

Greenhouse gas emissions from digestate in soil

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Abstract

Purpose Biogas residues, digestates, contain valuable nutrients and are therefore suitable as agricultural fertilizers. However, the application of fertilizers, including digestates, can enhance greenhouse gas (GHG) emissions. In this study different processes and post-treatments of digestates were analyzed with respect to triggered GHG emissions in soil.

Methods In an incubation experiment, GHG emissions from two contrasting soils (chernozem and sandy soil) were compared after the application of digestate products sampled from the process chain of a food waste biogas plant: raw substrate, digestate (with and without bentonite addition), digestates after separation of liquid and solid phase and composted solid digestate. In addition, the solid digestate was sampled at another plant.

Results The plant, where the solid digestate originated from, and the soil type influenced nitrous oxide (N₂O) emissions significantly over the 38-day experiment. Composting lowered N₂O emissions after soil application, whereas bentonite addition did not have a significant effect. High peaks of N₂O emissions were observed during the first days after application of acidified, liquid fraction of digestate. N₂O emissions were strongly correlated to initial ammonium (NH₄⁺) content.

Conclusion Fertilization with dewatered digestate (both fractions) increased N₂O emission, especially when applied to soils high in nutrients and organic matter.

Keywords Greenhouse gas, Nitrous oxide, Digestate, Food waste, Organic fertilizer

Abbreviations

C	Compost	RH	Relative Humidity
CH ₄	Methane	RBA	Romerike Biogas plant – mesophilic process
CO ₂	Carbon dioxide	S	Substrate
D	Digestate	SO	Soil Only
DB	Digestate and Bentonite	THP	Thermal Hydrolysis Process
D-s	Digestate - solid fraction	TK	Tukey Kramer
D-l	Digestate – liquid fraction	WFPS	Water Filled Pore Space
D-s-l	Digestate – solid and liquid fraction		
GHG	Green House Gas		
HRA	Hadelandog Ringerike Avfallsselskap AS – <i>thermophilic process</i>		
NH ₄ ⁺	Ammonium		
N ₂ O	Nitrous Oxide		
NO ₃ ⁻	Nitrate		
N _{tot}	total Nitrogen		
PS	Pore Space		

Introduction

The atmospheric concentration of gases with an effect on the earth's radiative forcing is increasing and climate is changing. The gases affecting radiative forcing are primarily carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O). CO₂ produced from combustion of fossil fuel is considered as a net addition to GHG emissions, whereas CO₂ emitted from agricultural soils is considered as neutral in the IPCC 2014 report (Smith et al. 2014) because it is part of the short-term cycles of carbon. The agricultural sector is also contributing a large part of anthropogenic non-

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CO₂ GHG, mainly N₂O and CH₄ emissions (USEPA 2012).

Methane is produced under anoxic conditions and the most important sources are ruminant digestion and flooded soils (Mapanda et al. 2011; Nusbaum 2010). The largest source of N₂O emissions in the agricultural sector are agricultural soils and several factors are known to contribute to N₂O emissions, primarily available N, pH, organic matter content and soil moisture (Šimek and Cooper 2002; Maag and Vinther 1996; Ranucci et al. 2011). However, because N₂O can be produced by a number of biological processes (Wang et al. 2017), emission rates are difficult to predict.

Biogas is produced by anaerobic digestion (AD) of organic matter in a reactor. The gas produced is a mixture of CO₂ and CH₄ and the CH₄ can be used to replace fossil fuels (Horschig et al. 2016; Dahlin et al. 2015). The partly degraded organic material left after AD is called digestate. Degradation of organic material depends on feedstock and on retention time, pre-treatment and process types. Digestates are rich in plant available nutrients (Albuquerque et al. 2012b) and usually have a high pH. Many biogas plants mechanically separate digestate into a liquid and a solid fraction which can more easily be transported and stored. Often polymers are added to improve separation. The solid phase is known to contain mainly organic N and the liquid fraction holds usually the major part of ammonium. In some cases, also further treatments are used, such as acidification to reduce ammonia volatilization from liquid phase of digestate. Because of the high concentration of plant available nitrogen (N), digestates are good fertilizers and can replace mineral fertilizers (Holm-Nielsen et al. 2009). Hence, the substitution of synthetic fertilizer can save energy and money by reduced use of mineral fertilizer, while still adding N to the soil (Sigurnjak et al. 2017).

Digestates have characteristics that set them apart from both mineral fertilizers and most other organic fertilizers. Most of the organic carbon in the substrate is used up for producing CH₄ and CO₂ during the biogas process. Digestates are rich in mineral nitrogen, almost exclusively on ammonium (NH₄⁺) form, prone to losses via NH₃, and they also contain some residual organic matter with organic N (Albuquerque et al. 2012b). They usually have high pH and, at least untreated, high water content.

Various studies have already investigated the effect of application of digestate and digestate products on soil quality and fertility (Möller and Müller 2012), soil biological properties (Albuquerque et al. 2012b), soil

chemical properties (Losak et al. 2014), crop yield and other effects on crops (Nkoa 2014; Albuquerque et al. 2012a, b) and leaching losses (García-Albacete et al. 2014). However, the effects of digestate on GHG emissions from soil have not been much studied (but see Senbayram et al. 2014; Johansen et al. 2013). It is not straight forward to predict how the special properties of digestate and digestate products will affect emissions. Assumptions can be made regarding composition affecting emissions: High ammonium content and high pH mean a high risk of N losses due to ammonia volatilization during spreading and storage, how pH affects GHG emissions from soil is more uncertain. It is e.g. well recognized that high organic matter content enhances N₂O emissions as the availability of carbon and N usually increases denitrification rates. On the other hand, increased denitrification rates do not necessarily lead to higher N₂O emissions. N₂O can be produced both during nitrification and denitrification, although denitrification is usually thought to be dominant. Digestate was shown to increase nitrification-derived N₂O emission (Senbayram et al. 2009). It is unclear how the high NH₄⁺ and low NO₃⁻ combined with relatively high organic matter content as is found in digestate will affect emission. Methane emissions would be expected to be low when digestates are applied to aerobic soils, but as they contain methanogenic bacteria, they may still induce emissions.

Acidification of digestate makes it possible to reduce water content by evaporation with minimal ammonia losses. However, it is unclear how pH affects N₂O emissions (Šimek and Cooper 2002; Mørkved et al. 2010) and therefore, how acidification affects subsequent N₂O emissions when digestate is added to soils (Raut et al. 2012).

Composting the solid fraction of digestate could give a high-quality soil conditioner containing less N than e.g. mineral fertilizer but more recalcitrant organic matter leading to humus formation. Composting is an aerobic decomposition process where smell is reduced or eliminated and some NH₄⁺ is transformed to NO₃⁻. However, substantial amounts of N are usually lost as ammonia during composting, and some GHG emission will also occur (Jiang et al. 2013; Fillingham et al. 2017; Hellebrand and Kleinke 2000). It is not clear, however, if compost also increases GHG emission when applied to soil.

NH₄⁺ in digestates is easily sorbed to clay particles, so soil properties determine availability for plants and leaching losses. Sorbents like clay particles can

improve N retention and optimize efficiency of use (Ma et al. 2011; Wang et al. 2014). They can also be used to concentrate nutrients in the solid phase of liquid waste streams, e.g. digestate. Bentonite is a clay whose properties are determined by smectite minerals and it is known to be a good sorbent for NH_4^+ . It is not well known how sorbents affect GHG emissions. If N availability for microbes is decreased by sorption, it could reduce N_2O emissions.

In this study GHG emissions from digestate and digestate products after application to soil were investigated. The digestates were incubated in two soils with contrasting properties and N_2O , CH_4 and CO_2 emissions were measured. Effects of digestate processing and post-treatment on GHG emissions were determined. We hypothesized that i) separation might lead to lower N_2O emissions in the solid phase and to higher emissions in the liquid phase, that ii) composting of the solid fraction of digestate will reduce N_2O and CH_4 emissions and that iii) the application of digestate with bentonite might reduce N_2O emissions.

Materials and Methods

Laboratory incubation

Based on an equal volume of 50 ml, 71.33 g of sandy soil (low carbon, nutrient and clay content) and 59.70 g of chernozem (high carbon, nutrient and clay content) were transferred to lab bottles (250 ml) each. The soils were pre-incubated at 40 % water filled pore space (WFPS) in a climate chamber at 20°C for 4 days. After pre-incubation, the treatments were added to the soils in the bottles according to Table 1.

The amount of digestates and compost used was chosen based on the total amount of nitrogen (N_{tot}). The total nitrogen applied (8.20 mg N_{tot} per bottle) was calculated based on the amount of fertilizer- N_{tot} known to be applied in the county Akershus, Norway

(164 kg N_{tot} /ha) (Deelstra and Greipsland 2017). A depth of 10 cm ($0.1 \text{ m}^3 / \text{m}^2$) soil was assumed to be influenced by the fertilizer application.

The added material was mixed with the soil to mimic the procedure of working in the fertilizer after application. The water content was adjusted to 60 % WFPS at the beginning of the incubation (Table 6 in Appendix), as this is reported to cause high N_2O emissions from nitrification and denitrification (Del Prado et al. 2006; Van Lent et al. 2015; Senbayram et al. 2009). During the incubation, the temperature was set to 20°C at a high rel. humidity (RH) of 90 % to minimize evaporation.

Digestate sampling

Romerike biogas plant (RBA) and Hadeland og Ringerike Avfallsselskap AS (HRA) are two biogas plants to the north of Oslo, both only treating food waste mainly collected from households but also originating from food industry. The organic waste was grinded, sieved, screw pressed and heated up to 80°-100°C before it was used as a substrate in the digestion process in the biogas plant.

The biogas plant in Romerike uses Thermal Hydrolysis Process (THP) as pre-treatment and a mesophilic process (38°C), whereas HRA is running a thermophilic process (52-53°C). At RBA, samples were taken at different stages of the process (Fig. 1): before anaerobic digestion, after the digestion process, after the de-watering process (liquid and solid fraction) and from composted digestate. The compost was collected from a different site (ØRAS - Miljøstasjon) which received solid digestate from RBA for composting. Wood chips were added as structural material. Only the solid fraction after de-watering was collected from HRA. All the samples were stored at a temperature of 4°C and analyzed by Eurofins and NIBIO (Table 2).

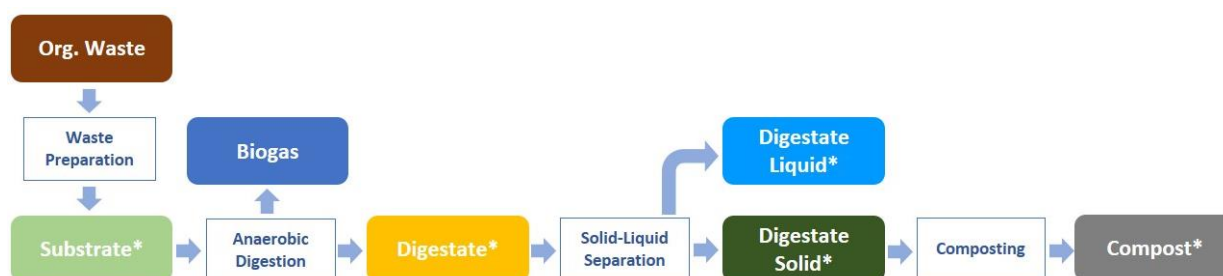


Fig. 1 Anaerobic digestion process including composting (*Sampled stages)

Measurements

Measurement of N₂O, CH₄, CO₂

For gas measurements, bottles were closed for +/- 1 hour in the beginning and up to +/- 2 hours towards the end of the experiment to increase the concentration (time schedule in Table 9 in Appendix). Then, 12 ml of gas was extracted through a septum with a syringe. This gas was injected into an evacuated vial and the samples were analyzed by gas chromatography mass spectrometry (GC-MS) to determine concentrations of N₂O, CH₄, and CO₂. The analysis was performed using an Agilent Technologies 7820A GC System gas chromatograph, coupled to a mass detector Agilent Technologies 5875 Series MSD and a Gilson 222 XL auto sampler. The sample was injected by a 5 ml sample loop, through a 0.5 m x 0.32 mm deactivated precolumn, into a 25 m x 0.32 mm CP-PoraPLOT Q-HT column (Chrompack), kept at 40 °C. Helium was used as carrier gas at 1.0 ml min⁻¹. Measurements were performed twice on the first day after starting the incubation, then every day at the beginning and at longer time intervals towards the end of the

experiment. The total number of sampling days was 24 during a time period of 38 days.

Measurements after experiment

The pH of all used soil samples was measured after the incubation. A slurry of the wet soil (equivalent to 10 ml of dry soil) and 25 ml deionized (DI) water was shaken and left overnight. Samples were shaken again before measurement with a Thermomix electrode.

Furthermore, NO₃-N and NH₄-N contents of all the samples after incubation were determined by KCl-extraction method. 100 ml 1 M KCl was added to a weighed sample of soil (10 g organic soil / 40 g mineral soil) and stirred for 1 hour and then filtered before analyses. The NO₃⁻ was determined according the ISO 14256-2/NS-EN ISO 11732 and NH₄⁺ according the NS-EN ISO 11732 part 3.

Treatments

The different treatments are shown in Table 1. All of them were repeated on the two soil types with three replicates. Bottles with the same amount of inert sand (also 50 ml) were used as a standard.

Table 1 Treatments of the 2 soils (19 combinations x 3 replicates = 57 bottles in total)

Treatments	Abbrev.	Sand	RBA		HRA	
			Chernozem	Sandy soil	Chernozem	Sandy soil
Substrate + soil	S		x	x		
Digestate + soil	D		x	x		
Digestate + bentonite + soil	DB		x	x		
Solid fraction digestate ^a + soil	D-s D-s-HRA		x	x	x	x
Liquid fraction digestate ^a + soil	D-l		x	x		
Liquid ^a + solid ^a fractions digestate + soil	D-l-s		x	x		
Composted digestate ^a + soil	C		x	x		
Soil only	SO		x	x		
Inert sand		x				

^aAfter centrifugation

Substrate (S) – Grinded food waste, sieved and screw pressed (max. diameter 10 mm), heated up to 80° - 100°C for about 20 min; samples were taken before THP, liquid;

Digestate (D) - Taken directly from the digester, sieved through a 2-mm sieve, liquid;

Digestate with bentonite (DB) - Bentonite material mined in Pétervására (Hungary) applied in bentonite digestate ratio (volume) of 1:9 before mixing (Makadi and Nemeth-Borsanyi 2016) it into the soil. The montmorillonite content was ca. 50 % and the material was ground to a particle size range of 0.05–1 mm and

dried in a cyclone at 70°C. Its chemical composition was determined by XRF (fluorescence x-ray spectrometry) from scanning electron microscopy (SEM) in weight % and it can be seen in (Table 7 in Appendix).

Solid fraction of digestate (D-s and D-s-HRA) - Solid organic material separated from the water by decanter centrifuge after adding a polymer for flocculation; sampled at both biogas plants (D-s: RBA and D-s-HRA: HRA).

Liquid fraction of digestate (D-l) – Concentrated liquid fraction of digestate after addition of polymer,

acidification and separation by centrifugation.

Liquid and solid fraction (D-l-s) - Based on dry matter content of the concentrated fraction of digestate after centrifugation (D-l), a concentration by factor 10 was assumed. The concentrated liquid was diluted with DI water to the initial water content of digestate and then mixed with the corresponding mass of the solid fraction.

Compost (C) - Solid fraction of digestate composted together with structure material, mostly wood chips. The age of the compost was ca. 8 months when it was sampled.

Soil only (SO) - Chernozem or sandy soil without treatment.

Table 2 Characteristics of treatments: substrate (S), digestate (D), digestate and bentonite (DB), solid digestate (D-s), solid digestate from HRA (D-s-HRA), liquid digestate (D-l) and compost (C)

Treatment	pH	Conductivity [mS/m]	TS _t [%]	TOC [% TS]	N _{tot} [mg N _{tot} /kg TS]	NH ₄ -N [mg NH ₄ -N/kg TS]	NO ₃ -N [mg NO ₃ -N/kg TS]	C/N ratio
S	3.9	140	8.6	50.6	28000	404.7 ^a	38.7 ^a	18.1
D	8.5	140	3.1	40.5	94000	34193.5 ^a	322.6 ^a	4.3
DB			3.1	40.5	94000	34193.5 ^a	322.6 ^a	4.3
D-s	8.80	100	28.3	43.5	63000	3044.0	3.7	6.9
D-s-HRA	8.90	190	23.4	37.8	37000	9388.0	4.6	10.2
D-l	5.30	830	9.9	35.1	68000	58131.3 ^a	101.0 ^a	5.2
C	7.8	120	61.3	16.2	17000	14.7 ^a	1065.0 ^a	9.5

^aAnalyzed by NIBIO

Soil characteristics

A chernozem and a sandy soil, both originating from Hungary, were used for the experiments. These soils were chosen for the experiment because of their contrasting properties (Table 3). The properties were measured by Eurofins standard methods, except pore space (PS) and texture (wet sieving method) that was measured at Norwegian Institute of Bioeconomy Research (NIBIO).

Table 8 in the Appendix shows the grain size distribution (weight %) of chernozem and sandy soil.

Pore space (PS) - Water filled pore space (WFPS)

The PS of the soils was measured based on the method of Blume et al. (2011) (Table 3). In addition to the measurement, the pore volume was calculated by assuming a real density of 2.65 g/cm³ (reference value of quartz) for the mineral particles and a lower value of 1.4 g/cm³ for the organic matter. The fraction of the organic material was interpreted to equal loss on ignition (PS: 50.0 % - chernozem and 41.5 % sandy soil). All samples were adjusted to 60 % WFPS at the beginning of the incubation considering the moisture of the treatments.

Table 3 Major soil properties of the sandy soil and the chernozem

Soil properties	Sandy soil	Chernozem
Dry matter DM (%)	99.4	96.9
pH	7.0	6.4
Conductivity (mS/m)	2.5	5.4
Loss on ignition (% DM)	1.2	5.7
Sand (% DM) ^a	92.0	11.6
Silt (% DM) ^a	2.3	60.7
Clay content (% DM) ^a	5.7	27.7
Pore space (%) ^a	42.0	49.8
Total carbon (% DM)	0.22	1.9
Nitrogen (% DM)	0.04	0.21
Phosphorous (% DM)	0.023	0.071
Potassium (% DM)	0.067	0.31
Ammonium-N (% DM)	0.00076	0.00187
Nitrate-N (% DM)	0.00029	0.00317
Phosphorous (% DM)	0.0047	0.01
Potassium (% DM)	0.01	0.036

^aAnalyzed by NIBIO

Calculations and statistical analysis

Cumulated emissions and GWP (global warming potential)

The production rates for CH₄-C, CO₂-C and N₂O-N per 14 days were calculated for each of the three replicates and then averaged. The trapeze approach was applied to calculate the gas produced between the time steps before cumulating the emissions. The GWP calculation was based on the IPCC 2013 factors (Myhre et al. 2013) for 20 years: 264 for N₂O, 84 for CH₄ and 1 for CO₂.

Statistical Analysis

SAS 9.4 TS Level 1M1 statistic software for Windows was applied to perform statistical analysis for cumulative emissions. A linear model was used for each response variable (CH₄-C, CO₂-C and N₂O-N), which was assumed to be nearly normally distributed. The assumptions of normality, homogeneous variance etc. were checked by an analysis of the residuals from the fitted model. The explanatory factors were soil (with two levels: Chernozem and Sandy soil), treatment (with nine levels: S, D, DB, D-s, D-s-HRA, D-l, D-l-s, C and SO), and their interaction. When there were significant ($p \leq 0.05$) effects of a factor, Tukey's multiple comparison method was used to order the groups, interactions or main effects, as far as the data allowed. Before these final analyses, some outlier observations were deleted, based on a preliminary similar analysis, to avoid that a few observations totally dominate the results.

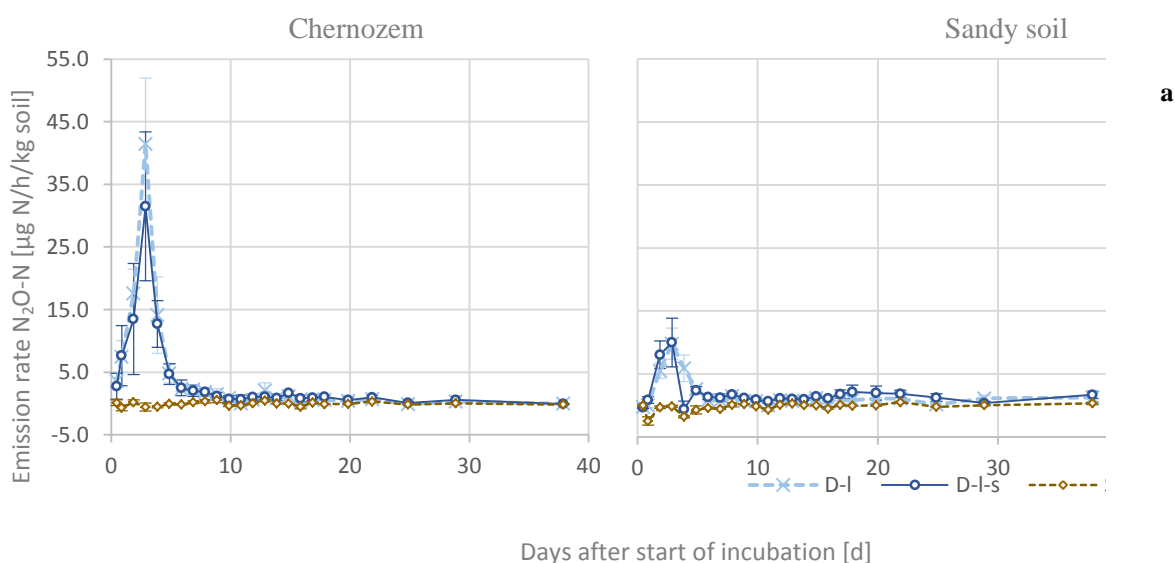
The same software was used to analyze data using a model for repeated measurements. This analysis was done separately for each soil and gas. The model contains treatment, hours from start, and their interaction as independent variables, treatment as a factor and hours from start as a covariate. The model considers that observations from the experimental unit may be correlated. The residuals indicate good approximation to the usual assumptions of normality, etc. 4 treatments were compared in this analysis: C, D-s, D and D-l. Because of large differences between replicates, it was not possible to carry out this analysis for all treatments.

A multiple linear regression was performed (Minitab v18). Measured values of NO₃⁻, NH₄⁺ and total organic carbon in the mixtures were used as predictors. Average values of cumulative gas emissions were used as response variables (one regression for each gas, CO₂, N₂O and CH₄).

Results and Discussion

N₂O and CO₂ emissions

The application of liquid, acidified digestate resulted in peak N₂O emission within the first 5 days for both soils (Fig. 2a). There was no or only little effect of bentonite addition on N₂O emission rates (Fig. 2b), but a clear reduction after composting the solid digestate (Fig. 2c and d). The solid digestate from HRA (D-s-HRA) caused high emission rates during the first 20 days, whereas the same fraction from RBA (D-s) resulted in enhanced emissions on a much lower level (Fig. 2d).



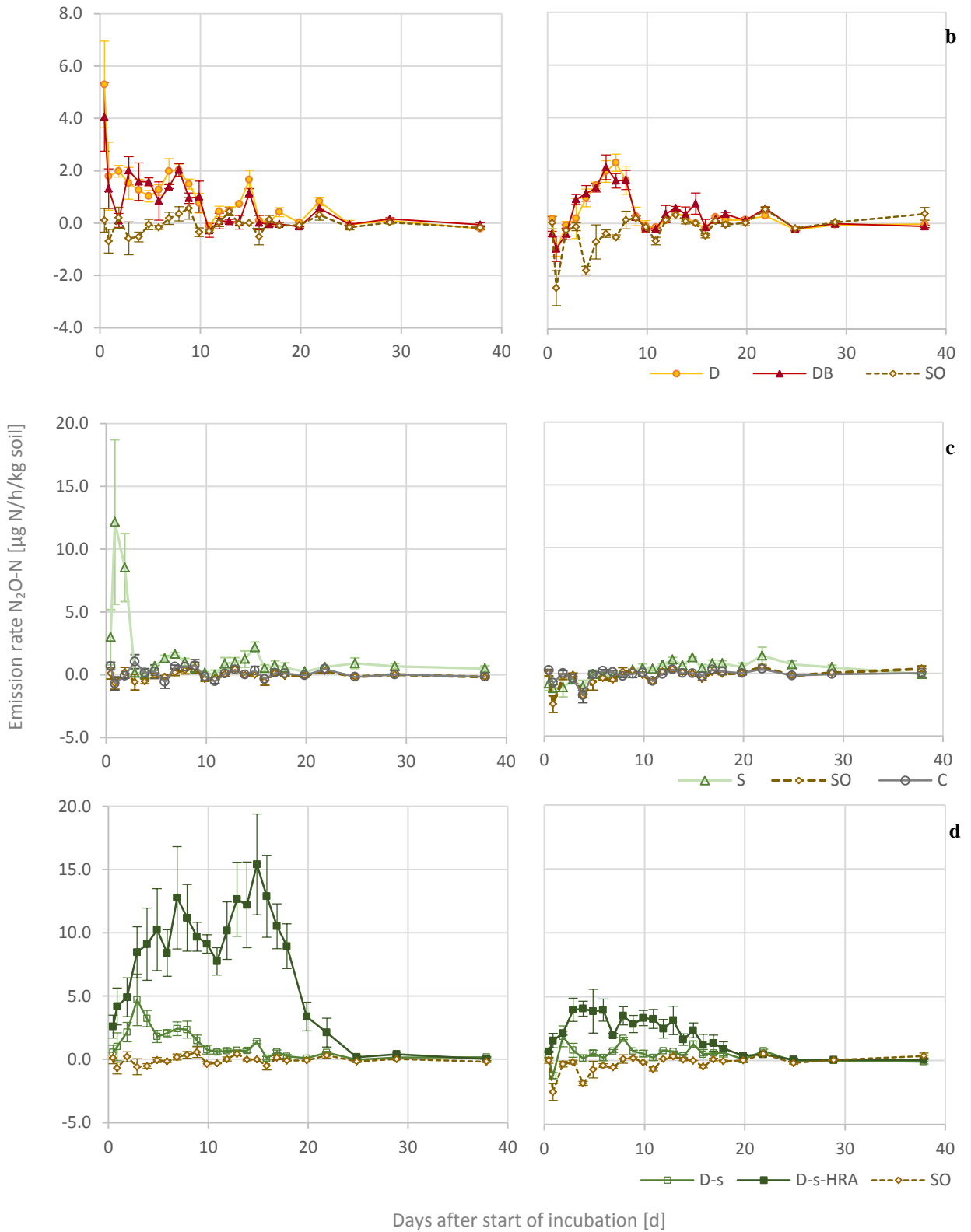


Fig. 2 Comparison of N₂O-N emission rates (means) from chernozem (left) and from sandy soil (right) after adding D-l or D-l-s (a), D or DB (b), S and C(c), and D-s and D-s-HRA (d) or no treatment (SO), respectively. Error bars indicate standard error (n=3)

Hence, five treatments had high N₂O emission rates that are cause for concern: sustained high emissions from the soils treated with D-s-HRA, high peaks of emissions from soils with D-l and D-l-s (peaking on 4th day) and initial emissions right after application of the liquid treatments D and DB to the chernozem.

Soil effects

N₂O and CO₂ emission rates were found to be higher from the chernozem than from the sandy soil in almost all treatments, throughout the 38-day time run (Fig. 2 and 3). However, the patterns of emission rates over time were similar for both soils in most treatments.

The enhancing effect of chernozem compared to sandy soil on N₂O can be attributed to its differing characteristics and carbon and N contents. It contained 2.5 times more NH₄⁺, 10 times more NO₃⁻ and 8.5 times more total organic carbon (TOC) than the sandy soil.

The differing texture and structure of the soils could have promoted N₂O emissions in the chernozem by creating more microsites with partially anoxic conditions at 60 % WFPS than in the sandy soil. There were also substantial differences in texture between the two soils, especially a clay content of 27.7 % vs. 5.7 % (chernozem vs. sandy soil) (Table 3). NH₄⁺ can be immobilized by microbes (bacteria and fungi) in the soil and it can be adsorbed to soil particles to a certain extent. Wang and Alva (2000) found clay soils to be more capable to sorb NH₄⁺ than sandy soils. Pivato and Raga (2006) stated that NH₄⁺ was sorbed well to bentonite. Sorbed NH₄⁺ is immobilized and therefore could mitigate N₂O emissions. In our experiment, there was no evidence that the higher clay content in the chernozem had mitigating effects on N₂O emissions due to their sorption capacity, but the effect may have been overshadowed by e.g. the high N content. The assumption is corroborated by no effect of the bentonite.

CH₄ emissions were generally low and the effect of soil type on CH₄ emission was less clear as the treatments showed no pattern over time.

Separation and acidification

The D-l and D-l-s treatments had the second and third highest cumulative N₂O emissions during the first 14 days in both soils (Table 4). There was a statistically significant effect on N₂O emissions in both soils treated with D-l compared to the D treatment (CH: p=0.0141, Ss: p=0.0007). This result confirmed our

hypothesis that separation and acidification leads to higher emissions in the liquid phase. However, cumulative N₂O emissions from the D-s was not found to be significantly lower than for the unseparated D.

N₂O emissions on day 1 after application of D-l and D-l-s were lower than for D or DB. This could have indicated some inhibition of nitrification in the D-l and the D-l-s due to initial low pH (5.3 for D-l) caused by acidification after separation. Optimal nitrification rates in slurries are reported to take place at pH values between 7.5 and 8.0 (Tchobanoglous et al. 2014). At pH values in the range of 5.8 to 6.0, ammonia oxidation rates are only 10 to 20 percent of the rate at pH 7.0 (USEPA 1993). The CO₂ emissions within the first 3 days indicate a high microbial activity (Fig. 3 and Fig. 4) and the subsequent peaks (between first and 5th day) in N₂O emissions could be interpreted as a result of increasing nitrification (Albuquerque et al. 2012b) and denitrification after O₂ was depleted in the soil (Parkin 1987). D-l and D-l-s treatments contained largest amounts of NH₄⁺ (7 and 5.7 mg, resp.) which was probably nitrified and denitrified to a large extent within the first 3-6 days. These nitrification and denitrification processes lead to high N losses as N₂O. High N₂O emissions from liquid fraction after separation of the digestate were also observed in other studies (Aguilera et al. 2013).

Digestate addition resulted in an immediate but low N₂O production (D: maximum of 5.3 µg N₂O-N/h/kg soil after 0.5 days). In contrast, enhanced N₂O production was observed later for the treatments containing a liquid fraction of digestate (D-l and D-l-s: maximum of 41.5 and 31.5 µg N₂O-N/h/kg soil, respectively, after 3 days). However, these initial emissions from D only occurred in the chernozem, whereas in the sandy soil there was low release or even an uptake of these gases. High total carbon content of the soil itself leads to high microbial biomass. This relation is also reflected in the initially increased CO₂ production (Fig. 4). Soil content of NO₃⁻ and more easily available carbon in the chernozem could have increased N₂O emissions compared to sandy soil, which is similar to reported results of Möller and Stinner (2009) and Sosulski et al. (2017). Combined with the quick depletion of oxygen in the fine textured chernozem N₂O emissions may have been enhanced in microsites of the chernozem whereas the structure of the sandy soil was still providing a mainly aerobic environment and low NO₃⁻ contents. In addition, D contained slightly more nitrate than e.g. only the liquid fraction (D-l) of it, which

could be denitrified immediately after application, enhancing N₂O emissions (priming effect).

Effects of different origin of solid digestate

Two solid digestates of different origin (D-s vs. D-s-HRA) were compared. The sustained high N₂O emissions from the solid fraction of HRA (D-s-HRA) caused the highest emission over time in the chernozem, the second highest in the sandy soil and substantially higher emissions than the solid fraction from RBA (D-s) in both soils (Table 4). NH₄-N and total organic carbon values as well as the texture of the two solid products diverged greatly. This emphasizes the impact of different anaerobic processes and pre-treatments (e.g. mesophilic vs. thermophilic process) and separation processes on GHG emissions.

The amounts of NH₄-N, NO₃-N and TOC added by each treatment were rather different (Table 6 in Appendix). D-s-HRA had high inputs of NH₄-N and TOC. The nitrogen of D-s was organically bound to a larger extent and contained only 19 % of the NH₄⁺ added by D-s-HRA treatment. O₂ concentration might have been low due to high WFPS and also additional biological O₂ depletion by microbial respiration, both enhancing denitrification. The large reservoir of organic carbon and NH₄⁺ combined with low O₂ concentrations could have resulted in the long lasting, and especially in the chernozem, high emission rates of N₂O. This in turn lead to the highest cumulative N₂O emissions (14 days) of all treatments in both soils. D-s application resulted in long lasting N₂O emissions, too, but the cumulated N₂O values were only 21 % (sandy soil) and 19 % (chernozem) of the D-s-HRA emissions.

These results are in line with findings of Garcia-Ruiz and Baggs (2007) who stated that adding organic matter to the soil makes carbon available as substrate for denitrification and enhances N₂O emissions at low O₂ concentrations due to high WFPS.

Parkin (1987) found that hot-spots of high specific denitrification activity were associated with particulate organic carbon material in the soil. A more compact structure in D-s-HRA than in D-s may mean that organic carbon was exposed for longer periods, releasing more N₂O than the more porous D-s. Therefore, it is evident that treatment and soil characteristics like structure and available organic

carbon content are important influencing factors not only at the time of application but also up to 2 weeks after application.

Overall, the thermophilic vs. the mesophilic temperature also affects organic matter composition and degradability.

Furthermore, microorganisms from the mesophilic process at HRA might e.g. have been better adapted to temperatures during the experiment than the thermophilic process in RBA. This might have induced a quicker depletion of O₂.

Composting

Composted digestate gave only low N₂O emissions in the chernozem and even an N₂O uptake was observed in the sandy soil. D-s showed significantly higher CH₄ emissions than C in the sandy soil (p<0.0001). This indicates that the carbon in the compost was already stabilized and lower NH₄-N contents than in D-s (0.01 mg vs. 0.40 mg), which did not lead to enhanced N₂O emission anymore. Nevertheless, composting might not be the best GHG mitigation option for solid digestate, as during composting itself GHGs are released (Ermolaev et al. 2015; Boldrin et al. 2009; Amlinger and Peyr 2008; Lim et al. 2016). GHGs are not only produced by the microbial degradation process itself but also the energy and machinery used causes emissions. Further research is needed to get data on GHG emissions from compost processing and further usage to provide a basis for comparison of different digestates and composted digestate.

CO₂ emission rates

CO₂ emissions from treatments with substrate were much higher than all the others and higher in the chernozem soil than in the sandy soil (up to 15927 and 6199 µg CO₂-C/h/kg, resp.) (Data not shown). This is because the material is not already degraded like digested material and therefore it contains much more available carbon causing high CO₂ emissions. The treatments containing liquid digestate (D-l and D-l-s) showed higher emission rates than all others, except substrate. In comparison, C was always very close to the samples from SO and only showing low emission rates (Fig. 3). C had significantly lower CO₂ emissions than D-s (p<0.0001 for both soils).

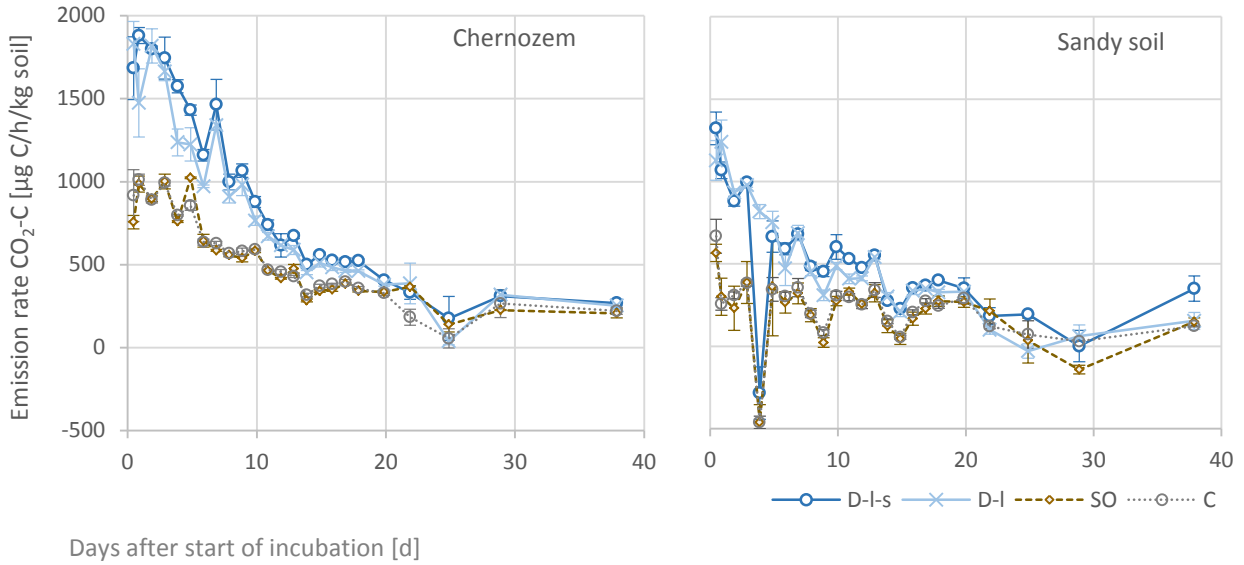


Fig. 3 Comparison of CO₂-C emission rates from chernozem after adding D-l, D-l-s, C or soil only (SO). Error bars indicate standard error (n=3)

Fig. 4 Shows the relation between CO₂-C and N₂O-N emission rates. The high CO₂-C release of D-l corresponds to a substantial N₂O-N release during the first days. On the contrary, the emissions of the two

gases are not correlated for the D treatment. The SO samples exhibited enhanced CO₂-emissions on a lower level during the first days without producing N₂O.

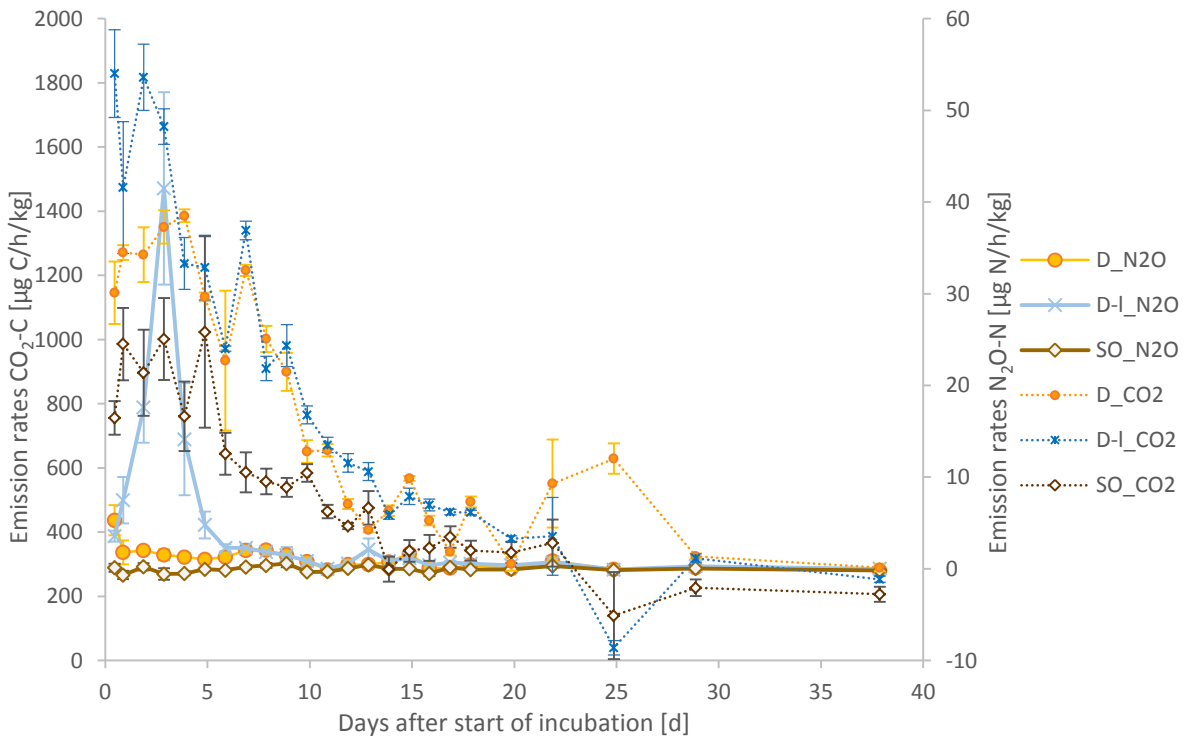


Fig. 4 Comparison of CO₂-C and N₂O-N emission rates from chernozem after adding D, D-l or SO. Error bars indicate standard error of means (n=3). Left y-axis corresponds to CO₂-C and right y-axis to N₂O-N emissions

There was no or little correlation between CO₂ and N₂O production rates directly visible in this experiment. Anaerobic denitrification is known to be high when respiratory consumption of O₂ exceeds the rate of replenishment within the soil (Smith et al. 2018). The release of large amounts of CO₂ from D, DB and SO was not necessarily accompanied by high N₂O emissions at the time (Fig. 4). This means that high CO₂ and microbial activity does not necessarily induce high N₂O emissions.

CH₄ emissions

The CH₄ emissions were low for all treatments and, especially in the beginning, negative. To compare effects of different processes (thermophilic vs. mesophilic) and to see a probable influence of methanogens originating from those digestates, CH₄ emissions of D-s, D-s-HRA and SO are shown in Fig. 5. The treatments containing D-s showed, particularly in the sandy soil, enhanced CH₄ emission compared to D-s-HRA during a long-time period.

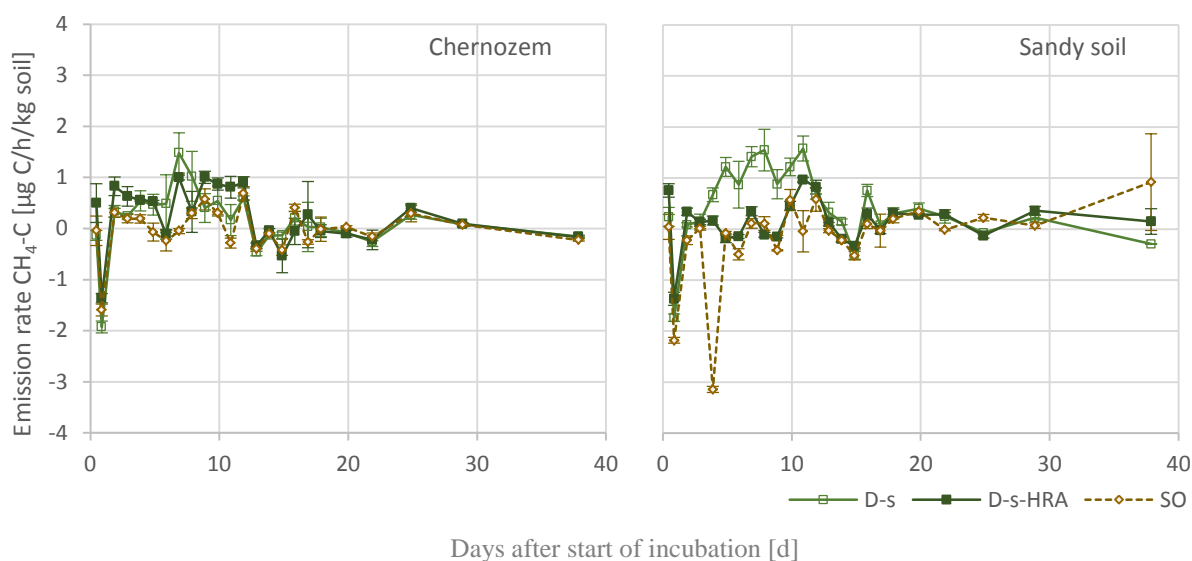


Fig. 5 Comparison of CH₄-C emission rates from chernozem (left) and sandy soil (right) after adding D-s and D-s-HRA or nothing (SO). Error bars indicate standard error (n=3)

Aerobic soils are usually sinks for CH₄ and only acting as a source when anaerobic conditions prevail (Topp and Pattey 1997; Ridgwell et al. 1999; IPCC 2007). CH₄ emissions varied on a low level and uptake and release were alternating for most of the treatments during the whole incubation (data not shown). This suggests that the anaerobic microbes that are added with the digestates stay alive for a considerable time and can start producing CH₄ whenever they find appropriate conditions. It was also observed that the D-s containing mesophilic bacteria from the RBA plant produced more CH₄ in the sandy soil than the D-s-HRA containing thermophilic bacteria from HRA. This indicates that the temperature the microbial community in the digestate is adapted to is important for emissions after application in the field.

Cumulative emissions

The cumulative GHG production was calculated for a time period of 14 days (corresponding to 333 h), the assumed period with minimal plant effects in the field, and for the whole experiment period of 38 days (corresponding to 909 h). The main results of the statistical model are given in Table 4 (measured data in Table 10 in Appendix). The influence of the different soil types on GHG production over 14 days was highly significant for all gases measured (CH₄ (p=0.0003), CO₂ and N₂O (p<0.0001 each)). This was also the case for the treatments (p<0.0001 for all 3 gases) and the interaction of soil and treatment (p<0.0001 for all 3 gases).

Again, the differences between treatments with and without bentonite were not significant (Table 4).

Table 4 Estimates of N₂O-N, CH₄-C, and CO₂-C emissions cumulated over the first 14 days (top) and the whole experimental period of 38 days (bottom) for both soil types (CH=chernozem and Ss=sandy soil) and all treatments: S=substrate, D=digestate, DB=digestate & bentonite, D-s=digestate-solid, D-s-l=digestate solid and liquid fraction, D-s-HRA= digestate solid from HRA, C=compost, SO=soil only). SE means Standard Error of the estimate of the Least Squares Means. Tukey Kramer Grouping (TK - same letter means not significantly different); outliers excluded

14 days										
Soil	Treatment	cum. N ₂ O-N [μg N/kg soil]	SE [μg N/kg soil]	TK (α<0.05)	cum. CH ₄ -C [μg C/kg soil]	SE [μg C/kg soil]	TK (α<0.05)	cum. CO ₂ -C [mg C/kg soil]	SE [mg C/kg soil]	TK (α<0.05)
CH	S	447.1	70.6	d,e,f	46.5	19.6	b,c,d	1555	16	a
	D	433.3	70.6	d,e,f	1.6	19.6	c,d	313	9	d
	DB	338.4	70.6	e,f,g	51.5	19.6	b,c,d	350	9	c,d
	D-s	580.6	70.6	d,e	50.1	24.0	b,c,d	379	9	c
	D-s-HRA	3146.8	122.3	a	149.8	19.6	a,b	317	9	d
	D-l	2398.4	122.3	b	18.2	19.6	c,d	357	9	c,d
	D-l-s	-	-	-	74.7	19.6	b,c	395	9	c
	C	58.0	70.6	f,g,h	-17.2	19.6	c,d,e	2	9	e
	SO	-16.4	70.6	g,h	7.3	19.6	c,d	219	9	e
Ss	S	-26.6	70.6	g,h	8.8	19.6	c,d	771	9	b
	D	201.2	70.6	f,g,h	-29.5	19.6	c,d,e	181	9	e,f
	DB	203.0	70.6	f,g,h	-46.2	19.6	d,e	179	9	e,f
	D-s	-	-	-	230.4	19.6	a	189	9	e,f
	D-s-HRA	979.1	70.6	c	49.0	19.6	b,c,d	157	9	f
	D-l	726.7	70.6	c,d	-4.6	19.6	c,d	211	9	e
	D-l-s	722.1	70.6	c,d	41.6	33.9	b,c,d	194	9	e,f
	C	-73.1	70.6	h	-109.5	19.6	e	78	9	g
	SO	-132.5	70.6	h	-112.2	19.6	e	72	9	g
38 days										
Soil	Treatment	cum. N ₂ O-N [μg N/kg soil]	SE [μg N/kg soil]	TK (α<0.05)	cum. CH ₄ -C [μg C/kg soil]	SE [μg N/kg soil]	TK (α<0.05)	cum. CO ₂ -C [mg C/kg soil]	SE [mg N/kg soil]	TK (α<0.05)
CH	S	1040.8	94.4	c,d,e	33.5	32.0	b,c	1949	22	a
	D	538.1	94.4	e,f,g	44.7	32.0	b,c	550	22	c
	DB	405.2	94.4	f,g,h	93.0	32.0	b,c	574	22	c
	D-s	720.8	94.4	d,e,f	104.5	32.0	b,c	598	22	c
	D-s-HRA	4470.8	115.6	a	151.8	32.0	a,b	529	22	c
	D-l	2593.8	163.5	b	5.5	32.0	b,c	535	22	c
	D-l-s	-	-	-	66.1	32.0	b,c	590	22	c
	C	60.9	94.4	g,h	-11.3	32.0	b,c	361	22	d
	SO	-43.0	94.4	h	2.7	32.0	b,c	368	22	d
Ss	S	304.2	94.4	f,g,h	-34.4	32.0	c	1071	22	b
	D	201.7	94.4	g,h	-46.4	32.0	c	299	22	d
	DB	245.0	94.4	f,g,h	-32.2	32.0	c	304	22	d
	D-s	326.1	94.4	f,g,h	282.3	32.0	a	298	22	d
	D-s-HRA	1219.9	94.4	c,d	148.7	32.0	a,b	255	22	d,e
	D-l	1277.3	94.4	c	-	-	-	293	22	d
	D-l-s	1127.4	115.6	c,d	-	-	-	317	22	d
	C	-70.3	94.4	h	-2.8	32.0	b,c	147	22	e,f
	SO	-81.1	94.4	h	-63.2	39.2	c	117	22	f

Both soils had cumulative N_2O uptake in SO for 14 days and the whole experimental period (Table 4). An N_2O uptake was also observed for S and C applications for the first 14 days in sandy soil, whereas in the chernozem they lead to emissions. D-s-HRA resulted in higher N_2O emissions than the D-s from RBA. The difference was higher in the chernozem than in the sandy soil. The reduction of N_2O emissions due to composting (D-s vs. C) was significant in chernozem and visible in the sandy soil. The differences in N_2O emissions from D and DB was not significant and the composted digestate C did not show significantly different N_2O emissions from SO in both soils. The low and stabilized organic carbon and NH_4-N content could be a reason for this. The D-s treatment resulted in significantly lower CH_4 emissions than the D-s-HRA in the sandy soil,

whereas in the chernozem the difference was not significant. S had the highest CO_2 emissions over 14 days in both soils. This was expected as the easily degradable carbon was still available in the undigested substrate, whereas in digested materials it has been used up e.g. for methane production. CO_2 emissions from D, D-l and D-l-s were significantly higher than from C or SO in both soils. TOC contents had a significant influence on CO_2 emissions over 38 days but cannot be seen as a predictor for CH_4 emissions.

The global warming potential (GWP) of N_2O and CH_4 emitted during the first 14 days are shown in Fig. 6. Especially cumulated emissions and their GWP showed that N_2O emissions of digestates applied to soil posed an environmental problem, whereas CH_4 emissions had a low impact in this experiment as expected.

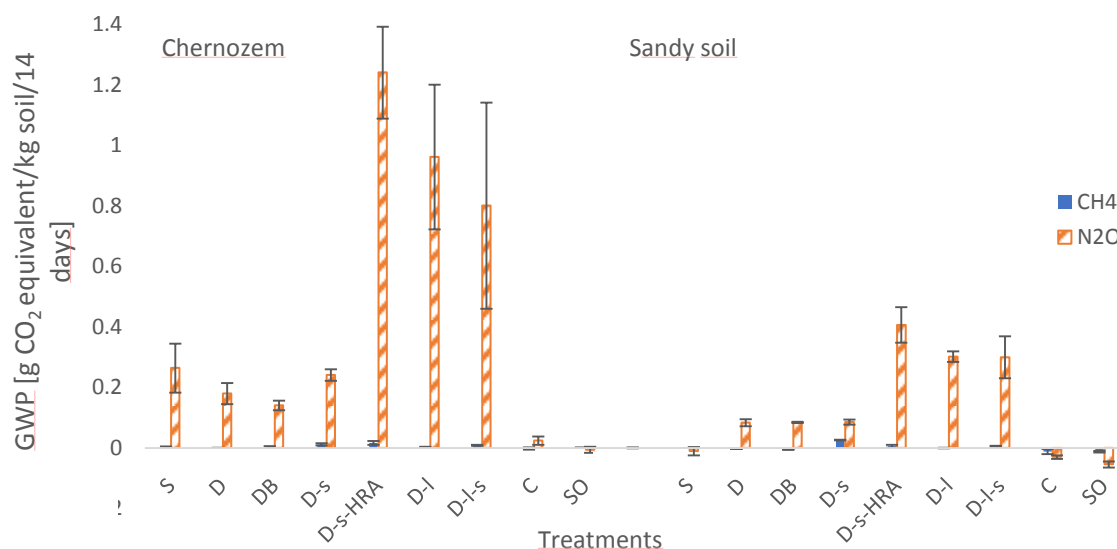


Fig. 6 GWP of mean cumulated CH_4 and NO_2 emissions from the chernozem (left) and the sandy soil (right) during the first 333 h of the experiment

Changes in pH, NO_3-N and NH_4-N contents in the soil during the experiment

The D-l caused the biggest decrease in pH (-2.1 in sandy soil and -1 in chernozem soil) during the entire incubation period of 38 days. This is not surprising as chernozem most probably had a higher buffering capacity. D with the highest pH (8.5) also resulted in a decreased pH in sandy soil after the incubation period (-0.3) (Table 5). This could be a result of acidification by nitrification, when H^+ is released during the transformation of NH_4^+ to NO_3^- . After the 38 days, a mineral N loss was observed for D-s-HRA, D-l and D-l-s in both soils, for C and SO only in sandy soil (Table 11 in Appendix).

The majority of the treatments had pH values in the alkaline range, but S and D-l and D-l-s were acidic, (pH=3.9 and 5.3, resp.). Šimek and Cooper (2002) reported in their research review that in pH below 7, the total N gas emission would be decreased. In contrast to this, in our study the acidic D-l and D-l-s treatment showed a peak in N_2O emission rates during the first 4 to 5 days, whereas emissions from e.g. D (pH=8.5) were lower. The enhanced emissions from D-l and D-l-s could indicate that N_2O was the main product of denitrification at low pH as reported by Morkved et al (2010).

Table 5 pH of soils and the treatment materials before addition to the soil, and all samples at the end of the incubation

Soils	pH		
	Before incubation	After incubation	
Chernozem	6.2	6.6	7.1
Sandy soil	6.8	6.7	6.7
Treatments	Before incubation	Chernozem	Sandy soil
Substrate <i>S</i>	3.9	6.6	7.1
Digestate <i>D</i>	8.5	5.9	6.7
Digestate & bentonite <i>DB</i>	8.5 7-7.5	5.9	6.7
Digestate solid <i>D-s</i>	8.8	6.1	7
Digestate solid HRA <i>D-s-HRA</i>	8.9	6.2	7.2
Digestate liquid <i>D-l</i>	5.3	5.2	4.7
Digestate liquid & solid <i>D-l-s</i>	5.3 8.8	5.2	5
Compost <i>C</i>	7.8	6.1	6.7
Soil only <i>SO</i>		6.1	6.8

The multiple regression showed that N_2O emissions are significantly increasing with higher NH_4 contents in the mixture of soil and treatment over 14 and 38 days. Cumulated N_2O -N emissions (14 days) and amount of NH_4 -N application were positively correlated in material from the RBA plant (sandy soil: $R^2 = 0.90$; chernozem: $R^2 = 0.78$) (Fig. 7). This result is supported by findings of Möller and Stinner (2009),

suggesting that a higher supply of readily available NH_4 -N was the main driving force for N_2O emission. N_2O emissions from sandy soil were, especially at high NH_4 -N contents, lower than from chernozem. Material from the HRA plant was different, D-s-HRA contained only slightly over 2 mg NH_4 -N but resulted in comparatively high N_2O -N emissions.

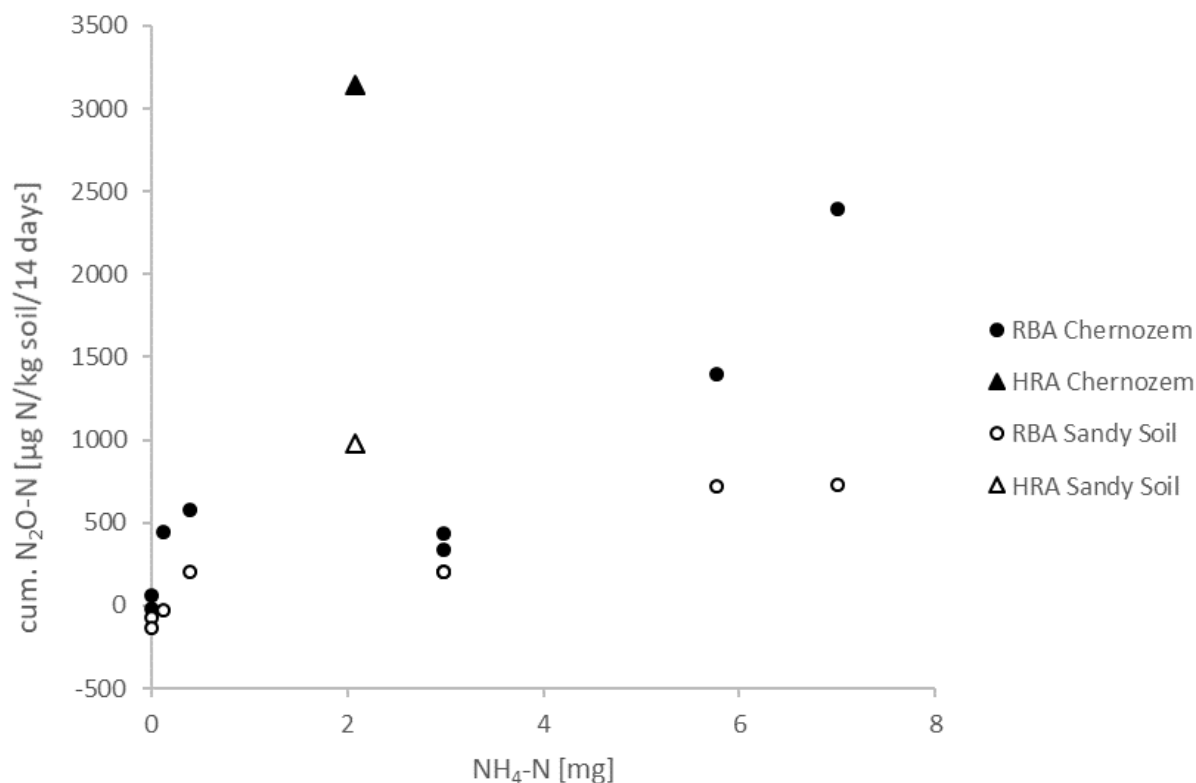


Fig. 7 Correlation between cumulative N_2O -N emissions over 14 days and added mass of NH_4 -N in treatments originating from RBA (circle) and HRA (triangle) in chernozem (filled) and the sandy soil (empty)

Conclusion

Finding GHG mitigation options for digestate application to soil should focus on N₂O emissions while CH₄ emissions were found to be negligible. The fact that we observed some CH₄ emissions suggests that microbes from the digestate survived for a long time in aerobic soil. High N₂O emission rates were detected for 5 treatments: Sustained high emissions from soil treated with the compact solid fraction from HRA (D-s-HRA), peak emissions from treatments containing liquid fraction of digestate (D-l and D-l-s) and initial emissions right after application of unseparated digestate (D and DB) to chernozem (ranking in decreasing importance).

The high N₂O emissions of D-s-HRA emphasizes the influence of different AD and separation processes of the two different plants. The separation process, including concentration and acidification of digestate, are not only energy intensive, but also trigger additional GHG emissions. The benefit of acidification reducing NH₃ emissions during storage of liquid digestate should be compared to additional emissions from the soil. The bentonite addition did not result in a significant difference in N₂O emissions, possibly it was added to the digestate right before soil application. Composted digestate had no or insignificant emissions when applied to soil, but potential emissions during composting were not addressed in this study.

NH₄-N content was found to be an important factor of emission potential as well as the type of anaerobic digestion process. The emission peaks occurred at the same time and had similar shapes in the 2 soils, but the peaks in the sandy soil were much smaller than in the

chernozem. The sandy soil was found to emit less GHGs and therefore, to be better suited for the digestate treatments than the chernozem regarding only N₂O and CH₄ emissions. For a complete and comprehensive picture, one would have to, for example, include also NH₃ emissions, nitrogen use efficiency, etc.

Further research should focus on understanding the processes that trigger N₂O emissions after application of digestate, including emissions of N₂ and NO. In addition, focus on application strategies and other measures to maximize quick plant uptake of N in digestate could be ways to reduce emissions of GHG emissions and optimize plant utilization of nutrients in digestate.

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Compliance with ethical standards

Conflict of interest The authors declare that there are no conflicts of interest associated with this study.

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Appendix

Table 6 Mass of added treatments, bentonite and DI water (m_t , $m_{\text{bentonite}}$ and m_w resp.) and added mass of total org. carbon (TOC), total nitrogen-Kjeldahl mod. (N_{tot}), NH₄-N and NO₃-N per bottle

Treatment	m_t [g]	$m_{\text{bentonite}}$ [g]	m_w [g]	TOC added [mg]	N_{tot} added [mg]	NH ₄ -N added [mg]	NO ₃ -N added [mg]
S	3.41		11.82	148.19	8.20	0.12	0.01
D	2.81		12.21	35.33	8.20	2.98	0.03
DB	2.81	0.31	12.21	35.33	8.20	2.98	0.03
D-s	0.46		14.60	56.62	8.20	0.40	0.00
D-s-HRA	0.95		14.21	83.77	8.20	2.08	0.00
D-l	1.22		13.84	42.33	8.20	7.01	0.01
D-l-s	1.08		14.20	44.99	8.20	5.77	0.01
C	0.79		14.63	78.14	8.20	0.01	0.51

Table 7 Characteristics of crude bentonite added to digestate

Particle size	0 - 300	mm
Water content	25 - 32	%
Specific gravity	2.5 - 2.6	t/m ³
Loose bulk density	1.2 - 1.3	t/m ³
Solid volume weight	1.6 - 1.65	t/m ³
pH	7 - 7.5	
Mohs hardness	1	
Ion exchange capacity		
Ca	0.25 - 0.6	meq/g
Na	0.04 - 0.06	meq/g
K	0.01 - 0.02	meq/g

Table 8 Grain size distribution (weight %) of chernozem and sandy soil. Standard error (n=3) in brackets

Grain size [mm]	Chernozem [%]	Sandy soil [%]
0.6-2.0	0.1 (0.03)	0.0 (0.01)
0.2-0.6	0.7 (0.02)	18.4 (0.27)
0.1-0.2	5.3 (0.09)	62.6 (0.21)
0.06-0.1	5.6 (0.08)	11.1 (0.42)
0.02-0.06	33.3 (0.57)	1.1 (0.12)
0.006-0.02	20.2 (0.59)	0.7 (0.22)
0.002-0.006	7.1 (0.1)	0.4 (0.13)
<0.002	27.6 (0.07)	5.7 (0.17)

Table 9 Time schedule of the 24 measurements and additional treatments

Measurement	Hours after incubation [h]	Date	Additional treatment
1	11	5.4.	
2	21	5.4.	
3	45	6.4.	
4	69	7.4.	
5	93	8.4.	
6	117	9.4.	
7	141	10.4.	
8	165	11.4.	
9	189	12.4.	
10	213	13.4.	
11	237	14.4.	
12	261	15.4.	
13	285	16.4.	
14	309	17.4.	
15	333	18.4.	
16	357	19.4.	
17	380.5	20.4.	
18	405	21.4.	
19	429	22.4.	
20	476.7	24.4.	
21	525	26.4.	+2ml DI water
22	597	29.4.	
23	693	3.5.	
		5.5.	Filling up to 60 % WFPS
24	909	12.5.	End of incubation

Table 10 Measurements of N₂O-N, CH₄-C and CO₂-C emissions cumulated over the first 14 days for both soil types (CH=chernozem and Ss=sandy soil) and all treatments: S=substrate, D=digestate, DB=digestate & bentonite, D-s=digestate-solid, D-s-l=digestate solid and liquid fraction, D-s-HRA= digestate solid from HRA, C=compost, SO=soil only). SE means Standard Error of the measurements of the Least Squares Means and Tukey Kramer Grouping (TK – same letter means not significantly different); including outliers

		14 days								
Soil	Treat- ment	cum. N ₂ O-N [μg N/kg soil]	SE [μg N/kg soil]	TK (α<0.05)	cum. CH ₄ -C [μg C/kg soil]	SE [μg C/kg soil]	TK (α<0.05)	cum. CO ₂ -C [mg C/kg soil]	SE [mg C/kg soil]	TK (α<0.05)
CH	S	635.9	195.6	c,d	46.5	7.3	b,c	1530	52	a
	D	433.2	84.1	d	1.6	2.2	b,c,d	313	5	d
	DB	338.4	37.5	d	51.5	30.5	b,c	350	13	c,d
	D-s	580.6	46.3	c,d	103.8	56.7	a,b,c	379	12	c,d
	D-s- HRA	2990.2	366.1	a	149.8	18.3	a,b	317	4	c,d
	D-l	2318.1	577.5	a,b	18.2	12.5	b,c,d	357	14	c,d
	D-l-s	1930.3	821.0	a,b,c	74.7	32.4	b,c	395	12	c,d
	C	58.0	33.7	d	-17.2	12.0	c,d	220	3	e
	SO	-16.4	24.3	d	7.3	12.1	b,c,d	219	21	e
Ss	S	-26.6	32.3	d	8.8	6.1	b,c,d	771	11	b
	D	201.2	28.3	d	-29.5	3.4	c,d	181	4	e
	DB	203.0	5.7	d	-46.2	9.0	c,d	179	1	e
	D-s	205.2	20.5	d	230.4	42.9	a	189	4	e
	D-s- HRA	979.1	141.3	b,c,d	49.0	7.4	b,c	157	10	e,f
	D-l	726.6	41.6	c,d	-4.6	24.7	b,c,d	211	5	e
	D-l-s	722.1	167.3	c,d	37.2	77.7	b,c,d	194	5	e
	C	-73.1	14.3	d	-109.5	19.8	d	78	4	f,g
	SO	-132.4	23.3	d	-112.2	11.5	d	72	3	g

Table 11 NO₃-N and NH₄-N of soils and the treatment materials together with soil before and after the incubation

Soils	Before incubation							
	NO ₃ -N		NH ₄ -N					
	[mg/kg soil]							
Chernozem	1.83		1.08					
Sandy soil	0.54		0.21					
Treatments with soils	Before incubation				After incubation			
	Sandy soil		Chernozem		Sandy soil		Chernozem	
	NO ₃ -N		NH ₄ -N		NO ₃ -N		NH ₄ -N	
	[mg/kg soil]							
Substrate S	0.55	0.33	1.85	1.20	1.73	0.15	3.03	0.26
Digestate D	0.56	3.19	1.86	4.06	4.75	0.05	6.73	0.12
Digestate & bentonite DB	0.56	3.19	1.86	4.06	4.58	0.04	6.71	0.14
Digestate solid D-s	0.54	0.60	1.83	1.48	1.51	0.08	4.42	0.15
Digestate solid HRA D-s-HRA	0.54	2.29	1.83	3.16	1.47	0.06	2.71	0.11
Digestate liquid D-l	0.55	7.22	1.85	8.09	5.34	0.39	9.03	0.25
Digestate liquid & solid D-l-s	0.54	5.98	1.83	6.85	4.84	0.19	8.10	0.21
Compost C	1.05	0.22	2.35	1.09	1.52	0.05	3.16	0.10
Soil only SO	0.54	0.21	1.83	1.08	1.02	0.04	2.56	0.09

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