MAPPING AVAILABLE EU BIO-CO_2 SOURCES AS FEEDSTOCK FOR METHANOL SYNTHESIS

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ABSTRACT: The scope of this work is to provide a mapping of available biogenic CO_2 sources to produce methanol that could eventually be used as marine fuel. In contrast to fossil CO_2 emissions, biogenic CO_2 does not add additional stress to the carbon cycle and is therefore investigated as a feedstock for methanol synthesis. In this work, the main investigated sources of biogenic CO_2 are biogas, bioethanol, food and beverage, pulp and paper, as well as biomass combustion plants. Furthermore, indications on the points of generation within the plant, the geographical distribution of the plants in Europe, indicative stream compositions/impurities in the bio-CO_2 stream as well as potential seasonality aspects of each sector. The influence of the contained impurities in the sources is assessed, whereas the current uses of biogenic CO_2 and the total amount of the biogenic CO_2 produced are also included to the study.

Keywords: carbon dioxide (CO_2), CO_2 emissions, fuel, methanol, maritime

1 INTRODUCTION

The International Maritime Organization (IMO) reports that the shipping industry is responsible for about 3% of global CO_2 emissions. In 2018, the IMO approved an initial strategy to reduce greenhouse gas (GHG) emissions from ships, aiming to cut annual GHG emissions from international shipping by at least 50% by 2050 compared to 2008 levels, which reflects the industry's acknowledgment of the importance of addressing CO_2 emissions. Currently, the maritime sector relies mostly on various types of fossil fuels for propulsion and power generation. The choice of fuel in maritime operations is influenced by factors such as operational patterns, vessel size and type, regulatory requirements, and fuel availability. Commonly used fuels include heavy fuel oil, marine diesel oil, and marine gas oil. Alternative fuels include methanol, liquefied natural gas, liquefied petroleum gas, biofuels, ammonia, and hydrogen. In some cases, these alternative fuels can be blended with conventional fuels to improve ignition performance, known as "dual fuels" [1].

Some alternative fuels, such as methanol and liquefied natural gas, are produced from carbon sources. Ships using these carbon-based alternative fuels may face emission charges, under the European Union's Emissions Trading System. To this end, vessels using carbon-based fuels must purchase carbon credits to compensate for emissions during voyages to and from Europe, regardless of their starting point. Although these fuels may emit less carbon emissions than conventional fossil fuels, they are often viewed as temporary solutions [2].

Biogenic CO_2 emissions originate naturally within the carbon cycle since they involve organic materials such as biomass utilized in processes, such as combustion, fermentation, or digestion. These differ from fossil CO_2 emissions, which are derived from burning fossil fuels and release carbon that has been stored underground for millions of years.

Methanol (MeOH, CH_3OH) is the simplest alcohol, attracting significant interest for its potential as both an energy source and a hydrogen carrier. Methanol is soluble in water, easily degradable, and liquid at atmospheric conditions make it easier to store and transport compared to gaseous fuels like hydrogen Existing fueling facilities can store MeOH with minimal modifications to their infrastructure [3]. Thus, using methanol requires relatively small investments to adapt storage, transportation, and refueling infrastructure, unlike hydrogen-based transportation fuels [3]. Currently, methanol is available at over 120 ports worldwide, including European ports such as Antwerp and Rotterdam. While it is primarily used as a feedstock, its main production comes from fossil sources like natural gas and coal, highlighting the need for environmentally-friendly and renewable methanol production methods [4,5].

For methanol to be adopted globally as marine fuel, it must be safely bunkered to ships and supported by a global network of bunkering facilities at numerous ports [5]. Shipowners must also be assured of the long-term availability and economic viability of switching to this fuel. Ensuring the availability of green maritime methanol fuel at ports requires sufficient amounts of carbonaceous feedstocks and renewable hydrogen at reasonable costs. Estimating total bio-CO_2 emissions in the EU is highly uncertain due to the diverse bio-CO_2 sectors and varying emission reporting data from different countries and plants, with differing estimation methodologies across reports [6]. Despite these challenges, critical aspects include the technical and economic feasibility of capturing and utilizing this CO_2 for producing high-value products like methanol.

To address this, the EU funded project M^{2}ARE (Grant Agreement Nr. 101136080) is exploring the use of bio-CO_2 resources combined with renewable hydrogen to produce methanol specifically for maritime use. By producing methanol with bio-CO_2 instead of fossil-derived CO_2 and incorporating renewable hydrogen, it is expected that this approach will eventually do not impose additional stress on the carbon cycle. This report is focused on conducting a mapping of the availability and locations of different bio-CO_2 streams across Europe and assessing their suitability as feedstock for methanol synthesis.

This work is organized as follows. Section 2 describes the potential impurities of CO_2 streams, the different CO_2 capture, storage, separation and transportation methods as well as the methodology of the mapping of bio-CO_2 sources that has been followed in this work. Section 3 shows the results of the investigated biogenic CO_2 sources: biomass power plants, pulp and paper plants, biogas plants, bioethanol plants and food and beverage industry. Finally, Section 4 shows the conclusions and discussions of this work.

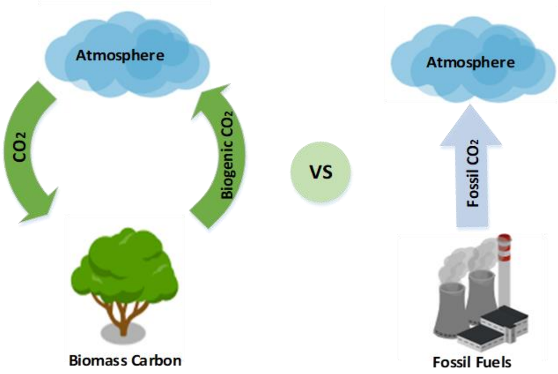

Figure 1: Biogenic and fossil-based carbon cycle

2 BIOGENIC CO_2 OVERVIEW AND METHODOLOGY

2.1 Biomass conversion processes

Bio-CO_2 is produced as a by-product from various biomass conversion technologies. In the sectors examined in this study, bio-CO_2 and the associated biogenic carbon emissions are not the primary products but rather by-products that often remain unutilized and pose an environmental burden. The main biomass conversion technologies that generate bio-CO_2 can be classified into three main categories: combustion, digestion and fermentation.

The biomass feedstock could be combusted for the production of heat, electricity or combined heat and power production. Biomass combustion is the main CO_2 emitting process in biomass power plants and pulp and paper plants, which use biomass or bio-based by-products for heat and power production [7]. The related bio-CO_2 streams, due to the combustion process will involve low CO_2 contents (<15 wt.%), with high inert contents mostly nitrogen, oxygen due to the incomplete combustion process as well as NO_x, SO_x and other traces inherent to the combustion process and the biomass feedstock

Biomass digestion, or anaerobic digestion, is a biochemical process that decomposes organic materials, like biomass, organic/domestic waste, manures, and sludges, in the absence of oxygen to produce biogas and nutrient-rich digestate [7]. This process is well established, with industrial-scale biogas and anaerobic digestion facilities operating in Europe and globally.

Biomass fermentation is a biochemical process where microorganisms decompose organic materials, such as sugars or starches, to produce useful products like bioethanol. This process includes pretreatment to break down the biomass structure, followed by hydrolysis to produce fermentable sugars, which are then converted into ethanol. Widely used in the bioethanol industry and the food and beverage industry for products like wine and beer, the fermentation process generates a significant amount of pure CO_2 as a by-product. The stoichiometric equation of ethanol fermentation is: $C_6H_{12}O_6 \rightarrow 2\ C_2H_5OH + 2\ CO_2$ [8].

2.2 Potential impurities

To examine the potential impurities that might be included in the bio-CO_2 feedstock that will be used for methanol synthesis, it is essential to examine the different biomass feedstocks that were initially used in the upstream processes. Impurities and components that are contained in the biomass feedstock could be also contained in the off-gases of those processes, in trace contents as well as at different forms/compounds. Most common impurities are moisture, sulfur-containing compounds, such as SO_2, H_2S and COS, Nitrogen compounds such as ammonia, hydrogen cyanide nitrogen oxides (NO_x), halides like HCl, HF, and HBr, which are possible of forming toxic by-products like polyhalogenated dioxins and furans [9]. Off-gases may also contain siloxanes, which are common impurities for biogas plants, oxygen, trace elements and heavy metals, as Zn, Pb, Hg, Cd, Pb, and Cr. All these elements and compounds could poison the methanol synthesis catalyst and lead to catalyst deactivation. In addition, they could contribute to corrosive effects on the equipment and they are hazardous to human health and the environment. As a result, further gas purification processes are necessary before utilization of the bio-CO_2 streams.

2.3 CO_2 capture and separation techniques

Three primary methods of CO_2 capture can be employed to reduce CO_2 emissions: post-combustion CO_2 capture, pre-combustion CO_2 capture, and oxy-fuel combustion. For all capture methods (pre-, post-combustion and oxy-combustion), similar CO_2 separation technologies can be used to isolate the CO_2 from the flue gas stream [10]. Many separation technologies are commercially available for separating CO_2 from gas mixtures. The selection of the most appropriate technology depends on the required purity of the product and the conditions of the gas stream being treated, including CO_2 pressure, temperature, concentration, and the presence and level of trace species or impurities.

2.4 CO_2 storage and transportation

There are several scenarios involving the storage of CO_2 containing industrial off-gases. CO_2 streams are mainly stored on-site in gas holders. CO_2 is extracted from the off-gases or available from dedicated processes, and then stored in relatively pure form. Some contaminants are removed from the stream, but additional provisions for gas cleaning are necessary. However, it is essential to clean the off-gas stream from any inhibiting species that could affect the catalyst used in methanol synthesis [6,11,12].

Carbon dioxide can be transported in different states: as a gas, liquid, dense form, or as a supercritical fluid. Typically, CO_2 is transported in liquid form for shipping, trucking, or rail transport, and as a supercritical fluid for pipelines. For large volumes exceeding one million tons per year, pipeline and maritime transport are generally deemed feasible [11]. Road transport is typically reserved for smaller volumes and shorter distances, especially when installing a pipeline is not practical. Liquid CO_2 transport occurs below the critical point and above the triple point, ranging between temperatures of -56.6 to -31°C and pressures of 5.2 to 73.8 bar [13]. The choice of transportation modes and methods is influenced by various factors, including the volume of being transported, costs involved, geographic location and terrain, and specific usage requirements.

2.5 Methodology

To categorize and map the different bio-CO_2 sectors, a series of different aspects directly related to each sector must be taken into account. The methodology of categorization will provide information regarding the following aspects that are directly linked to the suitability of those streams for MeOH synthesis:

- Bio-CO_2 emissions points in the plant: Bio-CO_2 could be generated at various points within a particular plant. This work will outline different points of generation within those plants but will focus mainly on the most feasible points of capture and utilization.
- CO_2 concentration (%) in the bio-CO_2 stream: this aspect is related to the content of CO_2 in the total bio-CO_2 streams and among others depends on the upstream process type as well as feedstock composition.
- Seasonality: certain bio-processes are operating within a certain period in a year such as grape fermentation. A wide range of biomass feedstocks are directly affected by seasonality, meaning that the annual yearly emissions of particular industries have peaks at certain seasons/months depending on each industry.
- Current uses of the bio-CO_2 stream: The bio-CO_2 stream could be stored, re-used in other applications or released to the atmosphere. This factor outlines the percentage of the stream that can be used for methanol synthesis afterwards.
- Geographic distribution: the different bio-CO_2 emitters could be geographically scattered across different locations, whereas a critical factor is the CO_2 emissions per plant. Higher total sector emissions with low emissions per plant indicates the decentralized nature of this sector, with challenges in achieving significant cost reductions through the economies of scale. This is a critical aspect in bio-CO_2 emitters in contrast to the fossil- and energy-intensive industries, that will also affect the logistics of CO_2, H_2 and MeOH transportation contributing to the final cost of methanol production.
- Potential impurities and other compounds: this refers to the potential impurities as well as other compounds that might be contained in the bio-CO_2 stream and could affect the methanol synthesis catalysts and the relevant equipment of the plant. The contained impurities will also dictate the required gas cleaning of the stream before utilization in the synthesis units
- Total sector emissions: the cumulative emissions of each bio-CO_2 sector and its contribution to the overall emissions. This value provides a first indication about the sectors that are mostly contributing to the EU bio-CO_2 emissions to assess the affect that could be achieved when applying CO_2 utilization technologies such as methanol synthesis.

3 RESULTS

This section presents the results of the investigated CO_2 sources, biomass combustion, pulp and paper, biogas, bioethanol and food and beverages industry, based on the criteria that are shown in Section 2.

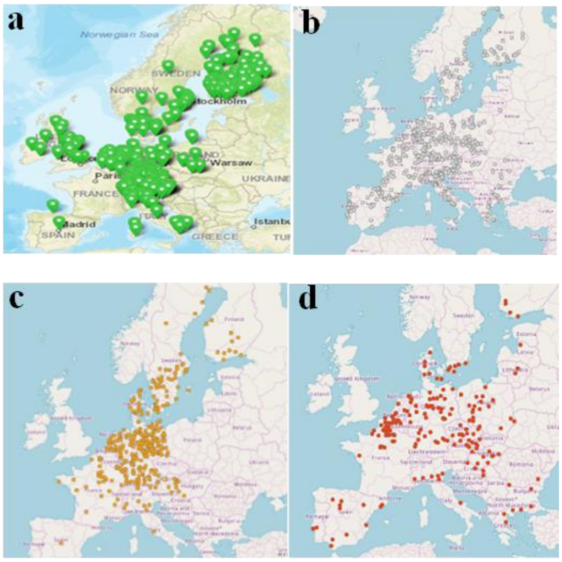

Figure 2: Geographical distribution of a) biomass power plants, b) pulp and paper plants, c) biogas plants, d) sugar & starch industry[14,15]

3.1 Biomass combustion

Biomass power plants harness the chemical energy contained in organic matter to generate electricity. A wide array of biomass feedstocks can be utilized for power production, including wood waste, agricultural residue, animal waste, and energy crops. Biomass combustion stands at the forefront for bioenergy production, occurring across both small-scale and large-scale combustion setups for heat, electricity, or Combined Heat and Power (CHP) applications. This technology is well-established commercially, with biomass being combusted in steam boilers connected to steam turbines and power generators. Biogenic CO_2 can be extracted from the combustion flue gas, where the CO_2 concentration is estimated to range from 3% to 12% [16]. For biomass power plants, seasonality could affect their operation since the availability and price of certain biomass feedstocks could vary within the year. The fuel switching flexibility of power plants could provide a solution to the seasonal fluctuation, where power plants can use a variety of locally available bio-feedstocks to produce electricity [17]. Apart from feedstock seasonality, there are additional seasonality aspects inherently linked to the operation of the power plant. For instance, regarding biomass district heating plants, the heat demand for the residential sector will vary during summer and winter affecting also the associated CO_2 emissions. In 2021, the number of large combustion plants (all fuel-based plants) in Europe is estimated to be 3,414 plants [18]. This sector illustrates a large distribution in sizes (ranging from small- to large-scale) and consequently with either small or large amounts of CO_2 emitted. The emissions from biomass plants are primarily influenced by factors such as the fuel composition, combustion process employed, and flue-gas treatment methods. The key emissions from biomass combustion include $PM_{2.5}$, PM_{10}, dust, NO_2, NO, CO, H_2S, HCHO, HCl, NH_3, SO_2, aromatic compounds and heavy metals. In total, the estimated emissions from biomass power plants for electricity and heat production in EU could range from 225.5-313.3 MT/year.

3.2 Pulp and paper

Paper is manufactured using pulp, a soft and moist material primarily composed of cellulose sourced from materials such as wood, plants, and recycled paper. Pulp can be obtained through chemical processes or by grinding the raw material and mixing it with water. This mixture of pulp, water, and chemicals is then flattened, dried, and cut into sheets and rolls to create paper. Paper serves various purposes, including writing, printing, packaging, and sanitation [19]. Chemical pulping processes, particularly the Kraft pulping method, are the predominant means of pulp production. This process leads to significant point source emissions, primarily from the combustion of lignin in the recovery boiler, which is separated from the cellulose during pulping. Other notable emission sources at pulp and paper plants include the lime kiln, where lime mud is heated to recover lime, and potentially a bark- or auxiliary boiler [20]. The recovery boiler stands out as the largest emitter of CO_2 at a Kraft pulping mill. The recovery boiler uses concentrated black liquor from the pulping process, recovering sodium-based pulping chemicals and producing biogenic CO_2 emissions. Typically, the CO_2 concentration in the recovery boiler ranges between 10-15 vol. % [21,22]. The lime kiln is usually fueled with heavy fuel oil or other fossil fuels, with CO_2 emissions originating from both the fuel itself (fossil CO_2) and the wood raw material used in the pulp mill (biogenic CO_2). The overall CO_2 concentration in the lime kiln tends to be higher, often around 20 vol. % [21]. Many pulp mills also feature an additional power boiler for combustion of materials like bark and hog fuel. The CO_2 concentration in this biomass-fired boiler is similar to that of the recovery boiler, ranging between 10-15 vol-% [22,23]. In this case, the seasonality of the feedstock does not affect the operation of plants due to the year. There is a large distribution of plants in Europe whereas pulp and paper plants are large emitters of CO_2. The key emissions from biomass combustion include SO_2, NO_2, NO, CO, H_2S, total reduced sulfur and dust. The total biogenic CO_2 emissions of the EU pulp & paper sector were estimated at 92 MT in 2022 [24].

3.3 Biogas

Biogas is produced by the microbial decomposition of biomass under anaerobic conditions. Its main components are methane, carbon dioxide and small amounts of hydrogen, nitrogen, oxygen, and hydrogen sulfide. Biogas finds extensive applications in electricity generation, heating, and fuel sectors [25], whereas various organic materials such as crops, agricultural waste, manure, biowaste and sewage sludge are used as biogas plants feedstocks. Anaerobic digestion emits significant quantities of bio-CO_2 which mostly remains unexploited after the biogas upgrading process; since methane is the main recovered product of interest and carbon dioxide is emitted to the atmosphere. Depending on the type of organic material (feedstock), the produced biogas consists of 45-85 % methane and 25-50 % carbon dioxide [26]. The seasonality has a minor impact in the biogas production and operation. According to the European Biogas Association there are around 20,000 biogas plants operating in Europe and around 1,322 of those upgrade and inject biomethane into the natural gas grid [11]. Biogas plants present a large distribution with a small amount of CO_2 per plant emitted. The main emissions of biogas production are N_2, O_2, H_2, H_2O, CO, NH_3, H_2S, HCN, HF, HCl, halogens, terpenes, BTX, siloxanes. In total, 24 MT biogenic CO_2 were released from biogas and biomethane upgrading production across Europe [27].

3.4 Bioethanol

Bioethanol stands as a renewable liquid biofuel synthesized through the fermentation of various feedstocks, including corn, soybeans, crops, wheat straw, sugar beet, sugar cane and woodchips [27]. Bioethanol can be directly utilized in vehicles, functioning in a similar manner to conventional fuels. The bio-CO_2 produced from this process is relatively pure (>99 wt.%) and would also allow a relative feasibility for capturing and reusing it. However, due to the relative purity of the bio-CO_2 emissions from this process they are reused within the same industry or sold to other FAB industries. Bioethanol plants illustrate large geographical distribution among Europe with small plant emitters. The key emissions from bioethanol plants include alcohols (propanol, butanol, amyl alcohol, glycerol, phenethyl alcohol), acids (acetic, caproic, caprylic, lactic, pyruvic, succinic), esters (ethyl acetate and any other combination of acids and alcohols), acetaldehyde, diacetyl and H_2S [28]. In total, the biogenic CO_2 emissions of bioethanol plants in Europe was 4.3 MT in 2023 [29].

3.5 Food and Beverage

Fermentation is a controlled process performed by specific microorganisms to alter the texture of foods, preserve them through acid or alcohol production, or enhance flavors and aromas. It plays a crucial role in the production of various food and beverage items, such as beer and wine (alcoholic fermentation). During alcoholic fermentation, simple sugars are broken down into alcohol by yeast, typically in an anaerobic environment at temperatures ranging from 8 to 30°C. The bio-CO_2 produced from this process is relatively pure (>99 wt.%) due to the relative purity of the bio-CO_2 emissions from this process they are reused within the same industry or sold to other FAB industries, whereas the total bio-CO_2 emissions from this sector represent a lower percentage of the total EU bio-CO_2 emissions sectors. Nevertheless, wine fermentation is widely affected from the seasonality of the feedstock. This sector's plants are large distributed across Europe with a small amount of CO_2 per plant emitted. The key emissions from wine and beer fermentation plants include alcohols (propanol, butanol, amyl alcohol, glycerol, phenethyl alcohol), acids (acetic, caproic, caprylic, lactic, pyruvic, succinic), esters (Ethyl acetate and any other combination of acids and alcohols), acetaldehyde, diacetyl and H_2S [28]. In total, the biogenic CO_2 emissions of food and beverage plants in Europe was 2.8 MT in 2023.

4 CONCLUSIONS

This study involved the mapping of various EU bio-CO_2 sources according to their suitability for methanol synthesis. This study is focused on biomass conversion processes that emit bio-CO_2, such as combustion, fermentation, and digestion. The bio-CO_2 sectors investigated in this report are biomass combustion, biogas, pulp & paper, bioethanol, and the food & beverages industry. Each sector was analyzed for CO_2 purity, total emissions, seasonality, impurities, geographic distribution/plant size, and current CO_2 uses. The study's results indicate that the bio-CO_2 sector is highly distributed, utilizing diverse feedstocks that produce varying off-gas amounts and compositions. For example, biogas upgrading results in pure, already separated CO_2, not used in other processes but with highly dispersed locations, making it a promising sector requiring special attention to feedstock logistics. Biomass power plants and pulp and paper plants are less distributed but are large bio-CO_2 emitters with highly diluted CO_2 streams, necessitating CO_2 capture technologies, which could increase methanol production costs. Fermentation-related sectors, such as bioethanol and the food and beverages industries, produce highly pure bio-CO_2 streams ready for use but represent a smaller percentage of total bio-CO_2 emissions. In some cases, this stream is reused within the same industry or sold as a market product, indicating a lower potential for methanol synthesis. A critical concern is that impurities in the biomass feedstock can be present in those off-gases, potentially damaging the methanol synthesis catalyst and to this end, gas cleaning methods are required.

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