

Article

# The Biogas Potential of Oxytree Leaves

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**Abstract:** This article describes the characteristics of the Oxytree (*Paulownia*) plant, both in terms of its impact on GHG emissions and its potential use to produce biofuel, i.e., biogas. The described research involved the physico-chemical and elemental analysis of the Oxytree leaf composition and its biogas efficiency depending on the harvesting method. Three different scenarios were considered: the freshest possible leaves—processed immediately after stripping from the living tree; after the first day of collection from pruned or harvested wood; after the first week of collection from pruned or harvested wood. The best results were achieved for the harvest of the freshest leaves—on average 430 m<sup>3</sup>/Mg (biogas) and 223 m<sup>3</sup>/Mg (methane) per dry organic mass. The highest yield of biogas in terms of fresh mass (FM) was obtained for leaves fallen and collected after 1 day—123 m<sup>3</sup>/Mg FM, and 59 m<sup>3</sup>/Mg FM (methane). Processing Oxytree leaves through anaerobic digestion will contribute to reducing the carbon footprint of wood biomass production and is an additional source of renewable energy and fertilizer product.

**Keywords:** Oxytree; *Paulownia*; leaves; biogas; methane; fertilizer; biogas plant; fermentation; digestate; GHG



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## 1. Introduction

“Global energy consumption continues to grow, but it does seem to be slowing—averaging around 1% to 2% per year” [1]. Generally, since the first half of the 20th century, global energy consumption has increased almost every year, except in special cases such as the beginning of the 1980s and just after the world financial crisis (2009) (Figure 1). The explanation for the previous quote is the continued compensation of energy efficiency to the significant demand, especially from developing countries [2,3]. However, the tech mechanism may not be enough. Additionally, the burden on the environment is not reduced. Other sources present the above thesis as follows—each year, the total energy consumption increases by 2%, while energy production decreases by 1.1% [3]. This outcome affirms that energy efficiency has not kept pace with consumption. In addition, it indicates that CO<sub>2</sub>-equivalent emissions are increasing in proportion to the demand for primary energy, especially due to the rapid increase in coal consumption in China and other Asian countries (mechanisms to reduce environmental impacts are increasingly introduced there, but the balance is still negative) [3].

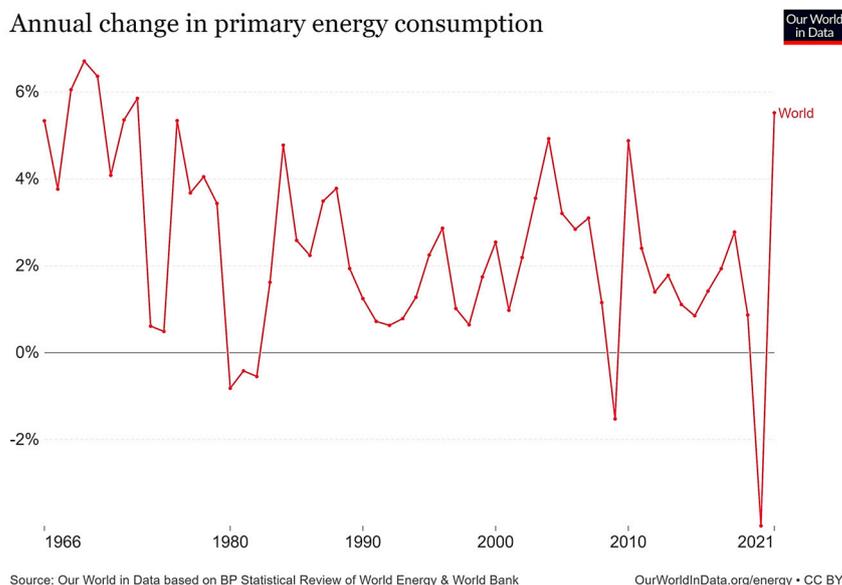
In order to mitigate global warming and counter the current energy crisis in 2022 (mainly as a result of the war in Ukraine), further, more environmentally friendly and economically justified searches for energy sources are necessary.

One of the most important aspects here is reducing the use of fossil fuels and balancing this stream through the use of renewable sources [4–7].

Biomass, the resources of which are very large and still not used to a large extent, can play an important role [8–17].

Plantings with Oxytree could be a representative of a renewable energy source (such as bioenergy biomass) that further improves air quality and at the same time reduces the effect of global warming [18]. Oxytree has the capacity to absorb from 10 to even 30 kg of CO<sub>2</sub> and exhales 6 kg of O<sub>2</sub> per year, which equates to thousands of cubic meters of purified

air per year for only one plant [19,20]. Such efficiency is due to the high carbon binding factor and significant increases in biomass, which result in the absorption of approximately 1250 Mg CO<sub>2</sub>/ha/year [19].



**Figure 1.** Global primary energy consumption change (the change is expressed as a percentage of the previous year's consumption) [1].

*Paulownia* sp. (Oxytree) is a family of fast-growing woody plants that have a very good ability to adapt to diverse climatic conditions [21]. *Paulownia* is one of the fastest-growing species in the world; this plant contains little nitrogen (N) and sulfur (S), but it has a relatively good calorific value (mass for generated energy) with low ash content [18].

Depending on the taxonomic division, up to 17 *Paulownia* species are distinguished; the most important are *Paulownia albiphloea*, *australis*, *catalpifolia*, *elongata*, *fargesii*, *fortunei*, *kawakamii*, and *tomentosa* [18,21,22]. *Paulownia* has many common names such as Oxytree, Princess tree, Empress Tree, Royal tree, Kiri tree, Phoenix tree, Anna Pavlovna Romanova, Grand Duchess of Russia, and later, Queen of the Netherlands, etc. [18,23,24].

*Paulownia* wood is very light, with a density of 0.22–0.35 g/cm<sup>3</sup> [18,23,25,26]. It has a deep and well-developed root system with a radius of almost three times larger than the crown [23]. Usually, the roots reach approximately 2 m, but cases with larger ranges are known—9 m [18,27]. Oxytree leaves usually measure 15–30 cm and 10–20 cm (length and width, respectively), which is approximately 0.9 m in diameter on average [19,21,23]. Such large leaves enable very efficient assimilation of solar energy, hold a high level of organic mass, store significant amounts of CO<sub>2</sub> and also release a large amount of oxygen [18,19,28,29]. Oxytrees have the ability to carry out photosynthesis with the use of C<sub>4</sub> (mainly) and C<sub>3</sub> cycle enzymes [23,29,30]. It is mainly thanks to these processes that the plants can quickly increase their mass. Oxytrees, although highly adaptive, need a lot of water and light to grow quickly [18,19,23,31]. As the nitrogen content (and thus protein) in the leaves of Oxytrees is similar to that of legumes, and as they contain a large amount of fats and sugars, as well as phosphorus and potassium, they are used as so-called “green fertilizer” or as animal feed. Of interest, research indicates that adding Oxytree leaves to feed can reduce methane emissions from cattle. This and other methods are currently being actively analyzed in the international project “Mitigating emissions from livestock systems” (MELS), which gathers knowledge on greenhouse gas mitigation measures in livestock production that are currently available or technically well-established [19,21,29,32–34].

Oxytree plantings are mainly determined by how they are used. If it were to serve as a source of good-quality wood biomass, there should be fewer than 600 trees/ha; for energy purposes, the planting density may be even several times higher at 2000–3500 trees/ha [18,35,36].

As the production of biomass from Oxytree ranges from 0.4 m<sup>3</sup>/piece to even 1 m<sup>3</sup>/piece, on this basis (with the aforementioned density of 0.22–0.35 g/cm<sup>3</sup>), it can be estimated that Oxytree productivity is from several dozen to even over 330 Mg/ha. This type of planting enables the production of from 30 kg to even 100 kg of dry leaves [18,21,22,37].

Apart from its employment as wood for general construction purposes, Oxytree is used to produce medicinal substances [21,38–40], feed substances [21,23,41–43], and fertilizers [19,21,29,32,33], and for the phytoextraction of toxic elements from post-industrial waste [30,44]. Recently, there have been more and more attempts to use Oxytree industrially through technologically advanced processes to obtain biofuels. Examples of such processing include the pyrolysis of wood and the acquisition of pyrolysis gases [45–47], low-temperature thermal conversion (torrefaction) that results in carbonized solid fuel [4], and alcoholic fermentation for the production of bioethanol [48,49] or biohydrogen [50–52].

Co-fermentation processes using various fallen leaves have already been described in several publications. In one such process, Chinese scientists used fallen leaves and sewage sludge at a ratio of 80/20 (volatile solids—VS) [53]. From this mixture of substrates, they produce (in the so-called dark fermentation) over 65% more biohydrogen compared to monofermented leaves and about 18% more compared to leaves alone. This means that sewage sludge in small quantities could be a good supplement, the synergistic effects of which increase biogas production. Accordingly, the increase in efficiency after adding 20% of sludge (incl. VS) is due to the enhanced C/N ratio, the provision of more bioavailable carbohydrates, the dilution of inhibitors, and the diversity and enrichment of the microbial mix in the fermenter, mainly due to such microorganisms as *Bacillus* spp., *Rummelibacillus*, and *Clostridium* spp. [53–57]. More about the characteristics of sewage sludge and its various uses in biological processes can be found in many sources [58–61].

The purpose of this study was to test the biogas potential of leaves from Oxytree plants. Often, leaves are unnecessary waste in Oxytree vegetation production and during its cutting. Such leaves are a fairly large source of biomass that can be used for energy purposes—for the production of biogas. Moreover, as digestate, it can fulfill the fertilizing function. The article provides data on the impact of the harvesting method on biogas efficiency.

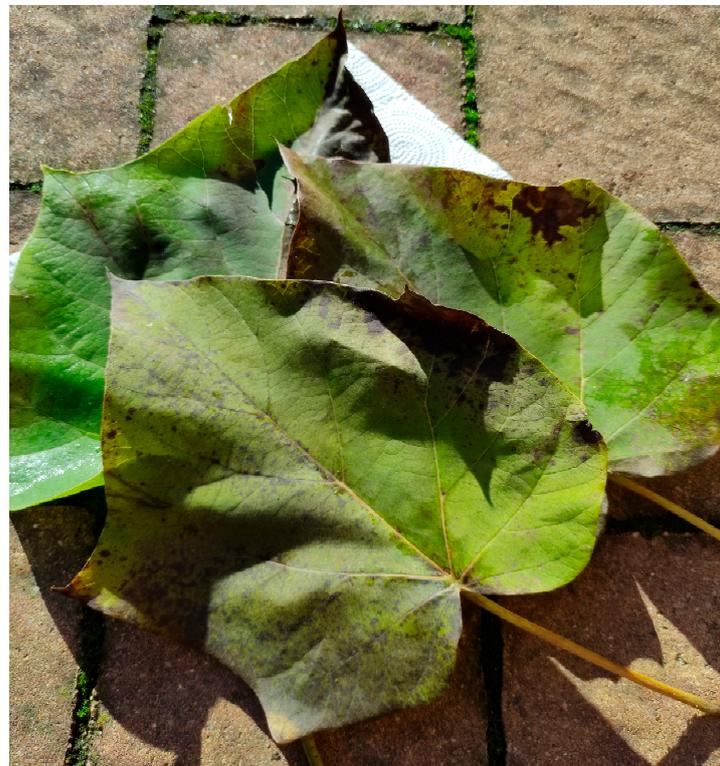
This study fills a gap in Oxytree leaf management practices, which fits in with the idea of sustainable development and the circular economy. The author has not found similar publications on Oxytree leaves, which until now have been treated as waste, sometimes as compost, from which products with fertilizing properties are produced.

## 2. Materials and Methods

As part of the work, 3 different types of collections of Oxytree leaves were analyzed. The first, the so-called “Growing”—freshly harvested leaves (stripped from the living tree), the second, the so-called “Fresh fallen”—collected on the next day after the plant is pruned or harvested for its wood, and the third so-called “Collected (1 week)” —gathered one week following wood harvest. In Figures 2–4, photographs of the appearance of the leaves used in the experiments are shown sequentially. The same samples were separated into 3 replicates, which were analyzed separately, both in terms of physicochemical and the entire methane fermentation process. The anaerobic digestion process was performed on all samples and replicates with both leaves substrate and inoculum for a period of 60 days. Such a long period was adopted due to dealing with certain variables within the Oxytree leaves substrate harvest, including that it came from various crops, and in order to eliminate the possible error caused by the repeated stage of substrate decomposition.



**Figure 2.** Pictures of leaves from the harvest “Growing”.



**Figure 3.** Pictures of leaves from the harvest “Fresh fallen”.



**Figure 4.** Pictures of leaves from the harvest “Collected (1 week)”.

### 2.1. Determination of Dry Mass and Organics and Elemental Composition

The course of the determination of dry mass (or, in other words, total solids—TS) is as follows: samples to be collected are weighed and then dried at 105 °C for about 24 h (depending on the size and moisture of the collected sample). After 24 h, the samples are removed from the dryer and then weighed.

The formula used to calculate the dry mass is:

$$TS = m_2/m_1 \times 100\% \quad (1)$$

where:

$m_1$ —sample mass before drying [g],  
 $m_2$ —sample mass after drying [g],  
 TS—total solids (dry mass) [%].

Each sample was analyzed in three replications.

The determination of the dry organic mass (VS) content consisted of the following steps: after drying and weighing the samples, they were placed in an oven heated to 550 °C, and at such a high temperature, the organic substances volatilize, and, after burning, only ash remains, which is a mineral mass. The samples are completely burnt within 3–4 h. After this time, they are taken out and weighed again.

The final value can be calculated from the formula:

$$VS = (m_1 - m_2)/m_1 \times 100\% \quad (2)$$

where:

$m_1$ —sample mass before combustion [g],  
 $m_2$ —sample mass after combustion [g],  
 VS—volatile solids (dry organic mass) [% TS].

Each sample was analyzed in three replications.

The analysis of the elements was carried out with the use of an X-ray spectrophotometer XRF Niton XL5, manufactured by Thermo Scientific, (Boston, MA, USA) with an SDD detector GOLDD technology (the GOLDD technology allows for significant improvements

in measuring accuracy and time). The analyzer enables the assessment of the content of the following elements: Sb, Sn, Cd, Pd, Ag, Mo, Nb, Zr, Y, Sr, Rb, U, Th, Bi, As, Se, Hg, Au, Pb, W, Zn, Cu, Re, Ta, Hf, Ni, Co, Fe, Mn, Cr, V, Ti, Ca, K, Ba, Al, P, Si, Cl, S, and Mg according to the manufacturer's method. A CHNS/O Thermo Scientific Flash Smart analyzer was employed to determine C and N, based on the national standard PN-EN 15,104:2011.

## 2.2. Determining the Quantity and Quality of Generated Gases

The biogas and methane efficiency analysis carried out at the Ecotechnology Laboratory at the Department of Biosystem Engineering at the University of Life Sciences in Poznań is based on the commonly used modified DIN 38414-S8 and VDI 4630 standards. Our Ecotechnology Laboratory was the first facility in the country to receive a certificate confirming the high quality of research on methane fermentation, issued by the German organization Verband Deutscher Landwirtschaftlicher Untersuchungs- und Forschungsanstalten—VDLUFA [62] and Kuratorium für Technik und Bauwesen in der Landwirtschaft—KTBL [63].

The test of biogas and methane efficiency (in a “batch culture” mode) was carried out in a 21-chamber stand designed and fabricated by employees of the Ecotechnology Laboratory (Figure 5), while the calculations were performed in a specially prepared calculation sheet working in the MS Excel environment.



**Figure 5.** View of batch culture test stands (designed and created by employees of the Ecotechnology Laboratory).

In such work, the appropriate dose of the substrate is placed in the reactor with a capacity of 2.0 dm<sup>3</sup> and mixed with the microbial inoculum (the so-called inoculum), and then placed in a water bath that maintains a constant temperature of 39 °C. The produced biogas is stored eudiometrically in graduated tubes filled with water, wherein the solubility of the gases in water is prevented by the use of a liquid–gas barrier at the liquid–gas interface.

Each test was run in triplicate, including control samples, for the final balance and calculation of the biogas production from the substrate alone.

The daily biogas production was measured, while the composition identification was performed using a GeoTech GA5000 gas analyzer. This device allows for the analysis of 5 gases at the same time in the following concentration ranges: CH<sub>4</sub> 0–100%, CO<sub>2</sub> 0–100%,

O<sub>2</sub> 0–25%, NH<sub>3</sub> 0–1000 ppm, and H<sub>2</sub>S 0–10,000 ppm. Additionally, the fermentation of the inoculum (modifiers) alone was performed as a control.

### 3. Results

#### 3.1. Initial Parameters of Substrate

The examined Oxytree leaves were analyzed in terms of their physicochemical properties: dry mass (TS), organic dry mass (VS), total carbon (C), total nitrogen (N), ratio C/N, and pH, obtained from three different harvesting periods, which are presented in Table 1.

**Table 1.** Characteristics of the investigated substrate—the initial parameters.

Leaves/Parameters->	TS	SD; VC	VS	SD; VC	C	N	C/N	pH
Unit	% FM	%	% TS	%	% TS	% TS	-	-
Growing	17.66	0.53; 2.97	93.60	0.46; 0.49	50.90	2.19	23.20	5.38
Fresh fallen	36.67	0.89; 2.42	86.11	1.07; 1.25	47.02	2.19	21.45	5.42
Collected (1 week)	24.78	0.58; 2.32	85.80	0.15; 0.17	46.01	2.20	20.96	5.39

SD—standard deviations VC—variation coefficient.

The dry mass content, depending on the harvesting method, was at least 17.66% (growing leaves) to 36.67% (for freshly fallen leaves). The organic mass content was greatest in the freshly growing leaves—93.6%, while that for the remaining leaves was approximately 86%. The total nitrogen and carbon contents were similar for all leaves and were 2.29% of N and 47.98% of C, respectively, giving a C/N ratio of almost 22. The average pH for the mixture of leaves was 5.49.

The elemental composition is presented in Table 2. Table 2 shows the results only for those elements for which the content in the sample was above the detection limit. For: Zn, Zr, Al, As, Au, Ba, Bi, Co, Cr, Cu, Hf, Hg, Mg, Mn, Ni, Pb, Sn, and Ta, their share was negligible.

**Table 2.** Elemental composition of the investigated substrate.

Leaves	Growing	Fresh Fallen	Collected (1 Week)
Elements	Units—ppm		
Ag	3829	4416	4043
Bal	937,610,750	936,310,750	936,856,063
Ca	19,800,084	20,248,988	20,401,322
Cd	12,186	11,738	13,297
Cl	453,866	500,118	662,751
Fe	265,755	300,924	318,112
K	32,943,508	33,162,426	32,103,723
Mo	5433	5992	5976
Nb	7853	8004	8122
P	1,795,822	1,917,396	2,027,264
Pd	2935	3587	3454
Rb	7706	7847	7931
Re	<LOD	<LOD	<LOD
S	4,735,686	4,629,216	4,630,707
Sb	12,475	12,895	13,065
Se	<LOD	<LOD	<LOD
Si	2,228,237	2,768,440	2,727,961
Sr	14,286	14,164	13,514
Th	1531	1688	1513
Ti	56,956	53,306	65,129
U	1418	<LOD	<LOD

### 3.2. Biogas and Methane Efficiency

The cumulated value of biogas and methane efficiency, as well as the methane content in the biogas, were determined for the leaves as determined for the harvest parameter (Table 3). Productivity was calculated in terms of fresh mass (FM), dry mass (TS), and dry organic mass (VS).

**Table 3.** Methane content in biogas, biogas, and methane efficiency.

Leaves/Parameters->	CH <sub>4</sub> ratio	CH <sub>4</sub>	Biogas	CH <sub>4</sub>	Biogas	CH <sub>4</sub>	Biogas
Unit	[%]	[m <sup>3</sup> /Mg FM]		[m <sup>3</sup> /Mg TS]		[m <sup>3</sup> /Mg VS]	
Growing	51.86	36.81	70.99	208.45	402.07	222.70	429.46
Fresh fallen	48.09	59.19	123.09	161.42	335.66	187.46	389.81
Collected (1 week)	50.66	36.56	72.18	147.56	291.31	171.98	339.51
Mean value	50.20	44.19	88.75	172.48	343.01	194.05	386.26

For each method of leaf harvesting, the share of methane in the biogas oscillated around 50% (mean value—50.2%). The biogas yield from the fresh leaf mass was the highest for “Fresh fallen”—over 123 m<sup>3</sup>/Mg, and comparable for “Growing” and “Collected”—slightly more than 70 m<sup>3</sup>/Mg, which is almost twice as low. In terms of dry mass and organic dry mass, the highest values were unequivocally obtained for the “Growing” harvest—approximately 402 m<sup>3</sup>/Mg and 430 m<sup>3</sup>/Mg, respectively. This corresponded to the highest initial solids and organic content in the leaves.

In the case of the produced methane, the results are symmetrical, i.e., the yields for the “Growing” and “Collected” sets are very similar and amount to slightly over 36 m<sup>3</sup>/Mg, and about 60% more was obtained for “Fresh fallen” (approximately 59 m<sup>3</sup>/Mg). Using dry mass conversion (TS), methane production is the highest for the “Growing” harvest—208 m<sup>3</sup>/Mg; for “Fresh fallen”, it is about 40 m<sup>3</sup>/Mg less, i.e., 161 m<sup>3</sup>/Mg, and only about 147 m<sup>3</sup>/Mg for “Collected”. The classification according to the methane efficiency calculated for dry organic mass (VS) appeared to be similar—close to 223 m<sup>3</sup>/Mg, 187 m<sup>3</sup>/Mg, and 172 m<sup>3</sup>/Mg, respectively, for “Growing”, “Fresh fallen”, and “Collected”.

The exact course and differences in the substrate gasification over time can be observed in Figures 6–8, wherein the curves of the cumulative methane production over the entire experimental period (equal for all analyzed cases) are presented. It should be noted that the presented data have been converted into units used in practice—m<sup>3</sup>/Mg, respectively, fresh mass, dry mass, and dry organic mass.

The slowest increase in methane (in terms of fresh mass—Figure 6) in the first weeks of the anaerobic digestion process was observed in the leaves from the “Growing” harvest. For this substrate, methane production was completed within 4 weeks. In turn, the highest productivity in the first days was recorded for “Fresh fallen” and “Collected”. While in the first week, in the case of the “Collected” harvest, the increase in the volume of methane was very intense, it stopped within the subsequent week (productivity ceased within a dozen or so days). Leaves from the “Fresh fallen” harvest, on the other hand, had quite sharp methane production characteristics for almost 3 weeks, after which, the further methane generation capacity terminated at a level of about 60 m<sup>3</sup>/Mg FM (as a cumulative yield).

In Figure 7, the course of methane production (in terms of dry mass) can be observed. The volume of CH<sub>4</sub> generated by the “Fresh fallen” set was increasing the slowest, but the process lasted over 2 weeks. The “Collected” set revealed similar productivity, but more intensively (in the first week). The cumulative yield of CH<sub>4</sub> in both cases (“Fresh fallen” and “Collected”) was similar at about 150 m<sup>3</sup>/Mg TS.

The “Growing” collection was characterized by an almost identically fast increase throughout the methane production process. In this case, the cumulative yield was almost 25% higher than for other sets, and the CH<sub>4</sub> production was maintained for over 4 weeks to reach a level of over 200 m<sup>3</sup>/Mg TS.

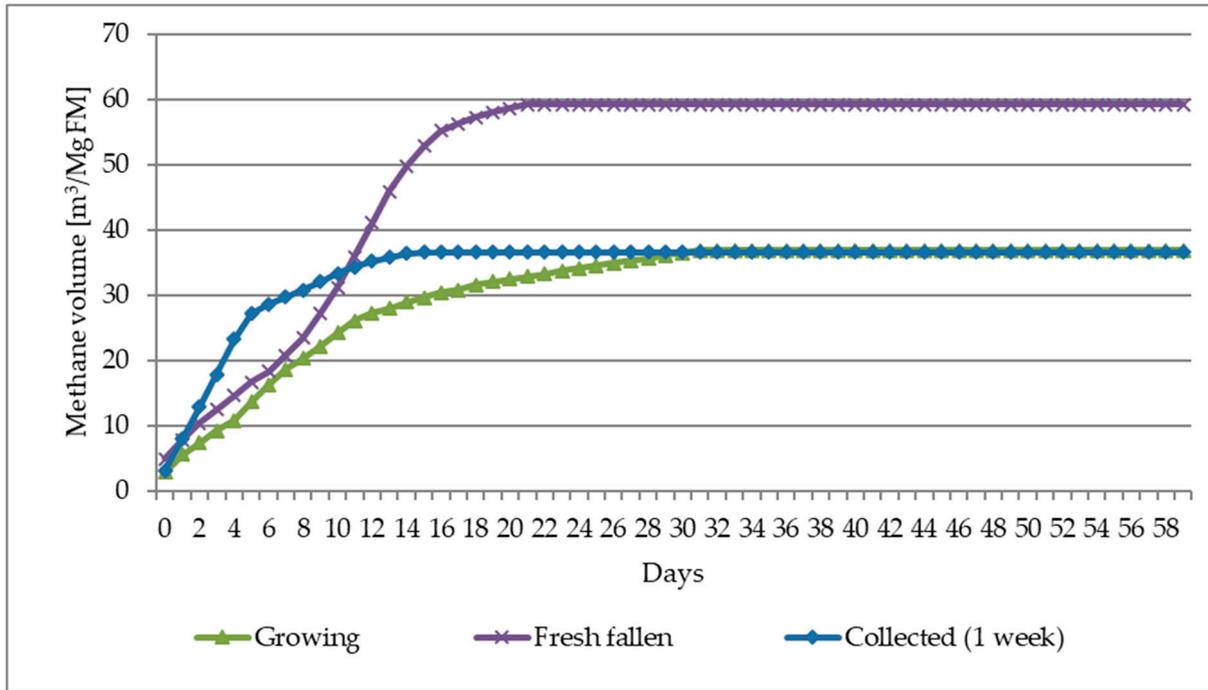


Figure 6. Methane production from fresh mass in time.

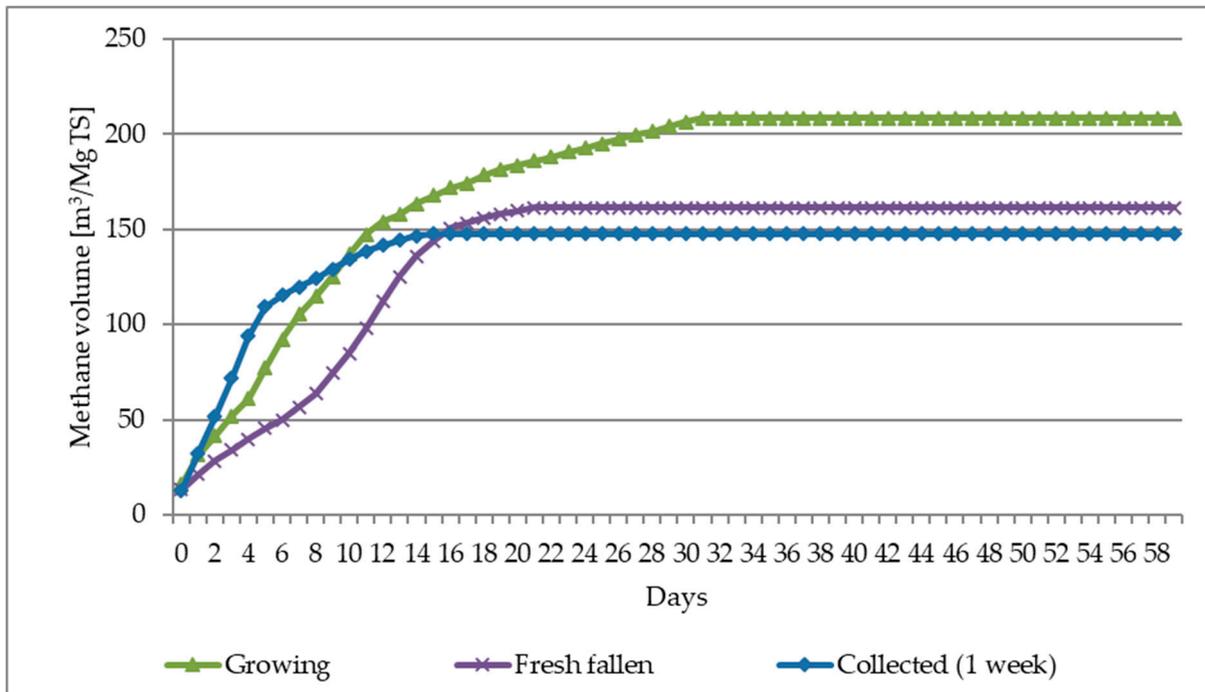


Figure 7. Biogas production from dry mass in time.

In the case of the analysis of methane production obtained from the dry organic mass of the substrate, the situation was very similar to the previous. The cumulative curves had similar sharp characteristics, and although the “Collected” set was the most intense, after about only two weeks, the methane production stopped. For the “Fresh fallen” harvest, this came about almost a week later, while the “Growing” set had a relatively long methane production process, with a fairly high process intensity. The time to complete the production for this experiment was around 4.5 weeks. During this time, about 223 m<sup>3</sup> of methane (from organic dry mass-VS) was generated, converted into Mg.

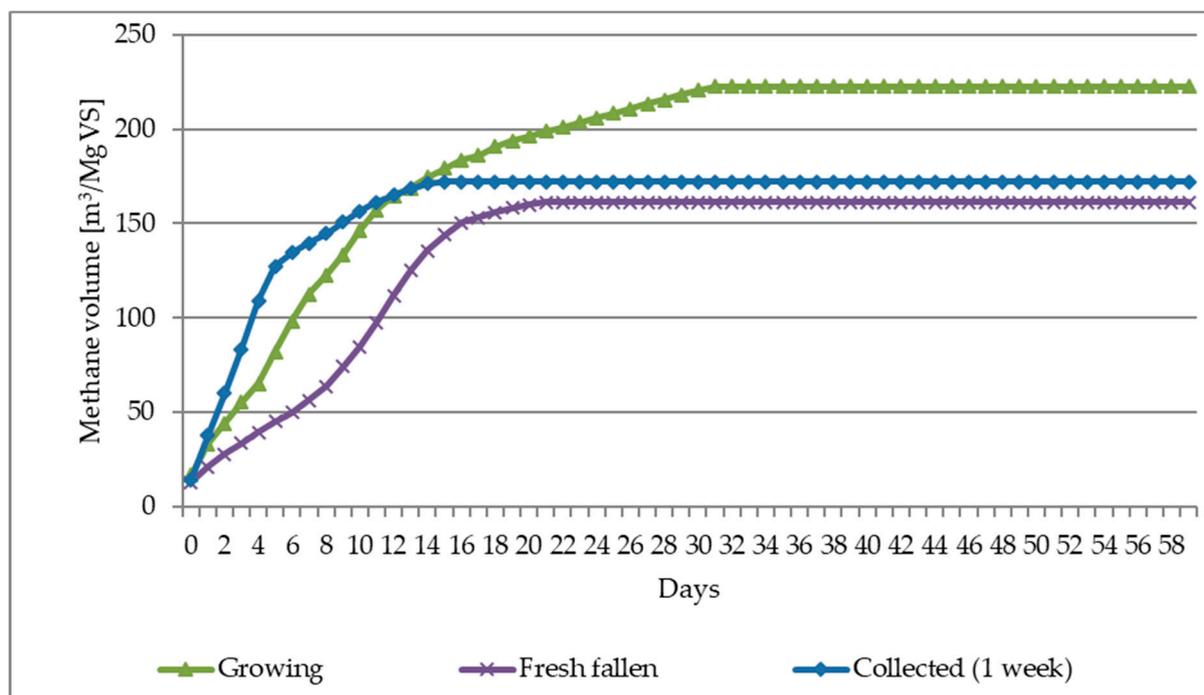


Figure 8. Biogas production from dry organic mass in time.

#### 4. Discussion

In Poland, the current use of biomass from Oxytree leaves to obtain biogas is quite limited, mainly because the plant is not currently planted to a great extent. This is mainly due to the climatic conditions in the country, which means that the wood productivity is not as high as in other parts of the world (Asia or southern Europe) [19,23].

However, the primary purpose of Oxytree crops is the production of wood biomass for the energy and cellulose industry, and the broadly understood construction industry. In most cases, leaves are waste that could be successfully used in the production of biofuel gas, i.e., biogas.

The results of the average yield of Oxytree leaves biogas and methane, carried out and described in the current publication, turned out to be promising. The production level of this biofuel (importantly—generated from raw materials often considered byproducts) corresponds to that of other plant feedstocks for biogas plants.

It should be noted that scientific reports can be found that state that different types of leaves may have the potential to reach up to 600 m<sup>3</sup>/Mg of biogas [64]. However, it depends on many factors—both its own characteristics and the method of cultivation, climatic conditions, and physical and chemical properties during the research period (e.g., dry mass, dry organic mass, C/N ratio, etc.). Nevertheless, compared to the yields of other substrates shown in the aforementioned publication [64], Oxytree leaves have a higher methane yield than, among others, straw from cereals; biowaste from household or overstored food, sewage sludge, or vegetable extraction residues; waste from paper and carton production; and liquid manure from cattle, pigs, chicken, and sheep (in some studies, the values were in a similar range [65–67]).

Contrary to the value given within the previous compilation (i.e., 600 m<sup>3</sup>/Mg VS), other literature sources show a much lower productivity from fallen tree leaves: 204 ± 19 m<sup>3</sup>/Mg VS, similar to that reported in the current research—on average, 194 m<sup>3</sup>/Mg VS (and for the average up to 223 m<sup>3</sup>/Mg vs. from the “Growing” harvest) [66]. Compared to the production of methane from chopped barley straw—280 ± 39 L Mg VS, the value from the current research is not much lower, and almost twice as high as the biomass of Baltic Sea algae (119 ± 62 m<sup>3</sup>/Mg VS) [66].

Research on the methane efficiency [68–71] of poplar waste has shown that it is in the range from 81.1 m<sup>3</sup>/Mg VS to 271.9 m<sup>3</sup>/Mg VS, which corresponds to the results of the current research: for “Growing”, “Fresh fallen”, and “Collected”, respectively: 222.70, 187.46, and 171.98 m<sup>3</sup>/Mg VS, which are also close to other popular substrates, including untreated rice straw (178.3 m<sup>3</sup>/Mg VS) [72], as well as 3% H<sub>2</sub>O<sub>2</sub> treated corn straw (216.7 m<sup>3</sup>/Mg VS) [73]. It is to be expected that biogas efficiency would be significantly increased by grinding and extrusion prior to addition to the fermenter, as this positively affects other substrates also with high lignin content. In the case of maize straw research, the lignin content is approximately 22% which is the same or a similar composition as that of Oxytree [74,75]. The treatment of Oxytree leaves and other pretreatment techniques for this type of lignin-rich substrate reported in the literature could also significantly increase biogas productivity [76–78].

Comparative studies of methane productivity carried out on potato peelings, corn waste, and fallen leaves showed 91.66, 81.81, and 63.13 m<sup>3</sup>/Mg per dry mass, respectively [79]. For all methods of leaf harvesting from the current research, much higher values of the produced methane were achieved—on average, approximately 173.5 m<sup>3</sup>/Mg TS.

During the research on grape leaves obtained from Polish crops, up to 59.0 m<sup>3</sup>/Mg of fresh mass was obtained, which means a much lower yield than in the current research, in which the average value was almost 89 m<sup>3</sup>/Mg [80]. The authors of that study, however, note that from herb production residues, usually much more can be obtained—70–300 m<sup>3</sup>/Mg.

Compared to the biogas (and biomethane) efficiency studies from olive leaves and green and dry palm leaves, the production achieved in the current study is much lower [81]. The analysis of both of these studies is, however, not complete because the international team of researchers reported the yields of biogas and methane only in terms of fresh mass. Freshly picked olive leaves, green palm leaves, and dry palm leaves were, respectively (in m<sup>3</sup>/Mg) 107 and 71.29; 142 and 94.75; 132 and 90.46 [81]. According to my study, the harvest of Oxytree leaves as “Growing” and “Collected” showed much lower efficiencies—just over 70% m<sup>3</sup>/Mg FM for biogas and just over 36% for methane, which is about 30% less than for olive leaves, almost twice less for biogas, and up to three times less for methane than the figures stated for palm leaves (green and dry) [81]. The outcomes are clearly more for the collection of “Fresh fallen” Oxytree leaves. It should be noted that the dry mass of olive leaves and green palm leaves was almost 50%, and of dry leaves, almost 80%. This would correspond to almost two–three-fold differences for this parameter in Oxytree leaves, which could explain such low values when compared to dry mass.

The biogas plant was fed with mixtures of leaves (6% TS), crushed, and directly followed by aerobic pretreatment (mainly, by mass, consisting of: mahogany leaves—75%, eucalyptus leaves—10%, and rain tree leaves—15%) and with 2% cow manure (converted into 8% TS), a maximum yield of 199 m<sup>3</sup>/Mg was achieved [82]. This value is comparable to the best “Growing” value. When comparing those studies in the version without leaf treatment mixed with manure, the biogas production was at an even lower level—only 106 m<sup>3</sup>/Mg, which is a worse result than the least efficient collection demonstrated in my work—“Collected” (147.56 m<sup>3</sup>/Mg). Although the content of methane in the biogas was quite high—around 69.3% (current research maximum 51.86%)—this was partly due to the addition of cow manure.

Other studies, in which the leaves were an addition to the substrate mixture, were carried out with a different share of co-substrates in the periodic fermentation of cultures in mesophilic conditions. For this purpose, the researchers used sugar cane leaves (SL) and solid fractions of cow manure (CM) and food waste (FW). The optimal mixing ratio of FW:SL:CD (relative to VS) was 85:11.25:3.75, respectively, achieving a methane yield of about 297 m<sup>3</sup>/Mg VS, and a methane content of over 73% [83]. In addition, the researchers showed that the cumulative methane production was higher by about 110% and 445% than in the monofermentation of sugar cane leaves and cow dung.

Some publications have shown that in the case of fermentation, whether for the production of methane or biohydrogen, a synergistic effect is obtained using sewage sludge as a co-substrate [53–57]. It should be noted, however, that these processes usually require much more advanced methods of substrate preparation (and thus more energy and chemicals). They include, e.g., high-temperature sterilization to kill hydrogen-consuming bacteria and periodic cooling (mainly sewage sludge) or thermochemical pre-decomposition using 1% HCl (*w/w*) at 100 °C [53,84,85].

In terms of carbon and nitrogen contents, the tested Oxytree leaves showed similar or better values than those of other commonly grown energy and fast-growing plants that are often colored—e.g., *Sida hermaphrodita*, *Miscanthus x giganteus*, *Arundo donax* or *Populus x euroamericana*, *Salix viminalis* clone TORA—they had from 43–50% of carbon and 0.3–1.3 nitrogen, while the Oxytree leaves tested in the current study had an average of 48% carbon and as much as 2.2% nitrogen [18]. The composition of the leaves of Oxytrees varies greatly, in particular with regard to the growing climate and species. In the Irish publication, the N/P/K is 2.8%/0.6%/0.4%, respectively, and in the current study, it is 2.2%/2.0%/3.3%, whereas in the case of Ca/Fe, it was 2.1%/0.6%, and in my work, 2.0%/0.3% [19]. Compared to sugarcane leaves (SL), Oxytree leaves contain approximately 5% more carbon and twice as much nitrogen and a closer-to-optimum C/N ratio—Oxytree is about 22, and SL, 36.5, with similar contents of dry mass and dry organic mass [83]. The elemental composition, including NPK macronutrients, and the high content of organic mass make Oxytree leaves a good fertilizer product both directly and after the fermentation process as digestate (the N content does not decrease slightly).

Even if we adopted the variant without biogas use, apart from highly advanced management methods (expensive and complicated), only the composting of leaves remains. However, as research conducted on leaves from Berlin trees has shown—this solution is three times more harmful to the environment. In this study [86], three scenarios were analyzed to assess the use of leaves: (a) composting (business-as-usual); (b) biogas production; and (c) the pre-treatment of the leaves prior to use in methane fermentation processes. For the considered variants, GHG and energy potential were estimated. As part of the calculation, a model of the impact of the use of biological resources was applied, and the locations and efficiencies of the existing agricultural biogas plants were taken into account (which should be disadvantageous—the long transportation of leaves increases exhaust emissions and thus GHG). Based on the results, it was found that in terms of GHG emissions, the biogas scenarios gave the best ecological effect in the form of a negative carbon footprint, from  $-140.1$  kg CO<sub>2</sub>eq/Mg of leaves to even  $-167.4$  kg CO<sub>2</sub>eq/Mg of leaves. The composting solution (business-as-usual) resulted in emissions of 49.0 kg CO<sub>2</sub>eq/Mg of leaves. In addition, a reduction in leaf rot, such as increased biogas plant loading or ensilage, resulted in even lower net GHG emissions and higher energy efficiency. The production of biogas by using the substrate as leaves from urban trees has enabled the production of a renewable energy source in the form of biofuel. It has been estimated that around 7.5 Mg of processed leaves would correspond to an average electricity consumption of one person for a full year. Composting leaves and other green waste can cause even many times higher emissions, significantly exceeding 100 kg CO<sub>2</sub>eq/Mg [86–88].

## 5. Conclusions

Based on the presented calculations and considerations, the following conclusions can be drawn:

1. Considering the climatic conditions and growth requirements of Oxytree, it is one of the best species to grow as an energy crop plantation. Its leaves, which are usually waste material, regardless of harvest, are a valuable source of chemical energy as biogas (liquid biofuel).
2. Its elemental composition and physico-chemical characteristics are very similar (often even greater) to other fast-growing or energetic plants.

3. The collection of leaves is quite convenient because they fall at the same time and they are large and heavy (they are in close proximity to the plant in large clusters).
4. The average biogas yield from all leaf-harvesting methods was approximately 386 m<sup>3</sup>/Mg of dry organic mass (VS), and for the most effective biogas harvesting—“Growing” (fresh leaves)—even 430 m<sup>3</sup>/Mg VS.
5. The average methane yield from all methods of leaves harvesting was about 194 m<sup>3</sup>/Mg of dry organic mass (VS), and for the most effective biogas harvest—“Growing” (fresh leaves), it was even 223 m<sup>3</sup>/Mg VS.
6. The average biogas yield from all methods of leaf harvesting in terms of fresh mass (FM) was approximately 44 m<sup>3</sup>/Mg, and for the most biogas-effective harvest—“Fresh fallen” (recently fallen leaves—maximum 1 day)—even 59 m<sup>3</sup>/Mg FM.
7. The average methane yield from all methods of leaves harvesting in terms of fresh mass (FM) was approximately 89 m<sup>3</sup>/Mg, and for the most biogas-effective harvest—“Fresh fallen” (recently fallen leaves—maximum 1 day)—even 123 m<sup>3</sup>/Mg FM.
8. The use of Oxytree leaves in anaerobic digestion processes contributes to reducing the carbon footprint of Oxytree crops productions.

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