

Green Hydrogen for the CO₂ Neutral Production of Sustainable Biogas at Stormossen's Plant, Vaasa, Finland.

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Eventually, I would like to make a statement about how these kinds of projects contribute to creating a net of knowledge that fosters sustainability and respect for the environment by showing commitment to the development of a circular economy and the reduction of human carbon footprint.

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Abstract

This thesis aims to study a newly designed system for the production of biomethane by the utilization of the CO₂ gas emitted into the atmosphere at Stormossen Oy, a waste management plant in Vaasa, a western coastal city of Finland. This can be achieved by studying how to use this CO₂ and convert it again into methane with green hydrogen production and storage from renewable resources.

Literature research has been done to obtain information about the current biomethane production process that the company has; the common processes of carbon dioxide usage and hydrogen production and storage and its maturity and availability, especially by electrolysis methods; and to study electricity prices to determine its viability.

As a result, an analysis of the most appropriate electrolysis method for the production of green hydrogen and the best conversion into biomethane technique has been determined. Regarding the results discussed in this thesis, Stormossen Oy's plant can therefore decide to invest in this type of system for the desired increase of biomethane production, by implementing the methodology and technology analysed.

Language: English Keywords: Biogas, biomethane, green hydrogen, CO₂ reduction, power-to-gas

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Glossary

Information was obtained directly from the European Environment Agency (EEA, 2022).

Biowaste: This kind of waste is formed mainly by food and garden waste. It is considered a key waste stream with a high potential for contributing to a more circular economy.

 CO_2 eq: Universal value used to indicate in terms of CO_2 the equivalent of any greenhouse gases regarding its potential for global warming.

GHG: Greenhouse gases are gases that contribute to the natural greenhouse effect. The Kyoto Protocol covers a basket of six greenhouse gases (GHGs) produced by human activities: carbon dioxide, methane, nitrous oxide, hydrofluorocarbons, perfluorocarbons, and sulphur hexafluoride. An important natural GHG that is usually not covered by most protocols is water vapour.

P2G: Power to gas is the process of converting surplus renewable energy into hydrogen gas through PEM electrolysis technology.

WEM scenario: The projections scenario 'with measures' (WM) or 'with existing measures' (WEM) means projections of anthropogenic GHG or air pollutant emissions by sources that encompass the effects of currently implemented or adopted policies and measures.

1. Introduction

Today, a large part of the world's pollution comes from industries, either directly emitting contaminating gases to the atmosphere in production processes, or indirectly due to the degradation of subproducts in the environment (Neagu & Teodoru, 2019). Nevertheless, European governments and institutions have been recently fostering sustainable policies as well as different measures that can achieve several goals like the reduction of greenhouse gas emissions (Albulescu et al., 2020).

In this sense, Vaasa, a municipality in the coastal western region of Finland with a population of about 60,000 (City of Vaasa, 2022) has been active in establishing a circular economy from the utilisation of municipal waste for waste to energy (Wte).

Vaasa's waste recycling plant called Stormossen Oy has been producing biogas since 1990 (Stormossen Oy, 2020). This company follows a clear strategy to achieve a 65% quantity of materials recycled by the year 2030. As part of the desire to contribute to the creation of a circular economy, they are making efforts to minimize their greenhouse gas emissions by optimising their biogas production process.

This can be achieved in several ways. In this thesis, the combination of CO_2 emissions from the current biogas process and the production and storage of green hydrogen to convert it into biomethane which is eventually reintroduced in the main production process is the subject analysed.

2. Aims and objectives

This thesis aims to develop an intensive analysis and research of biomethane production by the utilization of the CO_2 gas emitted into the atmosphere at the current Stormossen plant. By studying how this CO_2 is emitted and converting it again into biomethane with green hydrogen production from renewable resources to reintroduce it into the main biogas pipeline.

Therefore, to achieve the aim of this thesis several objectives have been defined:

- Study and comprehension of the current biomethane production process at Stormossen Oy.
- Investigation of the range of available green hydrogen production techniques based on electrolysis.
- Determination of the most adequate hydrogen storage method.
- Identification of the most suitable conversion technique for synthetic methane production.
- Evaluation of the economic and environmental impact of the newly designed process.

3. Stormossen Oy

Literature states that recycling is a key factor in a circular economy (Ragossnig & Schneider, 2019). Therefore, Stormossen Oy plays an important role in this field. This is a company that currently employs around 40 people and is responsible for treating bio-waste and sludge from six different municipalities in the Ostrobothnia region, Finland, as shown in Figure 1 extracted from (Digital Office, 2022).



Figure 1. Finland and Ostrobothnia's region map.



Figure 2. Owner municipalities of Stormossen Oy.

As observed in Figure 2, extracted from Stormossen Oy's webpage, and marked by a red dot, their main plants are located close to Vaasa, the principal municipality of the region because of logistical reasons (Stormossen Oy, 2022). Here, both the waste treatment centre and the incineration plant are situated, even though a total of 13 different waste reception stations, marked in dark blue in the previous figure, can be found all around the region. By paying a specific fee, Inhabitants and companies can bring sorted waste to these specific reception stations.

In 2018, Stormossen Oy's annual report discloses a total of 147.200 tonnes of waste were managed (Stormossen Oy, 2018), representing an increase of 4% from the former year (Stormossen Oy, 2017), from which 62.300 were municipal waste. That year, municipal waste amounts to 452 kg per person, which is significantly below the mean value of the country in 2017 which amounted to 510 kg/person (Official Statistics of Finland, 2017).

Taking into account that municipal waste mainly consists of combustible waste, biowaste and recyclable waste received at the recycling stations. The report states a recovery rate of 98.7%, which can be then divided by 52.8% of material recovered and 45.9% recovery in the form. The rest, approximately 800 tonnes, which represents 1.3% of the total, was deposited at the landfill site.

This previous sorting in recycling stations allows Stormossen Oy to separate mainly biowaste from the rest, and to proceed with the conversion of it into compost soil and biogas vehicle fuel at the waste treatment centre, observable in Figure 3.



Figure 3. Stormossen Oy's waste treatment plant scheme.

This scheme shows the different elements that set up this plant. In numerical order: 1 is the biowaste delivery point; 2 is the crusher; 3 is the 1st screw press; 4 is the 2nd screw press; 5 is the magnet; 6 is the Westenergy Oy Ab's building; 7 is the buffer tank; 8 is the gas storage; 9 is the upgrading facility; 10 is the refuelling station; 11 is the gas engine; 12 is the steam plant; 13 is the central heating, 14 is the torch; 15 is the reception silo for sludge; 16 is the homogenisation process; 17 is the reactors; 18 is the intermediate tank; 19 is the drum sight; 20 is the centrifuge; 21 is the process water treatment plant and 22 is the composting. Moreover, in green the biowaste, in orange the combustible waste, in dark blue the metal, in yellow the gas in light blue the process water and in brown the sludge.

In this sense, the biogas from Stormossen's plant is refined into vehicle gas with the name BIG, an umbrella brand for three biogas producers in Finland which make vehicle fuel (BIG Biogas, n.d.). It is remarkable that the first refuelling station Stormossen inaugurated not long ago, in 2017 (City of Vaasa, 2017).

As Stormossen Oy's 2018 report asserts, that year, Stormossen sold a total of 407.000 kg of vehicle gas achieving double refuelling compared to the previous year. Its main use is for public transport as currently, the Vaasa region has twelve gas-powered buses operating with it and it is planning to increase this number up to nineteen buses at the end of summer 2022. Therefore, Stormossen plans to increase its biogas fuel production to supply the demand by inspecting different methods.

3.1. Production process

The biogas production process follows a strict methodology. Biowaste is daily disposed of at Stormossen's plant, with around 10 garbage trucks delivering all kinds of food leftovers per day at the delivery points, as observable in Figure 4. Secondly, a pre-treatment facility opens all bags and packages where this waste is contained, as shown in Figure 5.



Figure 4. Biowaste delivery point at Stomrossen Oy.

Figure 5. Screwtransporters feeding biowaste to crusher at Stormossen Oy.

After the biowaste is crushed into smaller pieces, warm water is introduced, as seen in Figures 6 and 7. This process is crucial for the biowaste slurry to be transported by pipes to the reactor.



Figure 6. Ventilation pipelines at Stomrossen Oy.



Figure 7. Biowaste crushing process at Stormossen Oy.

A separator then eliminates some extra residues that might be found mixed with the biowaste such as metals or plastics that could affect the process, as literature states (Mudhoo & Kumar, n.d.). The pipes lead to the buffer tank before the biomass is fed into the reactor which holds up to 1,7 million litres of biomass, see Figure 8.



Figure 8. Buffert tank for biowaste at Stomrossen Oy.

Figure 9. Metal coil in bioreactor at Stomrossen Oy.

Here, the biomass and biogas are mixed constantly with huge metal spirals in the reactor for about 3 weeks, as observable in Figure 9. In this reactor, different microorganisms and bacteria are eating the biomass and therefore creating methane gas (Li et al., 2011).

This gas from the reactor moves to the gas storage, seen in Figure 10, and continues to an upgrading unit where the gas is washed with the amminoscrubber that gives a final gas with a methane content of 98% as required, observable in Figure 11.



Figure 10. Gas storage at Stormossen Oy.

Figure 11. Gas upgrading unit at Stormossen Oy.

After adding the required substances like its characteristic odour, the biogas is ready to be filled up in any of Stormossen's gas stations. Nevertheless, the main drawback is that nowadays the bioreactor is working at maximum capacity and thus research in new innovative technologies must be done to solve it. Here is the point that this thesis emphasizes.

3.2. Biomethane production and emissions

Biogas is a mixture of gases, basically a combination of two-thirds of methane (CH₄) 50%-75% with carbon dioxide (CO₂) 50%-25% and traces of hydrogen sulphide (Bharathiraja et al., 2016). It is a great alternative source of energy adding to water, wind, and solar energy (Khanal et al., 2019). Its production process consists of an anaerobic fermentation which can be accomplished by specific water treatments, and most certainly for the matter at hand by sludge stabilization and organic waste treatment (Laca et al., 2019). This generation process employs bacteria that work under anaerobic conditions, as oxygen kills these bacteria that are naturally found in the raw materials (Thomas, 2003).

Nevertheless, to get biomethane CO₂ needs to be eliminated, as unwanted CO₂ reduces the quality of biogas, and then expensive upgrading processes to purify it is required (Sangeetha et al., 2020). Moreover, to use the natural gas transport system higher pressure and the absence of CO₂ must be ensured (Muth et al., 2021). Biogas, once purified by removing CO₂, is used as a renewable and low-carbon fuel for electricity generation and transportation (Yu et al., 2018). CO₂ removal is currently based on different physicochemical processes such as absorption, adsorption membrane and cryogenic techniques (Scholes, 2020).

Stormossen's CO₂ emissions take place after the anaerobic digestion in the process of biogas cleaning, as mentioned, in the upgrading to reach the desired 96% of biomethane, by the application of some separation techniques (Zeppilli et al., 2019). These emissions must be reduced or avoided in order to ensure a CO₂ neutral production process. Therefore, as observable in Figure 12, a new method of production is introduced by the reuse of the emitting CO₂.



Figure 12. Diagram of the biomethane production process with the CO₂ conversion method.

All inputs and outputs of the diagram can be identified, illustrating how the emitting CO_2 from the main process would be converted into new biomethane with the use of green hydrogen from renewable resources.

As observable in Figure 13, a schematic view of how Stormossen Oy plans to use this emitted CO_2 has been already thought and planned by the company. This figure represents the future vision the company has in order to get to a more sustainable future without polluting the atmosphere.



Figure 13. Scheme of Stomossen Oy's future plans.

In the scheme, the storage of CO₂ and the use of green hydrogen are presented, but not detailed. This thesis aims to crumble the specifications and calculations behind this process in order to analyse its viability and practicality. Therefore, the application of this methodology would perfectly fit into Stormossen Oy's future plans of reducing its carbon footprint and fostering a circular economy.

4. Research and Design

4.1. The role of Carbon dioxide (CO₂) in global warming

Carbon dioxide is also known as carbonic anhydride. It is a gas formed by a carbon atom covalently double bonded to two oxygen atoms as seen in Figure 14, extracted from (Peter Schreiber, 2020).



Figure 14. Carbon dioxide molecule.

This gas exists in the atmosphere by a proportion of 280 ppm in the carbon planetary system. This system includes all active reserves: atmosphere, biosphere, hydrosphere, and lithosphere (Herrick & Lipták, 2003). Although this molecule is less found in the atmosphere, it is in this stage where it plays the main role.

During the last 800.000 years, atmospheric CO_2 concentration has been fluctuating between 170 and 330 ppm which are the acceptable levels for the sustainability of the earth (Lüthi et al., 2008). Nevertheless, during the last 170 years with a surprisingly increased way in the last decades, it has reached values of 415 ppm.

CO₂ emissions have increased drastically, and this comes with consequences, as it is a gas that contributes to global warming. Other natural gases like methane and nitrous oxide or artificial ones like fluorinated gases are part of the well-known greenhouse gases (GHG) (Darkwah et al., 2018). Their rise triggers climate change, climate crisis and climate emergency, different terms to describe the effects on The Earth of global warming.

Latest official statistics (United Nations, 2022) state that CO₂ emissions have not reduced during the last years, excluding lockdown months and the drastic activity drop many countries had because of the Covid-19 pandemic. In 2018, as an example, the European Union (UE) emitted around 3,9 Gton of CO₂eq (equivalent carbon dioxide), representing up to 7% of GHG. Therefore, if the EU managed to accomplish climate neutrality it would have a huge impact on the climate challenge.

Uncontrolled carbon dioxide emissions are one of the main causes of global warming (Goel & Agarwal, 2014), and some studies certify it's the largest contributor to it (Mardani et al., 2019). The cause of it is not only the high and unrestrained human activity but also worsen due to the long prevalence CO_2 has on the atmosphere.

4.2. The role of Hydrogen (H₂) in energy storage

Hydrogen is the first element in the periodic table, being the lightest existing element in the known universe. In normal conditions i.e., standard pressure and temperature (SPT), it is found as a gas (McCay & Shafiee, 2020a). Its atom is formed by a proton and an electron, and its stable form is done in the diatomic molecule H₂, as observable in Figure 15.



Figure 15. Electron shell - Hydrogen atom.

Literature states that hydrogen exhibits the highest heating value per mass of all chemical fuels and is not only regenerative but also environmentally friendly (Kayfeci et al., 2019). Furthermore, in energy terms, 1kg of hydrogen has the same energy as 3 kg of gasoline, but the latter emits around 9kg of CO₂ when combusted (Siyal et al., 2015). Historically hydrogen has been obtained using different methods, the most common ones use fossil fuels as the main source (Scott, 2022), especially steam reforming of methane. In this sense, its production methods can be classified depending on the primary fuel used (hydrocarbons, ammonia, methanol, ethanol, water) and depending on the chemical reactions that take place (steam reforming, partial oxidation, decomposition, gasification, electrolysis) (Kothari et al., 2008).

As part of fostering a circular economy, a good method to be studied is electrolysis because of its main fuel capability, which is water. Electrolytic hydrogen production based on electricity generated from renewable resources is commonly called green hydrogen and can contribute to the global need for sustainable energy supply methods (Bhandari et al., 2014). Nevertheless, this technique is also not free from some environmental burdens.

4.2.1. Electrolysis of water

Electrolysis is the process in which two elements of a chemical compound are separated using an electrical current (Chi & Yu, 2018). Electrons are liberated by the anions in the anode, producing oxidation and therefore these electrons are captured by the cations in the cathode producing a reduction. This can be obtained by water and renewable energies for the obtention of electricity (Kakoulaki et al., 2021). In these terms, the process consists of the decomposition of water (H₂O) into oxygen (O₂) and hydrogen (H₂). This production of storable hydrogen by water electrolysis can solve the discontinuity of electricity prices and its fluctuating utilization (McCay & Shafiee, 2020a). The electrolysis process utilizes the DC power from sustainable energy resources for example solar, wind and biomass. But, at present only a sum of 4% of hydrogen is obtained by electrolysis of water mainly due to the economic issues (Shiva Kumar & Himabindu, 2019a). It is expected an increase in this value in the near future, because of the usage of renewable energies (solar, wind, nuclear), following the already European Energy Directive goal fixed that stated to utilize 20% of the energy requirements from renewable energy sources by 2020 (Roadmap et al., 2011).

In this sense, water electrolysis can be classified into different types based on their electrolyte, operating conditions, and ionic agents (OH⁻, H⁺, O²⁻), while the operating principles are the same (Kumar, S. & Himabindu, 2019). The four kinds of electrolysis methods are (i) Alkaline water electrolysis (AWE), (ii) Solid oxide electrolysis (SOE), (iii) Microbial electrolysis cells (MEC) and (iv) Proton exchange membranes (PEM) water electrolysis.

The more conventional alkaline water electrolysis operates at temperatures of 60-80°C, having an operating pressure value that varies from atmospheric to 5 bar in conventional electrolysers and goes up to 10 to 30 bar in advanced ones (Koponen, 2015). Nevertheless, PEM water electrolysis technology is often demonstrated in the literature as a possibly very effective alternative to the former mentioned (Rashid et al., 2015). Its main advantages are flexibility in operation and higher energy efficiency. Moreover, it not only requires low current, partial load and low-pressure operation (about 2 bar) but also works well with hydrogen density. In this technique, water is electrochemically split into hydrogen and oxygen at their respective electrodes such as hydrogen at the cathode and oxygen at the anode, as shown in Figure 16, extracted from (Shiva Kumar & Himabindu, 2019).



Figure 16. PEM electrolysis production method.

As observable, in the anode, the water molecule is split forming oxygen (O_2) as a subproduct, and on the cathode side, the protons and electrons recombine to produce hydrogen (H_2).

The overall electrolysis reaction is the sum of the two electrochemical half-reactions, that occur at the electrodes in an acidic environment according to the succeeding reactions:

$$H_2 O \rightarrow 2H^+ + \frac{1}{2}O_2 + 2^{e^-}$$
 (1)

$$2H^+ + 2^{e^-} \to H_2 \tag{2}$$

As a result:

$$H_2 O \rightarrow H_2 + \frac{1}{2}O_2 \tag{3}$$

According to (Hinkley et al., 2016) 54kW of electrical energy is demanded to reach a capacity of 900kW. Moreover, the minimum water electrolysis can consume is about 9 kg of water per kg of hydrogen (Lampert et al., 2016). Nevertheless, as the literature states, taking into account the process of water de-mineralisation, the ratio can range between 18 kg and 24 kg of water per kg of hydrogen. The exact amount of hydrogen from PEM electrolysis can be calculated following the equation:

$$M_{H_2} = \frac{E_{ED} \cdot \eta_{rec}}{E_{ez}} \tag{4}$$

Where:

 E_{EO} is the electrical energy demand of the conversion system (e.g. Wind, solar, hydropower).

 η_{rec} is the rectifier efficiency taken as 0,9 (Mohsin et al., 2018).

 E_{ez} is the energy demand of the electrolyser, taken as 54 kW/kg (Hinkley et al., 2016).

4.3. CO₂ capture and H₂ storage

At Stormossen Oy's biogas production process, CO₂ is already separated from biogas in a clean state. Biomethane can meet a low average sulphur value (e.g. 10 mg/m3), according to the company's annual reports (Stormossen Oy, 2017), containing some percentage of H₂S from the amminoscrubber (Carver Pump, 2021).

As CO₂ then, is already obtained daily from the production process, hydrogen must be available to proceed with the conversion. Therefore, H₂ viable storage methods must be analysed to maintain the process running without the need to stop because of the lack of hydrogen availability.

Hydrogen can be stored with several methods, overall, 6 different methods were found in the literature (Graetz, 2008; Züttel, 2004a).

- High-pressure gas cylinders (800 bar).
- Liquid hydrogen cryogenic tanks (21K).
- Adsorbed hydrogen on materials (<100K).
- Adsorbed on interstitial sites in a host metal (ambient conditions).
- Chemically bonded in covalent and ionic compounds (ambient conditions).
- Oxidation of reactive metals.

Hydrogen can be stored at high density in different forms but not all of them have reached commercial maturity for a large-scale level (Andersson & Grönkvist, 2019). Some of the mentioned storage technologies are nowadays being vigorously investigated, meaning that considerable progress still has to be made (McCay & Shafiee, 2020b).

It is clear that innovative storage methods are still in relatively early stages of development. Despite the several methods mentioned, the volumetric density of hydrogen of 0,089886 kg/m³ makes it a challenge to store that must be overcome (Rivard et al., 2019). To achieve the reduction of this volume, hydrogen must be compressed.

This is the reason why high-pressure gas cylinders are the most common method (Ayodele & Munda, 2019). They seem to be the most appropriate too, by not only reaching a maximum pressure of 200 bar but also because new technology in this area allows these new so-called lightweight cylinders to bear pressures up to 800 bar reaching a volumetric density of 36 kg/m³ (Züttel, 2004b).

The volume of this compressed hydrogen can be determined by the equation.

$$Q_{H_2} = \frac{M_{H_2} \cdot \eta_{cm}}{\sigma_c} \tag{5}$$

Where:

 M_{H_2} is the quantity of hydrogen obtained in kg.

 σ_c is the density of the compressed hydrogen.

 η_{cm} is the compression efficiency taken as 0,95 (Lagorse et al., 2008).

4.4. Conversion to biomethane

The conventional biogas plants using older technologies should be updated with recently developed biological technologies. Literature shows that biological technologies have not been used for full-scale or pilot plants yet. Currently, the primary challenge with biological methods is their upscaling from lab scale to full-scale because of their complexity and various abiotic and biotic parameters should be considered for upscaling (Hagos et al., 2017).

These processes are also time-consuming, and they require extra equipment (Tabatabaei et al., 2019). Besides this, biological technologies also require more experimental data to generate a full mass and energy balance needed for a reliable techno economics analysis along with a life cycle analysis. Upscaling from laboratory to full scale can be tricky especially since these processes rely on mass transfer of H₂ as well as CO₂ into the liquid phase, which has very different kinetics (Bharathiraja et al., 2018). In this sense, Table 1 gives a comparison between conventional biogas upgrading technologies.

	Membrane separation	Pressure Swing Adsorption	Water Scrubbing	Chemical (Amino) Scrubbing	Organic Solvent Scrubbing
Maximum recovery (%)	96 - 98	>96	>97	99,5	>99
Heat demand	-	-	-	100 - 180 ºC	55 – 80 ºC
Operation pressure (bar)	6 - 8	4 - 10	4 - 10	Atmospheric	4 - 8
Outlet pressure (bar)	4 -6	4 -5	7 - 10	4 - 5	1,3 - 7,5
Energy demand (kWh/m³)	0,25 – 0,43	0,46	0,46	0,27	0,49 – 0,67
N_2 and O_2 removal	Partial	Possible	No	No	No

Table 1. Conventional biogas upgrading techniques.

On the other hand, conventional biogas upgrading methods are commonly used today and account for 99% of all upgrading plants (Ardolino et al., 2021). However, these conventional technologies have limitations that can increase the cost of upgrading raw biogas. For instance, water scrubbing is the most commonly used conventional technique for upgrading raw biogas used in 41% of the upgrading plants. Nevertheless, this process consumes large amounts of water, and water regeneration will greatly increase the cost of water treatment (Mwacharo et al., 2020). Therefore, the water issue may be resolved by using emerging biological processes in which water is not required during the upgrading of the raw biogas (Petersson & Wellinger, 2009).

Therefore, Table 2 shows a comparison between different emerging biogas upgrading techniques.

	Chemoautotrophic	Photoautotrophic	Fermentation	Cryogenic	Hybrid
Methane content (%)	>96	90	95	99	99
Temperature	30 - 88	15-35	25 - 30	-196	-
рН	6,5 - 8	7 - 10	5,5 - 6,6	-	-
CO₂ removal	43,3 - 100	89-93	-	-	-
CH₄ recovery	78,4 - 96,1	97,2	-	97 – 98	>99,5
H ₂ S removal	Yes	Yes	-	Yes	-

Table 2. Emerging biogas upgrading techniques.

As observable, biological conversions of CO₂ and hydrogen to methane are made without the need for high pressure and temperature, and it is basically carried out by hydrogenotrophic methanogens.

Besides hydrogenotrophic methanogens converting carbon dioxide into methane, microbial electrolytic cells capturing carbon dioxide have also attracted attention (Christodoulou et al., 2017; Zhang et al., 2020). The method of removing carbon dioxide by converting it into methane is called electro-methanogenesis, as indicated by the following equation (Nakasugi et al., 2017).

$$CO_2 + 8e^- + 8H^+ \rightarrow CH_4 + 2H_2O$$
 (6)

This production of biomethane from CO₂ with the usage of hydrogen solves two different environmental problems and stays ahead of the recently developed sector of carbon capture and utilization.

Most biochemical removal systems for carbon dioxide rely on hydrogen fed into the biogas to generate more methane by conversion of CO₂. However, recent literature shows that some methanogens, such as Methanosaeta, can be involved in direct interspecies electron transfer (Rotaru et al., 2014; Zhao et al., 2015). Therefore, compared to the electron transmission between species via H₂, this former methodology is "cheaper" as the production of hydrogen requires energy for electrolysis (Gahlot et al., 2021), but that would be another topic of discussion.

4.4.1. Methodology

As mentioned, in order to make the process economically viable cheap energy to produce H₂ must be used, and to ensure the availability of the process a suitable CO₂ source must be selected, which in this case is the main methane production unit. Among the different options mentioned previously to upgrade biogas, such as chemical/physical absorption, membrane and cryogenic technologies, the Power to Gas (P2G) concept has received increased interest in recent years (Michailos et al., 2020).

The idea of using electricity to hydrolyse water and the produced hydrogen will react with the CO_2 in the biogas to form methane through the biological Sabatier reaction, Eq. (7) (Götz et al., 2016).

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad \Delta H^0 = -165 \, kJ/mol$$
 (7)

Two approaches exist to produce methane via the Sabatier reaction like chemical and biological synthesis. The latter operates at much lower process conditions (e.g., temperature and pressure) and can treat biogas of less strict quality (Yentekakis & Goula, 2017).

4.5. Calculations

In general, a mass and energy balance should be calculated in order to identify and give value to all the important agents of the reactions.

4.5.1. Data collection

Annual production and emission data for Stormossen Oy are annually published and can be found at the following source (Stormossen Oy, 2018a). Nevertheless, data not found in the public domain was obtained via personal communication with Johan Saarela (Project leader at the Stormossen Oy Wte plant).

In this sense, all specific data about chemistry is extracted from ECHA, the European Chemicals Agency, as it is a reliable and trustable information source (ECHA, n.d.).

4.5.2. Mass balance

Mass balances are the most appropriate way to find the desired results. In this sense, the Sabatier equation mentioned in Chapter 4.4.1. shows the stoichiometry between the different chemical species of the main conversion process.

At first, the molar mass of all the elements in the reaction must be identified.

$$M(CO_2) = 44,01 \frac{g}{mol}$$
$$M(H_2) = 2,02 \frac{g}{mol}$$
$$M(CH_4) = 16,04 \frac{g}{mol}$$
$$M(H_2O) = 18,02 \frac{g}{mol}$$

Therefore, following the reaction mentioned before:

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad \Delta H^0 = -165 \, kJ/mol$$
 (7)

By this reaction, important data can be extracted. Knowing 1 mol of CO_2 equals 4 mols of H_2 ; 1 mol of CH_4 , and 2 mols of H_2O .

Knowing that the mass flow of CO_2 emissions is 200 Kg/h by internal data (Stormossen, 2022), the following calculations considering the IS (International System) can be done.

$$200\frac{Kg}{h} of CO_2 \cdot \frac{1000 g}{1 Kg} \cdot \frac{1 mol}{44,01 g} = 4545,45 \frac{mol}{h} of CO_2$$

Once the molar flow is calculated, other values can be found by the stoichiometric ratio.

$$4545,45\frac{mols}{h} \text{ of } CO_2 \cdot \frac{4 \text{ mol } H_2}{1 \text{ mol } CO_2} = 18181,82 \frac{mol}{h} \text{ of } H_2$$

$$4545,45 \frac{mols}{h} \text{ of } CO_2 \cdot \frac{1 \text{ mol } CH_4}{1 \text{ mol } CO_2} = 4545,45 \frac{mol}{h} \text{ of } CH_4$$

$$4545,45\frac{mols}{h} of CO_2 \cdot \frac{2 mol H_2O}{1 mol CO_2} = 9090,91 \frac{mol}{h} of H_2O$$

Therefore, in mass flow values:

$$18181,82 \frac{mol}{h} of H_2 \cdot \frac{2,02 \ g}{1 \ mol} \cdot \frac{1 \ Kg}{1000 \ g} = 36,36 \frac{Kg}{h} of H_2$$

$$4545,45 \frac{mol}{h} of \ CH_4 \cdot \frac{16,04 \ g}{1 \ mol} \cdot \frac{1 \ Kg}{1000 \ g} = 72,73 \frac{Kg}{h} of \ CH_4$$

$$9090,91 \frac{mol}{h} of \ H_2O \cdot \frac{18,02 \ g}{1 \ mol} \cdot \frac{1 \ Kg}{1000 \ g} = 163,64 \frac{Kg}{h} of \ H_2O$$

To prove the results are right, the sum of each mass flow must be made on each side of the reaction.

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \tag{7}$$

$$200 \ \frac{Kg}{h} + 36,36 \ \frac{Kg}{h} = 72,73 \ \frac{Kg}{h} + 163,64 \ \frac{Kg}{h}$$

$$236,36 \ \frac{Kg}{h} \approx \ 236,37 \ \frac{Kg}{h}$$

As observable, both values (236,36) and (236,37) are nearly identic, verifying in this way, the correct procedure of the calculations.

Knowing these values, the determination of how much oxygen is produced and how much water is used in the electrolyser can also be determined, following the reaction mentioned before in Chapter 4.2.1.

$$M(O_2) = 31,98 \frac{g}{mol}$$
 $H_2O \rightarrow H_2 + \frac{1}{2}O_2$ (3)

$$18181,82 \frac{mol}{h} of H_2 \cdot \frac{\frac{1}{2} mol O_2}{1 mol H_2} \cdot \frac{31,98 g}{1 mol} \cdot \frac{1 Kg}{1000 g} = 290,00 \frac{Kg}{h} of O_2$$

$$18181,82 \ \frac{mol}{h} \ of \ H_2 \cdot \frac{1 \ mol \ H_2 O}{1 \ mol \ H_2} \cdot \frac{18,02 \ g}{1 \ mol} \cdot \frac{1 \ Kg}{1000 \ g} = 327,64 \ \frac{Kg}{h} \ of \ H_2 O$$

4.5.3. Energy balance

Therefore, the former data can be developed as energy flows, by using the specific energy of the compounds. This value represents the energy per unit of mass, also called gravimetric energy density, it can be calculated by the quotient of the standard enthalpy of combustion and the molar mass of the desired compound.

Therefore, taking the lower heating value (Billy Wan, 2004; Michelle Fung, 2005):

Specific energy of $H_2 = 120,0 \text{ MJ/kg}$ Specific energy of $CH_4 = 50,0 \text{ MJ/kg}$

$$36,36 \frac{Kg}{h} of H_2 \cdot \frac{120,0 MJ}{1 Kg} = 4363,2 \frac{MJ}{h} of H_2$$

This value would be correct if the electrolyser efficiency was 100%. Nevertheless, the literature states that PEM electrolysers work with an efficiency of around 60-70% (Barbir, 2004; Carmo et al., 2013; Hernández-Gómez et al., 2020). Therefore, the proper calculation must be done, taking by reference the lower efficiency value found, of 0,60.

$$\frac{4363.2 \frac{MJ}{h} of H_2}{0,60} = 7272 \frac{MJ}{h} of H_2$$

7272
$$\frac{MJ}{h}$$
 of $H_2 \cdot \frac{2,78 \cdot 10^{-4} MW}{1 MJ} = 2,02 MW of H_2$

Therefore, the electricity demand would be at least 7271,67 MJ/h, which is about 2 MW. Moreover, by the application of the previous conversion formula.

72,73
$$\frac{Kg}{h}$$
 of $CH_4 \cdot \frac{50,0 MJ}{1 Kg} = 3636,5 \frac{MJ}{h}$ of CH_4

A total of 3636,5 MJ/h of CH_4 would be generated with this reconversion method of CO_2 with H_2 .

$$3636,5 \ \frac{MJ}{h} \ of CH_4 \cdot \frac{2,78 \cdot 10^{-4} \ MW}{1 \ MJ} = 1,01 \ MW \ of \ CH_4$$

Being around 1 MW

Moreover, by using the previous energy formula from chapter 3.2.1. and isolating the energy demand, the consumption power of the electrolyser can be determined.

$$M_{H_2} = \frac{E_{ED} \cdot \eta_{rec}}{E_{ez}} \tag{4}$$

$$E_{ED} = \frac{E_{ez}}{M_{H_2 \cdot \eta_{rec}}} = \frac{54 \frac{kg}{kg}}{36,36 \frac{kg}{h} \cdot 0,9} = 1,65 \, kWh = 5,9 \, MJ$$

The value of how much power the electrolyser should receive is displayed, as it should get 1,65 kWh, which is the same as 5,9 MJ.

On the other hand, the methanation reaction is quite exothermic. Supposing a volumetric flow velocity of the gas of 5000 h⁻¹ and a total CO_2 conversion of about 2 MW heat per m³, the catalyst bed needs to be removed. Respectively, chemical methanisation has a significant issue, as good temperature control in the reactor in order to prevent thermodynamic limitation and catalyst sintering has to be ensured (Ridzuan et al., 2022). Nevertheless, the biological methanation process, known as the Sabatier reaction, mentioned before, acts as an anaerobic metabolic pathway where hydrogen and carbon dioxide are converted into methane, without the existence of the former problem. In the Sankey diagram observable in Figure 17, which is adapted from (Götz et al., 2016), two improvement potentials for P2G can be identified.



Figure 17. Sankey diagram of the P2G process efficiency.

In this sense, not only water electrolysis efficiency can be improved but also the heat from the methanation process could be used so less heat gets lost during the process. The main advantages then as part of the chain value would imply the possibility of using the heat from methanation and the oxygen from the electrolysis (Miltner et al., 2012; Varone & Cremonese, 2022).

5. Legislation

Legislation is, as far as important as all the calculations and research behind a project. In the end, it's the obligated regulations that must be followed in order to develop correctly the desired industrial activity. In this sense, the focus of this chapter is to take a look at the actual GHG restrictions and a little discussion of what is expected for the near future.

5.1. European regulations

This specific industrial activity can be defined as the production of "Natural gas and biomethane for use in transport and biomethane for injection in the natural gas network". It is based on the European Standard EN 16723-1, approved on 16 September 2016, as observable in *Appendix I*.

In it, a set of quality specifications for biomethane to be used as a fuel for vehicle engines and to be injected into natural gas pipelines is carefully described and standardized. This aims to increase the security of the energy supply, as well as to contribute to reducing the GHG emissions accepted by the European Union. Moreover, a special focus is set on the development and use of energy from renewable sources in this context.

Values as the max limit of total volatile silicon (0,3 to 1 mg/m³) and the max amount of CO (0,1% mol), NH_3 (10 mg/m³) and Amine (10 mg/m³) are defined strictly. Furthermore, the methodology of data collection is clearly specified for taking measurements of the different parameters mentioned and a risk assessment table is presented with the designated control procedures.

5.2. GHG emissions

In this sense, Finland has a clear and defined strategy to stick to in the following years. Finland's long-term strategy (LTS) is based on the agreement of the twenty-first session of the Parties to the United Nations Framework Convention on Climate Change in Paris, 30 November to 11 December 2015, also known as the 'Paris Agreement' (United Nations, 2015).

Finland's long-term strategy, which can be found in *Appendix II* lays out scenarios and impact assessments concerning the national carbon neutrality target set for 2035 and developments in greenhouse gas (GHG) emissions and removals by 2050 (Ministry of Economic Affairs and Employment, 2005). In it, the use of biological products is highlighted in the results of the low-emission scenarios in terms of biogas production.

The use of municipal waste for energy is expected to remain at the current level of about 20 PJ (representing about 6 TWh) by 2030 in all of the scenarios, as can be observed in Figure 18, and start declining slightly thereafter as a result of enhanced recycling (Ranta et al., 2021).



Figure 18. GHG emissions from waste management in Finland.

The system model includes the low-emission scenarios of methane emissions from biological treatments of solid waste to decline by 15%. As a combined effect of these, the level of emissions from waste treatment over the 2035–2050 period is about 0.1 Mt CO₂eq lower under the low-emission scenarios than under the WEM (with existing measures) scenario.

In addition, waste management emissions are also expected to be reduced to some extent by improving biological waste treatment processes, but additional measures have quite limited impacts on the overall GHG balance.

Overall, the investments required to achieve the climate neutrality objective particularly concern the industry sector. Therefore, several studies of great importance have been developed to reach this goal. For example, an investment survey 1/2020 published by the Confederation of Finnish Industries (EK) suggested that industries were expected to invest a total of about 10 billion euros in 2020 to accomplish this objective (Karoliina Rasi, 2022).

6. Results and discussion

6.1. System design and integration

The current system consists of a methane production unit that emits CO_2 into the atmosphere. Therefore, a method for its conversion with the hydrogen stored must be designed. This process can be outlined as shown in Figure 19.



Figure 19. Diagram of the biomethane production process with the CO_2 conversion method with the calculated values.

In it, the different mass flows, calculated previously in chapter 4.5.2., are observed. Therefore, this system creates a close cycle for the biowaste by using the main process CO_2 emissions to ensure another conversion process with the use of green hydrogen and later reintroduction of the biomethane.

The newly defined system can be therefore installed as mentioned, by the addition of the several parts of the new conversion process, which are basically the electrolyser, the hydrogen storage tank and the converter reactor. For the PEM electrolysis cell, literature shows the major components are membrane electrode assemblies (MEAs), current collectors (gas diffusion layers), and separator plates (Office of Energy Efficiency and Renewable Energy, n.d.).

6.2. Products valorisation

So far, all the elements that are considered in the reactions and processes from the new system, have their valorisation. Considering Figure 19, from Chapter 6.1. the only outputs are biomethane, water and oxygen; without taking into account heat, which was previously discussed in 4.5.3.

Biomethane is reintroduced in the main production process, achieving an improvement in the whole biomethane production system capacity without affecting the original bioreactor. This would represent an increase of 1,01 MW or 3636,5 MJ/h, as calculated in chapter 4.5.3 accomplishing the main aim of the thesis. Moreover, water from the conversion process can be used in electrolysis, as it is the main reactant. This substance should go through a cleaning process in order to eliminate possible impurities that could have remained on it. Therefore, a decrease in the need for water consumption would be achieved.

Last but not least, literature states no O_2 valorisation has generally been carried out in P2G facilities (Laurent Lardon et al., 2018). However, several options for O_2 valorisation exist such as oxycombustion in power plants, other gasification processes, medical care, or plasma industrial treatments (Zhu et al., 2020). Nevertheless, O_2 could also be used for the primary desulphurisation of biogas as some literature suggests (Ahamparam, S. & Stephen Harrison, 2012); or used in Stormossen Oy's reject water treatment plant, where compressors put air (O_2) to the microorganisms to control the nitrification process. As O_2 is usually transported in its liquid form, its liquefaction would give extra costs for the new system taking into account that O_2 would be used at the electrolysers location. Another key aspect of O_2 valorisation is the intermittency of the electrolyser, as O_2 would only be produced when it is in operation.

6.3. Financial impact

An analysis of electricity prices must be done to guarantee the viability of this procedure. As this is one key factor that determines the viability of the newly designed system due to the electrolysis process for the production of hydrogen, which requires electrical power as mentioned and calculated before in Chapter 4.5.3. Therefore, a study of whether electricity prices are at an affordable price or not can be done, determining a cheap price of around 25-30€/MWh, which would be considered acceptable due to the gain/losses value.

This can be achieved by comparing data from previous years in Nord Pool, a company that offers intraday power market data in Europe (*Market Data | Nord Pool*, n.d.). But focusing on Finland as it is where Stormossen Oy's plant is located, an excel spreadsheet is created in order to get a clearer vision of the electricity value.



Figure 20. Weekly price of electicity in Finland from years 2020 (green), 2021 (black) and 2022 (blue).

By observing Figure 20 and focusing on the black line which represents the year 2021, a big fluctuation can be appreciated, due to the offer demand for electricity during the day and weeks. These numbers can be found in a more detailed way in Appendix III, but by observing the graphic we get an idea of how electricity prices have been gradually increasing since 2020.

Ideally, this must have been made hourly, but the webpage does not allow data to show hourly during a whole year, therefore daily values are shown. Therefore, a simplified calculation is undertaken in excel, the full table can be found in *Appendix IV*.

MONTH	DIC	NOV	ОСТ	SEPT	AUG	JUL	JUN	MAY	APR	MAR	FEB	TOTAL
	2021	2021	2021	2021	2021	2021	2021	2021	2021	2021	2021	
MEAN VALUE	193,38	85,90	64,85	89,27	68,20	78,76	56,16	45,94	36,76	38,32	57,13	74,06

As observable, the Table shows the different calculated mean monthly values of the electricity price. Therefore, the average annual price is determined, being around 75 €/MWh, which is significantly high. This is due to the circumstances and the big demand for electricity in the modern world (Rokicki et al., 2021).

7. Conclusion

Some challenges were discussed in this paper not only related to hydrogen storage methods and infrastructure; electrolysis methods and efficiencies; conversion methods and P2G capability.

Therefore, some statements can be made:

Electrolysis methods require a cost reduction, not only because of the high electricity prices that already make the process not the most viable one financially speaking but also in terms of reliability. On the other hand, the literature showed better PEM electrolysis performance will be achieved concerning transient operations in P2G plants. Moreover, solid oxide electrolysis, which is in a development phase, shows a big potential to carry out exothermic reactions at steady-state operation.

Additionally, as mentioned, the biological methanation has a bigger impurity tolerance, having some actual examples of P2G systems that work with bio-methanation, like The MicrobEnergy plant in Germany and the BioCat project in Denmark (Zavarkó et al., 2021). In this sense, catalytic methanation requires smaller reactor sizes for the same feed gas flow (Materazzi & Foscolo, 2019). Therefore, the use of catalytic methanation leads to higher efficiencies because of the not requirement of using a stirrer and the reusability of the waste heat. Taking everything into account, a big potential is seen in P2G, even though some economic and technical obstacles have to be solved before P2G can be successful at a commercial level (Saccani et al., 2020).

Overall, by the application of this system, a full close cycle of the initial biowaste is created, ensuring not only a CO₂ neutral system but also improving Stormossen Oy's current biomethane process by increasing its production capacity.

With regards to recommendations and suggestions for more research, this thesis states more work should be done in studying profoundly the financial impact this newly designed system would have. Moreover, analysing another available method for hydrogen production or executing a different action plan in which the new system is introduced in a different phase of the biomethane production process would be beneficial in order to compare results with the ones obtained in this paper.

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9. Appendices

Appendix I. European Standard EN 16723-1

https://standards.iteh.ai/catalog/standards/cen/55154af1-529b-407d-890cb9e16c935898/en-16723-1-2016



Appendix II. Finland's long-term low greenhouse gas emission development strategy.

https://unfccc.int/sites/default/files/resource/LTS Finland Oct2020.pdf

Ministry of Economic Affairs and Employment

Finland's long-term low greenhouse gas emission development strategy

October 2020

Unofficial translation

Appendix III. Nordpool 2021 Daily Electricity Prices Finland.

<u>https://www.nordpoolgroup.com/en/Market-data1/Dayahead/Area-</u> <u>Prices/FI/Daily/?view=table</u>

EUR/MWh

	Dec-21	Nov-21	Oct-21	Sep-21	Aug-21	Jul-21	Jun-21	May-21	Apr-21	Mar-21	Feb-21
01	167,91	53,81	61,89	74,36	53,97	82,72	59,91	46,56	35,19	34,34	86,93
02	196,43	55,65	44,77	81,96	75,63	91,02	63,95	45,65	30,18	40,61	68,16
03	178,47	72,24	12,56	67,67	56,78	68,44	56,69	60,72	20,57	41,04	54,97
04	163,59	81,45	56,93	66,08	68,29	63,66	58,46	62,31	10,46	45,25	60,19
05	148,28	56,80	64,74	70,51	75,22	82,01	42,59	45,52	7,10	53,40	79,38
06	290,06	13,85	69,23	98,37	71,06	81,14	41,04	61,04	32,43	22,13	45,60
07	469,03	21,30	94,89	73,22	52,59	90,98	65,96	65,54	46,75	26,94	40,66
08	439,69	107,68	100,08	62,20	26,08	77,70	66,47	47,42	48,99	56,13	59,13
09	267,44	55,90	79,14	88,74	78,88	87,41	65,55	25,94	20,02	54,57	71,74
10	212,45	29,90	34,47	100,07	82,33	76,76	69,60	43,13	19,88	63,74	73,59
11	178,24	24,50	81,33	92,90	83,96	67,92	57,98	41,36	26,10	35,00	99,72
12	167,90	69,17	117,84	94,90	89,34	92,31	31,64	46,67	40,77	35,54	82,48
13	205,26	47,94	157,57	70,44	77,03	98,29	15,99	35,32	49,72	29,79	53,22
14	193,07	54,27	73,04	90,02	58,53	108,47	65,80	38,41	56,59	31,88	46,50
15	79,08	67,48	40,74	138,62	56,60	102,78	63,43	21,61	53,95	53,02	67,99
16	121,87	63,96	9,10	123,92	79,96	89,51	85,06	29,65	47,89	53,25	63,70
17	152,56	47,75	47,29	103,23	68,26	67,32	70,09	59,68	26,49	50,16	79,56
18	83,05	59,05	94,13	85,26	53,32	36,96	57,01	61,60	25,08	48,92	68,69
19	66,06	72,47	139,25	78,95	75,22	81,81	41,61	58,99	48,12	51,27	60,55
20	269,82	15,04	39,02	122,11	69,91	73,42	33,61	46,17	47,95	22,90	47,57
21	405,52	61,99	60,77	114,02	51,62	69,12	47,49	35,05	30,63	18,44	35,03
22	360,78	139,44	41,76	94,84	52,82	58,52	71,48	34,78	32,64	48,17	46,17
23	175,43	129,12	75,99	61,35	66,24	77,68	75,69	21,48	25,13	35,96	41,47
24	136,51	104,58	31,08	78,20	92,76	69,67	62,85	54,63	17,91	30,03	37,44
25	128,97	108,65	78,68	77,90	58,90	66,17	33,61	57,88	16,28	43,29	33,35
26	154,19	165,59	71,21	80,41	64,20	83,59	31,02	46,64	56,75	39,19	34,58
27	158,04	175,08	90,48	88,05	71,40	87,30	31,41	54,24	61,30	18,50	32,52
28	131,90	188,09	28,47	97,68	57,54	84,04	70,28	55,17	60,49	19,49	28,74
29	154,86	263,96	40,62	107,89	63,81	84,76	80,01	42,26	56,05	23,50	
30	73,35	170,20	20,11	94,31	95,73	76,51	68,41	28,31	51,49	27,12	
31	65,04		53,31		86,17	63,48		50,54		34,46	
	Dec-21	Nov-21	Oct-21	Sep-21	Aug-21	Jul-21	Jun-21	May-21	Apr-21	Mar-21	Feb-21

Appendix IV. Electricity prices data calculations developed in a Microsoft Excel spreadsheet.

	Dec-21	Nov-21	Oct-21	Sept-21	Aug-21	Jul-21	Jun-21	May-21	Apr-21	Mar-21	Feb-21	
1	167,91	53,81	61,89	74,36	53,97	82,72	59,91	46,56	35,19	34,34	86,93	
2	196,43	55,65	44,77	81,96	75,63	91,02	63,95	45,65	30,18	40,61	68,16	
3	178,47	72,24	12,56	67,67	56,78	68,44	56,69	60,72	20,57	41,04	54,97	
4	163,59	81,45	56,93	66,08	68,29	63,66	58,46	62,31	10,46	45,25	60,19	
5	148,28	56,8	64,74	70,51	75,22	82,01	42,59	45,52	7,1	53,4	79,38	
6	290,06	13,85	69,23	98,37	71,06	81,14	41,04	61,04	32,43	22,13	45,6	
7	469,03	21,3	94,89	73,22	52,59	90,98	65,96	65,54	46,75	26,94	40,66	
8	439,69	107,68	100,08	62,2	26,08	77,7	66,47	47,42	48,99	56,13	59,13	
9	267,44	55,9	79,14	88,74	78,88	87,41	65,55	25,94	20,02	54,57	71,74	
10	212,45	29,9	34,47	100,07	82,33	76,76	69,6	43,13	19,88	63,74	73,59	
11	178,24	24,5	81,33	92,9	83,96	67,92	57,98	41,36	26,1	35	99,72	
12	167,9	69,17	117,84	94,9	89,34	92,31	31,64	46,67	40,77	35,54	82,48	
13	205,26	47,94	157,57	70,44	77,03	98,29	15,99	35,32	49,72	29,79	53,22	
14	193,07	54,27	73,04	90,02	58,53	108,47	65,8	38,41	56,59	31,88	46,5	
15	79,08	67,48	40,74	138,62	56,6	102,78	63,43	21,61	53,95	53,02	67,99	
16	121,87	63,96	9,1	123,92	79,96	89,51	85,06	29,65	47,89	53,25	63,7	
17	152,56	47,75	47,29	103,23	68,26	67,32	70,09	59,68	26,49	50,16	79,56	
18	83,05	59,05	94,13	85,26	53,32	36,96	57,01	61,6	25,08	48,92	68,69	
19	66,06	72,47	139,25	78,95	75,22	81,81	41,61	58,99	48,12	51,27	60,55	
20	269,82	15,04	39,02	122,11	69,91	73,42	33,61	46,17	47,95	22,9	47,57	
21	405,52	61,99	60,77	114,02	51,62	69,12	47,49	35,05	30,63	18,44	35,03	
22	360,78	139,44	41,76	94,84	52,82	58,52	71,48	34,78	32,64	48,17	46,17	
23	175,43	129,12	75,99	61,35	66,24	77,68	75,69	21,48	25,13	35,96	41,47	
24	136,51	104,58	31,08	78,2	92,76	69,67	62,85	54,63	17,91	30,03	37,44	
25	128,97	108,65	78,68	77,9	58,9	66,17	33,61	57,88	16,28	43,29	33,35	
26	154,19	165,59	71,21	80,41	64,2	83,59	31,02	46,64	56,75	39,19	34,58	
27	158,04	175,08	90,48	88,05	71,4	87,3	31,41	54,24	61,3	18,5	32,52	
28	131,9	188,09	28,47	97,68	57,54	84,04	70,28	55,17	60,49	19,49	28,74	
29	154,86	263,96	40,62	107,89	63,81	84,76	80,01	42,26	56,05	23,5		
30	73,35	170,2	20,11	94,31	95,73	76,51	68,41	28,31	51,49	27,12		
31	65,04		53,31		86,17	63,48		50,54		34,46		
Mean	193,38	85,90	64,85	89,27	68,20	78,76	56,16	45,94	36,76	38,32	57,13	74,06
	Dec-21	Nov-21	Oct-21	Sept-21	Aug-21	Jul-21	Jun-21	May-21	Apr-21	Mar-21	Feb-21	