + LG, while it remained elevated in W + LG until September 2003. The average soil temperature increase was around 4 °C and more or less constant with depth. Outside temperature strongly influenced soil temperature. Soil temperature correlated negatively with CO_2 and CH_4 concentrations in W + LG, especially in the lower soil depths.

The soil moisture content under landfill leachate irrigation remained high at 45–50 vol% in 2002 and about 40 vol% in 2003 and ponding of water on the soil surface was observed. In contrast, the soil moisture generally dropped under water irrigation in summer. The decrease was restricted to the upper horizons in W + LG.

O_2 decreased and CO_2 increased significantly with increasing soil moisture content in W, while an opposite correlation was observed in LL. The association between soil moisture and gas composition was stronger at greater depths. Besides soil moisture, the ambient air temperature (Figure 1a) appeared to influence the soil gas composition. An increase in temperature decreased the O_2 and increased the CO_2 content in W and LL. A multivariate general linear model, type III, indicated a stronger impact of air temperature than soil moisture on the gas distribution especially in the upper horizons. The impact of soil moisture and air temperature on the gas concentrations was less clear under landfill gas fumigation.

3.3. Soil gas distribution

Monthly averages of O_2, CO_2 and CH_4 concentrations in the soil are listed in Tables III–V. Within the first irrigation month, landfill leachate application increased O_2 concentrations in the upper horizon (Figure 4a) and subsequently in the lower horizon in non fumigated lysimeters (Figure 4b). No significant differences between irrigation regimes were observed in summer 2002, while in autumn, landfill leachate irrigation showed decreased O_2 and increased CO_2 concentrations especially at lower depths (Figure 4c). In spring 2003, CO_2 concentrations were lower and O_2 concentrations were higher in the upper horizons of LL (Figure 4d), which reversed in summer (Figure 4e). Generally, the change in CO_2 and O_2 concentrations with depth was more linear in LL than W throughout the experimental year 2003 (linear regression, $R^2_{\text{water}} < R^2_{\text{landfill leachate}}$).

Full landfill gas migration through the lysimeters (0.6 m) was detected after less than 24 h from the onset of landfill gas fumigation in May 2002. The CH_4 and CO_2 concentrations line in the soil profile crossed at 0.1 m after 3 days under both irrigation regimes, but differences between CO_2 and CH_4 were not significant until 3 weeks after the onset of fumigation. In LL + LG CH_4 concentrations decreased

TABLE III

Monthly averages and standard deviations (in brackets) of the O_2 concentration in vol% in the soil

		2002							2003					
Treatment	Depth (m)	April $n = 6$	June $n = 5$	July $n = 5$	Aug. $n = 5$	Sept. $n = 4$	Oct. $n = 5$	Nov. $n = 4$	April $n = 3$	May $n = 2$	June $n = 2$	July $n = 2$	Aug. $n = 2$	Sept. $n = 3$
W	0.1	17.1 (1.3)	16.4 (1.1)	15.1 (0.7)	13.1 (1.7)	16.8 (1.3)	19.6 (0.3)	19.6 (0.2)	19.9 (0.3)	18.0 (0.6)	18.9 (0.6)	18.2 (0.8)	18.2 (0.4)	18.7 (0.5)
W	0.2	11.6 (1.6)	12.3 (1.3)	11.0 (0.8)	9.1 (1.7)	14.9 (1.5)	18.7 (0.4)	18.8 (0.3)	18.9 (0.2)	15.3 (0.7)	17.5 (1.0)	16.1 (0.7)	17.2 (0.1)	18.0 (0.4)
W	0.3	3.9 (1.6)	4.8 (1.5)	6.5 (0.9)	5.2 (1.6)	13.5 (2.0)	17.6 (0.6)	17.9 (0.4)	17.4 (0.2)	12.1 (0.6)	16.5 (1.6)	14.6 (0.5)	16.4 (0.2)	17.5 (0.5)
W	0.4	2.2 (1.7)	1.8 (0.7)	4.4 (1.0)	3.7 (1.3)	13.0 (2.2)	17.2 (0.8)	17.4 (0.4)	16.5 (0.2)	10.4 (0.9)	16.0 (1.7)	13.7 (0.5)	16.0 (0.3)	17.3 (0.5)
W	0.5	1.7 (1.6)	1.0 (0.6)	2.5 (0.7)	2.4 (1.1)	12.5 (2.4)	16.8 (1.0)	17.0 (0.6)	15.8 (0.1)	8.9 (1.1)	15.7 (1.8)	13.2 (0.5)	15.6 (0.3)	17.1 (0.5)
LL	0.1	17.6 (0.5)	17.0 (0.6)	16.7 (0.3)	15.6 (0.5)	17.6 (0.4)	19.1 (0.2)	19.7 (0.3)	20.2 (0.1)	18.4 (0.5)	17.7 (0.6)	17.8 (0.9)	17.9 (0.8)	19.1 (0.4)
LL	0.2	10.7 (1.5)	12.6 (1.1)	12.6 (0.7)	12.7 (1.1)	15.1 (0.7)	17.8 (0.3)	18.5 (0.4)	19.0 (0.3)	15.6 (0.8)	14.2 (0.7)	14.6 (1.1)	15.2 (0.6)	17.2 (0.7)
LL	0.3	3.7 (1.1)	6.0 (1.7)	7.4 (0.8)	6.6 (1.5)	11.2 (1.4)	15.6 (0.6)	16.5 (0.9)	17.6 (0.4)	12.2 (0.6)	10.0 (0.8)	9.9 (1.4)	10.6 (1.2)	14.3 (0.7)
LL	0.4	1.8 (0.9)	2.4 (0.7)	4.8 (0.9)	4.2 (1.0)	9.2 (1.5)	14.4 (0.6)	15.2 (0.9)	16.5 (0.5)	10.1 (0.7)	7.7 (0.7)	7.3 (1.0)	9.3 (2.5)	12.6 (0.4)
LL	0.5	1.2 (0.7)	1.5 (1.0)	3.5 (1.1)	3.0 (1.1)	9.5 (0.8)	13.8 (1.2)	15.1 (1.3)	15.2 (1.1)	8.1 (0.4)	6.0 (1.0)	6.0 (1.4)	6.7 (0.1)	10.8 (1.3)
W+LG	0.1	17.3 (1.3)	6.2 (1.9)	4.5 (3.6)	5.5 (3.3)	4.1 (2.3)	5.4 (3.1)	1.8 (–)	8.4 (4.2)	5.8 (3.1)	3.9 (2.2)	3.6 (2.3)	5.6 (2.2)	5.0 (3.3)
W+LG	0.2	12.2 (1.5)	1.6 (1.0)	1.2 (0.5)	0.9 (0.5)	0.5 (0.1)	0.5 (0.4)	0.3 (–)	1.9 (1.0)	1.5 (0.7)	1.1 (1.1)	0.8 (0.6)	0.5 (0.7)	0.0 (0.0)
W+LG	0.3	5.0 (1.1)	0.3 (0.2)	0.3 (0.1)	0.2 (0.2)	0.0 (0.0)	0.0 (0.0)	0.1 (–)	0.2 (0.2)	0.2 (0.1)	0.2 (0.2)	0.1 (0.1)	0.0 (0.0)	0.0 (0.0)
W+LG	0.4	2.3 (1.0)	0.1 (0.1)	0.0 (0.0)	0.2 (0.2)	0.0 (0.0)	0.0 (0.0)	0.0 (–)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)
W+LG	0.5	1.8 (0.9)	0.0 (0.0)	0.0 (0.0)	0.0 (0.1)	0.0 (0.0)	0.0 (0.0)	0.1 (–)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)	0.0 (0.0)
LL+LG	0.1	18.0 (1.0)	8.9 (2.2)	7.4 (2.9)	6.1 (2.3)	7.3 (2.9)	5.4 (1.6)	6.3 (2.3)	7.3 (2.4)	5.4 (3.6)	8.6 (2.7)	9.2 (2.1)	–	14.9 (2.2)
LL+LG	0.2	12.2 (2.4)	2.7 (2.3)	1.8 (1.4)	1.6 (0.5)	2.3 (1.3)	1.0 (0.3)	1.0 (0.5)	1.7 (0.9)	1.2 (0.8)	1.2 (1.0)	1.9 (1.9)	–	0.9 (1.8)
LL+LG	0.3	3.9 (1.3)	0.4 (0.3)	0.4 (0.2)	0.5 (0.4)	0.6 (0.5)	0.2 (0.1)	0.1 (0.1)	0.1 (0.1)	0.0 (0.0)	0.0 (0.1)	0.2 (0.1)	–	0.0 (0.0)
LL+LG	0.4	2.5 (1.2)	0.1 (0.1)	0.1 (0.1)	0.8 (0.8)	1.3 (1.5)	0.2 (0.1)	0.1 (0.1)	0.0 (0.0)	0.2 (0.5)	0.0 (0.0)	0.0 (0.1)	–	0.0 (0.0)
LL+LG	0.5	1.9 (1.3)	0.1 (0.0)	0.0 (0.0)	0.0 (0.1)	1.0 (0.8)	0.3 (0.2)	0.2 (0.2)	0.0 (0.0)	0.0 (0.0)	0.0 (0.1)	0.0 (0.0)	–	0.0 (0.0)

n: Number of measuring dates.
–: Missing value.

TABLE IV

Monthly averages and standard deviations (in brackets) of the CO_2 concentration in vol% in the soil

		2002							2003					
Treatment	Depth (m)	April $n = 6$	June $n = 5$	July $n = 5$	Aug. $n = 5$	Sept. $n = 4$	Oct. $n = 5$	Nov. $n = 4$	April $n = 3$	May $n = 2$	June $n = 2$	July $n = 2$	Aug. $n = 2$	Sept. $n = 3$
W	0.1	3.3 (1.3)	4.5 (1.3)	6.3 (1.0)	8.1 (2.0)	4.4 (1.6)	1.6 (0.5)	1.2 (0.2)	1.2 (0.2)	3.4 (0.9)	2.3 (0.9)	2.9 (0.9)	2.2 (0.3)	1.9 (0.3)
W	0.2	8.2 (1.3)	8.5 (1.4)	11 (1)	12 (2)	6.4 (1.8)	2.5 (0.6)	2.0 (0.4)	2.3 (0.2)	6.7 (1.1)	3.8 (1.5)	5.1 (1.0)	2.9 (0.1)	2.3 (0.3)
W	0.3	14 (0)	15 (1)	15 (1)	15 (2)	8.0 (2.4)	3.5 (1.0)	2.7 (0.4)	3.9 (0.3)	10 (1)	4.9 (2.1)	6.9 (0.9)	3.8 (0.2)	2.7 (0.4)
W	0.4	15 (1)	18 (1)	18 (1)	17 (2)	8.4 (2.5)	3.9 (1.1)	3.1 (0.4)	4.8 (0.2)	12 (1)	5.5 (2.3)	7.7 (0.7)	4.1 (0.3)	2.9 (0.4)
W	0.5	15 (1)	19 (1)	19 (1)	18 (2)	9.0 (2.8)	4.3 (1.5)	3.4 (0.6)	5.5 (0.2)	14 (2)	5.8 (2.2)	8.1 (0.6)	4.5 (0.4)	3.1 (0.5)
LL	0.1	2.7 (0.5)	3.9 (0.6)	4.5 (0.4)	5.3 (0.6)	3.2 (0.4)	1.7 (0.2)	1.0 (0.2)	1.0 (0.1)	3.1 (0.6)	3.6 (0.9)	2.9 (0.9)	1.9 (0.8)	1.4 (0.5)
LL	0.2	8.8 (1.3)	8.5 (1.0)	9.1 (0.8)	8.9 (0.8)	5.9 (0.8)	3.0 (0.3)	2.2 (0.4)	2.2 (0.2)	6.8 (0.8)	7.6 (0.9)	6.1 (1)	4.3 (0.6)	2.9 (0.6)
LL	0.3	14 (1)	15 (2)	15 (1)	14 (1)	10 (1)	5.2 (0.9)	3.8 (0.7)	3.5 (0.5)	10 (1)	12 (0.9)	11 (1)	8.5 (0.9)	5.3 (0.6)
LL	0.4	16 (0)	18 (1)	17 (1)	16 (1)	12 (1)	6.5 (0.8)	4.9 (0.8)	4.7 (0.7)	13 (1)	15 (1)	13 (1)	10 (2)	6.8 (0.3)
LL	0.5	16 (0)	19 (1)	19 (1)	17 (2)	11 (1)	6.7 (1.6)	5.0 (1.1)	5.8 (1.1)	15 (1)	16 (1)	14 (1)	13 (0)	8.2 (0.8)
W+LG	0.1	3.0 (0.8)	18 (2)	17 (5)	19 (9)	22 (6)	20 (8)	34 (–)	17 (7)	20 (8)	19 (4)	24 (6)	20 (5)	25 (5)
W+LG	0.2	7.9 (1.4)	31 (3)	28 (3)	27 (5)	29 (4)	28 (6)	36 (–)	32 (3)	29 (3)	26 (3)	33 (3)	29 (3)	36 (2)
W+LG	0.3	13 (1)	36 (1)	33 (2)	31 (5)	32 (3)	32 (4)	37 (–)	37 (1)	35 (3)	31 (3)	37 (3)	32 (4)	36 (2)
W+LG	0.4	15 (1)	36 (1)	35 (2)	34 (2)	35 (1)	34 (2)	37 (–)	39 (2)	36 (2)	34 (2)	38 (2)	34 (3)	37 (2)
W+LG	0.5	16 (1)	37 (1)	35 (2)	35 (1)	36 (1)	36 (1)	37 (–)	38 (1)	38 (1)	37 (2)	38 (2)	34 (2)	37 (2)
LL+LG	0.1	2.4 (0.9)	16 (4)	15 (3)	20 (1)	21 (5)	25 (3)	25 (4)	18 (3)	24 (6)	18 (4)	20 (4)	–	10 (2)
LL+LG	0.2	7.6 (2.1)	27 (4)	28 (3)	30 (5)	30 (2)	35 (2)	34 (1)	30 (2)	34 (2)	31 (4)	34 (3)	–	32 (3)
LL+LG	0.3	14 (1)	34 (3)	34 (3)	31 (8)	32 (6)	36 (1)	35 (1)	36 (2)	37 (1)	35 (3)	36 (0)	–	34 (1)
LL+LG	0.4	15 (1)	35 (1)	36 (2)	36 (3)	32 (5)	36 (1)	36 (1)	37 (2)	37 (1)	36 (2)	37 (0)	–	35 (1)
LL+LG	0.5	15 (1)	35 (2)	36 (1)	38 (3)	37 (3)	36 (2)	35 (1)	37 (2)	37 (1)	37 (1)	37 (1)	–	35 (1)

n: Number of measuring dates.
–: Missing value.

TABLE V

Monthly averages and standard deviations (in brackets) of the CH_4 concentration in vol% in the soil

		2002							2003						
Treatment	Depth (m)	April $n = 6$	June $n = 5$	July $n = 5$	Aug. $n = 5$	Sept. $n = 4$	Oct. $n = 5$	Nov. $n = 4$	April $n = 3$	May $n = 2$	June $n = 2$	July $n = 2$	Aug. $n = 2$	Sept. $n = 3$
W+LG	0.1	0.0 (0.0)	10 (3)	6.7 (6.3)	10 (18)	17 (15)	13 (13)	50 (–)	10 (8)	14 (15)	10 (18)	12 (18)	12 (7)	31 (18)
W+LG	0.2	0.0 (0.0)	33 (5)	24 (10)	19 (22)	28 (18)	25 (19)	57 (–)	28 (4)	26 (13)	19 (12)	26 (13)	29 (5)	55 (6)
W+LG	0.3	0.0 (0.0)	56 (2)	46 (10)	31 (21)	39 (16)	38 (17)	60 (–)	45 (4)	40 (14)	33 (15)	39 (12)	40 (10)	57 (4)
W+LG	0.4	0.0 (0.0)	59 (0)	55 (7)	45 (12)	51 (8)	51 (8)	60 (–)	54 (2)	47 (11)	44 (11)	49 (6)	49 (5)	59 (2)
W+LG	0.5	0.0 (0.0)	60 (0)	58 (2)	56 (1)	59 (2)	60 (1)	60 (–)	58 (1)	57 (3)	55 (5)	57 (1)	56 (3)	60 (1)
LL+LG	0.1	0.0 (0.0)	6.1 (3.8)	5.7 (1.9)	19 (6)	30 (6)	36 (5)	34 (5)	8.1 (2)	24 (12)	17 (5)	24 (14)	–	13 (3)
LL+LG	0.2	0.0 (0.0)	27 (11)	30 (12)	36 (15)	46 (7)	56 (2)	56 (3)	27 (3)	47 (11)	54 (5)	53 (8)	–	55 (6)
LL+LG	0.3	0.0 (0.0)	52 (9)	51 (8)	51 (9)	56 (9)	60 (1)	60 (2)	46 (3)	55 (6)	59 (1)	58 (2)	–	59 (1)
LL+LG	0.4	0.0 (0.0)	58 (1)	56 (1)	55 (3)	56 (8)	60 (1)	60 (1)	53 (2)	57 (3)	59 (1)	58 (2)	–	59 (1)
LL+LG	0.5	0.0 (0.0)	60 (0)	57 (1)	59 (1)	58 (5)	59 (1)	59 (1)	58 (2)	59 (1)	59 (0)	58 (1)	–	59 (0)

n: Number of measuring dates.
–: Missing value.

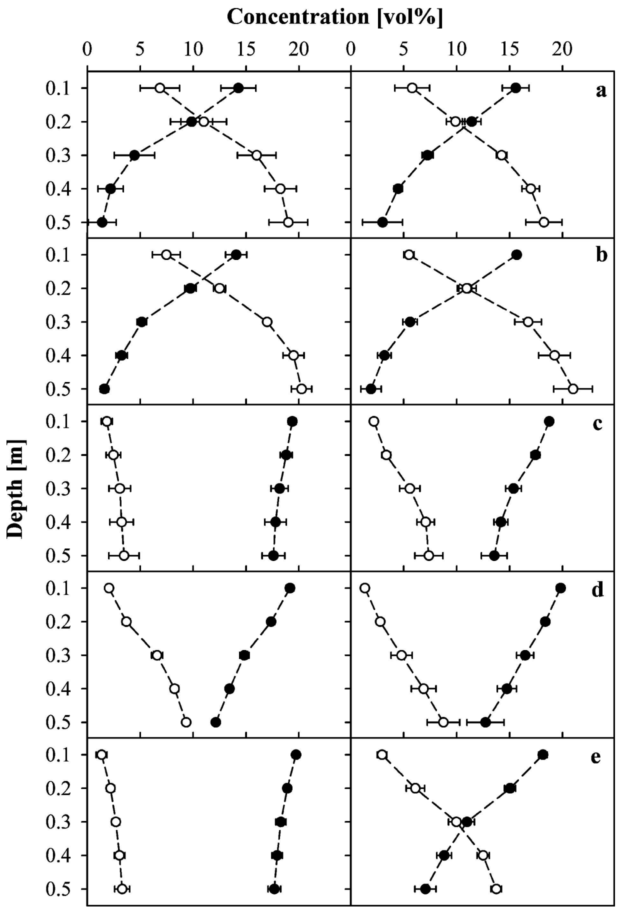

Figure 4. O_2 (full circles) and CO_2 (open circles) concentrations in W (left) and LL (right) (a) two weeks after irrigation commenced in 2002, (b) after five weeks irrigation, (c) two weeks after irrigation was stopped in autumn 2002, (d) just before irrigation commenced in spring 2003 and (e) after eight weeks irrigation in summer 2003. Error bars indicate the standard deviations.

after two weeks (Figure 5a), but this effect vanished after four weeks irrigation. On the contrary, CH_4 concentrations were observed to increase under long term landfill leachate irrigation (Figure 5b). Even after cessation of irrigation in autumn 2002, CH_4 concentration remained high under LL + LG, while it decreased under W + LG (Table V) (Figure 5c). Consequently, some W + LG lysimeters became gas impermeable, as it had been already observed in summer. In winter, CH_4 and CO_2 concentrations matched the incoming landfill gas composition throughout the soil profile, except at 0.1 m depth, independently of its irrigation treatment (Figure 5d). In spring 2003, before irrigation commenced, CH_4 concentrations decreased again in both irrigation treatments (Figure 6a). After irrigation was commenced, CH_4 concentrations increased in LL + LG in comparison to W + LG after four to

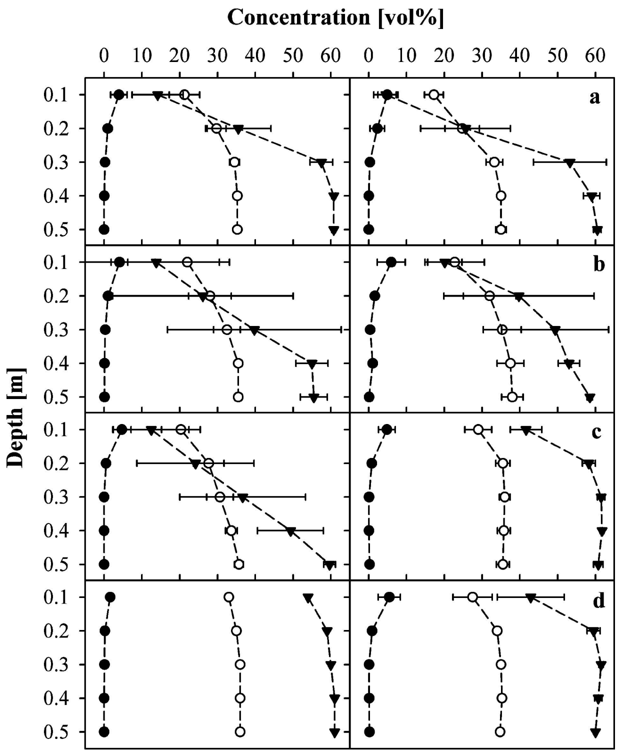

Figure 5. O_2 (full circles), CO_2 (open circles) and CH_4 (full triangles) concentrations in W + LG (left) and LL + LG (right) in 2002 (a) two weeks after irrigation commenced, (b) in late summer, (c) two weeks after irrigation was stopped in autumn, (d) in winter. Error bars indicate the standard deviations.

eight weeks (Figure 6b), and the shape of the gas distribution curve in the soil profile changed as follows: The soil gas composition matched the input gas composition up to 0.2 m in LL + LG, then sharply diverged. In contrast, CH_4 concentrations decreased constantly over the whole depth in W + LG. Similar gas distribution patterns have previously been noted in September and October 2002 (Figure 5c). From September 2003 and onwards, CH_4 concentrations in W + LG increased, and the gas distribution pattern approximated the distribution pattern of LL + LG. At the end of the irrigation year 2003, no significant differences between LL + LG and W + LG were monitored (Figure 6c). Production of CH_4 was occasionally detectable in LL + LG.

N_2O concentrations were only elevated in LL, where the concentrations of N_2O increased with depth and duration of landfill leachate application (Table VI). N_2O concentrations of all other treatments were low, averaging 1.4, 0.5 and 1.7 $\mu L\ L^{-1}$ in W, W + LG and LL + LG, respectively. A trend of N_2O increase with depth was observed in W. When N_2O was formed under landfill gas fumigation, higher

TABLE VI

Averages and standard deviations (in brackets) of the N_2O concentrations in $\mu L\,L^{-1}$ in LL. The significances of difference between depths (p_{depth}) and dates (p_{date}) have been calculated

Depth (m)	Sept. 2002	June 2003	July 2003	Sept. 2003	p_{date}
0.1	16 (8)	26 (15)	47 (23)	61 (35)	0.067
0.2	27 (17)	40 (26)	79 (34)	122 (57)	0.014
0.3	49 (20)	69 (50)	137 (56)	243 (117)	0.003
0.4	68 (27)	93 (77)	181 (84)	310 (191)	0.041
0.5	71 (22)	157 (117)	334 (123)	589 (406)	0.061
p_{depth}	0.007	0.106	0.001	0.024	

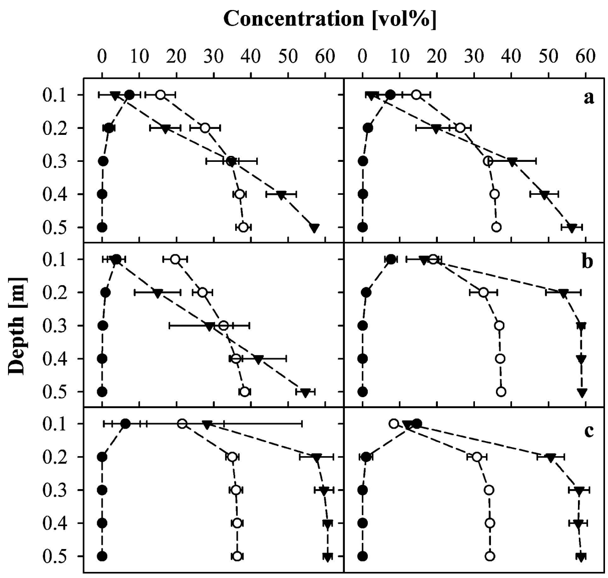

Figure 6. O_2 (full circles), CO_2 (empty circles) and CH_4 (full triangles) concentrations in the soil fumigated with landfill gas and irrigated with water (left) and landfill leachate (right) in 2003 (a) just before irrigation commenced, (b) after eight weeks irrigation and (c) at the end of irrigation. Error bars indicate the standard deviations.

concentrations were monitored in the upper horizons. H_2S formation was associated with landfill gas fumigation. H_2S concentrations were highest in LL + LG (85 $\mu L\,L^{-1}$ at 0.3 m in October 2002). In comparison, the maximum concentration was only 28 $\mu L\,L^{-1}$ in W + LG at 0.4 m in August 2002. In non fumigated soil, some H_2S was occasionally detected at 0.4 and 0.5 m during summer 2002. The variation of both H_2S and N_2O concentrations was high over time and between replicates.

4. Discussion

4.1. Characteristics of methane oxidation

During the chemical reaction of methane oxidation 2 mol CH_4 are consumed and 1 mol CO_2 is produced. Considering incorporation of carbon into the microbial biomass, the crossing of CH_4 and CO_2 concentrations in the soil profile presents a confident indicator for the start of MMO. Accordingly, MMO was already found 3 days after adding artificial landfill gas, which was only 2 days after the full migration of landfill gas through our lysimeters. Similarly low lag phases were found by Christophersen *et al.* (2000) and Kightley *et al.* (1995) following interruption of methane supply. Accordingly, our original compost might have contained a population of rather inactive methanotrophs and was a good substrate for microbial growth.

Soil temperature increased under MMO owing to the microbial activity, but temperature was still strongly influenced by the outside temperature, especially in the upper horizons. Due to the low self heating potential and a low methane oxidation depth, MMO ceased in winter under temperatures below 0 °C. Accordingly, Börjesson *et al.* (2001) suggested that, MMO does not take place at temperatures below 0 °C and Boeckx and Van Cleemput (2000) suggested higher susceptibility of MMO to extreme temperatures deriving from a low MMO depth. In contrast, MMO continued in biofilters during winter (Streese and Stegmann, 2003).

A more linear decrease of CH_4 concentration was measured under high MMO. Similar gas distributions to ours were measured by Kightley *et al.* (1995) and Hilger *et al.* (1999), who suspected a rather wide band of methanotrophic activity resulting from a good aeration of the soil. However, in our experiment no O_2 was detected below 0.3 m, and discolouring of the soil, which indicated the presence of methane oxidizers according to Wilshusen *et al.* (2004), was found in a defined layer at about 0.10 to 0.15 m depth. Anaerobic oxidation of methane (AOM) would explain the decrease of CH_4 in anoxic horizons. However, up to date AOM has been found via reduction of SO_4 and consequent production of H_2S in marine sediments only (Valentine, 2002). In our experiment, H_2S production was too low to fully account for AOM and the time of H_2S production did not correlate with the occurrence of a linear CH_4 concentrations profile.

The decline of MMO in summer and October 2002 and September 2003 along with a decrease of gas permeability following weeks of high methanotrophic activity might have been attributed to production of exopolymeric substances (EPS) by methane oxidisers. EPS may act as a microscale diffusion barrier coating the base biofilm (Hilger *et al.*, 2000b) and as a macroscale diffusion barrier preventing gas transport in the soil (Wilshusen *et al.*, 2004). A reddish-brown band, which was related to EPS formation according to Wilshusen *et al.* (2004), was found in W + LG. Coating of the roots with EPS indicated improved living conditions for methanotrophs in the vicinity of roots possibly owing to better O_2 availability, as willows are known to transfer O_2 into the rhizosphere (Marschner, 2002; Maurice *et al.*, 1999).

4.2. The effect of landfill leachate irrigation

Small amounts of landfill leachate irrigation (<150 mm) promoted microbial methane oxidation (MMO) due to good plant performance, low soil moisture content and hence increased aeration. O_2 transport into the soil was inhibited after the moisture content increased and the soil structure deteriorated under long term landfill leachate irrigation. Methanotrophs, especially the more efficient type I, are sensitive to low O_2 concentrations (Mancinelli, 1995). Consequently MMO and soil redox potential decreased in LL + LG, which caused high H_2S production and sometimes methanogenesis. In addition, landfill leachate application added organic and reduced inorganic substances to the soil. Oxidation of these compounds also consumed O_2. Unlike respiration, MMO activity recovered fully in spring 2003 corresponding to improved plant performance. Our results are clearly in contrast to the positive effect of landfill leachate irrigation on MMO reported by Maurice *et al.* (1999). However, the landfill leachate used in that study was more diluted and tree growth was promoted.

Besides decreased O_2 availability, the high electric conductivity (EC), high concentrations of NH_4, Na, Cl and to a lower extent B in the landfill leachate might have inhibited MMO. Mancinelli (1995) and Boeckx and Van Cleemput (2000) reported cooxidation of NH_4 and consequently competition with MMO. Controversially, in later studies done by De Visscher and Van Cleemput (2003) NH_4 has been described as stimulating MMO by adding nutrients. In our study the importance of the presence of NH_4 was probably overruled by high amounts of Na and Cl. Gebert *et al.* (2003) reported decreased methane consumption at EC values >6 mS cm^{-1}. In their study, MMO declined by a factor of 3, but adaptation of the methanotrophic community to high salt concentrations occurred. De Visscher and Can Cleemput (2003) found an inhibition of the MMO in soil amended with 126 mg kg^{-1} Cl. In our experiment, about 70 g Cl was added with the landfill leachate until MMO diminished. Special emphasis should be given to Na, as Na is not only known to be toxic for microorganisms, but also disperses soil aggregates and hence diminishes water conductivity and gas permeability (Scheffer and Schachtschabel, 1998). Little is known about the toxicity of B on methanotrophs, but added B concentrations are above threshold values reported for irrigation water (Adriano, 2001).

In contrast to MMO, long term landfill leachate irrigation also showed decreased respiration at times of good aeration, as it was observed from non landfill gas fumigated lysimeters, indicating a direct negative effect of landfill leachate constituents on soil microorganisms. In W, respiration sometimes seamed to suffer from lack of water e.g. in summer 2003, which could not be verified for MMO.

4.3. Formation of nitrous oxide

Reduction of greenhouse gases through MMO might be partly counteracted if N_2O production is enhanced at the same time. Boeckx and Van Cleemput (2000) reported a reverse relationship between N_2O emissions and MMO and Bogner *et al.* (1999) found N_2O production in a zone below the optimum CH_4 oxidation depth. In contrast, no or only little N_2O emissions were measured at landfill sites in Finland (Einola *et al.*, 2003) and Börjesson *et al.* (1998) stated that both N_2O and N_2 production were only slightly influenced by the extent of CH_4 oxidation, but positively related to soil moisture. Accordingly, our results indicated no or little N_2O formation under landfill gas fumigation or if N_2O was produced, it was probably reduced to N_2.

High N_2O production (up to 85 $\mu L\,L^{-1}$) occurred in LL possibly owing to the high soil moisture content and limited aeration. Contrary to fumigated soil, N_2O concentrations decreased in the upper horizons. Consequently, N_2O emissions might be lower than soil gas concentrations. Long term landfill leachate application increased N_2O production owing to increasing input of N and decreasing redox potential. Lee *et al.* (2002) reported low N_2O generation by nitrification in landfill leachate treated soil, therefore, nitrification of NH_4 added with the landfill leachate in the upper 0.1 m of the soil, as monitored by Ankers and Ruegg (1991), followed by denitrification of nitrate at lower depths might have been the primary source of N_2O in our experiment.

5. Conclusions

Independently of irrigation treatment, MMO started readily after landfill gas addition and ceased in winter, at temperatures below 0 °C. The visual methane oxidation depth was at 0.10–0.15 m depth, but CH_4 concentrations was also decreased below that depth during periods of high MMO. High MMO rates were often followed by decreased MMO probably due to EPS formation.

The availability of O_2 strongly influenced MMO and N_2O formation in a compost - gravel substrate. Under landfill leachate application, gas permeability and hence O_2 availability diminished, mainly because the soil water content increased as plants deteriorated and transpiration decreased. In addition, microbial activity (especially respiration) might have been directly affected by toxic compounds in the landfill leachate, primarily by high NaCl concentrations. Minor N_2O concentrations were detected under landfill leachate fumigation owing to the low soil redox potential. In contrast, landfill leachate irrigation in non fumigated soil led to a substantial formation of N_2O with increasing amounts of applied leachate. In conclusion, it is suggested that landfill leachate irrigation must be dosed at low amounts (<200 mm) if adverse effects on greenhouse gas emissions (CH_4 and N_2O) are to be avoided.

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