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Review

Possibilities and Limitations of Anaerobic Co-Digestion of Animal Manure—A Critical Review

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Abstract: Anaerobic digestion is a well-known and long-used biological method for stabilizing organic materials. Among the benefits of this process in waste management are the reduction of greenhouse gases, the production of alternative energy, and the acquisition of valuable digestate that can be used in the form of biogas, thereby closing the cycle of elements in nature. For some materials, such as manure, which is heterogeneous in terms of morphology and chemical composition, digestion of a single substrate may not be very efficient. Therefore, more and more studies on the co-digestion process are appearing in the literature. This solution allows higher biogas production and the possibility of processing several wastes simultaneously. The prospect of the future effective application of anaerobic co-digestion depends on regulations, work regime, and access to raw materials. Therefore, there is a need to systematize the available knowledge and results, as well as to identify the possibilities and limitations of the discussed process, which is undertaken in this paper.

Keywords: anaerobic digestion; co-digestion; biogas; animal manure; renewable energy; methane

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1. Introduction

Rational management of the Earth's resources is becoming more and more important in widely understood environmental protection. Scientists are undoubtedly facing the challenge of reducing global warming, which has the consequence of ozone depletion and ecosystem decline. Recent reports by Greenpeace Southeast Asia and the Centre for Research on Energy and Clean Air (CREA) indicate that air pollution from the burning of fossil fuels (coal, oil, and natural gas) is responsible for approximately 4.5 million deaths each year worldwide, and the global economic loss from fossil fuel air pollution is estimated to be $2.9 trillion per year or about 3.3 percent of global GDP. In the US alone, fossil fuel combustion emissions are associated with approximately 230,000 deaths and $600 billion in economic losses annually [1]. There is increasing focus on the integration of energy, water, and environmental systems, including aspects related to the development of transport, industry, and agriculture. Therefore, it is important to take a holistic approach to the presented problem, and above all to adopt a policy aimed at the efficient use of alternative energy and rational waste management [2,3].

Reducing greenhouse gas emissions and developing a reliable strategy for saving fossil fuels is of particular interest to policymakers. In the area of climate policy, the European Union aims to achieve the so-called net-zero emission by 2050, which is the main postulate presented in the European Green Deal project. Over the 21st century, the proposed thresholds for reducing the emissions of gases responsible for global warming and the use of renewable energy sources are increasing [4]. Figure 1 shows the direction of the EU authorities' action in the field of climate and energy policy [5].

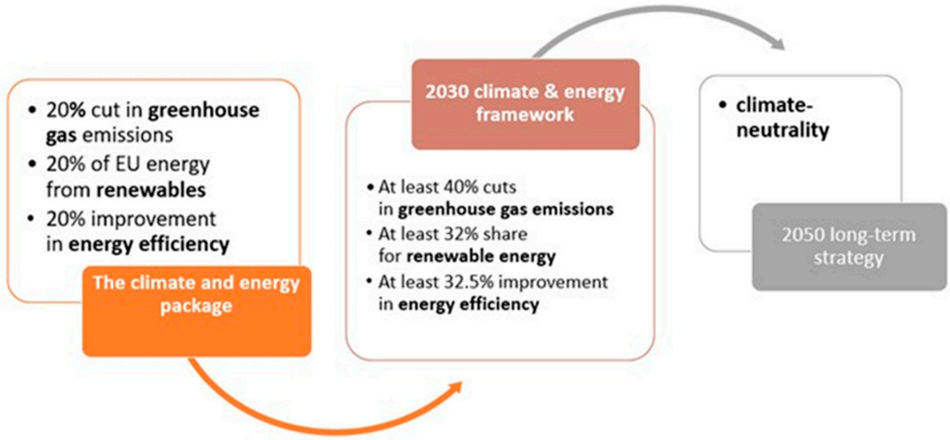

Figure 1. The most important demands of the 21st century regarding climate policy proposed by the European Parliament [5].

In the face of progressive climate change and the need for rapid reduction of conventional energy sources, the search for appropriate waste treatment methods is extremely important. One of the major producers of waste is the agri-food sector, which generates, among other forms of waste, animal excrement. Animal husbandry is one of the main sources of atmospheric pollution and greenhouse gas (GHG) emissions globally. Methane (CH_4), nitrous oxide (N_2O), carbon dioxide (CO_2), ammonia (NH_3), hydrogen sulfide (H_2S), methyl mercaptan (CH_3SH), di- and trimethyl sulfide, volatile organic compounds, endotoxins, and poisons can be emitted from livestock manure, causing serious environmental pollution and health problems [6]. GHGs discharged from animal production account for 18% of total global emissions [7]. Globally, cattle farming is leading the way (Figure 2, Tables 1 and S1), but due to the expected growth of the global human population, the increase in global total meat production is estimated to increase from 330 million tons (2017 data) to 465 by 2050 [8]. As a result, intensive livestock farms will increase, leading to an increase in the production of different types of manure, which, if untreated, can lead to serious environmental problems [9]. The slurry is a heterogeneous mixture of feces, urine, food residues, a small amount of litter, and technological waters (from animal washing, flushing, positions, and canals). Factors affecting manure properties include species and age of animals, type of feed used, and the content of litter and food residues, as well as ambient temperature (Table 2) [10]. Improper manure management can lead to ground and surface water pollution, deterioration of the biological structure of the soil, release of animal pathogens, attracting of pests, and generating odors [11,12]. In Poland, which ranks among the leaders in poultry farming in the world, this industry is developing extremely rapidly, resulting in increased manure production [13]. Environmental problems related to poultry production have been a concern in the last century. Farm animals are usually enclosed for most of their lives in large clusters in small rooms, which leads to large volumes of feces accumulating in concentrated areas [14]. Farmers face the challenge of tuning their livestock operations to increasingly stringent regulatory requirements [15]. Poultry manure is organic matter, rich in valuable elements such as nitrogen, phosphorus, and potassium, and is traditionally stored and used in agriculture (Table 2). However, direct application to arable fields leads to various environmental problems related to their physicochemical, hygienic, and sanitary properties [16]. This method also causes great irritation on the part of local society because of the unpleasant odor. Stored and unused chicken droppings emit large amounts of methane, carbon dioxide, and ammonia into the atmosphere. It should be emphasized that in the event of storing poultry manure in heaps, the temperature often increases spontaneously, reaching the level of 30–40 °C after the beginning of the aerobic decomposition process. Later, in turn, rapid oxygen consumption and the transition to anaerobic digestion occur. During this time, there are high emissions of methane (even up to 80 kg per 100 birds per year), which has a 21-fold stronger greenhouse effect than carbon dioxide [17].

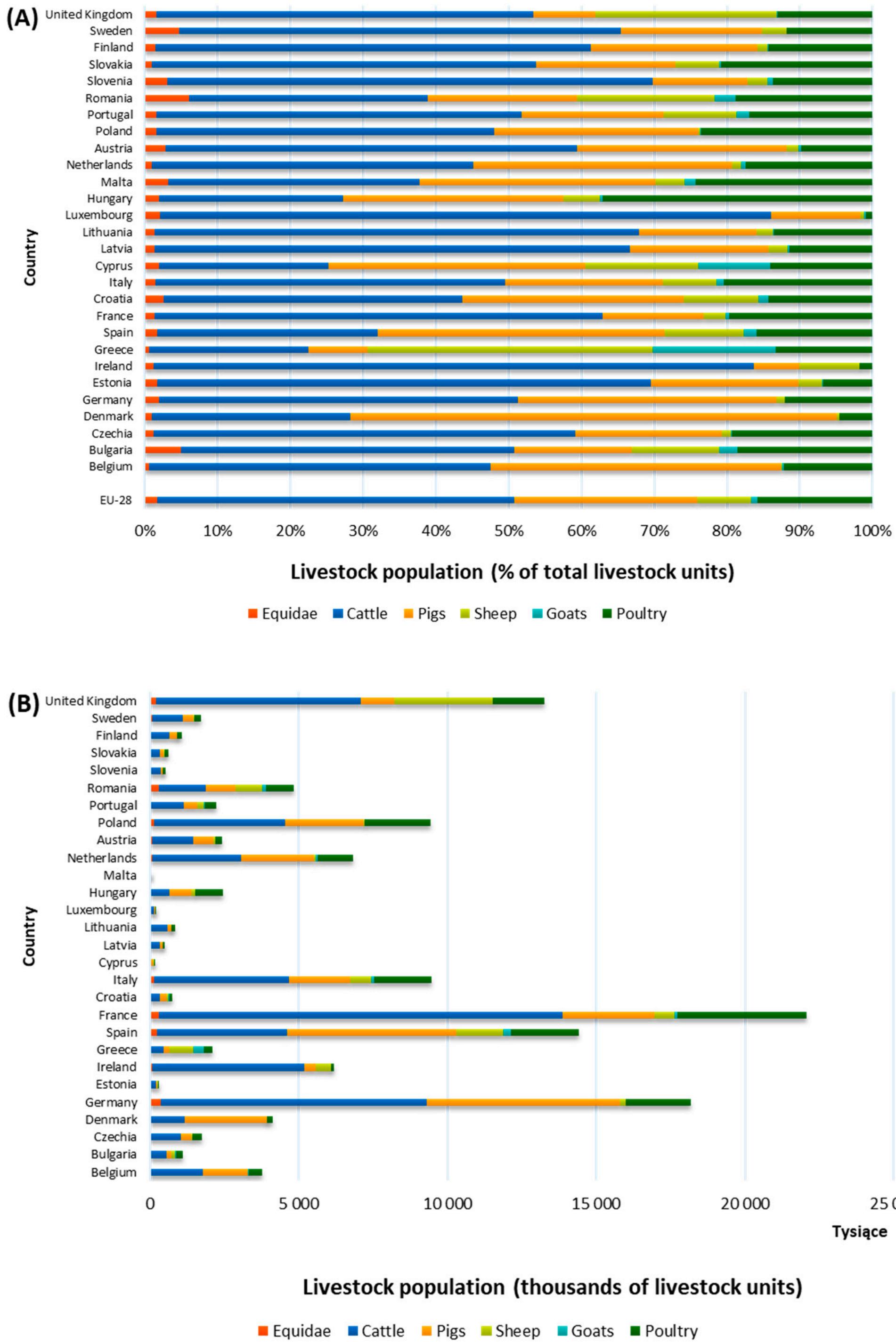

Figure 2. Livestock population in livestock units by type in Europe in 2016: (**A**) in % of total livestock unit; (**B**) total livestock unit (heads) [18].

Table 1. The quantities of manure produced by different animals [19–21].

Animal	Unit	Value
Laying hens	kg/(1000 birds·d)	120–150
Chicken broilers		80
Turkeys		200–350
Duck		150–190
Geese		200
Calves	kg/(head·d)	8.0
bovine		20.0
Male bovine		25.0
Dairy cows		53.0
Other cows		25.0
Piglets		0.5
Other pigs		4.5
Sows		11.0
Sheep		1.5
Goat		1.5
Broilers		0.10
Laying hens		0.20
Other poultry		0.3

Table 2. Characteristics of different types of raw manure [22].

Type of Manure	Manure	TS (%)	C (%)	N (%)	P (%)	K (%)
BM	Cattle, Horse, Sheep, or pig	20.9–69.9	11.9–12.0	0.4–2.2	0.2–4.0	0.9–4.0
	Poultry	33.3–78.5	12.6–50.4	1.1–5.9	1.1–3.2	2.0–3.3
SM	Cattle, Horse	24.4–65.0	10.4–48.1	0.6–4.6	0.1–2.5	0.1–3.2
	Pig	28.0–29.0	35.3–41.0	1.3–2.7	1.5–3.2	0.7
	Poultry	33.0–79.4	24.9–46.2	1.7–7.1	0.7–6.7	1.9–5.0
SL	Cattle	0.5–8.3	17.5–36.5	0.2–2.8	0.04–0.1	0.4–0.5
	Pig slurry	0.3–8.3	16.3–41.4	0.1–3.4	0.01–3.1	0.1–2.5
	Cattle, Horse	4.9	NA	NA	0.05	0.2
	Pig	<1.6	NA	0.1	1.0	NA

BM—bedding manure; SM—solid manure; SL—semi-liquid; NA—not available.

However, the high energy potential of animal manure can be used in a controlled manner [9]. The use of biomass for energy purposes and its conversion into fuels—for example, anaerobic digestion—has increased rapidly over the last decade. The anaerobic digestion market is growing at a rate of around 10–12% across Europe and is expected to exceed $8 billion by 2024 [23]. Anaerobic digestion is now a well-known and well-established technology on a large scale in Europe, primarily in rural areas such as Germany and Denmark [24,25]. The motive for using this method is primarily to save operating costs on farms. However, research on improving the efficiency of operating systems should be intensified, and policies should be implemented to encourage the use of biogas. Animal excrements are an attractive raw material for this process, but they are heterogeneous in size, composition, structure, and properties, and show different degradability by enzymes or bacteria. They often contain high concentrations of lignocellulose, which reduces their biodegradability and increases the required retention time in the anaerobic digestion process [24,26,27]. Characteristic in the case of animal droppings, especially chicken droppings, is a high content of ammonia, which is a process inhibitor. Diluting manure to 3–6% of the total solids eliminates the problem and ensures good mixing conditions for anaerobic tanks, but the biogas yield (and methane content) is often too low for profitable production due to the larger reactor volumes required, the water consumption, and also the production of large volumes of digestate slurry [28].

Hence, the need to use new solutions related to the proper treatment of the substrate and obtaining greater efficiency of methane production, which can be achieved by adding other substances to the charge (anaerobic co-digestion) [17], among other methods. The anaerobic co-digestion process is increasingly being studied, with the involvement of various materials and wastes, as confirmed by the literature. In the case of animal manure, a large number of results have been published, while only a few papers have attempted to systematize the knowledge and evaluate the prospects of using the method. This work provides an overview of the available information and collected results on manure processing through co-digestion. Particular attention has been paid to identifying opportunities for intensification of biogas production from manure and optimization of the process. In addition, this issue is covered in broad terms, taking into account the factors influencing the high production of animal manure in the world, its processing by biological technologies, and the management of the resulting products.

2. Anaerobic Digestion in the Face of Rational Animal Manure Management

The principles of storage and management of manure are regulated via a variety of legal acts. The correct method of recycling this waste is important for sustainable development and environmental protection. EU member states are primarily obliged to apply EU law, which in this situation prevails over national legislation. One of the legal frameworks that take into account the use of animal manure as fertilizing product is the Nitrates Directive [29], which obliges member states to create a code of good agricultural practices. In Poland, the Code of Good Agricultural Practice is in force, containing a set of environmentally friendly solutions for the use and collection of natural fertilizers [30]. The green light for the effective and safe use of manure as a natural fertilizer came with the entry into force of Regulation (EU) 2019/1009 of the European Parliament and of the Council on 5 June 2019 [31]. This document allows the natural use of manure, classified as category 2 animal by-products, provided it is first properly treated.

Due to the intensive production of animal excrement, there is a problem with their current use, hence the need for their proper storage and use. Slurry storage methods include manure storage on heaps in a field, a deep barn under an inventory, or deposited on a liquid manure pit with a sealed bottom and a slope that allows water to drain off. However, in organic farming, composting or anaerobic digestion is recommended [30,32,33]. For example, the storage of fresh manure takes about 4–6 months. During this time, its humidity decreases even to 20–30%, and gases produced due to microbial activity migrate to the atmosphere. Mroczek et al. [34] report that thermal drying is a better solution, where the dryer capacity fluctuates between 0.8–1.5 tons per hour. Unfortunately, odors are a significant problem in this process. According to Best Available Techniques (BAT), it is recommended to dry poultry manure to a content of 90% dry matter. The resulting product can be stored in closed rooms and further processed in the pelleting process. Pellets obtained from compressed manure can be used for energy or fertilizing purposes. It has the form of a briquette or pellets and has a calorific value similar to wood, and little ash is produced during combustion.

Combustion is a relatively simple method of manure management and is economically advantageous, due to high electricity production. The BMC Moerdijk power plant in the Netherlands processes 430,000 tons of poultry litter per year, generating 285,000 MWh of green energy and 60,000 tons of high-quality fertilizer in ash form [35]. Combustion, despite its simplicity, is an extremely invasive method in relation to the environment because the process generates compounds such as carbon monoxide, volatile phosphorus compounds, dioxins, and furans, which require the use of special filters. Another disadvantage of this process is the loss of nitrogen [36].

Therefore, more and more emphasis is placed on biological utilization techniques, among which composting and anaerobic digestion stand out. Organic recycling such as the composting process is a natural process of decomposing organic matter by aerobic bacteria, fungi, and nematodes. Thanks to organic recycling, a product with a higher organic nitrogen content and less odor nuisance is obtained. Particularly noteworthy is the vermicomposting technology, which involves earthworms. However, due to the sensitivity of the organisms, in this case, a mixture of feces undergoing the process to obtain a neutral pH is recommended and about 12–16 months of aging of the substrate are required. Among the mentioned methods, the greatest hopes are associated with digestion related to biogas production [34].

Anaerobic decomposition of organic substances is one of the oldest biological processes and the beginnings of its use were associated with a lack of thorough knowledge of the microbiological activity of the organisms involved. Sewage sludge is a common waste subject to anaerobic digestion. The positive effects of processing the waste in this way prompted the development of this technology and the attempt to use other types of organic substrates, both solid and liquid. Virtually any such organic matter can be a raw material for biogas production, provided it does not contain inhibiting substances [37]. Recycling animal waste reduces the amount of manure and uncontrolled greenhouse gas emissions and prevents the migration of pollutants into ground and surface waters. In addition, it contributes to the sustainable development of energy production. Anaerobic digestion is an economically viable process and is socially acceptable due to its environmental friendliness. In 2020, biogas production in Europe reached 191 TWh, of which 32 TWh was upgraded, while the rest was used to produce local heat and electricity [38]. Figure 3 shows the realistic potential of methane production from manure in different European countries based on data from 2018 estimated by Scarlat et al. [21]. The realistic biogas potential, calculated based on total collectible manure and specific biogas yield, is about 16.1 billion m^3 biomethane in the EU and 17.8 billion m^3 in the whole of Europe. For example, this is equivalent to the natural gas consumption of Belgium each year [21]. The economic efficiency of biogas production largely depends on the cost of the substrate, which seems to be a cost-effective solution in the case of waste [39,40]. Figure 4 shows the main advantages of the digestion process [26].

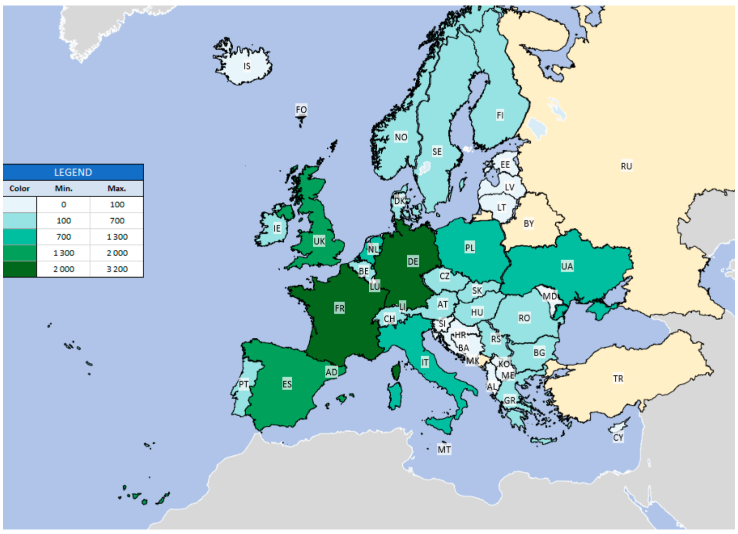

Figure 3. The realistic potential of methane production from manure in different European countries based on data estimated by Scarlat et al. [21]; value at mln m^3 methane (yellow—not available).

Figure 4. Advantages of the anaerobic digestion process [26].

The benefits of using anaerobic digestion to utilize animal manure are primarily waste stabilization, odor control, energy production, reduction of pathogenic organisms, preservation of biogenic elements, inactivation of weed seeds, compliance with progressive legal restrictions, and social acceptance [26]. The carbon in organic waste is part of the renewable carbon cycle so that CO_2 from the combustion of waste biogas does not represent additional GHG emissions, unlike conventional management practices where carbon from waste is oxidized to CO_2. Therefore, the use of waste-derived biogas should be considered climate neutral, so that replacing fossil fuels with biogas mitigates GHG emissions, provided that fugitive CH_4 emissions are properly managed. Furthermore, during anaerobic digestion, the organic nitrogen present in the feedstock is converted to nitrate (NO_3) and NH_3 and is retained in the digestate residue. Digestate has low levels of pathogens and associated odors compared to untreated animal manure and contains nutrients that are readily absorbed by plants. Sigurnjak et al. [41], in their studies, have shown that digestate can be used to replace synthetic fertilizers without the loss of crop yield. The use of digestate as a fertilizer and soil improver also helps to maintain soil carbon content. In addition to the traditional use of biogas, the process of biogas production can be integrated into other agricultural activities: providing energy to farms and using the digestate to fertilize crops for food, feed, and other value-added by-products [42,43].

In the anaerobic digestion process occur complex biochemical reactions. Nevertheless, from a process engineering point of view, this method is considered relatively simple because no sterilization steps are required ("mixed culture" enrichment of ubiquitous organisms) and there is no need to separate the biogas product as it separates from the aqueous phase [26]. Anaerobic digestion is a relatively simple method of manure management and is economically advantageous, due to high electricity production.

3. The Most Important Factors Affecting the Anaerobic Digestion of Animal Manure

Biogas consists of methane (CH_4), carbon dioxide (CO_2), and trace amounts of nitrogen (N_2), hydrogen sulfide (H_2S), ammonia (NH_3), and water vapor. The exact composition of biogas depends on the biomass sources and the used technology [44]. The effectiveness of anaerobic digestion as a biological process depends primarily on the activity of microorganisms, among which should be distinguished methanogenic species extremely sensitive to environmental conditions. The stages of anaerobic degradation are hydrolysis, acidogenesis, acetogenesis, and methanogenesis [45]. Hydrolysis decomposes polymers such as cellulose, starch, and proteins into monomers by exoenzymes [9]. During acidogenesis, acetate, H_2, CO_2, and volatile fatty acids (VFAs) are formed, while acetogenesis produces acetic acid. The last stage, methanogenesis, runs in parallel to the third to convert CO_2 and H_2 into methane [46]. Thus, the course of the process depends largely on the chemical composition of the substrate, its amount and frequency of introduction as well as parameters like temperature, digestion time, pH value, and the presence of toxic substances such as ammonia, ammonium nitrogen, hydrogen sulfide, or heavy metals. Current control of the above-mentioned factors allows for the effective work of bioreactors and effective biogas production [10]. Below is a more detailed description of the main factors affecting the process of animal manure digestion.

3.1. pH

Organisms participating in individual stages of the anaerobic digestion process show different tolerance to reactions. However, it is assumed that their increase is optimal for a pH value in the range of 6.8–7.5. Undoubtedly, the buffer capacity of the substrate is an important parameter related to the reaction. When the acid–base balance is disturbed, process-inhibiting substances are formed. The buffer capacity of the charge is affected by alkalinity (amounts of carbonates and acid carbonates) and the content of undissociated organic acids. High concentrations of acids in the undissociated form lead to an increase in the carbon dioxide content, resulting in a decrease in the pH value [10]. A sharp drop in pH usually results in a complete collapse of the process. Animal manure is characterized by a relatively high pH (even up to 10), but also a large buffer capacity [47].

3.2. Volatile Fatty Acids, Alkalinity

A useful indicator for controlling acid–base digestion conditions is the ratio of volatile fatty acids (VFAs) to alkalinity. An increase in this ratio precedes the appearance of critically low pH, which can prevent the process from falling faster [10]. For proper digestion, the content of volatile fatty acids is between 100–500 g/m^3 [10], while the VFAs/alkalinity ratio should not exceed 0.3 [48]. Wang, Xang et al. [49], in their work on the impact of VFAs on methane production efficiency, also point to the important role of the acidogenesis phase on the anaerobic digestion product. Volatile fatty acids are first converted to acetic acid before the degradation to methane. The incorrect conversion rate of volatile fatty acids can lead to changes to the desired order: acetic acid > ethanol > butyric acid > propionic acid; this change can cause the accumulation of propionic acid, which fails in the methanogenesis stage. Methanogenic species responsible for the production of the final product of anaerobic degradation are therefore the most exposed to the toxic effects of acids [45]. It is also worth mentioning that low pH also increases the mobility of heavy metals, which are process inhibitors [48].

3.3. Ammonia

In the opposite case, namely when the reaction is alkaline, ammonia harmful to methanogens is formed [10]. During anaerobic digestion, some organic nitrogen is bound by organisms in biomass, which depends on the C/N ratio of the substrate. On the other hand, the unbound nitrogen changes into the ammonium form. However, it should be noted that the higher the dissociation rate of ammonia, the lower its toxicity. Low pH causes a higher proportion of undissociated ammonia. At a concentration of 50–200 mg/L, this compound affects anaerobic processes because ammonium nitrogen is the basic element in the synthesis of amino acids, acids, and nucleic acids. In addition, ammonium nitrogen, due to its chemical properties, neutralizes the acids present in fermentative bacteria and thus helps maintain the neutral pH conditions that support cell growth. If ammonia is present in high concentrations, it can lead to the inhibition of the process [23]. It is generally recognized that the inhibition of ammonia consists mainly of the inhibition of the methanogenesis phase [23]. The adaptability of methanogens to the increasing concentration of ammonia depends mainly on the rate of its formation, which is associated with the substrate load, process temperature, and hydraulic retention time [16,50,51]. It is difficult to determine the limit value of ammonia concentration, which causes inhibition of digestion, due to the discrepancy in the results of studies of various authors. The mechanism underlying the inhibition is still not sufficiently defined, with reported concentrations ranging from 1500–7000 mg/L [23]. In aqueous conditions, ammonia occurs mainly in two forms, as ionized ammonium ion (NH4+) stable in the aqueous phase and in gaseous form as free ammonia (FAN), which is represented by Equation (1) [23]:

$$NH_3 + H^+ \underset{k_{-1}}{\overset{k_1}{\rightleftarrows}} NH_4^+ \quad (1)$$

The relative fraction of free ammonium nitrogen (FAN) relative to the total ammonium concentration is related to the pH and temperature of the solution, as shown in Equation (2) [23]:

$$FAN = TAN \times \left(1 + \frac{10^{-pH}}{10^{-\left(0.09018 + \frac{2729.92}{T(K)}\right)}}\right)^{-1} \quad (2)$$

FAN = concentration of free ammonium nitrogen (mg L^{-1})
TAN = total ammonium concentration (mg/L)
T(K) = temperature (Kelvin)

Although the inhibitory concentrations of ammonia reported in the literature are different, when converted to free ammonia, they are more consistent, indicating the main reason for the inhibition of this form. For example, for a pH 8 solution, only 4% of TAN is available as FAN at 20 °C; while at 40 °C, 13% becomes available as FAN. The discrepancy in results thus illustrates the difficulty of administering ammonia inhibition based on TAN rather than free ammonia, as total ammonia inhibitory concentrations reported in different studies are not comparable unless pH and temperature conditions are also reported [23].

High ammonia concentration is a major problem in animal manure digestion, especially poultry manure. In addition, longer manure storage results in increased ammonia content, which is associated with the need for rapid waste management or effective removal of this toxic substance [9,10,52]. The simplest method of ammonia neutralization is to dilute the raw material with water; however, this involves a decrease in biogas yield, water consumption, and a large amount of secondary waste, which increases handling costs (pumping, storage, solids/liquid separation, and transport) [28]. Therefore, it seems right to use solutions that are less invasive to the environment, such as stripping with air or water vapor [53], adsorption processes [54,55], filtration techniques, nitrification [56], precipitation [57], ion exchange [58], or anaerobic oxidation [59,60] (Figure 5). Ammonia removal also promotes the recovery of nutrients from the feedstock by capturing ammonia from the carrier gas with scrubbers or traps such as sulfuric acid, creating high-end fertilizers that can be used for agricultural purposes [61]. Due to the possibility of digesting several wastes at the same time, which heterogeneously translates into process economics, the co-digestion technique seems to be particularly promising [11,16].

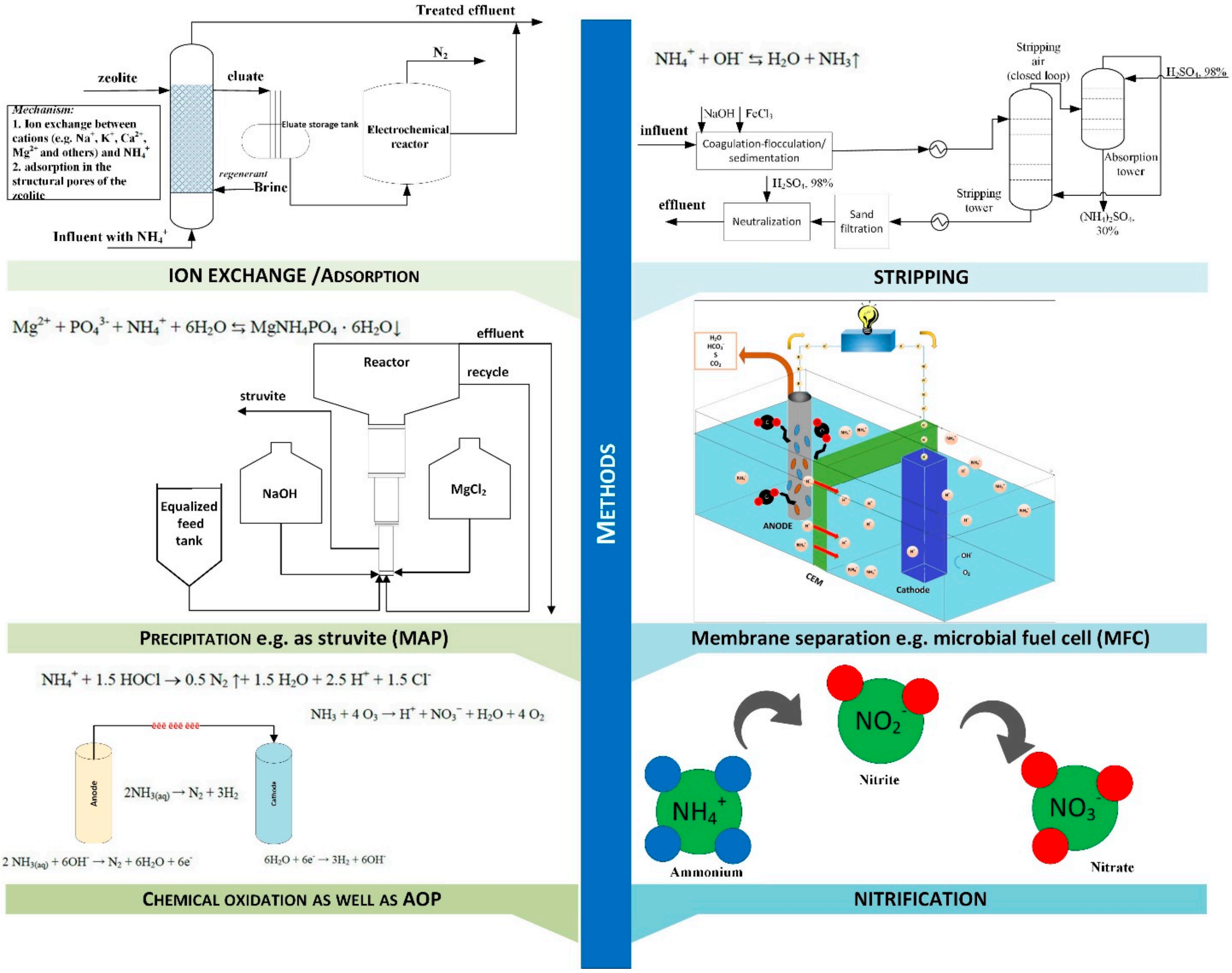

Figure 5. Methods used to remove ammonia from the substrate [53,56–59,62].

3.4. C/N Ratio

The proper ratio of elements such as C and N in the substrate is extremely important from the point of view of the anaerobic digestion process. The optimal ratio of these nutrients is given in the range of 10:1–25:1, while 100:3 is indicated as the maximum value. If the threshold of this ratio is exceeded, nitrogen will be used by organisms participating in digestion, which will translate into a decrease in methane production efficiency. In turn, the decrease in the C/N ratio results in the formation of toxic ammonia and an increase in the pH value [63]. In the case of animal manure, this ratio is usually insufficient for effective anaerobic digestion [64–66]. The appropriate C/N ratio may be the main reason for improving biogas production from this waste [65]. Increasing the amount of C can be achieved by using co-substrates with a high content of this element. There have been many publications in recent years in which the positive impact of a mixture of several wastes on the efficiency of anaerobic digestion has been presented. Wang et al. [67] also pay attention to the interactive effect between the C/N ratio and temperature on methane production efficiency.

3.5. Temperature

Rapid temperature changes cause the death of organisms involved in anaerobic digestion, which is why optimization of this parameter is extremely important in the course of the process. Depending on the psychrophilic (<25 °C), mesophilic (30–40 °C), and thermophilic

(>40 °C) types of digestion, the latter two are commonly used. The influence of temperature on process efficiency is a subject of constant research. Mesophilic conditions favor all operational activities and require less financial effort [27]. Despite the need for heating, thermophilic digestion determines the increased metabolism of organisms, and thus faster degradation of volatile solids and reduction of pathogenic microorganisms [4,68,69]. However, the issue of methane production efficiency at elevated temperatures is divergent. Böske et al. [27] indicate in their research on horse manure digestion 59.8% higher methane yields and 58.1% higher methane rates in the case of thermophilic process in relation to mesophilic conditions. On the other hand, Hansen et al. [70] noticed that methane production efficiency decreases with increasing temperature. Maranon et al. [71] also showed that the co-digestion of cattle manure, food waste, and sewage sludge causes a lower methane gain at 55 °C than at 36 °C. In turn, Mata-Alvarez et al. [40], in their paper presenting a critical review of the literature in the years 2010–2013 regarding anaerobic co-digestion, conclude that the efficiency of the process under thermophilic conditions is slightly higher. However, in fact, maintaining a relatively high temperature in reactors, especially on small farms, can be a problem due to large differences in external temperature. Therefore, the issue of this parameter should be further examined, taking into account other factors that may correlate with temperature, such as mixing intensity, reactor type, or substrate properties.

3.6. Mixing

By mixing the contents of the reactor, a homogeneous fermenting mass is obtained throughout the entire volume of the chamber. The choice of mixing intensity is closely related to the process temperature. At higher digestion temperatures, the mixing of the substrate should also be increased. For mesophilic conditions, the daily capacity of devices used for mixing should be 6–20 times higher than the capacity predicted for the volume of the chamber. In the bioreactor, the difference between the dry matter content of the substrate at different depths should not exceed 5 kg total solids/m^3 [10]. In the case of manure, especially horse manure, the mixing process is hindered by the presence of bedding materials that are used to create dry and clean spaces for animals [38]. It is also worth mentioning that mixing should be adapted to the specific type of reactor used [72,73].

3.7. Reactor Type

For most studies on anaerobic digestion, the process is carried out in a CSTR (continuous-stirred tank reactor). However, technology performance can be improved by using other bioreactor configurations [40]. Animal manure is a relatively dry waste and its digestion in a single-stage reactor can be difficult to carry out. Solids tend to float on the surface of the liquid phase, which contributes to the clogging of equipment. For this reason, works are devoted to the separation of individual stages of anaerobic degradation; the aim of which is to optimize the process depending on the substrates used. Smith and Almquist [72], in a study focused on the co-digestion of horse manure and food waste, proposed a technology based on two-stage digestion, where in the first phase the pH value is close to 4 and includes the stages of hydrolysis and acidogenesis, and in the second phase the pH is neutral during acetogenesis and methanogenesis. The main motive of the presented solution was to prevent the accumulation of fibrous biomass in the second phase reactor, which is directly responsible for the production of methane [72]. A similar concept was also presented by Zhang et al. [73] who used a three-stage digester for the co-digestion of horse manure and food waste, broken down into hydrolysis, acidogenesis, and wet methanogenesis. As a result, they obtained greater hydrolytic and acidogenic efficiency of the solid organic substance, thereby accelerating the subsequent stage responsible for the production of methane.

3.8. Hydraulic Retention Time

The presence of the substrate in the bioreactor until it is replaced with a new charge is defined as the hydraulic retention time. This parameter is particularly important at the time of start-up, where the slow increase of the substrate gives the possibility of a maximum growth rate of microorganisms [74]. Changes in hydraulic retention time (HRT) can affect the structure of the microbiome community. The imbalance between fast-growing microorganisms (hydrolytic and acid-forming bacteria) and slow-growing methanogens as a result of inappropriate HRT causes problems such as insufficient utilization of hydrolysis/acidogenesis products in subsequentdigestion stages and/or methanogen leaching effects [75]. HRT should be adapted to the type of substrate so that it allows for complete degradation, with different organic substances decomposing at different rates. Usually, the hydraulic retention time is from 20 days in the case of slurry, and up to 60 days for energy crops and hardly degradable compounds such as cellulose or lignin. This parameter depends on the temperature because organic substances decompose faster under thermophilic conditions [76]. As previously mentioned, animal manure digestion is usually carried out at an HRT equal to 20 days. However, Grosser, in her work [77] on the co-digestion of sewage sludge, grease trap sludge, and the organic fraction of municipal waste at different HRTs (12–20 days), indicates that, despite the best process efficiency at the longest HRT, carrying it out at an HRT lower than 20 days is also possible, and daily biogas production was about 46% higher compared to the period with the highest HRT.

3.9. Water Content

Water, as the compound that is the basis of the life of organisms, is important in the course of the process both from the point of view of biological activity and the structure and properties of the charge. To assess the moisture content of the substrate, its dry organic matter content is determined. Depending on the hydration of the biomass, digestion can be divided into wet, semi-dry, and dry, where for the first type the dry matter content does not exceed 15% and the second is about 20%. However, dry digestion occurs in the case of total solid (TS) values above 20%. The optimal amount of TS is considered to be 12–15% because in such conditions the substrates can be easily pumped between devices, and an efficient decomposition process takes place [63,78]. Higher moisture content also promotes the growth of methanogenic bacteria and improves the transfer between substrate molecules and organisms during the last stage of anaerobic digestion [73].

3.10. Pre-Treatment

Due to the structure and composition of animal manure, various pre-treatment techniques can improve its anaerobic digestion and affect the greater efficiency of methane production. Pre-treatment methods include mechanical and chemical techniques as well as biological techniques such as bioaugmentation [39,64]. The selection of the right technique is largely determined by capital expenditure. The year 2011 was a breakthrough in terms of the number of articles published on the preliminary treatments to which digestion substrates are subjected. The increase in publications was undoubtedly associated with research interest in the co-digestion process and the possibility of anaerobic degradation of hardly biodegradable and inaccessible substances [27,40]. Pre-treatment of such substrates is often encouraged to accelerate or increase the methane production potential due to the availability of organic matter or the removal of process inhibitors [64,79].

One of the popular methods in this area is the use of ultrasound, commonly used to break down complex polymers in the treatment of sewage sludge, which can lead to an increase in methane production by up to 34% [71]. Castrillon et al. [80] have shown that the use of this treatment in the case of cattle manure with glycerol causes an increase in the amount of biogas by 121%. However, Maranon et al. [71] noted that the better anaerobic digestion efficiency of cattle manure with food waste and sludge caused by ultrasound treatment does not compensate for the energy that it requires. Similar observations are also made by Azman et al. [81] using ultrasound to treat the manure digestate. Other effective pretreatment methods associated with large financial outlays and physical manure include aeration [10].

Thermal pre-treatment seems to be a promising solution, often integrated with other treatment methods. Research conducted by Gonzalez-Fernandez et al. [82] has shown that treating pig manure with a temperature of 170 °C for 30 min at a pressure of 7 bar increases methane production by 35%. For comparison, the same authors also pre-treated this substrate using a strong base, but the effect was an increase in methane efficiency by 13%. An interesting strategy was proposed by Rodriguez-Verde et al. [64], combining the thermal treatment of chicken manure at 90 °C with the simultaneous stripping of ammonia (Figure 6). Liquid manure, especially dust, is abundant in nitrogen, hence the removal prevents process inhibition. The solution of the researchers made it possible to reduce the content of ammonia in the substrate and increase its biodegradability.

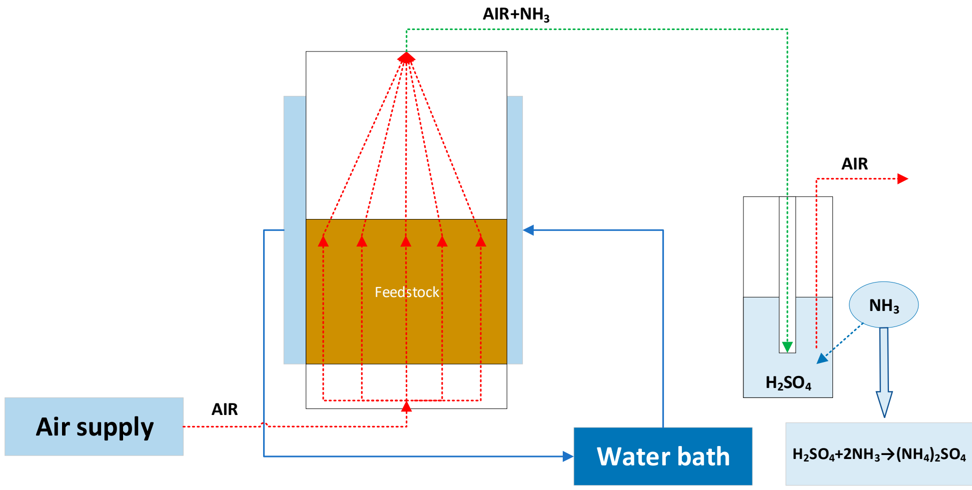

Figure 6. Scheme of thermochemical pretreatment proposed by Rodriguez-Verdee et al. [64].

It is also worth mentioning that it is important to properly secure and quickly manage the manure. The long-term storage of animal waste can significantly reduce biogas production. The use of this feedstock after two months results in an almost 6% decrease in gas production, while after four months it's 17% decrease compared to fresh material. [83].

3.11. Other Process Inhibitors

In addition to the ammonia and fatty acids described above, many other substances interfere with the anaerobic digestion process. They include, among others, heavy metals that are extremely dangerous in a mobile and digestible form for methanogenic organisms. The main factors affecting the migration of these elements to cells are pH, oxidation–reduction potential, and the sorption capacity of the substrate. To determine the mobility and bioavailability of heavy metals, speciation analysis is commonly performed, such as the BCR sequential extraction technique, which is based on the elution of elements with reagents of increasing aggressiveness [84]. Hydrogen sulfide, which is an inherent component of biogas, is another compound that can inhibit the digestion process and can induce harmful effects at 50 mg/L [50]. It is worth mentioning, however, that its presence may affect the neutralization of heavy metals in soluble form due to the formation of more soluble metal sulfides [64]. Other elements that inhibit the process also include sodium, potassium, calcium, and magnesium.

4. Importance of Anaerobic Co-Digestion in the Treatment of Animal Manure

The anaerobic degradation process has been investigated in numerous studies, and special attention is paid to the processing of organic waste while maintaining efficiency in biogas production [72]. Figure 7 shows the number of publications on the anaerobic digestion of animal manure from 2001–2023 [85]. However, the anaerobic digestion of animal manure may not be successful due to the specific properties of this material. Wet anaerobic digestion is commonly used due to the low level of formed sludge, ease of use, and greater methane production efficiency per volatile solids [78]. Animal feces contain a high content of large fibrous particles, causing problems with the clogging of devices [27]. The digestibility and efficiency of horse manure methane production are the lowest compared to other farm animals due to the presence of bedding materials (for example wood shavings) that contain hardly degradable compounds such as lignin or cellulose [73]. Cattle manure is also characterized by relatively low biodegradability [9]. In turn, poultry manure contains a higher concentration of nitrogen compared to other organic waste, which is associated with the risk of process inhibition by the release of ammonia [17,64]. Table 3 presents the basic parameters of anaerobic digestion for the most digested feces: cow, poultry, swine, and other manure.

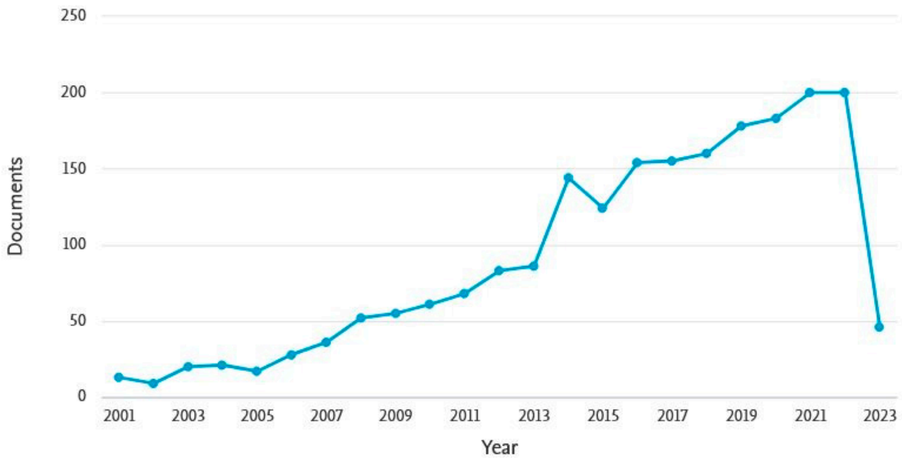

Figure 7. The number of publications on anaerobic digestion of animal manure in 2001–2023 (data status: March 2023) [85].

Due to the potential for energy recovery from liquid manure and its high buffer capacity, attempts were made to remove problems related to process inhibitors. In addition to substrate pre-treatment and stripping of ammonia, joint digestion with animal manure and other materials is a promising method. The main motive for introducing such a solution is processing several wastes at the same time and balancing the content of nutrients in the material, which translates into the optimization of the C/N ratio [52,65,86]. Moreover, this modification of the charge allows for reducing the negative impact of toxic compounds on the process and causes the succession of microbial communities and system stability [87,88]. Co-digestion is therefore defined as a combination of the decomposition of various types of substrates to obtain greater biogas efficiency [24]. Joint treatment of several wastes is also economically advantageous, as it is possible to obtain materials from one source, e.g., the household. Co-digestion is also the most cost-effective and easiest way to improve digestion efficiency for farmers [25].

When choosing suitable co-substrates, factors such as price, access, material composition, methane production efficiency, and pre-treatment and machining costs should be taken into account [9]. Defining the optimal substrate mix is based on trials, but also using modeling of the ratio of co-substrates in batch experiments can maximize methane production [64]. In the case of co-digestion, it is important to properly balance the composition of the substrates and the process parameters. Usually, a solution is observed where the proportion of one of the substrates is above 50%. In agricultural biogas plants, agricultural waste, as well as liquid and solid animal excrements, should be digested first [37]. The anaerobic decomposition of the mixture of municipal bio-waste with liquid manure can also be important in the aspect of sustainable waste management in local conditions [89]. During the processing of animal excrements, co-substrates with a high C/N ratio, low buffer capacity, and, depending on biodegradability, the ability to release large amounts of volatile fatty acids are sought [40,90]. Table 4 summarizes examples of materials characterized by different C/N ratios [37]. A particularly high carbon-to-nitrogen quotient is found in wood (700), as well as paper (170–800), scobs (200–500), and bark (100–130). Slightly less is contained in straw (80–100), leaves and weeds (90), corn cobs (40–80), and hay (40). Materials with low C/N, are usually those that contain a lot of protein, such as manure (15–18) or legumes (18–20). Below <25 also include kitchen waste, green and food waste, or other non-legumes.

Table 3. Basic technologic parameters for different animal manure digestion.

Substrate	Type of Reactor (Total Volume, L/ Working Volume, L)	Description of Process	VS Removal (%)	Biogas or Methane Production (Increase *)	Methane (%)	Ref.
		COW MANURE				
FW + CM	CSTR (140/86)	55 °C; 16 rpm; Recirculation rate: 11.40 m^3/h OLR: 1, 2, 3, 4 kg VS/(m^3d)	63.01–82.81	0.60–0.8 [1] (up to 88.6%) [2]	61.34–65.89 (up to +4.7%) [2]	[91]
CM + barley	Batch (1/0.75)	55 °C; 100 rpm; CM to barley mixing ratio equal 1:1, VS basis; trials inoculated with sewage sludge (SS) last trial co-inoculation of CRF with inoculum	NA	0.278 [1] (+18%) [3]	53–66	[92]
CM + a trace metals solution	Batch (0.120/-)	53 d; 35 °C	NA	0.148 [1] (+24%) [4]	NA	[93]
CM	Batch (0.5/0.2)	36.5 °C; I/S 0.5; manure loading a factor was 3.5 g VS/L	58.6	0.204 [2]	69.1	[94]
CM + steel slag	Batch (0.5/0.4)	36 ± 1 °C, 35 d concentrations of steel slag: 0.5, 1.0, 1.5, and 2.0 wt%	58.62 [5] (+15.5%) [6]	0.275 [1] (+153%) [6]	51.12	[95]
CM + APW	Batch (0.5/0.375)	36 ± 1 °C, APW/DM wet weight ratios: 1:0, 3:1, 1:1, 1:3, and 0:1.	55.9–59.91 (up to +7.4%) [7]	0.195 [1] (up to 23.6%) [7]	61.4–67.1 (up to +12.6%) [7]	[96]
CM + BS [8]	CSTR (20/15)	49 ± 1 °C, HRT = 20 d; 5% of shredded straw and 95% of CM of fresh matter	NA	0.213 [1] (+28.9%) [7]	NA	[97]
	CSTR (20/15)	49 ± 1 °C, HRT = 20 d; 5% of briquette straw (BS) and 95% of CM of fresh matter		0.217 [1] (+30.9%) [7]		
	CSTR (30 m^3/-)	50 °C, BS concentration—9% of fresh matter		0.351 [1] (+33.1%) [7]		
CM + ESBS-DP	Batch (-/2)	35 ± 0.5 °C ESBC-DP:CM mixture ratios were tested: 0:100, 25:75, 50:50, 75:25, and 100:0	65.3–77.5 (up to +33.2%) [7]	0.323–0.557 [1] (up to+24.6) [7]	NA	[98]
the lactating CM + FeW	Batch (0.05/-)	37 °C added to feed at 30% of the total sample VS weight, 88 d	45.45 (+22.9%) [7]	0.374 [1] (−9.4%) [7]	NA	[99]
CM from young cow + FeW			42.98 (+23.2%) [7]	0.349 [1] (+5.1%) [7]		
Dry CM + FeW			41.60 (+26.9%) [7]	0.257 [1] (−5.7%) [7]		
the lactating CM + WM		37 °C, manure with waste milk was tested at two mixing ratios, 70:30 and 30:70; 88 d	45.44–47.3 (up to +27.9%) [7]	0.413–429 [1] (up to +3.9%) [7]		
CM from young cow + WM			40.03–43.08 (up to +20.9%) [7]	0.408–0.470 (up to 41.6%) [7]		
Dry CM + WM			40.22–42.17 (up to +28.7%) [7]	0.301–0.335 [1] (up to 22.3%) [7]		
FR + CM	Pilot scale (-/850)	35 °C, 27% radish and 73% dairy manure (ww); 13% radish and 87% dairy manure (ww),	NA	0.208–0.210 [1] (up to 38.7%) [7]	NA	[100]
CM + MS	Batch (1/0.8)	35 ± 1 °C, mixing ratio of 3:1, 2:1, 1:1, 1:2 for CM/MS	NA	0.534–0.614 [1] (up to +39.8%) [7]	51.21–58.66 (up to +39.5%) [7]	[101]
Ss + CM		35 ± 1 °C, mixing ratio of 3:1, 2:1, 1:1, 1:2 for Ss/CM		0.352–0.470 [1] (up to +7.1%) [7]	48.4–58.7 (up to +39.6%) [7]	
POME + CM	SABr (5/3.5)	35 °C, 25:75, 50:50, 75:25, and 100:0 mixing ratios of POME and CM	41–63 (up to +90.9%) [7]	357–1005 [9] (up to +292%) [7]	NA	[102]
CM + ShM	CSTR (-/2.4)	HRT: 25 d; 37 ± 1 °C, 120 rpm Ratio 1:1	NA	0.179 [1] (+22.6%) [7]	61 (+8.9%)	[103]

Table 3. *Cont.*

Substrate	Type of Reactor (Total Volume, L/ Working Volume, L)	Description of Process	VS Removal (%)	Biogas or Methane Production (Increase *)	Methane (%)	Ref.
CM	Batch (2/0.25)	Mechanical Pre-treatments: shredded (SP), then mixed (MP), and finally blended (BP).	NA	0.216–0.235 [1] (up to +11.9%) [10]	NA	[104]
CM + WS	Reactor (23.6/20.9 m^3)	35 ± 1 °C, daily flow of feedstock on the level of 0.39 m^3/d ratio of 1:1 *w*/*w*; Ultrasonic pretreatment		0.460 [1] (+24.6%) [10]	53 (+1.3%) [10]	[105]
		35 ± 1 °C, daily flow of feedstock on the level of 0.39 m^3/d ratio of 1:1 *w*/*w*; hydrodynamic cavitation		0.430 [1] (+16.5%) [10]	54.1 (+3.4%) [10]	
CM + CRS + SBP	Batch (0.5/-)	Mixing ratio: 2:1:1; 39 ± 2 °C Thermal pre-treatments: at 100, 120, 150 and 180 °C with 10, 20, 30, 60, and 120 min	NA	AcD: 0.180 [11] (+11.4%) [7] (+100.6%) [10]	NA	[106]
CM + CST	Batch (2/1)	Mixing ratio 1:1 *w*/*w* 35 ± 2 °C, 60 rpm Pre-treatment: 1.5% $Ca(OH)_2$ and 120 °C	NA	0.290 [1] (+31.82%) [10]	NA	[107]
	EGSB (3.4/2)	HRT: 1–16 d, 35 ± 2 °C, OLR: 2.18–35.21 kg SCOD/(m^3d) Mixing ratio 1:1 *w*/*w* 35 ± 1 °C, Pre-treatment: 1.5% $Ca(OH)_2$ and 120 °C	85.12–96.41 [12]	0.23–0.31 [13]	48.21–69.32	
CM + tea waste	Batch (0.6/-)	Mixing ratio 1:1 *w*/*w*; 40 d, 25–35 °C; Pre-treatment: 4% NaOH g/g TS	NA	43.85 [14] (+55.9%) [10]	NA	[108]
		Mixing ratio 1:1 *w*/*w*; 40 d, 25–35 °C; Pre-treatment: microbial consortium		52.55 [14] (+86.8%) [10]		
CM:RS	Batch (0.25/-)	Mixing ratio of 1:1, based on TS mass, 35 °C Pre-treatment: limonite concentrations of 1%, 5%, and 10%	NA	1351–1462 [15] (+18.5–30.3%) [10]	NA	[109]
CM	Batch (0.5/0.4)	37 ± 1 °C, 0.18 wt% microwave pyrolytic carbon material	NA	0.380–0.502 [14] (up to +70.7%) [7]	NA	[110]
CM + acorn slag waste	Batch (0.5/0.4)	36 ± 1 °C; 3:1wet weight ratio Additive: biochar dose: 0.72, 1.08, 1.44, 1.80, and 2.16 g/L	57.4–67.75 (up to +27%) [7]	0.431–0.581 [16] (up to +42%) [7]	62.3–66.4 (up to +11%) [7]	[111]
CM	Batch (1/-)	38 °C, 30 d Additives: microscale waste iron powder or iron oxide nanoparticles	46.39–55.06 (up to +77.8%) [7]	0.67–0.222 [1] (up to +39.6%) [10]	54.33–58.94 (up to +11.6%) [7]	[112]
CM	Batch (-/0.4)	36 ± 1 °C; Additives: nano-scale tungsten (WC, W2N, and W18O49)	50.08–71.11 [5] (up to +73.9%) [7]	0.426–0.580 [16] (up to +58.5%) [7]	NA	[113]
CM + APW	Batch (0.5/0.4)	36 ± 1 °C; 35 d, ratio CM:APW 1:3 *w*/*w* Additive: Ti-sphere core-shell structured (0.03 g/L); the magnetic field	53.03–78.25 [5] (up to +73.9%) [7]	0.366–0.510 [1] (up to +65.53%) [10]	NA	[114]
CM + Cereal crops	Batch (1/0.75)	37 ± 1 °C, 100 rpm Pre-treatment: 10% *v*/*v* of *Orpinomyces* sp. (anaerobic fungus) and spent medium	NA	0.115–0.430 [1] (up to +33%) [10]	NA	[115]
CM	CSTR (3.0–3.5/-)	37 ± 1 °C, 120 rpm; HRT = 30–40 d, Pre-treatment: bioaugmentation culture containing *Bathyarchaeota*	NA	0.179 [1] (+20.1%) [10]	NA	[116]
CM	Batch	A meta-analysis AD, 160 of case studies.	NA	Mean:0.204 [1] (+38.5%) [7]	NA	[90]
CM	Continuous mixed	A meta-analysis AD, 72 of case studies.	NA	Mean:0.299 [1] (+70.9%) [7]	NA	
		POULTRY MANURE				
PM + a trace metals solution	Batch (0.120/-)	53 d; 35 °C	NA	0.407 [1] (+12%) [3]	NA	[93]
PM + B	Batch (0.5/0.2)	36.5 °C; I/S 0.5; manure loading the factor was 3.5 g VS/L	81.4	0.259 [1]	61.1	[94]

Table 3. *Cont.*

Substrate	Type of Reactor (Total Volume, L/ Working Volume, L)	Description of Process	VS Removal (%)	Biogas or Methane Production (Increase *)	Methane (%)	Ref.
PM + RS	Batch (0.120/-)	SS-AcD, 35 °C; 180 rpm, I/S: 0.5–4.0	80.92–93.25 [17]	0.123–270 [1]	NA	[117]
PM + CC			54.55–88.89 [17]	0.131–0.291 [1]		
PM + PS			56.66–75.94 [17]	0.084–0.157 [1]		
PM + SW			49.89–87.61 [17]	0.098–0.262 [1]		
PM + CH			30.67–81.03 [17]	0.116–0.155 [1]		
PM + SB			33.82–91.7 [17]	0.140–230 [1]		
PM			32.20–89.03 [17]	0.123–0.302 [1]		
PM + CST	CSTR (2.5/2)	HRT:20 d; VS ratios of CST/CM or UPCS/CM were 1:2; OLR: 2.1 g VS/(L d) Pre-treatment: Urea Pretreated CST (UPCS) Additive: 10 g/L of biochar (B)	NA	0.449 [1] (PM:CST) 0.499 [1] (PM:UPCS) 0.513 [1] (PM:CST+B) 0.530 [1] (PM:UPCS+B)	57.1 (PM:CST) 60 (PM:UPCS) 61.4 (PM:CST+B) 62.5 (PM:UPCS+B)	[79]
PM	Batch (0.5/0.4)	35 ± 1 °C; A: Manure loading (g VS/L):31.0–58.1 Additives: Biochar dosage (%): 1.8–5.2; Cellulose loading (g VS/L): 40.0–158.1	NA	0.177–0.292 [1]	NA	[118]
PM	Batch (0.5/0.4)	37 ± 2 °C; 95 rpm, 35 d Additive: pumice	66.83 [17]	8796 [9]	68.46	[119]
PM + AWS	Batch (0.5/-)	SS-AcD (TS 20%); 35 ± 2 °C; control AD of PM	NA	0.406 [1] (+195%) [7]	NA	[120]
		SS-AcD (TS 20%); 55 ± 2 °C, control AD of PM		0.323 [1] (+150%) [7]		
TPM + AWS		SS-AcD (TS 20%); 35 ± 2 °C, control AD of TPM Pre-treatment: stripping ammonia from PM		0.562 [1] (+63%) [7]		
		SS-AcD (TS 20%); 55 ± 2 °C; control AD of TPM Pre-treatment: stripping ammonia from PM (treated PM -TPM)		0.298 [1] (+70%) [7]		
PM	Batch (1/-)	37 ± 1 °C; enzymatic pretreatment (a mixture of Onozuka R-10 enzyme and Macerozyme R)	NA	0.537 [18] (+35%) [7]	NA	[121]
PM + VW	Batch (0.25/-)	SS-AcD, 37 °C, 50 d	NA	0.244 [1] (+2.8%) [7]	NA	[122]
PM	CSTR (15/12)	OLR: 1.6 and 2.5 g VS/(ld) 55 °C	42–62	0.094–0.220 [19]	56–67	[123]
		OLR: 1.6 and 2.5 g VS/(ld) 37 °C	44.5–46.1	0.245–0.252 [19]	67–68	
PM	Batch (0.5/0.4)	35 ± 1 °C Additive: biochar made up of wheat straw, discarded fruitwood, and chicken manure at temperatures of 350 °C, 450 °C, and 550 °C	NA	0.214–0.294 [1] (up to +69%) [4]	NA	[124]
PM + BPS	CSTR (3/2.5)	4:1 based on VS; OLR: 0.8–3.2 gVS/(ld)	NA	0.193 [1]	NA	[125]
PM	Batch	A meta-analysis AD, 36 of case studies.	NA	Mean:0.260 [1] (+22.4%) [7]	NA	[90]
PM	Continuous mixed	A meta-analysis AD, 20 of case studies.	NA	Mean:0.169 [1] (+71.1%) [7]	NA	
SWINE MANURE						
SM	Batch (0.25/-)	37 °C, manually mixed once a day, manure loading factors: 8, 16, 32, and 64 g VS/L, pH adjusted to 7.0	54.4 (VS/L = 8) 54.2 (VS/L = 16) 52.2 (VS/L = 32) 49.4 (VS/L = 64)	409.57 [20] (VS/L = 8) 384.66 [20] (VS/L = 16) 361.30 [20] (VS/L = 32) 318.01 [20] (VS/L = 64)	72.8–78.8	[126]
SM	Batch (0.5/0.4)	37 °C, I/S: 1:1, manually mixed once a day Additive: zeolites (natural and sodium), at rates of 0, 10, 40, 70, and 100 g/L of SM	NA	(SM + NZ 40g/L SM) (+35% biogas, and +29% methane)	NA	[127]

Table 3. *Cont.*

Substrate	Type of Reactor (Total Volume, L/ Working Volume, L)	Description of Process	VS Removal (%)	Biogas or Methane Production (Increase *)	Methane (%)	Ref.
SM	Batch (1/0.8)	Pre-treatment: use of in situ formed graphene in an electric methanogenesis system, 38 °C, 28 d, I/S: 1:5	NA	356.49 [21] (+41.49%), 222.17 [22] (+60.89%)	NA	[128]
SM + a trace metals solution	Batch assay (0.120/-)	53 d; 35 °C	NA	0.180 [1] (+22%) [3]	NA	[93]
CM + SM + a trace metals solution	Batch assay (0.120/-)	53 d; 35 °C	NA	0.511 [1] (+9.7%) [4]	NA	[93]
SM	CSTR (5.5/4)	196 d, 60 rpm, HRT: 20 d, mesophilic conditions	35.7–41.0	1.06–1.16 [23]	NA	[129]
SM + G	CSTR (5.5/4)	196 d, 60 rpm, HRT: 20 d, mesophilic conditions	74.1–77.7	5.44–5.58 [23]	NA	
SM	Batch (-/0.4)	Additive: ferrous chloride in the amount characterized by final elemental iron concentrations of 5, 10, 25 and 40 mmol/L, 37 °C, I/S: 1:3, 41 d	NA	269.1 [20] (+21.5)	NA	[130]
SM + CST	Batch (1/-)	Substrate combination ratios (SM/CST): 30:70, 50:50 and 70:30 (% *w*/*w*); Initial pH values adjusted to 6.0, 6.5, 7.0, 7.5, and 8.0 using 5 mol/L NaOH and 5 mol/L HCL; 35±1 °C, I/S: 1:2.5	7.5 (SM:CST = 30:70) 16.7 (SM:CST = 50:50) 23.8 (SM:CST = 70:30)	11.92 [20] (SM:CST = 30:70) 14.08 [20] (SM:CST = 50:50) 220 [20] (SM:CST = 70:30)	NA	[131]
Dry SM	Semi-continuous (2/1.2)	Additive: wrapped granular activated carbon: 50 g, acclimated sludge (inoculum): 1200 g, HRT: 60 d, 35 ± 1 °C	6.6	1.1–1.67 [24] (+10.6%)	58.8–73.2	[132]
SM	Batch	A meta-analysis AD, 73 of case studies	NA	Mean:0.287 [1] (+20.6%) [7]	NA	[90]
SM	Continuous mixed	A meta-analysis AD, 23 of case studies	NA	Mean:0.322 [1] (+52%) [7]	NA	
		OTHER				
HM	Batch assay (0.5/0.2)	36.5 °C; I/S 0.5; manure loading the factor was 3.5 g VS/L	52.9	0.155 [1]	70.1	[94]
HM	Batch (0.5/-)	35 °C, 35 d, HM solid ratios: 0.5, 1, 2, and 4%	80–90	339 [25]; 203 [20] (TS: 0.5%) 374 [25]; 239 [20] (TS: 1%) 370 [25]; 236 [20] (TS: 2%) 381 [25]; 247 [20] (TS: 4%)	60 (TS: 0.5%) 64 (TS: 1%) 63 (TS: 2%) 65 (TS: 4%)	[133]
HM + Ss	Batch (0.5/-)	AcD, 35 °C, 35 d, HM TS ratios: 2 and 4%, HM:Ss = 9:1	90	410 [25]; 270 [20] (TS: 2%) 425 [25]; 280 [20] (TS: 4%)	65 (TS: 2%) 66 (TS: 4%)	
HM + Ss	Continuous digester (5/-)	AcD, 35 ± 2 °C, TS ratio: 4%, HM:Ss = 9:1	>50	NA	66–68	

Table 3. *Cont.*

Substrate	Type of Reactor (Total Volume, L/ Working Volume, L)	Description of Process	VS Removal (%)	Biogas or Methane Production (Increase *)	Methane (%)	Ref.
GM	Batch assay (0.5/0.2)	36.5 °C; I/S 0.5; manure loading the factor was 3.5 g VS/L	46.4	0.159	65.8	[94]
RM	Batch (0.25/-)	37 °C, manually mixed once a day, manure loading factors: 8, 16, 32, and 64 g VS/L, pH adjusted to 7.0	49.5 (VS/L = 8) 48.9 (VS/L = 16) 47.5 (VS/L = 32) 46.2 (VS/L = 64)	323.22 [20] (VS/L = 8) 296.87 [20] (VS/L = 16) 261.46 [20] (VS/L = 32) 211.48 [20] (VS/L = 64)	68.3–76.5	[126]

Substrate: FW—food waste; CRF—cow rumen fluid; APW—aloe peel waste; WS—wheat straw; ESBS-DP—dried pellets of exhausted sugar beet cossettes; FeW—feed waste; WM—waste milk; FR—forage radish; MS—maize straw; Ss—sewage sludge; POME—palm oil mill effluent; ShM—sheep manure; CRS—corn silage; SBP—sugar beet pulp; CST—corn straw; SCOD—soluble chemical oxygen demand; RS—rice straw; CC—corn cob; PS—peanut shell; SW—sawdust; CH—coffee husks; SB—sugarcane bagasse; B—biochar; AWS—agriculture wastes; VW—vegetable waste; BPS—banana pseudo-stems; NZ—natural zeolite; G—glycerol; CM—cow manure; PM—poultry manure; SM—swine manure; HM—horse manure; GM—goat manure; RM—rabbit manure; NA—not available. Reactors: CSTR—Continuously Stirred Tank Reactors; SABr—solar-assisted bioreactor; EGSB—expanded granular sludge blanket. Process description: OLR—organic loading rate; I/S—inoculum to substrate ratio; AcD—anaerobic co-digestion; SS-AcD—solid phase anaerobic co-digestion. Other: [1]—specific methane yield, $m^3/kg\ VS_{add}$; [2]—increase in comparison to mono-digestion of FW; [3]—AcD of CM + barley; [4]—without supplementation; [5]—COD degradation rate; [6]—control check, namely cow manure and sewage sludge; [7]—increase in comparison to mono-digestion of CM or PM; [8]—shredded and briquettes; [9]—mL; [10]—untreated manure; [11]—biogas yield m^3/kg TS; [12]—SCOD removal; [13]—m^3/kg COD; [14]—the accumulation of biogas production, mL/g TS; [15]—methane production, mL; [16]—cumulative biogas yield, m^3/kg VS); [17]—COD removal; [18]—methane yield, $m^3/kg\ VS_{removed}$; [19]—methane yield, m^3/kg TS; [20]—methane yield, $mL/g\ VS_{add}$; [21]—biogas production, m^3/t dry swine manure; [22]—methane yield, m^3/t dry swine manure; [23]—biogas production, $L/g\ VS_{add}$; [24]—biogas production, L/d; [25]—biogas yield, $mL/g\ VS_{add}$; *—comparison to control reactor.

Despite the fact that the agricultural industry is the most convenient source of obtaining such materials, the need to overcome seasonality and increase the efficiency of methane production has caused great interest in other biodegradable waste [40]. In recent years, a lot of research has been done on the anaerobic co-digestion of animal manure and by-products of various industries. In addition to the aspect of improving process efficiency, an important criterion for selecting appropriate substrates is their availability and production in a given region. For example, rice straw is one of the most abundant wastes generated in Valencia, Spain. Traditional methods of processing this material, namely combustion and landfills, generate high emissions of toxic compounds into the environment and, when stored in soil, an uncontrolled digestion process. Sillvestre, Gómez, et al. [134] used 1, 2, and 5% rice straw addition (on a mass basis) to digest cattle manure, which is also widely generated in Spain [71]. As a result, the largest increase in biogas production in relation to controls (anaerobic mono digestion of cattle manure), amounting to 54%, was achieved with a 5% share of rice straw.

Other substrates used to co-digest cattle manure are food or distillery waste. Zhang, Xiao, et al. [65] showed that with a ratio of food residues to liquid manure of 2 to 41.1% methane production increased. In turn, El-Mashad and Zhang [135] determined that for a digestion time of 20 days, a mixture of 60% food residues and 40% dairy manure is recommended. Callaghan, Wase, et al. [51] also stated that fish and whole solid offal from a brewery could be successfully used for the anaerobic digestion of waste from cattle farming. An interesting experiment was carried out by Westerholm et al. [136] using the joint processing of cattle manure with whole stillage. This waste is also characterized by a low C/N ratio; however, the co-digestion of these substrates has significantly stabilized the process. However, it should be taken into account that the properties of stillage produced in different plants differ, which translates into methane production efficiency.

Table 4. C/N value for exemplary substrates [37].

High C/N Content Materials		Low C/N Content Materials	
Substrate	**C/N**	**Substrate**	**C/N**
Paper	170–800	Kitchen waste	12–20
Scobs	200–500	Green waste	10–25
Wood	700	Fresh grass	12–20
Bark	100–130	Legumes	18–20
Straw	80–100	Non–legume plants	11–12
Leaves and weeds	90	Manure	18
Maize cobs	40–80	Poultry manure	15
Hay	40	Food waste	15

Another waste added to cattle manure digestion may be crude glycerin. This compound is mainly produced in the production of biodiesel, but the market is not able to absorb a large increase in this by-product [40,80,129]. The optimal amount of glycerol as a supplement for digestion is in the range of 4–6% [9]. In their paper, Astals et al. [129] also notice the positive effect of raw glycerin on the processing of swine manure. The authors showed that the addition of this substrate increased the organic loading rate, optimized the C/N ratio, and reduced the free ammonia concentration in the feedstock.

In the case of swine manure, which is characterized by high nitrogen concentration, co-digestion can be performed, for example, with energy crop residues. Cuetos, Fernandez, et al. [24] used for this purpose maize, rapeseed, and sunflower residues. Based on the results obtained, these authors concluded that the best results were obtained with the co-substrate in the form of maize. In the case of the remaining mixtures, however, they received worse results, probably as a result of higher lignin content in rapeseed and sunflower residues. The use of more than two substrates, including other types of excreta, can also be a promising method. Liu, Tang, et al. [33] successfully co-digested swine manure with cattle manure and solid waste. Such an undertaking allows for the treatment of waste in animal husbandry areas.

A lot of the research focuses on poultry manure because it possesses the largest methane gain that can be obtained from 1 kg of dry matter compared to other manure [9]. However, as in the case of waste from pig farming, the proper digestion of this raw material disturbs the high concentration of nitrogen [51]. The suggested share of poultry manure in co-digestion with other materials is from 10–40% of the mixture [137]. As co-substrates for poultry manure, popular agricultural waste, [14,45] like corn stover [78] may be used. Bayrakdar, Molaey, et al. [16] were the first to co-digest poultry manure with used poppy straw, whose annual production in Turkey is around 20,000 tons per year. The result of the research was a methane yield of 0.36 L/g VS when the total ammonium nitrogen concentration did not exceed 4000 mg/L. Borowski et al. [138] also presented satisfactory results in a study on the anaerobic co-digestion of chicken manure and sugar beet pulp residues. By mixing these substrates in a 1:1 weight ratio, a organic loading rate with stable pH and optimal nutrient balance was achieved. A greater proportion of manure in the co-mixture caused process inhibition, mainly as a result of the toxic effect of ammonia and, to a lesser extent, volatile fatty acids. Cocoa pod husk is another waste that can be used for the anaerobic digestion of poultry manure [3]. Cocoa is intensively produced, especially in Ivory Coast and Ghana (over 50% of world production). However, the by-product is difficult to decompose due to the presence of lignin components. Dahunski et al. [3] suggest the pre-treatment of cocoa pod husks with alkaline hydrogen peroxide before co-digestion. In turn, Gelegenis et al. [139] considered whey as a material that could help in the processing of chicken droppings. Whey, produced as a result of precipitation and removal of casein from cheese, is characterized by a high content of organic matter and biodegradability. The results of these authors' research indicate good effects of the co-digestion of whey with chicken droppings; however, this finding only applies in the case of whey as a component percentage below 50% (based on VS). In the case of a 1:1

ratio of these substrates, a decrease in biogas production was observed. Carlini et al. [52], using cheese whey wastewater, obtained the correct course of co-digestion with 50% shares of whey and chicken manure. Additionally, Wang, et al. [25] also presented interesting results regarding the processing of chicken manure. The authors co-digested dairy manure (DM) and poultry manure (CM) with wheat straw, which was added to optimize the C/N ratio. They reached their maximum methane potential at DM/CM 40.3:59.7 by weight and a carbon-nitrogen ratio of 27.2:1.

However, there are still few studies on the possibilities of processing horse manure. The key factor conditioning the course of digestion of this waste is the type and amount of bedding material present in it, such as wheat straw, flax, hemp, and wood chips. For example, softwood bedding hardly decomposes and hinders the anaerobic digestion process, while straw has a higher biochemical methane potential [38,140]. Hadin and Eriksson [141] draw attention to the fact that, despite the low biodegradability of litter, it still makes a positive contribution to the energy balance. In the group of other types of manure, horse manure can have a total solid (TS) content of 20% or more and is therefore exactly suitable as a substrate for handling high-solid or solid-state anaerobic digestion, which usually requires a TS above 15% [38]. Carlos-Pinedo and Wang [38] ran simulations of several scenarios with different feedstock component combinations in a full-scale solid-state process. Their results suggested that the replacement of green waste by horse manure with wood chips as bedding material in a co-digestion mixture with organic waste gave the best improvement in terms of energy turnover.

Undoubtedly, the aspect of using sewage sludge for co-digestion with animal feces deserves special attention. Production of this waste is still increasing and, as in the case of liquid manure, its rational management is important. It is a substrate with non-specific properties, but also with high energy and fertilizing potential [86]. At present, sewage sludge digestion is a thriving process on a global scale. Sludge often contains toxic compounds, so it may be beneficial to dilute it by processing it with other materials. It would seem that, due to the relatively low C/N ratio of sewage sludge, anaerobic co-digestion with animal manure is not a good solution. However, Borowski and Wheatherley [142] demonstrated that a 30% addition of poultry manure to sewage sludge caused an increase in biogas production by 50% and higher efficiency of VS removal. In another work, Borowski et al. [143] also studied the co-digestion of sewage sludge with the manure of pigs and poultry. The experiment showed that a 30% addition of pigsty waste caused an increase in biogas production by almost 40% compared to the anaerobic digestion of sewage sludge alone. However, by supplementing the co-digestion mixture with 10% poultry manure, the efficiency of the process decreased as a result of the high concentration of ammonia. It should be mentioned that the latest EU Regulation [31] excludes the use of sewage sludge as a fertilizer. Nevertheless, research into the possibility of their treatment and a better understanding of the risk associated with their management may contribute to the development of new solutions for their fate.

5. Ecological Potential of Digestate

The product of anaerobic digestion is not only a valuable fuel in the form of biogas but also a post-digestion mass (digestate). Its composition depends primarily on the substrates used in the process. Knowledge of the individual properties of the substrate is important from the point of view of monitoring the quality of the resulting product. The introduction of some co-substrates can lead to the production of unstable substances. The use of digestate as a fertilizer or soil conditioner seems to be the most sensible development direction due to the significant amounts of organic carbon in its composition. The components of the digestion product are mainly organic and mineral compounds as well as the biomass of organisms that have not decomposed [26,65]. However, in areas of intensive animal breeding and manure production, the amount of waste generated often exceeds the plant's nutrient requirements. Therefore, a reasonable solution seems to be the separation of liquid manure into a liquid fraction that can be managed within the farm and a solid fraction that can be transported to areas poor in fertilizers [24].

Limited soil resources constitute a significant barrier to acquiring new places for plant cultivation. Mineral fertilizers, which are easily available and have good solubility in the environment, are commonly used. However, the negative effect of their application, related primarily to their high nitrogen content, has been noticed. The invasiveness of these substances towards the natural environment is often observed already at the production stage. In addition, their price is not affordable. The advantage of organic fertilizers over mineral fertilizers is not only related to their economic benefits, but also in line with the principles of the circular economy. The digestate contains basic elements and other various micro- and macro-elements necessary for plant development. For example, phosphate rock, which is the only source of P, has been declared a critical raw material by the European Union (EU) (EU Report COM/2014/0297) due to its low substitutability. The EU pays particular attention to critical raw materials within the framework of sustainable development principles. The recovery of phosphorus from phosphorus-rich wastes such as poultry manure, sewage sludge, and their incineration ashes is one of the most promising ways to improve the security of P resources [144]. Anaerobic digestion does not affect P content, meaning that the P content of the digestate is completely determined by the input streams. Similarly, the process does not change the heavy metal content. However, during digestion, dry matter is reduced, resulting in increased P and heavy metal concentrations in the digestate. Only easily degradable organic matter is decomposed, while complex substances such as lignin, remain in the digestate [145]. The high content of organic substances resistant to rapid degradation, which are found in the products of anaerobic digestion, promotes the formation of caries. The use of fresh natural fertilizers such as manure has long been widespread in various European countries. However, this is controversial, especially due to the penetration of toxic compounds, such as pathogens, antibiotics, veterinary drugs, and heavy metals [146] into the soil environment, ground, and surface water, and uncontrolled greenhouse gas emissions. Unlike raw waste, digestate is a stabilized and sanitized material, and its production has no generally negative impact on the environment [147]. Nevertheless, the digestion product still needs to be tested for the presence of antibiotics and their degradation by-products. Dosing manure on agricultural land may also contribute to the dissemination of antimicrobial resistance in the environment through bacterial mechanisms such as transformation, conjugation, or transduction [148,149]. However, the literature suggests that the anaerobic digestion process eliminates, or reduces, the presence of antibiotics and resistance genes in manure [8]. Another contaminant present in manure that has attracted particular attention in recent years is microplastics. Wu et al. [150] demonstrated that the direct application of pig and poultry manure may be a new pathway for this substance in agricultural soils. There is growing evidence that microplastics have a negative impact on the microbial community, as carriers of mobile genetic elements and pathogenic microorganisms promote the persistence of antibiotic-resistant genes. Therefore, greater monitoring of this xenobiotic in digestion-processed manure is suggested.

The high quality of digestate obtained has been confirmed in research by, among others, Recebli et al. [151] using as a substrate a mixture of bovine and chicken manure, or Bohdziewicz et al. [89] in the digestion of swine manure and municipal bio-waste.

The digestate may also be an alternative source of water. For instance, Gao and Li [152] used anaerobic digestate effluent collected from a biogas plant as a source of fresh water and nutrients during bioethanol production. In comparison to the production using fresh water, a higher fermentation yield and ethanol concentration in the product was achieved. Depending on the biogas plant technology, the weight of the digestion product may be less if some of the liquid in the form of process water is recycled to the bioreactors. Usually, however, legal and logistical problems arise in managing such a large amount of digestate. High hydration of the mass also affects its transport costs. These limitations can be overcome by drying and concentrating the product or by separating it into solid and liquid fractions using centrifuges, screw presses, or sieves. Isolation of a solid fraction can also be achieved using processes such as coagulation, flocculation, or flotation. The dehydrated mass can be directly introduced into the soil or subjected to other treatments, e.g., composting or pelleting [153]. Figure 8 shows the main options for the management of digestate.

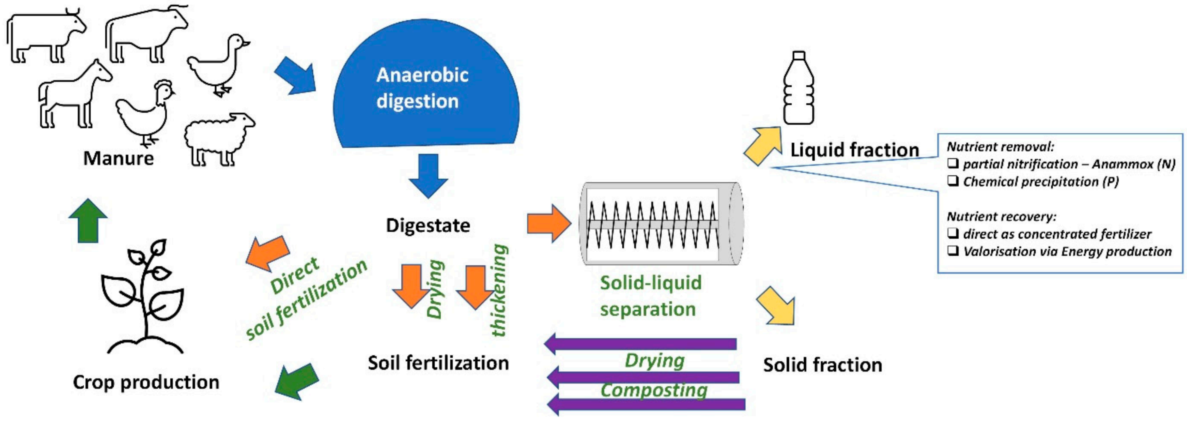

Figure 8. Digestate management methods [153].

6. Conclusions

In connection with the progressive legal restrictions on waste and energy management, it is necessary to implement optimal techniques enabling sustainable development of the agricultural sector. Anaerobic digestion is an attractive solution for processing many raw materials, including animal manure. However, the specific properties of this waste which may disturb the process should be taken into account. The main problem that accompanies the anaerobic digestion of animal excrements is the low C/N ratio and the inhibitory effect of ammonia, which translates into low efficiency of biogas production. The solution in this case may be the adequate pre-treatment of the substrate, but above all its co-digestion with other materials rich in organic carbon. Manure, due to its high buffer capacity, can be successfully decomposed along with raw materials of opposite properties, including onerous waste, such as raw glycerin, stillage, or cheese waste.

The greatest energy potential among all manure is found in poultry manure, which is why a lot of research focuses on it. However, it has a low C/N ratio, which leads to various modifications of the classic anaerobic digestion of this waste. In turn, horse manure seems to be the most difficult to digest because of the bedding materials that accompany it, which is associated with a limited number of publications in this area. Nevertheless, the prospects of conducting the process in different conditions and configurations of bioreactors leave room for further consideration.

Special attention in future research should be paid to the economics of the process based on local conditions and availability of raw materials, as well as a full physicochemical analysis of the substrates used and their biodegradability. Both empirical and modeling methods allow the selection of appropriate process parameters, reactors, and the proportion of individual materials in the feedstock. Due to the progressive regulatory restrictions on the stabilization of biowaste and the possibility of its reuse, it is necessary to characterize the products of the process and assess their further fate based on the available legislation, while keeping in mind other toxic substances such as antibiotics or microplastics, the presence of which in the environment is still subject to research and attempts to establish permissible limit concentrations in the environment.

Among the available methods of managing animal manure, the choice of its anaerobic digestion with other substrates is argued not only by its effectiveness in producing alternative energy but also by obtaining high-quality fertilizer and the possibility of recovering the water and valuable elements, which are part of a sustainable circular economy.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/en16093885/s1, Table S1: Livestock population in Europe in 2016. Table S2: Total livestock unit (heads) in Europe in 2016.

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