Methane Production during Storage of Anaerobically Digested Municipal Organic Waste

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ABSTRACT

Anaerobic digestion of source-separated municipal organic waste is considered feasible in Denmark. The limited hydraulic retention in the biogas reactor (typically 15 d) does not allow full degradation of the organic waste. Storage of anaerobically digested municipal organic waste can therefore be a source of methane (CH_4) emission that may contribute significantly to the potential global warming impact from the waste treatment system. This study provides a model for quantifying the CH_4 production from stored co-digested municipal organic waste and estimates the production under typical Danish climatic conditions, thus quantifying the potential global warming impact from storage of the digested municipal organic waste before its use on agricultural land. Laboratory batch tests on CH_4 production as well as temperature measurements in eight full-scale storage tanks provided data for developing a model estimating the CH_4 production in storage tanks containing digested municipal organic waste. The temperatures measured in separate storage tanks on farms receiving digested slurry were linearly correlated with air temperature. In storage tanks receiving slurry directly from biogas reactors, significantly higher temperatures were measured due to the high temperatures of the effluent from the reactor. Storage tanks on Danish farms are typically emptied in April and have a constant inflow of digested material. During the warmest months the content of digested material is therefore low, which limits the yearly CH_4 production from storage.

DURING ANAEROBIC DIGESTION of municipal organic waste in biogas plants, 75 to 85% of the organic matter is typically degraded (Davidsson et al., 2006), leaving 15 to 25% that may be degraded in the storage tank where the digested waste is often kept for months before application to agricultural land. According to Danish regulations, organic waste and manure can be spread on land only twice a year to enhance nutrient uptake and limit loss to surface water and ground water. Since CH_4 has a global warming potential 23 times that of CO_2 , CH_4 emissions from storage tanks for anaerobically digested organic waste may considerably affect the global warming impact from the waste management system. These emissions have not previously been investigated and the significance of CH_4 emissions from digested organic waste is unknown. Therefore, quantification of CH_4 emissions from storage tanks is essential for environmental assessment of treatment of municipal organic waste in biogas plants.

The CH_4 production from storage of digested waste is governed by degradability of the material, temperature,

and retention time in the storage tank. Whether the produced CH_4 is emitted to the atmosphere depends on such factors as the physical features of the storage tank and whether the tank is covered and has gas collection. The Intergovernmental Panel on Climate Change (IPCC) has not developed guidelines for estimating methane emissions from storage of digested organic waste. However, according to the IPCC Good Practice Guidelines (Intergovernmental Panel on Climate Change, 2001), reliable estimates of CH_4 emissions from storage of manure should consider the amount of volatile solids (VS), time of storage, CH_4 potential of the material (VS basis), and a CH_4 conversion factor depending on the manure management system and climate region. Hence, these factors should also be included when determining the potential CH_4 emissions from storage of anaerobically digested municipal organic waste.

Methane emissions from storage of anaerobically digested municipal organic waste could not be measured directly, since co-digestion with either manure or sewage sludge is the most common anaerobic treatment technology used for municipal organic waste in Denmark. Accordingly, investigations of emissions from storage tanks have typically concentrated on digested manure and not on municipal organic waste.

The objective of this study was to model possible CH_4 production in storage tanks. This modeling was based on measurements of temperature and filling degree in full-scale storage tanks as well as the results of laboratory batch tests, where the methane production of digested municipal organic waste was determined at seven different temperatures (5–55°C).

MATERIALS AND METHODS

Methane Production Tests

The CH_4 production of anaerobically digested municipal organic waste was determined by batch tests on the effluent from pilot-scale biogas reactors treating only source-sorted municipal organic waste collected in the city of Aalborg. The stirred pilot-scale reactors of 35 L were thermophilic (55°C) and continuously fed with waste diluted to 5% dry matter. The hydraulic retention time was 15 d, which is a normal hydraulic retention time for Danish biogas plants. The reactors were run for 2 to 3 mo and the effluent for the batch tests was sampled during periods with stable gas production. The pilot-scale biogas experiments are further described in Davidsson et al. (2006). Table 1 shows the chemical composition of the digested waste with respect to dry matter, VS, ash, fat, fibers, nitrogen, phosphorus, potassium, carbon, hydrogen, sulfur, chloride, and calorific value. Furthermore, the enzyme degradable organic

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Abbreviations: EDOM, enzyme degradable organic matter; Nm^3 , normal cubic meter (one cubic meter at 0°C and 1 atmosphere); VS, volatile solids (the loss from oxidation of a dried sample at 550°C, % of dry matter).

Table 1. Composition of effluent from anaerobic pilot-scale digestion of source-sorted municipal organic waste (collected and pre-treated in the Danish city of Aalborg) used in the methane production batch tests.

Property	Unit	Value
Dry matter	% of wet wt.	1.3
Volatile solids	% of dry matter	60.3
Ash	% of dry matter	39.7
Fat	% of dry matter	4.1
Fibers	% of dry matter	9.0
N (total)	% of dry matter	8.9
N (NH ₄ ⁺)	% of dry matter	4.7
P	% of dry matter	1.0
K	% of dry matter	4.9
C	% of dry matter	32.0
H	% of dry matter	4.3
S	% of dry matter	0.6
Cl	% of dry matter	4.3
Calorific value	MJ/kg dry matter	14.5
EDOM†	% of VS‡	56.5

† Enzyme degradable organic matter.

‡ Volatile solids.

matter (EDOM) was determined by treating the waste samples with acid and heat followed by washing and treatment with three types of enzyme to determine the enzyme degradable fraction. The analytical procedures are described in detail in Hansen et al. (2006b).

The batch tests for determining the CH₄ production of the anaerobically digested waste were performed in triplicate in 2-L glass bottles according to the procedure described in Hansen et al. (2004). The tests covered a broad spectrum of seven different temperatures representing possible conditions in storage tanks in Denmark: 5, 10, 15, 22, 28, 35, and 55°C. No inoculum was used, since the tests should determine the actual CH₄ production in a storage tank for anaerobically digested organic waste. For each temperature, parallel tests were performed where cellulose was added to the effluent samples (triplicates) to test whether slow or missing CH₄ production was caused by lack of degradable organic matter or microbial inactivity due to low temperature. The batch tests lasted 125 d.

The results from the batch tests were given as the accumulated CH₄ production over time (Nm³ CH₄/g VS, where Nm³ is one cubic meter at 0°C and 1 atmosphere) per gram of VS in the digested material.

Temperature and Filling Degree of Storage Tanks

To estimate the CH₄ production from stored anaerobically digested municipal organic waste, the temperature and amount of stored waste must be known. At Danish biogas plants municipal organic waste usually constitutes less than 5% of the material digested. Manure, sewage sludge, and other organic wastes constitute the main part. Therefore, it was assumed that there were no differences in temperature and filling degree between storage tanks containing only anaerobically digested manure and those containing anaerobically digested manure mixed with municipal organic waste. The temperature and depth of digested manure was measured with 3-wk intervals throughout 1 yr in eight full-scale storage tanks containing anaerobically digested manure.

Three of the eight tanks were separate tanks, tank volumes of 2800 to 4000 m³, located on farms receiving anaerobically digested manure by truck from the biogas plants (Blåskærvej, Lintrup, and Kolstrupvej). The tanks all had a floating surface cover of straw, but no gas collection.

Five of the eight storage tanks were located at biogas plants and directly connected to the main reactor (Linkogas, Filskov,

and Århus 1–3). These tanks were all covered and the gas was collected together with gas from the biogas reactor. The volumes of the tanks were 1000 to 3000 m³.

The temperature was measured with thermocouple sensors (Type T, data collection unit Type 1033-130K; Eltek, Haslingfield, UK) mounted on an 8-m pole at 0.1, 1.1, 2.1, 3.1, and 4.1 m from the tip inserted in the storage tank. Calibration of the sensors showed variations of 0.5%. The vertical, horizontal, and 45° temperature profiles were determined; in tanks with low slurry content the horizontal and 45° angle temperature profiles could not be determined. The depth of the slurry was also measured to determine the volume of digested material in the tanks at each measurement.

RESULTS AND DISCUSSION

Methane Production Tests

Methane production from the digested waste was slow in batch tests at temperatures lower than 28°C. At 5°C, the CH₄ production was almost undetectable, both for the digested waste and the cellulose sample (Fig. 1), indicating that the microorganisms in the reactors were not capable of degradation at this low temperature. At 10, 15, and 22°C, the CH₄ production was low and the accumulated CH₄ production over time was linear (see Fig. 1). At 10 and 15°C, the CH₄ production from the digested waste and the cellulose was of the same order of magnitude, whereas at 22°C the cellulose produced 10 times as much CH₄ per kg VS as did the digested waste. This indicates that low degradability of the organic matter in the digested waste limited the degradation process at 22°C. At 28 and 35°C, the CH₄ production was substantial in the early phases, but decreased after about 70 and 50 d, respectively, indicating depletion of all easily degradable matter in the digested material. Furthermore, within 50 d at 35°C, the accumulated CH₄ production from the cellulose samples reached the theoretical maximal CH₄ potential determined by the Buswell formula (Symons and Buswell, 1933). At 55°C, the CH₄ production rate decreased after 10 d, but continued for another 60 d. The cellulose samples reached the theoretical CH₄ potential within 15 d at this temperature. The microorganisms present in the effluent were already adapted to 55°C and therefore fast and effectively degraded easily degradable matter at this temperature. The degradation of slowly degradable organic matter (in this case biogas plant effluent) was, as expected, slower.

The average CH₄ production for the effluent material obtained in the batch tests at 55°C was 225 Nm³ CH₄/Mg VS (see Fig. 1). In Hansen et al. (2006a) the measured average CH₄ potential for municipal organic waste at 55°C was found to be 460 Nm³ CH₄/Mg VS with the average EDOM value being about 90% of VS (equaling 510 Nm³ CH₄/Mg degradable VS). Using the EDOM value in Table 1 as a measure of degradability, the maximal obtainable CH₄ production for the digested waste is 285 Nm³ CH₄/Mg VS. Thus, around 80% of the degradable VS in the digested waste, as determined by the VS and EDOM of the material, was degraded in the batch tests at 55°C.

Fig. 1. Accumulated methane production over time (days) for anaerobically digested organic municipal waste and cellulose. The methane production was measured in batch tests (triplicates) at seven temperatures: 5, 10, 15, 22, 28, 35, and 55°C. Nm³, cubic meter at 0°C and 1 atmosphere pressure; VS, volatile solids in the organic waste.

The Methane Production Model

The results from the batch tests showed an approximately linear CH₄ production rate over time in the beginning of the tests for the digested waste samples. As the easily degradable organic matter is degraded, the CH₄ production decreases until finally no degradable VS is present. The CH₄ production rate (Nm³ CH₄/Mg VS h) was determined from the linear part of the CH₄ production curves for the investigated temperatures (5–55°C) and is, therefore, valid only within the period where substrate for CH₄ production is abundant. For the temperature range 5 to 35°C an exponential correlation between temperature and CH₄ production rate was

found (see Fig. 2 and Eq. [1]). The rate for 55°C was not included, since this temperature is normally not relevant for storage tanks under Danish conditions:

$$E_{\text{CH}_4} = 0.0004e^{0.159t}, R^2 = 0.98 \quad [1]$$

where E_{CH_4} is the production rate for methane (Nm³ CH₄/Mg VS h) and t is the temperature of the digested waste within the interval 5 to 35°C.

The results of the batch tests showed a clear linear relation between the log-transformed estimated CH₄ production and the inverse temperature in Kelvin ($R^2 = 0.98$) as was also proven for measured CH₄ emissions by Husted (1994), Sommer et al. (2000), and Khan et al. (1997).

Fig. 2. Estimated correlation between temperature and methane production rate for anaerobically digested organic municipal waste based on laboratory batch tests. Nm³, cubic meter at 0°C and 1 atmosphere pressure; VS, volatile solids in the organic waste.

At the lowest temperatures (5 and 10°C) the production of CH₄ within the first 24 h was relatively high compared to the overall production. When the effluent entered the batch reactors it still had the temperature of the pilot-scale reactors (55°C) and, therefore, a comparably higher microbial activity. This activity decreased as the material cooled. However, this initial production was included in the linear determination of the CH₄ production rate.

Figure 3 shows the calculated cumulative CH₄ production from storage of 1 Mg of VS over 1 yr at different temperatures, assuming constant CH₄ production as determined by Eq. [1]. The horizontal line represents the obtainable CH₄ potential (VS basis) assuming a maximal CH₄ potential of 510 Nm³ CH₄/Mg degradable VS (Hansen et al., 2006a) and a maximal degradability of 56% of the VS (EDOM value in Table 1). With storage of digested waste over 1 yr at temperatures at 22°C or lower, the degradable organic matter is unlikely to be the limiting factor for the CH₄ production. At these temperatures, the CH₄ production can, therefore, be assumed to be linear over time, which is a basic assumption for the model estimates. At 28°C the degradable organic matter will be depleted within approximately 7 mo and at 35°C within 4 mo. With storage at these temperatures the linear CH₄ production cannot be assumed to be valid beyond these time limits.

Fig. 3. Linear estimation of methane production from stored anaerobically digested organic municipal waste (1 Mg VS) over time. The horizontal curve represents the maximal obtainable methane production from the waste. Nm³, cubic meter at 0°C and 1 atmosphere pressure; VS, volatile solids in the organic waste.

When using the batch test results for estimation of CH₄ production in full-scale storage tanks, it should be considered whether the microorganisms in the full-scale tanks, which are well adapted to the low temperatures, would be more productive than those in the batch tests adapted to 55°C. It was, however, assumed that the length of the tests (125 d) allowed adaptation of the microorganisms to the lower temperature and this is, therefore, not considered a crucial uncertainty.

Temperature and Filling Degree

The horizontal, vertical, and 45° temperature profiles in the storage tanks showed little variation in space and no clear patterns. Wind and temperature gradients in the stored slurry may enhance natural convection, thereby contributing to homogeneous temperatures in the storage tanks (Olesen and Sommer, 1993). Therefore, the average temperature from each occasion of measuring temperature in the stored slurry was used (Fig. 4) and related to the average monthly Danish air temperature based on 30 yr of monitoring (www.dmi.dk, verified 31 Jan. 2006). Figure 4a shows that the temperature of the separate storage tanks for digested manure was generally slightly higher than the air temperature. This indicates that the temperature in farm tanks may be slightly elevated due to microbial activity in the digested material, but that the temperature is mainly affected by

Fig. 4. Temperature in storage tanks measured over 1 yr in Denmark. Figure 4a shows measured temperatures in storage tanks for anaerobically digested manure in separate storage tanks, while Fig. 4b shows measured temperatures in storage tanks for anaerobically digested manure directly connected to the biogas reactor.

Fig. 5. Correlation between temperature measured in separate storage tanks for anaerobically digested manure and the Danish average monthly temperature.

average air temperature. A linear regression of the temperature of the material in the tanks as a function of the average monthly Danish air temperature (Fig. 5) showed the following correlation:

$$t_{sl} = 0.75t_{air} + 6.3, R^2 = 0.8 \quad [2]$$

where t_{sl} is the slurry temperature in the tank (°C) and t_{air} is the air temperature (°C, monthly Danish average, www.dmi.dk).

The measured temperatures in storage tanks directly connected to biogas reactors are shown in Fig. 4b. These were significantly higher than the average monthly temperature and mainly depended on the temperature of the effluent from the reactor. The temperature variations in these tanks were larger than in the separate storage tanks due to intermittent inflow of heated effluent from the biogas reactors.

The digested material is applied to agricultural fields in April and the tanks are then emptied. From April the storage tanks are gradually filled until the following spring. This pattern reflects the Danish regulations enforcing farmers to apply slurry in spring at the beginning of the growth season. The addition of slurry to the storage tanks generally varies only slightly, since the biogas plant treats the same amount of manure throughout the year.

Fig. 6. Filling degree of separate storage tanks for anaerobically digested manure measured over 1 yr.

Estimation of Methane Production in Storage Tanks

The CH_4 production from storage tanks for anaerobically digested organic waste was modeled from Eq. [1] and assumptions based on the field measurements: (i) the temperature of the digested material in the separate storage tanks follows the average air temperature with respect to Eq. [2], (ii) the tanks are emptied in April, and (iii) the tanks have a constant monthly inflow (see Table 2). Over 1 yr the CH_4 production from the digested material was estimated to be $0.08 \text{ Nm}^3 \text{ CH}_4/\text{average Mg digested waste}$. Assuming 1.3% dry matter and 60.3% VS (see Table 1), this CH_4 production equals $10 \text{ Nm}^3 \text{ CH}_4/\text{Mg VS}$ in the digested waste or $2 \text{ Nm}^3 \text{ CH}_4/\text{Mg VS}$ in the waste entering the biogas reactor [assuming 80% degradation as measured in the pilot-scale reactors in Davidsson et al. (2006)]. This equals 0.4% of the CH_4 potential of the waste going into the biogas reactor ($460 \text{ Nm}^3 \text{ CH}_4/\text{Mg VS}$). The estimated CH_4 production represents the maximal potential (worst case) emissions, assuming no cover of the storage tank and no oxidation of CH_4 in the tank. In Denmark, slurry in open tanks must be covered by a layer of straw, floating granules of leca pebbles, or similar to reduce ammonia emission. It has been shown that these porous covers may sig-

Table 2. Estimated methane production in a 1200-m^3 storage tank for anaerobically digested organic waste. The tank is emptied in April, while the inflow of digested material is distributed over the year.†

Month	Volume of digested material	VS‡ in storage	Temperature, Danish mean	CH_4 production rate	Potential CH_4 emission
	m^3	Mg	°C	$\text{Nm}^3 \text{ CH}_4/\text{Mg VS month}$	$\text{Nm}^3 \text{ CH}_4$
January	850	6.2	0	0.78	4.8
February	950	6.9	0	0.78	5.4
March	1050	7.6	2.1	1.01	7.7
April	1150	8.3	5.7	1.55	12.9
May	50	0.4	10.8	2.84	1.0
June	150	1.1	14.2	4.26	4.6
July	250	1.8	15.6	5.04	9.1
August	350	2.5	15.7	5.10	12.9
September	450	3.3	12.7	3.57	11.6
October	550	4.0	9.1	2.32	9.2
November	650	4.7	4.7	1.37	6.5
December	750	5.4	1.6	0.95	5.2
					90.9 m^3
					0.08 m^3/Mg

† Dry matter content = 1.3%; volatile solid content = 60.3% of dry matter.

‡ Volatile solids.

§ Normal cubic meter (one cubic meter at 0°C and 1 atmosphere).

Table 3. Estimated methane production in a 1200-m³ storage tank for anaerobically digested organic waste. The tank is emptied in September, while the inflow of digested material is distributed over the year.†

Month	Volume of digested material	VS‡ in storage	Temperature, Danish mean	CH ₄ production rate	Potential CH ₄ emission
	m ³	Mg	°C	Nm ³ § CH ₄ /Mg VS month	Nm ³ CH ₄
January	850	3.3	0	0.78	2.6
February	950	4.0	0	0.78	3.1
March	1050	4.7	2.1	1.01	4.7
April	1150	5.4	5.7	1.55	8.4
May	50	6.2	10.8	2.84	17.5
June	150	6.9	14.2	4.26	29.3
July	250	7.6	15.6	5.04	38.3
August	350	8.3	15.7	5.10	42.4
September	450	0.4	12.7	3.57	1.3
October	550	1.1	9.1	2.32	2.5
November	650	1.8	4.7	1.37	2.5
December	750	2.5	1.6	0.95	2.4
					155.0 m ³
					0.13 m ³ /Mg

† Dry matter content = 1.3%; volatile solid content = 60.3% of dry matter.

‡ Volatile solids.

§ Normal cubic meter (one cubic meter at 0°C and 1 atmosphere).

nificantly reduce CH₄ emissions (Petersen et al., 2005; Sommer et al., 2000).

Emptying the tanks in spring is an important factor for the magnitude of the CH₄ production. Figure 6 shows the lowest filling degree of the storage tanks during the warmest months, where the potential CH₄ production rate is the highest. Application of the digested material in the autumn (September) would increase the estimated yearly CH₄ production for the average digested material by up to 70% to 0.13 Nm³ CH₄/Mg of digested waste. These data are shown in Table 3.

The importance of the relatively low Danish monthly average temperature is shown in Table 4. The CH₄ production for a storage tank similar to that in Tables 2 and 3 is estimated assuming the highest monthly average air temperature measured in Denmark since 1874 (www.dmi.dk). The temperature difference is approximately 4°C compared to the average temperatures in Table 2 resulting in a 60% increase in the yearly CH₄ production. Assuming elevated temperatures in January, February, and December only (average temperatures for the rest of the year) increased the CH₄ production by around 10% compared to production at average temperature.

The CH₄ produced from the digested organic waste could be collected and could contribute to the gas production of the main reactor. This type of gas collection is already in place at many biogas plants, especially for storage tanks directly connected to the biogas reactor. The amount of gas collected in storage tanks may be a significant part of the gas production at the plant. Typically, the retention time in these tanks is 1 to 2 mo. Assuming 30°C in the storage tank, Eq. [1] estimates a CH₄ production of 68 Nm³ CH₄/Mg VS in the digested waste or 13.6 Nm³ CH₄/Mg VS in the waste entering the biogas reactor. This equals approximately 3% of the CH₄ potential of the waste delivered to the biogas plant (460 Nm³ CH₄/Mg VS).

If the degradation in the biogas reactor is less than the assumed 80%, due to decreased retention time, overload, or operational problems, CH₄ production in the storage tanks may be significantly higher than estimated above.

Environmental Assessment

Environmental assessment of biogas treatment of municipal organic waste, including electricity produced

Table 4. Estimated methane production in a 1200-m³ storage tank for anaerobically digested organic waste assuming a monthly average temperature corresponding to the highest measured monthly averages in Denmark since 1874 (www.dmi.dk). The tank is emptied in April, while the inflow of digested material is distributed over the year.†

Month	Volume of digested material	VS‡ in storage	Temperature, Danish mean	CH ₄ production rate	Potential CH ₄ emission
	m ³	Mg	°C	Nm ³ § CH ₄ /Mg VS month	Nm ³ CH ₄
January	850	6.2	4.9	1.41	8.7
February	950	6.9	5.5	1.51	10.4
March	1050	7.6	6.1	1.62	12.3
April	1150	8.3	8.4	2.14	17.8
May	50	0.4	13.8	4.07	1.5
June	150	1.1	18.2	6.87	7.5
July	250	1.8	19.5	8.02	14.5
August	350	2.5	20.4	8.93	22.6
September	450	3.3	16.2	5.41	17.6
October	550	4.0	12	3.28	13.1
November	650	4.7	7.7	1.96	9.2
December	750	5.4	5.1	1.44	7.8
					142.9 m ³
					0.12 m ³ /Mg

† Dry matter content = 1.3%; volatile solid content = 60.3% of dry matter.

‡ Volatile solids.

§ Normal cubic meter (one cubic meter at 0°C and 1 atmosphere).

from the gas and utilization of the effluent on agricultural fields, will often show savings in the impact category "global warming" due to substitution of energy based on fossil fuels and substitution of commercial fertilizers. Kirkeby et al. (2006) evaluated the environmental impacts from different treatment methods of the organic waste from the municipality of Århus (Denmark). Based on their data and assumptions, the savings in global warming from biogas treatment of 1 Mg of municipal organic waste could be estimated to be 261 kg CO₂ equivalents. They did not include the CH₄ emissions from storage of the digested waste. Estimated according to the proposed model, these emissions would potentially constitute around 8 kg CO₂ equivalents/Mg waste (based on the production of 10 Nm³ CH₄/Mg VS in the digested waste, 80% degradation in the biogas reactor as well as 80% VS, and 30% dry matter content in the waste entering the biogas reactor). If emitted to the atmosphere, the potential savings in global warming, resulting from the biogas treatment, will be reduced by 3%.

CONCLUSIONS

This paper presents a model for estimating the CH₄ production from stored digested municipal organic waste. The model takes into account typical patterns of managing the storage tanks and the average temperatures during the months where the digested waste is actually stored.

For a storage tank under typical Danish conditions, the model estimated the CH₄ production to be 0.4% of the CH₄ potential of the waste delivered to the biogas plant. Whether the produced CH₄ is emitted to the atmosphere depends on the physical features of the storage tank (e.g., cover and gas collection).

For storage tanks directly connected to biogas reactors the temperature was higher and the estimated potential CH₄ production corresponded to 3% of the CH₄ potential of the organic waste treated in the biogas plant. The gas produced in these tanks is typically collected and, therefore, contributes to the overall gas

production of the biogas plant. Storage in these tanks before transfer to the separate storage tanks may, therefore, decrease the content of degradable VS in the digested waste and thereby limit the methane production and potential emissions from later storage in the separate tanks on the farms.

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