

han CO<sub>2</sub> in 2015 by sector (right).

s are evaluated. The focus of this paper is on non-fossil sions from fossil power generation and the construction shown to illustrate the gap between non-fossil and con- $O_2$  potential, and in the case of the cement industry, where lons are immanent to the production process. Within this process  $CO_2$  generated by the construction industry can be a sustainable if the possible input energy for the process is able (e.g. biochar or waste streams).

he steel industry, one of the major CO<sub>2</sub>-emitting industries, is not fidered in this study. Currently, fossil oil, gas and coal are used to be duce steel. Unlike the cement industry, there is a CO<sub>2</sub>-free operation to to produce steel [5,6]. The present day use of fossil fuel can be completely substituted by H<sub>2</sub> derived from renewable energy. This is technically feasible, as several studies have proven. Similar studies investigating CO<sub>2</sub> sources have also found the steel industry to be a depleted CO<sub>2</sub> source by 2050 [7]. By implication, in such a scenario the necessary amount of renewable energy must be provided, which corresponds to economic barriers. To address these would be beyond the scope of this study, and therefore we refer to other studies [5].

All the necessary data considered in this study were collected by official publication databases or calculated on that basis.

### 2.1. Emissions for CO2 from fossil sources

For decades, there has been an increase in energy consumption, combined with an increase in  $CO_2$  emissions; a countervailing trend is currently not observable [8]. Current global yearly anthropogenic  $CO_2$  emissions are 32.3 Gt/a (2015) [2]. These emissions mainly originate from the power, transportation and manufacturing industries. In Fig. 1, global anthropogenic  $CO_2$  emissions are illustrated. With 3.64 Gt/a (2015), the EU contributes contribute approximately 11% to global emissions and thus are a leading  $CO_2$  producers [9]. Germany emitted 0.79 Gt, approximately 22% of all EU-28 emissions in 2015, and is therefore the leading emitter in the EU (followed by the U.K. and France) and is at rank 6 in the world [10].

For Germany, the greenhouse gas emissions are shown in more detail in Table 1. The total emissions ( $CO_2 + CO_2$  equivalents) in the year 2015 were  $\tilde{\phantom{0}}$  887.4 Mt without LULUCF (land use, land use change, forestry) [10].

The majority of greenhouse gas emissions are caused by  $CO_2$  (~88%), followed by methane and nitrous oxide. From 1990 until today, there was a steady decrease in  $CO_2$  emissions, with a slowed reduction rate in recent years. To evaluate the theoretically possible amounts of  $CO_2$  for utilization, the following restrictions were applied to national trend tables for the German atmospheric emission [10]:

or for hyde. If d, it concuse cycle. Industry. To ations and inthe theoretical s was calculated. ble non-fossil CO<sub>2</sub>

'n

p<sub>2</sub> emissions into the atmoneasures for the prevention of capture and utilization of CO<sub>2</sub> d point sources with already high terms and economically-speaking, from unnecessary CO<sub>2</sub> concentration -scale units. However, decentralized evant for CCU technologies regarding a

er describes current and future quantities of . Hence, conventional (i.e. fossil power genruction industry) as well as renewable (e.g.

of all CO<sub>2</sub>equivalent emissions originate from non – CO<sub>2</sub>

emissions [16–18](VDZe.V.ÉÉ. These emissions are ngst 53 cement plants in Germany, of which 18 do not a clinker production. The highest regional concentration ne northern part of North Rhine-Westphalia (Ruhrgebiet)

 $_2$  emissions, incidental to the cement production, can be to three different sources: electricity-related (~13%), thermal elated (17%) and chemical reaction-related (70%) [20]. The hare of chemical reaction-related emissions is due to the calcineaction of calcium carbonate (CaCO<sub>3</sub>) to produce clinker (CaO), in in reaction Eq. 1 [10].

$$CO_3 \leftrightarrow CaO + CO_2$$
 (1)

The clinker produced with around 70 mass-% – depending on cement quality [21] – accounts for the largest component of cement.

Since most  $CO_2$  emissions are due to the immanent reactions that take place during clinker production, the only possibility to significantly reduce emissions in the cement industry is by means of  $CO_2$  sequestration [22].

For instance, the CEMCAP project [23] is dedicated to researching  ${\rm CO}_2$  capture from cement plants. For this purpose, several different  ${\rm CO}_2$  capture technologies suitable for integration into cement production are assessed theoretically as well as experimentally, namely amine scrubbing, the oxyfuel process (as a full and partial concept), membrane-assisted liquefaction, chilled ammonia process (CAP) and calcium looping (CaL). The goal of this project is to raise the TRL (technology readiness levels) of these capture technologies to at least 6. Furthermore, all technologies should be analysed in a techno-economic manner to decide which technologies are most suitable and promising for implementation in existing or future cement plants. [24]

In the following, the technical potential for the sequestration and utilization of  $CO_2$  from the German cement industry will be assessed. The technical potential is the amount that can be obtained with reasonable technical effort, while the economic potential lies within the technical potential and is restricted by economic constraints.

#### 2.2.1. Future CO2 potential

To evaluate the potential for  $CO_2$  sequestration and utilization in the cement industry, an inventory of the current emission status will first be made. The European Pollutant Release and Transfer Register (E-PRTR) requires that all pollution sources of air, water or soil exceeding a certain limit report their yearly emissions based on standardized measurements or calculation methodologies [25]. For  $CO_2$  emissions, all facilities with emissions above  $100~\rm kt_{CO2}/a$  are required to report. According to the E-PRTR, the total amount of  $CO_2$  emitted by cement production plants in 2015 totals  $24.63~\rm Mt_{CO2}$ . In contravention of the

cenarios
in the sobehaviour/
eduction scey 2050 against
s of two of these
are emitted by the
ons of these studies
as innovative future
retation systems, highly
thus, they show a high
iten do not give detailed
overview.

nes were identified as possible nat with a policy targeting 80% 0–78% fossil CO<sub>2</sub> and in 2050 CCU processes are available, with a O<sub>2</sub> is fossil-based and a further demistic scenario assumptions.

# onstruction industry

und 19 million tons of CO<sub>2</sub> a year, the German a significant contribution to anthropogenic

re. the

tudies

rd correction for the recession of total cented to be  $15\%_{2010}$  for 2030 and  $25\%_{2010}$  for of production shown in Table 3, above. Adare reduced to account for measures of  $CO_2$  action values of  $5\%_{2010}$  for 2030 and 2050 are  $b_2$  emission values for cement production have ast years [20]. The last columns show the amount utilization, with an assumed capture rate of 90%, with today's state of the art technologies [28]. As a r future development, the value for 2015, as reported 25] is given.

s state the technical potentials for the utilization of CO<sub>2</sub> e cement industry. Another downward adjustment must an considering economic feasibility. From a technical point of these sites are suitable for sequestration and the further of CO<sub>2</sub>. However, an economic analysis of CO<sub>2</sub> sources is the scope of this paper.

## CO<sub>2</sub> potential from biomass

Regarding the sustainability and long term perspective, renewable ources of  $\mathrm{CO}_2$  are evaluated within this study. Typical renewable sources of  $\mathrm{CO}_2$  are the biochemical conversion (biogas/biomethane pathway) and thermochemical conversion (gasification and methanation pathway) of biomass. In this study, we focus on the commodity potential of  $\mathrm{CO}_2$  from biochemical conversion, which is already well-established in the market.

The biogas/biomethane process shows high potential for future CO<sub>2</sub> capture and utilization. The typical biogas process consists of substrate pre-treatment (mainly crushing), fermentation (anaerobe digestion) and a post-treatment (mainly the removal of sulphur compounds like H<sub>2</sub>S). The produced biogas is a mixture of mainly methane and CO<sub>2</sub>, while methane is commonly the major part, constituting between 50% and 70% [29]. The biogas can either be used directly within a CHP process (combined heat and power) or upgraded to biomethane [30]. The typical bio-SNG (synthetic natural gas) process of thermochemical conversion is based on substrate pre-treatment (mainly crushing and drying), gasification, syngas treatment, methanation and upgrading [31]. The products of biomethane and bio-SNG are chemically identical (pure methane) and can be used as a substitute for natural gas. Within the upgrading process of both pathways (biogas/biomethane and bio-SNG), the CO<sub>2</sub> is separated as a by-product. Depending on the upgrading technology, it is diluted with air or is highly concentrated. While the biogas and biomethane processes are already established in the market, the bio-SNG process is still at the research and demonstration stage [32].

als or all and as emismore, the integration Although the proach, its apsolar radiation,

on could already be ing the fact that the have an effect on the

emitted and available for for the considered years of s, selected for further analysis

ature CO2 emissions by cement production.

	2030		2050	
CO <sub>2</sub> after	Total emissions after corrections	Available CO <sub>2</sub> after	Total emissions after corrections	Available CO <sub>2</sub> after
90%]	[-5%; -15%]	capture [90%]	[-5%; -25%]	capture [90%]
a	Mt <sub>CO2</sub> / a 19.70	Mt <sub>CO2</sub> / a 17.73	Mt <sub>CO2</sub> / a 17.24	Mt <sub>CO2</sub> / a 15.12

hechanism for the biogas plant shutdown, market models, are applied. Within this illability is evaluated by implying an optifuture for Germany, primarily based on the substrate.

itions for waste stream-focused biogas/biotil 2050 are as follows:

umber of biogas plants until 2024 (status quo) ase in the number of biogas upgrading plants through polated trend)

political framework for biogas and biomethane plants waste streams

ditation of the potential biomass resources (waste ams) until 2050

ation of energy crop-based biogas and biomethane plants ugh 2024

tween 2024 and 2034, the shutdown of energy crop-based ants to zero, a continuation of 90% of all existing biogas plants that are already based on waste streams (10% of the waste stream plants are closed down due to missing reinvestment)

- Potential biomass resources according to [37]
- Biogas-relevant streams (thus far unused fractions) were applied in this study
- Mainly manure and organic waste from kitchens, canteens, weekly markets as well as cereal straw
- In total, between 16 and 20.5 Mt sof biomass (dry matter), technical potential
- Assuming a constant amount of waste streams through 2050
- Exploitation of 50% of the average waste streams until 2050

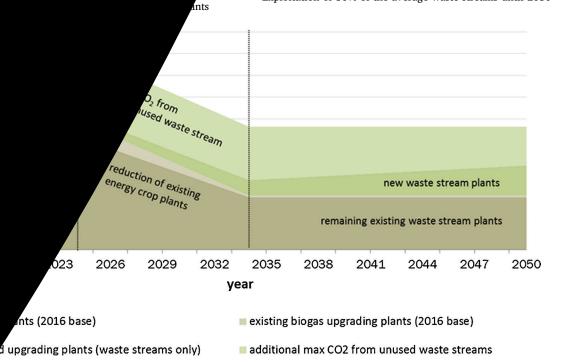


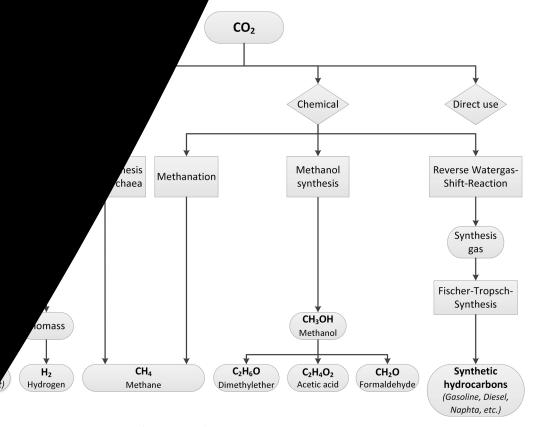
Fig. 3. Development of CO<sub>2</sub> (in Mt) from biogas and biogas upgrading through 2050.

le

example. The advantage of this kind of utilization of sun light without bypassing, the artificial photosynthesis is in an early hus out of the scope of this study.

s only little demand for CO<sub>2</sub> as a resource. all global emissions (32.3 GT CO<sub>2</sub> in 2015), the ation is fairly limited. The most suitable sources for heentrated with a high volumetric flow rate, such as from power plants or cement facilities (compare

dions in the literature for the yearly global demand of stock for CCU vary. In a review of publications dealing CCU potential, Aresta et al. (2013) found that all range ame maximum of 200 Mt CO<sub>2</sub> p.a. which are currently uti-46]. Within these limits, the majority of this demand is syninto urea (58%), inorganic carbonates (25%) and methanol lowever, already today there is further demand in the direct tion and CO<sub>2</sub> as a source for fuels or other materials. The share of t utilization (e.g. beverage carbonization, food packing and intrial gas) is about 9%. Table 5 gives a detailed overview of the trrent and near term (within the next 10 years) demand of CO<sub>2</sub> as a esource. The near term demand was calculated with ~ 250 Mt [44,45], with the highest increase in the material branch. There are also studies



**Fig. 4.** CCU pathways [39–41].

esis of chemical energy carriers. This of producing a broad range of fuels for nobile applications. For reasons of scope, Ill be considered in this paper. Described as nree – technically and economically – most of methane-, methanol- and Fischer-Tropsch ed and evaluated.

esses at hand based on energetic performance, described in the following will be used. These derived for the rating of energy conversion in and have been used in various publications [53–55]. ersion efficiency ( $\eta_{CCE}$ ) only balances energy flows ctant (H<sub>2</sub>) and product (fuel). The plant efficiency all energy flows inside the synthesis plant. Therefore, in reactant and product energy flows, electrical ( $\dot{E}_{el}$ ) and by ( $\dot{E}_{th}$ ) demand for the operation of the plant are also the energy flows of chemical streams are calculated via the  $\dot{m}$ ) and the lower heating values (LHV) of the respective

ical conversion efficiency:

$$\frac{LHV_{fuel} \cdot \dot{m}_{fuel}}{LHV_{H_2} \cdot \dot{m}_{H_2}}$$
 (2)

Plant efficiency:

$$\int_{plant} = \frac{LHV_{Fuel} \cdot \dot{m}_{fuel}}{LHV_{H} \cdot \dot{m}_{H_2} + \dot{E}_{el} + \dot{E}_{th}} 
 \tag{3}$$

#### 3.3.1. Methanation

Methane is a very common energy carrier worldwide. It is the main component of most resources of "natural gas". The most common use of methane or natural gas in Germany is for the supply of heat in households. Also, natural gas power plants have gained an increasing share of Germany's power production mix due to their good dynamic characteristics compared to the predominant existing coal power plants. Furthermore, natural gas can be used in cars as a substitute for gasoline in Otto engines. Because of its higher H:C ratio, this technology has the potential to reduce direct  $\mathrm{CO}_2$  emissions in passenger cars in comparison to conventional gasoline or diesel fuel.

The production of methane on the basis of sustainably-produced  $\rm H_2$  via electrolysis would therefore quickly find use in the existing infrastructure and markets.

Bailera et al. [56] compiled a list of existing power-to-gas projects and plants in 2017. The largest plant, in operation since 2013, is the Audi e-gas plant in the northern German town of Werlte. The plant operates with offshore wind, powering  $3\times 2.0~\text{MW}_{el}$  alkaline electrolyzers. It is expected to produce around 1000 tons of methane per year [57].

# Process description

The most prominent reactions taking place inside the methanation reactor are the following:

$$CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O\Delta_R H^0 = -206.3 \, kJ/mol$$
 (4)

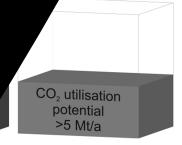
$$CO_2 + H_2 \leftrightarrow CO + H_2O \Delta_R H^0 = 41.2 \, kJ/mol \tag{5}$$

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \Delta_R H^0 = -165.2 \, kJ/mol$$
 (6)

The actual quality of the reactions taking place in the reactor is dependent on pressure, temperature and the infeed ratio of the

proxiituting ompares ) with the s upgrading on surpasses

kets for possible CO<sub>2</sub> compared to ally-reasonable imes must be identified. energy, as is used for



an CO<sub>2</sub> potential from biogas and biogas upgrading

1.02 1 19.918

O<sub>2</sub> conversion to methanol reaches a value 4-gas phase-separation, nearly all of the unecirculated. Table 7 shows the specific energy the methanol synthesis. [63] yield the following efficiency factors:

(12)

(13)

reports a range of 69–89% efficiency for methanol on a literature review, in accordance with the results

#### -Tropsch

ydrocarbons are considered an option for storing renewably energy. Due to their high volumetric energy density, liquid the main energy source for heavy load transportation, shipiation, as well as long-distance traffic [65]. In the near future, ain transportation concepts will continue to be based on liquid ocarbons. The increasing amount of CO<sub>2</sub> in the atmosphere must be taken into account. At this point, climactic changes due to exsive CO<sub>2</sub> content can only be countered with modern technological nterventions. There are many different technologies applied for the production of chemicals from CO<sub>2</sub>. Amongst these, Fischer-Tropsch (FT) synthesis is the most prominent route to producing liquid fuels [66].

# **Process description**

FT synthesis is a heterogeneously-catalyzed hydrogenation of CO with a polymerization character. Mainly liquid hydrocarbons are produced like diesel fuel, kerosene and gasoline [67]. Due to the catalytic process, the products of the synthesis are free of sulphur and produce less soot during combustion [68]. Thus, the diesel dilemma - soot or NO<sub>x</sub> emissions – can be leveraged. Synthetic fuels can be used to fulfill EU6 standards. The catalysts used for the FT synthesis are typically Febased or Co-based. For the FT synthesis, the feed is synthesis gas in a composition of H<sub>2</sub>/CO of slightly less than 2:1. Synthesis gas is conventionally provided through the steam reforming of natural gas. This process is called the gas-to-liquid (GtL) process. When solid biomass is gasified, instead of using natural gas as a feedstock, biomass-to-liquid (BtL) is the technical term. If CO2 and power are applied to producing liquid fuels, power-to-liquid is the relatively new wording [69,70]. All of these processes mainly differ in the generation of the synthesis gas (CO/H<sub>2</sub> mixture) and the adaptation of the right hydrogen to CO ratio.

When  $CO_2$  is considered a carbon source, a two-step process typically combines reverse water gas shift (rWGS) and FT synthesis as a potential process combination. For the reduction of carbon dioxide, additional hydrogen is required apart from the hydrogen for FT synthesis. The hydrocarbons produced by the FT process are typically separated from unreacted feed and gaseous hydrocarbons and, finally, upgraded via hydrocracking and isomerization to obtain the final product.

In the rWGS reaction,  $CO_2$  is converted into CO in accordance with the following equation:

$$CO_2 + H_2 \leftrightarrow CO + H_2O \Delta H^{\circ}_{r298K} = 415 \text{ kJ/mol}$$
 (14)

The reaction is endothermic and therefore thermodynamically-favored at high temperatures. In the rWGS stoichiometry, the number of

can al for thanol of mecommon

a chemical inly produced iso be made via umerous research and CO<sub>2</sub> feedstock. to-fuel plant is the ing International) in year of climate-friendly

on  $CO_2$  in Eq. 9 is commonly 0 and 11:

$$-130.98 \, kJ/mol$$
 (9)

$$.2 \, kJ/mol \tag{10}$$

$$28.13 \, kJ/mol \tag{11}$$

s designed and simulated by Otto [63] will in the following. The synthesis reactions take all reactor under 250 °C and 80 bar. With a Cu/ stem described by Pontzen [64] and a molar

lat twice the current German CO<sub>2</sub> demand supplied by the CO<sub>2</sub> from biogas and biogas at theoretical amount, see Section 2.3). Adfossil CO<sub>2</sub> through the cement industry could be current production capacity of non-fossil CO<sub>2</sub> sur-CO<sub>2</sub> demand. For a technically- and economically-entation of CCU products, novel market niches must hising possibility is the market for chemical energy sed, for example, as transportation fuels.

shows the high potential of the chosen CCU products thane and Fischer-Tropsch fuels) combined with the redemand in Germany. These products were chosen because dy well-established state of the art and good data available, er technologies like the enzymatic reaction and artificial thesis were not considered due to the early stage of developed lack of data, but are also a possible future pathway for CCU. Section 4.1, a detailed analysis of the related H<sub>2</sub> demand is ucted and evaluated. Concluding, Section 4.2 gives insight into the cent markets for the chosen CCU products and evaluates its potential lization for non-fossil CO<sub>2</sub> within this market.

# 4.1. Hydrogen demand

As is described above, hydrogen is necessary in order to produce methane, methanol or Fischer-Tropsch fuels from  $CO_2$ . Within this study, the potential sustainable  $CO_2$  in Germany today and for the years 2030 and 2050 has been calculated and analysed. For each technology process described in chapter 3.3, the resulting production potential (methanol, methane and Fischer-Tropsch fuels) were calculated, as well as the related  $H_2$  demand, as is shown in Table 9. The conversions were then computed under the assumption of ideal conditions, while losses due to non-ideal reaction conditions were not considered.

For 2030 and 2050, the amount of  $CO_2$  from biogas and biogas upgrading was merged due to higher uncertainties, which each process alone would have induced. Here, the total amount of biogas and biomethane, and thus of  $CO_2$ , decreases through 2050 because of the assumed shutdown of energy crop-based plants (compare Section 2.3). Within the considered scenario, the installation of new plants (mainly based on organic waste as a substrate) cannot compensate for the decrease in capacity and  $CO_2$  production. Regarding the cement industry, a decrease of capacity and  $CO_2$  production due to process optimization can also be seen. Thus, non-fossil  $CO_2$  sources, as well as fossil  $CO_2$  sources, decreased from 2015 through 2050 (within the framework of an achievement of the Paris agreement).

As is shown in the table above, the largest production potential today arises from the cement industry, which therefore would require the largest amount of hydrogen. The sustainable and economic production of  $H_2$  is one of the limiting factors to actually process sustainable  $CO_2$  into methane, methanol and Fischer-Tropsch fuels.

The highest  $H_2$  demand is caused by methanation (0.18 kg per kg of  $CO_2$ ). The methanol production and Fischer-Tropsch fuel production show almost the same rate of  $H_2$  demand (0.14 kg of  $H_2$  per kg  $CO_2$  for the methanol process to 0.13 kg of  $H_2$  per kg of  $CO_2$  for the Fischer-Tropsch fuel).

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adapmicroized apble 8 are

accordance

(18)

(19)

rs in Fischer-Tropsch be increased to 70% or hore specifically, re-comis pressure can be reduced reased conversion is also

# ropsch.

Mass [kg]	Energy [MJ]
0.533	96.682
4.185	-
_	2.238
1	44.121

	5.5
2.1	2.8
8.2	11.0
1.6	2.1
2.7	3.6
1.4	1.9

ata, no national demand for methanol be evalyearly production of methanol in Germany is 015 [86], which indicates a large national market. per ton of methanol, this resulted in a turnover of 015.

yearly demand of methane was around 77.2 Mt. Thus, if duated and available CO<sub>2</sub> from biogas and cement were d to methane, around 16% of fossil-based methane could with non-fossil product. Therefore, synthetic methane alone tion as a substitute for fossil methane. The same conclusion awn for the Fischer-Tropsch fuels. The substitution of these ed products with products based on non-fossil CO<sub>2</sub> is not an te solution alone. It must go along with decreasing consumption ssible other substitutes to meet the targeted GHG emission sav-

In contrast to methane and FT fuels, methanol has a much lower nsumption-respective production rate in Germany. The non-fossil  $O_2$  produced through methanol could substitute German production several times over. It must be kept in mind that for this scenario, the  $CO_2$  from highly decentralized point sources must be collected and utilized, and this relates to thus far unknown costs. In conclusion, the local methane production could be technically substituted by non-fossil  $CO_2$ , but with hitherto unknown costs.

For a future outlook of the product demands and production rates, no reliable data was available. However, trends indicate an increased consumption in the near term [80,86]. A study commissioned by the German Federal Ministry of Transport [80,87] give a range of 1275 to 1688 PJ energy demand for fuels in road transport (excluding electrified vehicles), depending on the scenario for the development of vehicle electrification. This corresponds to 26.6–39.2 Mt of diesel fuel.

# 5. Conclusions

This paper has shown that  $CO_2$  from non-fossil sources is a source of carbon that will continue to be available in the future. On the one hand, it functions as a resource, while on the other, it (temporally, with the option of a constant loop) reduces  $CO_2$  emissions into the atmosphere. So far, only a minority of the globally available  $CO_2$  is used for synthetic conversion into other commodities, despite the technical feasibility of so doing (see chapter 3.2). This indicates what will most likely be a barrier due to economic, political or acceptance reasons. Here, further research must be conducted to identify the specific reasons for hindrances and to develop counter measures. Additionally, more efforts must be made to promote the substitution of fossil-based carbon with non-fossil carbon, e.g. by biochemical conversion or cement production, as indicated in this paper.

This study highlights the limits but also the chances of biomass-respective non-fossil  ${\rm CO_2}$  as future carbon sources. It focuses on high volume products with high turnovers (see Table 11). In a future work, products with lower turnover but much higher value could also be

and in able 11).

with natural rischer-Tropsch the represented aluated in Section

In Germany for methanol, in [79–85].

	price in €/t**	turnover in bil. €	
	317 342	24.5 0.32	
	1,864 1,393 452	35.3 51.3 3.8	

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