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Greenhouse gas emissions from a sandy loam soil amended with digestate-derived nitrogen fertilisers – a microcosm study
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Abstract:	Nutrient recovery from anaerobic digestion systems provides several side streams that are useful as biobased fertilisers (BBFs). A microcosm approach was employed to assess the short-term greenhouse gas emissions from a sandy-loam soil enriched with 18 BBFs in comparison with mineral fertilisers (urea and calcium ammonium nitrate). In total, 20 different fertilisers were homogeneously incorporated into an arable sandy loam soil at a rate of 170 kg nitrogen (N) ha $^{-1}$ and incubated at 80% water-filled pore space. Over 18 days, the fluxes of nitrous oxide (N_2O), methane (CH_4), and carbon dioxide (CO_2) released in the headspace of the microcosms were measured using a Gasera One Multi-gas analyser. Cumulative N_2O emissions from the BBF treatments were either comparable or lower (0.04 – 0.09 %N applied) than the mineral fertilisers (0.10 – 0.14 %N applied). Nitrification of the initial ammonium-N present in the BBFs was likely the dominant biological process driving N_2O production. The application of digestate and evaporator concentrates led to an increase in CO_2 emissions (8–51% of applied carbon (C)), mostly in the first days of the incubation. Meanwhile, the solid fraction of digestate exhibited slow mineralisation patterns (3–7 % of applied C). The variability in CO_2 respiration was strongly influenced by the availability of labile C. Fertilisation had no effect on soil-borne CH_4 emissions. Estimation of global warming potential, with respect to added N, suggests that BBFs obtained from the post-digestion treatment of digestate have a lower environmental impact compared to the unprocessed digestate due to lower N_2O emissions.
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Professor Judith Ascher-Jenull
Editor-in-Chief
Agriculture, Ecosystems & Environment

17 March, 2022

Dear Professor Ascher-Jenull,

I am pleased to submit an original research article entitled "**Greenhouse gas emissions from a sandy loam soil amended with digestate-derived nitrogen fertilisers – a microcosm study**" for consideration for publication in the *Applied Soil Ecology* Journal.

In our study, we assessed and compared the short-term emissions of N_2O, CO_2, and CH_4 from a sandy-loam soil enriched with 18 different fertilisers obtained from treated and untreated digestates. We adopted a microcosm approach to closely monitor and quantify the fertilisation effects on soil GHG emissions and C and N turnover processes. Our results showed that the differences in fertiliser characteristics, linked to the types of digestate treatment techniques, can influence the soil N_2O and CO_2 emissions after fertilisation. In short, our findings provide a first look at the GHG emission potential of several biobased fertilisers derived from novel digestate processing techniques.

We believe that this manuscript is appropriate for publication in *Applied Soil Ecology* because it discusses the potential effects of digestate fertilisation on environmental quality and the dynamics of nitrogen transformations in soils.

This manuscript has not been published and is not under consideration for publication elsewhere.

Thank you for your kind consideration.

Sincerely,

Caleb Elijah Egene, MSc.
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Highlights

- Nitrification was the dominant biological process driving N_2O production.

- CO_2 respiration was strongly influenced by the availability of labile C.

- Fertilisation had no effect on soil-borne CH_4 emissions

- Global warming potential of biobased fertilisers did not exceed that of mineral fertilisers.

1   Greenhouse gas emissions from a sandy loam soil amended with digestate-derived nitrogen

2   fertilisers – a microcosm study

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13

Abstract

Nutrient recovery from anaerobic digestion systems provides several side streams that are useful as biobased fertilisers (BBFs). A microcosm approach was employed to assess the short-term greenhouse gas emissions from a sandy-loam soil enriched with 18 BBFs in comparison with mineral fertilisers (urea and calcium ammonium nitrate). In total, 20 different fertilisers were homogeneously incorporated into an arable sandy loam soil at a rate of 170 kg nitrogen (N) ha^{-1} and incubated at 80% water-filled pore space. Over 18 days, the fluxes of nitrous oxide (N_2O), methane (CH_4), and carbon dioxide (CO_2) released in the headspace of the microcosms were measured using a Gasera One Multi-gas analyser. Cumulative N_2O emissions from the BBF treatments were either comparable or lower (0.04 – 0.09 %N applied) than the mineral fertilisers (0.10 – 0.14 %N applied). Nitrification of the initial ammonium-N present in the BBFs was likely the dominant biological process driving N_2O production. The application of digestate and evaporator concentrates led to an increase in CO_2 emissions (8–51% of applied carbon (C)), mostly in the first days of the incubation. Meanwhile, the solid fraction of digestate exhibited slow mineralisation patterns (3–7 % of applied C). The variability in CO_2 respiration was strongly influenced by the availability of labile C. Fertilisation had no effect on soil-borne CH_4 emissions. Estimation of global warming potential, with respect to added N, suggests that BBFs obtained from the post-digestion treatment of digestate have a lower environmental impact compared to the unprocessed digestate due to lower N_2O emissions.

Keywords: biobased fertilisers, digestate, N_2O emission, microcosm, global warming potential

1. Introduction

Anaerobic digestion (AD) of organic wastes enables the production of biogas as a renewable energy carrier and provides a promising alternative to the use of fossil fuels (Holm-Nielsen et al., 2009). By converting food and animal wastes to energy, AD plays a significant role in reducing greenhouse gas (GHG) emissions from conventional energy sources. In addition, evidence suggests that the digestate, which is the residual matter after AD, may be useful as a fertiliser or soil improver because it retains most of the plant nutrients contained in the initial feedstock (Möller, 2009). Therefore, its use in agriculture is expected to improve soil fertility while reducing farmers' input of fossil-dependent mineral fertilisers.

The increase in soil available nitrogen (N) from the application of N fertilisers enhances nitrification and denitrification, which are soil processes that release nitrous oxide (N_2O) into the atmosphere (IPCC, 2006; Senbayram et al., 2010). N_2O is critical from the climate perspective because it is an ozone-depleting gas, with a global warming potential (GWP) 296 times that of carbon dioxide (CO_2) and 12 times that of methane (CH_4) in a 100-year time horizon (IPCC, 2007). Recent data revealed that transformations of N in agricultural soils by nitrifying and denitrifying microorganisms account for approximately 70% of the annual N_2O budget globally (Tian et al., 2019). This represents a major increase compared to the 50% contribution estimated in the early nineties (Bouwman, 1990). Therefore, for digestate-derived bio-based fertilisers (BBFs) to be used sustainably in agriculture as N fertilisers, it is necessary to ensure that their environmental impact is minimal. Moreover, fertiliser N lost from soils in gaseous form (N_2O, nitric oxide, and dinitrogen) reduces the fertiliser efficiency (Holland and Weitz, 2003).

Besides N availability, other factors that influence the magnitude of N_2O emission from soil include moisture content, pH, and carbon (C) supply (Akiyama et al., 2005; Dalal et al., 2003). In particular, the soil moisture content plays a crucial role as it impacts soil

aeration, diffusion of dissolved nutrients, and the rate of microbial activities (Hütsch et al., 1999). Nitrification by heterotrophic microorganisms proceeds under sufficiently aerobic conditions while heterotrophic denitrification increases as oxygen availability become restricted (Hütsch et al., 1999; Le Roux et al., 2013). The addition of organic substrates with high organic C concentration is likely to favour denitrification since heterotrophic denitrifiers utilize organic C as electron donors (Hattori, 1983). Digestate, however, have lower availability of labile C since a large fraction of the easily degradable organic matter in the feedstock is consumed during AD (Alburquerque et al., 2012). This partly explains why some studies found that N_2O emissions from the soil after the application of digestate are generally lower than those after application of untreated feedstock (Cayuela et al., 2010; Johansen et al., 2013; Möller, 2009). Differences in N_2O emission rates from those studies were related to the form of N in the digestate (mineral N or organic N) as well as the quantity and quality of organic C.

Bodelier and Laanbroek (2004) demonstrated that the application of N fertiliser could indirectly stimulate the oxidation of CH_4 because the increase in available N increases the activity of methanotrophic and nitrifying bacteria in the soil. Existing literature on CH_4 emissions after digestate fertilisation is often focused on paddy fields since they are the main sources of CH_4 from soils (Odlare et al., 2012; Singla and Inubushi, 2014; Walling and Vaneeckhaute, 2020). In the few studies that have explored CH_4 emission dynamics in digestate-fertilised upland soils, emission rates were related to the soil moisture content and the availability of labile C (Czubaszek and Wysocka-Czubaszek, 2018; Pezzolla et al., 2012; Rosace et al., 2020). As such, organic C mineralization into CO_2 and CH_4 is useful as a measure of soil microbial activity and the intensity of N transformations (Liu et al., 2019; Shao et al., 2014).

In recent years, emphasis has been placed on nutrient recovery and reuse (NRR) technologies which enable the processing of digestate into concentrated end-products with high fertilisation value (Vaneeckhaute et al., 2017). At the basic refinement level, phase partitioning of digestate is carried out to obtain a phosphorus- (P) rich solid fraction (SF) and an N-rich liquid fraction (LF). The SF is sometimes subjected to drying or composting. Meanwhile, further processing of the LF using advanced NRR technologies such as N-stripping, vacuum evaporation, membrane filtration, among others, can deliver multiple cascades of nutrient-rich BBFs (Brienza et al., 2021; Logan and Visvanathan, 2019). Examples of such BBFs include ammonium sulphate solution (Brienza et al., 2020; Sigurnjak et al., 2019), ammonia water (Jamaludin et al., 2018; Vondra et al., 2019), evaporator concentrate (Vondra et al., 2019), reverse osmosis concentrate and permeate water (mineral Logan and Visvanathan, 2019). Based on the compositional differences of these novel BBFs, compared to the unprocessed digestate, it can be assumed that their behaviour as fertilisers could differ considerably. While a few studies have investigated the soil fertilisation potential of these novel BBFs (Ehlert et al., 2019; Sigurnjak et al., 2017), there is a paucity of data on their GHG emission potential when applied to the soil.

To close this knowledge gap, the short-term and long-term effects of the application of digestate-derived BBFs on soil GHG production ought to be evaluated. In this context, this work aimed to evaluate and compare the short-term N_2O, CO_2 and CH_4 emissions from a sandy-loam soil fertilised with (i) untreated anaerobic digestate (ii) BBFs derived from different post-digestion treatment processes of digestate (iii) two mineral N fertilisers – urea and calcium ammonium nitrate (CAN). It has been previously shown that soil processes such as respiration, nitrification, and denitrification respond sensitively to C and N availability as well as changes in redox conditions (Le Roux et al., 2013). Therefore, it is predicted that the differences in the forms and availabilities of N and C in the digestate-derived BBFs will

influence the production of GHGs after their addition to soil. We hypothesized that the short-

term emissions of N_2O, CO_2, and CH_4 from different BBF-amended soils are variable and this

variability is due to differences in concentrations of ammonium-N (NH_4^+-N) and labile C.

2. Materials & Methods

2.1 Origin of the biobased fertilisers

Digestate samples were obtained from five full-scale AD plants. The digestates were

treated to produce thirteen different biobased fertilisers, depending on the type of NRR

technology present at each plant (Table 1).

Groot Zevert Vergisting (GZV, Beltrum, the Netherlands) operates a mesophilic (~35

ºC) AD plant that is fed with pig slurry (81% w/w) and residue from the agro-food industry

(a.o. grain and rice husk, potato skins and coffee grounds, 19% w/w). A decanter centrifuge is

used to dewater the digestate (D-GZV) to obtain the solid fraction (SF-GZV). Part of the SF is

further processed in an installation to remove and recover P which includes washing the SF

with water and sulphuric acid to obtain an SF with a reduced P content and a fibrous structure

(SF-GZV$_{P\text{-poor}}$). The N-rich LF is treated through micro-filtration followed by reverse osmosis

to produce an N-potassium (K)-rich concentrate (RO-GZV) and permeate water.

Am-Power (AmP, Pittem, Belgium) treats residues from the food processing industry

and source-segregated food waste via thermophilic (~55 ºC) AD with a retention time of

about 60 days. The digestate (D-AmP) from this installation is dewatered to obtain a solid

fraction (SF-AmP). The solid fraction is passed through a fluidized bed dryer at 60°C to

obtain a dried solid fraction (SFD-AmP). The LF of digestate is sent to a vacuum evaporator

to evaporate water, leaving behind an evaporator concentrate rich in N and K (E-AmP).

Ammonium-rich condensed water from the vacuum evaporator is passed through a reverse

osmosis unit resulting in a reverse osmosis concentrate (RO-AmP) and permeate water.

6

137   Waterleau NewEnergy (WNE, Ieper, Belgium) operates an AD plant used for the

138   mesophilic (~35 ºC) digestion of residues from agro-industry (potatoes and grain, 40% w/w),

139   sludge from industrial wastewater treatment plants (15% w/w) and animal manure (45%

140   w/w). Digestate (D-WNE) is passed through a decanter to obtain a SF which is then dried in a

141   Hydrogone dryer to obtain a solid organic fertiliser (SFD-WNE). The evaporated water from

142   the dryer, together with the LF of digestate (LF-WNE) is treated in a biological aerobic

143   reactor to reduce the chemical oxygen demand. Ammonium is then transferred to the gas

144   phase via evaporation resulting in a K-rich concentrate (E-WNE). The ammonia-rich gas is

145   condensed with water vapour and condensed ammonia water (AW-WNE) is recovered.

146   Benas (BNS, Ottersberg, Germany) treats energy crops (silage maize, silage rye and

147   corn, 85% w/w) and poultry litter (15% w/w) using thermophilic digestion. The ammonia

148   content of a side stream of digestate (D-BNS) is lowered using a modified stripping process to

149   obtain ammonium nitrogen to which gypsum is added to produce a concentrated marketable

150   ammonium sulphate solution (AS-BNS).

151   Acqua & Sole (A&S, Vellezzo Bellini, Italy) processes sewage sludge from

152   wastewater treatment plants (86% w/w) and coproducts (digestate from anaerobic treatment of

153   source-segregated domestic food waste, 14% w/w) in a thermophilic (~55 ºC) AD. The

154   process is equipped with an ammonia stripping unit, whereby biogas acts as a stripping agent.

155   Ammonia is extracted from biogas by adding acid (H_2SO_4) resulting in an inorganic

156   ammonium sulphate solution (AS-A&S). The digestate from the plant is denoted as D-A&S.

157   GZV, AmP, WNE, BNS, and A&S are demonstration plants within the EU project

158   SYSTEMIC and more information is available on www.systemicproject.eu and in Brienza et

159   al. (2021).

2.2 Soil characteristics

The soil samples for the incubation were collected from the top layer (0–25 cm) of an arable field in Bottelare, Belgium. Composite samples of the sandy-loam (55% sand, 6% clay, 39% silt) soil was taken in April 2020 with the following characteristics: pH-$H_2$0 7.5, total N (TN) 0.76 g/kg dry weight (DW), total carbon (TC) 10.4 g/kg DW. This soil was used in the incubation experiments with BBFs from GZV and AmP. Soil was taken again from the same location in April 2022 having a slightly different composition (pH-$H_2$0 7.11, TN 1.30 g/kg DW, TC 12.8 g/kg DW) and used for the incubation experiments with BBFs from WNE, BNS, and A&S. The soils were air-dried until constant mass, sieved (2 mm) then stored in a cool dry room before being used in the experiments.

2.3 Analytical methods for fertiliser characterisation

The dry matter (DM) content was determined by drying to constant weight (48 h) at 80 °C and was calculated as a percentage of wet weight. Organic matter (OM) was measured on dried solids by incineration at 550 °C in a muffle furnace for 4 h (Dean, 1974; Santisteban et al., 2004). TC was determined using a PRIMACS100 Analyzer series (Skalar B.V., Netherlands). TN was determined using the Kjeldahl destruction method (EN13654-1, 2002). Ammonium-N (NH_4^+-N) was determined spectrophotometrically after 1M KCl extraction at a sample to solution ratio of 1:10. For the determination of dissolved organic carbon (DOC) and total dissolved nitrogen (TDN) in the BBFs, fresh samples were weighed into a 50 cm^3 centrifuge flask and extracted with 0.01 M $CaCl_2$ (sample to solution ratio of 1:10) by shaking for 2 h followed by centrifugation for 10 min at 3000 rpm (Houba et al., 1990). TDN and DOC in the extracts were measured using the Dumas Dry Combustion Method for TN and TC content (Bertsch and Ostinelli, 2019).

2.4 Microcosm setup

The incubation experiments were conducted in soil microcosms which enable studying the effects of amendment addition on soil respiration under controlled conditions. Each microcosm consisted of a 1L Duran bottle adapted with a GL45-thread Smart Cap (model: SW45-2A). The smart cap has two 2 mm threaded openings that can either be closed with blind plugs or fitted with valves that enable gas sampling. The incubation experiments were carried out in four batches from February to June 2021 at a mean room temperature of 20 ºC (diurnal temperature range: 18.5 – 21.5 ºC). Each batch included four or five BBFs from the demonstration plants, one blank (unfertilized control soil), and two mineral fertilisers (urea and CAN as positive controls). The same urea (46% urea N; Yara Benelux B.V.) and CAN (30% N) was used in all four incubation batches.

The soil was pre-incubated for one week at 40% water-filled pore space (WFPS) to activate the soil microorganisms. Next, 568 g of pre-incubated soil was thoroughly mixed with a biobased or mineral fertiliser in a steel bowl and then transferred into the microcosm. Prior to mixing, the mineral fertiliser granules were ground to <0.5 mm. The soil-fertiliser mixtures were carefully packed to attain an equivalent bulk density of 1.3 kg m^{-3}. All fertilisers were applied at a rate of 170 kg N ha^{-1}. The equivalent amounts of NH_4^+-N, organic N (N_{org}), TC applied in each treatment are summarized in Table 2. The moisture content in each bottle was brought to 80% WFPS and maintained throughout the experiment according to Cayuela et al. (2010). A 2 mm opening at the top of the microcosm was left uncovered to allow for aerobic respiration. The microcosms were laid out in a randomised block design with three replicates per treatment.

2.5 Measurement

Over an incubation period of 18 days, emissions of CO_2, N_2O, and CH_4 were measured using the Gasera One Multi-gas analyser (Turku, Finland) equipped with a photo-acoustic infrared analyser. Measurements were performed on days 0, 1, 2, 4, 7, 9, 11, 14, 16, 18. Measurements on day 0 were taken on average 2.5 hours after fertiliser incorporation. The analyser was connected to the microcosm using two 1 m-long non-reactive Teflon tubes with a 2 mm internal diameter. During measurement, the gases were pumped out from the headspace (at 800 mL min^{-1} flow rate), passed through the analyser then returned to the microcosm in a closed loop. This method ensures non-intrusive sampling and reduces the risk of systematic errors. The gas concentration in the headspace of the microcosms was measured at 4, 8, 12, and 16 minutes after connecting the tubes to the microcosm. During each 4 min time step, the analyser detected the change in concentration of the measured gases. Fluxes of CO_2, N_2O, and CH_4 were then calculated from the change in concentration over time considering the volume of the headspace, tubing, and area of the soil surface. Sample sets with a linear regression value of $R^2 < 0.90$ were rejected. At the end of the incubation period, soil mineral N (NH_4^+-N and NO_3^--N) in each treatment was analysed in a 1:10 ($w\ v^{-1}$) suspension of soil and 1 M KCl and shaken end-over-end for 30 min. The extracts were filtered (Whatman No. 45) and analysed for their NH_4^+-N and NO_3^--N contents with a continuous flow auto-analyser (Chemlab System 4, Skalar, the Netherlands).

2.6 Calculations

Gas concentrations measured in ppm were converted to emission flux using the ideal gas law according to Equation (1) (Comeau et al., 2018):

$$Flux = \frac{\Delta gas}{\Delta t} \times \frac{P \times M \times n}{R \times T} \times \frac{V}{A} \qquad \text{Equation (1)}$$

229   Where flux is the elemental flux which is released as gas in µg m^{-2} h^{-1}; $\frac{\Delta gas}{\Delta t}$ is the

230 slope of the linear regression of gas concentration (ppm) vs. time (h); P is the sampling

231 pressure of the device (0.838 atm); M is the elemental molar mass (e.g. 12 for C, 14 for N); n

232 is the number of atoms of the element in the gas (e.g. 2N in N_2O); R is the ideal gas constant

233 (0.08206 L atm mol^{-1} K^{-1}); T is the average atmospheric temperature (294 K); V is the sum

234 volume of the headspace, tubing, and analyser cell (0.623 L); A is the surface area of soil in

235 the microcosm (0.0069 m^2).

236   The cumulative flux for each gas was calculated using a linear interpolation between

237 two consecutive measurement days (Cai et al., 2013). Cumulative fluxes obtained with the

238 soil control were subtracted for all cumulative fertiliser emissions.

239   Net N release (N_{rel}) in the soil at the end of the incubation was calculated according to

240 Equation (2) (De Neve and Hofman, 1996):

241 $$N_{rel}\ (\%) = \frac{[(Mineral\ N_{fertiliser}) - (Mineral\ N_{control})]}{total\ N\ applied} \times 100 \qquad \text{Equation (2)}$$

242   where mineral $N_{fertiliser}$ is the soil mineral N content of the fertiliser treatment and

243 mineral $N_{control}$ is the soil mineral N content of the unfertilised control soil.

244 **2.7 Statistical analysis**

245   The data were subjected to one-way analysis of variance (ANOVA) and when

246 significant ($p < 0.05$), means were compared using Tukey's test. In addition, a principal

247 component analysis (PCA) was performed to evaluate the relationships between the

248 fertilisers' characteristics (pH, TN, NH_4, OM, TC, C/N, DOC, DOC/TC, TDN/TN) and

249 cumulative GHG emissions. PCA is an exploratory statistical tool that is used to quickly

250 visualize and analyse correlations between variables. Pearson's correlation analysis was

251 performed to test the relationships between product characteristics and cumulative GHG

252 emissions. All analyses were performed using XLSTAT 2021 software (Addinsoft, 2021).

253

3. Results

3.1 Composition of the biobased fertilisers

The studied BBFs differed considerably in their physicochemical composition (Table 3). The pH of most BBFs was alkaline (7.0 – 9.4) and any deviations from this range were attributed to the use of sulphuric acid during digestate processing, e.g., in SF-GZV$_{P-poor}$ with pH 5.5. On a fresh weight (FW) basis, OM constituted between 17 and 67% of the SFs of digestate. The low OM content (<3% FW) in the R, LF, AW, and AS fertilisers indicates these BBFs are dominantly mineral in nature and are, hereinafter, collectively referred to as organo-mineral BBFs. DM content in the whole digestate varied between 5.2% in D-WNE and 9.3% in D-A&S. For the organo-mineral BBFs, DM was below 5% except in the ammonium salt solutions with 21 and 38 %DM due to their high salt content. The large variation in DM content of the SFs of digestate (26 – 94%) was attributed to the type of dewatering equipment used and whether a drying step was included during processing.

Nitrogen was present mostly (>95%) in mineral form in the R, AW, and AS fertilisers. This, in addition to their high total N content, makes these BBFs comparable to CAN and urea in terms of potential N availability to crops. Unlike in AmP, the LF of digestate fed to the evaporator implemented at WNE is not acidified, resulting in a lower NH_4-N content in E-WNE, compared to E-AmP (Table 3). Organic N was the dominant fraction in E-WNE, SFD-AmP, SFD-WNE, and SF-GZV$_{P-poor}$, corresponding to 97%, 95%, 77%, and 73% of total N, respectively.

All five SFs of digestate had high TOC content, ranging from 85 g/kg in SF-AmP to 335 g kg^{-1} in SFD-WNE. Lower TOC levels were measured in the whole digestate (15 – 30 g/kg) while the organo-mineral BBFs were characterised by very low amounts of TOC (<5 g kg^{-1}). The variation in TN and TC content among the BBFs was reflected in the C/N ratio. As

278 expected, the C/N ratio was high in the SFs of digestate $(11 - 27)$, low in the whole digestate

279 and evaporator concentrates $(2.3 - 8.4)$, and less than 1 in the organo-mineral BBFs.

280   A higher relative concentration of labile C, indicated by the DOC/TC ratio, was

281 observed in D-GZV, D-WNE, D-BNS, and E-AmP, with 0.19, 0.33, 0.17, and 0.34,

282 respectively. Meanwhile, in the SF fertilisers, DOC constituted a much lower fraction of TC,

283 ranging between $0.01 - 0.03$. The TDN concentration of AW and AS fertilisers were not

284 analytically determined since N in those fertilisers is present almost entirely in dissolved form

285 as NH_4^+-N. Concentrations of TDN in the BBFs varied widely and ranged between 1.3 to 76 g

286 kg^{-1} FW. This variation was also reflected in the TDN/TN ratio.

287

288   **3.2 N_2O emission**

289   N_2O emission dynamics differed considerably among the different treatments.

290 However, the overall trend of N_2O fluxes indicated a gradual decrease towards background

291 levels (Figure 1), in agreement with studies by Cayuela et al. (2010) and Dietrich et al.

292 (2020). Daily mean N_2O fluxes ranged between 5 to 246 µg N_2O-N m^{-2} h^{-1} and peaked within

293 the first four days after fertiliser application in all treatments, except E-WNE which peaked on

294 day 11. Average daily N_2O emissions from the CAN and urea followed similar flux patterns

295 in the four incubation batches and were not significantly different from each other ($p<0.01$).

296 Within each plant, the highest N_2O peaks were observed in the D fertilisers except for AmP

297 where the highest peak was measured in E-AmP on day 0.

298   Cumulative N_2O (N_2O_{cum}) emissions among the different groups of fertilisers were not

299 easily differentiated, at least not statistically. Soil amended with urea showed a significantly

300 higher N_2O_{cum} release than most other treatments, except for CAN, D-AmP, D-WNE, E-AmP,

301 E-WNE, and AW-WNE (Table 4). It is important to note that the N_2O_{cum} data for the mineral

302 fertilisers are means of the measurements in the four incubation batches which ranged

303   between 0.11 to 0.14 % of urea N applied and 0.10 – 0.12 % of CAN N applied. The SF with

304   depleted P (SF-GZV$_{P-poor}$) and the AS fertilisers induced the lowest N$_2$O$_{cum}$ emissions (0.04 %

305   of applied N) compared to the other fertilisers. However, this value was not significantly

306   different from SF-GZV, AS-A&S, AS-BNS, D-BNS, LF-WNE, and R-AmP. N$_2$O$_{cum}$

307   emissions from the D fertilisers from the five plants varied between 0.06 – 0.09 % of applied

308   N. When considering the end-products originating from the individual biogas plants, the D

309   fertilisers emitted more N$_2$O$_{cum}$ compared to the fertilisers derived from D processing,

310   although the difference was not statistically significant in A&S and BNS (Table 4).

311   The net N$_{rel}$ from the BBFs and mineral fertilisers expressed as a percentage of applied

312   total N is shown in Table 4. This parameter indicates the amount of fertiliser-derived available

313   N present in the soil at the end of the incubation period under the conditions of the

314   experiment. There were large variations in the net N$_{rel}$ within the different fertiliser groups.

315   The highest N$_{rel}$ was observed in the mineral fertilisers (102 – 108%) and AW-WNE (108%).

316   N immobilisation occurred in the SF with depleted P (SF-GZV$_{P-poor}$). The net N$_{rel}$ was

317   significantly negatively correlated with C/N ratio (r = 0.67; $p < 0.01$).

318

3.3 CO$_2$ emission

320   Figure 2 shows the cumulative CO$_2$ emissions from BBFs where greater than 300 kg

321   C/ha was applied. At the end of the incubation, the D fertilisers mineralised between 13 and

322   52% of applied C. The highest cumulative CO$_2$ emission was from the D-WNE treatment

323   ($p<0.01$). Soils amended with SFs of digestate showed a significantly lower release of CO$_2$

324   than other soils, with 3 to 8% of applied C mineralised. CO$_2$ emissions of treatments with

325   AmP, A&S, and BNS fertilisers were not significantly different from each other, probably due

326   to the high variability in the measurements. The CO$_2$ emission data for CAN, R, LF, AW and

327   AS fertilisers were not shown because they contain little or no organic C, therefore, amounts

328 of C added to soil after their application was deemed to be negligible. Any additional CO_2

329 emitted was attributed to the positive priming effect of native soil organic C which stimulates

330 mineralisation (Fontaine et al., 2003). Urea which contains 20% C showed rapid

331 mineralisation (>70%) within the first 2 days of incubation in all the batches due to its fast

332 hydrolysis after application to soil.

333

334 **3.4 CH_4 emission**

335 Table 4 indicates that for most of the fertilisers studied, cumulative CH_4 emissions

336 were lower than the control. The exceptions were E-AmP and E-WNE with net positive

337 emission of 4 and 1 mg m^{-2}, respectively. However, no significant difference ($p<0.05$) was

338 observed among the treatments due to high variability in the flux measurements.

339

340 **3.5 Principal component and correlation analyses**

341 The overall grouping of individual observations and variable correlations is depicted in

342 Figure 3. CH_4 was not included as a variable in the principal component analysis (PCA) since

343 there was no significant difference in CH_4 emission among the treatments. The first two

344 factors explained 61% of the total variance in all variables. The grouping of individual

345 observations shows a separation of the different BBFs into three distinct groups based on their

346 properties in relation to cumulative N_2O and CO_2. Separation along F1, which accounts for

347 24% of the total variation, was explained by differences in TN, TC, NH_4, OM, C/N, DOC,

348 TDN/TN. The second factor (F2) which accounts for 36% of the total variation, was described

349 by differences in N_2O_{cum} and CO_{2cum} emissions.

350 The D and E fertilisers were grouped at the upper part of the diagram indicating BBFs

351 with the highest average DOC concentrations, pH, and N_2O_{cum} and CO_{2cum} emissions. The SF

352 fertilisers were clustered along the positive quadrant of the F1 axis where the fertilisers with

353   the highest average TC and C/N ratio are represented. The AS, AW, R and LF fertilisers were

354   found more displaced along the left side of the F1 axis, where TN, NH_4^+, and TDN/TN are the

355   dominant variables.

356   Significant linear correlations were established between N_2O_{cum} emissions and the

357   characteristics of the different fertiliser groups. When omitting fertilisers LF, R, AW, AS,

358   which are outliers regarding TC (i.e., <5%), N_2O_{cum} correlated negatively with C/N ratio (r = -

359   0.68, $p < 0.05$) and positively with TDN/TN ratio (r = 0.75, $p < 0.01$). Also, N_2O_{cum} emission

360   from the SF fertilisers was positively related to TN (r = 0.92, $p < 0.05$) and DOC (r = 0.70, p

361   < 0.05). In the dominantly mineral fertilisers (i.e., LF, R, AS, AW), pH appeared to be

362   positively correlated with N_2O_{cum} emissions (r = 0.81), but this relationship was not

363   statistically significant ($p = 0.10$).

364   Cumulative CO_2 emission in SF, D, and E fertilisers was correlated positively with TN (g

365   kg^{-1} DW) (r = 0.85, $p < 0.01$), TDN/TN ratio (r = 0.57, $p < 0.05$), and DOC/TC ratio (r = 0.83,

366    $p < 0.01$), and negatively to C/N ratio (r = 0.60, $p < 0.05$).

367

368   **4. Discussion**

369

370   **4.1 Physicochemical properties of the biobased fertilisers**

371   The high OM and/or N contents of the studied BBFs underline their potential as soil

372   improvers or fertilisers. Elevated OM contents in the SFs of digestate make them better suited

373   as organic soil improvers (Egene et al., 2020; Peters and Jensen, 2011) with the potential to

374   increase C sequestration in soils (Veeken et al., 2017). Drying of the SF of digestate (as in

375   SFD-AmP and SFD-WNE), concentrates the N (24 and 31 g kg^{-1}, respectively), making them

376   also applicable as N or P fertilisers (Regelink et al., 2021). On the other hand, ammonia

377   stripping of the LF of digestate produces pure mineral N fertilisers – as in AW-WNE, AS-

378   A&S and AS-BNS with total N contents of 53, 76, and 41 g kg^{-1}, respectively. The large

379   variation in total N within the D fertilisers was attributed to differences in the N contents of

380   the feedstock. The D with the highest total N content was obtained from GZV where animal

381   waste constituted a high proportion (81% w/w) of the feedstock. On the other hand, D

382   obtained from the processing of industrial food waste had the lowest total N content.

383   The pH of the studied BBFs was mostly alkaline which can influence soil pH and

384   processes, including nutrient availability and nitrification rate. Fertilisers with a strong

385   alkaline character, as was measured in AW-WNE (pH 9.7), indicates a high risk of NH_3

386   volatilisation when applied to soils. Low pH in some BBFs was related to the use of sulphuric

387   acid during digestate processes. For example, SF-GZV$_{P-poor}$ (pH 5.5) was treated with

388   sulphuric acid to lower its P content. Similarly, the digestate acidification step before vacuum

389   evaporation explains the slightly acidic pH of E-AmP (pH 6.2).

390   For most of the BBFs, the TDN concentration, which represents the sum of dissolved

391   organic N and mineral N (Christou et al., 2005), was higher than the NH_4^+ concentration. This

392   indicates the presence of a sizeable dissolved organic N pool in the BBFs, especially in E-

393   WNE. As explained in section 2.1, E-WNE is the concentrate that is obtained after NH_3 is

394   evaporated from the LF of digestate. This explains the low NH_4^+ content in the fertiliser.

395   The PCA of variables related to the cumulative N_2O and CO_2 emissions showed a clear

396   separation among the different groups of BBFs. TC and C/N ratio were the most important

397   variables for separating the SF fertilisers from the others (along F2), while DOC and DOC/TC

398   ratio differentiated the D and E fertilisers from the LF, R, AW and AS fertilisers. Except for

399   SF_GZV$_{P-poor}$, the TOC/N ratio in the BBFs was below 20 which is favourable for N

400   mineralisation (Mendham et al., 2004; Wagner and Wolf., 1999). In organic substrates with a

401   high C/N ratio as in SF_GZV$_{P-poor}$ (27), mineralisation is slow and N immobilisation may

402   dominate (Egene et al., 2020).

4.2 N$_2$O emissions and N mineralisation

The principal factors that regulate soil-borne N$_2$O emissions are soil mineral N

concentration, availability of decomposable organic C, soil moisture, soil temperature, soil

pH, and the activity of (de)nitrifying organisms (Dalal et al., 2003; Šimek and Cooper, 2002;

Wang et al., 2021). In this study, we aimed to create the conditions optimal for N$_2$O

production by using soil with a neutral pH, pre-incubating the soil for one week to activate

microorganisms, and maintaining a high WFPS (80%) (Cayuela et al., 2010). As such,

differences in N$_2$O flux from the different treatments may be attributable to differences in

mineral N and labile organic C availability. The N$_2$O emissions in the first days of incubation

were likely driven by nitrification of the initial NH$_4^+$ present in the fertilisers (Alburquerque

et al., 2012a; Askri et al., 2016; de la Fuente et al., 2013; Egene et al., 2020). Some studies

(Askri et al., 2016; Pampillón-González et al., 2017) on digestate fertilised soils reported

positive correlations between N$_2$O$_{cum}$ emissions and the initial NH$_4^+$ content in the digestate.

However, this was not observed in the current study. This may be because NH$_4^+$ was not a

limiting factor in most of the fertilisers investigated, except for E-WNE which has a low

NH$_4^+$/TN ratio of 0.026. Moreover, the mineral N content at the end of the incubation in all

treatments mainly consisted of NO$_3^-$-N (Table 4) which supports our assumption that NH$_4^+$ in

the treatments was nitrified (or immobilized) during the 18-day incubation.

The urea fertilised soil produced the highest N$_2$O$_{cum}$ emission with 0.12% of applied

N, although this value was not statistically differentiated from CAN, D-AmP, D-WNE, E-

AmP, E-WNE, and AW-WNE. Generally, mineral N fertilisers quickly dissolve after addition

to soil leading to increased NH$_4^+$ availability followed by nitrification and N$_2$O production

(Saggar et al., 2013a; van der Weerden et al., 2016). The hydrolysis of urea or the dissolution

of CAN in the soil can result in the loss of NH$_3$ which is a precursor to the formation of N$_2$O

(Forrestal et al., 2016; Huang et al., 2014; Saggar et al., 2013b). It is, therefore, conceivable

that the high N_2O emissions observed in urea and CAN may be due to the combination of

direct N_2O production (from nitrification of NH_4^+) and the indirect N_2O release (due to NH_3

oxidation). This phenomenon could also explain the high N_2O emission from the soil

amended with NH_3-water (AW-WNE).

Despite their high mineral N contents (~100%), significantly lower N_2O emissions

were measured in AS-BNS and AS-A&S compared to the mineral fertiliser treatments. This

was attributed to the low pH of the AS fertilisers which inhibits N_2O production from

nitrification (Dalal et al., 2003). In contrast, the hydrolysis of urea may have caused

alkalinisation of the soil, especially in the microsites close to the urea granules (Clayton et al.,

1997), thereby creating conditions more favourable for N_2O production from nitrification

(Dalal et al., 2003). Similar observations were found in other studies that compared N_2O

emissions between mineral AS, urea, and nitrate fertilisers (Clayton et al., 1997; Tierling and

Kuhlmann, 2018). According to Clayton et al. (1997), N_2O_{cum} emissions from a clayey-loam

soil fertilised with AS, urea, and calcium nitrate over 12 months were 0.2, 0.8, and 0.5% of

applied N, respectively. Meanwhile, Tierling and Kuhlmann (2018) performed a 21-day

incubation of a loamy-sand soil supplemented with AS, urea, and potassium nitrate fertilisers

and reported N_2O_{cum} emissions of 0.07, 0.26, and 0.02% of applied N, respectively. In both

studies, differences in N_2O production between the fertilisers were also attributed to pH

effects in the soil.

Cumulative N_2O emissions from the D, SF, E, LF and R fertilisers could hardly be

differentiated, at least not statistically. The exception was the SF with depleted P (SF-GZV$_{P-poor}$) which induced significantly lower N_2O emissions compared to the other fertilisers. As

previously described, the low emissions may have been caused by the acidic pH (5.5) of SF-

GZV$_{P-poor}$. However, its high C/N ratio (27) may have also played an important role in

limiting nitrification (Egene et al., 2020) and consequently, N_2O production. Elevated C/N

453 ratio in organic substrates slows down their decomposition and subsequent release of DOC

454 and NH_4^+ through mineralisation, both of which are linked to increased N_2O emissions

455 (Huang et al., 2004). Among the SF fertilisers, differences in C/N ratio (r = -0.87) and DOC (r

456 = 0.70) could explain the variation in N_2O emission. Similar results were reported by Huang

457 et al. (2004) who found that N_2O_{cum} of N_2O were negatively correlated with C/N ratio and

458 positively correlated with DOC of solid organic residues after a 21-day incubation study.

459 The pH effect described above was not apparent in E-WNE which has a slightly acidic

460 pH of 6.2 but produced N_2O_{cum} emission comparable to the mineral fertilisers (Table 4).

461 Remarkably, soil amended with E-WNE emitted less than the control at the start of incubation

462 before showing rapid N_2O release from the ninth day (Figure 1). This was attributed to its low

463 initial NH_4^+ (0.26 g kg^{-1}) meaning there was almost no readily available N to be nitrified. This

464 N deficiency enhanced microbial decomposition of labile organic matter to obtain N,

465 otherwise known as "microbial mining" (Craine et al., 2007; Moorhead and Sinsabaugh,

466 2006) which resulted in temporary N immobilisation in the first days of the incubation. The

467 high TDN concentration in E-WNE (7.9 g kg^{-1}) indicates that microorganisms could easily

468 access the dissolved organic N in the fertilisers to release NH_4^+. At this point, nitrification of

469 the released mineral N could progress rapidly.

470 Despite the short duration of our incubation experiment (18 days), the N_2O_{cum} values

471 for D fertilisers were strongly agreed with results from a 110-day field study by Baral et al.

472 (2017) who measured N_2O_{cum} of 0.10% of applied N due to digestate fertilisation applied at

473 167 kg N ha^{-1}. Nitrification of NH_4^+-N in fertilised "hotspots" within the soil, characterized

474 by enhanced microbial activity and oxygen demand, was determined as the principal factor

475 controlling N_2O production, with most of the emissions occurring in the first 35 days. After a

476 one year investigation of N_2O emissions from mineral and organic fertilisers, Meijide et al.

477 (2009) also reported N_2O_{cum} emissions of 0.12 and 0.11% of applied N from digestate and

478 urea fertilised agricultural fields, respectively. The authors determined that nitrification and

479 denitrification occurred at different stages of the experiment but concluded that environmental

480 factors, mainly WFPS, strongly influenced N_2O emission rates.

481   Overall, the results from our incubation study show that the differences in N_2O

482 emission flux among the BBFs were small and in some cases, marginal. Therefore, the rate of

483 N_2O emissions was probably more affected by the soil type and condition than by the fertiliser

484 properties. In a study by Abubaker et al. (2013), large differences in N_2O emissions were

485 found between different biogas residues when incubated for 24 days a sandy and clayey soil.

486 However, the same digestate showed comparable N_2O_{cum} emissions when they were

487 incubated in loamy soil, with values between 0.08 to 0.09% of applied N. The characteristics

488 of the soil used in our study closely resemble the loam soil used in Abubaker et al.'s (2013)

489 study, at least in terms of soil texture. This suggests that the BBFs discussed in this study may

490 induce different N_2O emission fluxes when incubated in differently textured soils.

491

492   **4.3 CO_2 emissions**

493   Cumulative CO_2 emissions from the soils amended with the D, E, and SF fertilisers

494 were related to the proportion of readily available C, as evidenced by the strong positive

495 correlation between CO_{2cum} emission and DOC/TC ratio (r = 0.83). Cysneiros et al. (2008)

496 and Jacobi et al. (2009) have previously shown that considerable amounts of volatile fatty

497 acids are formed as intermediates during AD. These organic compounds are easily

498 metabolized by soil microorganisms within a few days, releasing CO_2 in the process. The

499 slower C mineralisation in the SF treatments (3.5 – 7% of applied C) suggests that the organic

500 C in SF fertilisers is more stable than those in the D and E fertilisers. This may be because

501 solid-liquid separation of the whole digestate resulted in the separation of the stable

502 particulate C, mainly associated with the solid phase, from the easily degradable C which

503   remained mostly in the liquid phase. These results reaffirm the findings from other incubation

504   studies with biogas residues, that the availability of labile C favours the production of soil-

505   borne CO_2 (Askri et al., 2016; Cardelli et al., 2018; Mukherjee et al., 2016).

506   It is worth noting that the enhanced degradation of native soil C (priming effect)

507   and/or the reduction of carbonates in the fertilisers may have contributed to the CO_2 emissions

508   in the BBF-fertilised soils (Kuzyakov et al., 2000; Yoshida et al., 2015). However, the

509   differentiation of the origins of soil-borne CO_2 emissions was not investigated in this study.

510

511   **4.4 CH_4 emissions**

512   The low CH_4 emissions from the treatments highlight the fact the incubations were

513   carried out under sufficiently aerobic conditions. Our results are in agreement with studies by

514   Odlare et al. (2012) and Pampillón-González et al. (2017) who reported negative or negligible

515   CH_4 emissions from the soil after the application of biogas residues. In this study, CH_4

516   oxidation was likely driven by the presence of methanotrophic (CH_4 oxidizing) bacteria in the

517   soil whose activity was stimulated by the addition of N fertiliser. Conrad (1996) and Steven et

518   al. (2006) testified that agricultural soils are common habitats for methanotrophic bacteria

519   who, in the presence of oxygen, utilize CH_4 as a source of carbon and energy.

520

521   **4.5 Global warming potential**

522   The global warming potential (GWP) of N_2O and CH_4 emissions were determined and

523   expressed as CO_2 equivalents per 100 grams of N added, using a conversion factor of 298 for

524   N_2O and 25 for CH_4 (IPCC, 2007). CO_2 emission was not considered for GWP emissions

525   calculation since, from the life cycle assessment perspective, the biodegradation of organic

526   matter releases biogenic carbon (USEPA, 2010; WRI, 2014). Therefore, the mineralization of

527   BBFs in soils does not contribute to the net increase of CO_2.

528   As shown in Figure 4, the GWP of the BBFs was either equal to or lower than the

529   GWP of the mineral fertilisers. In all the fertilisers, N_2O emissions contributed significantly

530   more to GWP than CH_4 emissions. Generally, CH_4 was taken up rather than emitted,

531   however, the benefit gained by CH_4 consumption was offset by the increase in N_2O

532   emissions. The fertilisers derived from the NRR processing of digestate appear to decrease the

533   GWP relative to the unprocessed digestate fertilisers. This trend is particularly noticeable in

534   fertilisers from WNE and AmP.

535   GWP values varied between 0.08 and 0.35 kg CO_2 eq kg^{-1} N and are hence low as

536   compared to values obtained from long-term field studies. Over ten months, Zilio et al. (2022)

537   measured net N_2O_{cum} emissions between 1.2 and 3% of N applied, from digestate+AS and

538   urea+AS fertilised fields, corresponding to 3.80 and 9.84 kg CO_2 eq kg^{-1} N, respectively. The

539   authors concluded that the environmental impact in terms of GHG emissions from fertilisation

540   with the tested BBFs or urea were comparable, in agreement with findings from this study.

541   Meanwhile, Walling and Vaneeckhaute (2020) found in their review that N_2O-derived GWP

542   due to digestate fertilisation can range between 0.15 to 17.6 kg CO_2 eq kg^{-1} N, depending on

543   the application technique. This means that the impact of GHG emissions related to the

544   application of digestate N fertilisers could still exceed the impact of synthetic N fertilisers and

545   hence, N_2O emissions from N fertilising products from digestates remain a point of

546   environmental concern.

547   **4.6 Limitations of the laboratory incubation approach**

548   The microcosm approach employed in this study enables the close monitoring and

549   quantification of fertilisation effects on soil GHG emissions and C and N turnover processes.

550   However, the standardized conditions (for temperature and moisture) under which the

551   incubations were performed greatly differ from field conditions. Generally, field

552 measurements give higher N_2O emissions due to pulses of N_2O release after a rainfall event or

553 from management practices such as tillage (Wang et al., 2021). The method of fertiliser

554 application also influences the N_2O production dynamics as reported by Velthof and

555 Mosquera (2011) who found that injection of slurry increased the average emission factor of

556 N_2O in comparison to surface application.

557 Furthermore, our incubation was performed only on sandy-loam soil even though

558 fertilisation effects on GHG emissions are modulated by soil texture (Pelster et al., 2012). A

559 meta-analysis by Charles et al. (2017) found that N_2O emissions from fertilisation were 2.8

560 times greater in fine- than in coarse-textured soils. Future studies should investigate the effect

561 of soil texture on GHG emissions from BBF enriched soils through similar microcosm

562 incubation experiments. Finally, other pathways of N losses such as NH_3 and N_2 emissions as

563 well as NO_3^- leaching were currently not explored. Quantifying these loss pathways is

564 necessary to get a full picture of soil N cycling after fertilisation with BBFs and should be

565 investigated in future studies.

566 **5. Conclusions**

567 The key finding from this study was that none of the biobased fertilisers (BBFs)

568 emitted more N_2O than the mineral N fertilisers (urea and calcium ammonium nitrate). Soil-

569 borne N_2O emission from the BBFs was attributed to nitrification of the initial ammonium N

570 shortly after the fertilisers were applied. Differences in C/N ratio and TDN/TN ratio could

571 partly explain the variation in N_2O emissions in the unprocessed digestate, evaporator

572 concentrates, and solid and liquid fractions of digestate. CO_2 respiration in the BBF

573 treatments was strongly influenced by the availability of labile C while CH_4 emissions due to

574 fertilisation were negligible. Global warming potential (GWP) of the BBFs was comparable

575 or lower than that of the mineral fertilisers. Furthermore, the GWP of BBFs derived from the

processing of digestate was generally lower than that of whole digestate. These findings not

only revealed the important factors driving short-term GHG emissions in BBF-fertilised soils,

but also suggested that the refinement of digestate into concentrated fertilising products does

not increase the risk of soil-borne GHG emissions.

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856
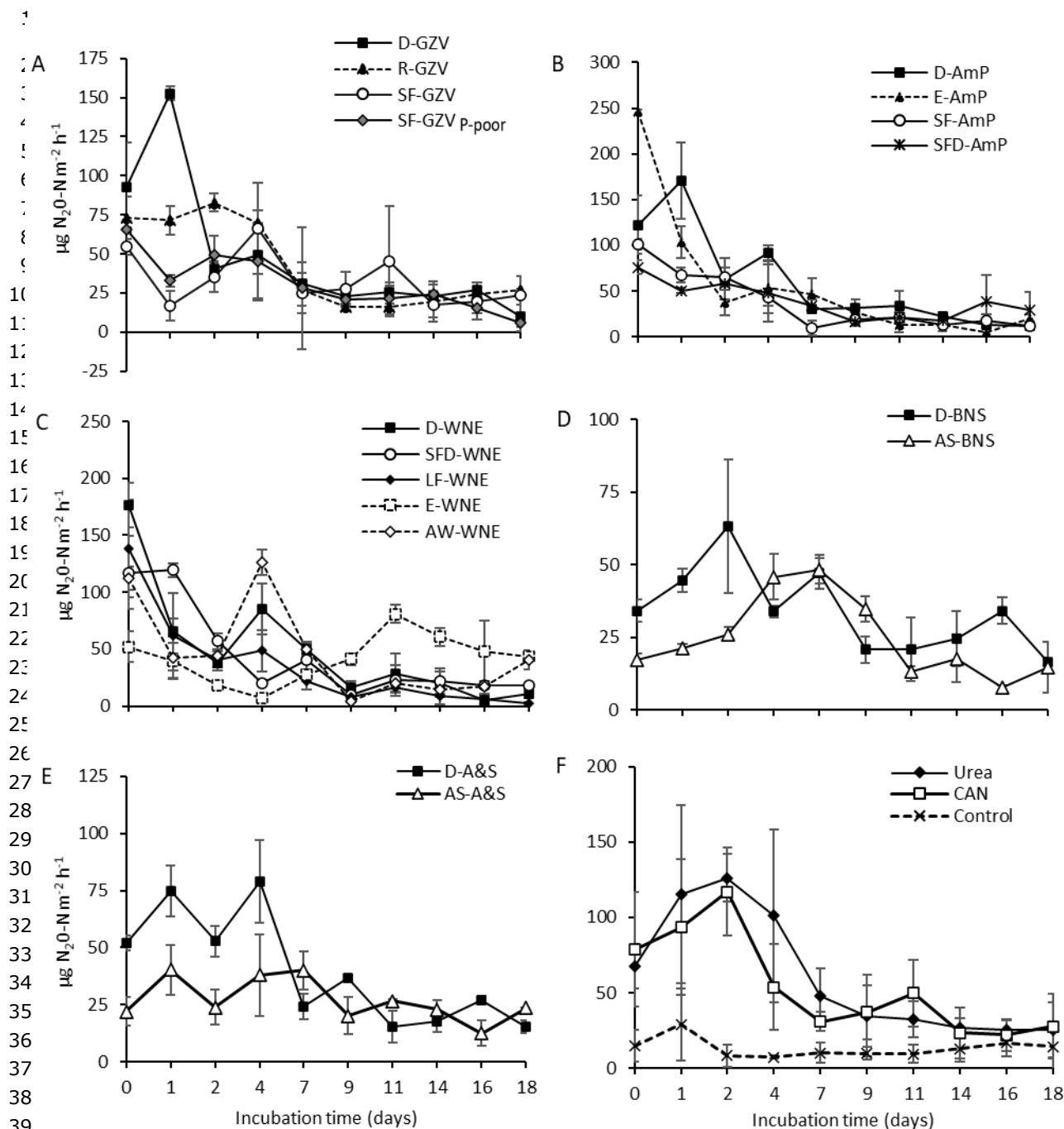
Figure 1. Kinetics of N2O emission rates measured during incubation of digestate-derived biobased fertilisers (A-E) and mineral N fertilisers (F), applied to a sandy-loam soil. Values are means with standard deviation represented by vertical bars; n = 3 for the BBFs; For control, urea and CAN, data points represent the means of measurements in the four incubation batches, n = 9). For abbreviations, refer to Table 1 in Materials and methods.
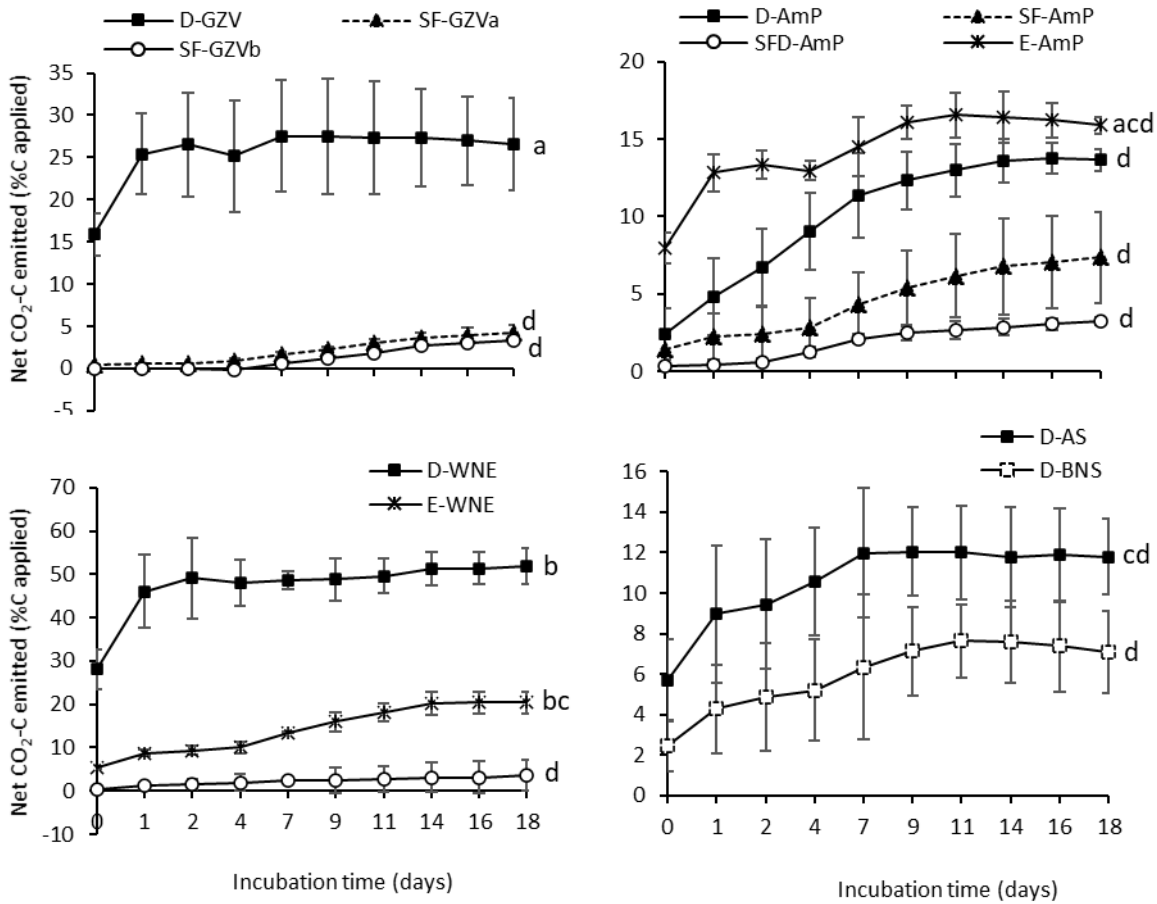
Figure 2. Cumulative CO_2 emission measured during incubation of digestate-derived biobased fertilisers applied to a sandy-loam soil. Different lower-case letters indicate significant differences between C mineralisation means according to Tukey's test ($p<0.05$). Values are means with standard deviations represented by vertical bars (n = 3). For abbreviations, refer to Table 1 in Materials and methods.
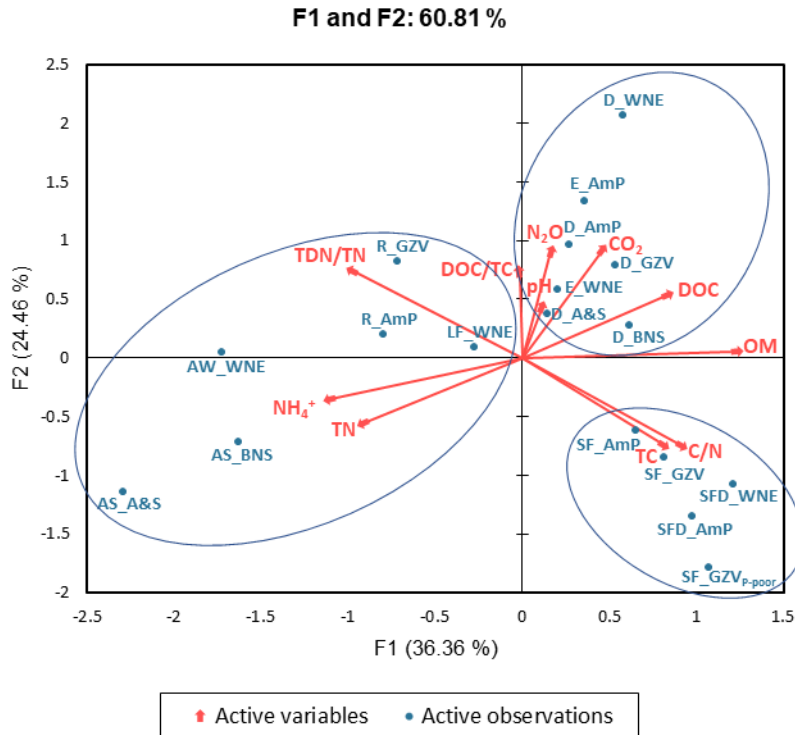
F1 and F2: 60.81 %

Figure 3. Biplot showing ordination of the BBFs (blue text) based on a PCA of variables (red text) related to cumulative N_2O and CO_2 emissions. F1 explains 36% of the variance in the data and F2 explains 24% of the variance in the data. Variables far from the centre and close to each other are significantly positively correlated (Pearson's correlation; r close to 1); Variables in orthogonal positions are not correlated (r close to 0); variables on opposite sides of the origin are significantly negatively correlated (r close to -1). The arrows point in the direction of treatments with an above-average signal. Variable abbreviations: OM: organic matter; TN: total N; TC: total C; DOC: dissolved organic carbon; TDN: total dissolved N; C/N: carbon to nitrogen ratio. For abbreviations of BBFs, refer to Table 1 in Materials and methods.
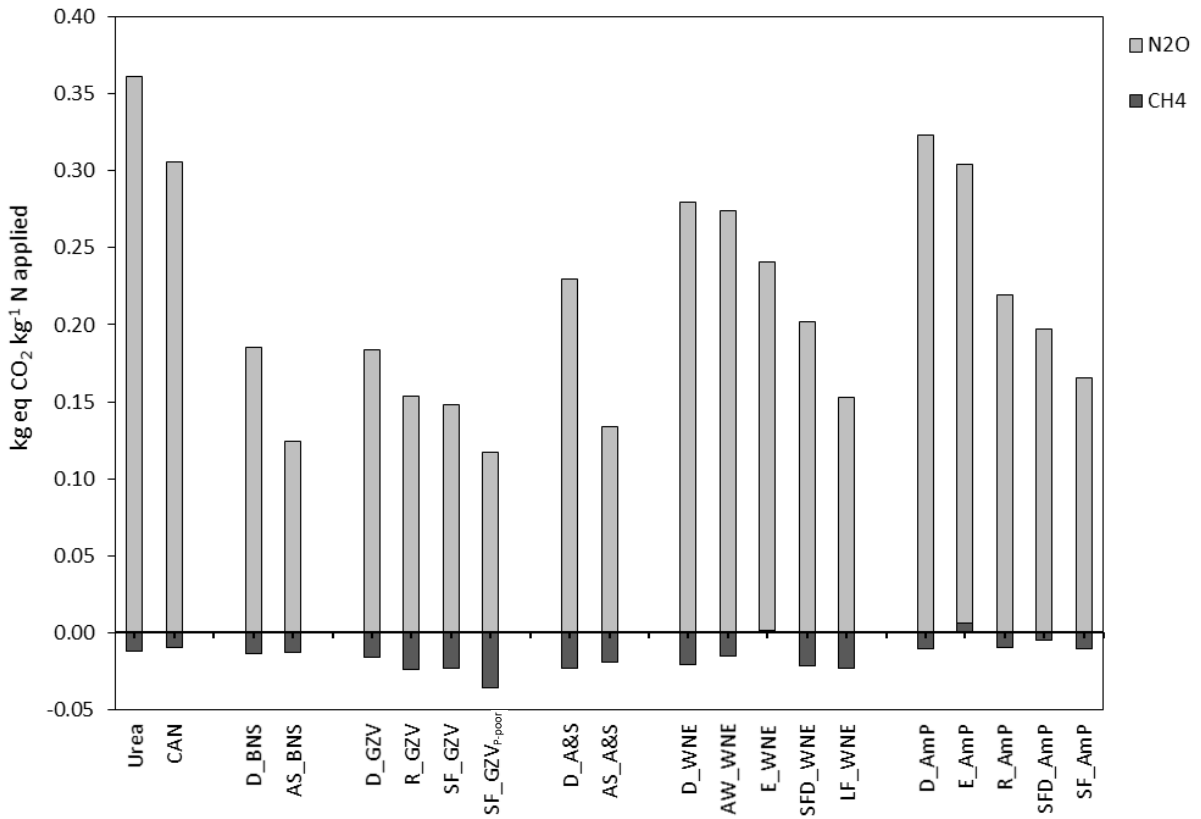
Figure 4. Global warming potential of mineral and biobased fertilisers calculated as the sum of grams of cumulative N_2O and CH_4 (expressed as grams of CO_2 equivalents) per 100 grams of N applied to the soil. Results are based on aerobic incubation experiments conducted over 18 days. For abbreviations, refer to Table 1 in Materials and methods.

1   **Table 1.** Overview of feedstock and produced biobased fertilisers at five biogas plants

Inputs	Code	Fertiliser and processing information
Pig slurry (81% w/w) and residues from agro-food industry (a.o. grain and rice husk, potato skins and coffee grounds, 19% w/w).	D-GZV	Digestate, unprocessed
	SF-GZV	Solid fraction (SF) of digestate, separated via decanter centrifugation
	SF-GZV$_{P-poor}$	Low phosphorus (P) soil improver, obtained after leaching P from GZV-SF with water and sulphuric acid
	R-GZV	Reverse osmosis concentrate, obtained after the liquid fraction (LF) of digestate is treated via dissolved air floatation and membrane filtration
Residues from food processing industry and source-segregated food waste	D-AmP	Digestate, unprocessed
	SF-AmP	Solid fraction of digestate, separated via decanter centrifugation
	SFD-AmP	Dried SF of digestate, obtained after drying AmP-SF in fluidized bed dryer at 60 °C
	E-AmP	Evaporator concentrate, obtained after the LF of digestate is passed through a vacuum evaporator
	R-AmP	Reverse osmosis concentrate, obtained after ammonia water from vacuum evaporator is treated via reverse osmosis.
Residues from agro-industry (potatoes and grain, 40% w/w), sludge from industrial wastewater treatment plants (15% w/w) and animal manure (45% w/w)	D-WNE	Digestate, unprocessed
	LF-WNE	LF of digestate, separated via decanter centrifugation
	E-WNE	Evaporator concentrate, the fraction of LF of digestate retained after NH_3 removed via evaporation
	SFD-WNE	Dried SF of digestate, obtained after drying the SF of digestate in a Hydrogone dryer
	AW-WNE	Condensed ammonia water, obtained after NH_3-rich gas is condensed with water vapour
Sewage sludge from wastewater treatment plants (86% w/w) and coproducts (digestate from anaerobic treatment of source-segregated domestic food waste, 14% w/w)	D-A&S	Digestate, unprocessed
	AS-A&S	Ammonium sulphate solution, obtained after ammonia is extracted from biogas using H_2SO_4
Energy crops (silage maize, silage rye and corn, 85% w/w) and poultry litter (15% w/w)	D-BNS	Digestate, unprocessed
	AS-BNS	Ammonium sulphate solution, obtained after gypsum is added to ammonia stripped from digestate

2

Table 2. Fertiliser and equivalent amounts of NH_4^+-N, organic N (N_{org}), and total carbon (C) added to soil at the total N application rate of 170 kg ha^{-1}

Fertiliser groups	Fertiliser	NH_4^+-N	N_{org}	C added
		kg ha^{-1}		
Digestate (D)	D-GZV	113	57	595
	D-AmP	109	61	1334
	D-WNE	89	81	392
	D-A&S	108	62	671
	D-BNS	81	89	1000
Solid fractions of digestate (SF)	SF-GZV	154	16	2838
	SF-GZV$_{P-poor}$	46	124	4489
	SF-AmP	39	131	1778
	SFD-AmP	8.0	162	2047
	SFD-WNE	39	131	1814
Evaporator concentrates (E)	E-AmP	138	32	570
	E-WNE	4.0	156	748
Liquid fraction of digestate (LF)	LF-WNE	95	75	151
Reverse osmosis concentrates (R)	RO-GZV	162	8	86
	RO-AmP	143	27	197
Ammonia water (AW)	AW-WNE	170	0	0
Ammonium sulphate solutions (AS)	AS-A&S	170	0	0
	AS-BNS	169	1	0
Mineral fertilisers	CAN	170	0	0
	Urea	170	0	74

19 **Table 3.** Main physicochemical characteristics of biobased and mineral fertilisers included in

20 the study. DM: Dry matter; FW: fresh weight; OM: organic matter; TN: total N; TC: total C;

21 DOC: dissolved organic carbon; TDN: total dissolved N

Fertiliser	pH	DM	OM	TN	TC	NH_4^+	TDN	DOC	C/N	DOC/TC	TDN/TN
	-	%	%FW	\multicolumn{5}{l}{g kg^{-1} FW}				-	
D-GZV	8.20	7.9	5.7	8.12	30.0	5.42	5.30	5.13	3.7	0.19	0.63
D-AmP	8.10	8.1	5.0	3.44	29.0	2.20	2.44	3.20	8.4	0.09	0.93
D-WNE	8.57	5.2	3.2	6.50	17.0	3.41	4.91	5.05	2.7	0.33	0.78
D-A&S	8.60	9.3	5.5	6.95	29.0	4.40	1.27	4.40	4.1	0.05	0.63
D-BNS	8.30	8.1	6.5	5.10	33.0	2.42	5.19	2.80	6.5	0.17	0.55
SF-GZV	8.80	33	25	11.8	136	7.24	2.97	3.30	17	0.02	0.41
SF-GZV$_{P-poor}$	5.50	27	24	4.50	120	1.22	0.53	1.32	27	0.00	0.29
SF-AmP	8.30	26	17	9.10	100	2.10	3.01	2.40	11	0.03	0.26
SFD-AmP	8.10	47	47	24.0	310	1.10	2.44	1.00	13	0.01	0.04
SFD-WNE	7.90	94	67	31.4	343	7.14	8.73	3.20	11	0.03	0.10
E-AmP	6.20	6.3	6.3	7.80	29.0	6.30	8.77	7.70	3.7	0.34	0.99
E-WNE	7.00	17	9.5	10.0	51.0	0.26	2.96	7.90	3.4	0.07	0.81
LF-WNE	8.85	2.4	1.2	4.94	5.90	2.77	1.10	4.34	1.2	0.25	0.88
R-GZV	8.30	3.6	1.0	9.10	5.70	8.65	8.85	-	0.6	-	0.97
R-AmP	7.30	1.5	1.4	3.10	3.60	2.60	3.04	-	1.2	-	0.98
AW-WNE	9.70	<1	0.0	53.2	0.50	53.2	-	-	-	-	1[**]
AS-A&S	5.90	38	0.0	76.1	0.10	75.9	-	-	-	-	1[**]
AS-BNS	6.30	21	0.0	41.3	0.26	41.0	-	-	-	-	1[**]
CAN	7.39	99	0.0	300	-	-	-	-	-	-	-
Urea	7.91	99	0.0	460	200[*]	-	-	-	-	-	-

22 [*]based on the molecular composition of urea

23 [**]assumed values since NH_4^+-N≈TN
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Table 4. Soil concentrations of NH_4^+ and NO_3^- and net* mineral N release (N_{rel}) at the end of the incubation (18 days); and net* cumulative fertiliser-derived N_2O and CH_4 emissions

Fertiliser	NH_4^+	NO_3^-	N_{rel}	N_2O_{cum}	CH_{4cum}
	mg kg^{-1}		% N applied		mg m^{-2}
D_GZV	1	122	88	**0.06**[bcde]	-1[a]
D_AmP	1	151	97	0.11[ab]	-7[a]
D_WNE	2	105	75	0.09[abcde]	-14[a]
D_A&S	2	77	45	*0.08*[bcdefg]	-16[a]
D_BNS	2	67	34	**0.06**[efgh]	-9[a]
SF_GZV	1	93	57	**0.05**[fgh]	-16[a]
SF_GZV$_{P\text{-poor}}$	0	38	-3	**0.04**[h]	-24[a]
SF_AmP	1	91	54	**0.06**[fgh]	-7[a]
SFD_AmP	1	58	23	*0.07*[defgh]	-3[a]
SFD_WNE	1	60	26	*0.07*[cdefgh]	-15[a]
E_AmP	0	118	84	0.10[abc]	4[a]
E_WNE	2	90	56	0.08[abcdef]	1[a]
LF_WNE	1	125	89	**0.05**[fgh]	-16[a]
R_GZV	1	117	83	**0.05**[fgh]	-16[a]
R_AmP	1	121	87	0.07[bcdefgh]	-6[a]
AW_WNE	2	135	108	0.09[abcde]	-8[a]
AS_A&S	2	112	82	**0.04**[fgh]	-13[a]
AS_BNS	2	117	88	**0.04**[gh]	-9[a]
CAN	1	135	108	0.10[abcd]	-6[a]
Urea	1	130	102	0.12[a]	-8[a]

In a column, values followed by the same letter do not differ significantly according to Tukey's test ($p<0.05$).

N_2O_{cum} values in bold are significantly lower than CAN and urea; italicized values are significantly lower than urea but not different from CAN.

*After subtracting the control

Declaration of interests

☒The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: