



Emission of Nitrous Oxide and other Trace Gases during Composting of Grass and Green Waste*

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The emission of trace gases during composting of green waste from land maintenance (fresh cuttings of mixed herbage from fallow land) were studied. Concentrations of nitrous oxide and other trace gases were measured in experimental compost heaps by means of an infrared gas analyser and a high-resolution FT-IR spectrometer. It was verified that the maintenance of aerobic conditions is essential to keep emissions of methane and nitrous oxide at a low rate. Estimates made using a simple air transport model, indicated that the emission of nitrous oxide during composting of green waste from land maintenance was about 0.5% of the total nitrogen content of the initial material. Carbon monoxide was also detected in the compost air and its emission during biodegradation corresponded to about 0.04% of the initial carbon content of green waste.

For a detailed evaluation of the findings during field experiments, laboratory apparatus for measuring the composition of compost air was developed. The measurements were performed at a temperature of 35°C and at different ventilation rates. Moistened plant material (dry cuttings of mixed herbage from fallow land) amended with lime saltpetre was used as compost substrate. At a ventilation rate of 100 cm³/min of air per kg of substrate, the maximum emission rate of nitrous oxide was 2.2 mg/h per kg of substrate. The maximum emission rate increased to 13.3 mg/h kg, when the ventilation rate was lowered to 20 cm³/min kg. The emission rate of carbon monoxide was about 40 µg/h per kg of substrate at the low ventilation rate and rose above 200 µg/h kg at the higher ventilation rate.

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1. Introduction

Biomass from managed land and grass land, which cannot be used as fodder, can serve as green manure or should be a source for other uses. One possibility is composting, since this is considered to be an environmentally friendly procedure. Emissions to ground water during composting can be avoided by appropriately designed composting facilities, but knowledge about emissions of trace gases into the atmosphere during composting is not sufficient at present.¹

Carbon dioxide, methane and nitrous oxide are the main components of agricultural greenhouse gas emissions. The generation of carbon dioxide during the transformation of biomass does not contribute to the global greenhouse effect because of the assimilation of CO₂ by plants. Methane may be generated by microorganisms when biomass is stored under anaerobic conditions. Ammonia also contributes to the greenhouse effect indirectly through the following processes. Deposition of ammonia, followed by nitrification, enriches the soil with nitrate. Through denitrification, nitrate is progressively reduced to pure nitrogen gas. Small amounts of nitrous oxide volatilize during nitrification and denitrification.² This volatilization of N₂O is an undesirable emission during the biodegradation of biomass.

Since it is recommended that grass and other plant materials with a high C/N ratio be amended with nitrogen fertilizers in order to achieve good rotting (see e.g. Ref. 3), the influence of such additives on emission of nitrous oxide should be known. Nitrous oxide has been found during composting (see e.g. Refs 4–8), and emissions of nitrous oxide during composting of solid cattle manure (dry matter content between 14 and 23.6%) have been studied on a laboratory scale by gas chromatographic measurements.⁹ The emissions measured were between 10 and 100 ml gas per kg of dry matter.⁹ This corresponds to about 0.1–1% of the total nitrogen content, if an average nitrogen content of 0.5% is taken for

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solid cattle manure.¹⁰ Emission values of 10 mg (equivalent to 5 ml) nitrous oxide per kg of compost mixture (grass cuttings with leaves and potato peelings) have been reported in preliminary studies⁶ under laboratory conditions using a thermally insulated, closed compost container on a small scale (20 l). During composting of farm waste (bedding with horse manure and poultry manure),⁸ 2.19% of the total nitrogen, was emitted in terms of N_2O-N within 60 d. In the case of yard waste (mostly grass clippings with a batch of fallen leaves and no nitrogen addition), only 1.18% of the total nitrogen was released as N_2O-N within the same period. The N_2O fluxes were measured using a gas flux chamber and a gas chromatograph.⁸

At the beginning of the study reported in this paper, field measurements of trace gas concentrations were made in compost heaps, where the aim of composting was to improve rotting of grass by amending with soil or manure.⁵ An increase in the concentration of nitrous oxide in the heaps was found with manure as additive. A high variability of concentrations of methane and nitrous oxide, was observed in compost heaps of municipal green waste (grass cuttings, leaves, bush and tree trimmings), where the aim was to improve the aeration by horizontal channels.^{11,12} The present work reports systematic measurements of emissions of trace gases into the atmosphere from field scale compost heaps (15 t of biomass) to evaluate the environmental impacts during composting of green waste from land maintenance (i.e. fresh cuttings of mixed herbage, with no amendments), when simple compost procedures are applied.

In addition to this field study, in order to study the influence of amendments and of aeration on the emission of trace gases in more detail, a laboratory experiment was devised, such that the ventilation rate and temperature of a container, filled with substrate and amendments, could be controlled. In contrast to other works, where the object was to simulate composting¹³ or to model the natural composting process,¹⁴ the object of this experiment was to measure emissions at constant temperatures and at constant ventilation rates.

2. Materials and methods

2.1. Field-scale experiments

2.1.1. Measurements of compost air by a multi-gas monitor

In the first experiment (July–September 1994), the compost was contained in wooden boxes with dimensions of 4 m × 2 m × 1.75 m high. The boxes had been filled with alternate layers of grass cuttings (thickness 25 cm) and soil (thickness 5 cm) in one box and layers of

grass cuttings (thickness 25 cm) and layers of solid manure (thickness 15 cm) in the other box. Three polyethylene pipes were horizontally embedded at heights of 0.75, 1.20 and 1.60 m (Fig. 1a). The effect of different shaped channels on aeration of a compost heap was studied in a second experiment (August 1994). The heat insulated compost box was 7 m × 4 m × 2 m high and was filled with grass cuttings, leaves, bush and tree trimmings. Four polyethylene pipes were pushed vertically into the compost material in line with the aeration channel (Fig. 1b).

In all compost experiments, the pipes were open at the inside end and closed by a rubber stopper at the outside end. For each measurement, about 800 cm³ of compost air was drawn from the central part of the heap through the polyethylene pipe via a stainless-steel cooling coil (diameter 5 mm, length 160 cm) and a Teflon pipe (inside diameter 3 mm, total length 10 m) to the multi-gas monitor (Brüel & Kjaer 1302). Since moisture affects the performance of the multi-gas monitor, the compost air was cooled to the measuring temperature (room temperature) to avoid water condensation in the measuring chamber of the monitor. Calculations indicated that about 50 mg of water would condense at the surface of the cooling coil during one measurement. After each measurement, the coil was dried in preparation for the next measurement. Due to this condensation, measurements of ammonia were affected because of the high solubility of ammonia in water. In addition, preliminary studies showed that ammonia had a high adsorption rate at different kinds of surfaces. Therefore, the measured concentrations were less than the real ammonia concentrations. Especially at low concentrations of ammonia

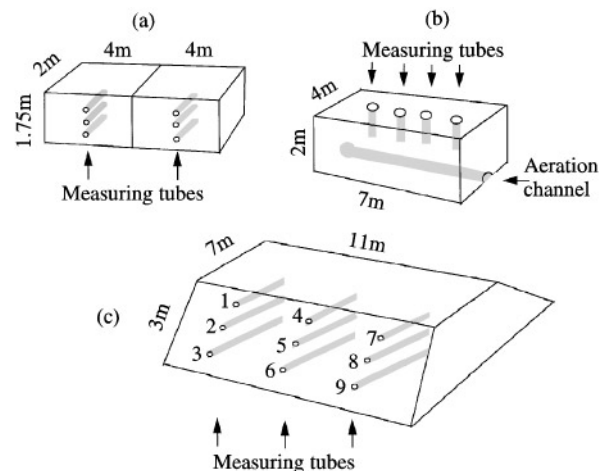


Fig. 1. Scheme of compost heaps with measuring tubes: (a) wooden compost boxes, filled with layers of grass and soil (left) and layers of grass and manure (right); (b) thermally insulated compost box, filled with municipal green waste; (c) trapezoidal compost heap of grass cuttings from land maintenance on bare soil

(below 10 p.p.m.), erroneous readings by up to a factor of two could not be excluded, whereas at high concentrations (above 100 p.p.m.), laboratory studies suggested that this error dropped below 30%.

The multi-gas monitor had a sensitivity of about 0.05 p.p.m. for N_2O , 0.2 p.p.m. for NH_3 , 0.3 p.p.m. for CH_4 and 3 p.p.m. for CO_2 . Because of the infrared filter principle in combination with a photoacoustic detector, it was possible to measure values over four decades. The disadvantage of the filter principle is that wrong measurements can be made when signals are superposed with unknown components having infrared absorption lines in the filter regions (so-called cross-sensitivity), as is the case with organic acids, which are generated sometimes in low concentrations during the initial stage of decomposition of biomass. In such a case, the CH_4 measurements would be incorrect. With air having high contents of CO_2 and CH_4 , trace concentrations of NH_3 and N_2O could not be determined accurately with the multi-gas monitor. The reason was that at high concentrations of CO_2 (>5%) and of CH_4 (>500 p.p.m.), additional absorption lines occurred in the infrared-filter regions resulting in incorrect measurements. Therefore, to achieve sufficient accuracy and reliability in the analysis of compost air, a high-resolution Fourier transform infrared (FT-IR) spectrometer was used for the third field experiment. Since the monitor was a totally portable instrument, while the FT-IR measurements could be performed in the laboratory only, the monitor served as back-up and control system. Measurements with the multi-gas monitor made it possible to estimate errors caused by transport and storage of compost air in gas bags.

2.1.2. Measurements of compost air by an FT-IR spectrometer

For the third field experiment (September 1995–March 1996), a trapezoidal compost heap (Fig. 1c) was made on bare soil with initial dimensions of 11 m × 7 m × 3 m high. The initial mass of the material (green waste from land maintenance) was 14.8 ± 0.2 t. The mass was measured by means of a trailer and two axle balances with a resolution of 20 kg and an accuracy of 50 kg at maximum load of 25 t. The mass was determined at the outset, and at turning over after 32 and 70 d (weighing on the trailer and setting at a new place) and at the end of composting after 194 d. The carbon and nitrogen content of the material were analysed by means of an element analyser (VARIO EL, Elementaranalyse GmbH Hanau) at the same times. On each occasion, three mixed samples were taken out of the compost material. The initial C/N ratio was 27 ± 2 (carbon content $40.3 \pm 0.7\%$, nitrogen content $1.48 \pm 0.06\%$, on a dry mass basis). The initial carbon mass of 4.3 t

decreased to 0.73 t after rotting and the C/N ratio to 13 in the course of composting.

The gas measurements started one day after setting up the compost heap. On gas measuring days, the compost air was pumped by a small electric diaphragm compressor (3 l/min) through polyethylene pipes at nine different measuring points with initial heights 0.7, 1.5, and 2.3 m (Fig. 1c) into evacuated polyethylene gas bags each of 10 l volume. Three gas bags were filled with natural air (before, during and at the end of each field measurement day). For calculation of emission rates, concentration differences between the compost and outside air were used. There was no influence of nearby sources of trace gases, because the compost heap was set in the countryside with distances of more than 3 km to buildings and streets.

The trace-gas analysis was made by means of a high-resolution FT-IR spectrometer (Perkin-Elmer, System 2000, resolution 0.2 cm^{-1}). To achieve sufficient sensitivity for quantitative FT-IR detection, a heated measuring cell (80°C , measuring volume of 5 l) with an optical path length of 20 m was utilized in combination with an HgCdTe-detector (MCT). The measuring cell and all pipes were evacuated to 0.3 mbar before each measurement. The filled gas bags were warmed up at 60°C for about one hour, in order to transform condensed water into the gaseous phase. Then the measuring cell was filled with compost air. The high resolution of the FT-IR spectra was essential to measure the wide span of concentrations of nitrous oxide, ammonia, carbon monoxide and methane in air that had high levels of humidity and carbon dioxide, as was the air from the compost. The detection limit was better than 0.05 p.p.m. for the gases methane, nitrous oxide and carbon monoxide and 0.2 p.p.m. for ammonia. The systematic error in the determination of concentrations was about 20% for these gases. It depended on the type of gas and the concentration span to be evaluated (line positions and changes in line intensities), on calibration and spectrum evaluation, and on measurement procedure. Errors, caused by electronic noise, gas handling and other stochastic sources were less than 5%. Because of the high adsorption potential of ammonia, a dynamic calibration was necessary. This meant that all conditions, which influenced the adsorption such as time of transport, storage and measurement, had to be equal. Also the type and initial chemical state of all materials such as pipes and bags had to be identical as far as possible (e.g. absence of NH_3) during the measurement and calibration procedures.

All gases with absorption lines in the infrared region contribute to the infrared spectrum with specific lines and intensities according to their concentrations. An unexpected result was that comparatively high concentrations of carbon monoxide of up to 100 p.p.m. were

found in compost air, when the spectra were analysed. Therefore, carbon monoxide was also included in the evaluation of the measurements.

2.1.3. Estimation of the emission rates by a transport model in connection with the carbon balance

In order to derive values for the total emissions on the basis of the measured concentrations, a simple transport model was chosen. As long as the mass transport is flow-controlled (air speed is higher than the mean transport speed caused by the diffusion of the molecules), the release rate $\Delta m / \Delta t$ of the component m is the product of the concentration $[m]$ and of the volume flow rate ($\Delta V / \Delta t$),

$$\Delta m / \Delta t = [m] (\Delta V / \Delta t) \quad (1)$$

In a compost heap, convection currents are caused by temperature gradients diffusion of gases is caused by concentration gradient and air flow is caused by air pressure difference due to wind. There is no uniform flow rate and the flow distribution is not known. An approximation for the gaseous mass transport can be derived, if the gas generation inside the heap is considered to be uniformly distributed. The decrease of the total carbon content per time in the compost material is equal to the carbon mass flow of the carbon containing gases ($\Delta C / \Delta t$) of the air leaving the compost, if carbon losses in terms of liquid components can be neglected. Then the mean concentration of carbon containing gases inside the heap $[C]$ is related to the gaseous carbon emission rate ($\Delta C / \Delta t$) of the heap.

$$\Delta C / \Delta t = [C] F(\Delta V / \Delta t) \quad (2)$$

The transport function $F(\Delta V / \Delta t)$ is determined by the structure of the heap and does not depend on the type of molecules, if losses inside the heap due to dissolving or other sinks can be neglected. In the case of mass transport by diffusion, the coefficient of diffusion must be included. The diffusion constants of carbon dioxide and nitrous oxide are equal. With the assumption that volatilized nitrous oxide, generated in the liquid phase by microorganisms, cannot be trapped or dissolved as part of the compost air, the emission rate of nitrous oxide $\Delta N / \Delta t$ can be written as

$$\Delta N / \Delta t = [N] F(\Delta V / \Delta t) \quad (3)$$

with $[N]$ the measured concentration of nitrous oxide in the heap. Using Eqn (2), the emission rate of nitrous oxide can be estimated by

$$\Delta N / \Delta t = [N] (\Delta C / \Delta t) / [C] \quad (4)$$

According to Fick's law, the diffuse mass transport is directly proportional to the concentration gradient. As-

suming a constant concentration in the outside air and steady-state conditions for the flow inside, we find as approximation for the diffusion-controlled gaseous mass transport:

$$\Delta m / \Delta t = D_m / D_{CO_2} [m] (\Delta C / \Delta t) / [C] \quad (5)$$

with D_m the diffusion coefficient of the component m and D_{CO_2} the diffusion coefficient of carbon dioxide, the main component of gaseous carbon transport.

The estimated relative errors in the emission rates are believed to be lower than 50%. They arise from weighing of the compost during turning over (<2%), from the determination of the carbon content (<3%), from the FT-IR measurement (<25%) and from the approximations of the transport model. One main source of error of the model is the inhomogenous generation and transport of CO_2 , the reference gas in Eqns (4) and (5). Other sources of error are gas sinks inside the heap (solution, adsorption or consumption by microorganism) and disproportional changes of concentrations of N_2O , NH_4 , CO and CH_4 in relation to the concentration of CO_2 between the measuring days. The homogeneity of CO_2 generation over the heap can be estimated by the variation over the nine tubes for each measuring day. The mean coefficient of variation over all measuring days was 19%. If sinks and disproportional changes in concentrations are neglected between the measuring days, then an error of about 20% can be assumed for the transport model.

In the case of ammonia, an additional error could be caused by the solubility of ammonia in water. Bearing in mind that we are considering trace gases of a complex biological process and that the first aim was to get the right order of emissions, especially for the direct greenhouse gases nitrous oxide and methane, these errors in field measurements were considered acceptable.

2.2. Laboratory-scale experiments

The aim of the laboratory studies was to determine the percentage of nitrate-nitrogen, emitted as nitrous oxide, and its dependence on ventilation at a given temperature. Additionally, it was necessary to check the generation of carbon monoxide under laboratory conditions. The main factors which influence the emission of nitrous oxide are temperature, moisture content, pH value, partial pressure of oxygen and C/N ratio.^{2,15} The maximum of the nitrification rate is observed between 30 and 35°C.^{15,16} The denitrification rate decreases with decreasing temperatures, but the ratio of N_2O/N_2 increases.¹⁵ The C and N-mineralization accelerates with increasing temperatures.¹⁶ Maximal decomposition of urea by nitrification and denitrification was observed between 30 and 35°C.¹⁷

Therefore, a temperature of 35°C was chosen for the first laboratory measurements.

Laboratory apparatus was designed as shown schematically in Fig. 2. The rectangular substrate container made of aluminium had a length of 30 cm, a width of 20 cm and a height of 60 cm. The substrate was placed on a removable grid inside the container. This grid permitted uniform aeration from the bottom of the container.

Moistened strawy herbage (dry cuttings of mixed herbage from fallow land) with a moisture content of 70% w.b. was utilized as substrate. Measurements were performed with substrate amended by lime saltpetre, and for comparison, with substrate without additives. The carbon content of the substrate was $43.6 \pm 0.6\%$, on a dry mass basis and the C/N ratio of the substrate was in the range 58 ± 4 . Lime saltpetre as fertilizer is a mixture of calcium oxide and calcium nitrate with a content of nitrate nitrogen ($\text{NO}_3\text{-N}$) of 15.5%. The total nitrogen content of the initial substrate mixture related to dry mass was $1.6 \pm 0.1\%$, of which $0.86 \pm 0.05\%$ was nitrate-nitrogen and $0.74 \pm 0.05\%$ organic nitrogen. The errors here were caused by the inhomogeneity of the samples. The element analysis itself had a higher accuracy and contributed less than 0.01% (related to dry mass) to the error of nitrogen determination. The C/N ratio of the initial mixtures was 27 ± 2 . During the comparatively short times of the experiments (duration of two to three weeks), there was only a slight drop in the C/N ratio to about 25 ± 3 . The pH values of the mixtures were in the range of 6.5 ± 0.3 after the start. At the end of the experiments, the pH values had increased to 7.0 ± 0.2 .

A cryo-thermostat (HAAKE F3-CH, heating power of 2000 W, cooling power of 380 W at 20°C, external temperature sensor) kept the bath temperature at a constant level. The temperature in the centre of the container did not exceed 38.5°C at a bath temperature of 35°C (Fig. 3). The inside temperature was measured by a temperature

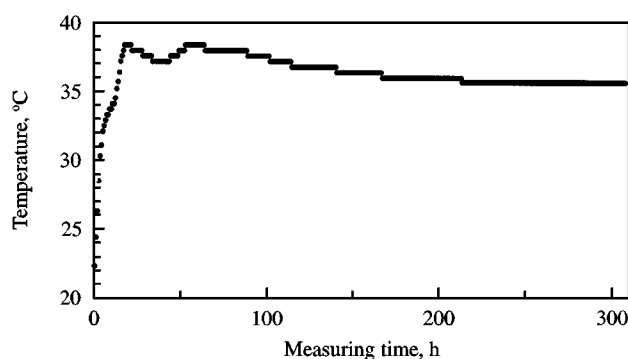


Fig. 3. Temperature versus time dependence in the centre of the container with an outside bath temperature of 35°C and an air ventilation rate of 50 cm³/min per kg of substrate (mixed herbage with moisture content of 70% w.b., 1.7% lime saltpetre)

data collector (TINYTALK-TEMP, Orion Components Ltd, temperature resolution of 0.4°C).

The ventilation rate could be varied between 10 and 1000 cm³/min. A peristaltic pump was used for flow rates up to 200 cm³/min. This type of gas pump ensured a constant volume flow independent of small fluctuations in the air resistance of the total system. The fluctuations in the resistance to air flow arose from the decomposition of the biomass and from the multiplication of microorganisms during the experiment. The changes of the aeration rate were less than 5% in the course of two to three weeks. At higher flow rates, the pipe in the peristaltic pump was not reliable and was destroyed after several days of pumping. Therefore, a critical orifice was used for the control of higher flow rates. Changes in the water level at the orifice had to be corrected daily to keep the changes to less than 10% of the chosen ventilation rate. The effect of condensation was minimized by utilizing a perpendicular pipe arrangement after the substrate container in such a way that the condensed liquid dropped back into the container. The air from the container had to pass a flow meter and filters, before entering the spectrometer. The filters were necessary to avoid dust layers on the mirrors of the gas cell of the spectrometer. The total air volume was registered by a wet-type drum gas meter (Ritter TG 3) after the FT-IR analysis. The water level of the gas meter was checked several times. Changes due to evaporation or condensation were not measurable, since the air leaving the container had saturated humidity.

The absolute emission rates were calculated according to Eqn (1) on the basis of differences between mean gas concentration in laboratory air (measured three times a week) and the gas concentrations of air leaving the container. The average values of the gas-meter readings between the measurements of compost air (three times a day) were used for the determination of the flow rate.

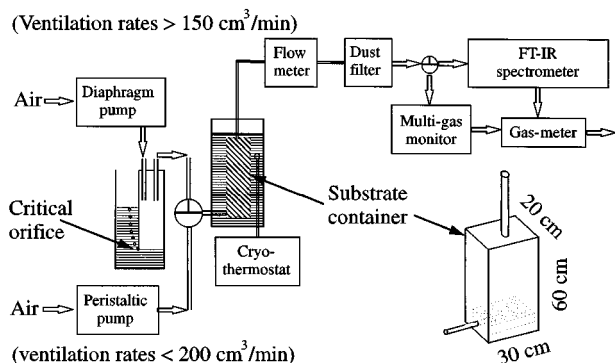


Fig. 2. Scheme of gas measurement for laboratory-scale determination of emission rates

The multi-gas monitor had the function of backing the data collection, in cases where the on-line FT-IR measurements had to be interrupted for air control or other measurements.

3. Results and discussion

3.1. Field-scale experiments

3.1.1. Application of the multi-gas monitor for the study of quality of composting

In the first experiment of 1994, measurements of trace-gas concentrations served as qualitative indicators of the emissions due to the treatments⁵ and also for control of aerobic conditions inside the heap. Anoxic conditions in the heap caused bad odours and methane emissions. Results are shown in *Fig. 4*. The peak concentrations of

ammonia and nitrous oxide generation were observed at the first measurement just after establishing the heap (14/07/94), whereas concentrations increased in the course of several days after turning over (19/09/94). The concentrations were higher in the treatment containing additional nitrogen (manure). These findings were interpreted as a result of nitrification followed by denitrification, and the availability of free nitrate after setting up the heap due to the higher nitrogen content of the grass manure mixture. In the grass/soil mixture, methane generation increased with time. In this case, the soil layers had compressed parts of the grass layers in the compost box resulting in anaerobic conditions. Since undesirable compost conditions can be detected by monitoring of methane concentrations in compost air it was concluded that soil was not a suitable amendment for composting of grass.

In the second compost experiment with heat insulated walls, measurements of methane and other gases with the multi-gas monitor were used to indicate the effect of aerating by channels (*Fig. 1b*). Since methane had been generated (*Fig. 5*), it was obvious that aeration by channels was not sufficient during the measurement period. In addition, variations in the nitrous oxide concentration were observed, which followed the variations in methane concentration. There are two possible interpretations. Different thinning of gas concentrations by variations of airflow and air convection inside the heap could have been one reason for synchronization effects. Alternatively, insufficient aeration may have resulted in anoxic regions in the heap where methane was generated, while in other regions with a low partial pressure of oxygen, the denitrification may have been enhanced. No distinction could be made in this experiment because of the arrangement of the measuring tubes.

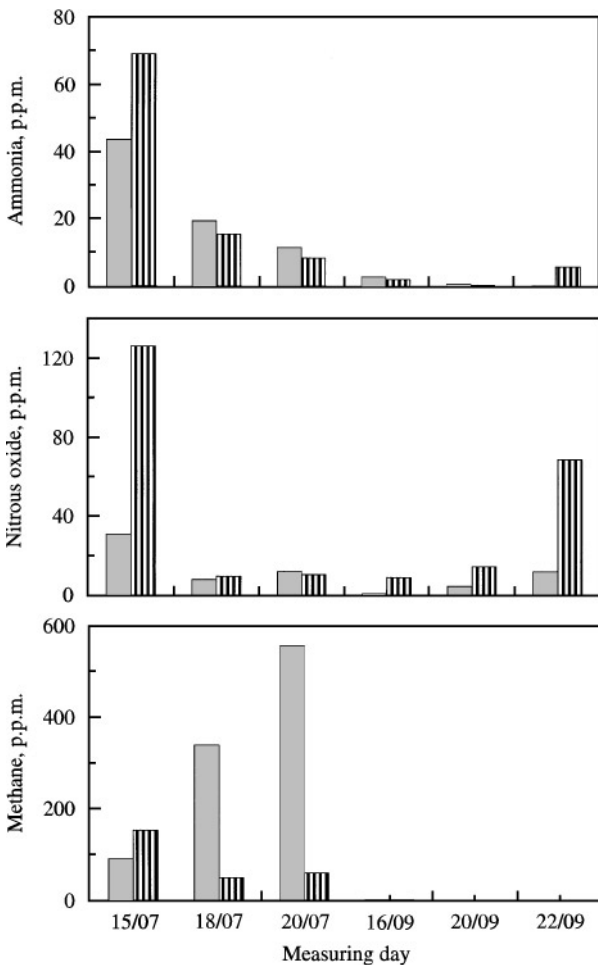


Fig. 4. Gas concentrations during composting of grass clippings with soil (■) and manure (▨) as amendments (set on 14/07/94, turnedover on 19/09/94)

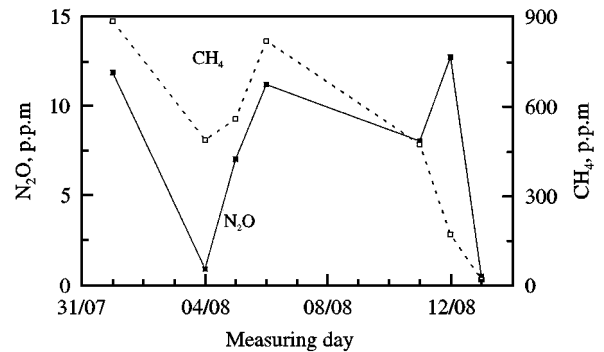


Fig. 5. Mean concentrations of nitrous oxide N₂O and methane CH₄ during composting of municipal green waste (grass cuttings, green leaves, bush and tree trimmings)

3.1.2. Application of FT-IR spectroscopy for emission measurements during composting

As shown in Fig. 6, methane was generated in some parts of the heap due to insufficient air exchange in the course of the composting process. After the first turning of the heap, the compost material collapsed, especially in the central part of the heap. Connected with this, the generation of methane reached values of the order of 1% by volume (measuring tubes 5 and 6; Fig. 6). The increased methane levels at the upper tubes 1 and 7, respectively, were probably a result of convection and diffusion from the central part, since at the lower left and right tubes (Fig. 1c) reduced methane concentrations were observed.

The generation of nitrous oxide reached a maximum in the central parts (measured at tubes 5 and 6 on 01/11; Fig. 7) after turning on 29/10. Thereafter, an increased level of the concentration of nitrous oxide was measured at all tubes for a period of five weeks, except at tubes 5 and 6. Since methane is the indicator for anaerobic degradation of biomass, it was evident that nitrous oxide had only been produced by microorganisms under aerobic conditions. On the other hand, in commercial practice it is argued that no nitrous oxide will be generated, if aeration of the compost is sufficient. Therefore, it was decided to examine the generation of nitrous oxide and its dependence on the ventilation rate through laboratory studies.

The average of the concentrations measured at the nine tubes (Fig. 8) was utilized for the determination of the total emissions in connection with weighing and carbon and nitrogen analysis of the rotting compost material. During composting, 3.6 t of carbon and 102 kg of nitrogen were released, and 90% of these emissions took place

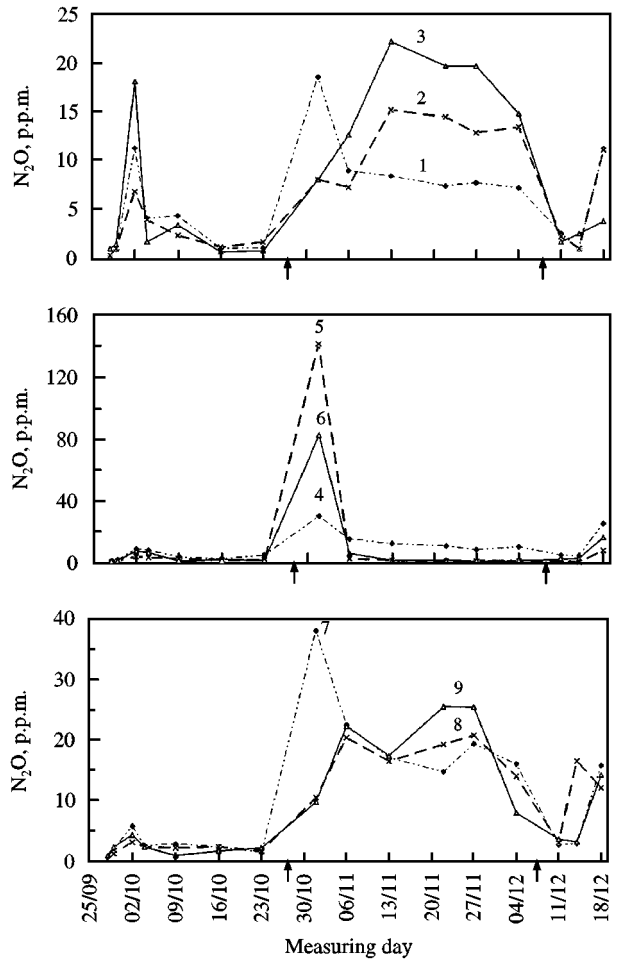


Fig. 7. Concentration of nitrous oxide at the different measuring tubes. The positions of the measuring tubes are shown in Fig. 1c. Arrows indicate dates of turning over of the compost

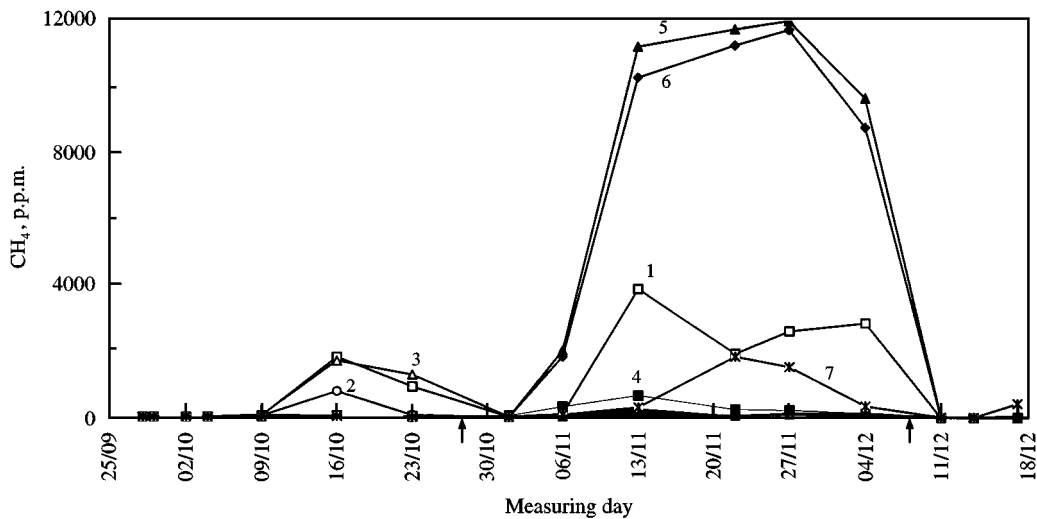


Fig. 6. Concentration of methane during composting. The positions of the measuring tubes are shown in Fig. 1c. Tubes 8 and 9 are not indicated. They are too close to the bottom line (less than 100 p.p.m.). Arrows indicate dates of turning over of the compost

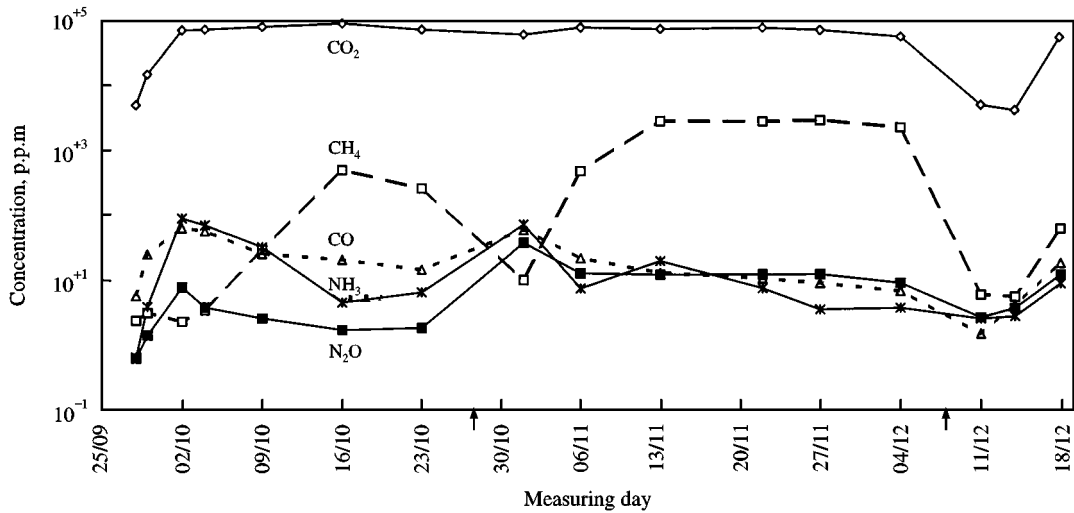


Fig. 8. Mean concentrations (logarithmic scale) of carbon dioxide CO_2 , methane CH_4 , carbon monoxide CO , ammonia NH_3 , and nitrous oxide N_2O in compost air during composting of green waste from landscape care. Arrows indicate dates of turning over of the compost

within the first three months. The emissions of gases were calculated using Eqns (4) and (5) on the basis of the carbon balance.

About 80% of the initial carbon mass was transformed by microorganisms into carbon dioxide. Methane contributed in the range of 1.5–2% of the carbon reduction and carbon monoxide had a share of about 0.04%. In the case of nitrogen, 0.8 kg nitrogen escaped as N_2O -N and 1.9 kg as NH_3 -N (Table 1). This means that about 0.5% of the total nitrogen content of the biomass was converted into nitrous oxide during the whole composting process. Similar values (0.5–0.8%) were observed in closed chamber measurements of large compost heaps of municipal green waste.¹⁸

It was surprising that concentrations of carbon monoxide up to 100 p.p.m. were measured, since CO is produced usually by combustion and not by composting. Carbon monoxide contributes only negligibly to the greenhouse effect of the atmosphere, but this toxic component with an atmospheric half-life of a few months is involved in the chemistry of the tropospheric ozone.^{19,20} The normal atmospheric concentration of CO is in the range 0.01–1 p.p.m. and can reach up to 100 p.p.m. during rush hours at some road crossings.²¹ No information concerning emission rates of CO during composting could be found in the literature.

Because of this unexpected result, all FT-IR spectra were carefully checked. Since only undisturbed IR absorption lines were used for the evaluation, the finding was confirmed. There are different biological sources of CO such as a few plants,²² certain algae,²³ and special microorganisms.^{24–26} The first assumption was that

Table 1
Emissions during composting of green waste from land maintenance

Component	Emission, kg	Percentage, % (related to initial element mass)
N_2O -N	0.8	0.5
NH_3 -N	1.9	1.2
CO_2 -C	3500	81
CH_4 -C	75	1.7
CO -C	1.7	0.04

The initial mass of the compost heap was 14 800 kg with a carbon content of 4300 kg and a nitrogen content of 158 kg.

a chemical reduction might have taken place, as the majority of CO production in soil seems to be caused by chemical processes.²⁶ More detailed results were therefore expected from the laboratory experiments.

3.2. Laboratory-scale experiments

The basic development of the emissions was similar at all three aeration rates tested (Fig. 9). At first, a peak of carbon monoxide appeared some hours after the start of the experiment. The maximum carbon dioxide concentration, with levels up to 20% by volume, was observed in the air leaving the substrate container after about 1 d. Then the generation of nitrous oxide increased. The induction period to reach the maximum was 4–10 d. The

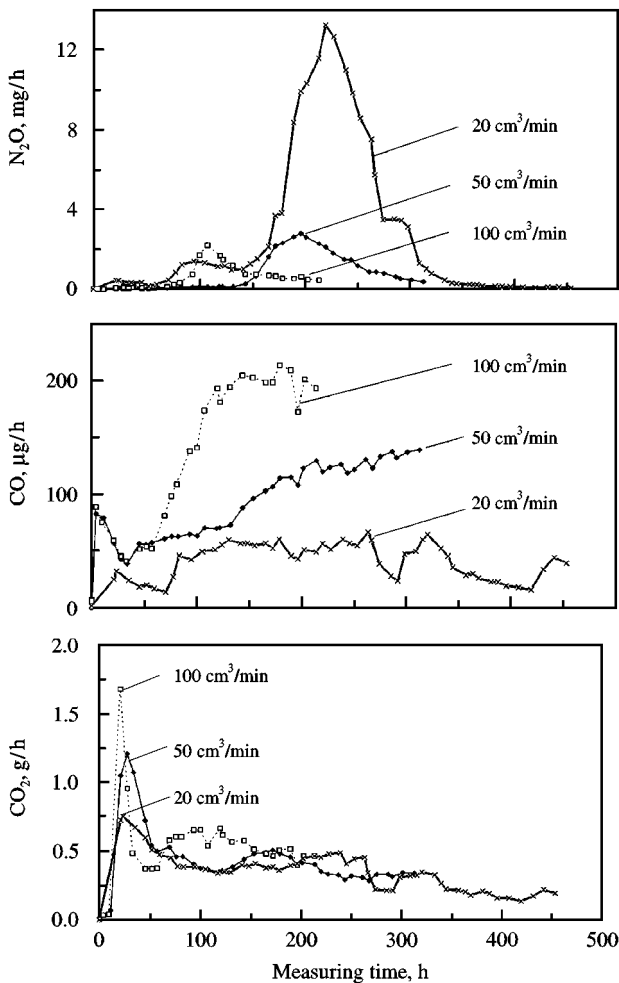


Fig. 9. Emission rates of nitrous oxide N_2O , carbon monoxide CO and carbon dioxide CO_2 per kg of substrate mass at a temperature of $35^\circ C$ and its dependence on ventilation rate per kg of substrate (mixed herbage with moisture content of 70% w.b., 1.7% lime saltpetre)

maximum concentration was in the range of 200 p.p.m. with a ventilation rate of $100\text{ cm}^3/\text{min}$ per kg of substrate. It rose to values of more than 4000 p.p.m. at a ventilation rate of $20\text{ cm}^3/\text{min}$ per kg of substrate. Then a decrease over a period of about 5 d was registered. When the concentration of nitrous oxide dropped below 40 p.p.m., the experiment was stopped. For comparison, the concentration in fresh air is about 0.3 p.p.m..

At the low aeration rate of $20\text{ cm}^3/\text{min}$ per kg of substrate mass, the maximum emission rate of N_2O was 13 mg/h per kg of substrate mass (Fig. 9) and the mean emission rate 2.9 mg/h kg. With increasing ventilation rate the average emission rate of nitrous oxide decreased. A ventilation of $50\text{ cm}^3/\text{min}$ kg gave an average of 0.85 mg/h kg. A further reduction of the mean emission rate to 0.55 mg/h kg was measured at the increased ventilation rate of $100\text{ cm}^3/\text{min}$ kg.

No emission of N_2O was observed in measurements without additives (pure herbage with water), although high carbon dioxide concentrations confirmed the biodegradation of the substrate. Therefore, the generation of nitrous oxide as a result of denitrification was caused by the availability of nitrate ions. With reference to the total nitrogen content, 0.4% was emitted as N_2O-N at a ventilation rate of $100\text{ cm}^3/\text{min}$ kg of substrate within the measuring period. The emission increased to 0.7% at an aeration rate of $50\text{ cm}^3/\text{min}$ kg and up to 3.2% at an aeration of $20\text{ cm}^3/\text{min}$ kg, respectively.

It is apparent from Fig. 9 that the emissions of nitrous oxide started after the peak in carbon dioxide generation had passed. Due to the activity of oxygen-consuming micro-organisms nearly all oxygen was converted into carbon dioxide. It appeared that, at this borderline between the aerobic and anaerobic state, specialized bacteria could utilize the denitrification chain as an oxygen source and, generate nitrous oxide. This was supported by the measured dependency on aeration, since at the lowest ventilation rate, the highest emission of nitrous oxide was measured.

Although the source of emission of carbon monoxide could not be identified in this experiment, the measured concentration dependencies on time and ventilation rate indicated a biological origin and not a chemical reduction of carbon dioxide. After about 50 h, the concentration of CO_2 decreased nearly continuously. Then an approximately equal level of CO_2 emission rates was measured for all ventilation rates. In the case of carbon monoxide, generation rates increased with time and with rising ventilation rate. The minimum CO generation corresponded to the time period just after the maximum CO_2 generation. Because of these dependencies, it is believed that the most probable assumption was that microorganisms were the source of carbon monoxide. Further studies are necessary to support this thesis. If this biogenic source of carbon monoxide can be verified, microorganisms involved in composting and decomposition of biomass would be identified as an additional source for the natural global level of carbon monoxide concentration in the atmosphere.

4. Conclusions

Emission of methane occurred, when insufficient aeration of the compost heap allowed anaerobic regions to develop inside the heap. Methane generation was observed during composting of mixtures of grass and soil and in the lower central parts of a heap of green waste without bush and tree trimmings. Monitoring of gas concentrations was used to detect this undesirable stage during composting.

The total release of N₂O-N, related to the initial nitrogen content, was about 0.5% in the field compost experiments with green waste from land maintenance. This was a comparatively low degree of emission of nitrous oxide, so that in this regard, low-cost composting procedures used were environmentally friendly.

Concentrations of carbon monoxide of up to 100 p.p.m. were detected in compost air. The emission of carbon monoxide corresponded to about 10⁻³ to 10⁻⁴ of the release of carbon dioxide. The generation of carbon monoxide was confirmed, by laboratory experiments. Since the concentrations of CO and CO₂ versus time had different dependencies, it was concluded that the carbon monoxide was of biogenic origin.

In laboratory studies, it was verified that increasing ventilation reduced the emission of nitrous oxide. Nevertheless, even at high ventilation rates of 100 cm³/min kg of substrate, nitrous oxide was generated. The emission of nitrous oxide was determined by the availability of nitrate ions, since there was no emission of nitrous oxide without lime saltpetre. During biodegradation of green waste, nitrate is produced by micro-organisms via the nitrification chain. It was concluded that emission of nitrous oxide could not be avoided, but could be kept at a low level with sufficient aeration.

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