Assessing Biogenic Carbon Dioxide Potentials in Europe for Valorisation

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ABSTRACT

This study investigates the theoretical potential and limitations of green carbon dioxide sources for technical valorisation approaches. The emission of greenhouse gases, especially CO_2, must be rigorously reduced in order to achieve the European and global climate objectives. As CO_2 is an increasingly valuable resource for industries and new disrupting technologies on CO_2 utilization, the potential of CO_2 obtained from different green and fossil sources in Europe is discussed for a comparative evaluation. Biogenic or green and fossil CO_2 sources are classified according to their emitting processes and industry sectors, respectively. The CO_2 potentials are then calculated from statistical data for CO_2 generating processes in Europe, complemented and verified by relevant papers and reports. This study demonstrates the European potential of capturing and utilizing the biogenic and fossil CO_2. In Europe, 69.7 Mt/a CO_2 are estimated to be produced by biogas upgrading, biogas combustion, as well as bioethanol and other fermentation processes. Additionally, 437 Mt/a CO_2 are produced by solid biomass combustion. This accounts for a theoretical potential of 506.7 Mt/a CO_2 currently available, which is nearly seven times the amount of the current European industrial CO_2 demand. The CO_2 from biomass combustion is more difficult to capture and is mixed with impurities, which potentially reduces its technical and economic potential, whereas the 63 Mt/a from other high-purity sources are already partially utilized, e.g., by breweries or dry ice producers.

KEYWORDS

Carbon Capture and Utilization (CCU); Green CO_2; Fossil CO_2, Stationary emissions, Potential analysis

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ABBREVIATIONS

Carbon Capture and Utilisation (CCU); Carbon Capture Storage (CCS); biogenic CCU (bio-CCU); Intergovernmental Panel on Climate Change (IPCC); Direct Air Capture (DAC); polyurethane (PUR); food and beverage (FAB); CO_2 Capture Project (CCP); European Industrial Gases Association (EIGA); International Society of Beverage Technologists (ISBT); European Biogas Association (EBA); pressure swing adsorption (PSA); biological compressed natural gas (bio-CNG); metal-organic frameworks (MOFs); European Union (EU); EU emission trading system (EU ETS); World Biogas Association (WBA); Gas Infrastructure Europe (GIE); combined heat and power (CHP)

1 INTRODUCTION

Carbon dioxide (CO_2) is mainly seen as a global hazard due to its properties as a greenhouse gas (GHG). In fact, it is also a valuable resource for various state-of-the-art and innovative technologies and processes. Carbon Capture and Utilisation (CCU) technologies are a way to reduce carbon emissions while tapping into this resource. This paper investigates the theoretical mass potential and limitations of “green” CO_2 sources for the implementation of utilization options in CCU. Many studies explored the technical opportunities for CCU [1–4], emphasizing the disruptive potential of some of the potential future applications [5]. Possible CCU pathways can be categorized based on the energy supply for activating the stable CO_2, the various synthesis processes or the marketable products. (Figure 1-1).

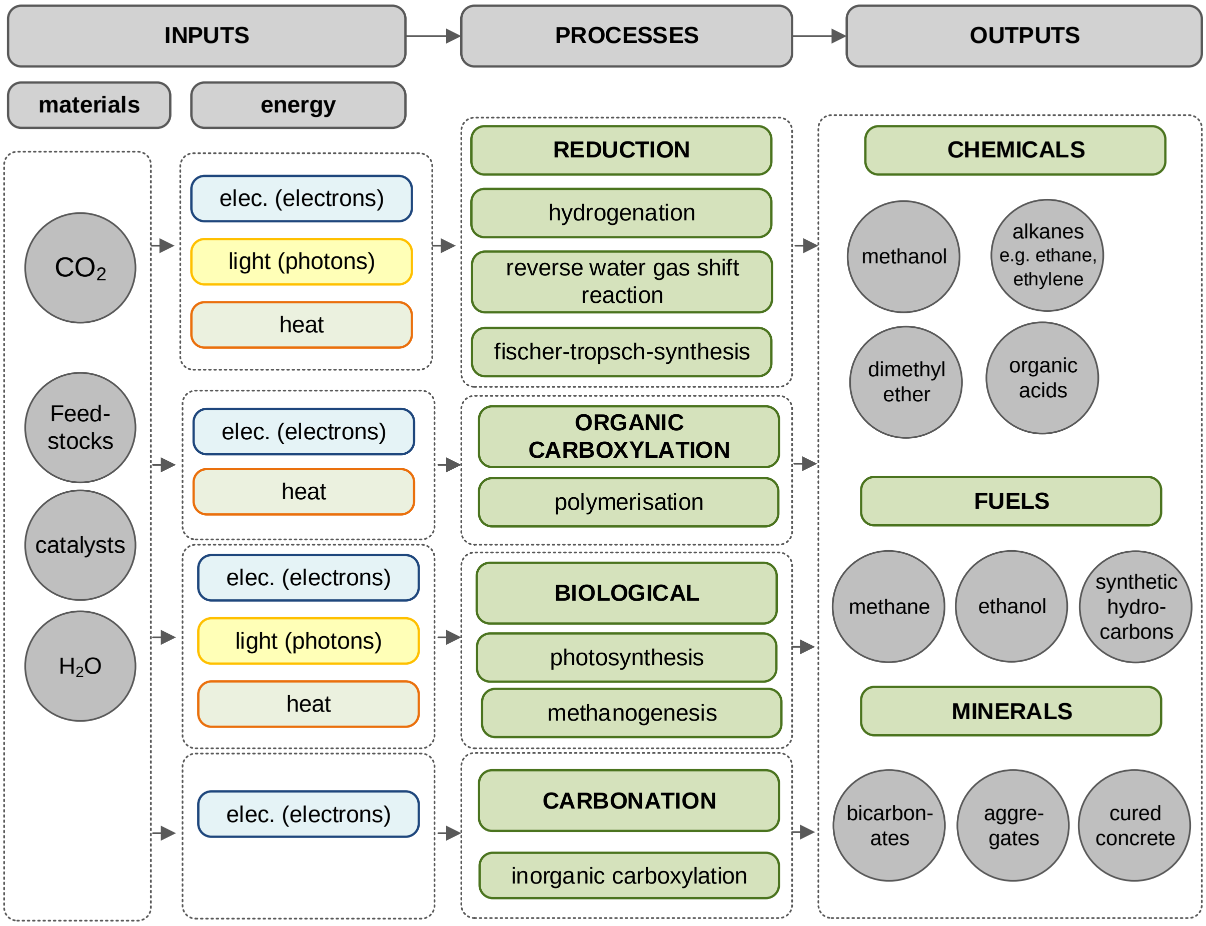

Figure 1-1: Classification of potential Carbon Capture and Utilisation pathways.

To ensure the climate neutrality of CCU products, the usage of biogenic CO_2 is preferred. Hence, the authors mainly focus on the use of already available biogenic CO_2 from industrial sources, as it has been defined as carbon neutral to the environment [6].

Here, quantifications of biogenic and fossil CO_2 sources are discussed and compared to the biogenic renewable sources based on the available quantities and qualities of CO_2. Processes involving direct CO_2 capture from the air are not the focus of this study, as the required processes are relatively energy and cost intensive at the current development stage [7].

1.1 *Literature review*

Publication history of studies on biogenic and fossil carbon capture

Since the 1990s, CCU and Carbon Capture and Storage (CCS) have become the centre of increasing scientific attention. A short key word search on “Carbon Capture” on sciencedirect.com shows an increase from just over 2,800 items in 1996 to more than 28,000 in August 2019. Within the subject, the possible methods to capture CO_2 from different sources is the most comprehensively discussed topic in scientific publications, followed by the storage and utilization technologies, fossil CO_2 emitting sources, such as the transport sector and the chemical industry and their CO_2 potential. Biogenic Carbon Capture is mentioned much less frequently: search results for “Biogenic Carbon Capture” increased from nearly 98 in 1996 to over 800 in August 2019; for “Bio-CCS”, results range from 34 to roughly 400 in the same time period. The lower frequency of studies on Bio-CCS can possibly be attributed to the concentration of biogenic CO_2 sources in just a few countries, such as a considerable number of biogas plants in Germany [8]. Another reason for the relatively low research interest in biogenic CCS could be the comparatively simple, far-advanced and low-cost capture and purification process of biogas. Most papers deal with case studies or specific Carbon Capture, Carbon Storage or Carbon Utilization pathways. Moreover, these studies often include new processes, which allow the use of CO_2 as a feedstock material or investigate methods of efficiently capturing CO_2 from fossil sources.

However, the available amount of CO_2 from fossil sources or from primarily biogenic sources at the European or global scale is seldom a core topic [9–11]. More often, the current and future CO_2 demand is discussed [12–16]. Comparisons of future CO_2 potential (theoretical, technical and economical) from biogenic and fossil sources, together with the potential CO_2 demand, is out of the scope of most publications. Billig et al. and Horschig et al. [12,17] compared the current biogenic CO_2 supply in Germany with the future demand, although this only accounts for one country’s CO_2 potential. Thus, there is a lack of studies on the holistic potential and sources of CO_2 at the European and global scale.

The current CO_2 demand is partially satisfied by CO_2 extracted from natural wells, which is considered to contradict the logical approach to (European) climate goals [18]. According to Naims’ CO_2 supply and demand analysis [19], in the USA, approximately 45 Mt/a were extracted from natural wells in 2012 for economic purposes. In addition to the future reduction of fossil CO_2 sources, it is argued that CCU could be used as a complementary technology in mitigation technologies, with a focus on local circular economic approaches. Aresta et al. [20,21] state that CO_2 recycling technologies, such as renewable fuel production, could become economically and environmentally feasible, with the support of renewable energies. It has been proposed that “spent carbon” emissions should be converted to “working carbon” emissions in order to reduce the total fossil carbon input in our economy and environment. The authors of the present paper are convinced that biogenic CCU (bio-CCU) complements this approach.

Naims [19] as well as most other authors [14,22,23] focus mainly on fossil CO_2 sources presumably because fossil CO_2 adds to the CO_2 content of the atmosphere to some extent, whereas biogenic CO_2 is seen as "neutral".

In this study, we focus on the potential of the capture and utilization of biogenic CO_2. Since the global economy can only become carbon neutral if fossil energy- and fossil resource-based chemical industries adapt to new, renewable energy driven bio-CCU processes, the utilization of CO_2 from green sources, such as biogas upgrading or bioethanol fermentation, is a strategic approach for CO_2 mitigation strategies.

Bioenergy-based CO_2

In 2005, the Intergovernmental Panel on Climate Change (IPCC) [24] estimated the CO_2 potential from bioenergy in North America and Brazil to be 73 Mt/a (based on the data from 2000 and 2003). In 2016, Naims [19] referred to the same data. Based on the 2014 data, Ericsson [9] estimated the European potential for biomass and renewable waste combustion to be 287 Mt/a, with municipal non-renewable and industrial waste combustion contributing an additional 81 Mt/a. As a future forecast, Pour et al. [25] estimated 2,800 Mt/a of CO_2 from municipal solid waste incineration on a global scale for the year 2100.

Fermentation-based CO_2

Fermentation processes are particularly attractive since this CO_2 is relatively easy to capture and is considerably more pure. According to IPCC [24], 17.6 Mt/a is estimated to be produced at North America and Brazil (data from 2000 and 2003). Naims [19] refers to the same numbers, whereas Ericsson estimates 4.4 Mt/a for Europe. Reiter et al. [26], Kouri et al. [27] and Marchi et al. [28] estimated the CO_2 potential from different fermentation processes for some European countries, with Marchi et al. also included an estimation for CO_2 from wine production on a North American and global scale.

Biogas (upgrading)-based CO_2

The potential of biogas upgrading, and biogas combustion has been highlighted in some studies. There is a concentration of biogas plants in several European countries, such as Germany, Austria, and Finland. Thus, the scenario for this type of CO_2 capture is mainly considered in research papers and studies for Germany, Austria and Finland and a few studies report on a global or European scale. Because of the reduced application of energy crops, Billig et al. and Horschig et al. [12,17] estimate 11.95–18 Mt/a of CO_2 generation from this source for Germany in 2016, and a reduction to 8–11.3 Mt/a by 2050 due to a significance reduction in the number of operating biogas and biomethane plants in Germany, based on a report from Scheftelowitz and Thrän [29]. Reiter et al. [26] estimate 0.013 Mt/a for Austria in 2013, and Ericsson [9] estimates 23 Mt/a for Europe in 2017. Pour et al. estimate that 1,160 Mt/a will be generated by landfill gas combustion (as an alternative to waste incineration) by year 2100, on a global scale.

Direct Air Capture

Wohland et al. [30] estimated the European and global Direct Air Capture (DAC) potential to be 500 and 7,000–22,000 Mt/a in 2018, respectively, provided that the DAC is based on carbon neutral renewable (surplus) energy.

Fasihi et al. [31] estimate the global demand for CO_2 capture from power, transport and industry (excluding iron & steel) and the CO_2 removal sector by DAC (or equivalent) to be 3 Mt/a in 2020 and up to more than 15,000 Mt/a in 2050, of which approximately 8,200 Mt/a is associated with the CO_2 removal sector. This estimation agrees with the range given in the estimation by Wohland et al.

Global bio-CCS potential

However, Ricci et al. [32] predict a global bio-CCS potential from the power sector of 5,800 Mt/a in 2050.

Résumé

The above listing of estimates and predictions for the potential of CO_2 capture shows that most available data are very inconsistent and thus hardly comparable. The potentials presented above mostly include theoretical potentials. Future economic and technical potentials are difficult to define, since they strongly depend on further economic and technological developments, such as trends towards decentralized or centralized energy supply systems of industries and municipalities and changing processes in the chemical industry. Overall, the results presented in this paper mainly correspond with the results from Ericsson [9].

1.2 *Actual CO_2 demand*

There are various CO_2 utilization pathways, some of which were established decades ago while others are still being investigated. Typical direct utilization pathways involve beverage carbonization and horticulture production (greenhouses), using CO_2 as a working fluid. Traditional chemical industries like that of urea, polyurethane (PUR) and various acid and carbonate production processes use CO_2 as chemical feedstock. More recent applications involve micro algae production or new processes to produce well-known products [33,34]. Mikulčić et al. [35] conducted an extensive review of CCU technologies and utilization pathways of captured CO_2.

According to Billig et al. [12], global CO_2 demand is estimated to increase from 197 Mt/a in 2013 to 250 Mt/a by around 2026. Chauvy et al. [14] estimated a global demand potential of 590 Mt/a based on a stoichiometric CO_2-uptake approach, excluding methane, which could account for 3,000 to 4,000 Mt/a. Patricio et al. [13] presented the current data for the potential CO_2 demand in Europe, which added up to 73 Mt/a for the industrial processes. Accordingly, in order to address the predicted increase in CO_2 demand, a comparative evaluation of the sources and limitations of biogenic CO_2 sources is needed.

2 METHODS

Different sources of biogenic CO_2 are classified and illustrated in Figure 2-1. The main biogenic CO_2 sources are combustion of biomass, biogas upgrading to biomethane and industrial fermentation, e.g., brewing and other fermentation processes in the food and beverage (FAB) industry. Although industrial bioethanol is also produced through fermentation [36], it has been classified in its own category. Differently from the FAB industry, bioethanol is mostly used as biofuel and as raw material in several industries. CO_2 from the atmosphere has not been considered as a source from existing industrial plants but, rather, as a diffuse source, which would demand a significant technical effort to be separated. Thus, for subsequent chemical conversion herein, ambient air is not included as an available existing source of biogenic CO_2 and not further discussed in this assessment. Nevertheless, CO_2 separation from ambient air may still play an important role in the sequestration of CO_2 from diluted and dispersed sources in the long-term, as the relevant technology has the potential for significant further development and optimization [37].

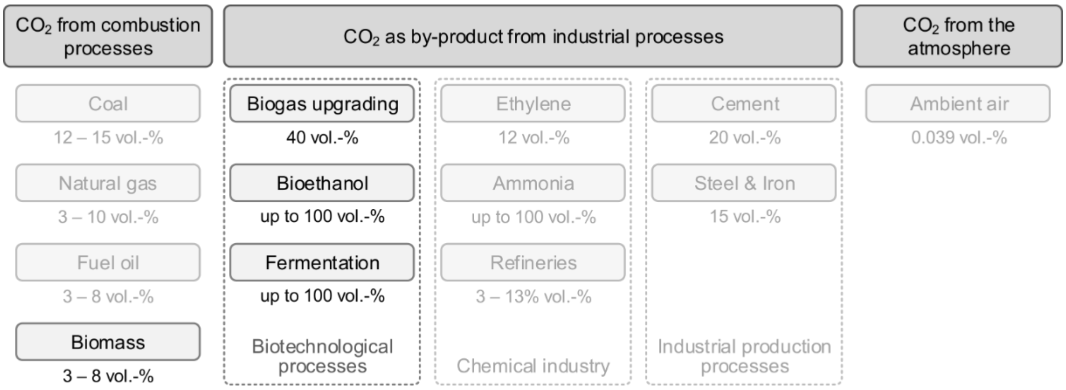

Figure 2-1: Classification of potential biogenic CO_2 sources including the available typical CO_2 concentration

Source: based on [26]

2.1 *CO_2 from solid biofuels – combustion of biomass and renewable waste*

Solid biofuels are defined here according to Eurostat:

"Solid biofuels covers solid organic, non-fossil material of biological origin (also known as biomass) which may be used as fuel for heat production or electricity generation. In energy statistics, solid biofuels is a product aggregate equal to the sum of charcoal, fuelwood, wood residues and by-products, black liquor, bagasse, animal waste, other vegetal materials and residuals and renewable fraction of industrial waste." [38].

As can be seen in Figure 2-1, flue gases from biomass combustion processes consist of only 3–8% CO_2. In flue gases, there are many other components that make the utilization of CO_2 in a pure form a technically challenging task. Because of this required technical effort, higher financial investments are expected, which result in a lower economic feasibility of CO_2 utilization from this source in comparison to CO_2 utilization from industrial bioethanol and biogas production.

An overview of existing and actual initiatives for the development of new CO_2 separation technologies for combustion processes can be found on the CCS browser [39] of the CO_2 Capture Project (CCP) [40]. Further information on CO_2 capture technologies are presented by Cuéllar-Franca and Azapagic [22] and in the IPCC special report on CCS [36].

2.2 *CO_2 from fermentation processes*

2.2.1 Liquid biofuels – fermentation of industrial bioethanol

Liquid biofuels are defined here according to Eurostat:

"Liquid biofuels includes all liquid fuels of natural origin (e.g. produced from biomass and/or the biodegradable fraction of waste), suitable to be blended with or replace liquid fuels from fossil origin. […]" [38].

There are several liquid biofuels like biodiesel, biogasoline, bio jet kerosene and bioethanol. The latter is produced by fermentation processes; additionally a considerable amount of CO_2 is produced. Concurrent to the stoichiometric equation, the gas produced during the fermentation consists of up to 99–100% CO_2 [9]. The basic equation of ethanol fermentation is:

$$C_6H_{12}O_6 \rightarrow 2\ C_2H_5OH + 2\ CO_2.$$

CO_2 from bioethanol production is pure enough to be directly utilized in the FAB industry, e.g., as carbon acid in beverages [41]; hence, this CO_2 source can also be expected to fulfil the fundamental requirements for CCU applications.

The quality requirements for CO_2 utilized in the FAB and pharmaceutical industries are very high [35]. Regulations for the quality of food grade CO_2 are released by the European Industrial Gases Association (EIGA) [42] and the International Society of Beverage Technologists (ISBT) [43] (Table 2-1). The aforementioned regulations are strict; however, for some impurities such as water, O_2, hydrocarbons, and CO, the limitations are not as demanding as those for CO_2 gases for the chemical industry according to EN ISO 14175: C1. As an example of the purity requirements for CO_2, the product data sheets from the Linde Group could be potentially considered: BIOGON® C flüssig E290 - Kohlendioxid 3.0 [44] for food grade CO_2 and Kohlendioxid 4.5 [45] for chemical industry CO_2. Depending on the requirements for the CCU pathway, further purification of food grade CO_2 or CO_2 for chemical industry may be needed, for example, by activated carbon technologies [46–49].

Component	**Concentration**
Assay	99.9% v/v min.
Moisture	20 ppm v/v max.
Ammonia	2.5 ppm v/v max.
Oxygen	30 ppm v/v max.
Oxides of Nitrogen (NO/NO_2)	2.5 ppm v/v max. each
Non-volatile residue (particulates)	10 ppm w/w max.
Non-volatile organic residue (oil and grease)	5 ppm w/w max.
Phosphine (only for CO_2 from phosphate rock sources)	0.3 ppm v/v max.

Total volatile hydrocarbons (calculated as methane)	50 ppm v/v max. of which 20 ppm v/v max. non-methane hydrocarbons
Acetaldehyde	0.2 ppm v/v max.
Aromatic hydrocarbon	0.02
Carbon monoxide	10 ppm v/v max.
Methanol	10 ppm v/v max.
Hydrogen cyanide (only for CO_2 from coal gasification sources)	0.5 ppm v/v max.
Total sulfur (as S for total content < 0.1 ppm v/v)[1]	0.1 ppm v/v max.
Taste and odor in water	No foreign taste or odor
Appearance in water	No color or turbidity
Odor and appearance of solid CO_2 (snow)	No foreign odor or appearance

Table 2-1: Limiting characteristics for CO_2 to be used in beverages.
Source: European Industrial Gases Association (EIGA) [cf. 42]

Notably, the FAB industry offers possible competing utilization pathways for CO_2, especially from industrial bioethanol production. According to ePURE, bioethanol producers in Europe commercialized 0.4 Mt of CO_2 utilization in 2016 [50]. The estimations within this paper are based upon the information available on the producers' websites [51] and on some approximations from ethanol outputs, indicating an amount of 1.52 Mt CO_2/year, which may possibly be commercialized by the European bioethanol producers in the mid-term future.

2.2.2 Other industrial fermentation processes

In addition to CO_2 derived from bioethanol industry, the CO_2 from fermentation processes in the FAB industry, such as brewing processes, is of interest. In the beverage industry, beer brewing and wine production lead to considerable amounts of CO_2. Furthermore, the fermentation of acids, e.g., citric acid, produces considerable amounts of CO_2.

According to [52] 41.1 billion litres of beer were brewed in Europe in 2016. Using an average value of 5 vol.-% of alcohol and 5 g/l carbon acid for beer, it can be estimated that 35 g CO_2/l were released during this fermentation process [53–55].

A comparable estimation could be conducted for the CO_2 potential from fermentation in European wine production. The average value of the European wine production is approximately 17 billion litres of wine annually [56]. The average alcohol content is 11 vol.-% while the carbon acid content is quite low, approximately 1 g/l, which corresponds to approximately 87 g CO_2/l released during the production process [57,58]. Marchi et al. [28] estimate 84.5 gCO_2/l_{must} to be released by wine production, which corresponds to the estimations in this paper. Furthermore, the carbon acid amount depends on the type of wine. Red wine in particular has a very low content of carbon acid and is decarbonated very often, whereas the carbonation of white wines and sparkling wines is quite common. Similar to beer brewers, wine producers protect their wine from air using CO_2; in practice most of this CO_2 is

[1] If total sulphur content > 1 ppm v/v, then: Carbonyl Sulphide 0.1 ppm v/v max., Hydrogen Sulphide 0.1 ppm v/v max., Sulphur Dioxide 1.0 ppm v/v max.

not available for chemical syntheses [28,59–61]. The small-scaled structure of producers in this sector is another barrier to the implementation of CCU, for economic reasons [28].

2.3 *Gaseous biofuels*

Gaseous biofuels are defined here according to Eurostat:

"Biogas is a gas composed principally of methane and carbon dioxide produced by anaerobic digestion of biomass or by thermal processes from biomass, including biomass in waste. In energy statistics, biogas is a product aggregate equal to the sum of landfill gas, sewage sludge gas, other biogases from anaerobic digestion and biogases from thermal processes." [38]

In principle, there are two ways to utilize CO_2 from biogas, independently of the source of the biogas. First, as biogas consists of approximately 60% methane and 40% CO_2 [9, 12, 62, based on 63], upgrading biogas to biomethane offers a large potential for the generation of biogenic CO_2. Second, the combustion of biogas and biomethane generates CO_2 as a compound of the flue gas during the generation of heat and power. The results section gives an overview of both basic possibilities.

2.3.1 Biogas substrates

Biogas from anaerobic digestion is derived from biogas plants with highly different biomass feedstock. Table 2-2 provides an overview of the possible compounds of biogas substrates. Most plants are supplied with varying mixtures of substrates. Some substrates demand special treatment and plant design. Depending on the substrates, the composition of the biogas varies, with regard to the methane concentrations, CO_2 concentrations and trace compounds. Since the ideal CO_2 stream needed for the various CCU synthesis options is highly pure, some biogas substrates may be not suitable for this application without extensive purification processes of the potentially utilized CO_2.

Category	Possible feedstocks	
Agricultural	energy crops	maize silage
		sugar beet silage
		silage from different grains
		crop residues in general
	manure	pig
		cow
		sheep
		poultry
FAB industry	liquor industry	grain stillage
		potato stillage
	sugar and starch industry	sugar beet residues
	beverage industry	fruit pomace
	food industry	production residues

		slaughter waste and blood
		dairy residues
Waste industry		municipal renewable waste
		industrial renewable waste
		sewage sludge
Textile industry	production residues	leather
		fur
		biological textiles
Wood industry	panels and furniture	wood residues
	paper industry	paper and cardboard residues
		pulp residues

Table 2-2: Selection of possible substrates for biogas plants.
Source: based on [64,65]

According to the Statistical Report of European Biogas Association (EBA) 2017 [66] the feedstock use for biogas production differs for every country. Using the substrate's mass percentage as an indicator for biogas production (excluding landfill gas), energy crops are the main substrates in Latvia, Austria and Germany, while in Greece, Cyprus, France, Serbia, Poland and Italy agricultural residues are the main feedstock. In the UK, Finland, Sweden, Spain, Denmark and especially in Switzerland [66], sewage accounts for the largest share. In some countries such as Belgium, Croatia and Hungary the distribution is more even.

Considering landfill gas, the statistics shift. Approximately one third of Estonia's feedstock origin is landfill waste [66]. In Greece, landfill gas accounts for two thirds of the produced biogas [66], and for Norway landfill gas accounts for nearly half [66]. Portugal is exceptional in using landfill gas; landfill gas accounted for over 95% of produced biogas in 2016 [66]. Other countries making significant use of landfill gas are the UK, Sweden, Romania, Poland, Ireland, France and Finland.

Figure 2-2 is compiled based on data from the EBA [66]; it shows the share of feedstock use for biogas according to the substrate's mass percentages and electricity production per substrate in each country for Europe. Agricultural waste and energy crops represent the highest share, with 39% each, followed by "other", which includes organic waste from households and industry, sewage, FAB and bio-waste/municipal waste. According to the EBA, the share of sewage is underestimated.

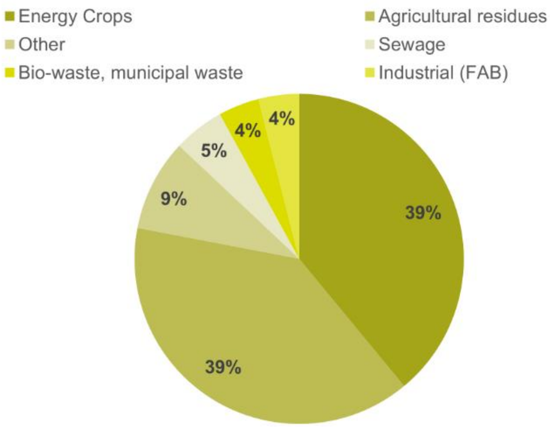

Figure 2-2: Estimation of relative significance of each feedstock in the biogas industry in Europe in 2016

Source: based on feedstock mass percentages and electricity production per feedstock in each country. The share of sewage sludge is underestimated due to missing information on the share of sewage sludge for some countries. The diagram is adapted from Figure 9-EU of the Statistical Report 2017 from EBA [66].

The main substrates by mass percentage, excluding landfill gas for current biomethane plants, are slightly different than the main substrates for biogases in general. In particular biowaste and municipal waste, agricultural residues and, to some extent, unknown feedstocks are of major relevance. In Germany most biomethane plants are based on energy crops, followed by agricultural residues and bio-/municipal waste, while in the UK "other" (municipal waste, etc.) and agricultural substrates play the biggest role. These two countries are the ones with the most biomethane plants (see Figure 3-3). For Sweden, which ranks third among European countries with the most biomethane plants, "other" and sewage are the feedstocks which account for the most biomethane plants [66].

2.3.2 Composition of biogas

Depending on the substrates, the plant system, operating temperature and various other parameters, different trace compounds can be found in biogas resulting from biological processes in anaerobic digestion. Typical impurities are water vapour, O_2, N_2, NH_3, H_2, H_2S, siloxanes and biogas specific volatile organic carbons [64, 67]. Table 2-3 provides a more detailed overview of possible impurities and their typical concentrations in biogas.

Components	Concentration range
Main components	
Methane (CH_4)	50 - 70 mol.-%
Carbon dioxide (CO_2)	30 – 50 mol.-%
Nitrogen gas (N_2)	0 – 3 mol.-%
Oxygen (O_2)	0.0 – 0.5 mol.-%
Hydrogen (H_2)	0.0 – 1.5 mol.-%
Water vapor (H_2O)	1 – 7 mol.-%
Carbon monoxide (CO)	0 – 1 mol.-%
Trace components	
Ammonia (NH_3)	0 – 308 ppm(mol)

Hydrogen sulphide (H_2S)	20 – 850 ppm(mol)
Terpenes	0 – 500 ppm(mol)
Benzene, Toluene, Xylene (BTX)	0 – 7 ppm(mol)
Hydrocyanic acid (HCN)	0 – 0.003 ppm(mol)
Fluorine compounds (R-F, incl. HF)	0 – 1.3 ppm(mol)
Chlorine compounds (R-Cl, incl. HCl)	0.1 – 5 ppm(mol)
Siloxanes (D4 & D5)	0 – 3.4 ppm(mol)

Table 2-3: Detailed overview of biogas components.
Source: adapted from [68]

Rasi et al. [69] provide a more detailed overview of biogas components, depending on the utilized substrates for biogas production. Landfill gas, in particular, often contains high amounts of H_2S and VOCs. Additionally, e.g., in industrial wastes, several potentially hazardous trace compounds can be part of the landfill gas, such as fluorinated and chlorinated hydrocarbons, aromatic compounds and higher hydrocarbons [70]. Other common trace compounds are siloxanes, which are also present in sewage sludge gas, since siloxanes originate from ingredients such as cosmetics, soaps and detergents. Depending on the substrate, agricultural biogas plants can also produce biogas with a very high H_2S content, for example with that derived from manure feedstock [71].

2.3.3 Upgrading technologies for biogas

There are several available technologies for upgrading biogas: water scrubbing, amine scrubbing, pressure swing adsorption (PSA), physical scrubbing, chemical scrubbing, membrane separation and cryogenic separation [8,67,72]. Since biogas upgrading mainly aims at the separation and purification of the methane content in the biogas, trace compounds are often removed together with the CO_2 stream as can be seen in Figure 2-3. Consequently, the CO_2 stream may contain considerable amounts of impurities and is potentially not suitable for most utilization applications for chemical synthesis without further treatment.

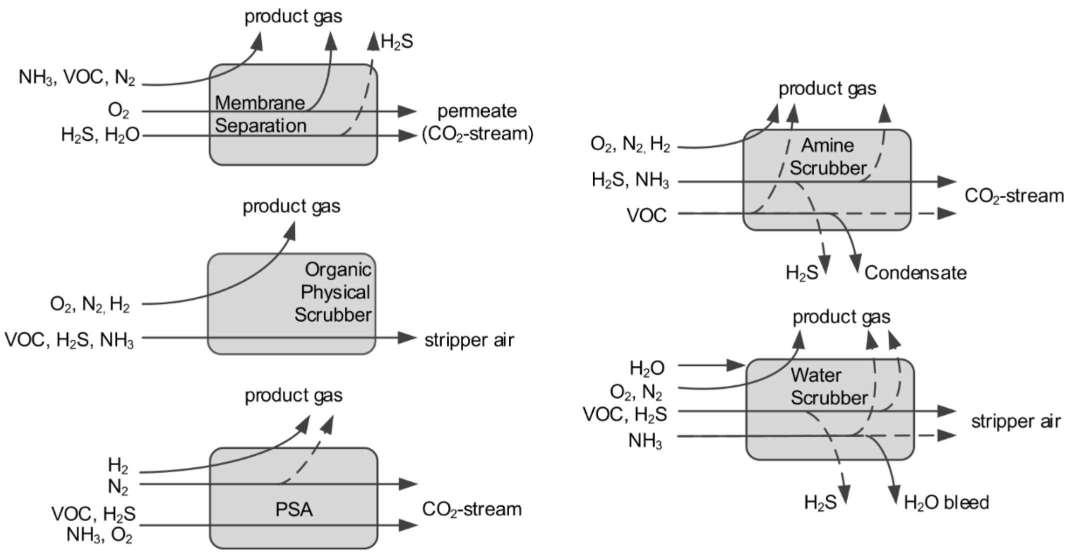

Figure 2-3: Some exemplary paths of impurities from biogas upgrading technologies
Source: based on Hoyer et al. [67, Fig. 12]

According to the European Biogas Association [66], most current biogas upgrading plants use water scrubbers, followed by chemical absorption, PSA, membrane separation and physical absorption. In Germany, the European country with the most upgrading plants, all upgrading technologies are represented; however, chemical absorption and water scrubbing are each implemented in 30% of the plants, and PSA in 22% of the plants. In Sweden, ranked third among European biomethane countries, 69% of the biomethane plants use water scrubbers, followed by chemical absorption, PSA and membrane separation [66]. For the UK, ranked second, no information on upgrading technologies is available [66]. The only European countries known for implementing physical absorption are Germany, Norway and Switzerland. Combining this information with the paths for impurities according to Figure 2-3, most CO_2 from upgrading plants requires further purification before it can be utilized for CCU pathways.

The available CO_2 concentration in biogas upgrading off-gases depends on the applied upgrading technology. In processes like membrane separation, amine scrubbing and pressure swing adsorption, relatively high CO_2 contents are reached in the off-gas streams (typically <99 vol.-%, CH_4 being the balance). Under certain circumstances, if the level of off-gas impurities is low, the off-gases of these processes can be readily used for CCU options.

In contrast, processes that involve stripping with air, such as with pressurized water scrubbing, produce more diluted CO_2 off gases. This means that CO_2 from such processes would need further gas upgrading steps to remove air components and increase the CO_2 content.

A review on upgrading technologies for biogas to biomethane from Vijayanand and Singaravelu [73] gives an overview of CO_2 separation techniques for biogas; additionally, Singhal et al*.* [74] gives an overview of the transformation of biogas to biological compressed natural gas (bio-CNG). A comprehensive review on biogas generation factors, enhancements of biogas production techniques, upgrading and cleaning techniques are given by Al Mamun and Torii [75] as well as Andriani et al*.* [76] and Sun et al*.* [70]. Pellegrini et al*.* [77] give an overview of the purification costs of biogas, depending on the source of biogas.

Cryogenic CO_2 Capture

A relatively new technique for biogas upgrading is the cryogenic CO_2 separation, which involves many different process steps using very low temperature processes. One example, which results in partially food grade CO_2, is the CO_2 Wash® process developed by US-based Acrion Technologies. It applies the effect of impurities solubility in liquid CO_2. After H_2S removal and drying, the biogas is mixed with liquid CO_2. This process results in biomethane, food grade CO_2 and a CO_2–VOC mixture as the products [72]. Depending on the requirements for the CCU pathways, further purification of the food grade CO_2 may be needed, for example, by activated carbon technologies [46,47]. Yousef et al. [78,79] improved cryogenic liquid CO_2 separation from Biogas and Tan et al. [80] present a system review and property impacts.

Examples of new CO_2 separation technologies for biogas

A new technique for CO_2 capture at room temperature using aqueous Na_2CO_3 has been presented by Barzagli et al. [81]. Chaemchuen et al. [82] and Cavenati et al. [83] presented metal-organic frameworks (MOFs) for upgrading biogas. Lim et al. [84] investigated clathrate-based CO_2 capture from biogas.

Because of the recent increase in popularity of CCS, but also because of the quality requirements of the chemical and FAB industries, purification processes of CO_2 are of high interest. For example, some impurities have corrosive properties, which is a problem in transport and long-time storage [85]; other impurities are harmful to chemical processes or toxic in terms of the FAB industry. For CCU, not only because of the transport but also because of the chemical conversion of the CO_2 and the possible poisoning of conversion equipment, very low impurity concentrations are required. Despite CO_2 purification processes' state-of-the-art nature, much development work is ongoing.

2.4 *Potential sources of fossil CO_2*

Although this study targets biogenic, and thus "green", sources of CO_2 as feedstock material, the main emitters of CO_2 are based on fossil fuels. Because of the increasing interest in the utilisation of CO_2 to attain ambitious goals of decarbonisation and closed carbon cycles, these fossil sources must be partly considered as relevant input sources as well. Therefore, this section gives a rough overview of potentially available carbon sources and their relevance to CCU process chains.

The potential sources for fossil CO_2 can be classified according to their emitting processes and industry sectors. This classification is shown in Figure 2-4.

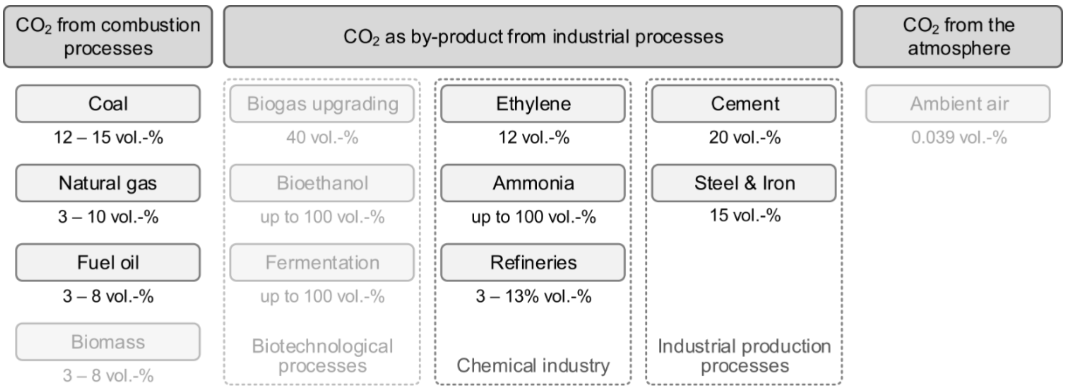

Figure 2-4: Classification of potential fossil CO_2 sources including the available typical CO_2 concentration

Source: based on data from [26]

For the European Union (EU), industrial CO_2 emissions are mainly registered in the EU emission trading system (EU ETS)[2]. The system records CO_2 emissions from power and heat generation as well as from energy-intensive industry sectors, including oil refineries, steel works and production of iron, aluminium, metals, cement lime, glass, ceramics, pulp, paper, cardboard, acids and bulk organic chemicals [86]. Beyond these, commercial aviation is also

[2] The EU ETS operates in 31 countries. This includes the 28 EU countries plus Iceland, Liechtenstein and Norway. These 31 countries are indicated as EU in the following context, if not stated otherwise.

included in the register; however, owing to the limited capability for the direct capture and separation of CO_2 emissions, these are excluded in the following investigations.

For the analysis of fossil CO_2 potentials in this study, the report data released by the European Commission in April 2018 [87] was used as a primary data source. Since data for 2017 was incomplete at the time of this report, year 2016 data was used as a reference for the subsequent calculations.

The EU ETS database for verified emissions allows for the categorization of registered emissions according to their originating industrial sector. Thus, the available data can be grouped into the following main categories:

- power & heat from fossil fuels; energy industry
- chemical industry
- iron & steel
- (other) metals processing
- cement, clinker, lime, ceramics
- production of glass & glass fiber
- pulp, paper & board
- other installations
- aircraft operator activities

In addition to these main categories, the chemical industry sector has been further subdivided into refinery, ammonia production and other chemicals. Within the category of cement, clinker, lime and ceramics, the manufacturing of ceramics by firing has been separated from the production of cement clinker and lime.

Adding additional detail categories in the other sectors according to the activity type codes provided in the data source is also possible; however, other than providing a clearer classification for large parts of the register, further categorization of the other sectors does not add any beneficial value to the investigations executed in the context of this potential analysis.

2.5 *Consideration of technological capture rates*

The amounts of CO_2 discussed in the previous sections 2.1 to 2.4 provide an overview of the overall direct emissions from different sources. To evaluate the real potentials for utilizable CO_2, the appropriate efficiencies of certain technologies used for separation, which limit the amounts of CO_2 that can be captured and used, must be considered. In the following sections, affordable capture rates and resulting utilisation values are analysed.

To obtain an overview of the technically affordable capture rates for industrial processes, an appropriate literature review was conducted. This review closely mimics the industrial sectors discussed in the sections 2.2 to 2.4. As some of these sectors include various technological processes, the analysis was performed on the even more fine-grained level of sector sub-categories, resulting in the categorization shown in Table 2-4.

Main industry sector	Sub-category
power & heat from fossil fuels; energy industry	coal
	natural gas
	energy industry
chemical industry	refinery
	ammonia production

	other chemicals
iron & steel	iron & steel
(other) metals processing	(other) metals processing
cement, clinker, lime, ceramics	cement, clinker, lime
	ceramics by firing (bricks, tiles, …)
production of glass & glass fiber	glass & glass fiber
pulp, paper & board	pulp, paper & board
biogenic processes	biogas upgrading
	bioethanol (fermentation only)
	bioethanol (fermentation & cogeneration)

Table 2-4: Categorization of CO_2 providing industrial processes.

2.6 *Costs for CO_2 capture*

Generally, the investment costs for CO_2 sequestration are not easy to define. It is reasonable to set a reference for specific costs according to the CO_2 source being used. Affordable sequestration rates strongly depend on the concentration of CO_2 in the (generally gaseous) source stream and the underlying emitting process. As the CO_2 sources and the reference values for assessing investment costs exhibit significant variance, it seems more practical to determine the value of the required CO_2 as an operating supply and therefore represent its costs as per ton (€/t) CO_2, depending on its source and sequestration technology, respectively.

2.7 *CO_2 from biogas or bioethanol plants*

Biogas plants which feed-in to the natural gas grid lend themselves as a source of otherwise unused CO_2 as characterized in section 2.3. Specific costs for CO_2 sequestration in such biogas plants are approximately €12 cents per standard cubic meter of methane. Assuming a CO_2 fraction of 40% in the raw gas flow, this unit price would lead to the cost of approximately €90 per ton CO_2 (for 2012). However, the sequestration, as well as the removal of impurities (like sulphur), is normally done for the retrieval of biomethane which can be fed into the gas grid, and hence costs are assigned to the methane production. In this aspect, the sequestration of CO_2 is neutral in terms of costs [26,88].

The costs of CO_2 from a bioethanol plant, as the source, would behave in a similar manner. In the fermentation process, a high-quality stream of CO_2 is accumulated as a by-product. If only this method is considered as a potential source, then the sequestration costs would be limited to the costs necessary for the compression of the gas, which can be assumed to be approximately €12–25 per ton CO_2 [23,26,89,90]. If the bioethanol plant uses cogeneration for energy provision and the CO_2 capture from the cogeneration process is also considered, then the costs would be between €42 [91] and €111 [89] per ton CO_2 for capturing and compression, respectively.

3 RESULTS AND DISCUSSION

3.1 CO_2 potential from solid biofuels in Europe

Solid biofuels are predominantly utilized by combustion but some amounts also statistically contribute to biogas production. However, the combusted amount is responsible for the highest biogenic CO_2 emissions in Europe, which account for approximately 437 Mt CO_2/year according to Eurostat statistics of 2016 and IPCC 2006 emission factors for stationary combustion [92,93]. Solid biofuel combustion takes place not only at large plants but also at various small-scale facilities such as household fireplaces and the central heating systems of buildings. Hence, it can be concluded, that if the global statistics for solid biofuels and the fraction of direct heat use is considered, the amount of CO_2, which could be utilized is therefore significantly lower than the theoretical potential of 437 Mt CO_2/year (see Section 3.1.1.).

3.1.1 Global CO_2 potential from solid biofuels

According to the Global Bioenergy Statistics 2017 of the World Biogas Association (WBA) [94], municipal waste, industrial waste and primarily solid biomass account for 54.72 EJ/year (for 2014) worldwide. Municipal waste and industrial waste include non-renewable fractions. 42.88 EJ of this solid biomass energy are converted to direct heat, meaning direct consumption of energy sources in the residential, agriculture and commercial sectors (not combined heat and power (CHP), heat or electricity-only plants) [94]. Therefore, only CO_2 derived from 11.84 EJ [3] could be utilized for CCU, but only 6.86 EJ[4] of solid biomass are utilized in CHP, electricity only and heat only plants. The respective CO_2 amounts are 1,184 Mt and 686 Mt per year[5].

3.2 CO_2 from fermentation processes

3.2.1 CO_2 potential from bioethanol industry in Europe

The total amount of CO_2 produced during bioethanol fermentation is approximately 5.71 Mt CO_2/year according to production statistics from 2016. Ericsson estimates 4.4 Mt CO_2/year for Europe [9]. Most European bioethanol fermentation plants are based in France (17 plants) followed by Germany (8 plants) and the UK (5 plants). Other European countries have no plants or at most, three plants. In total, there are approximately 57 plants in Europe, of which 43 are located in the EU [based on 50,51,93]. The total installed production capacity for bioethanol in Europe is approximately 9.2 billion liters a year (of which 6.3 billion litres a year account to ePURE Members, whereas total production of ePure Members accounted for 5.2 billion litres in the year 2016, i.e. 82.5 % of capacity. [50] It is assumed that the European share of real production is similar, i.e. 7.57 billion litres ethanol in 2016.

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[3] Difference of 54.72 EJ and 42.88 EJ

[4] Summed up the energy utilized 2014 in CHP (2.4 EJ), heat only plants (0.48 EJ) and electricity only plants (3.98 EJ), Source: [94].

[5] Conversion from energy to CO_2 with IPCC emission factors [92] Approx. 100,000 kg CO_2/TJ

The approximated theoretical amount of CO_2 produced in the European industrial bioethanol industry in 2016 is summarized in Figure 3-1.

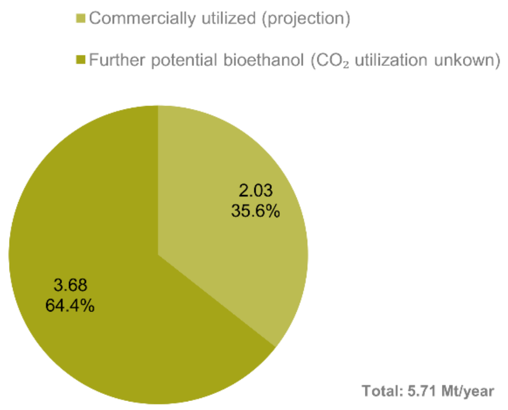

Figure 3-1: Amount of CO_2 in Mt/year produced in the EU bioethanol industry in 2016 including amount of possibly commercialized CO_2 according to projections.
Source: based on data from [50,and 51]

According to ePURE [50], the FAB industry as one group and other industries as another group each represent 11% of European industrial bioethanol consumption. Fuel accounts for the remaining 78% of consumption. The possibilities for yielding CO_2 "end-of-pipe" from the ethanol utilisation pathway in the FAB and other industries is complex and thus relatively unattractive. Similarly, while gaining CO_2 from ethanol flue gases would theoretically be possible, it is technically and economically not currently feasible, as bioethanol is usually used as a component of vehicle fuel.

3.2.1.1 Global CO_2 potential from bioethanol industry

However, the global bioethanol production is much higher; it accounted for 78 billion liters of bioethanol produced in 2014. In comparing ePURE figures from 2014 [95] to the 2014 global production, the European fraction is only 8.4%. Industrial bioethanol from America accounts for the biggest amount, with 88% of market shares, while Asia ranks similar as Europe, followed by Oceania and Africa. Since, as mentioned above, CO_2 from bioethanol is very pure and the capture process is relatively simple, establishing CCU production in North and South America, where large bioethanol production sites can be found, is an option for further consideration.

3.2.2 CO_2 potential from other industrial fermentation processes

Approximately 1.44 million tons of CO_2 are released from annual European beer production. The CO_2 amounts which can be utilized are smaller, since approximately 5,845 out of 8,130 European breweries in 2015 were so called microbreweries, with an annual beer output of, at maximum capacity, 1,000 hl [52], which corresponds to approximately 3.5 tons of CO_2/year. Therefore, the technical effort needed to gather large amounts of CO_2 is quite high. Furthermore, some breweries already utilize their own CO_2 as protective gas for filling [96–99].

Therefore, most of the CO_2 from this fermentation process is not applicable for other CCU processes.

For the EU wine industry, the annual CO_2 amount estimated from the average values presented in section 2.2.2 is 1.48 million tons CO_2. Marchi et al. [28] estimate an effective annual CO_2 potential of 1.065 Mt/a for Spain, Italy and France, who are the main wine producers in Europe.

3.2.3 CO_2 from biogas upgrading

3.2.3.1 Biogas upgrading in Europe

Approximately 17,783 biogas plants were operational in Europe at the end of the year 2017 [100] and approximately 497 upgrading plants were installed in Europe in early 2017 [8,100]. At the end of 2017, approximately 540 biomethane plants were operational in Europe [100]. Figure 3-2 gives an overview of the number of biogas plants per country in Europe at the end of 2017. Since then the distribution has changed only marginally.

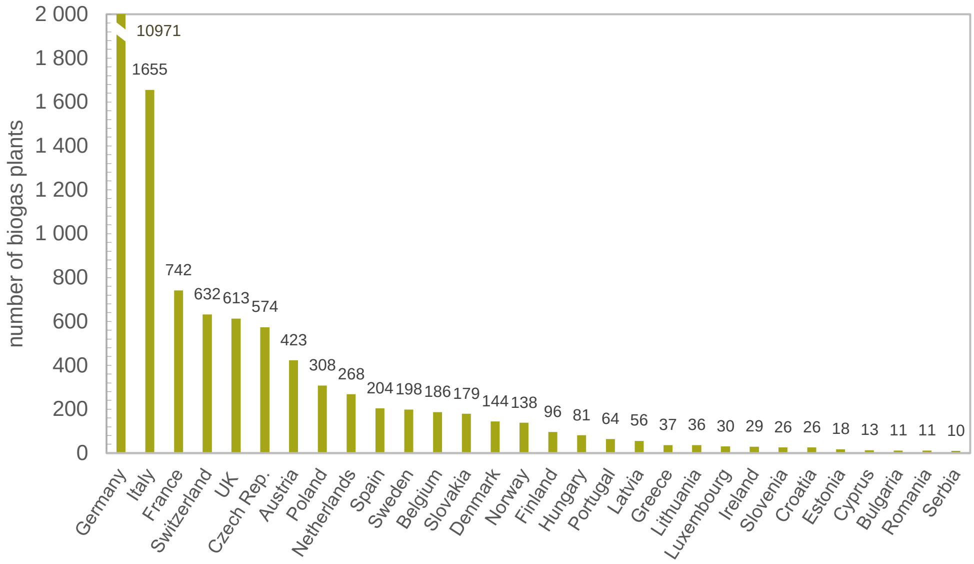

Figure 3-2: Biogas plant distribution in Europe at the end of 2017[6]

Source: based on European Biogas Association (EBA), Statistical Report on the European Biogas Association 2018, Figure EU-2 [100]

In 2018, EBA and Gas Infrastructure Europe (GIE), in collaboration with several partners, published the European Biomethane Map 2018 [8], which includes detailed data from all known European biogas upgrading plants. According to the map, Germany, the UK and Sweden are the pioneers of the field, in terms of the number of upgrading plants, as can be seen in Figure 3-3. By comparing Figure 3-2 and Figure 3-3, it can be seen that there is still a significant

[6] Data underlie constant change and are only an orientation guide for the development of European biogas economy.

potential for biogas upgrading. This potential indicates a significant CCU potential of CO_2 from biogas.

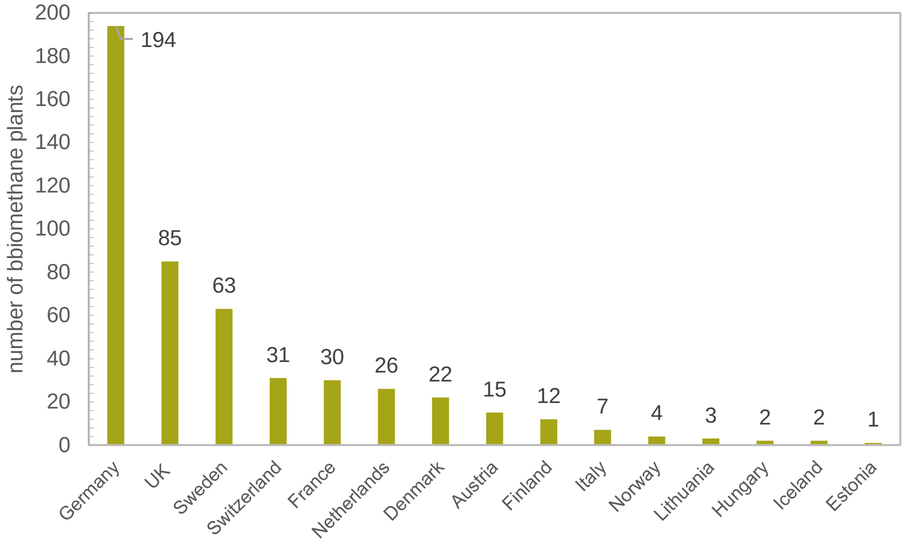

Figure 3-3: European country ranking according to the number of biogas upgrading plants in early 2017

Source: based on a diagram, which is part of the European Biomethane Map 2018 of the European Biogas Association (EBA) and Gas Infrastructure Europe (gie) [8].

Since not only the number of biogas and biomethane plants, but also the specific CO_2 potential or each are of interest, annual CO_2 amounts derived from biogas and biomethane production in Europe are presented here. The numbers include the CO_2 potential from the biogas composition (~ 40 vol.-% CO_2) and biogas upgrading (~ 99 vol.-% CO_2) and exclude the CO_2 emitted during the combustion of biogas in a CHP facility or utilisation of the upgraded biomethane. Figure 3-4 and Figure 3-6 show the approximated cumulative CO_2 potential in 2016 in the EU for biogas and biomethane plants. Figure 3-5 shows the CO_2 potential from biogas production per country for all of Europe. The minor difference in the volumes between Figure 3-4 and Figure 3-6 can be attributed to the different primary data sources. Germany is in first place for the most biogas produced except for landfill gas, for which the UK accounts for the largest current amount. Biogas from thermal processes accounts for a very small amount of CO_2 and is mostly derived from Finland.

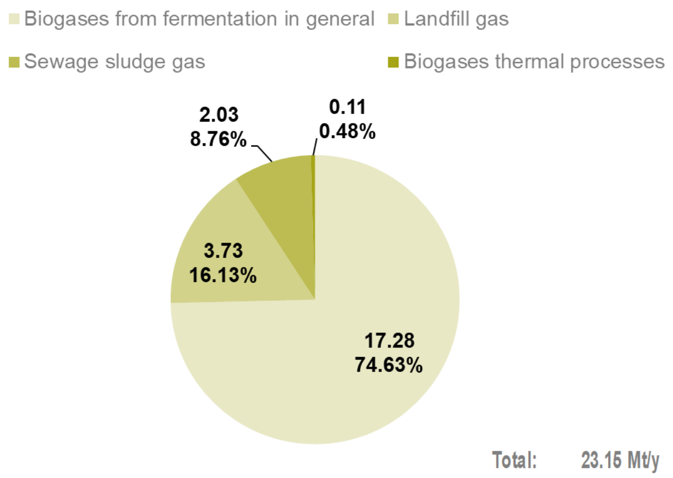

Figure 3-4: Cumulative CO_2 potential in Mt/year from biogas production for the EU-28 countries.
Source: based on Eurostat data on EU biogas production in 2016 [93]

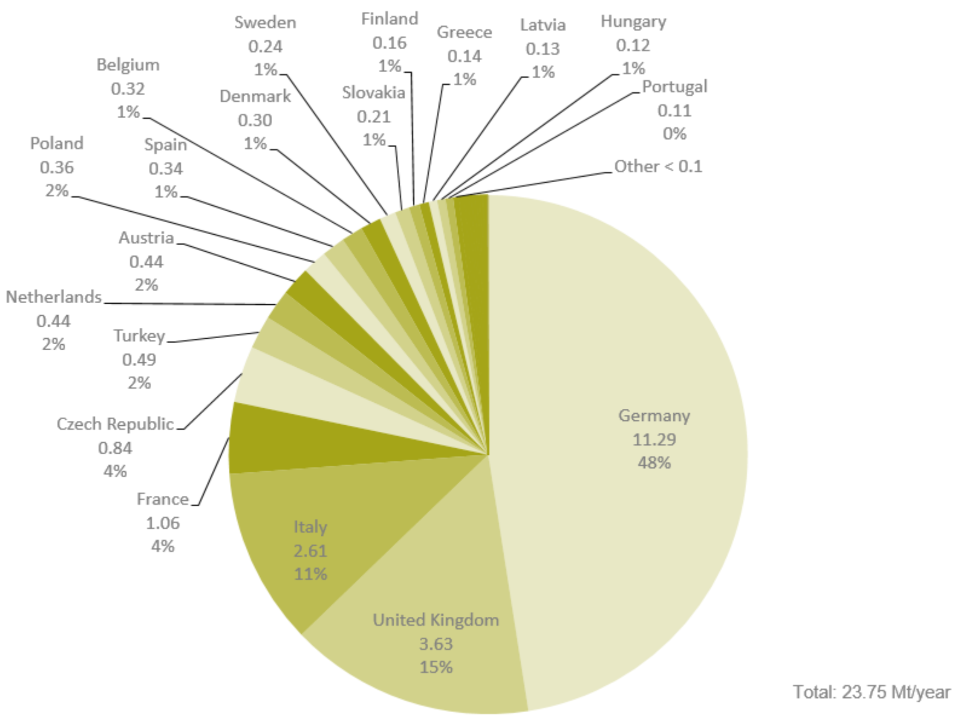

Figure 3-5: CO_2 potential from biogas production per country
Source: based on Eurostat data on European biogas production in 2016 [93]

As illustrated in Figure 3-6 the approximated CO_2 potential from biomethane upgrading represents only 14% of the cumulated CO_2 from biogas production, still excluding the CO_2 emitted during utilisation (combustion of biogas in a CHP facility or utilisation of upgraded biomethane in different applications, e.g., heat, electricity, or transport).

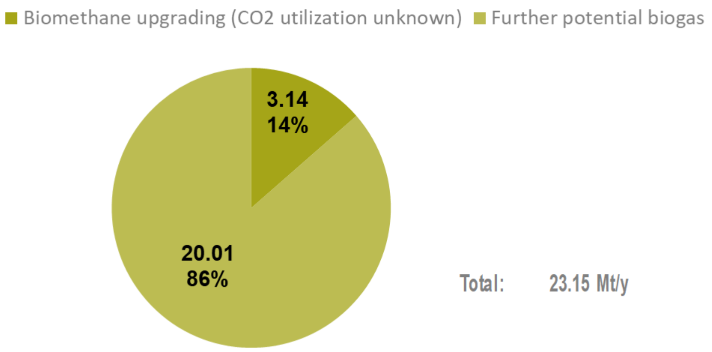

Figure 3-6: CO_2 potential in Mt/year in 2016 from biomethane upgrading as part of total CO_2 potential from biogas production for the EU-28.
Source: based on data from [8] [93]

3.2.3.2 Global potential of biogas upgrading

According to the data from the WBA [94] in 2014, approximately 1.27 EJ of biogas were produced globally. This accounts for a potential of approximately 42.3 Mt CO_2 from biogas production, assuming a CO_2 content of approximately 40% and a biogas heating value of 21.6 MJ/Nm^3 / 6 kWh/Nm^3. Approximately 50% of global biogas is produced in Europe, whereas Asia accounts for one third and America for roughly 17%.

3.2.4 CO_2 from combustion of biogas and biomethane

During the combustion of biogas and biomethane, other than some trace compounds, CO_2 and water are mainly produced. The exact amounts depend on the composition of the fuel gas and the air supply during combustion. Therefore, only approximate CO_2 amounts can be obtained.

3.2.4.1 CO_2 potential from biogas and biomethane combustion in Europe

Stoichiometric

In 2016, approximately 695 PJ of biogas were produced in the EU according to Eurostat [93]. Considering data from the EBA [100], 62 PJ biomethane were produced in 2016. This is equal to 8.9% of total biogas production. Therefore, 633 PJ, or approximately 26.5 billion m³, of biogas[7] are utilized in CHP and other plants, whereas the remaining amount is upgraded. The above estimations are based on a gas composition of 60 vol.-% methane and 40 vol.-% CO_2. This is equal to 52.5 Mt of CO_2 per year[8], considering a density of 1.98 kg CO_2/m³ CO_2.

Via emission factors

Emission factors can also be used in order to calculate CO_2 emissions. Considering a CO_2 fraction of 40 vol.-% of the biogas and the CO_2 from combustion, an emission factor of 91.5 g CO_2/MJ_{LHV} can be determined. This leads to an emission of 57.9 Mt/a.

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[7] Converted with higher heating value of 6.64 kWh/m³

[8] Stoichiometric calculation with pure oxygen as reagent. Source: Own calculation.

Basing the calculations on the emission factor from IPCC 2006 [92], which is equal to 54.6 g CO_2/MJ_{LHV} and is based on the calorific value, 34.6 Mt CO_2/year are released. The 34.6 Mt CO_2/year do not include the 40 vol.-% CO_2 fraction from biogas production, but only the CO_2 from combustion of biogas (approx. 40 vol.-% CO_2, 60 vol.-% CH_4). Summing up the latter and the CO_2 amount from biogas upgrading, which corresponds to the 40% CO_2 fraction (see section 3.2.3.2), it can be calculated, with the comparable order of magnitude, that approximately 55.6 Mt CO_2/year are emitted. This is nearly the same as using 91.5 g CO_2/MJ_{LHV} as the emission factor, and roughly higher than the stoichiometric result. Small deviations are derived from different data sources and rounding errors.

Potential limitation

In most cases, the upgraded biogas is injected into the gas grid. Therefore, in practice, the CO_2 derived from the combustion of biomethane is not available for CCU, unlike the CO_2 separated during the upgrading process. In 2017, 73 of 497 biomethane upgrading plants in Europe were not connected to the gas grid [8]. At these plants, the biomethane is directly utilized, which means that in some cases the CO_2 from combustion could be potentially harnessed for utilisation, except for places where the biomethane is used as biofuel for motor vehicles. In the latest Statistical Report from the EBA [66] the several incentives are presented to increase the use of upgraded biogas as a fuel. Especially in Sweden (88%) and Finland (25%) large amounts of the national production are already used as fuel for vehicles. Other countries, e.g., Estonia, Norway and Italy, are planning to strengthen this utilization pathway in the upcoming years.

3.2.4.2 Global CO_2 potential from biogas and biomethane combustion

Via emission factors

Considering the global biogas production of 1.27 EJ for 2014, approximately 116 Mt CO_2/year are released by biogas combustion, using the emission factor 91.5 g CO_2/MJ_{LHV}. Basing calculations on the emission factor from IPCC 2006 [92], 69.3 Mt CO_2/year are released. The 69.3 Mt CO_2/year do not include the 40 vol.-% CO_2 fraction from biogas production, but only CO_2 from the combustion of biogas. Summing up the latter and the CO_2 amount from biogas upgrading, which corresponds to the 40% CO_2 fraction (see section 3.2.3.2), it can be calculated that approximately 112 Mt CO_2/year are emitted. This is nearly the same as using 91.5 g CO_2/MJ_{LHV} as the emission factor. Small deviations are derived from different data sources and rounding errors.

3.3 *CO_2 potential from fossil sources*

The verified emissions registered in the EU ETS are allocated to their respective categories and their development in recent years compared, as illustrated in Figure 3-7. This figure shows that, on the one hand, the energy industry, and therefore the production of power and heat from fossil fuels, is by far the main emitter of fossil CO_2 in the EU. On the other hand, the energy industry is the only sector which continuously shows significant reductions in absolute emissions over recent years in the EU. Nevertheless, to achieve the goals of GHG emission reduction by 80–90% until 2050 compared to 1990 levels as stated by the European Commission [101], the use of fossil fuels in the energy sector must be reduced substantially or, rather, completely avoided. Therefore, the utilisation of fossil CO_2 from the energy industry is excluded from further analysis.

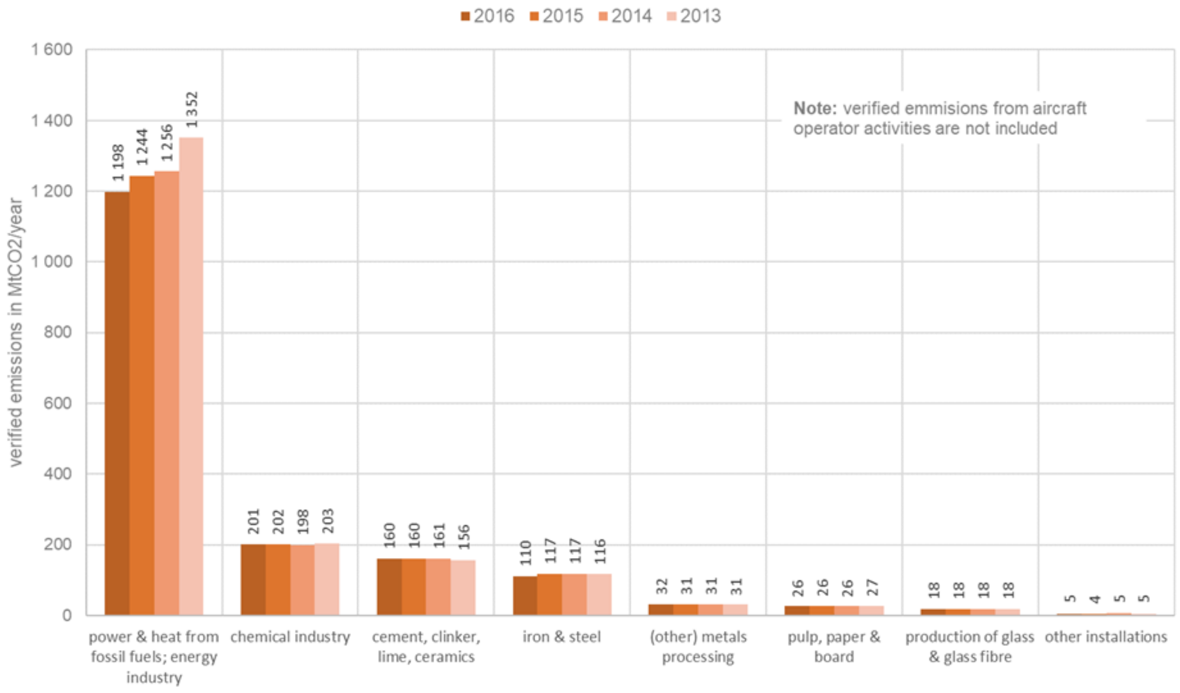

Figure 3-7: Total verified CO_2 emissions in the EU per industry sector.
Source: based on data from [87]

In this context, it must also be stated that the mobility sector has not been investigated according to its potential for serving as a carbon source for CO_2-based process chains. This is justified by the consideration that efficient capturing from the source in this sector is not expected to be feasible, with acceptable capture rates, in the mid-term. Additionally, the high decentralization of the emitters makes the industrial usage of captured CO_2 unviable.

3.4 *Incidence as point sources*

To establish Bio-CCU applications on an industrial scale, the centralized availability of resources is an important aspect. To maximize the economic and ecological advantages of such CO_2-based applications and reduce costs and efforts for transportation and storage, their operation near to the carbon emitting process is highly preferable. For an overview of which industry sectors provide highly centralized emissions of CO_2, the average per site emissions have been evaluated in this study based on the categorizations and data described above. The results are shown in Figure 3-8.

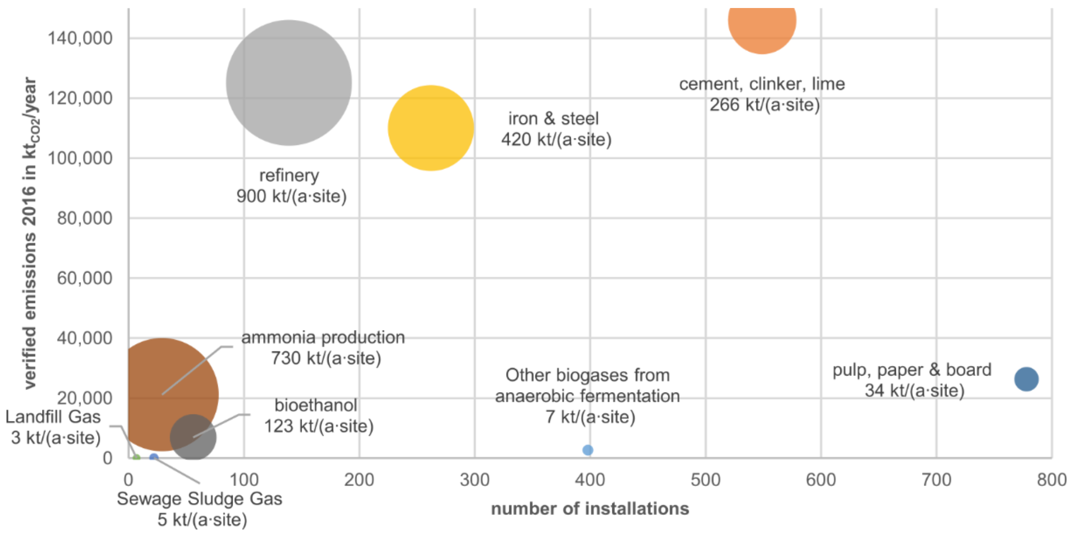

Figure 3-8: Average CO_2 emissions in the EU per site and industry sector.
Source: based on data from [87]

Figure 3-8 illustrates the highly centralized CO_2 emissions from iron and steel and the refinery industry, which provide an average amount of approximately 420 and 330 kilotons of CO_2 per year and site, respectively. Another remarkable sector is represented by the cement industry (including clinker, lime and ceramics production). While average emissions per site are well below the two major ones, the number of sites in total is significantly higher, which allows a more distributed installation of CO_2 utilizing technologies, and thus is advantageous in site selection.

With regard to these high amounts of centrally available CO_2, along with the fact that many industrial processes are highly established and efficient and are expected to still be available in the mid-term, it is reasonable to expect that these carbon sources be considered for renewable products. This especially applies to the steelmaking and cement industry, where a decarbonisation would imply a complete revision of the process chain. Therefore, when investigating resource potentials for future CO_2-based process chains at an industrial scale, these point sources should be considered as well.

3.5 *Technological capture rates*

The industrial processes covered by the categorization in Table 2-4 are significantly different from each other in terms of volume flows and purity of CO_2 in their flue or by-product gases. Additionally, each process allows the suitable use of one or more different capture technologies for efficient separation. This is particularly notable because efficiency often requires a compromise between the two, i.e., the process and capture efficiency. Consequently, highly different capture rates are technically and economically achievable, and thus may be actually implemented in existing processes and considered state of the art. These results are summarized in

Figure 3-9, showing the determined ranges and averages for the selected processes.

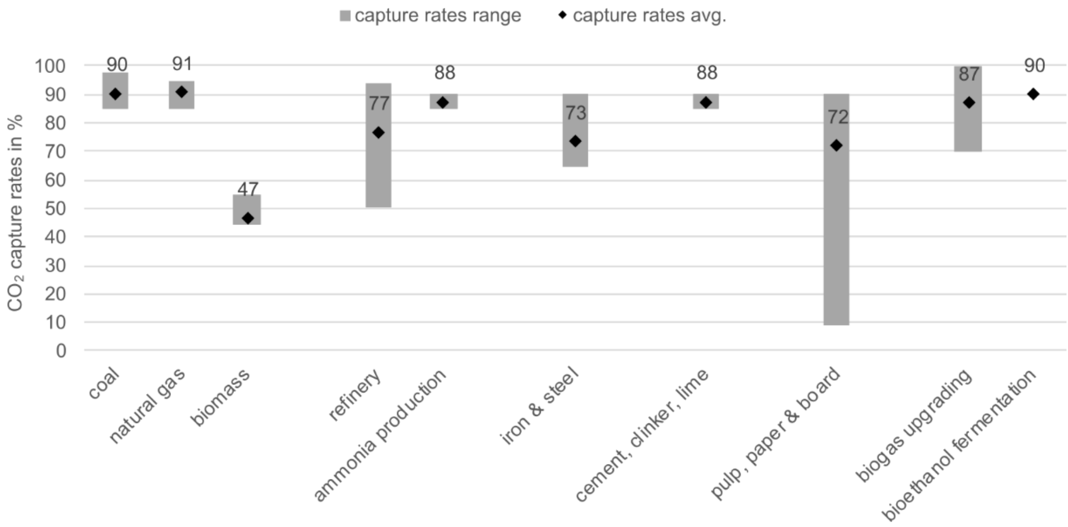

Figure 3-9: Affordable capture rates for various industrial processes (ranges and average).
Source: based on data from [23,26,88–91, 102–107]

As can be seen in

Figure 3-9, for some sub-categories listed in Table 2-4, appropriate values cannot be provided. On the one hand, this is caused by the rough definition related to the available data for CO_2 potentials, which does not further specify the underlying process (e.g., other chemicals, (other) metals processing). On the other hand, some processes and industrial sectors do not provide sufficient data to estimate appropriate capture rates (e.g., glass and glass fibre production, ceramics by firing) or do not allow feasible CO_2 capturing at all.

These capture potentials significantly reduce the amount of CO_2 that is utilizable from the total amounts emitted. Presuming the average capture rates as shown in

Figure 3-9, the resulting potentials for CO_2 from industrial sources are reduced to the amounts shown in Figure 3-10.

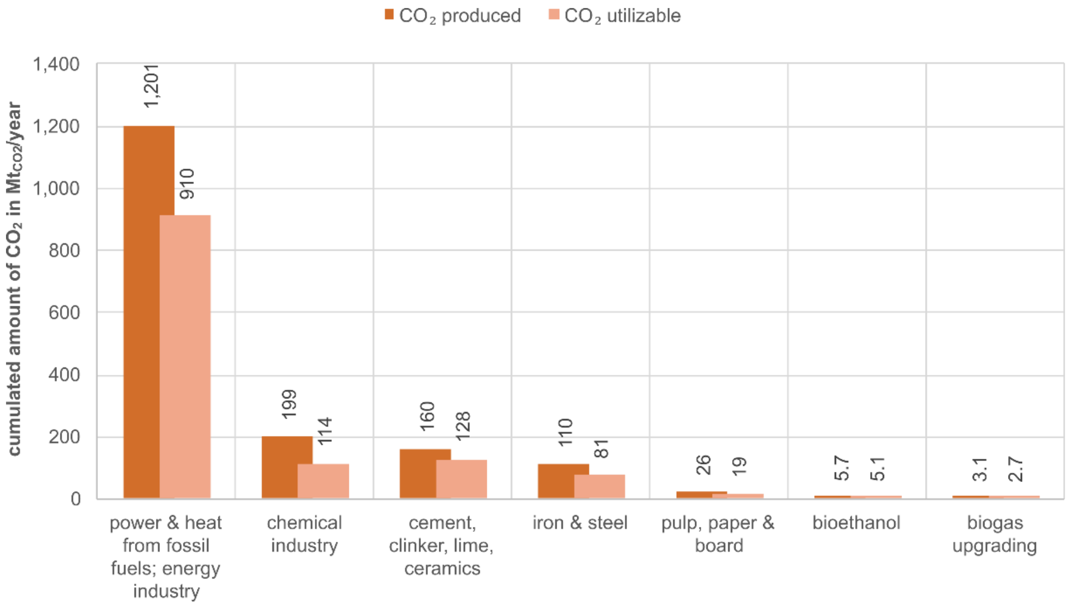

Figure 3-10: Comparison of produced and theoretically utilizable CO_2 per industrial sector.

The capture efficiency also has an impact on the utilizable CO_2 emissions per site as shown in Figure 3-11, according to the categorization given in Table 2-4. If fossil sources are considered, chemical industry processes, followed by iron and steel and cement production, would provide the highest amounts on utilizable CO_2 per site. In contrast, per site amounts for biogenic sources are rather low. Therefore, for the large-scale application of carbon capture and utilisation, fossil processes could be considered as a potential source for CO_2, as long as there are no environmental drawbacks and doing so does not support or elongate the deployment of processes that can and should instead be substituted by renewable approaches.

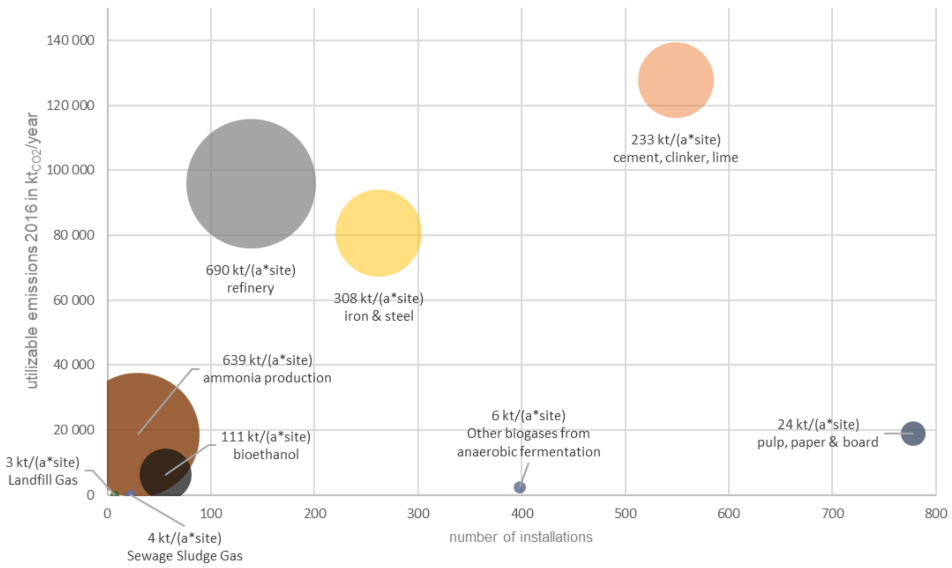

Figure 3-11: Utilizable CO_2 potentials from various industrial processes.

3.6 *Carbon capture cost*

Table 3-1 summarizes the gathered carbon capture costs for CO_2. Compared to the previous sections, the data herein was extended with CO_2 from fossil sources, though the acceptability of CO_2 from fossil sources for CCU must be further discussed (e.g., CO_2 may originate from waste gases from industrial processes which cannot be shifted to use as a renewable energy source, and therefore fossil CO_2 cannot be avoided).

CO_2 source		**capture costs** ***€/t_{CO2}***	**Year**	**exchange rate** ***USD/EUR***	**Ref.**
energy industry; power & heat from fossil fuels	coal	34 – 42	2017	0.83	[23]
		19 – 47	2015	-	[26]
		20 – 63	2015	0.72	[102]
	natural gas	63 – 83	2017	0,83	[23]
		54 – 101	2015	-	[26]
		35 – 75	2015	0.72	[102]
	biomass	54 – 101	2015	-	[26]
chemical industry	refinery	29 – 83	2017	0.83	[23]
		44 – 94	2015	-	[26]
		48 1)	2012	-	[103]
		97	2014	0.82	[90]
	ammonia production	12	2017	0.83	[23]
		23 – 54	2015	-	[26]
		22	2014	0.82	[90]
	other chemicals	12 – 52	2017	0.83	[23]
		21	2014	0.82	[90]
iron & steel production		19 – 33	2017	0.83	[23]
		16 – 41	2015	-	[26,103]
		81 – 83	2014	0.82	[90]
cement, clinker & lime production		22 – 35	2017	0.83	[23]
		33 – 69	2015	-	[26,103]
		17 – 37 1)	2012	-	[103]
		82	2014	0.82	[90]
pulp, paper & board production		18 – 27	2003	0.79	[91]
		57 – 87	2017	-	[105,107]
biogenic CO_2 sources	biogas upgrading	0 – 90	2012	-	[88]
		5 – 9	2015	-	[26]
	bioethanol fermentation	12	2017	0.83	[23]
		0 – 18	2011	-	[89]
		25	2014	0.82	[90]
		5 – 9	2015	-	[26]
	bioethanol fermentation (incl. cogeneration)	83 – 111	2011	-	[89]
		42	2003	0.79	[91]
Direct air capture		150 – 320	2012	-	[88]
		22 1)	2012	-	[88]
		150	2010	0.75	[108]
		331 – 423	2011	0.77	[109]
		268 – 309	2013	0.72	[110]
		341 – 475	2014	0.82	[111]
		81 – 201	2018	0.86	[112]

CO_2 source	capture costs *€/t*	Year	exchange rate *USD/EUR*	Ref.
	18 – 90 [1)]	2019	-	[31]
[1)] long term prediction				

Table 3-1: Average capture costs for CO_2 related to industrial sectors.

As can be seen, capture costs for CO_2 are highly dependent on the source used. Whereas capturing from diluted industrial flue gases (combustion of natural gas or solid biomass, refinery) ranges from €50–100 per ton, efforts for sources with high concentrations (biogas upgrading, industrial bioethanol fermentation, ammonia production, etc.) are substantially lower, reaching values clearly below €50 per ton. Because of low concentrations of CO_2, DAC shows the highest costs in conjunction with high uncertainties on account of the low maturity of DAC technology.

4 CONCLUSIONS

23.15 Mt CO_2 are derived from biogas production (anaerobic digestion process) as a by-product (~ 40 vol.-% CO_2 as by-product in biogas, ~ 60 vol.-% methane CH_4 in biogas) per year [93], of which approximately 3.14 Mt CO_2/year are already separated during biogas upgrading (> 95 vol.-% CO_2 off gas; CH_4 biomethane product gas) [8]. Considering the annual biogas production [93] and the annual upgraded biomethane [66,100], the utilisation of biogas, typically in combined heat and power plants, produces approximately 53 to 58 Mt CO_2/year including the CO_2 as a by-product from anaerobic digestion and the CO_2 derived from the combustion of methane content.

To sum this up, biogenic CO_2 from solid biofuel combustion (437 Mt CO_2), bioethanol fermentation (5.71 Mt CO_2), wine and beer production (1.48 and 1.44 Mt CO_2), biogas upgrading (3.14 Mt CO_2) and combustion of remaining biogas (53 to 58 Mt CO_2) amounts to approximately 506.7 Mt produced annually in Europe via assessed process routes. The assessment performed herein showed that, in reality, only part of this CO_2 potential is available for valorisation. Nevertheless, the amounts are vast.

Limitations of this study potentially are in the availability of up-to-date raw data, as comprehensive statistical data is difficult to access for some CO_2 sources due to missing recoding obligations. Data was validated and completed via desk research. Additionally, conversion factors from statistical raw data (for example "sewage sludge" in TJ) to yearly CO_2 mass amounts, might differ to other studies. Conversion factors were presented to preserve comparability to a maximum extent

There are several limiting factors for green CO_2 sources, as has been discussed in Section 2. First, solid, liquid and gaseous biofuels are distributed to many small applications, e.g., household fireplaces, motor vehicles and gas heating systems. The CO_2 emitted from these small consumers cannot be reasonably utilized, because of the high technical and economical effort required for collection and directed utilization. Furthermore, it is unlikely that the elaborate capture process would result in an ecological and sustainable system, which contradicts the intention of CCU. Additionally, CO_2 is already utilized as a raw material in the FAB and chemical industries. Other utilization pathways are offered by the water purification industry, the pulp and paper industry, the metal industry, welding, electronics, refrigerant gas and fire suppression technologies. A comprehensive review on the existing and emerging uses of CO_2 and their CO_2 demand was published in [113], [3] and [35].

The CO_2 produced during industrial bioethanol fermentation is already utilized to some extent, whereas the CO_2 accumulating in the beverage industry is reused nearly completely, especially in larger breweries and wine production sites. CO_2 from industrial bioethanol plants is often utilized as dry ice [114,115], gaseous fertilizer for green houses and food packing [12,35,116,117] or for carbonating beverages [12,35,41,114]. The CO_2 generated during fermentation processes in the beer and wine industry is often used for carbonating wine and beer directly [97,99,118] or as inert gas to preserve the beverages [35,59,61,119]. The major potential is therefore derived from biogas upgrading plants, the remaining CO_2 from bioethanol production and flue gases from biogas combustion, whereas the raw biogas (approx. 40 vol.-% CO_2 and 60 vol.-% CH_4) is combusted with air and the derived flue gas represents a lower CO_2 concentration (approx. 8-15 vol.-%). Consequently, this requires intense flue gas purification and separation, whereas biogas upgrading offers large amounts in high concentrations, but provides the auxiliary effect of potentially harmful trace compounds from potential CO_2 conversion technologies.

Depending on the biogas, e.g., landfill gas, sewage sludge gas, and biogas from anaerobic digestion with numerous substrates, various trace compounds can be present within the raw biogas. These impurities can be harmful in many ways to, e.g., CHP plants, as well as to other technologies and chemical reactions applied in CO_2 utilization. Section 3.2.3 provides more insight into these potential problems.

Capture costs for CO_2 are highly dependent on the source used. Whereas capturing from diluted industrial flue gases (combustion of natural gas or solid biomass, refinery) ranges from €50–100 per ton, efforts for sources with high concentrations (biogas upgrading, industrial bioethanol fermentation, ammonia production, etc.) are substantially lower, with values substantially below €50 per ton. Because of the low concentration of CO_2, DAC represents the highest costs in conjunction with high uncertainties on account of the low maturity of DAC technology.

Finally, further research should focus on the following questions: 1) How much of the European CO_2 potential can technically and economically be utilized via CCU? 2) How will the available CO_2 potential change with fulfilment of fossil energy/resource reduction goals and renewable energies/resources on the rise? 3) Which marketable CCU/CCS technologies will compete for CO_2 as a resource in the upcoming decades and how high is their CO_2 demand? 4) Which purity of CO_2 is needed for the different marketable CCU applications and how much does CO_2 purification influence the economic feasibility?

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