COASTAL BIOGAS

Cluster On Anaerobic digestion environmental Services and nuTrients removAL

A Report on Energy Recovery from Anaerobic Digestion

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Partners

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Preface

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Cover photo

Seaweed at Solrød beach September 2020, Photo by Nanna Skov Larsen, Roskilde University

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5. University of Rostock (UROS), Germany – project partner
6. Lithuanian Energy Institute (LEI), Lithuania – project partner

Table of Contents

List of figures

List of tables

List of abbreviations

BMP	Bio-methane potential
CH_4	Methane
CSW	Cast seaweed
DM	Dry matter
DW	Dry Weight
GUT	Gdansk University of Technology – project partner
HELCOM	The Baltic Marine Environment Protection Commission – also known as the Helsinki Commission
LEI	Lithuanian Energy Institute – project partner
Man	Manure
min	Minutes
mL	Millilitre
N	Nitrogen
NmL CH_4/g^{-1} VS	Accumulated methane over time
pH	Measure of how acidic/basic water is
P	Phosphorus
RUC	Roskilde University – project partner
SBP	Sugar beet pulp
t	Tonne
TN	Total Nitrogen
TP	Total Phosphorus
TS	Total Solids
VS	Volatile Solids
WFD	Water Framework Directive
WW	Wet Weight

Summary

The report focuses on the energy recovery from anaerobic digestion when adding seaweed to the biogas production. The intention of the study is to explore the possibilities for utilising seaweed as a feedstock for biogas. The study will explore the circular process of nutrients, which are recovered when the cast seaweed is collected and the digestate is used as fertiliser on farmland. The report has a special focus on investigating and documenting the energy recovery potential from anaerobic digestion.

Pre-treatment of seaweed and sand content: The pre-treatment of cast seaweed is necessary before using the seaweed as co-substrate in the production of biogas. It is mainly necessary because of the high sand content. Pre-treatment methods have been tested both during the collection of seaweed at the beach and at the biogas plant. The pre-treatments have shown a significant decrease in the sand content in most species of seaweed. Furthermore, pre-treatment of cast seaweed can greatly increase the methane potential. However, the increase depends on the pre-treatment method(-s) as well as the seaweed species.

Bio Methane Potential (BMP)-measurements: BMP-measurements have shown that the seaweed should be collected as soon as possible after it is washed ashore or while still in the water. Testing has shown that the pre-treatments have an effect on the methane yield. Some pre-treatment methods are showing an increase in the methane yield, but particular washing with heated water is showing a small decrease in the methane yield, compared to seaweed which has not been pre-treated.

Nutrients in seaweed: Seaweeds contain nutrients like nitrogen and phosphorus, which can be harmful to the marine environment and possibly end with eutrophication. To mitigate eutrophication, the nutrient levels can be reduced by collecting the seaweed. Studies are showing that to retrieve a nutrient level as high as possible, the seaweed needs to be collected while still in the water or soon after reaching the beach. If the seaweed is not collected in time, after reaching the beach, the nutrients are released and will most likely end up back in the sea.

Regulations: The EU Water Framework Directive (WFD) and the Baltic Sea Action Plan set by HELCOM aim to achieve good or high water quality in the Baltic Sea. The objectives set by the regulations aim to reach the goal towards a Baltic Sea unaffected by eutrophication. To reach the objectives, the focus needs to be at reducing the input of nutrients from inland rivers, agriculture and industries. Further actions are needed from the surrounding countries, which contribute to the overflow of nutrients into the Baltic Sea.

Reduction of nutrients at sea: The reduction of nutrients by collecting seaweed has a positive effect on the coastal and marine environment. By collecting seaweed, nutrients are reduced, which means seaweed collection could be an important tool for reducing the levels of nutrients at sea.

Overall conclusion: The removal of nutrients has the positive effect of a cleaner aquatic environment. The nutrients can be recycled via a biogas plant, and afterwards be used as fertiliser on nearby farmland.

1. Introduction to energy and nutrient recovery by utilising cast seaweed as feedstock in the biogas production.

The report focuses on the energy recovery from anaerobic digestion when adding seaweed to the biogas production. The intention of the study is to explore the possibilities for utilising seaweed as a feedstock for biogas. The study will explore the circular process of nutrients, which are recovered when the cast seaweed is collected and the digestate is used as fertiliser on farmland. The report has a special focus on investigating and documenting the energy recovery potential from anaerobic digestion.

The study firstly looks at previous and new seaweed test results. The tests include various pre-treatment methods, potential methane yield from different experiments based on different types of seaweed, collection location and pre-treatment methods, the levels and variations of sand content in collected seaweed and an assessment of an optimal raw material composition in a biogas plant using seaweed.

When collecting cast seaweed, pre-treatment is a necessary process for separating sand from the seaweed. The pre-treatment method has to be effective, in terms of reducing the sand content, but also has to be cost effective and should not have a negative impact on the cast seaweed (loss of nutrients and declining methane yield). Furthermore, the pre-treatment can greatly increase the methane yield.

Secondly, the study takes a look at the levels of nutrients and heavy metals, which occur in seaweed. Seaweed can have a high content of heavy metals depending on when and where it is collected, which both can have an impact on the possibilities of using the biomass at the biogas plant, as well as using it as fertiliser.

Lastly, the study looks at the circular use of seaweed, including nitrogen recycling, eutrophication, and the removal of nutrients in different coastal areas, regulations for removing cast seaweed at coastal areas and the reduction of nutrients at sea and at farmland, which includes an estimate of the potentials for the South Baltic Area (SBA).

Collection and the removal of cast seaweed can help to counteract eutrophication and improve the coastal water quality. The effects of eutrophication have caused that the natural balance of the Baltic Sea has been seriously disrupted by the excessive nutrient input, which originate from diffuse sources like over-fertilised farmland and air pollution [1]. The collection and utilisation of cast seaweed in the biogas production make it possible to contribute to the improvement of the marine environment. The utilisation of Seaweed in anaerobic digestion can replace artificial fertilisers and contribute to the production of renewable energy.

The purpose of this report is to provide an overview of different pre-treatments and their results in relation to sand content, the methane potential for different kinds of seaweed species, the advantages of co-digestion, the circular use of seaweed – the recycling of nutrients and finally the regulations which are related to the objectives of the Baltic Sea set by HELCOM and the Water Framework Directive.

2. Seaweed test results of anaerobic digestion

2.1. BMP-measurements from different experiments

The methane potential for cast seaweed varies depending on the species of seaweed, the collection location, ratio of co-digestion and for how long it has been lying on the beach.

Former experiments at RUC have shown a decrease in the bio-methane potential for seaweed when left on the beach for a longer period. The decreasing methane potential is caused by the degradation process [9]. When the seaweed degrades, it causes GHG emissions. Because of this, the seaweed should be collected as soon as possible when it reaches the coast, either when it is still wet (collected in the water) or damp (from piles on the beach).

Former studies have included macro algae species native to Scandinavian waters, where the methane potentials were measured to be 350-480 mL CH_4/g^{-1} VS [10]. Another study, which measured the methane yield of the species *Macro pyearifera* (Giant kelp) showed the methane yield to be 290-350 mL CH_4/g^{-1} VS [3]. In a recent study, which included unspecified seaweed collected from the coast in southern Sweden, the methane potential was measured to be 120 mL CH_4/g^{-1} VS [11]. For the pre-feasibility study at Solrød biogas plant the methane potential for seaweed collected at winter was 118 mL CH_4/g^{-1} VS [3].

2.1.1. BMP-measurements conducted at Roskilde University

A BMP experiment from RUC tested seaweed from two collection locations – one collected from piles on the beach and the other from the water edge, as well as the significance of pre-treatment when examining potential methane yield. The seaweed was collected at the start of September 2020 at Solrød beach, where the damp seaweed from piles on the beach (see Figure 1) was collected by hand, and the wet seaweed was collected by sieve in the water (up to 1 meter from the shore) (see Figure 2).

Figure 1: Seaweed in piles on Solrød beach [photo by RUC]

Figure 2: Seaweed in the water edge [photo by RUC]

Both types of seaweeds were afterwards pre-treated and mixed with manure. The pre-treatment consisted of heating the seaweed up from 23°C to 54°C and then blending the seaweed for 15 s (at level 4 on the HotmixPRO GASTRO). Hereafter, the seaweeds were transferred to a sieve where it was stirred for 60 s and lastly put aside for dewatering.

The BMP experiment was set up with the collected seaweed, both untreated and pre-treated, mixed with inoculum in a 1:3 ratio (by volume) collected at Solrød Biogas plant. The test was going for 36 days (see Figure 3). As shown in Figure 3 below, the BMP experiment shows a higher methane potential in the damp and wet seaweed, compared to the pre-treated seaweed. It shows that there is a decrease in the methane potential for the damp seaweed collected from piles on the beach after it has been pre-treated.

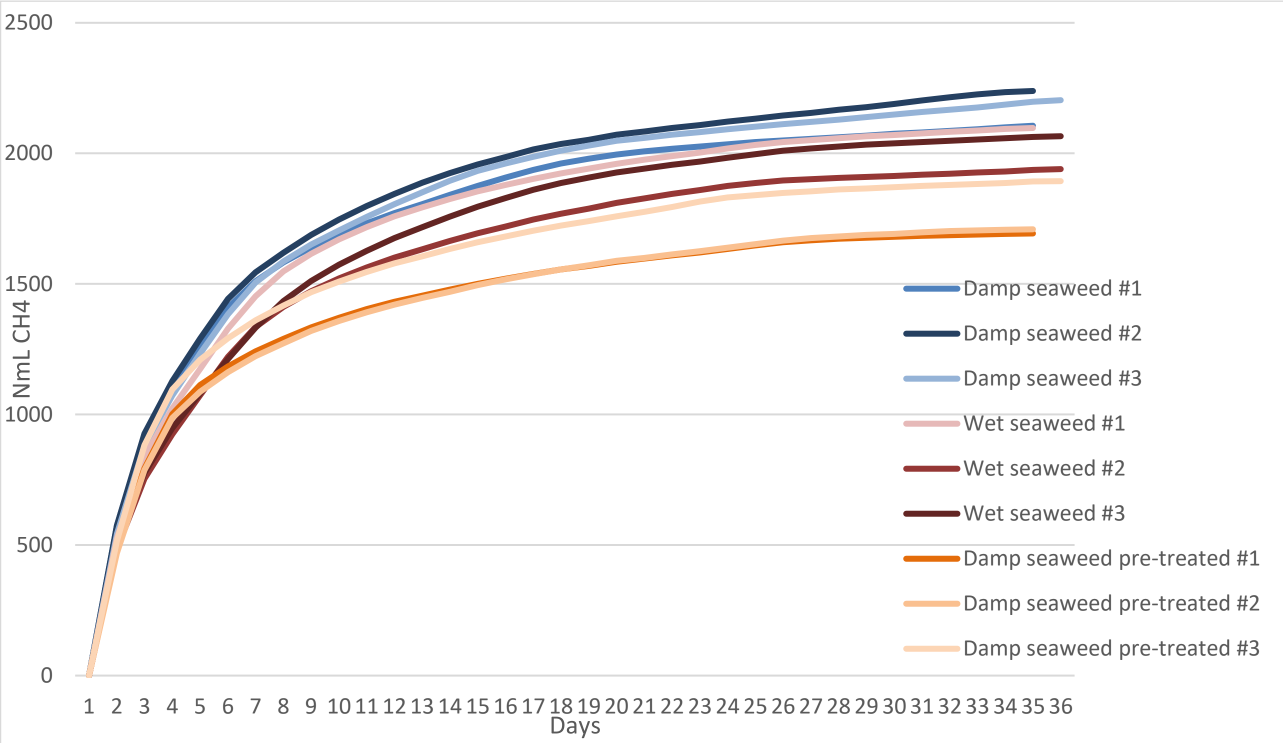

Figure 3: The bio-methane potential for damp, wet and pre-treated seaweed (RUC).

The BMP results for the damp and wet seaweed, which has not been pre-treated, are highly alike. This can be caused due to the circumstance that the damp seaweed, which was collected from piles, had not yet started degradation. The degradation, which is determined by conditions on the beach [12], can in this case be the reason of the high methane potential in the damp seaweed. Furthermore, it cannot be determined when the seaweed has been washed up on the beach, and therefore conclude how fresh it is, compared to the seaweed collected in water. Even though the pre-treatment showed a decrease in the methane potential, the study showed that the damp seaweed had a higher sand content than the wet seaweed, which means that a pre-treatment to decrease the sand content is necessary.

Another experiment from RUC has shown that the BMP depends on the type of seaweed species. The study included the seaweed species Eelgrass (*Zostera marina*), Toothed wrack (*Focus serratus*), Bladder wrack (*Fucua vesiculosus*) Dead man's rope/Sea lace (*Chorda filum*) and Sea felt (*Pylaiella littoralis*). The results are shown in Figure 4 below.

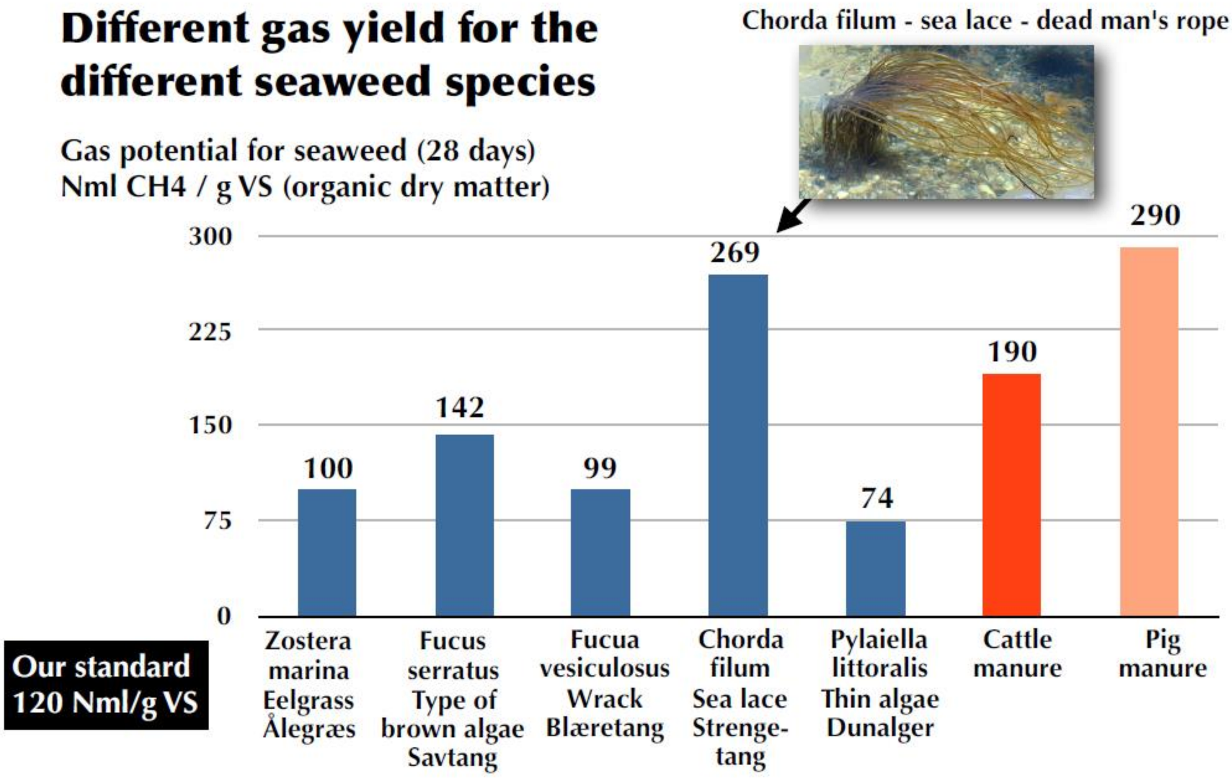

Figure 4: Gas yield for different seaweed species compared to cattle and pig manure (RUC).

The study showed a higher methane potential for the species Dead man's rope (269 NmL CH_4/g VS) and Toothed wrack (142 NmL CH_4/g VS) and the lowest methane potential for Sea felt and Eelgrass (74 & 100 NmL CH_4/g VS). The standard methane potential for seaweed is calculated to be 120 NmL CH_4/g VS. This calculation is based on the assumption that the collected cast seaweed will be a mix of different species and not exclusively one type. As Figure 4 is showing, the methane potential for seaweed is not as high as the methane levels found in cattle and pig manure.

2.1.2. BMP-experiments conducted by GUT

GUT conducted BMP-measurements of the seaweed species *Enteromorpha Compressa, Enteromorpha Plumosa, Potamogeton Pectinatus* and *Zostera Marina*. The seaweed was collected from water, in the Puck Bay near Rzucewo, Poland, and frozen right after at -18°C. The seaweed was mixed with cattle slurry in a 1:3 TS/TS ratio. The prepared feedstock was moved and weighted into a sealed glass bottle where it was kept at 36°C for 30 days.

After reaching certain pressure, biogas samples were collected into Tedlar bags from which they were further analyzed with a gas analyzer. The results are presented in Figure 5 below.

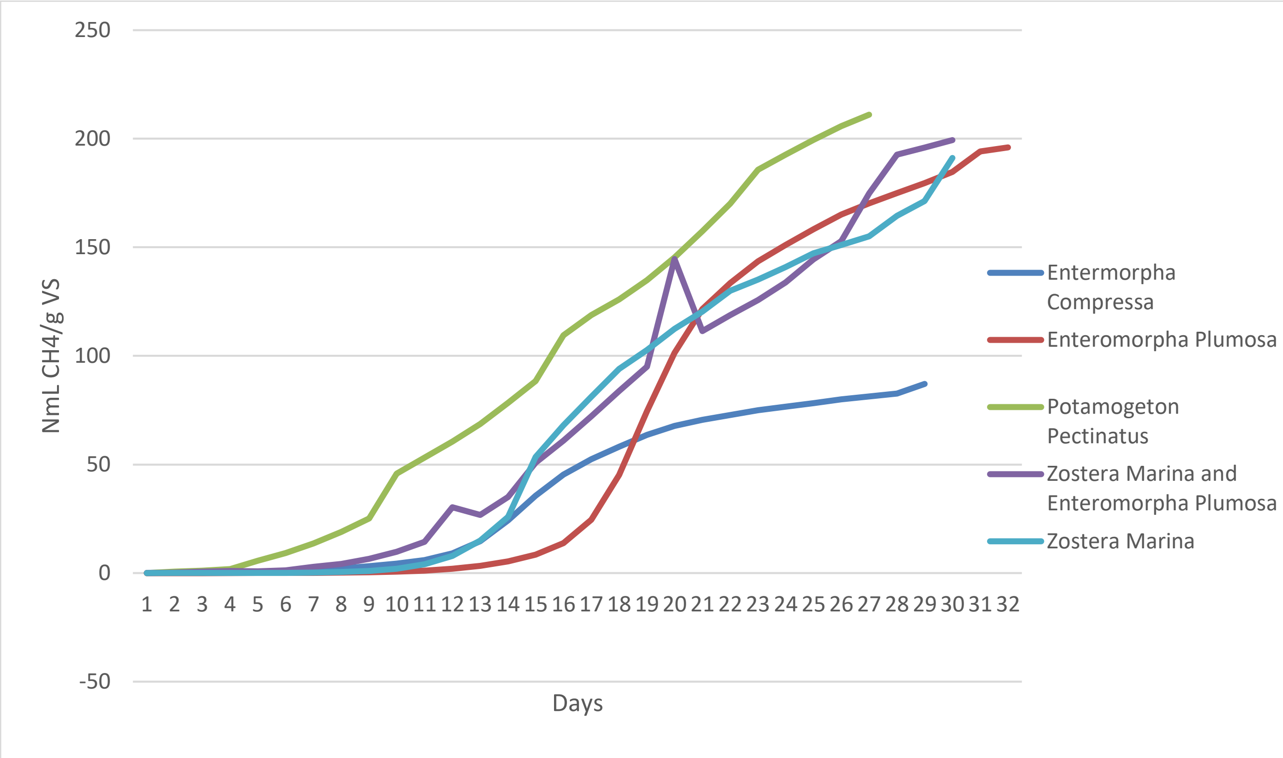

Figure 5: Methane yield in different seaweed species conducted by GUT (GUT).

As seen in Figure 5, the seaweed species *Potamogeton Pectinatus* showed the highest levels of methane yield and the species *Enteromorpha Compressa* showed the lowest. The mixture of *Zostera Marina* and *Enteromorpha Plumosa* showed an increase in methane yield, when compared to the experiments conducted on the species alone.

2.2. Pre-treatment of seaweed

Pre-treatment of cast seaweed is essential for the anaerobic digestion process and efficient operation of the biogas plant. Furthermore, the pre-treatment process can increase the quality of the methane yield as well as decrease the sand content [2]. This section will focus on different methods and experiences of pre-treatment to reduce the sand content and for increasing the methane yield.

A way of pre-treating seaweed is to wash the seaweed while collecting either on shore or in the sea. In Solrød, the seaweed, which visibly contains a high amount of sand (more than 50% of sand), is dumped in the water [3]. The seaweed is then rinsed and flows back towards the beach, where it is possible to collect it again. Furthermore, the seaweed is pre-treated again at the biogas plant, where it is put into a receiving tank and is then stirred to separate the sand from the seaweed.

An analysis at RUC has shown that it is possible to achieve a significant reduction of the sand content, by washing the cast seaweed with tap water at 52°C or above (see chapter 2.4 Sand content). The 52°C has been chosen due to the process temperature at Solrød Biogas plant.

A laboratory-based washing experiment was conducted by GUT and LEI, where two different methods were tested to reduce the sand content in cast seaweed collected from different places on the beach [4]. One of the methods consisted of mixing the sample with tap water in a beaker with a volume of 1 dm^3. The beaker was then set aside until the particles had completely sedimented. Next, the biomass was decanted and filtered on a paper filter. This process was repeated twice. The experiment showed a high decrease in sand content from the seaweed collected from shallow water. Furthermore, the experiment showed that it was easier to remove the sand if the algae were fresh, and much more difficult to wash off the sand if it had been lying on the beach for a longer period.

In Trelleborg Municipality, Smyge Pilot Biogas Plant tested a pre-treatment method, which consisted of a large mobile drum sieve, with the intention to remove or minimize the sand content [5]. Sieves with different mesh sizes were tested, and it was found that the optimal size dimension was 20 mm. In the test, the seaweed was treated twice through the sieve, but it was not possible to remove all of the sand. It was estimated that about 90% of the sand was reduced from the seaweed with the sieve method.

Some disadvantages with this method were found every time the equipment needed to be moved to another place. It required lying out iron plates on the beach, which ensured that the equipment did not damage or get stuck in the sand.

Methods of pre-treatment can be used to reduce the sand content as well as to enhance the methane yield in the cast seaweed. By pre-treating the seaweed, the organic matter becomes more accessible to the microorganisms by breaking down the complex biopolymers, enhancing the bio-digestibility of the algal biomass through accessibility of microbial enzymes, and disrupting cell walls by bringing out the chemical substances from polymers into more available compounds, to ultimately improve fermentation and the biofuel yield [6]. There are different methods to increase the biodegradability of seaweed: mechanical (cutting, drying), thermal (heating), alkaline (NaOH), acid (HCl) or enzymatic hydrolysis (cellulose or hemicellulose). Furthermore, a combination of the methods could significantly increase the methane production [6].

Mechanical pre-treatment can greatly enhance the accessibility for the microorganisms to the surface of biomass. The method involves the use of blades, knives and hammers, to chip, mill and shred the biomass into small particles prior to anaerobic digestion. The pre-treatment increases the surface to volume ratio and helps to improve the hydrolysis of complex carbohydrates to sugar, by making it more available for microorganisms. Ball milling of the biomass is the most commonly used pre-treatment process [7], but studies have shown that the method with some seaweed species resulted in lowered methane yield compared to the untreated seaweed [2].

Another method, which involves beating carried out by a modified Hollander beater, which is normally used in the paper industry, resulted in a 45% increase in methane yield compared to non-treated seaweed [8].

Experiments at GUT have tested mechanical, thermal and acidic pre-treatment methods, as well as combinations of some of the methods. Their tests showed a great increase in the average biogas production at thermal hydrolysis. The least effective pre-treatment method, when compared to untreated seaweed, was observed in mechanical disintegration pre-treatment. However, the average biogas production increased when combining mechanical and acidic pre-treatment methods (see chapter 2.3 BMP-measurements of cast seaweed after different pre-treatment methods).

2.2.1. BMP- measurements of thermally pre-treated seaweeds

Experiments conducted by GUT, tested the BMP of a mixture of different seaweed species, after thermal, mechanical and acidic pre-treatment methods had been applied. All the tests included cast seaweed collected from Gdansk Beach, which was mixed with cattle slurry at a 1:3 ratio by volume. The prepared feedstock was moved and weighted and was put into a sealed glass bottle where it was kept at a temperature of 36°C for 30 days. After reaching a certain pressure, biogas samples were collected into Tedlar bags, where it was further analyzed with a gas analyzer.

At the beginning, the collected seaweed was first thermally treated at elevated pressure. Afterwards the diluted seaweed was moved to a hermetic high pressure laboratory heater and heated in a set temperature, 160 °C, for 30 and 120 min respectively. After cooling, it was mixed with cattle slurry in a 1:3 TS ratio, with total solids of about 7.80% and 7.82% respectively.

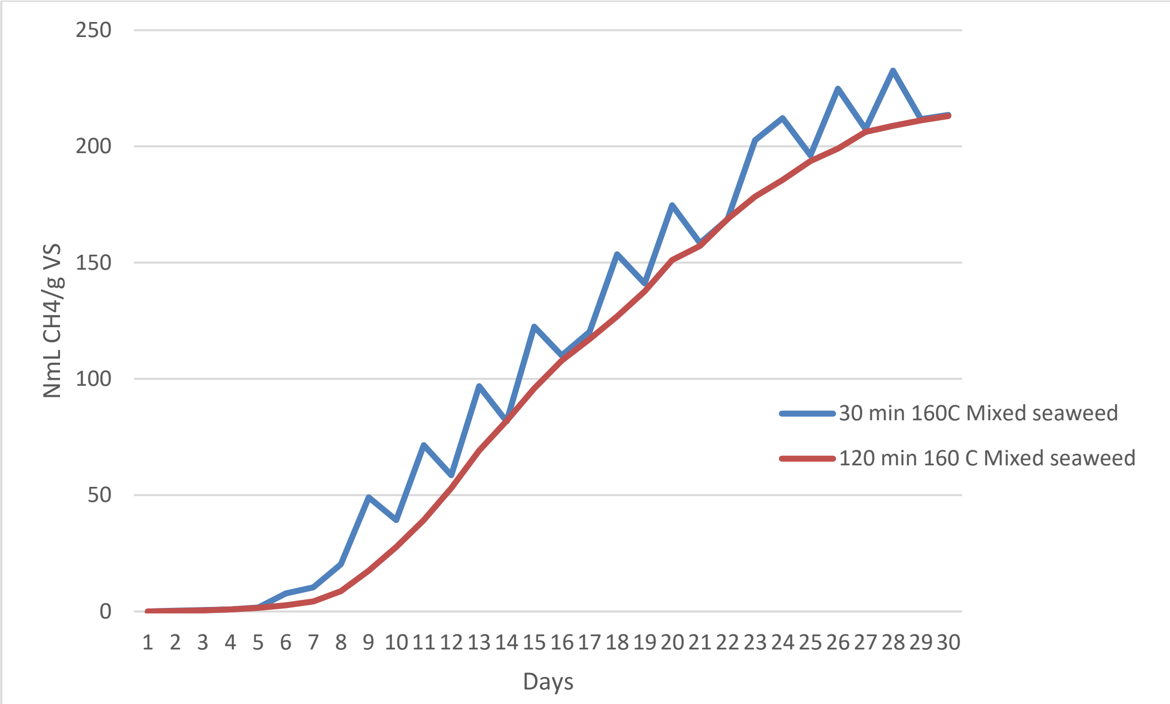

Figure 6: Methane yield after thermal pre-treatment, conducted by GUT.

Other experiments at GUT tested thermal pre-treatments, where the seaweed was heated at 160°C and 95°C respectively for 30, 120, 60 min and 24 hours. The test showed that in lower temperature conditions (95°C), a longer heating time increases the methane yield. In higher temperature (160°C) biogas and bio-methane yields result in higher values than at a lower temperature (95°C). However, a longer heating time (120 min) at 165°C shows lower yields than shorter time (30 min) (see Figure 7). This might occur because the high-pressure laboratory heater, used for this pre-treatment method, had a tendency to locally overheat the treated biomass, resulting in partial carbonisation.

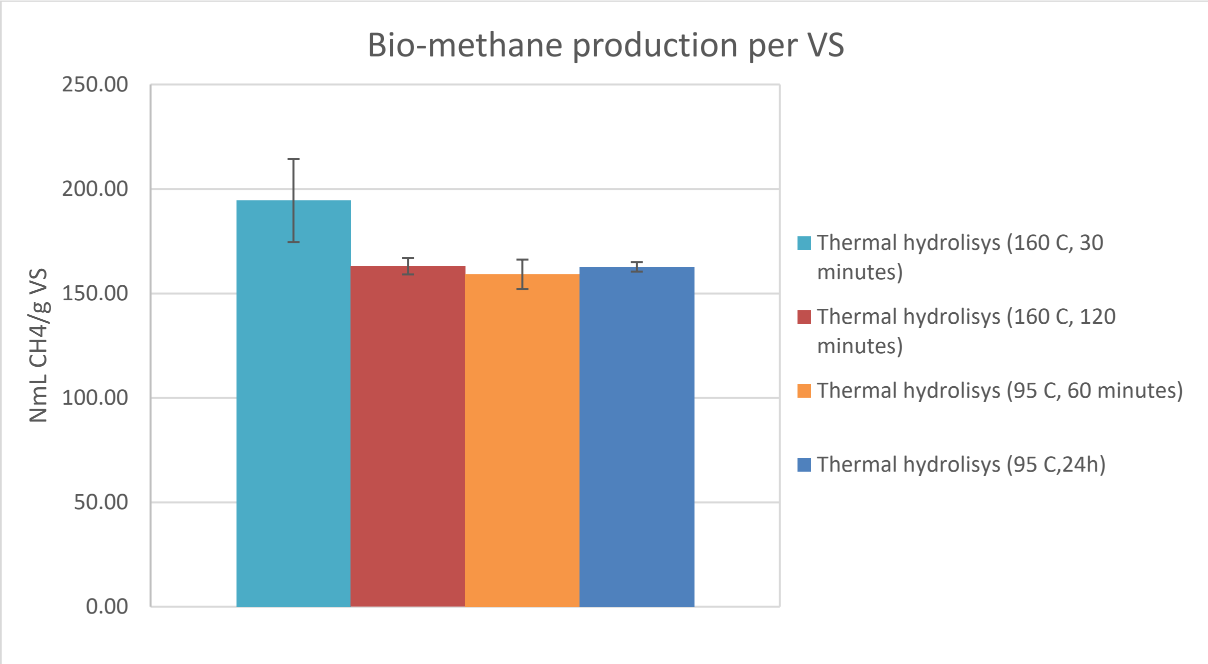

Figure 7: Average bio-methane yield per VS depending on the thermal pre-treatment (GUT).

2.2.2. BMP-measurements of mechanically pre-treated seaweeds

The seaweed (mixture of different species) was collected at the municipal beach in Brzezno, Gdansk. The seaweed was added to a laboratory grinder (power 1,200 W, rotary speed 24,000 min^{-1}, screen mash number 20 – 200 mesh) and treated respectively for 15, 60, 120 and 180 s. After disintegration, the seaweed was moved and mixed with cattle slurry in 1:3 TS ratio with a final total solids content of 6.53%, 6.50%, 6.40% and 6.97% respectively.

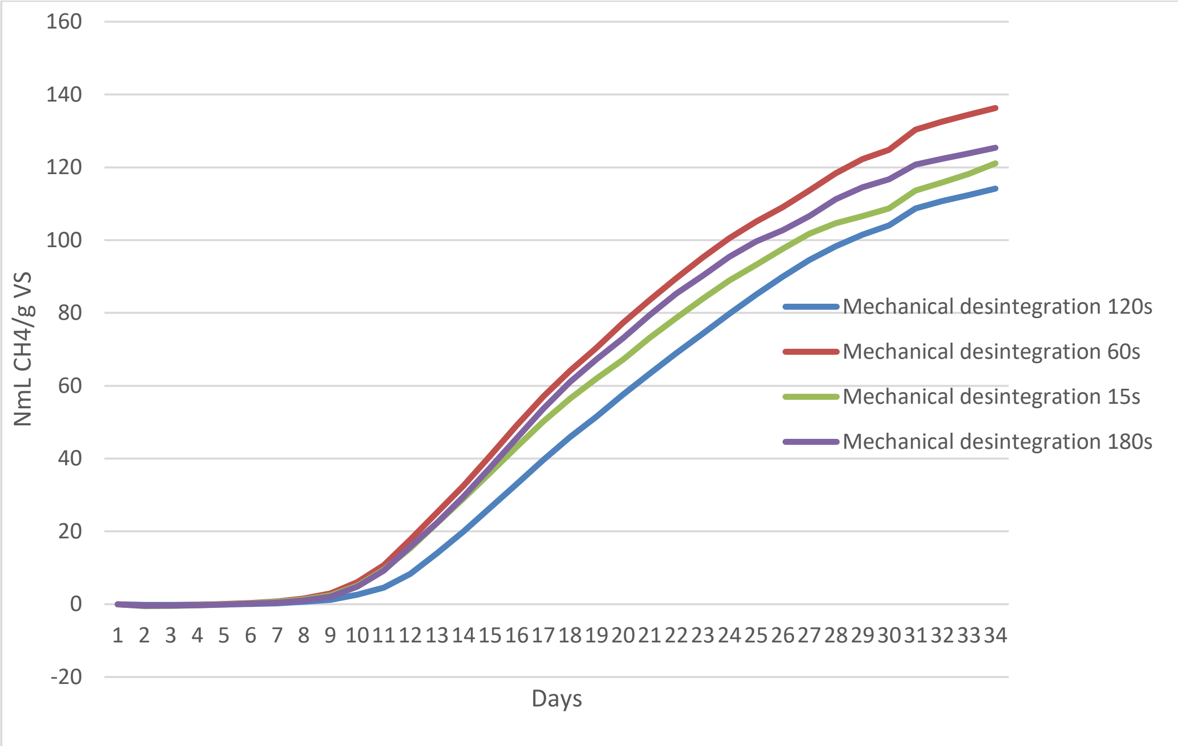

Figure 8: Methane yield for different mechanical pre-treatments, conducted by GUT.

The results of applying the mechanically pre-treatment methods for four different time spans showed the highest average biogas production at 60 s of pre-treatment time and the lowest average biogas production at 120 s.

Other experiments at GUT tested the biogas and bio-methane yields after mechanical disintegration of the seaweed for respectively 15, 60, 120 and 180 s.

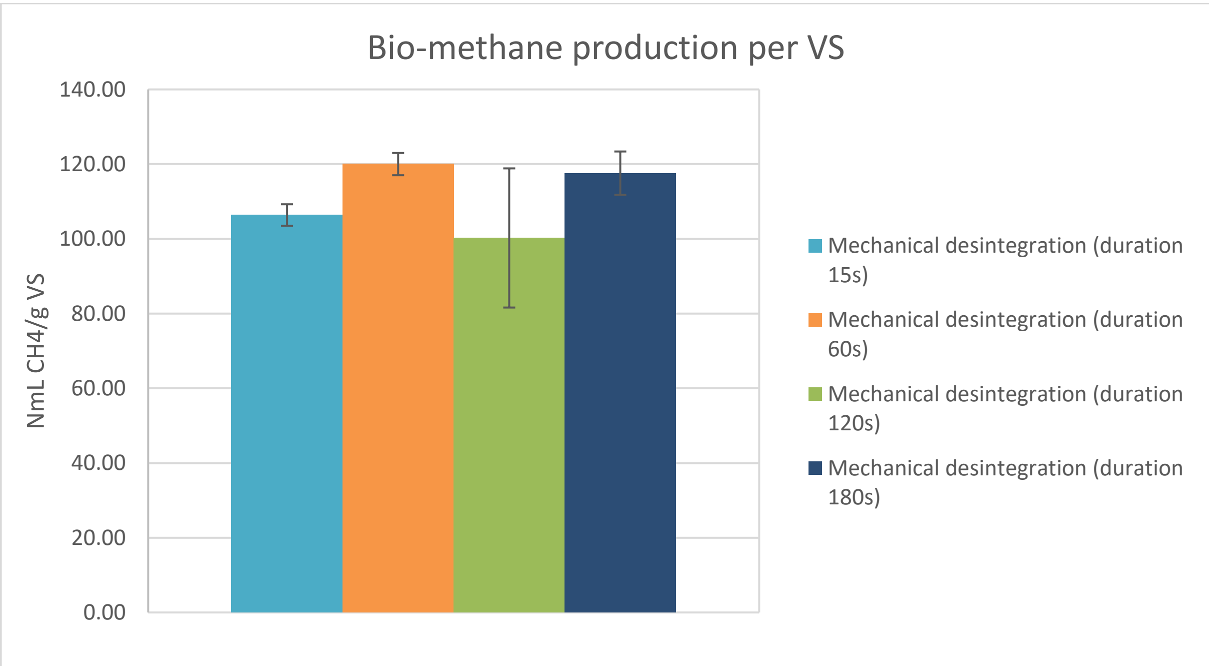

Figure 9: Average bio-methane yield per VS depending on the mechanical pre-treatment (GUT).

2.2.3. BMP-measurements of acidic hydrolysis pre-treatment of seaweeds

The seaweed collected at the municipal beach in Brzezno, Gdansk, was diluted after collection. Further, 2 M sulphuric acid was added to the diluted seaweed until pH 2 was reached. The hydrolysis was performed for 1, 6.5 and 25 hours respectively. Afterwards, sodium carbonite was added until the solution was fully neutralized to pH 7 level. After hydrolysis, the seaweed was moved and mixed with cattle slurry in 1:3 TS ratio with a final total solids content of 6.68%, 7.04% and 6.87% respectively.

Other experiments at GUT assessed the biogas and bio-methane yields after acidic pre-treatment methods had been applied, (chemical sulphuric acids (pH 2) for 1, 6.5 and 25 hours). It can be observed in Figure 10 that both 1 hour and 6.5 hour resulted in similar biogas and bio-methane yields. However longer period of time, e.g. 25 hours, resulted in slightly lower yield which might occur due to the formation of inhibitory by-products.

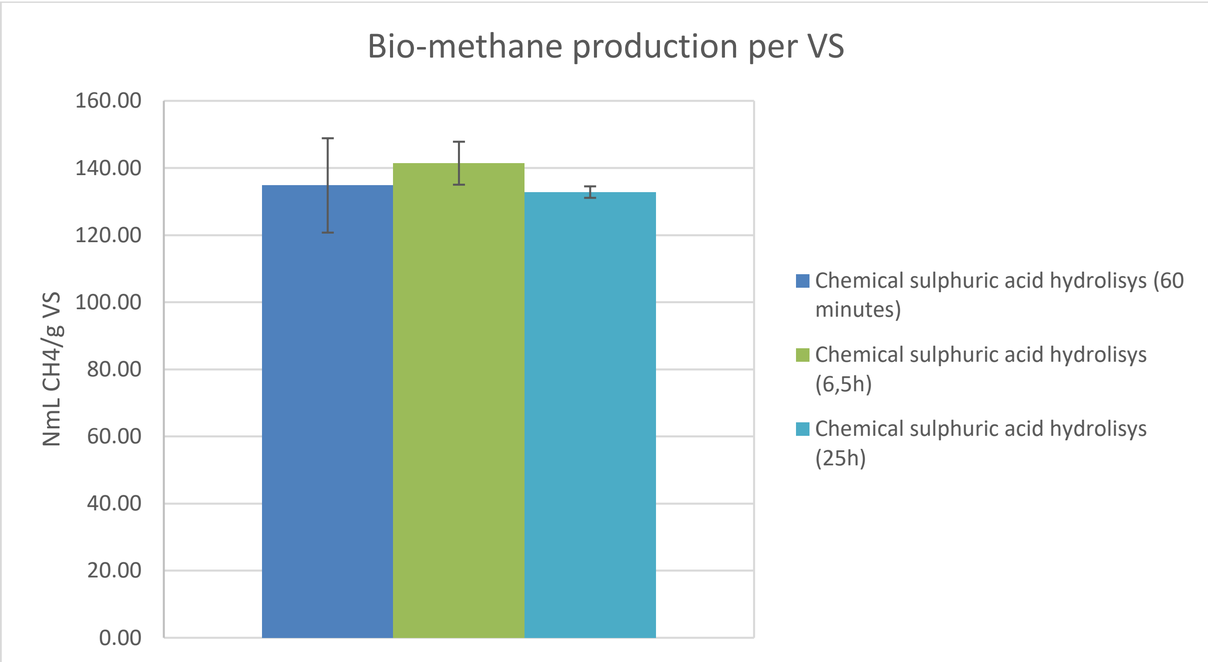

Figure 10: Average bio-metahen yield per VS depending on the acidic pre-treatment (GUT).

2.2.4. BMP-measurements of mixed pre-treatment methods

Experiments at GUT tested the biogas and bio-methane production after acidic hydrolysis and mechanical disintegration. At first the seaweed was mechanically treated for 120 s, and afterwards diluted. Further, 2 M sulphuric acid was added to the diluted seaweed until pH 2 level was reached. The hydrolysis was performed for 1, 6 and 24 hours. Afterwards, sodium carbonite was added until the solution was fully neutralised.

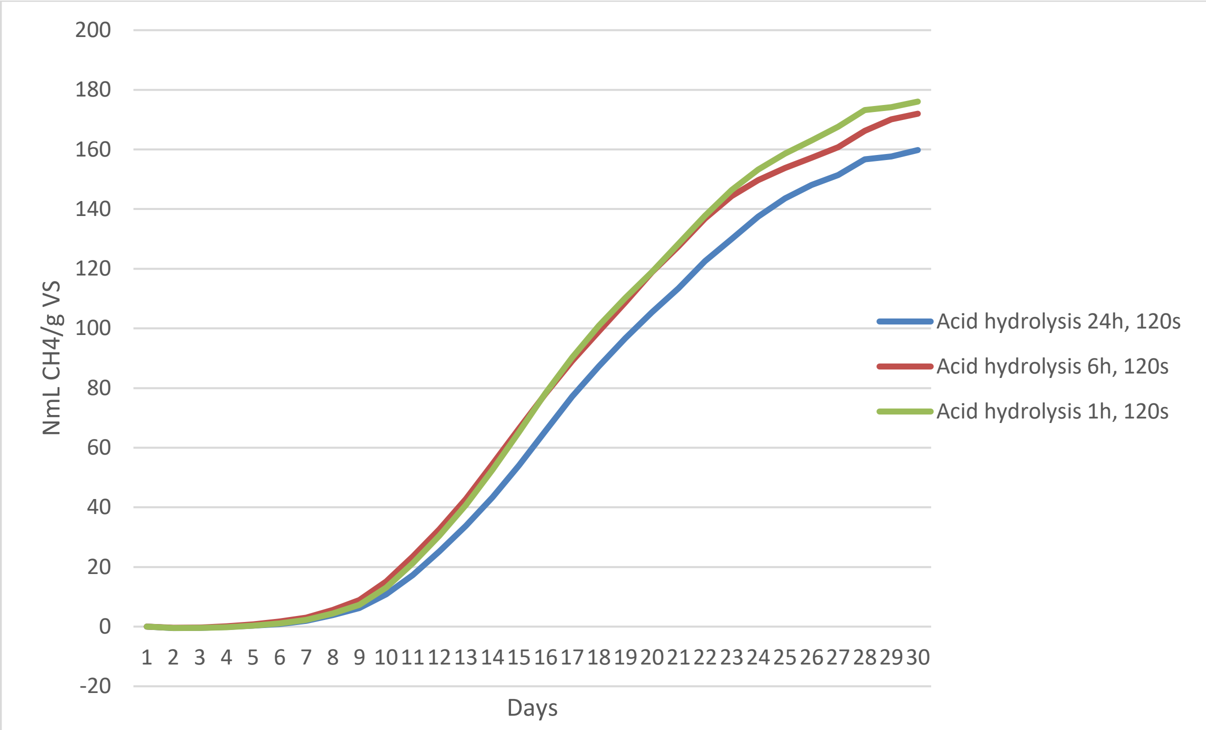

Figure 11: BMP measurements of acidic hydrolysis and mechanical pre-treatment conducted by GUT.

The experiments showed an increase in the average biogas production compared to the pre-treatment methods conducted alone.

As seen in Figure 11 above, the highest average biogas production is seen after 120 s (mechanical) and 6 hours (acidic). In comparison mixing of mechanical and acidic pre-treatment showed a percental increase of 49.7% compared to mechanical treatment alone. Likewise, an increase of 27.5% could be observed when comparing it to sole acidic pre-treatment. The experiments have shown that a mixture of pre-treatment methods can be an advantage when aiming for a higher methane production. A comparison of all pre-treatment methods is presented in Figure 12 below.

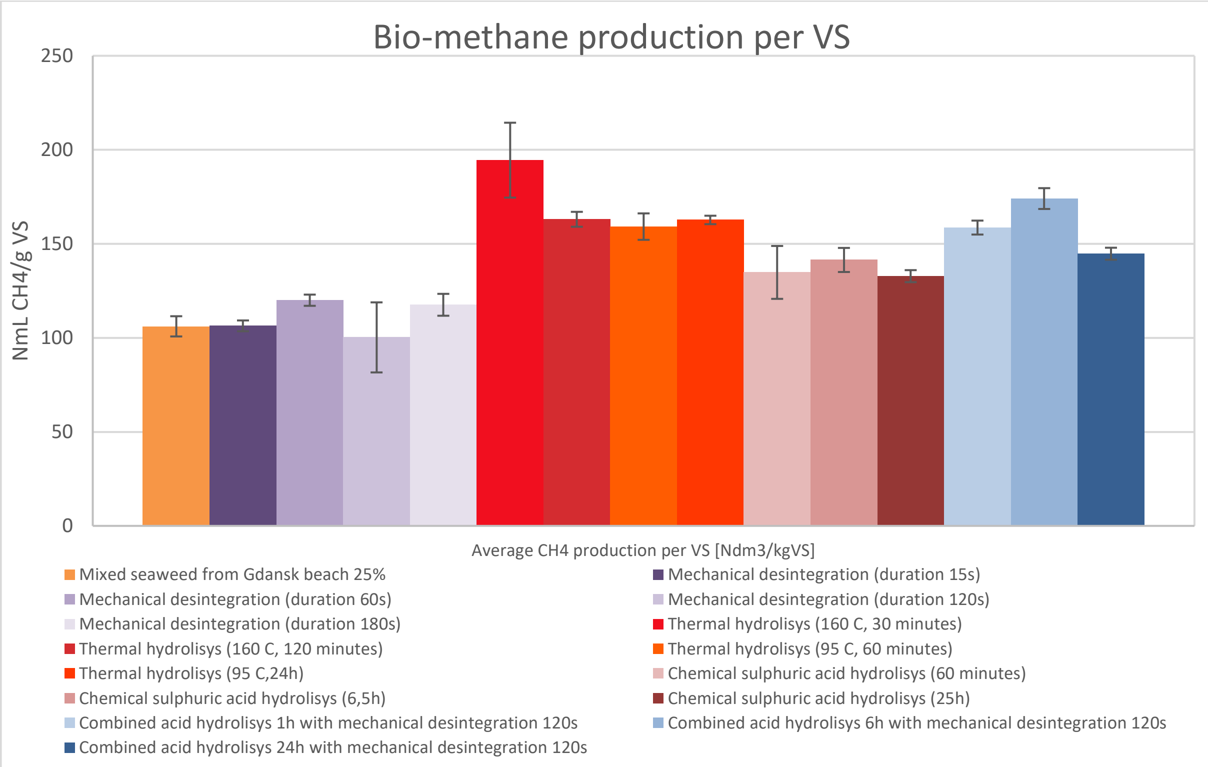

Figure 12: Average bio-methane yield of algae pre-treatment methods conducted by GUT.

The highest bio-methane yield was achieved after thermal pre-treatment (160°C, 30 min). Thermal pre-treatment using other time spans (160°C for 120 min, 95°C for 60 min) as well as the mixed pre-treatments of acidic and mechanical methods showed similar yields, which are significantly higher then using untreated seaweeds. Applying acidic hydrolysis resulted in slightly higher yields compared to untreated seaweeds. The least effect on bio-methane yield was observed when applying the mechanical disintegration pre-treatment.

2.3. Sand content

As mentioned, the sand content can be a challenge when collecting seaweed from the coastal areas. Depending on the collection technique, the cast seaweed can have a sand content up to 62% of the wet weight and 81% of the dry weight [13]. Furthermore, the sand content in the cast seaweed can depend on how long it has been lying on the beach.

Studies from Solrød have shown that fresh seaweed and seaweed that still floats in the water have a much lower sand content than the seaweed that has been on the beach for a longer period of time. The seaweed, which has been lying on the beach can have a sand content of 32-77%, whereas the fresh seaweed can contain as low as 14% sand [14].

Testing of sand content from seaweed collected by machine ("The Monster") (see Figure 13) has shown a sand content of 40% of DM for seaweed collected at the beach and 33% of DM for seaweed collected in the water.

Figure 13: "The Monster" collecting seaweed [RUC].

The testing of "The Monster" was better than other tests conducted with different types of collecting machines, but it was still not sufficiently well. It is desired to keep the sand content as low as possible in the collected seaweed, because a too high sand content can have a negative effect on the machinery in the biogas plant as well as causing higher transportation costs [15].

A study from RUC has tested the sand content in seaweed collected from three locations on the beach – fresh seaweed from the water, seaweed from the water edge (1-10 meters out) and seaweed from the beach. As shown in Table 1 below, the fresh seaweed collected in the water contained on average only 19% sand based on DM. Some of the tests were as low as 12%.

Table 1: Sand content in seaweed collected at Solrød beach

Collection location	Sand content DM%
Water (fresh)	12-25
Water edge (1-10 meters out)	42-49
Beach	37-53

The study from RUC corresponds with the results from the pre-feasibility study from Solrød Municipality [14], where the sand content of seaweed collected in water were 21.9% of DM.

Besides the location of collection, the type of seaweed can have an impact on the sand content, and furthermore on how easy it will be to clean it. This finding is supported by experiments conducted by GUT and LEI (see Table 2).

Table 2: Sand content in marine biomass after pre-treatment (GUT and LEI)

Type of algae	Sand content [%]	Place of sampling
Enteromorpha compressa	11.65	Shallow water
Enteromorpha plumosa	4.96	Shallow water
Potamogeton pectinatus	4.00	Shallow water
Zostera marina	20.88	Beach
Pheaophyta	7.80	Shallow water

As shown in Table 2, the collection location and the seaweed species has an effect on the sand content. *Enteromorpha plumose* and *Potamogeton pectinatus*, which were collected in shallow water, both have a low sand content of under 5%. The seaweed species *Enteromorpha compressa* has the highest sand content of the samples collected from shallow water.

An analysis of sand and seaweed from Denmark shows that the most common type of seaweed found in Solrød was Sea felt (*Pilayella littoralis*) and Filamentous brown algae (*Ectocarpus siliculosus*) [16]. These types of seaweed were stickier than others, which means that the sand content was higher. Pre-treatment will be necessary for these kinds of species, before they are used at the biogas plant.

The seaweed collected at the beach can contain up to 76.8% sand, whereas the sand content in seaweed collected directly from the water will be around 21.9% [14]. For efficiently using the cast seaweed in biogas plants, the sand content should be as low as possible and must not exceed 60% of DM – or even 50% as it is current code of practice at the Solrød biogas plant.

2.4. Co-digestion advantages

The methane potential in biogas production largely depends on the composition of the raw materials. The optimal choice of substrates and co-digestion ratio will depend on what feedstocks are available at the surroundings of each biogas plant. Former studies have shown that a co-digestion of the substrates can be of advantage for improving the gas yield [17].

In a pre-feasibility study conducted by Solrød municipality, the methane yield of pectin was tested [15]. The testing showed that pectin was responsible for 58% up to 68% of the methane production, which makes this co-substrate highly valuable for the biogas plant. However, pectin would not be suitable as sole substrate for biogas plants due to its low pH level. The mixture of manure, seaweed etc. is necessary to maintain an appropriate acidity level in the biogas plant. A similar test at Smyge pilot biogas plant concluded that maritime substrates should be co-digested with other organic substrates like manure and industrial residues to maintain a continuous and stable process [18].

A study from DTU – Department of Environmental Engineering [17], tested a set of co-digestion ratios to investigate the best co-digestion of cast seaweed and cattle manure. Among the ratios tested, 80% cattle manure/20% cast seaweed was identified as the best co-digestion ratio, when looking at potential methane yield (see Figure 14 below).

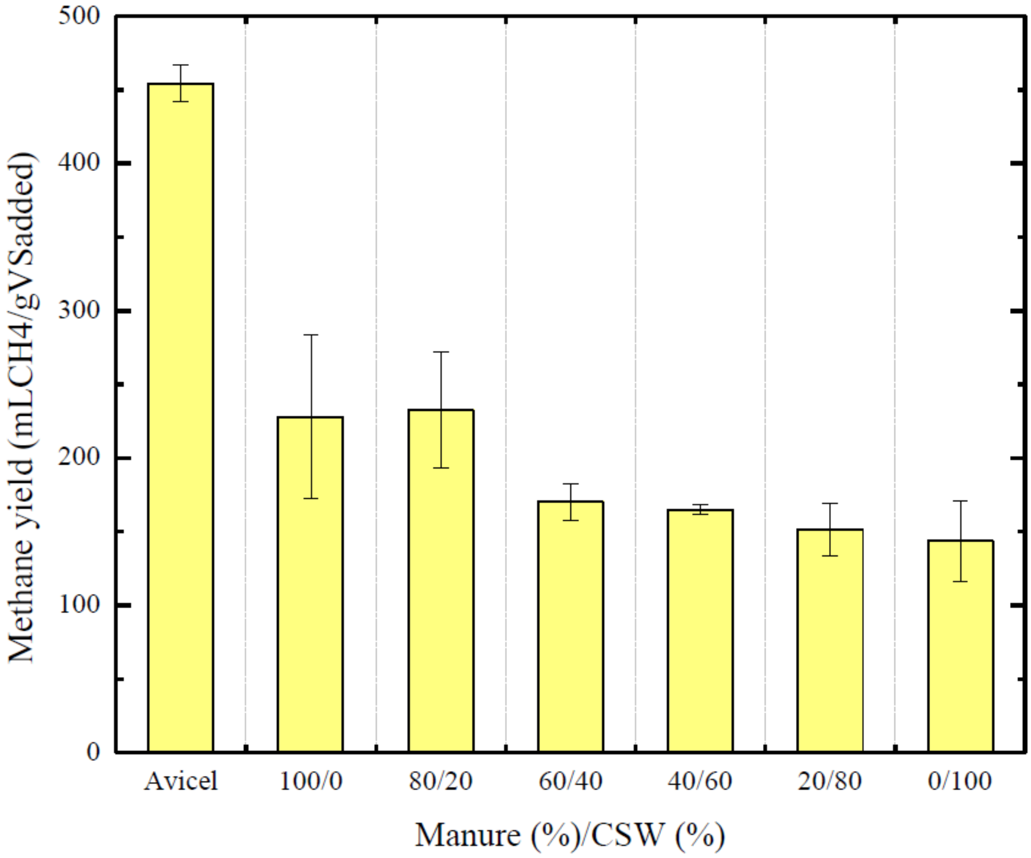

Figure 14: Methane yields of different manure-cast seaweed (CSW) co-digestion ratios [17]

The test showed that it was possible to achieve a higher methane production compared to a case where only manure is used as a substrate. A second set of experiments was performed to determine the best co-digestion ratios for sugar beet pulp, cattle manure and cast seaweed (see Figure 15 below).

The results of the co-digestion of different substrates at specific ratios seem to have a synergistic effect. This becomes obvious when comparing the results of the methane yields obtained from mono-digestion of each substrate and their co-digestion with cattle manure and cast seaweed.

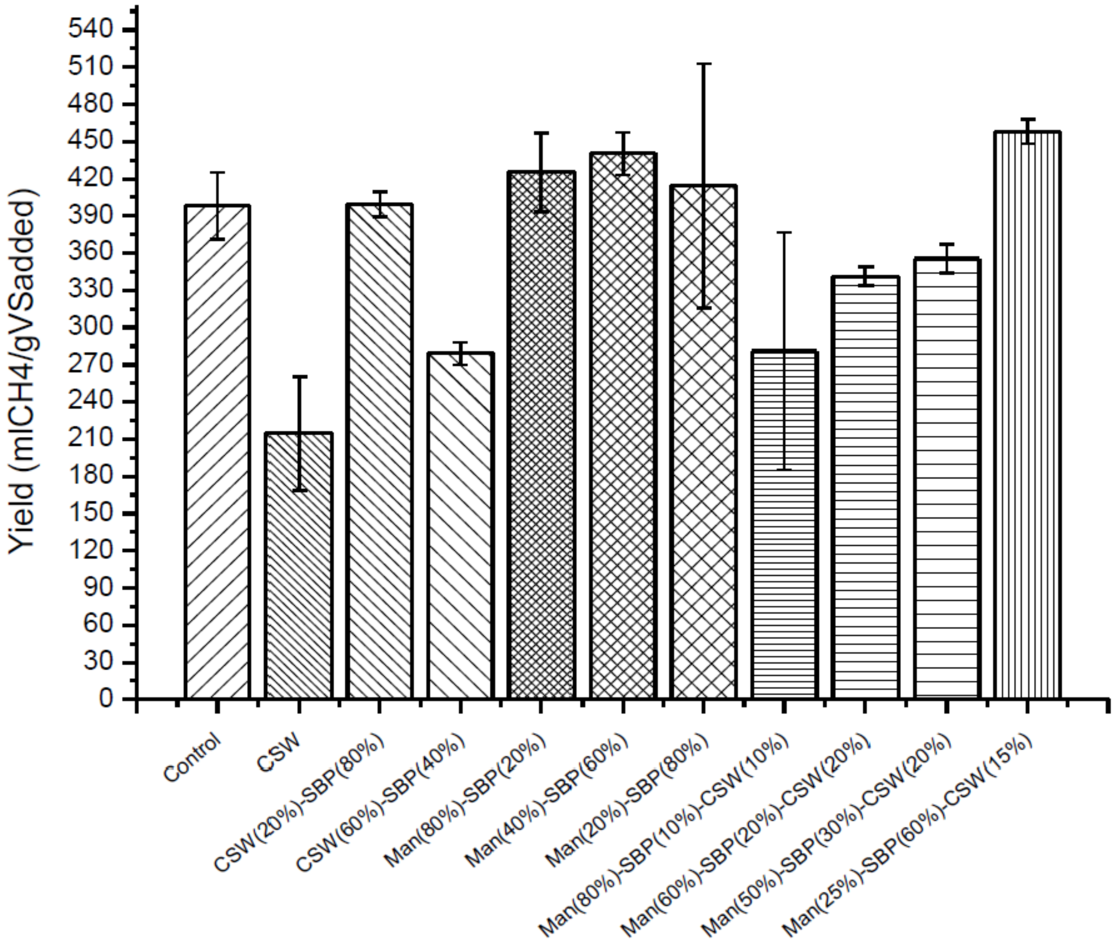

Figure 15: Methane yield of different sugar beet pulp (SBP), manure (Man) and cast seaweed (CSW) co-digestion ratios [17]

The best co-digestion ratios identified were Man (80%)/SBP (20%), Man (40%)/SBP (60%), Man (20%)/SBP (80%) and Man (25%)/SBP (60%)/CSW (15%). The study proved that co-digestion of sugar beet pulp, cattle manure and cast seaweed, results in a higher methane yield.

Co-digestion is necessary to maintain a stable methane yield over a longer period [17], which means that the ratios for the substrates at the biogas plant must be tested.

3. Nutrients and heavy metals in biogas production – the Solrød case

At Solrød Biogas plant, the material input consists of manure, pectin (a residual product at CP Kelco), eluate (a residual product from the production of lactic acid bacteria [19]) and seaweed collected from the coast of Køge Bay [3]. All of the materials contain different levels of nutrients and heavy metals, and both seaweed and manure contain sand. The material flow is shown in Figure 16 below:

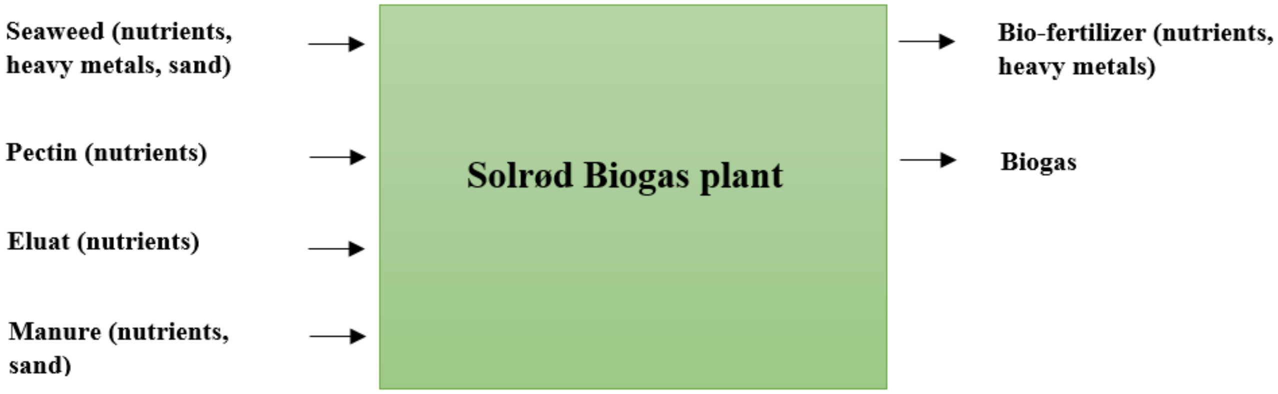

Figure 16: Material flow at Solrød Biogas plant (own illustration, data from Solrød Biogas A/S)

At Solrød Biogas plant, either the biogas is upgraded to methane, which is fed to the regional gas grid, or it is used for electricity and heat production. The residues of the process are used as organic fertiliser, which provided the manure as co-substrate. Furthermore, seaweed from the nearby coast is used as feedstock.

The levels of nutrients in the used seaweed and pectin, was tested by the Solrød Municipality [15]. According to the results, pectin contains 8.0 kg N/t and 0.74 kg P/t and seaweed on average 4.8 kg N/t and 0.69 kg P/t. The high level of these nutrients can also be measured in the process residues (the digestate) and is desirable when utilised as fertiliser on farmland. In the case of seaweed, an additional benefit is the removal of surplus nutrients from the marine environment.

3.1. Nutrient and heavy metal levels in seaweed

Although seaweed seems to have a great potential as feedstock in the production of biogas, the potentially high content of heavy metals constitutes a significant problem for both the digestion process, but also for further use as a fertiliser on farmland.

To use cast seaweed in the production of biogas, the levels of nutrients and heavy metals cannot exceed the current guidelines, which are set by the Danish Waste to Soil Regulation [20]. As for the heavy metal cadmium, for instance, the concentration cannot be above the limit of 0.8 mg/kg DM (in Denmark) in the collected seaweed [3]. Therefore, seaweed samples are taken once a month, to prevent using cast seaweed with an inadequate amount of heavy metals. The results of the cadmium content becomes available in the middle of the month, and if the cadmium content is below the limit value, the seaweed is supplied to the biogas plant. If the cadmium content is higher than the limit value, the seaweed is returned into the water. As seen in Figure 17 below [21], the cadmium content typically exceeds the limit value during the winter season.

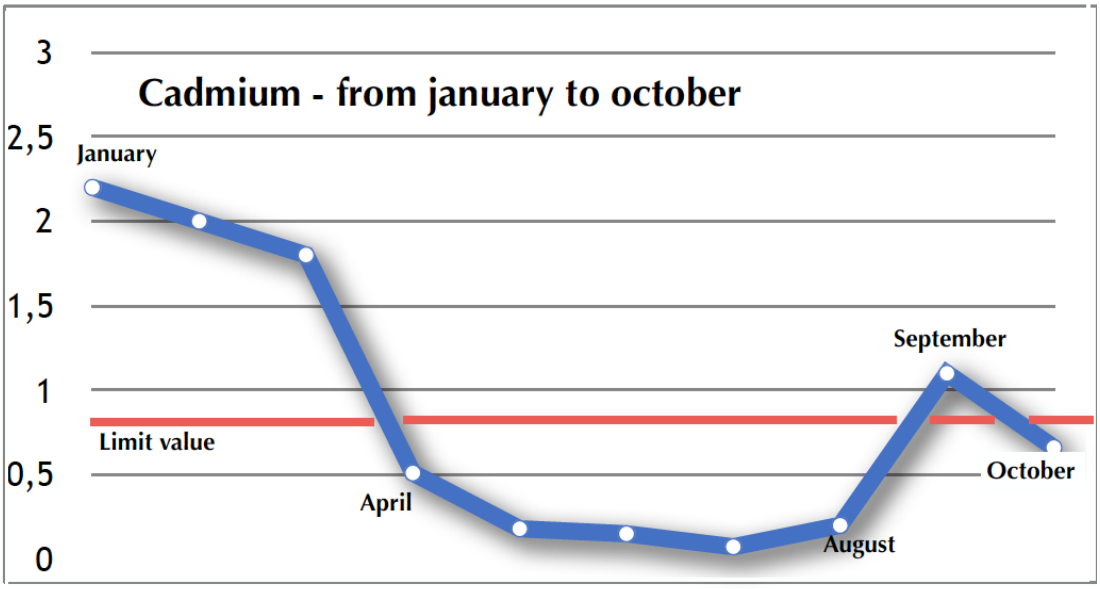

Figure 17: Cadmium content in cast seaweed collected at Solrød Beach, 2018

As depicted in Figure 17 it was only possible to use the cast seaweed in Solrød Biogas plant in the summer months of 2018 (April to August, and October). Because it takes seven to eight working days to analyse the seaweed and the seaweed collection is carried out until the next cadmium measurement, it is necessary to return the cast seaweed into the water, as it is not suitable for using in the biogas plant.

A study at RUC from September 2020 showed the levels of heavy metals in seaweed collected at Solrød beach. The seaweed samples were collected from piles on the beach and from the water edge. The analysis was performed by Højvang Laboratorier A/S (see Table 3).

Table 3: Levels of heavy metals in collected seaweed from Solrød beach, September 2020

Heavy metals	Damp seaweed from piles on the beach mg/kg DM	Wet seaweed from the water edge mg/kg DM	Limit values mg/kg DM
Lead	2.5	<2	120
Cadmium	0.44	0.17	0.8
Mercury	<0.03	<0.03	0.8
Nickel	3	1.76	30
Chromium	1.06	0.58	100
Copper	<5	<5	1,000
Zink	31.66	13.66	4,000
LAS	128	114	1,300
PAH	-	-	3
NPE	<0.1	<0.1	10
DEHP	<0.5	<0.5	50

The assessment showed that none of the samples exceeded the limit values set by the Danish Waste to Soil Regulation [20]. Both samples of seaweed did not exceed the limit value for any of the heavy metals stated in the regulation, which means that it would be suitable for use in the biogas plant.

The influence of seasons on the heavy metal content of seaweed is difficult to predict and makes it more difficult for the biogas plant operator to predict when this feedstock could be used. Experiences have shown that the cadmium level cannot be predicted depending on the month and that it will vary from year to year. A study from the bay of Køge showed [21] that seaweed cannot be included generally in the biogas production from November to April.

In addition to the heavy metals in seaweed, the nitrogen and phosphorus levels were analysed at Højvang Laboratorier A/S. The analysis showed the nitrogen levels in the seaweed collected in water amounted on average to 26.66 g/kg DM and 30.66 g/kg DM for seaweed collected from piles on the beach. The phosphorus levels for seaweed collected in the water were on average 0.63 g/kg DM and 0.76 g/kg DM for seaweed collected on the beach. The results of the two studies can be seen in Table 4 below.

Table 4: Nutrient levels for seaweed collected at Solrød beach in 2010 and 2020

Collection location	Total nitrogen g/kg DM	Phosphorus g/kg DM
Study conducted by Solrød, 2010		
Water	16.50	1.2
Beach	9.0	0.4
Study conducted by Roskilde University, 2020		
Water edge	26.66	0.63
Piles from the beach	30.66	0.76

In line with the cadmium contents that vary from month to month, the same can be observed concerning the levels of nitrogen and phosphorus in cast seaweed. The study from Solrød tested collected seaweed for nutrient and heavy metal levels at different times a year. The first batch from January 2009 showed a nitrogen level of 7.1 g N/kg DM and 1.2 g P/kg DM [14]. The second batch from May 2009 showed a nitrogen level of 4.1 g N/kg DM and 0.53 g P/kg DM. The third batch from January 2010 showed a nitrogen level of 3.10 g N/kg DM and 0.34 g P/kg DM. The study showed that the variation of the nutrient levels in cast seaweed cannot be predicted depending on the month when the seaweed is collected.

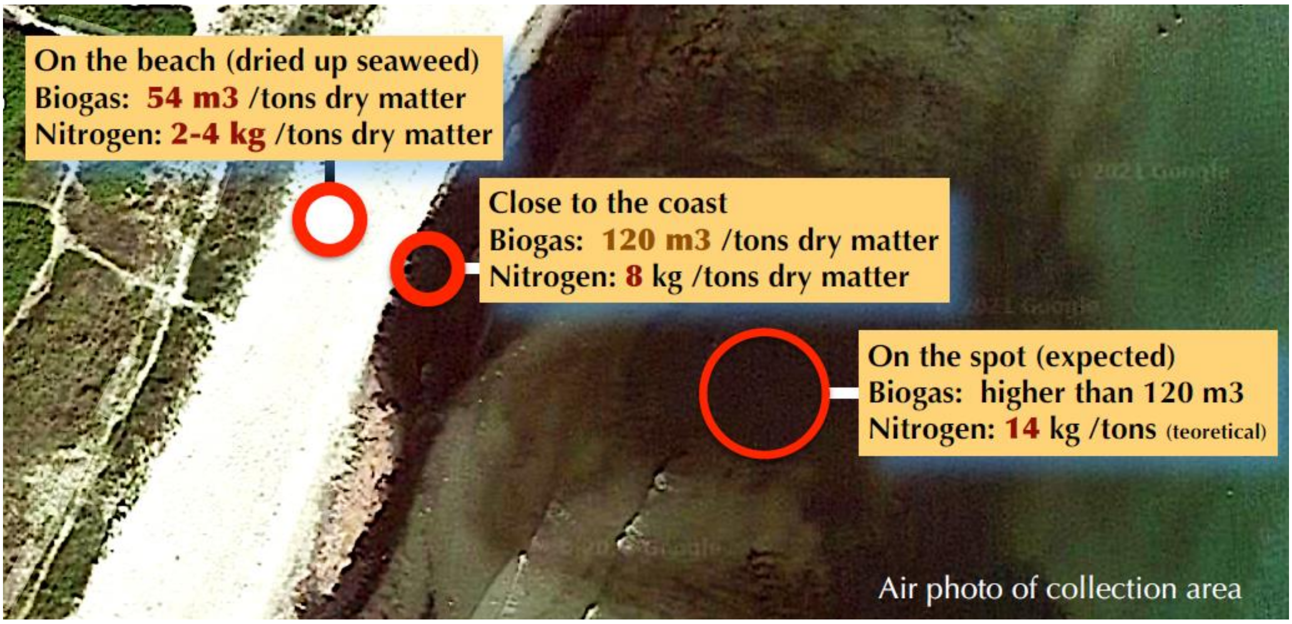

Figure 18: Nitrogen levels based on seaweed collected at different locations, on the beach, close to the coast and far out in water.

The analyses from Solrød Municipality showed that the nutrient content is higher in fresh seaweed [15]. When the seaweed is lying on the beach for a long period, the nutrients are mineralised and washed out. This means that the seaweed needs to be collected as fresh as possible to have an effect on the aquatic environment (and for producing and efficient organic fertiliser from the residues of the biogas production) (see Figure 18).

3.2. Nutrients in organic fertiliser

Digested biomass from the biogas plant could be used to substitute chemical fertilisers. In order to use cast seaweed for this purpose, the biomass must not exceed the given limit values for heavy metals. To ensure sufficiently low levels of heavy metals, chemical testing of the marine biomass is necessary. The following table shows the test results from Solrød Biogas plant [22], which was conducted in 2018. It lists the quantities of nutrients and heavy metals in the digestate for every month.

Table 5: Organic fertiliser harvest 2018, Solrød Biogas plant [22]

Month	Total Nitrogen (N), kg/ton	Phosphorus kg/ton	Ammonium (N), kg/ton
August	4.45	0.58	3.15
September	4.60	0.68	3.23
October	4.90	0.78	
November	5.75	0.60	3.96
December	5.77	0.70	4.04
January	4.69	0.64	3.27
February	4.79	0.78	3.30
March	4.71	0.50	3.30
April	3.94	0.57	3.00
May	4.33	0.67	2.85
June	3.86	0.64	2.80
July	4.14	0.44	3.05
Average	**4.66**	**0.63**	**3.27**

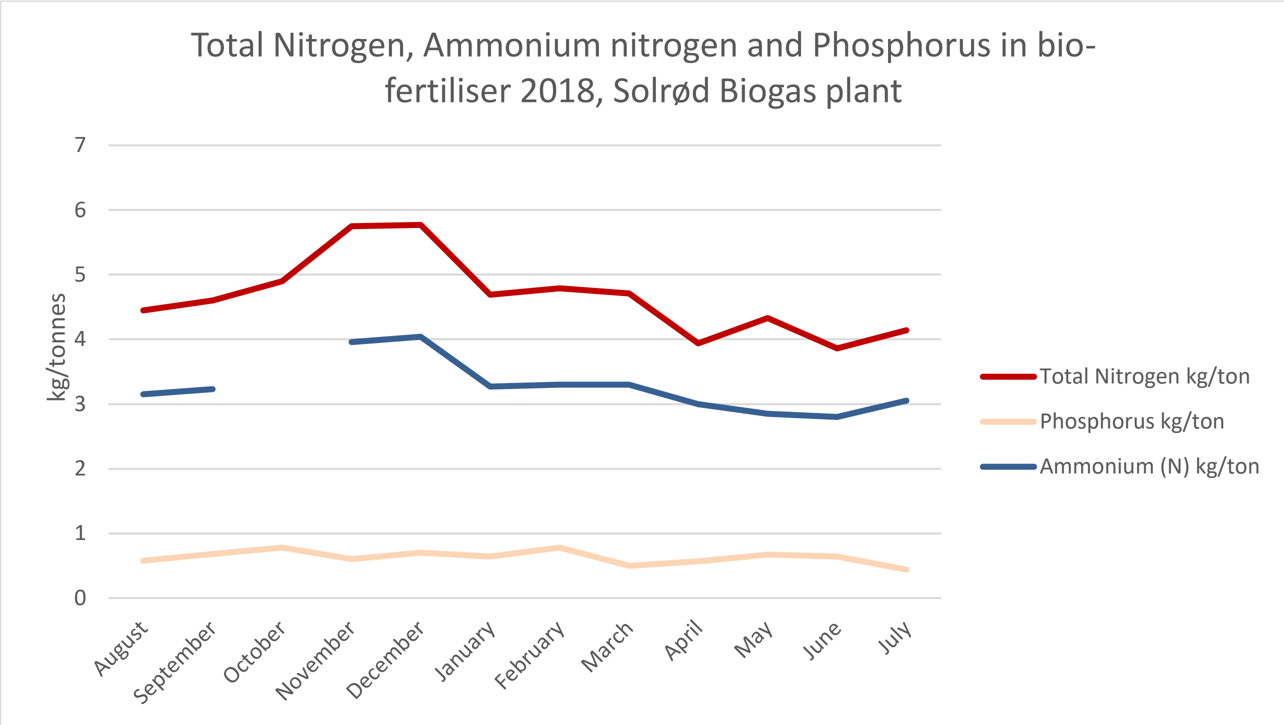

Figure 19: Nitrogen, ammonium nitrogen and phosphorus levels in bio-fertiliser at Solrød Biogas Plant, 2018

As Figure 19 above shows, the highest levels of total nitrogen and ammonium can observed in November and December. A spike in the total nitrogen level is happening from October to December, where the levels reached over 5 kg/t, afterwards the level declined and amounted to between 4 kg/t and 5 kg/t throughout the year. The phosphorus level was stable through the year (between 0.50 to 0.78 kg/t).

The bio-fertiliser has several advantages compared to chemical fertiliser. The bio-fertiliser is quickly absorbed by the plants, which means that the risk of nitrogen leaching into the environment is reduced. Furthermore, the nitrogen uptake of the crop increases by 10-25% [22]. For the organic fertiliser to be used on farmland, guidelines must be followed. Only 170 kg N/ha and 25 kg P/ha can be added to farmland [23].

4. Circular use of seaweed – nitrogen recycling

4.1. Eutrophication in the Baltic Sea

According to HELCOM, eutrophication is defined as *"one of the main threats to the biodiversity of the Baltic Sea and is caused by excessive inputs of nutrients to the marine environment"* [23].

Eutrophication is the result of enhanced inputs of nutrients and organic matter, leading to changes in primary production, biological structure and turnover and is resulting in a higher trophic state [24]. The secondary or indirect effects include increased or lowered oxygen concentrations, and changes in species composition and biomass. Furthermore, the low concentrations in the bottom water can affect the fish and plants.

According to HELCOM at least 97% of the Baltic Sea is assessed to be below a good eutrophication status, including all of the open sea area and 86% of the coastal waters, where much of the eutrophication occurs near the coastlines [25].

Eutrophication occurs when a significant amount of nutrients, including nitrogen and phosphorus, are added to the sea environment. In recent years, more than 700,000 t of nitrogen and 25,000 t of phosphorus have entered the Baltic Sea annually. Eutrophication starts with the supply of nutrients, which leads to a strong growth in biomass production, including elevated levels of macro vegetation, increased turbidity, oxygen depletion in bottom waters, changes in species composition and an increase of adverse blooms of algae, leading to oxygen deficiency [26].

The nutrients can be both airborne and waterborne and have a negative effect on the sea environment (see Figure 21). The waterborne sources are rivers and direct discharges from point sources and the airborne inputs include atmospheric deposition directly into the sea. According to HELCOM, about 75% of the nitrogen load and at least 95% of the phosphorus load enter the Baltic Sea via rivers or as direct waterborne discharges [23]. The other 25% of the nitrogen load comes from atmospheric deposition.

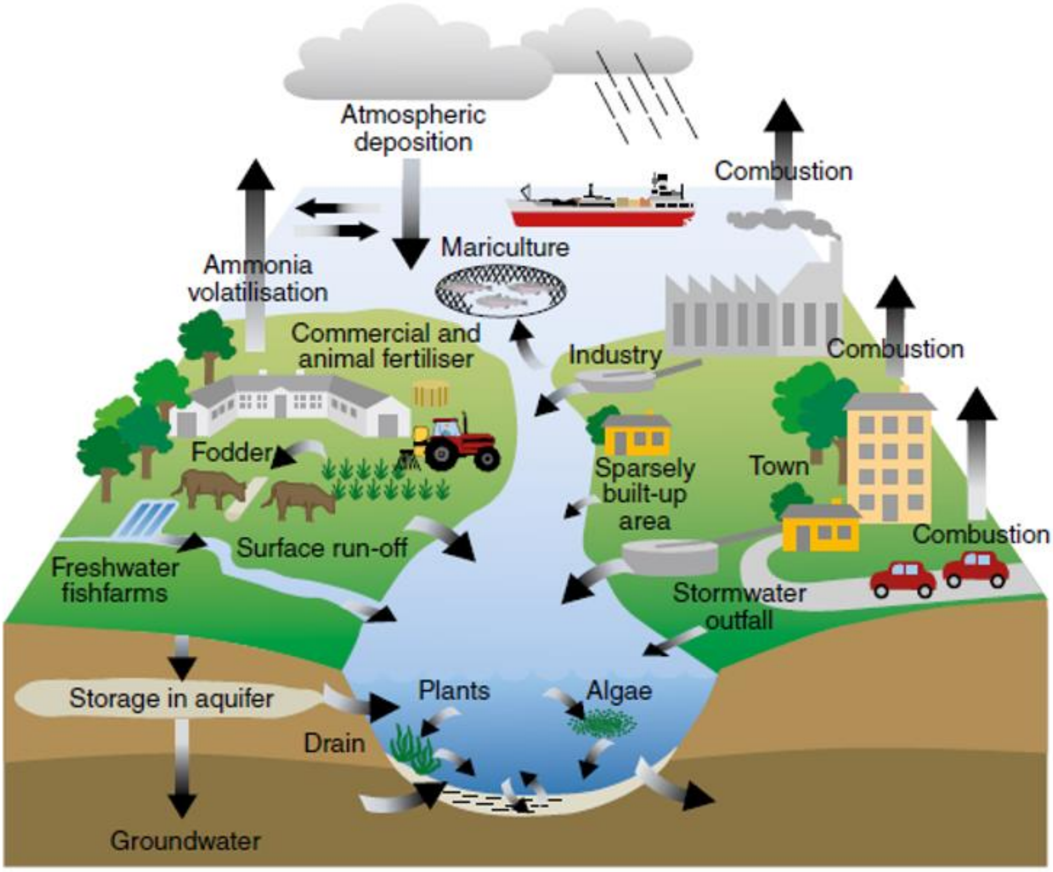

Figure 20: The nitrogen cycle in the aquatic environment [24]

Eutrophication particularly occurs in coastal areas, where the production and biological decomposition of seaweed plays a role [27]. The seaweed absorbs the nutrients, but with the biological decomposition of seaweed, the nutrients are released again. To stop this cycle, the seaweed needs to be collected before the degradation happens. By collecting the seaweed in coastal areas, a contribution can be made to counteract eutrophication and improve the coastal water and the quality.

4.2. Removal of nutrients

As stated, the removal of nutrients could help to prevent eutrophication in the Baltic Sea. Measurements by HELCOM depict the process of nutrient removal from the different areas in the Baltic Sea (see Figure 21).

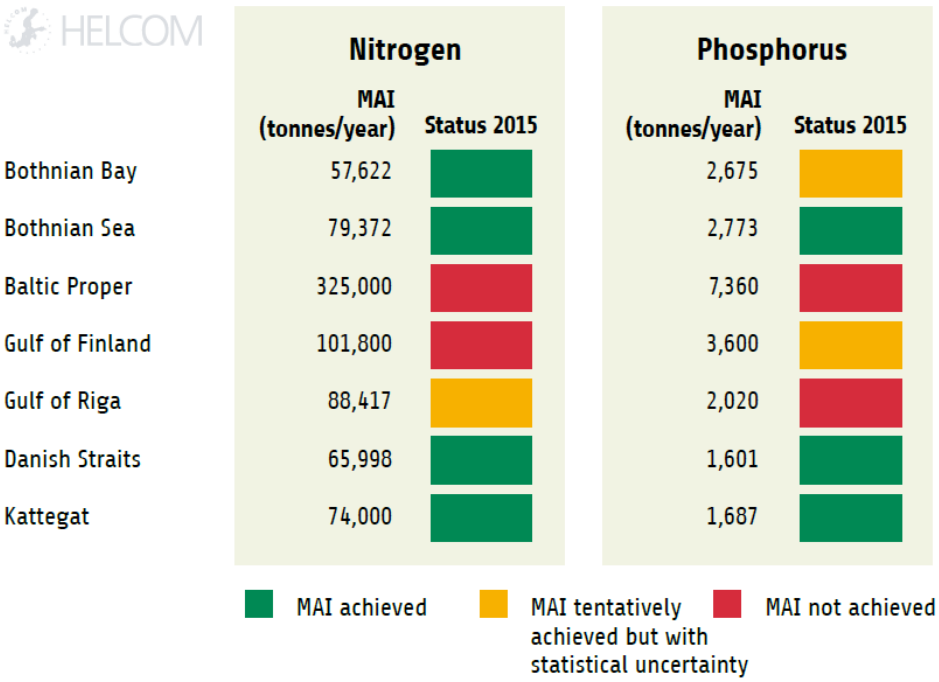

Figure 21: Progress of nutrient reductions in the Baltic Sea in relation to maximum allowable inputs (MAI) [29]

The removal of nutrients will be a necessary step to prevent further levels of eutrophication. To stay below the maximum allowable input of nutrients in the different areas, all surrounding countries have to map out the sources of nutrients, which are ending up in the Baltic Sea.

As Figure 22 below is stating, the total load of nitrogen (TN Total Nitrogen) and phosphorus (TP Total Phosphorus) in the year 2014 were 825,825 t and 30,949 t respectively [30].

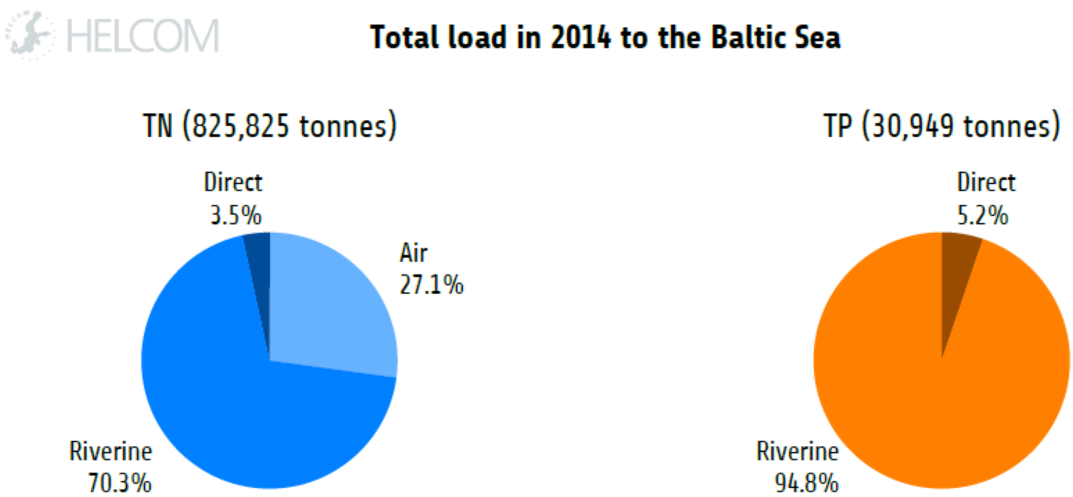

Figure 22: Total load of nitrogen and phosphorus in 2014 to the Baltic Sea [30]

Statistics from HELCOM show that the nutrient load from the surrounding countries has mostly been declining from 1995 to 2014 [31]. In **Germany**, the total nitrogen load has declined from 84,212 t in 1995 to 61,396 t in 2014. The nitrogen load in **Poland** has declined from 262,159 t in 1995 to 169,941 t in 2014. In **Sweden**, the total nitrogen load has declined from 133,681 t in 1995 to 109,596 t in 2014. In **Denmark**, the nitrogen load has declined from 85,611 t in 1995 to 56,630 t in 2014. The nitrogen load in **Lithuania** was declining from 1995 to 2000. In this period, the nitrogen load declined from 56,515 t to 42,080 t. From 2000 to 2014, the nitrogen load has increased to 56,426 t, which is almost the same level as in 1995.

Even though the statistics show that most of the countries have succeeded in declining the nitrogen load to the Baltic Sea from 1995 to 2014, most parts of the Baltic Sea are still assessed to be below a good eutrophication state [25]. Further decline in the nitrogen load will therefore be necessary.

As stated earlier, the collection of seaweed is contributing to the removal of nutrients, and at the same time the recirculation of nitrogen. For Solrød it is estimated that the removal of seaweed from the coast prevents 62 t of nitrogen and 7 t phosphorus to be released into the water [12]. This amount is not high enough to prevent the further development of eutrophication. However, it should be mentioned that the further utilisation of seaweed could have a positive effect on the nutrients cycle.

4.3. Regulations related to the reductions of nutrients in the South Baltic Sea

The **EU Water Framework Directive** 2000/60/EC (WFD) [32] incorporates the water quality standards and integrates the principles of effective and sustainable development. The WFD is the most substantial directive for water legislation in the EU. The WFD states that an effective water policy must take into account the vulnerablity of aquatic ecosystems in the costal areas, as these areas are strongly inclueneced by the inland waterways. Furthermore, it is stated that it is necessary to develop an integrated community policy on water. The WFD aims at maintaining and improving the aquatic environment. It aims to achieve the objective of a good water status in coastal areas. To achive the objectives and uphold the aquatic environment to a good or high status, the nutrient concentrations should not exceed the established levels.

The WFD is stating that the problem with the aquatic ecosystems should be solved jointly by the surrounding countries.

The Baltic Marine Environment Protection Commision (Helsinki Commision – HELCOM) aims to achieve the Baltic Sea Action Plan's goal of the Baltic Sea to be unaffected by eutrophication [23].

Nutrients overload from agriculture continues to be one of the biggest pressures on the aquatic and marine environment. This negative effect from farmland, rivers etc. needs to be addressed to achieve a good ecological status of waters as establised by the WFD. To achieve a Baltic Sea that is undisturbed by excessive inputs of nutrients, HELCOM has set a set of objectives [1] (see Table 6).

Table 6: Objectives set by HELCOM to achieve a Baltic Sea which is undisturbed by excessive inputs of nutrients.

Objectives
No excessive nutrient concentrations
Clear water
Natural oxygen levels
No excessive algal blooms
Natural distribution of plants and animals

To achieve this set of objectives, the actions required are to reduce the amounts of nutrients entering rivers from diffuse sources, especially farmland, to reduce nutrient pollution from remaining "hot spots", such as wastewater treatment plants and lastly to reduce airborne nutrient pollution.

In the Baltic Sea Action Plan from 2007 [23], the maximum nutrient input to the Baltic Sea can be allowed to be about 21,000 t of phophorus and 600,000 t of nitrogen.

Table 7: The maximum allowable nutrient inputs to reach good environmental status in the Baltic Sea [23]

Sub-region	Maximum allowable nutrient input (t)		Needed reductions (t)	
	Phosphurus	Nitrogen	Phosphorus	Nitrogen
Bothnian Bay	2,580	51,440	0	0
Bothnian Sea	2,460	56,790	0	0
Gulf of Finland	4,860	106,680	2,000	6,000
Baltic Proper	6,750	233,250	12,500	94,000
Gulf of Riga	1,430	78,400	750	0
Danish straits	1,410	30,890	0	15,000
Kattegat	1,570	44,260	0	20,000
Total	**21,060**	**601,720**	**15,250**	**135,000**

As shown in Table 7, some sub-regions in the Baltic Sea have already succeeded to reduce the nutrient input. Baltic Proper is the sub-region, which is in need of the highest reduction of both phosphorus and nitrogen. Over half of the sub-regions have not succeeded in reaching the needed reduction for nitrogen. Gulf of Finland, Baltic Proper, Danish Straits and Kattegat all need to endure further reductions to reach good environmental status.

As stated earlier, the total nitrogen load in 2014 to the Baltic Sea amounted to 825,825 t [31]. To reach the goal set by HELCOM in the 2007 Baltic Sea Action Plan further reductions will be needed. With a maximum allowable nitrogen input of 601,720 t, 224,105 t are still needed to be reduced.

By collecting cast seaweed and using it as a substate and utilising the digestate as a fertiliser, some nutrients will be removed from the Baltic Sea, and provide a tool to mitigate eutrophication.

4.4. Reduction of nutrients at sea and at farmland

In Køge Bay, the Danish Ministry of Environment has set an objective in the Water Framework from 2010 to reduce the supply of nitrogen with a minimum of 86.2 t and 5.9 t phosphorus [33]. The objective was applicable to the year 2015. To reach this, 22,200 t of cast seaweed would be needed to be collected yearly from the coast of Køge Bay. In 2019, only 1,522 t of cast seaweed were collected from Køge Bay. The nitrogen removal was 12.2 t. This is far less than what was described in the pre-feasibility study [14]. Furthermore, the biogas plant in Solrød is at the time only able to receive up to 7,400 t of cast seaweed.

A calculation by RUC (see Figure 23) shows the removal of nutrients at sea, as well as the added nutrients at farmland. Figure 23 below is based on the collection of 1,000 t of seaweed (fresh and sand free). By collecting 1,000 t of seaweed at the coast, 8,118 kg nitrogen and 197 kg phosphorus are removed from the coastal area and the sea. The same amount of nutrients – 8,118 kg nitrogen and 197 kg phosphorus – will be added to farmland.

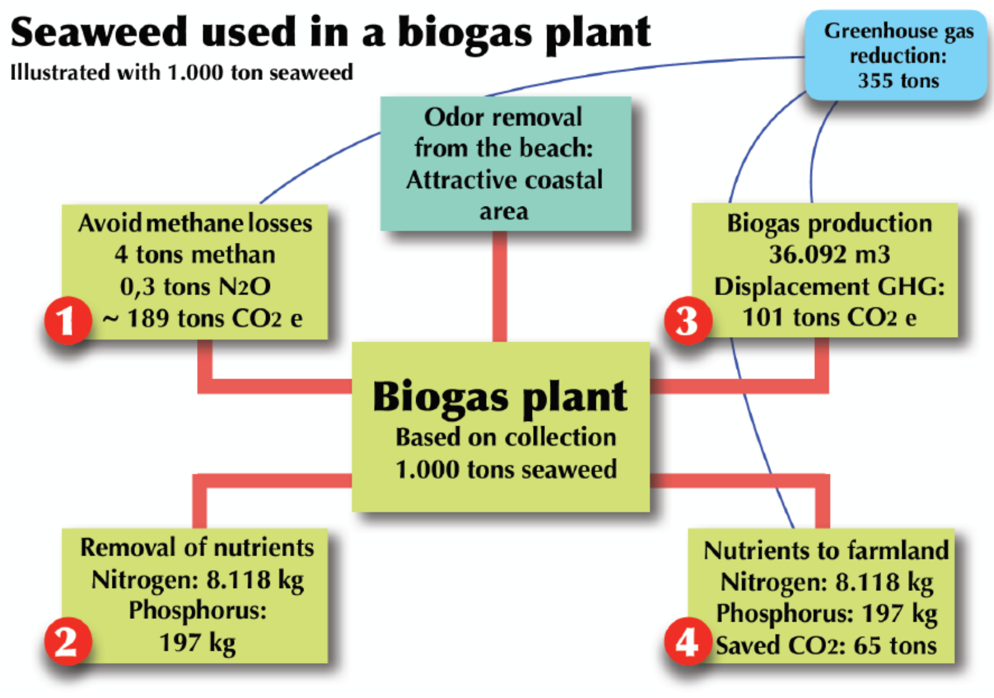

Figure 23: Benefits from removal of seaweed (illustration by Kjær, T.)

If the values as indicated in Figure 24 are being used to calculate the reduction of nutrients, the collected amounts in Solrød in 2019 (1,522 t) would mean a reduction of 12,355 kg N and around 300 kg P. If Solrød is able to collect the maximum 7,400 t of cast seaweed, as stated in the pre-feasibility study [34], the reduction would be 60,073 kg N and 1,457 kg P. This is still not enough to achieve the goal set in the Water Framework, but would still be a huge reduction by collecting seaweed alone.

Based on the earlier estimated seaweed amounts (made by RUC) in the partner countries, the nutrient reduction would be as shown in Table 8.

Table 8: Estimated amounts of nitrogen and phosphorus reduction, calculations based on Figure 24

Country	Seaweed amounts (t)	Nitrogen reduction (t)	Phosphorus reduction (t)
Denmark	607,646	4,932	119
Germany	140,278	1,138	27
Sweden	1,143,951	9,286	225
Poland	184,790	1,500	36
Lithuania	76,002	616	14
Total	**2,152,667**	**17,472**	**421**

The seaweed amounts are estimated based on suitable coastal areas for collecting seaweed in the partner countries. The description for the coastal areas, which are suitable, and how the seaweed amounts have been calculated can be found in Deliverable 4.1 of the COASTAL Biogas report – *A report on beach cleaning and pre-treatment of seaweed* [35]. To calculate the nitrogen and phosphorus reduction for each country, the local conditions need to be included.

To use the collection of seaweed as a tool for removing nutrients from the Baltic Sea, the seaweed amounts are still not high enough to have a significant effect. However, the seaweed collection could be an important tool for reducing nutrients in the coastal areas.

5. Concluding remarks

Nutrients overload from agriculture continues to be one of the biggest pressures on the marine environment. This needs to be addressed to achieve the objectives of good water quality as established by the WFD and HELCOM. The recommendation by the COASTAL Biogas project to collect and use cast seaweed as a substrate in biogas production could help to achieve parts of these objectives. By collecting seaweed, some of the nutrients are removed from the coastal marine environments, which can help to counteract eutrophication. The collected seaweed can be used at biogas plants and be included in the process of producing renewable energy.

Experiments have shown that the methane yield of digested seaweed depends on the species and when and where it was collected. The methane yield in cast seaweed is significantly lower than in other biomasses, such as cattle slurry, pectin residue etc. However, testing of pre-treatment methods and co-digestion with other biomass substrates have shown a significantly increase in the methane yield of cast seaweed, which makes the seaweed of higher value for biogas production.

Furthermore, the project recommendation to use the digestate as organic fertiliser as a substitute for chemical fertilisers could help to reduce the leakage of surplus nutrients from agriculture.

The collection of seaweed, as a way of reducing nutrients in the Baltic Sea, should be seen as one of the solutions, which could help to prevent eutrophication. It is not possible to use this method alone for reducing the total nutrient load in the Baltic Sea. However, it could be one efficient tool to face this problem. Besides, the methane emissions and odor nuisances, which are caused by cast seaweed, should be avoided by collecting and using this biomass as feedstock in the biogas production.

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