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Digestion of bio-waste - GHG emissions and mitigation potential

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Abstract

Background: For a precise description of the emission situation of the anaerobic digestion (AD) of the separately collected organic fraction of household waste (bio-waste), only a few data are available. The paper presents the greenhouse gas (GHG) emissions measured at 12 representative AD plants treating bio-waste. The results of the emission measurements were used to assess the ecological impact of bio-waste digestion and to describe possible mitigation measures to reduce the occurring GHG emissions. With respect to the climate protection, a quantitative assessment of the emissions of energy generation from biomass and biological waste treatment is important. Biogas plants need to be operated in a way that negative environmental effects are avoided and human health is not compromised.

Methods: GHG balances were calculated based on the measured emissions of the gases methane, nitrous oxide, and ammonia of bio-waste AD plants. The emission analysis supports the reduction of GHGs in biogas production and contributes to a climate-efficient technology.

Results: The results show that GHG emissions can be minimized, if the technology and operation of the plant are adjusted accordingly. The open storage of active material (e.g., insufficient fermented residues from batch fermentation systems), open digestate storage tanks, missing acidic scrubbers in front of bio-filters, or insufficient air supply during the post-composting of digestate can cause relevant GHG emissions.

Conclusions: Consequently avoiding open storage of insufficient fermented residues and using aerated post-composting with short turnover periods, smaller heaps, and an optimized amount of structure (woody) material can reduce GHG emissions.

Keywords: Anaerobic digestion; Emission measurement; Greenhouse gas balance; GHG mitigation

Background

Gaseous emissions are of great importance referring to the operation of biogas plants because they can affect the safety, the greenhouse gas (GHG) balance, and the economy of plants significantly. Depending on the used technology and the kind of operation, GHG emissions like methane, nitrous oxide, and ammonia are occurring. Methane emissions dominate GHG emissions of biogas plants.

Due to the global warming potential (GWP) of 25 relative to carbon dioxide [1], methane emissions have a strong effect on the climate change. Leakages, process disturbances, and unavoidable emissions during operation can influence the total GHG performance of the biogas plant negatively. Regarding measured emissions of biogas plants in operation, only a small number of detailed studies are available.

In former studies, the overall emissions of biogas plants usually have been estimated by assumptions, e.g., '1 % of diffuse methane emissions from the components of anaerobic digestion (AD) plants like digester, pipes,' etc. (e.g. [2,3]). However, in the recent years, several studies estimated methane emissions from biogas plants (e.g. [4-9]). Most of the published studies analyzed agricultural AD plants; if waste treating plants were investigated, only a few AD components were monitored as summarized by Dumont et al. [10]. Due to the fact that there are only few data describing the emission situation of AD plants based on bio-waste, in the study described here, 12 representative bio-waste treatment plants with AD process as part of

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the overall operation were analyzed. The overall objective of the study was a detailed analysis of GHG emissions generated from biogas production from bio-waste. This paper presents the results of a comprehensive measurement for GHG emissions at bio-waste digestion plants in operation during a long-term period of 3 years. Representative bio-waste digestion plants have been selected, and all relevant components of the process chain were investigated during two periods of a week per year on each of the selected plants to identify the main emission sources and the quantity of the emissions. The results of the emission measurements were implemented in an ecological assessment focused on GHG balances. The results of the examined biogas plants allow a description of possible mitigation measures to reduce GHG emissions. The results bring new aspects into the actual data base to support the assessment of environmental impacts of bio-waste digestion. Thus, the tests on practice biogas plants with respect to the whole process chain allow an optimization of the process in terms of reducing any identified emissions.

In Germany, approximately 9 million tons of bio-waste and green waste per year were collected separately in 2011 [11]. Most of this collected bio-waste and green cuts are used in composting processes. About 1.15 million tons of bio-waste per year and 0.05 tons of green cuts per year are used for digestion in biogas facilities [12]. By the end of 2013, there have been about 130 plants generating biogas from organic waste in operation. Compared to agricultural biogas plants, there is a higher share of dry fermentation processes in AD plants based on bio-waste. About one half of the bio-waste digestion plants are operated as dry fermentation plants in Germany, whereas half of the dry fermentation plants are operated discontinuously (batch system). Currently, there are 25 batch systems based on bio-waste in operation [13]. Due to the robustness of the process and the possibility to treat substrates which are hardly pumpable and contain disturbing materials (e.g., stones, metals, glass), the use of batch systems in case of dry fermentation processes of bio-waste is increasing. In the future, it will be more important to exploit additional potentials in the field of organic waste and residues from industry and municipalities. In the field of municipal bio-waste, the exploitation of additional potentials is in progress. The amount of municipal bio-waste that is available for digestion in biogas plants will increase considerably within the next years. Currently, a considerable trend to digestion of bio-waste and green waste, often integrated as so-called upstream systems into existing composting plants, can be assessed.

Methods

Twelve biogas plants were selected for the detection of plant-based emissions of methane (CH_4), nitrous oxide (N_2O), and ammonia (NH_3). Based on the measured emission rates, GHG balances in compliance with the analysis of GHG credits (e.g., for biogas production, fertilizer, and humus effect of fermentation products and composts) were prepared. Thus, the electricity production and heat utilization of biogas as well as the credits of the various fermentation residues were analyzed to estimate the specific GHG performance of the investigated facilities. Finally, the measurements with respect to mitigation of GHG emissions were analyzed and described.

Investigated biogas plants

The emission analysis includes four continuously operated wet fermentation plants (continuous stirred-tank reactor, CSTR), five continuous dry fermentation plants (plug-flow fermenter), and three batch fermentation processes (discontinuous operation, 'garage style' digesters). Table 1 shows the investigated 12 AD plants based on bio-waste with their specific characteristics. Table 2 presents the amount and kind of substrate treated at the bio-waste facility. The treated bio-waste is used completely for digestion in AD plant nos. 2, 4, and 5. Most AD plants operate with partial stream digestion of bio-waste. In these plants, just the bio-waste from separate collection is used for fermentation, whereas the green cut and structure (woody) material is added after digestion within the composting process.

AD plant nos. 1, 2, and 12 were operated with open, unaerated post-composting processes. AD plant no. 3 had a covered but no enclosed composting steps. In AD plant no. 4, larger quantities of sludge from wastewater treatment were treated. Thus, primarily liquid digestate was generated. The small amounts of solid digestate were stored on site and were used for external composting. The solid digestate of AD plant no. 5 were stored open after separation. Post-composting processes with active ventilation (pressure ventilation) and enclosed composting systems were used at AD plant nos. 7, 9, and 10. A defined step of aeration in which the air is integrated into the exhaust gas treatment (bio-filter) was considered at plant no. 10.

All investigated biogas facilities operated with bio-filters as gas treatment. However, most of plant operators did not use acidic scrubbers at biogas facilities. Only four of 12 plants operated with acidic scrubbers, and the proper operation was not always ensured. Five plants used the bio-filter combined with humidifier. The exhaust gas should be treated with acid scrubbers to deposit NH_3 and minimize N_2O formation in the bio-filter (e.g., plant nos. 5 and 9). It should be recognized that there were also diffuse emission sources which were not collected by bio-filters (e.g., open doors of delivery hall at AD plant nos. 6 and 7; post-composting at AD plant nos. 8, 9, 11).

Table 1 Characteristics of investigated AD plants based on bio-waste

Plant no.	Installed electrical capacity kW_{el}	Kind of fermentation[a]	Temperature[b]	Range of temp. in °C[c]	Mode of operation	HRT in days[d]	Residues storage tank	Post-composting[e]	Type of aeration (post-composting)	External heat utilization[f]
1	630	Wet	M		Multi-stage	8	Covered	x	Open, unaerated	
2	536	Wet	T		Multi-stage	20	Covered	x	Open, unaerated	x
3	986	Wet	M		Single-stage	17	Open			
4	1200	Wet	M	37-40	Multi-stage	25	Open, covered			
5	1790	Dry	T		Single-stage	25	Gas-proof covered			
6	1413	Dry	T	55	Single-stage	21	Covered	x	Open, unaerated	
7	816	Dry	T		Single-stage	28		x	Enclosed, aerated (pressure ventilation)	
8	625	Dry	T	55	Single-stage	14	Gas-proof covered	x	Enclosed, aerated and unaerated (pressure ventilation)	x
9	640	Dry	T		Single-stage	21	Covered	x	Enclosed, aerated (pressure ventilation)	
10	625	Batch	M	37-39	Single-stage	28		x	Enclosed, aerated (pressure ventilation)	x
11	680	Batch	M	40-42	Single-stage	21		x	Open, aerated, enclosed	x
12	370	Batch	M	40-42	Single-stage	21		x	Open, unaerated	x

[a]Wet = wet fermentation, dry = dry fermentation, batch = batch system (discontinuous). [b]M = mesophilic, T = thermophilic. [c]According to the information of plant operators (if available). [d]Hydraulic retention time. [e]x = post-composting process. [f]x = external heat utilization.

Table 2 Amount and kind of treated substrate of investigated bio-waste facilities

Plant no.	Total amount of substrate input treated at facility t/a (fresh matter)	Amount of input for AD t/a (fresh matter)	Percent share of AD (mass) related to total amount treated at facility	Kind of substrate[a]
1	83.840	32.000	38	Bio-waste, green cut
2	10.062	9.865	98	Bio-waste, catering waste, green cut
3	34.976	25.702	73	Bio-waste
4	35.388	35.388	100	Sludge from wastewater treatment, catering waste, food waste, manure
5	33.130	33.130	100	Bio-waste, catering waste
6	29.900	26.910	90	Bio-waste, green cut
7	35.450	17.725	50	Bio-waste
8	20.000	12.000	60	Bio-waste, green cut
9	23.000	17.250	75	Bio-waste, green cut
10	36.000	18.000	50	Bio-waste, green cut
11	73.333	22.000	30	Bio-waste, green cut
12	13.333	12.000	90	Bio-waste, green cut, food-waste

[a]Bio-waste = bio-waste from separate collection.

Often, digestate - whether separated or not separated - is stored open temporarily or for longer periods. Four of the seven examined plants which stored liquid digestate or process waters used covered storage tank (AD plant nos. 4, 5, 8, and 9). Two plants (nos. 5 and 8) with gas-proof covered storage tank are able to use the exhaust gas by involving into the CHP.

Emission measurements

There are in general two methods to determine the emissions of a large industrial facility or areas with diffuse emission sources. One way is to attempt to capture the overall emissions of the facilities by means of concentration measurements in the surroundings and the application of inverse dispersion models [7] or radial plume mapping [14]. These methods allow the determination of the overall emissions of a large area with uncertain sources of emission. They do not allow the localization of single sources and allocation of a certain quantity to them. However, for further efficient measures to reduce emissions, it is very important to identify and quantify the emission sources on site. For this reason, the methods used focus on the identification and quantification of single sources [5].

The emission analysis included two measurement periods in each plant (each 1 week in 2010 and 2011), in which all plant components from substrate delivery to storage of digestate and composting were investigated. The measured emissions of both periods were averaged. Several sampling points at AD plant and compost heaps were examined. Following the inspection of the biogas facilities on site, potential significant emission sources within the process chain were identified. The following emission sources were investigated: delivery and conditioning of substrate (material handling), storage of fermentation residues (digestate), fermenter, before and after exhaust gas treatment (acid scrubber and bio-filter), and exhaust of CHP unit (combined heat and power plant) as well as post-composting process of digestate. The emission measurements focused on the emission detection at the AD plant and post-composting processes - not the utilization of biogas in CHP units. Therefore, not all CHP were measured. With respect to the total GHG balance, the production as well as the utilization of biogas in CHP is important. Thus, an average of CHP emissions was considered (see 'Emissions from CHP'). For the emission measurements of the composting process, four or five sections of the windrow were selected for each measurement period, which differed in time of composting resp. age of rotting material.

According to the characteristics of the gases, the applied measurement techniques were adjusted. Leakage detection techniques were used to find the critical spots within the process; open and closed domes were used to determine the main emission sources. Regarding the methods of emission measurements, there are differences between captured and diffuse emission sources. Accordingly, different measurements for emissions from encapsulated areas (e.g., delivery hall with collection of exhaust) and diffuse emission sources during several measured periods were used. Waste treatment facilities often have gas collection systems that collect air from the captured process steps and deliver the gas after a cleaning stage into the atmosphere. In most cases, the cleaning step is a bio-filter. Because of that, in all investigated AD plants, the exhaust streams before and after treatment by bio-filters were examined. Depending on the plant system, further sampling points were analyzed. In case of encapsulated emission sources, the exhaust air flow was examined directly. Thereby, the volume flow and mass concentration within the investigated pipelines were determined. The volume flows were measured with vane anemometers. The quantity of the emission source was calculated from the concentration difference and the flow rate of the blower by using the following equation [5].

$$F = Q * \rho * (c_{out} - c_{in}) \quad (1)$$

F, emission flow rate (mg/h); *Q*, air flow rate (m^3/h); ρ, density of the target gas (kg/m^3); c_{out} exhaust gas concentration (mg/kg); c_{in}, background gas concentration (mg/kg).

Emissions of post-composting with active aeration (e.g., actively ventilated tunnel or container systems) were measured by using encapsulated areas with air extraction. In case of open windrows composting without active aeration, a wind tunnel as emission measurement was used. An air flow was generated by using a ventilator. The measurement methods, techniques, and technical guidelines used for the determination of emission concentrations are shown in Table 3. CH_4 was detected by gas chromatography with a flame ionization detector (FID), N_2O by gas chromatography, and NH_3 by absorption in an acid solution. The sampling for the determination of CH_4 and N_2O was carried out by a measuring gas line which is connected to a gas analysis with online data collection. The sampling for the determination of NH_3 occurs directly at the tunnel exit. The sample gas is guided without gas cooling through two wash bottles filled with sulfuric acid. Further information according to the methods of emission measurement at biogas plants are published in [4].

Residual gas potential

The residual gas potential of digestate from anaerobic treatment of bio-waste was considered. The gas potential can be analyzed by different temperature levels as described by [16]. The temperature of the stored digestate has a great influence on the emissions. Laboratory tests within the studies of [17] and [18] showed that depending on the temperature of the digestate during storage, the emission potential can be significantly reduced. In [5], it is shown that the average CH_4 potentials obtained at 20°C represent 39% of the CH_4 potential obtained at 39°C. According to [17], the CH_4 production at a temperature of 25°C is reduced to 40–50% of the value obtained at 37°C and at 10°C, the CH_4 production goes down to even 1% [5].

In this study, the residual gas potential of digestate was determined at a temperature of 38°C. The digestate samples were taken directly after the fermentation step and - in case of separation of digestate - after separation (see AD plant nos. 1, 2, and 7). With these samples, batch experiments were carried out according to the German technical guideline VDI 4630 [19]. Finally, relative residual gas potentials with respect to the used fresh matter were determined using the following assumptions: average CH_4 yield of 74 m^3 CH_4 (STP) per metric ton fresh matter bio-waste, 10% degradation of fresh matter by the fermentation stage, and a separation ratio of 20% solid digestate to 80% liquid digestate.

Assumptions - GHG balances

Based on a survey of plant operator, additional emission-related data (e.g., energy demand, amount, and kind of heat utilization) were collected to prepare the GHG balance of each plant. For the total GHG balances, the emissions as well as credits for the kind of products (combined heat and electricity from biogas; fertilizer and humus supply from fermentation residues) were considered. The overall GHG performance of each AD plant included in particular the following: GHG emissions according to the measured components of AD plant, calculated emissions of the electricity demand

Table 3 Measurement methods, techniques and technical guidelines for the determination of emissions at the investigated AD plants [15]

Compound	Kind of determination	Measurement methods	Measurement techniques	German technical guideline used for the calculation of emissions
Total carbon	Continuously, online data	Flame ionization detection (FID)	Bernath Atomic 3006	VDI 3481 - 3, VDI 3481 - 4, DIN EN 12619, DIN EN 13526
Methane	Continuously, online data	Infra-red (IR) method	ABB Advance Optima URAS 14	
Nitrous oxide	Continuously, online data	Infra-red (IR) spectroscopy	ABB Advance Optima URAS 14	DIN EN ISO 21258
Methane	Discontinuously, laboratory analysis	Gas chromatography (GC) with autosampler	Sampling with evacuated vials	DIN EN ISO 25139
Nitrous oxide	Discontinuously, laboratory analysis	Gas chromatography (GC) with autosampler	Sampling with evacuated vials	VDI 2469 - 1
Ammonia	Discontinuously, laboratory analysis	Wet chemical methods with sulfuric acid	Sampling with Desaga-pump and 2 wash bottles	VDI 3496 - 1

(AD plant and CHP), calculated emissions during the application of the fermentation residues, credits for the electricity production from biogas (substitution of fossil electricity supply), credits for the utilization of exhaust heat (substitution of fossil heat), and credits for the use of fermentation products (substitution of fossil fertilizer and peat, humus effects).

The considered GHG emissions for all processes of bio-waste digestion were converted into CO_2 equivalents (CO_2-eq) by using characterization factors.

The following factors according to the GWP for a 100-year time period were stated: $CO_2 = 1$, $CH_4 = 25$, $N_2O = 298$ [1]. With respect to the NH_3 emissions, it is assumed that 1% of the NH_3 is converted to N_2O emissions [1].

As a functional unit of GHG balances, 'ton input bio-waste treated at facility (fresh matter)' was used. This unit included the total amount of waste treated at the facility (bio-waste and green waste - if any) - not only the amount of bio-waste in the fermentation process. In few biogas plants, municipal bio-waste from separate collection and green waste from gardens and parks were treated, but only the bio-waste is used in the step of digestion. After the fermentation process, the digestate is often combined with the green cuts within the post-composting process. Thus, the measured emissions of post-composting processes based on the treated waste at the facility in total.

In addition to the measured GHG emissions of the AD plants, further assumptions to calculate the GHG performance were considered.

Emissions from CHP

Due to the fact that not all CHP units were measured, an average emission value for the CHP is assumed. According to measurements of gewitra (personal communications), the median of CH_4 and N_2O emissions of 161 measured CHP units in the range from 300 to 1,000 kW_{el} were determined with 1,760 g CH_4 per ton of bio-waste and 2.1 g of N_2O per ton of bio-waste treated at the facility. Considering the emission factors [1] for N_2O (298) and CH_4 (25), a GWP of 44.6 kg CO_2-eq per ton of bio-waste was estimated for all CHP units.

The energy demand of the investigated biogas plants was determined according to the data of plant operators. It was estimated to cover the electricity demand by using external electricity from the grid. The electricity production in Germany in 2011 produced in average 559 g CO_2-eq per kWh_{el} [20].

Electricity production

The electricity production from biogas replaces fossil fuels and can be considered as credit [21]. The amount of credit for the electricity production depends on the amount of produced electricity referring to the data of plant operators. The electricity mix of Germany in 2011 with 559 g CO_2-eq per kWh_{el} [20] was assumed to calculate the credit of electricity production.

Heat utilization

The exhaust heat of electricity generation in CHP units can - if used - substitute heat production based on fossil fuels [21]. The avoided GHG emissions of fossil heat supply by providing heat for external utilization (e.g., district heating, drying process) was stated as heat credits. The amount of heat credit may vary depending on the amount of heat and type of fossil heat, which is replaced in the specific case. With regard to the substitution of fossil heat, an average of the specified external heat mix of 291 g CO_2-eq per kWh_{th} [21] was used to calculate the heat credits.

Digestate - fertilizer and humus effects

Depending on the kind of digestate, respectively, the kind of treatment of the fermentation residues (e.g., with/without separation, with/without post-composting after fermentation process), different utilization pathways of digestate have been considered. According to the kind of digestate (finished compost, fresh compost, liquid fermentation residues, solid digestate), different GHG emissions can be saved and considered to the GHG balances as credits (Table 1). Referring to the kind of digestate, the following credits were determined: substitution of mineral fertilizer (nitrogen, phosphorus, potassium), substitution of peat (only in case of finished compost), humus accumulation (carbon-sink), and humus reproduction (i.e., for maintaining soil fertility).

According to the nutrient content (i.e., nitrogen, phosphorus, potassium amounts) of investigated digestates, the production of mineral fertilizer can be substituted and is stated in GHG balances as credit. The following emission factors for the production of mineral fertilizer were assumed according to [22]: 6.41 kg CO_2-eq per kg nitrogen (N), 1.18 kg CO^2-eq per kg phosphorus (P_2O_5), and 0.663 kg CO_2-eq per kg potassium (K_2O).

Humus effects of digestate at investigated AD plants were considered if applied on agricultural land. To evaluate the humus effects of fermentation residues, estimations according to [23] were used. That means, for the amount of finished compost, 20% substitution of peat and 80% agricultural use, thereof 20% of humus accumulation and 80% of humus reproduction was assumed. For the scenario of humus, reproduction was stated - in contrast to [23] - that the substitution of straw is considered and the credits for the fermentation of straw with recirculation of the digestate can be estimated. The humus reproduction (i.e., for maintaining soil fertility) of digestates depends on the content of dry matter and organic dry matter as well as the degrading stability of organic dry matter. Data regarding the humus reproduction of digestate from AD based on bio-waste are not available. The humus reproduction of digestate of investigated AD plants was calculated. The characteristics (e.g., dry matter, organic dry matter, amount of nutrients especially nitrogen) of each digestate were determined based on the 1-year certificate of digestate referring to the quality assurance of the Federal Compost Association.

According to the kind of digestate, the substitution effect compared to straw was analyzed. Therefore, the amount of straw was calculated which might be used for biogas production if the application of digestate on agricultural land is assumed. Differed to the kind of digestate, the amount of straw per ton of digestate (fresh matter) was calculated as follows: 2.11 (finished compost), 1.82 (fresh compost), 0.91 (digestate with post-composting), and 0.15 (liquid digestate). The electricity production of the assumed biogas production due to the fermentation of straw was considered as credit for humus reproduction of digestate.

The substitution of peat was estimated only in case of finished compost. According to the assumptions in [24], 1 kg dry peat (respectively, 2 kg fossil carbon dioxide) is replaced by 1 kg compost (organic dry matter). Referring to the humus accumulation (carbon sink) of composted digestate, the amount of organic carbon (Corg) as published in [23] was assumed as follows: 21.6 kg Corg per ton of digestate for fresh compost and 64.5 kg Corg per ton of digestate for finished compost. In consideration of the stoichiometric ratio of Corg relative to CO_2, 1 kg Corg can fix 3.7 kg CO_2.

Application of digestate

The application of digestate on agricultural land can cause N_2O emissions as well as NH_3 emissions [25]. With respect to the NH_3 emissions, it was assumed that 1% of the NH_3 is converted to N_2O emissions [1].

Results and discussion

GHG emissions

Various fermentation processes such as wet fermentation, dry fermentation, and batch fermentation were analyzed according to the emission situation. The results show that the emissions are dominated not by the kind of the fermentation process or the technology but by the manner of plant operation.

Figure 1 shows the measured emissions of CH_4, N_2O, and NH_3 (converted to carbon dioxide equivalents) of the investigated AD plants. The range of determined plant emissions varied between 40 and 320 kg CO_2-eq per ton of bio-waste. The detailed presentation on the type of GHGs shows that the CH_4 emissions - except for plant no. 6 - dominate the indicated GHG equivalents of biogas facilities.

Important sources of GHG emissions were identified. The component-specific GHG emissions of the bio-waste digestion plants are presented in Figure 2.

Especially, the inadequate aeration directly after fermentation (in order to interrupt the methanogenic activity) processes as well as unaerated or less aerated post-composting processes caused extremely high GHG emissions (see plant no. 1, no. 2, or no. 12). In case of some of the investigated biogas plants, the emissions of post-composting are summarized in the amount of 'emissions after bio-filter' (e.g., AD plant no. 10). The overall emissions of AD plant no. 10 was quite low because all parts of the fermentation and post-composting process were totally encapsulated.

Furthermore, AD plant no. 6 showed higher NH_3 emissions due to the drying of digestate at higher temperature and higher pH value. In this case, the existing downstream acidic scrubber was out of operation during the measurements. The operation of the bio-filters can also be problematic; extremely wet bio-filters for example can cause additional CH_4 production as observed at AD plant no. 8.

Finally, on almost all AD plants, emission sources were identified whose intensity can be reduced if the state-of-the-art treatment technology was used (e.g., acid scrubber before bio-filter, aeration of post-composting). The results show that the open storage of fermentation residues (with or without separation step) should be avoided. In addition to unaerated post-composting processes and open storage of active material (e.g., solid digestate), the CHP was one of the most important sources of CH_4.

According to the measured residual gas potential of digestate, a wide range from 4 to 23% was determined. Ten of 12 samples of digestate of investigated AD plants showed a relative residual gas potential higher than 10%. A high relative residual gas potential means insufficient fermentation of the substrate. The residual gas potential of bio-waste digestion achieved the same range as agricultural AD plants which were operated as single-stage processes, whereas in comparison to agricultural biogas plants with multi-stage process, the determined CH_4 potential of fermentation residues from bio-waste digestion provides basically higher values. Table 4 shows the gas potential of the investigated bio-waste plants compared to the gas potential of agricultural biogas plants as published in [17]. According to [17] where agricultural AD plants were investigated, discontinuous systems (batch) and single-stage systems have shown the highest residual gas potential. Moreover, multi-stage systems of agricultural AD plants achieved less than half of the residual gas potential of single-stage plants [17]. The results of [17] stated that single-stage processes achieve higher residual gas potential due to their generally shorter retention time. With respect to the investigated bio-waste AD plants

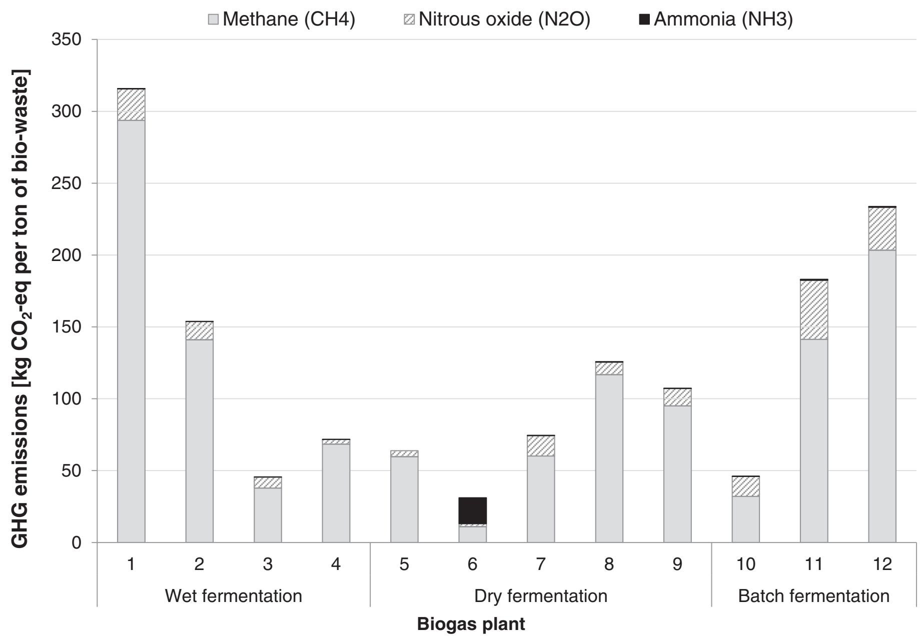

Figure 1 GHG emissions of the investigated biogas facilities (bio-waste) differed to the kind of GHG emission. The measured emissions of investigated AD plants are presented in kg CO_2-eq per ton of bio-waste differed to the kind of GHG emission and kind of fermentation process. Methane and nitrous oxide emissions = direct GHG emissions, ammonia = indirect GHG emissions.

hydraulic retention times (HRTs) ranged from 1 to 4 weeks. However, due to a great variability of other process parameters, the results do not give a clear answer regarding the estimation that lower HRT corresponds to lower gas potential (see Tables 5 and 6).

GHG balances

The overall GHG balance of the investigated AD plants depends on the measured GHG emissions on the one hand (see 'GHG emissions') and on the credits for the generated products (e.g., combined heat and electricity

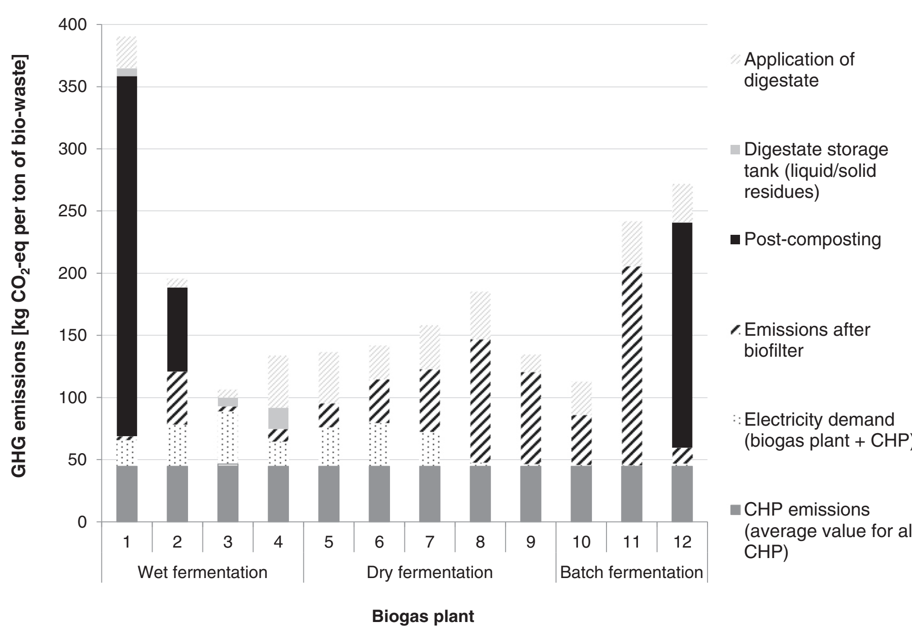

Figure 2 GHG emissions of bio-waste digestion plants differed to kind of plant components. The GHG emissions are presented in kg CO_2-eq per ton of bio-waste differed to the kind of fermentation process (wet, dry, or batch fermentation) and the main emission sources within the process chain.

Table 4 Investigated AD plants differed to kind of digestate and considered GHG credits (marked with 'x')

	Kind of digestate	Solid digestate				Liquid digestate	
		Substitution of mineral fertilizer	Substitution of peat	Humus accumulation	Humus reproduction	Substitution of mineral fertilizer	Humus reproduction
1	Fresh compost	x	-	x	x	-	-
2	Finished compost	x	x	x	x	-	-
3	Solid digestate (separated)	x	-	-	x	-	-
4	Liquid digestate	x	-	-	x	x	x
5	Separated digestate without post-composting	x	-	-	x	x	x
6[a]	Solid digestate	x		-	x	-	-
7	Fresh and finished compost	x	x	x	x	-	-
8	Fresh and finished compost	x	x	x	x	x	x
9[b]	Finished compost	x	x	x	x	x	x
10	Finished compost	x	x	x	x	-	-
11	Fresh and finished compost	x	x	x	x	-	-
12	Finished compost	x	x	x	x	-	-

[a]Assumption according to plant no. 5 (no data available). [b]Assumption according to plant no. 8 (no data available).

from biogas; fertilizer and humus supply from fermentation residues) on the other hand. The calculated GHG credits according to the AD plant concept are presented in Figure 3.

Finally, the highest amount of GHG credits of humus reproduction can be expected from composted digestate. In general, the following order of humus reproduction can be assumed: post-composted digestate (finished and fresh compost) > solid digestate > liquid digestate. In case of finished compost, additional GHG credits for the substitution of peat (by application in soil producing facilities, e.g.) can be considered.

If external heat (generated by the electricity production of CHP unit) is utilized, credits for avoided fossil heat production optimize the GHG balances as well (see plant no. 12). Nevertheless, in most cases (besides plants nos. 1, 7, 10, and 11), the credit for the electricity production based on biogas which was given for the substitution of fossil fuels dominates the GHG credits.

The total range of GHG balances (including credits) varied between −49 and 323 kg CO_2-eq per ton of bio-waste due to different plant concepts and measured emissions (see Figure 4).

Moreover, the emissions of each component have been set in relation to the amount of produced electricity in order to get an emission value according to the energy output (g CH_4/kWh_{el}). Compared to an assumed electricity mix in Germany (559 g CO_2-eq per kWh_{el} according to [20]), 8 of 12 AD plants show even lower values.

Table 5 Residual gas potential in percent related to the methane production

	Bio-waste AD plants 38°C	Agricultural AD plants 37°C	
		Single-stage	Multi-stage
Average	14.1	9.7	5.1
Min	3.6	3.2	1.2
Max	23.4	21.8	15

Investigated bio-waste AD plants in comparison to agricultural AD plants according to Weiland et al. [17] and temperature level for determination of gas potential.

Overall discussion of results gained in this study

The problem of increased emissions is not the anaerobic process itself, but a non-optimal after-treatment of the digestate. In general, the emission situation is not uniform; the plants show very different emission rates. The total emissions from AD plants no. 3, no. 6, and no. 10 were quite lower than the remaining. However, even those plants showed considerable potential for optimization. The best overall result of the analyzed AD plants belonged to a biogas facility with no external heat utilization and below-average credits for digestate. It can be stated that all investigated biogas facilities showed potential for optimization. Often, there are no incentives for a sufficient utilization of waste with respect to high CH_4 yields or reduction of emissions, due to the fact that the running costs of waste facilities has to be financed by the producers of the waste paying for the waste disposal. Moreover, there are no strict regulations to avoid uncontrolled emissions as for agricultural biogas plants for energy crops and for co-digestion of waste. Therefore, waste treatment plants show relevant potentials for optimization.

AD plant no. 12 showed that very high emissions can be covered by a very good energy concept combined with a good utilization of fermentation residues. The bad overall

Table 6 Hydraulic retention time and residual gas potential of investigated AD plants

Plant no.	HRT in days[a]	Relative residual gas potential in percent[b]		Kind of fermentation[c]	Mode of operation	Separation of digestate
		Minimum	Maximum			
1	8	11	15	Wet	Multi-stage	Yes
2	20	19	19	Wet	Multi-stage	Yes
3	17	-	-	Wet	Single-stage	No
4	25	-	-	Wet	Multi-stage	No
5	25	-	-	Dry	Single-stage	Yes
6	21	15	21	Dry	Single-stage	No
7	28	4	11	Dry	Single-stage	No
8	14	23	23	Dry	Single-stage	No
9	21	17	17	Dry	Single-stage	No
10	28	12	17	Batch	Single-stage	No
11	21	-	-	Batch	Single-stage	No
12	21	6	6	Batch	Single-stage	No

[a]Hydraulic retention time in days. [b]Residual gas potential of digestate according to the input material (fresh matter based). [c]Wet = wet fermentation, dry = dry fermentation, batch = batch system (discontinuous).

GHG balance of AD plant no. 1 evidences how certain factors may interact the GHG performance negatively. In this case, extremely high emissions occurring from the post-composting process and very low electricity generation caused high GHG emissions in total. Inadequate digestion of the substrate caused not only low gas production, respectively, electricity generation but also high emissions during the post-composting process of digestate.

Regarding the GHG credits, the highest importance of an efficient fermentation had the production of energy. A high share of electricity generation led to high GHG credits. As far as the utilization of exhaust heat of electricity production was possible, it had also a positive influence on the GHG performance of the AD plant. Moreover, the use of digestate showed positive effects on the GHG balances. In addition to the nutrient effect through the utilization of the fermentation residues as a fertilizer (substitution of mineral fertilizer), GHG emissions can be saved due to the humus effect of digestate. Especially, composted digestate like fresh and finished compost contributed to the humus accumulation

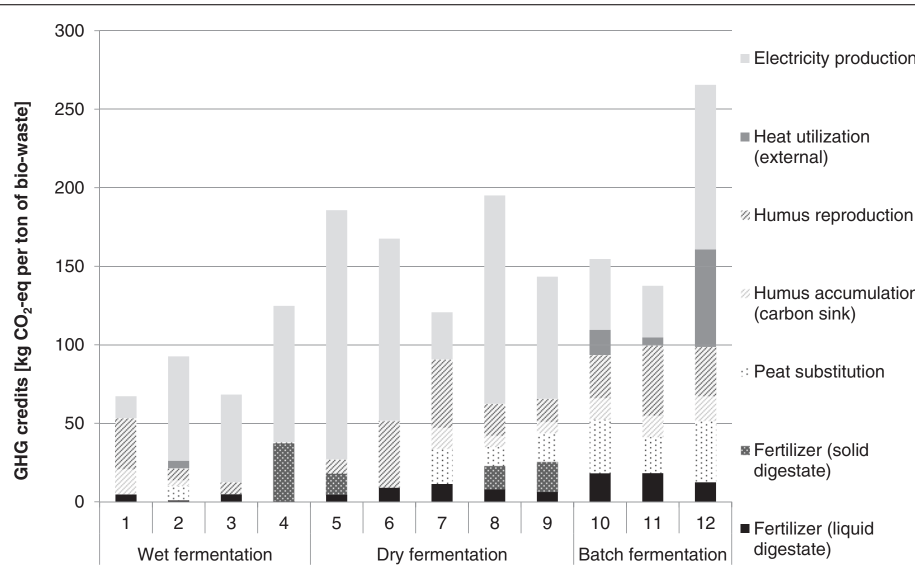

Figure 3 GHG credits of investigated bio-waste digestion plants. GHG credits depend on the amount of energy production or heat utilization (substitution of fossil electricity/fossil heat production) as well as the kind and amount of digestate (substitution of fertilizer, substitution of peat, humus effects).

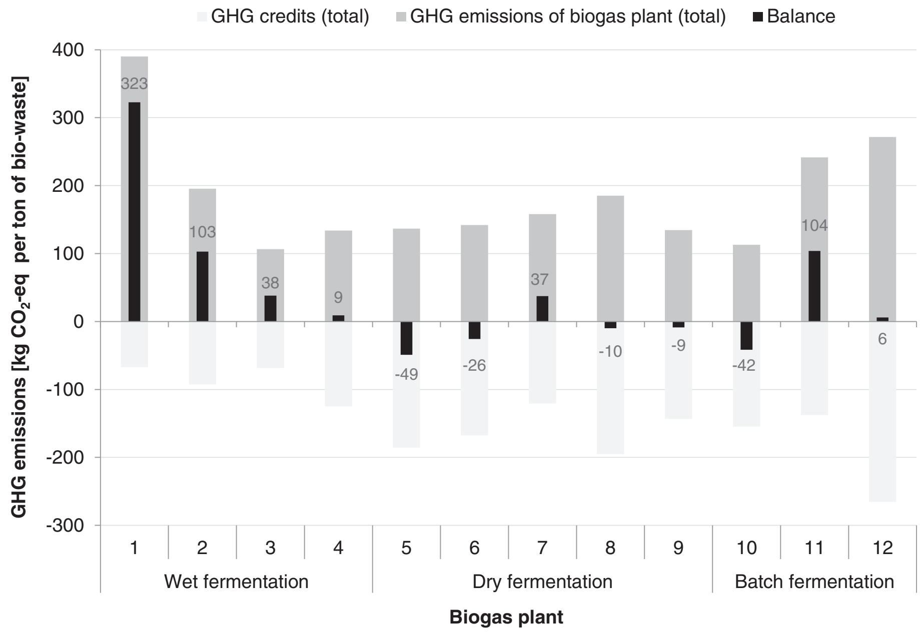

Figure 4 Total GHG balance of bio-waste digestion plants with GHG emissions of AD plant and GHG credits. The balance as a result of total GHG emissions of AD plant and total GHG credits (*black column*).

(carbon sink) and the humus reproduction of digestate. Compared to the production of fresh or finished compost digestate without post-composting process, which is used within the agriculture directly, less GHG credits were given. However, the risk of high emissions during the post-treatment of the fermentation residues was avoided.

The following measures are able to reduce GHG emission of bio-waste digestion: intensive aeration of the (solid) digestate after fermentation; gas-tight storage tank for fermentation residue and integration into biogas utilization; avoidance of any open storage of digestate and fermentation residues; and small, aerated compost windrows combined with sufficient structural materials and frequent turnover as well as the use of acidic scrubbers in front of the bio-filter.

With respect to the development of methodology of emission measurements and the standardization of procedure for the determination of emissions on biogas plants, further investigations are necessary. Further scientific data about the current emission situation and the ongoing development as well as reliable measurement methods are required to determine the CH_4 emissions from the plants in operation today. In this regard, the reliable measurement of stationary and diffuse emission sources is of high importance. Uncertain are the emissions sources that are not coupled to the gas system of the plant, but still cause GHG emissions as stated in [10]. As one example, no assessment of emissions from pressure relief valves could be carried out as part of this study. Concerning the emissions, the treatment and evaluation of temporary occurring emissions caused by certain operational conditions are still unclear. Moreover, the further development of ecological assessment of biogas pathways with respect to the humus effects of digestate in comparison to other pathways is of great importance.

Conclusions

Based on the emission measurements, significant sources of emissions were identified. The results show that GHG emissions can be minimized, if the technology and operation of the plant are adjusted accordingly. Basically, the kind of operation of the plant and the handling of digestate determine the amount of GHG emissions. The overall GHG balances of the investigated AD plants depend on the measured emissions as well as the amount of credits for the generated products (e.g., combined heat and electricity from biogas; fertilizer and humus effects from fermentation residues). The consideration of GHG credits can optimize the overall GHG performance of the biogas facilities.

Abbreviations

AD: anaerobic digestion; C: carbon; CHP: combined heat and power unit; CH_4: methane; CO_2: carbon dioxide; CO_2-eq: carbon dioxide equivalent; Corg: organic carbon; GHG: greenhouse gas; GWP: global warming potential; K_2O: potassium oxide; kW_{el}: kilowatt (electrical); kWh_{el}: kilowatt hours (electrical); kWh_{th}: kilowatt hours (thermical); N: nitrogen; NH_3: ammonia; No.: number; N_2O: nitrous oxide; STP: standard temperature pressure; t: metric ton.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

JDG carried out the total GHG balances including GHG credits and conceived of the papers structure. JL and CK conducted the emission analysis regarding the biogas plant and the components. JL, CK, and JDG described the mitigation potential. VD was responsible for the survey of plant operators and the assessment of these data. All authors drafted, read, and approved the final manuscript.

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Dedication

This publication is dedicated to Prof. Andreas Zehnsdorf on the occasion of his 50th birthday.

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