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Use of carbon dioxide as raw material to close the carbon cycle for the German chemical and polymer industries

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ARTICLE INFO

Article history:
Received 20 December 2019
Received in revised form 8 June 2020
Accepted 10 June 2020
Available online 29 June 2020

Handling Editor: Bin Chen

Keywords:
Carbon dioxide utilization
Circular economy
Chemical industry
Carbon recycling
Resource availability

ABSTRACT

This article explores how far the use of CO2 as raw material could enable the German chemical and polymer industries to contribute to a circular economy. Material Flow Analysis was conducted for all carbon flows for material use in Germany, comprising chemical production, polymer production, domestic use and waste management. For scenario modelling, Carbon Capture and Utilization technologies were included, and key parameters determining carbon flows were altered to show potential corridors for the future development. The results show that current carbon flows are dominated by fossil sources and are highly linear, with a secondary input rate of only 6%. Additionally, 12% (2 Mt/a) of the primary carbon input is lost due to dissipation. Currently available Carbon Capture and Utilization technologies would allow reaching a secondary input rate of 65% for the chemical industry. However, to achieve this rate between 80% (processes of direct synthesis) and 103% (methanol-based processes) of the total net supply for renewable electricity in Germany would be required in 2030 and between 41% and 50% in 2050. In contrast, the unavoidable substance related CO2-point sources in Germany could probably fill the carbon requirement for material use of the chemical industry in 2050. The authors conclude that the utilization of CO2 as a carbon source is necessary to close the carbon cycle where material or chemical recycling is technically not feasible or reasonable. The very high demand for renewable electricity indicates that the required production facilities for CO2-based chemicals will probably not be completely based in Germany.

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1. Introduction

The reduction of greenhouse gas (GHG) emissions and a more efficient usage of natural resources are two important goals to reduce environmental pressures of production and consumption. Both goals in combination with concrete regulations are part of national (Federal Government of Germany, 2018) and international political strategies (European Commission, 2015) and legislation (German Federal Parliament, 2019, 2017b). To reduce GHG emissions it is necessary to substitute fossil resources in production and consumption. To raise the resource efficiency, natural resources should be used in a circular and not linear way within an economy.

Carbon plays a key role for today’s chemical and polymer industries for energy as well as material purposes. Carbon carriers like coal or hydrocarbons are burnt to generate heat, electricity or both. Furthermore, carbon is the basic material in organic chemistry. It is necessary to produce products like polymers, solvents or hygienic products. In Germany, 15% of the consumed petroleum is used as material input to produce organic chemical products (VCI, 2019). In both cases, the carbon emitted into the atmosphere as CO2 at the end of its life cycle. Since around 90% of the used resources are fossil based (VCI, 2019), the use of carbon as material leads to net emissions of CO2 (>20 Mt/a in Germany) (UBA, 2018).

While options exist to substitute carbon for energy related purposes, the material use of carbon is more challenging. Combustion processes to generate electricity and heat can be substituted with renewable power sources like wind and solar together with the electrification of heat production (Bazzanella and Ausfelder, 2017). In contrast, alternatives for the substitution of fossil carbon used as material are lacking.

Biogenic resources could be an alternative, but the expansion of the currently used amount is critical. An increased use of biogenic carbon sources to produce chemicals would lead to similar problems caused by the production of 1st generation biofuels, like an

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https://doi.org/10.1016/j.jclepro.2020.122775
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increase in eutrophication (Weiss et al., 2012). Since the amount of global cropland is limited, an increased use of cropland for material use would raise the competition with food crops (UNEP, 2014) and counteract the necessary development towards sustainable land use rates (Bringezu et al., 2012). Additionally, recent publications show that the risk of biodiversity losses would increase globally (Di Fulvio et al., 2019; IRP, 2017). Hence, the expansion of the use of biogenic sources for material use is not regarded as option in this article.

The utilization of CO2 as a source for carbon used as material seems like a more promising option. First, the feasibility is well described in theory and shown in practice. There are multiple technological options to use CO2 in combination with additionally produced H2 as a carbon source to produce a variety of organic chemicals (Mikkelsen et al., 2010; Styring et al., 2015). Moreover, Bringezu (2014) qualitatively showed pathways for a circular use of carbon within an economy using CO2 as the carbon source. Von der Assen et al. (2016) analyzed the availability of CO2-sources in Europe. They concluded that there are enough CO2 sources to satisfy a demand of 500 Mt CO2/a in Europe. Recent technology assessments show that technologies to produce CO2-based chemicals, so called Carbon Capture and Utilization (CCU) technologies, already have a high Technology Readiness Levels (TRL) (Carbon-Next, 2017; European Commission, 2019). Second, the utilization of CO2 as a carbon source can reduce the GHG emissions compared to fossil-based production processes. Life cycle analysis showed that if fossil carbon sources are replaced by CO2 in combination with H2 to produce base chemicals like methanol and olefins (Hoppe et al., 2017) or formic acid (Sternberg et al., 2017) the respective GHG emissions can be reduced significantly. Katelhön et al. (2019) calculated a technical potential for CCU technologies to reduce GHG emissions of the global chemical industry by 3.5 Gt CO2-equivalents per year in 2030. However, GHG emissions can only be reduced if renewable electricity (REE) is used as the main electricity source. For methanol, Hoppe (2018) calculated a break-even for GHG-emissions when 86% of the electricity used to produce the necessary H2 comes from renewable sources.

The existing research shows and assesses options and possible barriers for the application of CCU technologies to produce base chemicals with a focus on the process level. However, it does not quantify the possible contribution to a circular use of carbon, especially if the actual and future availability of CO2 and REE is considered. This study aims to answer whether the utilization of CO2 can substitute the input of fossil based primary carbon on a sectoral level and thereby enhance the circularity of carbon for material use in the German chemical and polymer industries by the following steps:

1) The existing carbon flows for material use in the German chemical and polymer industries as well as the respective phases of domestic use and waste management are quantified using Material Flow Analysis (MFA). 2) The circularity of the carbon flows was assessed and a scenario analysis for a possible future use of CO2 as a carbon source is conducted. 3) The results of the scenarios were compared to the actual and future availability of REE and CO2-point sources in Germany.

2. Methods and data

MFA connects the source of a material, its pathways, intermediate conditions and final sinks (Brunner and Rechberger, 2017). Hence, to examine the circularity of material carbon flows in Germany, an MFA model for material carbon was developed. It covers the German chemical and polymer industries as well as further life cycle phases. The MFA model was then used to calculate the status quo of material carbon flows in Germany as well as future scenarios which include the use of CO2 as a carbon source. A mathematical description of the model is given in the Supplementary Information (SI-SI-1).

2.1. System definition and flow characterization

The system contains the process aggregates Chemical Production, Polymer Production, Domestic Use and Waste Management (CPUW-System) (Fig. 1). The carbon flows were determined for 2017. The geographical boundary for the system is Germany. Foreign trade balances were regarded for every carbon flow. The refinery sector was not included into the system since it is seen as a separate sector which provides fossil based raw material to the chemical industry. Hence, the flow analysis starts with the carbon flows from the refinery sector into the chemical industry. The definition of and delineation between the process aggregates Chemical and Polymer Production was done in accordance with production statistics (Destatis, 2018). The process aggregate Domestic Use describes the domestic consumption and storage of material in the economy. Finally, all post-consumer (pc) waste flows get processed in the Waste Management. Recycling processes were distinguished by their input (industrial or post-consumer waste flows).

To calculate the carbon input of the system, two sets of parameters are necessary. The first set contains the amount of non-energy-wise used energy carriers, i.e. energy carriers used as raw material for the production of chemicals or polymers instead of the use as fuel (Destatis, 2019a). The second set contains the stoichiometric carbon content of the respective energy carriers. Finally, the carbon input flows for material use are quantified by multiplying the amount of the non-energy-wise used energy carriers with the specific carbon content. This calculation procedure was done likewise for all following product and waste flows, within and out of the system.

In case of complex and highly variant material compositions or a lack of flow data, the carbon flows were estimated using mean values from literature or with the help of the physical balance of the process aggregates. Details of all input and output flows and the respective data sources are given in the SI-2.

2.2. Circularity indicators

The circularity of the carbon flows for the whole system and each aggregate was calculated and evaluated with three indicators derived from Helander et al. (2019). First, the Secondary Input Rate (SI-Rate) measures the percentage of secondary material within the total material input. Unlike primary material, the secondary material is extracted out of the technosphere and not the geosphere. Second, the Recycling Rate (R-Rate) measures the percentage of waste material which is actually used as material again. Third, the Loss Rate (L-Rate) measures the percentage of the total input which is lost due to emission, dissipation or foreign trade (Fig. 2).

2.3. Scenario analysis

To derive knowledge about future carbon flows and the impact of CCU technologies in the CPUW-System, a scenario analysis for the years 2030 and 2050 was conducted based on the described MFA model. In this section, the assumptions for the model parameters in the different scenarios are described. An overview of the scenario parameters is given in table 7 in the SI-3. The section continues with a description of the regarded CCU technologies and CO2-sources to supply secondary carbon to the CPUW-System. Finally, the future availability of the basic resources for sustainable CCU processes, CO2 and renewable electricity, is projected for Germany.
2.3.1. Scenario assumptions

The following parameters are identical in all scenario cases: (1) The development of production volumes and the trade balances is assumed to be constant, projecting the past trends between 2012 and 2018. The respective economic data is based on federal statistics (Destatis, 2019b) and production statistics from the German Association of Chemical Industries (VCI, 2018, 2017, 2016, 2015, 2014, 2013, 2012). (2) Biogenic carbon input is limited to the amount used today, due to sustainability reasons. (3) For the CO2 sequestration from industrial point sources a maximum sequestration rate of 90% (Bui et al., 2018) is considered.

In contrast, the development of material recycling rates, the application rate of CCU-technologies and net additions to the anthropogenic stock are modified to calculate the following scenarios for the years 2030 and 2050.

The Low Circularity (LC) Scenario represents a business-as-usual development. The rates for material recycling are developing as they did in the past five years (+0.5% per year). As a minimum goal, the recycling rates demanded by actual German legislation are reached (German Federal Parliament, 2017a). CCU technologies are slowly applied and the net additions to the anthropogenic stock gradually decline from 48% (2017) to 20% (2050).

The High Circularity (HC) scenario represents an ambitious development where a completely regenerative (secondary input + biogenic input) carbon supply for the system is reached by 2050. Therefore, the annual growth rates for material recycling are twice as high (+1% per year) compared to the LC scenario and the application of CCU technologies is accelerated. A flow equilibrium is reached for the net additions to the anthropogenic stock by 2050, as expected by Deilmann et al. (2009) as well as Gruhler and Böhm (2011) for the German building sector. To avoid an oversupply of carbon in the model, it was assumed that primary fossil carbon input is substituted by secondary carbon input. Also, increased material recycling substitutes the production of primary chemicals which leads to a reduced carbon demand of the chemical industry while the production volume of the polymer industry remains constant.

Fig. 1. Overview of the main carbon flows for material use in the CPUW (Chemical Production, Polymer Production, Domestic Use, Waste Management) system. (PC – Post Consumer).

Fig. 2. Overview of the used indicators to evaluate the circularity of the system and the subsystems. (PI – Primary Input; SI – Secondary Input; L – Losses; P – Products; W – Waste; SI-Rate – Secondary Input Rate; R-Rate – Recycling Rate; L-Rate – Loss Rate).
2.3.2. CCU-technologies

Base chemicals like methanol, olefins and aromatics represent the first production step in the chemical industry. They are synthesized out of crude oil and natural gas derivatives and serve as the basis for other synthesis steps and chemical products. Instead of using fossil energy carriers they can be synthesized directly or indirectly using CO₂, H₂O and electricity as process input (Fig. 3). According to recent studies the TRL of the available technologies lie between 1 and 8 (Carbon-Next, 2017; European Commission, 2019). This means, that there are already market ready technologies while at the same time fundamental research and upscaling of technologies is in progress.

To address the uncertainty of future development and to take possible future energy efficiency gains into account, two different CO₂-based process routes were considered. They substitute the fossil-based production of base chemicals in the model, where technologically feasible. For the first process route CO₂-based production of methanol in combination with the methanol-based production of olefins and aromatics is considered. This process route represents the state of the art (TRL 8). For the second process route, the direct synthesis of methanol, olefins and aromatics, using CO₂ and H₂ is considered. The respective synthesis processes require stoichiometrically less H₂ and therefore less energy but are currently only available at lab scale (TRL 4). Details about the considered processes are given in Tables 8 and 9 in the SI-4.

2.3.3. CO₂-sources

CO₂ can be sequestered from the atmosphere via Direct Air Capture (DAC) as well as from a variety of high concentrated industrial point sources. The availability of CO₂ in the atmosphere is hardly limited and the utilization can be considered as sustainable and circular. However, the sequestration process is comparably energy intensive because of the low CO₂-concentration in the atmosphere. DAC technologies using solid amine-based filters were already demonstrated on a high TRL (7–8) but only in cases where cheap excess heat is available (Fasihi et al., 2019). For CO₂-point sources, the CO₂ sequestration based on aqueous monoethanolamine absorbents is already used in various industrial applications with a TRL of 9 and a comparably high sequestration rate of 90% (Bui et al., 2018). Therefore, it was assumed that this technology or a different technology with a similar sequestration rate can be applied for all cases presented in Table 1. However, the availability of a specific point source for CCU technologies was determined by certain criteria to ensure that emissions avoidance has a higher priority than emission utilization. CO₂ point sources whose only function is the burning or processing of fossil energy carriers, like coal, lignite and natural gas fired power plants or refineries are not considered. Their future substitution in Germany is probable and necessary to reach the GHG reduction goals. In 2017, these sources accounted for a total emission volume of 478 Mt CO₂/a (DEHSt, 2018) or 60% of the total CO₂-emissions in Germany (UBA, 2019b). Furthermore, CO₂-emissions from the remaining point sources are distinguished into energy and substance related emissions according to UBA (2019b). Energy related emissions usually result from the incineration of fossil fuels, and thus are neither regarded as sustainable sources of CO₂. Substance related emissions have their origin in the processing of materials and the related stoichiometry (Table 1).

Currently, substance related emissions usually come along with energy related emissions (except for Biogas Production) because of the energy requirement of the respective process. The energy related emissions can be avoided by substitution of the energy supply (or higher energy efficiency). In contrast, substance related emissions can only be avoided if the basic technology for the provision of materials such as cement and biogas is substituted as well. In 2017, the substance related emissions in Germany accounted for 84 Mt of CO₂/a (Table 10 in SI-5) or 11% of the total CO₂-emissions in Germany (UBA, 2019b).

Three of the possible substance related point sources are also considered as avoidable in the longer term (Table 1). The respective emissions are considered as an available CO₂-source in 2030 but not in 2050. First, the substance related emissions of ammonia (NH₃)
production can be avoided by using electrolysis instead of methane to produce the required H₂. Therefore, the substance related emissions of the ammonia production are considered avoidable. Second, the non-energy related emissions of petrochemical processes will be avoided if CO₂ is used as a carbon source instead of fossil energy carriers. Third, substance related emissions of iron and non-iron metal production with coke as a reduction medium can be avoided using direct reduction technologies via H₂ (Arcelor Mittal, 2019).

The future amount of CO₂-emissions from the considered point-sources was derived from the economic development of the industries based on Billig et al. (2019) for biogas production plants and Pflüger et al. (2017) for the other sources except for waste incineration. For biogas production plants, the mean amount of emitted CO₂ is lower than 2 kt CO₂/a which is at least two magnitudes lower than for the other point sources. Therefore, these point sources are categorized as small point sources while the other point sources are categorized as large point sources.

The future development of waste flows is uncertain due to the development of the addition to the anthropogenic stock and recycling technologies. Therefore, a range for the emissions from waste incineration was calculated. Values from an actual study from the German Environment Agency serve as lower limits as they extrapolate the current development into the future (UBA, 2018). As the upper limit serve the values which have been calculated in the scenario analysis taking changes in the stock accumulation and recycling technologies into account.

Detailed information about the actual and future emission volumes of the considered substance related CO₂-point sources are given in the SI-5.

Since one objective of the scenario analysis is to assess whether sufficient long-term CO₂-sources are available to cover the CO₂-demand, there is no general economic or environmental merit order in the usage of the CO₂-point sources. All considered large sources are expected to get used by the same percentage of their total emission volume. The only assumptions are, that small point sources are getting used with a lower percentage because of their comparably very low emission volume. Furthermore, because of the comparably high energy demand, DAC is only considered if the CO₂-requirement is higher than the availability of point-sources.

2.3.4. Renewable electricity

To estimate the supply of REE a literature review of available energy models for Germany was conducted. The following criteria were applied to choose the respective energy models from which the REE availability was derived for 2030 and 2050:

- the fulfillment of GHG emissions reduction goals for Germany (-55% by 2030 and -95% by 2050 compared to 1990 (Federal Government of Germany, 2018))
- a phase out of coal and lignite fired power plants at the latest by 2038 (BMWi, 2019) in combination with a share of 65% renewable energy for the gross electricity consumption in Germany in 2030 (Federal Government of Germany, 2019)
- the consideration of non-energetic usage of energy carriers such as crude oil derivatives.

The last criterion could only be fulfilled for the energy model chosen for 2050 but not for 2030 since non-energetic usage is hardly considered in current energy models. For 2030 a domestic availability of REE (net amount) of 374 TWh/a was derived from an official forecast scenario of the German transmission grid operators (UBN, 2019). For 2050 an availability of 777 TWh/a was derived from an cost optimized and multisectoral energy model used in a recent study from the German environmental agency (UBA, 2019a).

3. Results

In this section, the status quo for material carbon flows in the CPUW-System as well as the results of the described scenario calculations are analyzed and assessed.

3.1. Status quo

In 2017, the carbon flow in the CPUW-System was mostly fossil-based as well as linear (Fig. 4).

In total 19 Mt of carbon (including recycled carbon) were converted and used as material in the system. Fossil resources accounted for 89% (crude oil derivatives: 81%; natural gas: 8%) and biogenic sources for 11% of the carbon input. Two thirds of the carbon are used to produce polymers and polymer products while one third is used in form of non-polymer products such as paints, coatings, carbon black, cosmetic products or washing powder. The net-addition to the anthropogenic stock of 7 Mt shows that a high share of the carbon input is additionally accumulated in the economy. The highest losses (5 Mt) occur in waste management due to waste incineration. The dissipation into the environment also accounts for significant losses (2 Mt). It occurs using chemical products, for example, when the solvent component of a coating evaporates after its application. Even though there is recycling of industrial and pc-waste, the amount of carbon recycled is rather low compared to the primary input.

Table 2 shows the results for the circularity indicators of the CPUW-System. They express the strong linearity of the system as well as differences between the industries. All values for the SI-Rate

### Table 1: Industrial Point Sources for substance related CO₂-Emissions.

| Substance Related CO₂-Source | Cause of Emission | Avoidable [yes/no] |
|-----------------------------|-------------------|--------------------|
| Ammonia Production          | Production of H₂ via Steam Reforming and Water Gas Shift Reaction | yes |
| | CH₄ + H₂O → CO + 3 H₂; CO + H₂O → CO₂ + H₂; | |
| | N₂ + 3 H₂ → 2 NH₃ | |
| Biogas Production           | The product gas of the fermentation process contains between 30% and 50% of CO₂ (Brosowski et al., 2014) | no |
| | Calcination reaction: CaCO₃ → CaO + CO₂ | no |
| Cement or Limestone Production | Carbonates are used as raw material. The contained CO₂ gets emitted during the melting process (UBA, 2019b) | no |
| | Use of carbon (coke) as reduction medium to reduce the metal-containing oxides (UBA, 2019b) | yes |
| Iron and Non-Iron Metal Production | Part of non-energetically used energy carriers which cannot be utilized for material production and gets burned without energy recovery (UBA, 2019b) | no |
| Soda Production             | CaCO₃ is used as raw material. The contained CO₂ is only partly bounded in the product (Na₂CO₃) (UBA, 2019b) | no |
| Waste Incineration           | Incineration of carbon containing solid waste | no |
are below 10% or even 1%. Comparison of the SI-Rates shows that the share of used secondary material is significantly higher in the polymer industry than in the chemical industry. The R-Rates for industrial recycling are higher with a factor 3.5 to 4 than for pc-recycling. This is a result of purer waste streams within the industry. However, the high recycling rates within the industry do not have a significant influence on the overall R-Rate (18%) because of their comparably low magnitude. Especially for the chemical industry, the existing recycling flows hardly have a relevance. Additionally, more than half of the carbon input is lost in the form of emissions caused by waste incineration, dissipation and foreign trade. The latter accounts for only 28% of the losses. Therefore, 72% of the losses, or 41% of the total system input, are definitively lost for further use if CCU technologies are not considered.

3.2. Scenario results

The scenario results can be structured in two parts. In the first part the resulting material flows as well as the respective circularity indicators are presented. In the second part the availability and the requirement of CO2 as well as renewable electricity are compared to each other.

3.2.1. MFA and circularity indicators

The scenario analysis outlines a broadening corridor of potential future development (Table 3). In the LC scenario only a moderate circularity for carbon used as material can be achieved. Although the circularity indicators are much higher in 2050 than in the status quo, about two thirds of the carbon input of the overall system would still be primary and fossil. Additionally, in none of the industries a SI-Rate greater than 50% is achieved. In the HC scenario a completely regenerative carbon input is achieved for the whole system. This means, that no fossil carbon input is necessary anymore. This is caused by the utilization of CO2 as carbon source as well as the increased rates for recycling in combination with the absence of a net addition to the anthropogenic stock. The latter implies higher amounts of waste streams which can be recycled.

Fig. 5 shows the carbon flows in the HC scenario for 2050. The carbon input of the chemical industry is reduced to 15 Mt with CO2 as the main carbon source. Only for the polymer sector material recycling plays a significant role as a carbon source. Since carbon losses are not completely avoided, a closed carbon cycle is not possible. Therefore other substance related point sources and DAC are used to compensate the losses.

3.2.2. CO2-demand vs. availability

To compare the CO2-availability with the CO2-requirement and to gain information about which CO2-sources would be necessary to fulfill the different scenario requirements, the CO2-availability is subdivided. For the subdivision, the emission volumes of the CO2-sources given in SI-5 (Table 10) are distinguished between their avoidability, the scale of the specific emission volume and the development of waste flows:
- **Substance related emissions — lower limit (large sources):** Unavoidable substance related emissions of large point sources + low amount of waste flows
- **Substance related emissions — lower limit (all sources):** Unavoidable substance related emissions of large and small point sources + low amount of waste flows
- **Substance related emissions — higher limit:** All substance related emissions of large and small point sources + high amount of waste flows

To calculate the CO$_2$-requirement, the efficiency losses in the sequestration process were added to the CO$_2$-demand of the applied CCU processes.

If the lower limit is considered, the CO$_2$-availability (33 Mt/a) would not suffice to fulfill the CO$_2$-requirement in 2050 in the HC scenario (53 Mt/a), even if small point sources are also considered (44 Mt/a) (Fig. 6). However, if the higher limit (59 Mt/a) is considered, the CO$_2$-availability of the respective point sources would be higher than the CO$_2$-requirement in all cases. Nevertheless, in the HC scenario DAC is considered as a CO$_2$-source in 2050 (5 Mt/a) since it is assumed that less than 100% of the point sources’ emissions are available for CCU processes out of technical reasons. For the LC scenario, the availability of the lower limit of large sources would suffice to fulfill the CO$_2$-requirement for both scenarios in 2030 (LC: 3 Mt/a; HC: 19 Mt/a) as well as for the LC scenario in 2050 (13 Mt/a).

### 3.2.3. REE-demand versus domestic supply

With the currently known CCU-Technologies to produce base chemicals a maximum SI-Rate of 65% can be achieved. The production of high-volume base chemicals like buta-1,3-diene and butylene or other chemical products like carbon black cannot be substituted with CO$_2$-based processes, yet. Thus, it was analyzed how high the SI-Rate could be using the available REE to produce the H$_2$-demand for the described CCU process routes.

The results show that the use of CO$_2$ as raw material at large scale would require significant amounts of REE. In 2018, the total
net REE-supply in Germany would have been required to achieve a SI-Rate between 44% and 50% (Fig. 7). Due to the growing national production of REE, in 2030 the maximum SI-Rate of 65% could be achieved only for processes of direct synthesis using 80% of the domestic supply. For methanol-based processes, the REE-demand lies at 103% and would therefore exceed total domestic REE-supply. In 2050, 50% of domestic REE-supply would be needed for the methanol-based processes and 41% for the processes of direct synthesis. In general, between 1.2 and 1.6% (2030) and 0.6–0.8% (2050) of the domestic REE-Supply would be necessary to raise the SI-Rate of the chemical production by 1%. At the same time, methanol-based processes require 22% more REE compared to the processes of direct synthesis.

4. Discussion

This article quantifies and evaluates for the first time the circularity for carbon used as material in the German chemical and polymer industries and the possible contribution of CCU processes to enhance the circularity. In general, the results of this article imply, that the utilization of CO₂ as a carbon source can lead to a more circular use for carbon used as material in the German chemical and polymer industries. However, the required resources are limited in Germany. The authors will shortly discuss the reliability of the used data, the comparability of the results with other studies and implications for practice.

The main data sources are federal statistics for Germany and

![Fig. 6. CO₂-Requirement vs. Availability in 2030 and 2050 in Germany. (LC – Low Circularity; HC – High Circularity).](image)

![Fig. 7. REE-Supply vs. Demand in Germany (SI-Rate – Secondary Input Rate).](image)
Europe, recent scientific publications or life cycle databases. Therefore, data reliability as well as temporal and geographical correlation can be regarded as very good. Nevertheless, due to the combination of databases with different system boundaries and data gaps several uncertainties had to be considered. They are described in detail in the SI-6. However, these uncertainties do not affect the order of magnitude and main ratios of the shown material flows in the system but should be addressed in further research.

The SI-Rates for carbon used as material of 0.2% (Chemical Production) and 9% (Polymer Production) together with the PC-Recycling Rate of 18% show the high linearity of the carbon flows. The linearity is also illustrated by a comparison with other material flows in Germany. For instance, the SI-Rate for steel in Germany of 45% (BDE, 2018) shows, that the SI-Rates for carbon in the polymer industry and especially in the chemical industry are significantly lower than for other materials.

The availability of CO₂ to fulfill the industries’ demand for carbon used as material depends on the selection criteria for the CO₂-sources. If all avoidable and non-avoidable substance related CO₂-emissions of industrial point-sources are considered, the CO₂-requirement for a largely closed cycle can be fulfilled within Germany. Even without considering fossil fired power plants and re-finedery! This confirms the results of von der Assen et al. (2016).

However, the CO₂ emissions from ammonia production, iron and non-iron metal production as well as petrochemical processes may not be considered as long-term CO₂ sources for CCU processes. Technologies for the substitution of carbon as reduction agent or hydrogen carrier already exist on low TRLs wherefore the long-term focus should be on emission avoidance instead of utilization. If only non-avoidable CO₂-emissions are considered for CCU processes, the CO₂-availability from point sources would not necessarily fulfill the CO₂-requirement to close the carbon cycle in 2050. In all other and less ambitious cases, the long-term non-avoidable CO₂ sources would be sufficient to fulfill the carbon demand for CCU processes. This shows that the necessity of DAC application depends on how effective the CO₂-emissions form non-avoidable point-sources are used, the development of waste streams as well as the targeted level of circularity.

For several reasons, the most critical factor for the utilization of CO₂ as a carbon source is the high requirement of REE. The results of this article confirm the results of previous studies. Geres et al. (2019) calculated an even higher energy requirement (665 TWh/a) for the German chemical industry to substitute the non-energetical use of fossil hydrocarbons in 2050. On global scale Kätelhön et al. (2019) concluded, that between 18 and 32 PWh/a would be required to produce the 20 base chemicals with the highest production volume on basis of CO₂ in 2030. This corresponds to 67% and 119% of the actual global electricity production of 27 PWh/a (IEA, 2019). Moreover, the use of REE for CCU may not provide the highest environmental benefit. For example, the direct utilization of REE for battery electric vehicles or heat pumps would have a higher GHG mitigation potential (Sternberg and Bardow, 2015). Another constraint for the planning of CCU installations is that in the current models for the future German energy system, the chemical industry is hardly seen as a consumer of REE to produce non-energetically used hydrocarbons. If CCU productions plants are in Germany, there will be a competition for REE between the chemical industry and other sectors like transport or heating. This needs to be considered in future scenarios for sustainable energy and material supply. Furthermore, the limited availability of REE will also have a limiting impact on the CO₂-requirement. Therefore it seems probable that the existing non-avoidable substance related point sources will fulfill the future CO₂-requirement for CCU processes based in Germany. In contrast, the application of DAC as a CO₂-source in Germany seems rather unlikely.

In general, the chemical and polymer industries will have to consider the possible future role of CO₂-utilization and the options for a more circular use of carbon. On the one hand, the analysis clearly showed that due to non-avoidable dissipative losses in the use phase and a net export, recycling of waste flows is by no means sufficient to secure a regenerative supply of carbon. The use of CO₂ as a carbon source is necessary to compensate these carbon losses. Furthermore, the expansion of material recycling processes is already critical and uncertain due to technical constraints (Rudolph et al., 2017). On the other hand, there are barriers for the application of CCU processes. First, they require high amounts of the scarce resource REE, since the energy hitherto bound in fossil energy carriers must be provided in a renewable way. Second, actual estimations of production costs for CO₂-based base chemicals are much higher than for fossil-based alternatives (Hoppe et al., 2018). This is mainly caused by high energy costs and the still small scales. Third, chemical recycling processes such as pyrolysis or gasification also offer the potential to recycle carbon in waste flows which cannot be materially recycled (Stapf et al., 2019). However, this technology is limited to waste flows and cannot compensate dissipative losses. Therefore, ecological and economical aspects of CCU as well as material and chemical recycling technologies need to be compared systematically to identify favorable application fields. Additionally, there are regions with higher availability and lower costs for REE than in Germany, for example the Maghreb region (Agora, 2018). Here, the production of CO₂-based chemicals is likely to be more competitive than in Germany. Hence, it is unlikely that the German chemical industry will produce all required CO₂-based chemicals on itself but import relevant shares. However, the targeted transition of the current energy system towards mostly renewable energy sources requires concepts for efficient and large-scale energy storages. Synthetic gases like H₂ or CH₄ as well as synthetic Hydrocarbons like Methanol are discussed as options to store volatile electricity production from renewable energy sources (Specht et al., 2009). Hence, the CO₂-based production of chemicals could be used to produce sustainable raw material for the chemical industry while delivering storage services to the energy system at the same time. The technical and economical parameters of such a system integration should be part of further research.

To achieve a broad market penetration of CCU technologies their future cost competitiveness will be crucial. High prices for crude oil or a carbon price on non-energetically used fossil hydrocarbons could accelerate the development and the application of CCU technologies. A respective carbon price could be introduced by integrating the non-energetical use of fossil hydrocarbons into existing emission trading schemes. However, the latest price decline on the global oil markets (IEA, 2020) as well as the general decline of the global economy due to the Covid-19 pandemic (Nicola et al., 2020) could hinder this development. Therefore, economic incentives become even more important to motivate the necessary investments. For that purpose, economic instruments such as carbon pricing for all fossil-based uses and funding schemes for sustainable carbon processing technologies might have to be integrated into policy programs.

5. Conclusion

The results in this article show, that the actual carbon flows for material use in the German chemical and polymer industries are highly linear. Compared with other industries, for instance metal production, the circularity for carbon used as material is significantly lower. Most of the carbon losses occur as emissions after waste incineration. Additionally, dissipative use of carbon
containing products cause relevant losses. This shows the requirement as well as the potential for enhanced and additional recycling of carbon used as material in order to develop towards a circular economy. CCU technologies can enhance the circularity of carbon used as material by creating additional recycling cycles and also compensate dissipative losses. However, the high REE-requirement of CCU processes implies that CO2 should only be used if material recycling, in particular of polymers, can hardly be applied.

With respect to the necessary resources in Germany, the situation is different for CO2 and REE now and in the future. Even if only non-avoidable substance related CO2 point sources are regarded, the CO2-requirement is mostly fulfilled while DAC technology or imports could fill the remaining gap. In contrast, REE-availability is limited and REE will remain a scarce resource in the future. A competition between chemical and polymer industries with other production sectors and with transport for REE can be expected. Probably, the application of CUC technologies to produce base chemicals for the German market will mostly take place at international locations with better conditions for REE production.

Further research is advisable to enhance the results of the MFA, and to shed light on dissipative losses of carbon used as material and about carbon stocks within the society. Furthermore, the environmental and economic performance of CCU processes should be compared to material as well as chemical recycling to identify the most efficient way to recycle carbon used as material. At the macro level, the production of base chemical supplies should be included in future integrated energy and material supply models, considering REE-availability and requirements of all economic sectors. Finally, possible international production locations should be identified and compared with locations in Germany with respect to environmental and economic conditions.

CRediT authorship contribution statement

Simon Kaiser: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Visualization. Stefan Bringezu: Conceptualization, Writing - review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

The authors thank the German Federal Ministry of Education and Research (BMBF) for their support within the framework of CO2Plus (Funding Number: 033RC001C).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.122775.

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