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Pelletized Straw for Biogas Production—Substrate Characterization and Methane Formation Potential

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Abstract: The use of agricultural residues in biogas plants is becoming increasingly important, as they represent an efficient and sustainable substrate alternative. Pelletizing straw can have positive effects on transportation, handling, and biogas production. In this study, different grain straw pellets from mobile and stationary pelleting plants in Germany as well as the corresponding untreated straw were characterized and investigated for their suitability for anaerobic digestion (AD). Therefore, tests on the biochemical methane potential (BMP) and the chemical–physical characterization of unpelletized straw and straw pellets were carried out. The characterization of the pellets and the straw revealed a high average total solid content of 91.8% for the industrially produced straw pellets and of 90.8% for the straw. The particle size distribution within the tested pellet samples varied greatly depending on the pelleting process and the pre-treatment of the straw. In addition, a high C/N ratio of 91:1 on average was determined for the straw pellets, whereas the average higher heating value (HHV) content of the pellets was 17.58 MJ kg^{-1}. In the BMP tests, the methane production yields ranged from 260–319 normal liter (NL) CH_4 kg^{-1} volatile solids (VS) for the straw pellets and between 262 and 289 NL CH_4 kg^{-1} VS for the unpelletized straw. Overall, pelleting increases the methane yield on average from 274 to 286 NL CH_4 kg^{-1} VS, which corresponds to an increase in methane yield of 4.3%. Based on the results, the feasibility of using straw pellets for AD could be confirmed, which can facilitate the possibility of increased biogas production from agricultural residues such as straw pellets and thus make the substrate supply more sustainable.

Keywords: straw pellets; biogas; anaerobic digestion; grain straw; pelleting; substrate characterization; methane

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1. Introduction

The global trend in energy production involves a circular economy and sustainable supply of energy sources. Some advanced methods support the economic development of energy production by the utilization of waste biomass, while limiting ecological effects [1–3]. An important part of generating renewable energy with respect to sustainability is the use of agricultural waste, which can be an environmentally friendly alternative to fossil fuels [4].

Agricultural residues are considered good candidates for energy production without competing with food or induce land use changes [5–7]. Straw is one of the most abundant crop residues produced in the world, making it an attractive substrate for biogas production via anaerobic digestion (AD) [7–10]. In general, lignocellulosic biomasses such as straw contain three main organic polymers called cellulose, hemicellulose, and lignin, which are intertwined, making them recalcitrant and hindering sugar release [11]. This leads to increased resistance to biochemical degradation processes and therefore makes lignin-rich straw a rather poor substrate for biogas production [12–14].

In order to improve the biochemical degradation processes, different pretreatment processes for AD were investigated in the last decades to make the substrate more accessible to microbial activity and improve the methane production rate [8,15–17]. In addition to chemical and biological treatments, there are various physical pre-treatments that are widely used in the utilization of lignocellulosic biomass, such as straw [8,18]. The most common physical treatments are mechanical pretreatment methods to reduce particle size of the straw and to facilitate handling prior to feeding anaerobic reactors [19]. Moreover, a particle size reduction of straw is required in order to enhance biomass biodegradability, avoid problems within the digesters, like clogging, and to provide a homogeneous mixture during the digestion process [20]. Although a physical reduction of particulate size can increase bulk density compared with baled straw, ground straw remains difficult to handle within the biogas production chain [21]. Accordingly, densification by briquetting or pelleting can overcome logistical problems [22] and help optimize the biogas production process [23,24], although this introduces an additional treatment step that requires additional energy and can lead to further difficulties [16,25–28]. The greatest challenge in using straw for biogas production is to break down its resilient structure in pre-treatment processes in order to enable subsequent AD [17].

A reduction in particle size and the application of high pressure and temperature during the pelleting process could both accelerate the biodegradation of the biomass in the anaerobic digestion process and lead to higher methane yields. However, the difficulty in assessing the impact of biomass pretreatment technologies like pelleting on biomass components relevant to bioenergy production remains a significant challenge for studying the impact of biomass pretreatment and factors affecting subsequent use [4,14,29]. Therefore, a correlation between the biogas production of physically pretreated straw and the commonly used parameters for biomass characterization is rather poor [3,30] and to the best of our knowledge, the effect of pelleting as mechanical pretreatment for cereal straws in an anaerobic digestion process chain has been scarcely evaluated [8,14,31].

Thus, the aim of this study was to evaluate the potential of pelletized straw in anaerobic digestion processes. Therefore, different straw pellets from industrial scale and pilot scale pelleting plants were investigated regarding their physical and chemical characteristics and their biochemical methane potential (BMP). Moreover, the effect of the pelleting was evaluated by comparing BMP of the native, untreated straw with the subsequent straw pellets.

2. Materials and Methods

2.1. Straw Pellets Material

The investigated straw samples came from farmers in various regions of the federal state of Lower Saxony, Germany. Thirteen straw pellet samples from stationary, semi-mobile, and mobile production were examined (Figure A1, Table 1). Overall, eleven samples of wheat straw pellets and one each of rye and barley straw pellets were tested.

Table 1. Investigated straw pellets with sizes and their origin.

ID	Grain	Scale of Pelleting Plant	Pelleting Process	Pellet Length [mm]	Pellet Diameter [mm]
P1	wheat	industrial	stationary pelleting, mobile plant (sm)	32–45	15
P2	wheat	industrial	stationary pelleting, mobile plant (sm)	25–35	15
P3	wheat	industrial	stationary pelleting, mobile plant (sm)	25–30	15
P4	wheat	industrial	stationary pelleting, mobile plant (sm)	20–30	15
P5	wheat	industrial	stationary pelleting, mobile plant (sm)	25–40	15
P6	rye	industrial	stationary pelleting, mobile plant (sm)	30–65	15
P7	barley	industrial	stationary pelleting (st)	15–30	8
P8	wheat	industrial	stationary pelleting (st)	8–52	8
P9	wheat	industrial	mobile plant, pelleting at field (m)	8–35	8
P10	wheat	industrial	mobile plant, pelleting at field (m)	15–40	8
P11	wheat	industrial	mobile plant, pelleting at field (m)	30–50	15
Ppp1	wheat	pilot plant	stationary pelleting (st)	5–20	8
Ppp2	wheat	pilot plant	stationary pelleting (st)	5–20	8

Two pellet samples were produced in an industrial scale stationary pressing process with an 8 mm diameter die at commercial pelleting plants in Germany (P7 and P8). Before pelleting, the straw was pressed into bales, stored and pre-shredded with a hammer mill, and dedusted. In contrast, mobile pelleting was carried out directly in the field without any straw pre-treatment using various mobile pelleting systems (Premos 5000 by Maschinenfabrik Bernard KRONE GmbH & Co.KG, Spelle, Germany, and METRITRON560 by Metritron GmbH, Pfronstetten, Germany) (Figure 1b). In total, three different straw pellet samples from the direct mobile production were examined (P9–P11). Six pellet samples were produced from square bales using the mobile pelleting systems as stationary pelleting plants, which were supplied with compressed and stored bales of straw (P1–P6) (Figure 1a).


(a) (b)

Figure 1. Industrial-scale stationary pelleting (**a**) and pelleting at field (**b**) with a mobile pelleting plant (Photos courtesy of Sören Mohrmann).

In addition to the pellets from industrial-scale pelleting plants, for comparison, two different pellets (Ppp1 and Ppp2) were produced from a bale-pressed wheat straw sample in the HAWK lab by a small-scale pellet mill (PM 200, Cissonius GmbH, Zehdenick, Germany) after crushing the straw with a hammer mill through a 6 mm sieve (CF-158, Cissonius GmbH, Zehdenick, Germany) (Figures 2 and A2).

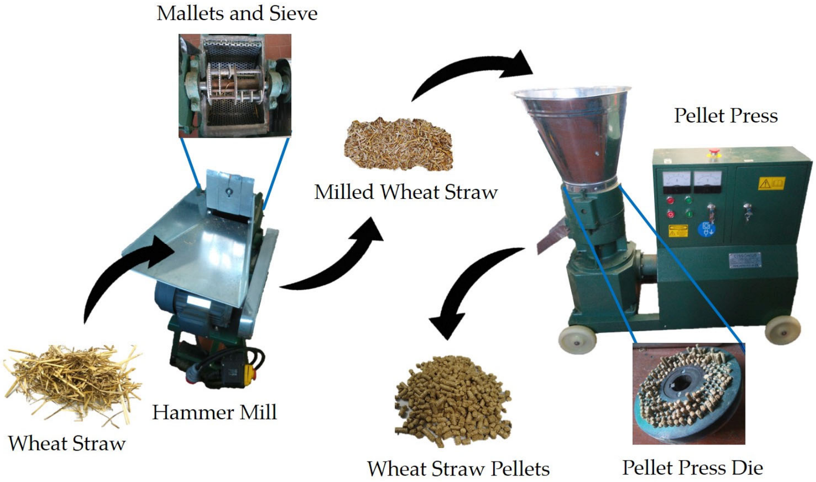

Figure 2. Workflow for grinding and pelleting straw in the pilot-scale trials for Ppp1 and Ppp2.

In total, seven samples with a diameter of 15 mm and six samples with a diameter of 8 mm were analyzed. The lengths of the individual pellets varied between 8 mm and 65 mm (Table 1) and had different colorations ranging from light beige to green–brown.

For eight pellet samples, the straw from which the pellets were produced was also analyzed. These were six wheat straw samples (S4, S5, S9, S10, S11, Spp) and one barley straw (S7) sample (Figure A3).

2.2. Analytical Methods

The pellets shape was described by determining the diameter and the minimum and maximum length of the pellets in a representative sample using a measuring device based on ISO 17829:2015 [32]. The appearance and color were described by means of a visual inspection. Various other analyses were carried out to determine whether pelleted straw is suitable as a substrate for AD. Parameters such as the total solids content, volatile solids and the carbon/nitrogen ratio are important for optimal anaerobic digestion and were therefore investigated accordingly. In addition, further properties of the straw pellets, such as bulk density and heating value, were examined.

The determination of total solids (TS) and volatile solids (VS) provides information about the water and ash content in the straw and straw pellet samples and thus about the storage and transport capacity as well as the quantity of organic matter. The TS of a sample was determined according to the DIN EN 12880:2001-02 [33] standard by drying at a temperature of 105 °C and weighing the difference in weight between the fresh and dried samples. The TS contains an organic component, which can be degraded to a certain extent in the biogas plant depending on its composition, and an inorganic component consisting of nutrients, sand, or stones. Easily degradable substrates with high VS contents are therefore preferred for treatment in biogas plants. Accordingly, the VS contents of the samples were determined by annealing at 550 °C in the presence of air according to DIN EN ISO 12879:2001-02 [34] in duplicate.

The influence of the particle sizes of the feedstock on biogas formation has already been investigated by various researchers and, in general, the size of the particles has a significant influence on the methane formation kinetics and the amount of methane produced [35–39]. The size distribution of the straw particles incorporated in the investigated pellets was determined by mechanical sieving (KS1000, Retsch GmbH, Haan, Germany) according to DIN EN ISO 17830:2016-11 [40]. In this process, the fractions were divided into the following categories using six different sieving trays: Fine fraction up to 2 mm, medium fraction from 2 mm to 5.6 mm, and coarse fraction > 5.6 mm.

The proportion of carbon (C) and nitrogen (N) as well as their ratio are also decisive for microbial degradation and the formation of methane. The amounts of C, N, and hydrogen (H) contained in the analyzed samples were evaluated through elemental analysis with a Vario MACRO cube (Elementar Analysensysteme GmbH, Langenselbold, Germany) according to DIN EN ISO 16948:2015 [41]. The C/N ratio was then calculated accordingly for each sample, as it plays a key role in the optimization of the AD process [42,43].

Bulk density of the straw pellets is an important storage and transportation parameter and was determined in accordance with DIN 51705:2001 [44]. The empty weight of a container was determined and then the container was filled with straw pellets. The excess bulk material was stripped off with a square bar and the container was weighed again with the leveled filling. The bulk density was calculated in triplicate using the mass of the container with specimen minus the empty weight of the container in relation to the container volume.

To determine the behavior of the straw pellets in the digester, the dissolution potential of the pellets in water as well as the formation of sediment and floating layers were investigated using a test developed by HAWK for this purpose. A glass cylinder was filled with 2200 mL of water (20 °C) and then 100 g of straw pellets were added. The dissolution and distribution of the straw pellets in the water were documented after 0, 60, 120, 180, and 240 min by photo.

The characterization of the straw biomass as feedstock for the straw pellets was performed using the standard methods for proximate, ultimate, and compositional analyses described above for the straw pellets. As a pre-treatment step for the investigations, the field-dried straw was ground using an ultra-centrifugal mill (ZM 300, Retsch GmbH, Haan, Germany).

2.3. Biochemical Methane Potential (BMP) of the Straw and Straw Pellets in Batch Tests

In order to assess the general suitability for anaerobic degradation of straw pellets, the BMPs of the samples were examined according to the standard VDI 4630 [45] with three replicates for each sample. For the investigations, 30 L PET barrels were used as reactors, with gasbags as storage for the produced biogas [46]. The biogas yield test was carried out in a climate chamber under mesophilic conditions at 37 °C until the termination criterion was reached (daily amount of gas formed <0.5% of the amount of gas formed up to that point, or 45 days if the termination criterion is reached earlier). For the preparation of the batch tests, the barrels were filled with an inoculum and the corresponding amount of straw pellets or straw. For handling reasons, during the BMP tests, the raw straw was shortened to a length of approx. 15 cm using household scissors.

Immediately after preparation, the headspace of each barrel was flushed with nitrogen to create anaerobic conditions. The liquid fraction of a freshly separated digestate from an agricultural biogas plant was used as inoculum. To assess the influence and general suitability of the inoculum used, the BMPs of the inoculum and the reference (microcrystalline cellulose) were also determined in triplicate. The ratio of VS from the inoculum and the amount of VS from the sample was set at 2:1. The resulting biogas was then collected in gasbags and the biogas amount was measured with a drum gas meter (Ritter Apparatebau GmbH Co KG, Bochum, Germany). To determine the biogas quality, the parameters methane (CH_4), carbon dioxide (CO_2), hydrogen (H_2), oxygen (O_2) and hydrogen sulphide (H_2S) were recorded with a gas meter with infrared sensors (visit 03, Messtechnik Eheim GmbH, Schwaigern, Germany). To convert the biogas to standard conditions (1013 mbar, 273 K), the temperature of the biogas and the air pressure were recorded for each measurement, and the biogas and methane yields were calculated using the measurement data converted to standard conditions by subtracting the biogas quantities from the inoculation substrate and taking the headspace volume into account. A total of thirteen different straw pellets and seven straw samples were tested in triplicate for their BMP.

3. Results

3.1. Straw Pellet Characterization

In order to assess the suitability of the straw pellets for AD, various physical and chemical parameters of the thirteen pellet samples were analyzed.

3.1.1. TS and VS

The TS content of the pellets tested was between 88.4% and 96.3% (except for one pellet produced in the lab with the addition of water, which led to a value of only 80.8%), whereas the TS content of the six straw samples was between 89.7% and 91.9%. Thus, the variation of TS content was higher for the pellets than for the straw (Figure 3). Overall, the average TS of the straw was 90.8% and that of the VS was 93.8% TS, while the industrially produced straw pellets had an average TS of 91.8% and a VS content of 93.8% TS (Table 2). Moreover, the TS content of the pellets (with the exception of P11) was around 1.1% to 2.8% higher than that of the corresponding straw sample in six of the seven samples analyzed.

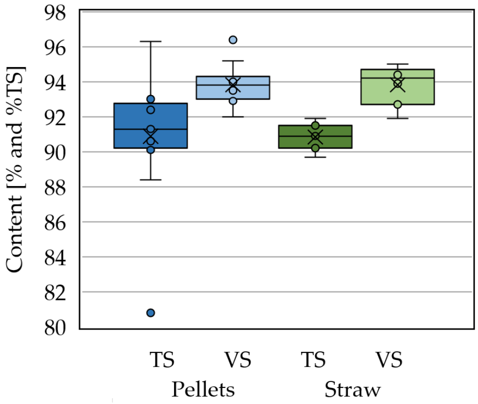

Figure 3. Results of the proximate analysis of the straw and the corresponding straw pellets.

Table 2. Characterization of the straw pellets analyzed.

ID	C	N	C/N	TS	VS	HHV	Bulk Density
	[%TS]	[%TS]	[-]	[%]	[%TS]	[MJ kg^{-1}]	[kg m^{-3}]
P1	43.4	0.44	98.5	90.3	92.0 ± 0.1	17.0	487 ± 30
P2	45.8	0.47	97.5	90.1	93.2 ± 0.5	17.7	595 ± 10
P3	45.1	0.38	118.7	96.3	94.4 ± 0.6	17.3	534 ± 1
P4	45.0	0.61	73.8	91.3	93.5 ± 0.0	16.7	562 ± 0
P5	44.0	0.56	78.5	92.4	92.9 ± 0.1	17.1	568 ± 17
P6	46.6	0.52	89.7	93.0	96.4 ± 0.1	17.9	546 ± 1
P7	45.4	0.31	146.5	91.3	94.7 ± 0.1	17.1	631 ± 6
P8	45.5	0.60	76.3	91.3	94.0 ± 0.0	17.5	645 ± 1
P9	45.3	0.65	69.6	92.5	94.2 ± 0.1	17.3	520 ± 3
P10	45.6	0.61	74.8	93.2	93.8 ± 0.0	17.4	500 ± 1
P11	44.8	0.42	106.7	88.4	93.1 ± 0.1	17.9	534 ± 1
Ppp1	45.9	0.50	91.7	80.8	92.9 ± 0.1	18.4	406 ± 2
Ppp2	47.0	0.72	65.8	90.6	95.2 ± 0.0	18.8	592 ± 1

3.1.2. C/N Ratio and Heating Value of the Pellets

The elemental analysis of the wheat straw pellets resulted in an average content of 45.32% C and 0.52% N on a dry weight basis. Accordingly, the nitrogen content in straw pellets is very low, with the lowest amount in barley pellets (P7) and the highest amount in the pilot plant wheat pellets (Ppp2). The average C/N ratio determined for the straw pellets examined was 91:1, ranging from 65.8:1 (Ppp2) to 145.5:1 (P7) (Table 2). For the higher heating value (HHV) of the pellets analyzed, an average energy content of 17.58 MJ kg^{-1} was determined, ranging from 16.72 to 18.80 MJ kg^{-1}. Interestingly, the self-produced pellets (Ppp1 and Ppp2) had a slightly higher calorific value than the industrially manufactured ones.

3.1.3. Bulk Density of the Pellets

The bulk density (BD) of the pellets, and therefore the transport and storage properties of the pellets, were largely dependent on the size of the presses used and the diameter of the dies selected. Overall, an average bulk density of 548 kg m^{-3} was assessed for the pellets, while the variation in BD was high, ranging from 406 kg m^{-3} (Ppp1) to 645 kg m^{-3} (P8) (Table 2).

3.1.4. Particle Size Distribution of the Pellets

The distribution of particle sizes within the pellets provides a distinct pattern: Pellets P1 to P5 and P11 were produced with a partly mobile or mobile pelletizer without prior preparation of the straw, which means that the proportions of the coarse and medium fractions dominate the particle size distribution (Figure 4). In particular, the fraction larger than 5.6 mm is notable. In comparison, the pellets P7 and P8 and Ppp1 and Ppp2, which were produced from chopped straw with a stationary pelletizer, are dominated by the fine fraction smaller than 2 mm. Accordingly, the particle size distribution of the tested pellets samples varied greatly depending on the pressing process and the pre-treatment of the straw.

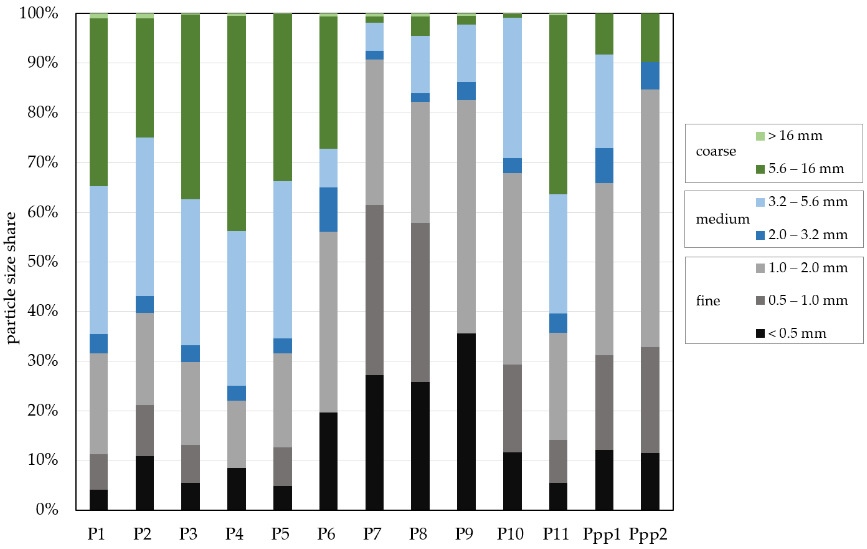

Figure 4. Particle size distribution of the straw pellets investigated.

3.1.5. Dissolution Potential of the Pellets

One advantage of pellets over straw is their better dissolution behavior in the digester and, thus, better process control. Without stirring, the substrate in the digester is usually stratified into three layers: the upper layer (floating layer), the middle layer (fermentation solution), and the bottom layer (sediment layer) [47]. All pellets tested dissolved immediately after addition ("start"). For most of the samples, the pellets were completely dissolved after 60 min, as shown in Figure 5b,c. In the case of P5 and P6, a slight floating layer formed after 120 min and persisted until the end of the experiment. With the pellets P2 and P7–P10 a clear floating and sediment layer formed after 60 min (e.g., P9 in Figure 5a). Pellets P9 and P10 showed a dark coloration of the water directly after the pellets were added. With P2 there was only a slight coloration, and with P7 and P8, the water remained clear.

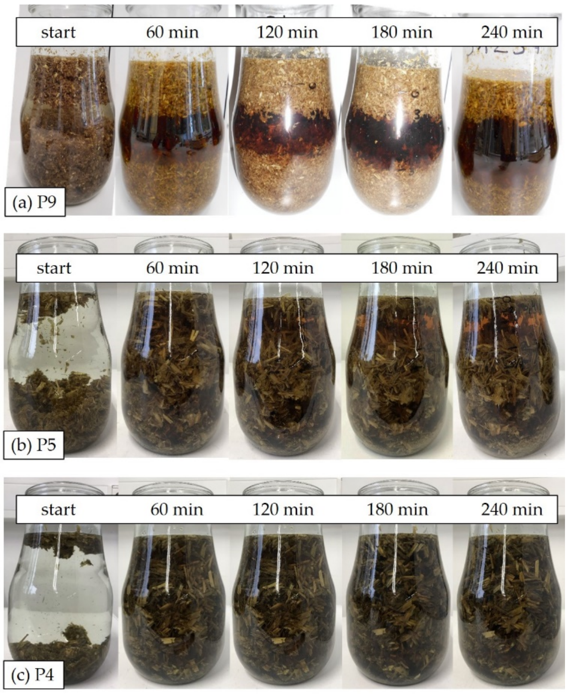

Figure 5. Examples of the dissolution tests of the pellets in water with (**a**) P9 forming floating and sediment layers directly after start, (**b**) P5 forming floating and sedimentation layer after 180 min. and (**c**) P4 showing a homogeneous distribution during the entire investigation.

3.2. Biochemical Methane Potential Tests

This study investigates the potential of grain straw pellets for energy generation through AD. Therefore, BMP tests were conducted for thirteen different straw pellets and seven associated, unpelletized straw samples (Section 2.1). The kinetics of the methane yield curves show a differentiated pattern. For some pellets, a clear difference between straw and straw pellets could be observed (Figure 6a). The straw pellets P4, P5, and P11, showed significantly faster decomposition and higher methane yields than the corresponding straw. In contrast, no difference was found between straw and pellets for P7, P9, and P10 (Figure 6b). In the case of pellets P1–P3, P6, and P8, slightly different kinetics were observed (Figure 6c). In the first 20 days of the test, P1–P3 and P8 showed slightly better degradation than the rye straw pellets P6. However, the BMP after 45 days showed slightly lower values for P6 (265.7 NL CH_4 kg^{-1} VS) and P8 (260.3 NL CH_4 kg^{-1} VS) than for P1–P3 (281.1-286.5 NL CH_4 kg^{-1} VS). Although the pellets from the pilot plant had a similar course as the other pellets and resulted in a BMP of 300.4 NL CH_4 kg^{-1} VS (Ppp1) and 281.6 NL CH_4 kg^{-1} VS (Ppp2), the associated straw (Spp) showed a significantly delayed degradation. However, at the end of the test period, the BMP of the straw approaches that of the pellets (276.5 NL CH_4 kg^{-1} VS).

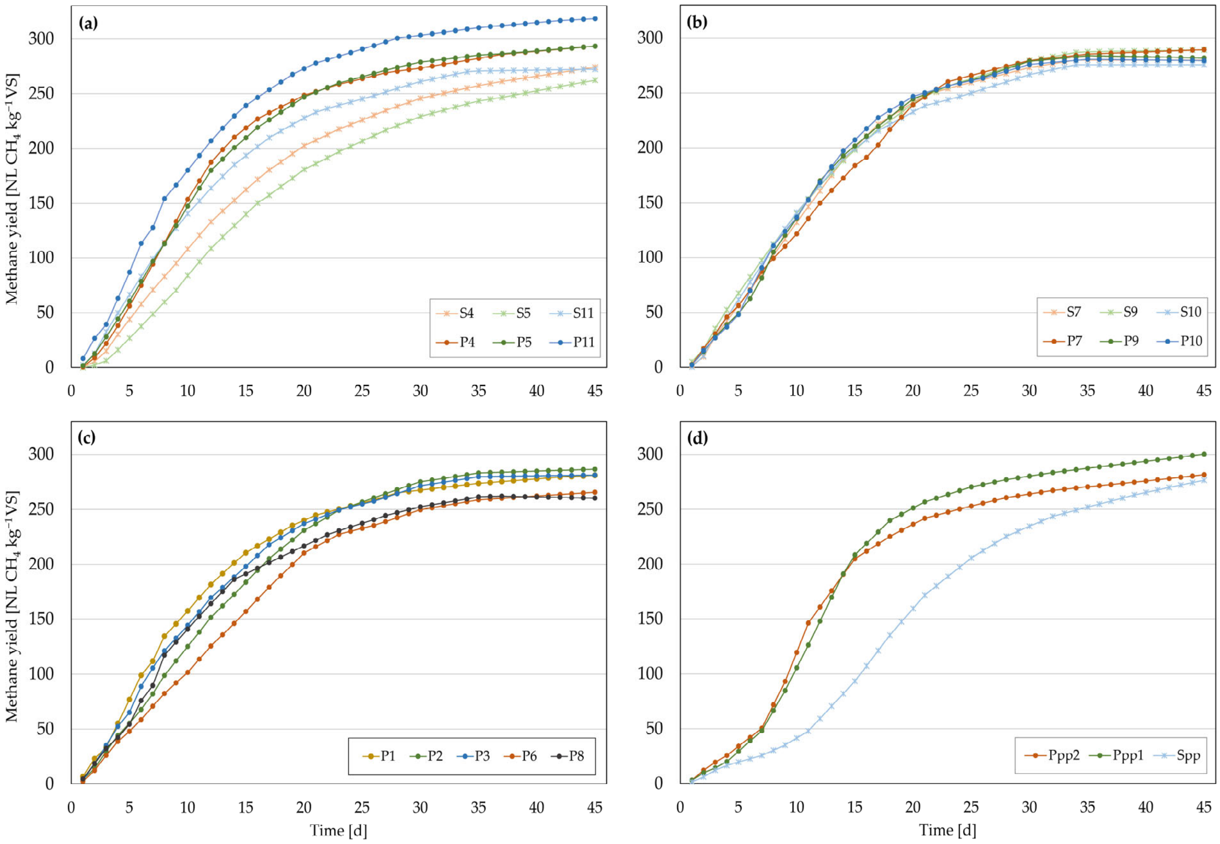

Figure 6. Methane formation graphs obtained in the BMP tests for: (**a**) the pellets P4, P5, P11, and their respective straw; (**b**) the pellets P7, P9, and P10 and their respective straw; (**c**) the pellets P1, P2, P3, P6, and P8; and (**d**) the pilot-scale pellets Ppp1 and Ppp2 and the straw Spp.

Overall, pelleting increases the methane yield, on average, from 274 to 286 NL CH_4 kg^{-1} VS, which corresponds to an increase in methane yield of 4.3% (Table 3). The methane potential ranges from 260 to 319 NL CH_4 kg^{-1} VS for the pellets and 262–289 NL CH_4 kg^{-1} VS for the unpelletized straw. The average yields related to the fresh mass (FM) were 243 NL CH_4 kg^{-1} FM (pellets) and 237 NL CH_4 kg^{-1} FM (straw). The corresponding increase due to pelleting in relation to the fresh mass of the substrate results in 3.7%. Moreover, a methane yield of around 51% was revealed for both the straw and the respective straw pellets.

Table 3. BMP of the straw and straw pellets investigated.

Substrate	Average BMP	Min.	Max.	Average BMP	Min.	Max.	Average Methane Content
		[NL CH_4 kg^{-1} VS]			[NL CH_4 kg^{-1} FM]		[%]
Straw pellets	286	260	319	243	223	262	51.2
Straw	274	262	289	236	222	255	51.4

4. Discussion

4.1. Substrate Characterization

The dry matter content of the pellets was generally higher than that of the raw straw, which can be attributed to the high temperatures during pelleting and the associated evaporation of part of the water in the straw [48]. However, the pellets produced at pilot scale (Ppp1 and Ppp2) have a lower TS value than the feedstock straw Spp from which the pellets were produced, which is due to the addition of demineralized water during pelleting. This was necessary to achieve a TS content in the input material of around 12–20% to enable the pelleting process [49–52].

The comparison of the average TS and VS contents showed that although water is lost during the pelleting, the organic content in the dry matter does not show any changes between the straw and the pellets, making pelleting a suitable pre-treatment process for AD.

The wide C/N ratio of the pellets was mainly due to their very low N content, where even a small change had a large effect on the respective ratio (Table 2). Barmina et al. (2016) demonstrated that elements such as C and N in wheat straw pellets were 47.4 and 0.74%, respectively, corresponding to a C/N ratio of 64:1, while the HHV was 17.54 MJ kg^{-1} [53]. These findings underline the results presented in this study, although the high variety is dependent on the type of grain, the time of harvest, and the growing conditions in general [54,55].

Nevertheless, all C/N ratios determined were relatively high and far above the ideal conditions for optimal fermentation, which is around 15:1 to 35:1 [56–59].

The high C/N ratio has a correspondingly negative effect on methane production, meaning that mono-digestion of substrates such as straw pellets is not an efficient way to produce biogas [60]. Therefore, co-digestion with nitrogen-rich substrates like manure could offer an advantage in the fermentation process [61–63], broaden the substrate range [64,65], contribute to emissions reductions [66], and reduce the use of cultivated biomass for energy generation [67,68].

The calorific values determined match those from the literature well. Guo et al. (2022) determined an HHV of 16.1 MJ kg^{-1} for wheat straw pellets [69] and Huang et al. (2009) examined an HHV of 16.2–18.0 MJ kg^{-1} for wheat straw [70]. Overall, straw pellets are therefore in the middle of the range of densified, agricultural residual biomasses [71].

The average bulk density of the straw pellets revealed in this study was 548 kg m^{-3}. Zhang et al. (2012) examined an average bulk density of milled wheat straws in the range of 97.5–177.2 kg m^{-3} [72], which corresponds to a volume reduction through pelleting of 68–82%. Assuming that uncompacted, unmilled straw is used, with a bulk density ranging from 24 to 111 kg m^{-3} [73], pelleting can achieve a volume reduction of 80–96%, representing an up to 23-fold compaction. As a result, the transportation costs, which dominate the total costs for the provision of substrate from agricultural residues [70], can be reduced accordingly. However, it has been shown that the bulk density of the pellets generally varies greatly and considerable differences can be observed depending on the pelleting process used, the type of grain, the pre-treatment, and the origin of the straw.

For the dissolution potential of the pellets, different behaviors could be observed. A homogeneous dissolution and only a slight floating layer formation for pellets with 15 mm and stationary pelleting (Figure 5b,c) indicate good mixing and thus a high biogas production. For pellets produced from pre-milled straw and therefore containing a higher share of fines (Figure 4), a clear formation of floating and sediment layers could be observed, which leads to an increased stirring effort in the digester.

4.2. BMP of Straw Pellets and Straw

In this study, the pelleting increased the methane yield of straw on average from 274 to 286 NL CH_4 kg^{-1} VS (Table 3). The variation in the test for the different samples was high, with values ranging from 260–319 NL CH_4 kg^{-1} VS for the pellets and 262–289 NL CH_4 kg^{-1} VS for the unpelletized straw.

Scherer et al. (2021) determined an average specific methane yield of 287.1 NL CH_4 kg VS^{-1} for milled wheat straw [74], which is slightly higher than the value determined in these experiments for unmilled straw and even higher than the value for pellets. However, the individual values of the different samples also fluctuated in a similar order of magnitude. A similar value (279 NL CH_4 kg VS^{-1}) was also determined by Hafner et al. (2020) for milled straw in a round robin test in 29 laboratories and with 47 BMP tests evaluated [75]. Slightly higher values (281-291 NL CH_4 kg VS^{-1}) were determined by Dumas et al. (2015) for their experiments with micronized straw smaller than 0.7 mm [39].

To date, there is very little literature comparing the values for pelleted straw. Sing et al. (2024) have investigated the influence of co-pelleting straw with cow manure on the methane potential [31]. Mönch-Tegeder et al. (2013) reported a higher final methane yield from pelletized straw (247 NL CH_4 kg^{-1} VS) than non-pelletized straw [76], which corresponds with the observed results in this study for P4, P5, P11, Ppp1, and Ppp2 (Figure 6). However, the average value reported is below the range observed in this study, and Victorin et al. (2020) found an even lower BMP of 239 NL CH_4 kg^{-1} VS for pelleted wheat straw [77].

The measured methane content of the biogas in the BMP trials of 51.2–51.4 % (Table 3) can also be found in literature [78], which underlines the validity of the results.

Considering the different production methods of the pellets (Table 1), it is not possible to derive a trend in terms of BMP (Figure 6), either for the scale of the pelleting plant (pilot scale and industrial scale), the pelleting process itself (stationary or mobile pelleting in the field), or for the pellet diameter (8 or 15 mm). In addition, the type of grain had no significant influence on the BMP, as both the rye pellets (P6) and the barley pellets (P7) with 265.7 and 289.4 NL CH_4 kg VS^{-1}, respectively, were in the range of the wheat pellets, although barley has a slightly higher theoretical methane potential than wheat [17].

4.3. Unpelletized Straw and Straw Pellets

In the study by Victorin et al. (2020), four mechanical methods for preparing wheat straw for use as a biogas substrate were investigated [77]. It was shown that pelleting leads to faster biogas formation than untreated straw in batch tests. However, a clear statement for the comparison of the biochemical degradation rate of straw and straw pellets could not be derived from the obtained results, since the boundary conditions for the digestion of straw play a crucial role.

Fernandez et al. (2022) performed lab-scale investigations of different substrates and found that shredding reduced the size of coarse particles, smoothly increased solubilization but did not much affect the methane yield in AD trials [79]. The main effect of mechanical pre-treatment, they observed, was the improvement in methane production rate rather than methane yield.

With regard to the particle size distribution of the tested pellets in this work, the pilot plant pellets show the influence of the particle size distribution on the methane formation kinetics (Figure 6d). It can be deduced that a higher proportion of fines in the pellets (Figure 4) increases the degradability, and thus the methane formation is higher at the beginning of the gas yield tests. Interestingly, the pellets P4, P5, and P11 (Figure 6a) showed increased BMPs compared to the corresponding straw materials without having a particularly high proportion of fines. In contrast, the kinetics of methane formation and the amount of methane of P7, P9, and P10 and their straws were quite similar (Figure 6b), although the proportion of fines in the pellets was relatively high in P7 and P9.

Other authors like Heller et al. (2023) [80], Dai et al. (2019) [81], Gallegos et al. (2017) [82] and Raud et al. (2020) [83] revealed a significant increase in the specific methane yields with different mechanical pretreatments. Interestingly, there appears to be an optimum particle size at which further crushing has little effect on the anaerobic degradability and thus on the BMP of the straw [38,39].

The trials conducted in this work indicate that the expected methane yields could be slightly increased by the obtained pressure during the pelleting process for some samples. This is clearly visualized in the BMP tests of pellets P4, P5, and P11 (Figure 6a) and the pilot scale pellets Ppp1 and Ppp2 (Figure 6d). On the other hand, the BMPs of P7, P9, and P10 revealed no difference in the BMP of the pellets compared to the untreated straw. This can be attributed to the fact that, despite the standardization of the test results according to the VDI 4630 guideline, considerable deviations in the resulting methane potentials can occur [17,74,77,78]. The reasons for these deviations can be manifold (e.g., varying straw compositions, thermal straw degradation during milling and pelleting, supply of trace elements, composition of the microbiome). Therefore, a clear correlation between biogas and methane production and the various physical and chemical properties of the biomass and the influence of various individual treatments is difficult to assess [30].

In addition to mechanical pre-treatments such as pelleting, there are also several studies on other pre-treatment methods and their combination, which increase the methane yield of straw and simplify the general use of straw as a biogas substrate. For example, grinding and ensiling the straw can also lead to an increased methane yield and facilitate the handling of the straw [25,82,84,85]. Other approaches pursue thermal pre-treatment, such as steam-exploding or hydrothermal processes, or chemical and or biological treatment as a practicable approach [8,14,38,86–89]. Therefore, a combination of the most suitable methods may further improve the energetic utilization of straw by AD.

5. Conclusions

The results indicate that the use of straw pellets for AD can achieve a positive result compared to uncompacted straw. The high bulk densities of straw pellets have an advantageous effect on transportation, storage, and feeding into the digester. In addition, dissolution tests were carried out to investigate the behavior of the pellets in the digester, which showed that pelleting has clear advantages in mixing the fermentation liquid. Straw pellets and straw in general have higher C/N ratios, making them suitable for co-digestion with nitrogen-rich substrates like manure. Accordingly, the further use of straw pellets can be facilitated by measures to raise awareness among farmers, increase the efficiency of work processes, and promote market demand through policy frameworks [90].

Finally, this study shows the usefulness of pelletized straw as an alternative substrate for biogas production due to the slight increase in degradation rates and the acceleration of anaerobic degradation, which was demonstrated for at least some of the pellet samples tested. Nevertheless, for the analysis of the best use of a biomass in a sustainable bioeconomy system, multiple stakeholders, applications, policies, and sustainability goals should be considered [91].

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Appendix A

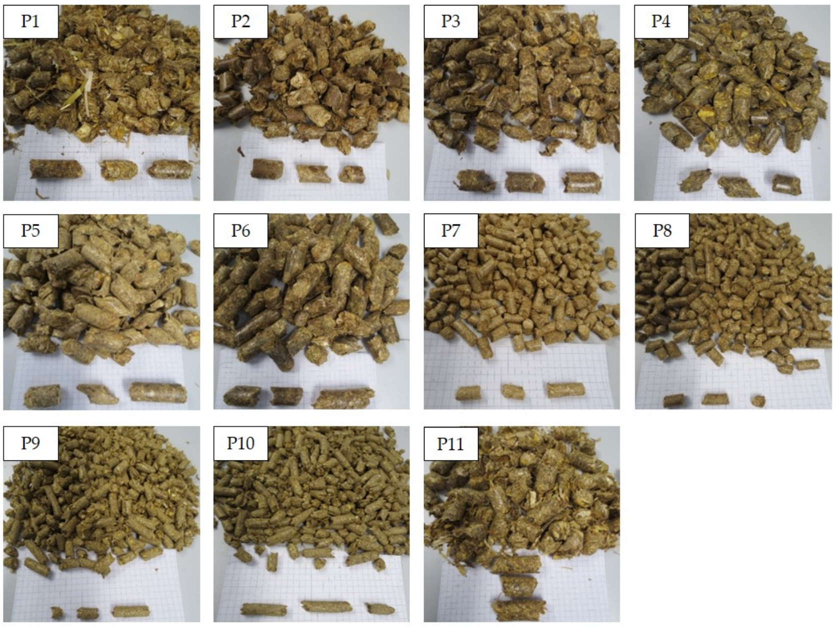

Figure A1. Appearance of the investigated straw pellets P1–P11.

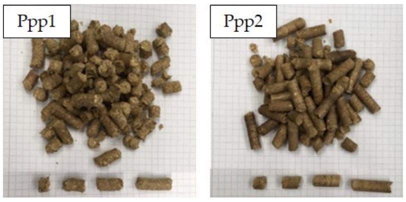

Figure A2. Appearance of the investigated straw pellets Ppp1–Ppp2.

Figure A3. Appearance of the investigated straw S4, S5, S7, S9, S10, S11, and Spp.

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