

Emissions Caused by Manure Composting

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The evaluation of the nitrogen balance of biological farming included the measurement of gaseous emissions during manure composting under practical conditions. The results show that ammonia- and methane emissions subside within three to four weeks. Nitrous oxide emissions exhibited significantly greater variability with regard to time and place. Maxima of nitrous oxide emission were observed mainly after ammonia and methane had decayed. The mean total flux per compost period amounted to 1,322 g m⁻² of methane, 76.9 g m⁻² of ammonia, and 19.8 g m⁻² of nitrous oxide. The relation of these average values to the mean flux of carbon dioxide per composting period was 4.6 10⁻² for CH₄, 2.7 10⁻³ for NH₃, and 6.9 10⁻⁴ for N₂O. Both the total average gas flux and the mean ratio of total gas emissions can serve to quantify the impact on the environment. The average gaseous nitrogen losses measured during manure composting amounted to 6% of the initial nitrogen content. Carbon losses account for 35% of the initial carbon content.

Keywords:

emission rates, composting, solid manure, nitrous oxide, ammonia, methane

Emissions from Agriculture

Agriculture is responsible for approximately one third each of the total emissions of the climate-relevant trace gases nitrous oxide and methane in Germany and is virtually the only source of the environmentally relevant gas ammonia. The development of suitable reduction measures requires knowledge about the quantities emitted by the individual sources and the influence parameters. Emission data are also necessary for the evaluation of the environmental impact of farming methods. The fundamental processes that lead to the formation of trace gases have already been examined thoroughly. Nitrous oxide emissions from soils can be influenced by varying the intensity and the kind of nitrogen fertilizing [1]. In general, nitrous oxide emissions from animal husbandry are lower than emissions caused by fertilizers in crop production. Depending on the kind of husbandry, however, they may exert a considerable negative influence on the environmental balance of the farm [1-4]. Methane is produced by microorganisms during the anaerobic degradation of organic substances. Cattle husbandry is the main source of agricultural methane emissions in Germany. The largest percentage of methane emissions is caused by food digestion in the rumen. The emission rate depends on the animal type and food composition and

ranges from between about 20 kg (heifers) and 125 kg (dairy cows) of methane per animal and per year [5]. Moreover, the microbial conversion of animal excrement is a source of methane. Animal excrement also causes the emission of ammonia as a result of enzymatic urea hydrolysis. The animal species and the kind of husbandry, as well as the storage and processing of the excrement, exert a significant influence on the formation rate of nitrous oxide, methane, and ammonia [6]. On ecological farms with littered husbandry, the aerobic conversion of organic substances is the preferred method for the production of healthy farm manure without protein rotting [7]. Precise nutrient balances for ecological farming require that, among other factors, emissions into the atmosphere during manure composting be established. In addition to N- and C-emissions, the intensity of methane emissions during the layer composting of manure had to be examined because layer composting allows the compost material to be ventilated sufficiently in order to avoid methane formation. However, excrement admixtures are expected to cause methane emissions, which may develop in anoxic zones of the excrement. Therefore, the environmentally relevant gaseous emissions (ammonia, nitrous oxide, and methane) caused by the composting of manure were measured and evaluated un-

der practical conditions during six composting periods in 1998 and 1999.

Current Knowledge

In recent years, emissions caused by the storage and processing of slurry and manure have been studied more intensively by several authors. Different factors such as water content, ventilation, pore distribution and bulk density, composition, and weather conditions influence microbial activity and the emission of gases. Methane formation can be reduced through appropriate processing of the compost material in combination with sufficient ventilation [8-10]. Ammonia emissions are determined by the quantity of ammonium ions, urea, and organically bound nitrogen. The pH value, temperature, ventilation, and the C/N-relation constitute other influencing factors. An increasing pH value, higher temperature, or better ventilation lead to greater emissions [8]. High C/N relations cause NH₃ emissions to diminish [11-13].

Nitrous oxide emissions are also determined by temperature, ventilation, nitrogen content, the C/N relation, and other factors [8]. Maximum N₂O formation rates are observed if the supply of oxygen during rotting is insufficient. This may occur, for example, if the partial pressure of oxygen in the rotting material drops to zero due to great biological activity [1, 14, 15]. Ventilation and the C/N relation alter the nitrogen conversion processes. Intensive ventilation in connection with low C-content lead to nitrite accumulation in slurry (up to 33% of the total nitrogen content) and incomplete ammonium oxidation [16]. Low ventilation rates and sufficient carbon supply support the formation of nitrous oxide during nitrification- and denitrification processes [1].

It was only a few years ago that knowledge about nitrous oxide emissions during composting was insufficient [17]. Since 1993, several authors have addressed this question. Hellmann [18] studied the emission of the gases CO₂, CH₄, and N₂O during the composting of domestic waste. Her results are based on sampling with gas flux chambers and the establishment of concentration using a gas chromatograph. The total CH₄-C and N₂O-N emissions were shown in relation to the dry mass of the basic material or the total

CO₂-C emission. Depending on the composting conditions, nitrous oxide emissions range from 12 to 114 g N₂O-N per tonne of basic dry mass for a composting period of 89 days. In relation to the nitrogen content of the basic dry mass (1% to 1.7%), these emissions account for approximately 0.1 to 0.8% of the initial total nitrogen content.

Ballesterio and Douglas [19] also used gas flux chambers and gas chromatography to measure the formation of nitrous oxide during the composting of manure and garden waste. Within 60 days, manure composting caused 2.19% of the initial nitrogen content to be emitted as N₂O-N. When garden waste was composted, the percentage of N₂O-N emissions was considerably lower (1.18%). Hüther [8, 9, 20] conducted defined laboratory experiments to measure emissions during the composting of solid manure. These measurements showed that N₂O-N emissions accounted for up to 6% of the initial nitrogen content. Gas chromatography served to establish the concentration of the gases CH₄, CO₂, and N₂O. NH₃ was measured using quantitative wet-chemical analysis (reaction with boric acid). When cattle manure was composted, NH₃ emissions generally amounted to less than 5% of the initial nitrogen content. The NH₃ emissions caused by manure composting are assumed to be largely dependent upon the C/N relation. When composting cattle manure (with C/N ranging between 58 and 64), no ammonia emissions were measured even though the reliable wet-chemical method was employed [11, 12]. However, when studying the composting process in a compost stall for fattening pigs [21, 22], Kaiser and Van den Weghe measured ammonia emissions of 4.47 g per animal and per day if wood shavings were used as litter (initial C/N relation 82). If spliced wheat straw with particle lengths of 2 to 3 cm served as litter (initial C/N relation 79), ammonia emissions amounted to 8.27 g per animal and per day. However, these emissions cannot be

directly compared with clamp composting [11] because in the compost stall [22] additional excrement is produced every day, while the emissions that occur during clamp composting [11] reflect the rotting process of the basic material. In addition to the C/N relation, the physical and chemical properties of the litter determine the rotting process. Thus, the use of wood shavings as litter caused the emission of a significant quantity of nitrous oxide (5.17 g per animal and per day), while straw litter did not lead to measurable nitrous oxide emissions. These different results are attributed to the development of anoxic conditions in the wood shavings litter [11].

Measurement Methods and Error Estimation

Sampling

The emission of gases was measured under practical conditions as part of the nitrogen balance of agricultural operations that apply biological farming methods. The studies were conducted on the farm „Hof Marienhöhe“ in Bad Saarow, where ecological farming has been practiced since 1928. The manure from the cattle- and pig stall (ca. 1,000 kg per day) is processed in trapezoidal compost clamps. The clamps are arranged in individual layers by adding fresh manure in 6-13 week intervals. The layers of fresh manure are about 30 cm thick and guarantee good ventilation during composting. Turning is not necessary. Due to the rotting process, a maximum of 5 layers results in a clamp height of up to 1.2 m. In the period under consideration, a compost clamp with four layers of fresh manure (1998) and one compost clamp with two layers of fresh manure (1999) were set up (**table 1**). While each layer of fresh manure was being composted, gas analyses were conducted in the part of the clamp where fresh manure had been deposited on seven successive days. Every week, two gas samples each were taken at these se-

ven measuring points. Hence, weekly sampling allowed one-day time resolution to be achieved when studying the emission behaviour. However, local fluctuations in clamp composition and manure structure led to increased variation in the direct course of the measurement values. The mass of the fresh manure at the seven measuring points was established (**table 1**). Random samples were used to analyze and calculate the dry mass and the composition [7, 23].

The gas flux chamber was used to establish emissions from compost. This measuring technique is based on a chamber (gas bell), which is put on the surface to be examined. The quantity of emissions at the chamber base can be deduced from the alterations of the gas concentration in the chamber. The cylindrical chambers out of PVC had a volume of 0.189 m³ and a base of 0.292 m². In ideally insulated chambers, concentration can be expected to exhibit reciprocal-exponential growth until the gas that streams in has reached its equilibrium concentration. The rise of this concentration curve at the beginning of the measurements corresponds to the undisturbed emission value at the measuring point. The speed of the concentration growth is determined by the diffusion resistance in the emitting substance and the relation of the volume to the base of the chamber. Moreover, the concentration distribution that develops in the chamber influences the increase in concentration. Therefore, the properties of gas flux chambers were first studied using CO₂ emissions from compost clamps, which are easy to measure. Multi-gas monitors and CO₂ sensors were employed to determine how the CO₂ concentration alters over time at different heights in the chambers. It turned out that linear approximation of the concentration increase is permissible if the measuring time is ca. 10 minutes (**figure 1**). Concentration measurements at different chamber heights yielded identical results and thus allowed the conclusion to be drawn that concentration

Table 1: Quantity and composition of solid manure during the individual composting periods

Component		Composting periods ¹⁾					
		18.04.1998	02.07.1998	20.08.1998	01.10.1998	08.04.1999	05.08.1999
		01.07.1998	19.08.1998	30.09.1998	18.11.1998	04.08.1999	13.11.1999
Fresh manure	[kg]	8933	6010	7076	7009	9961	5851
Dry mass	[kg]	2189	1472	1734	1717	2441	1433
Carbon	[kg]	985	663	780	773	1098	645
Nitrogen	[kg]	49	33	39	39	55	32,3

¹⁾ The first clamp comprised four layers of fresh manure (set up on 18 April 98, spread on 6 April 99). The second clamp was composed of two layers of fresh manure (set up on 8 April 99, spread on 13 November 99).

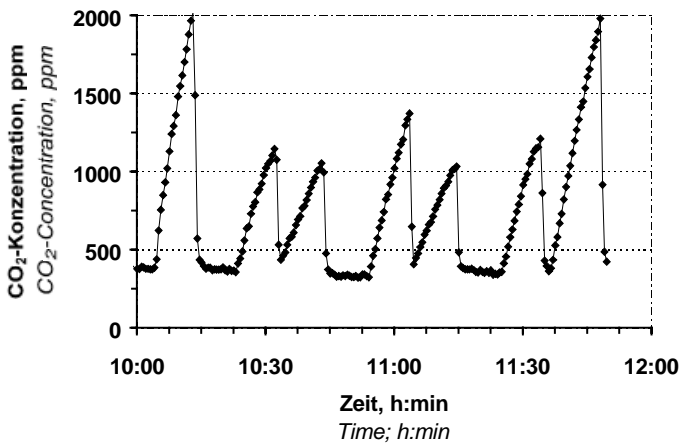


Figure 1: Increase in CO₂-concentration in flux chambers measured with CO₂-sensors during gas sampling on solid manure

distribution in the chamber is not caused by diffusion, but that convection leads to fast levelling of the concentration within the chamber. Moistened cloth collars were used for surface-adapted insulation of the gas flux chambers and for the limitation of gas losses due to the porous structure of the compost at the base of the chamber. To check tightness, the course of the CO₂ increase over time was evaluated. Incorrect measurements were noted and repeated or rejected afterwards. After a collection time of 10 minutes, gas bags were filled and analyzed after 24 h using Fourier transformation infrared spectroscopy (FTIR). On every measuring day, three samples of the surrounding air were taken at the site (before, during, and after gas sampling had been completed) because the calculation of the emission values includes concentration differences between the samples and the surrounding air.

FTIR Measurements and Error Assessment

The composition of samples of compost air and surrounding air was established with an FTIR spectrometer at a spectral resolution of 0.2 cm⁻¹. Thanks to this high resolution, undisturbed lines of all gases to be examined can be evaluated. Thus, higher measurement precision can be achieved as compared with routine spectrometers, whose resolution generally ranges from 1-4 cm⁻¹. To achieve high measuring sensitivity, a low-noise MCT detector (MCT: mercury-cadmium-tellurium alloy) and a long-way gas measuring cell with an optical way length of 20 m were used. The measuring volume of the gas cell amounted to five litres. The temperature of the gas measurement cell was kept at 80°C in order to avoid undesirable condensation on the cell mirrors.

The systematic error of the FTIR measurement for the gases CH₄, CO₂, and N₂O is ca. 20%. Depending on the concentration, the systematic FTIR error may vary because it originates during calibration in connection with the non-linear absorption

behaviour of the infrared lines and is determined by the concentration precision of the calibration gases. If the concentration of ammonia is measured, a higher systematic and stochastic error of up to 40% must be expected. This especially applies to lower concentrations in the ppm range. When ammonia is used for calibration, systematic errors are caused by the high adsorption of NH₃ on all surfaces of solid bodies. This adsorption reduces the concentration of the calibration gas during the measuring process. However, the surfaces release NH₃ again when calibration gases or samples with low NH₃ concentrations are measured. To keep this error small, calibration and sample measurement followed the same time regime (dynamic measurement method). This means that before the begin of the measurement the spectrometer must be „NH₃ free“. This can be achieved by evacuating for a sufficiently long time.

The number of spectrum accumulations (measuring time) should be identical for both the calibration- and the measuring process. When NH₃ emission measurements are conducted, stochastic errors are caused by sampling (adsorption on the walls of the gas flux chamber, solution of NH₃ in condensation water). In order to avoid errors due to condensation in the gas bags, the bags were warmed up before the measurement to a point where no condensation water was present.

Detector noise and alterations of the room temperature during the measuring time constitute other stochastic sources of errors in the FTIR measuring method. FTIR spectra are gained from the difference of two measurements. The first interferogram is taken in the evacuated state of the gas cell (background spectrum). The second interferogram is obtained from the gas to be examined (sample spectrum). After allowing for the frequency course of the measuring instrument, the Fourier transformation of the difference interferogram yields the desired FTIR spectrum of the sample. If a temperature drift occurs

between these two measurements, minimal length alterations of the interferometer may cause interference errors (phase errors) in the measuring instrument. These two last-mentioned sources of errors increase the relative error of concentration measurements, especially near the detection limit. While errors caused by noise can be reduced through spectrum accumulation, room temperature control or small time intervals between background- and sample measurements must be employed to minimize phase errors. Due to diffusion losses, additional stochastic and systematic errors result from sampling and the transport time of the samples. In order to allow the systematic error caused by diffusion to be estimated, the storage time-related change in concentration in gas bags was measured and evaluated using a linear drop-off function $C(t) = C_0 (1 - b \cdot t)$. The regression coefficient b amounted to 0.0039 h⁻¹ for CH₄ and CO₂. The value for N₂O and NH₃ was 0.0086 h⁻¹. If 24 hours pass between sampling and measurement, the concentration of CH₄ and CO₂ can thus be expected to diminish by about 10%. N₂O and NH₃ concentrations drop by approximately 20%. In the results of the analysis, this reduction in concentration was accounted for using compensation calculations. Sampling on compost is another source of stochastic errors. Compost is bulk material with a high percentage of air volume. Due to heat generation in the compost, convective air exchange is predominant. When the gas flux chamber is put on, the flow conditions change so that the values gained with gas flux chambers are lower than undisturbed emission.

Development of Emissions during Manure Composting

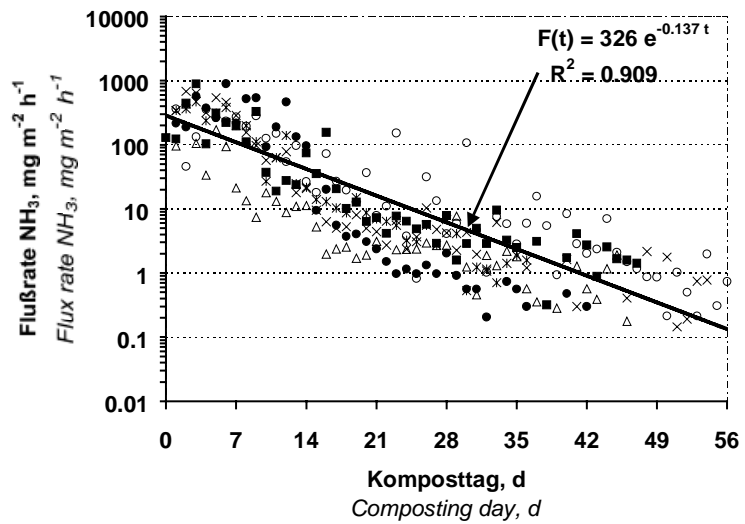
Over the course of a composting period, the emissions of ammonia and methane are reduced almost completely during the first three weeks. Nitrous oxide emissions exhibit great variation over the entire composting period. As the quantity of NH₃ and CH₄ diminishes, N₂O emissions show an increasing tendency. Maximum values are measured after two to six weeks. Afterwards, N₂O formation slowly decreases. These typical developments were observed during all composting periods examined. The highest ammonia emissions are measured two to five days after the manure has been put on. Afterwards, emission values drop. The maxima of the NH₃ emission rates range from between ca. 0.1-0.9 g m⁻² h⁻¹. The mean maximum value amounts to about 0.3 g m⁻² h⁻¹. By approximation, the reduction

behaviour of the ammonia emissions (**figure 2**) can be described as an exponential decrease. This form of development is the result of constant degradation reactions, i.e. proportionality between degradation rate (emission) and convertible material. Basic substances for NH₃ emission are solved NH₃, NH₄⁺-ions, and urea in addition to other, organically bound nitrogen in the substrate. For NH₃, the trend function provides a determinateness R² of 0.909 with a decay time (1/e decrease) of 7 days if the calculation is based on eight weeks of composting. In contrast to nitrous oxide and methane, it is not possible to evaluate ammonia emissions over a longer composting period. Due to the faster decrease, the detection limit for ammonia, which is 0.1 mg m⁻² h⁻¹, is reached after approximately eight weeks. This causes fluctuations around the zero point, which prevent exponential adaptation and logarithmic representation.

The detection limit of the chosen set-up of measuring instruments for methane and nitrous oxide is 0.02 mg m⁻² h⁻¹ so that evaluation was extended to comprise this range. Anaerobic degradation in excrement particles is assumed to be the methane source. Methane emissions exhibit an exponential decrease until about 10 weeks after their beginning (**figure 3**). The trend function for the CH₄ emissions has a decay time of 10 days and an R² of 0.737. Calculations over eight and ten weeks of composting both yield these trend function values. Therefore it can be assumed that methane formation continuously diminishes over this period.

Nitrous oxide emissions exhibit great variation over the entire composting period. N₂O emissions show an increasing tendency as NH₃ and CH₄ decrease. Maximum values are measured after two to six weeks (**figure 4**). Afterwards, N₂O formation drops continuously. The description of the course of the emissions with an exponential trend function shows a significantly longer decay time (26 days) as compared with methane and ammonia, as well as considerably greater variation (determinateness R² = 0.303).

Shortly after the start, CO₂ formation reaches its maximum values and then continuously decreases over a period of more than three months (**figure 5**). An exponential trend function enables the data gained in experiments to be described with sufficient precision. If measurement data are taken into account until the 70th composting day, the adaptation of this function reaches its highest determinateness of R² = 0.819. For the definition of the evaluation indicators of composting, it is necessary to determine the duration of a



Symbols for composting periods					
18.04.1998	02.07.1998	20.08.1998	01.10.1998	08.04.1999	05.08.1999
01.07.1998	19.08.1998	30.09.1998	18.11.1998	04.08.1999	13.11.1999
○	■	△	×	●	✱

Figure 2: NH₃-Emissions with an exponential trend function $F(t) = 326 e^{-0.137 t}$ for 56 composting days with a determinateness of $R^2 = 0.909$ on a logarithmic scale

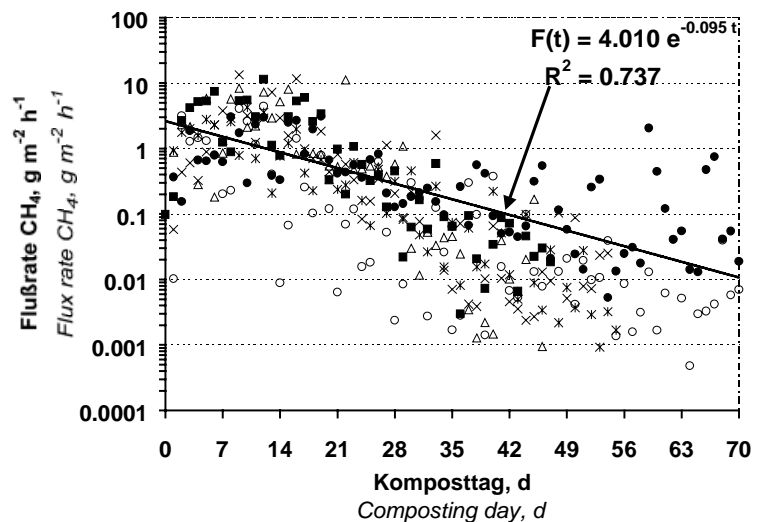


Figure 3: CH₄-Emissions with an exponential trend function $F(t) = 4.010 e^{-0.095 t}$ for 70 composting days with a determinateness of $R^2 = 0.737$ on a logarithmic scale (Symbols like Figure 2)

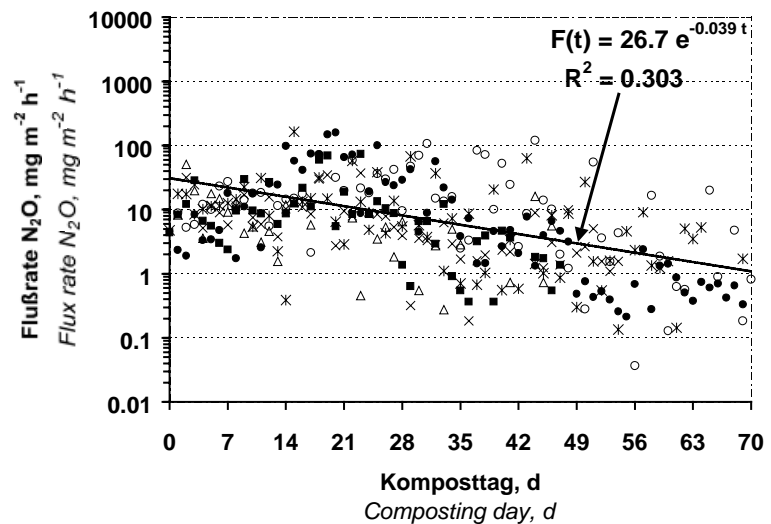


Figure 4: N₂O-Emissions with an exponential trend function $F(t) = 26.7 e^{-0.039 t}$ for 70 composting days with a determinateness of $R^2 = 0.303$ on a logarithmic scale (Symbols like Figure 2)

composting period with the aid of objective criteria. According to the results of the CO₂ emission measurements, the composting period was set at 10 weeks. In addition to the reduction of the CO₂ values, CH₄ decrease also speaks in favour of this period.

Average emission during the six composting periods amounted to 1,322 g m⁻² of CH₄, 76.9 g m⁻² of NH₃, and 19.8 g m⁻² of N₂O per composting period. If these total emissions are related to the average value of the quantities of CO₂ produced during the composting periods, indicator values amount to approximately 4.6 · 10⁻² for CH₄, 2.7 · 10⁻³ for NH₃, and 6.9 · 10⁻⁴ for N₂O. These emission conditions are not constant values, but quotients that depend on the development of the accumulated emissions (figure 6). Only after the end of the rotting process do the values become constant because at that point no further emissions can become part of the accumulation.

If one draws up a balance of the emissions during the composting periods (table 2), the CH₄-C emission ranged from between 2.1 and 7.7% of the carbon content in the dry mass of the fresh manure. NH₃-N emissions caused nitrogen losses of 2.2-7.0% of the initial N-content in the dry mass. N-losses through N₂O-N release ranged from 0.3 to 1.9%. As compared with the results cited above for manure composting [8], ammonia emissions are in the same order. Nitrous oxide emissions are lower. During the composting periods, 25-45% of the initial carbon content is used as a source of energy for the microbial conversion of the compost.

Conclusions

Manure composting leads to relatively high carbon losses. Ammonia emissions cause the largest part of the nitrogen losses. The emission values measured under practical conditions correspond with the findings set forth in the literature. The fact that nitrous oxide emissions are relatively low in relation to the initial content is proof of sufficient ventilation and the en-

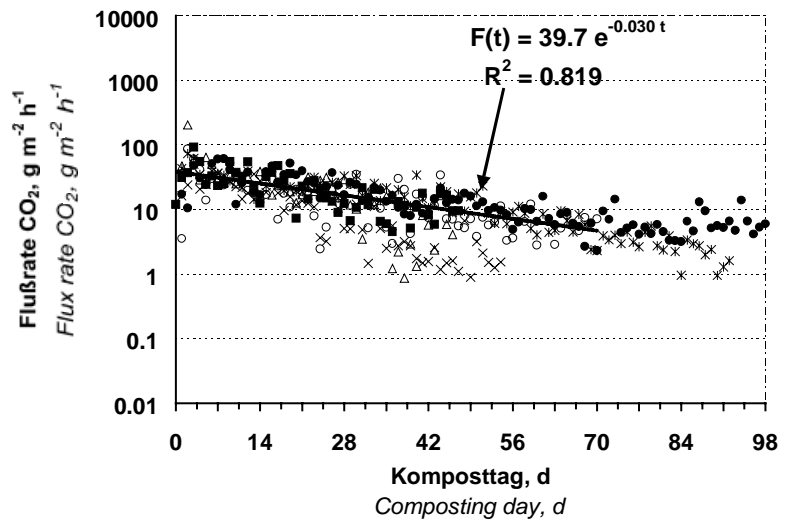


Figure 5: CO₂-Emissions with an exponential trend function $F(t) = 39.7 e^{-0.030t}$ for 70 composting days with a determinateness of $R^2 = 0.819$ on a logarithmic scale (Symbols like Figure 2)

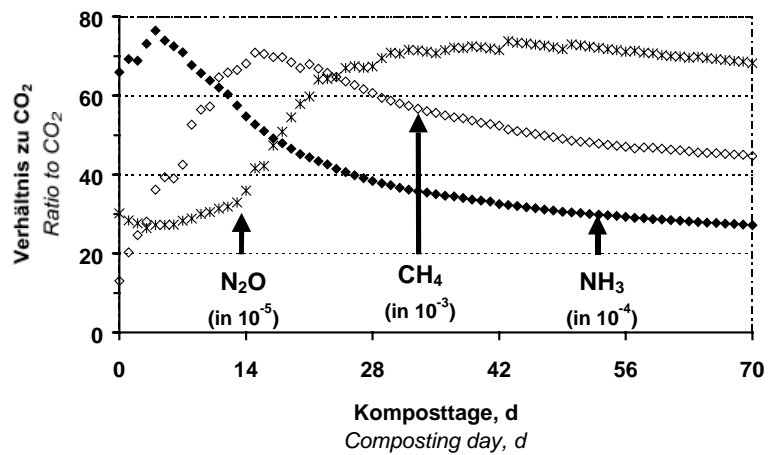


Figure 6: Accumulated emissions in relation to accumulated CO₂-emissions (mean values of 6 composting periods)

vironmental compatibility of layer composting. Even though fresh manure is put on in layers, methane emissions cannot be avoided. The excrement part of the manure is assumed to be the source of methane.

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Table 2: Total emissions during the individual composting periods

Gases	Composting periods						
	18.04.1998 01.07.1998	02.07.1998 19.08.1998	20.08.1998 30.09.1998	01.10.1998 18.11.1998	08.04.1999 04.08.1999	05.08.1999 13.11.1999	
CO ₂ -C-Percentage [%] ¹⁾	30,0	27,3	37,8	19,2	27,9	39,5	
CH ₄ -C-Percentage [%] ¹⁾	2,1	7,7	6,3	5,4	2,2	2,5	
NH ₃ -N-Percentage [%] ²⁾	5,6	7,0	2,2	7,0	4,6	4,9	
N ₂ O-N-Percentage [%] ²⁾	1,9	0,8	0,3	0,7	1,6	1,3	

¹⁾ Percentage of C in the dry mass

²⁾ Percentage of N in the dry mass

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