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Allocation procedures in life cycle assessment of negative emission technologies

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Abstract

Negative emission technologies (NETs) play a key role in the mitigation of climate change by removing CO₂ from the atmosphere. In this study a comparative life cycle assessment (LCA) of three NETs – Pyrolysis Carbon Capture and Storage (PyCCS), Bioenergy Carbon Capture and Storage (BECCS), and Direct Air Carbon Capture and Storage (DACCS) – is conducted, using a common functional unit of 1 ton of CO₂ captured and stored. To account for the co-production of electricity and/or heat in PyCCS and BECCS, different allocation procedures – mass-based, energy-based, and economic – are applied and their influence on the net carbon dioxide removal (CDRnet) potential is quantified. The results indicate that the investigated PyCCS system exhibits the lowest climate change impacts in this comparison, followed by DACCS and BECCS. The application of different allocation methods reveals substantial variability in impacts, underscoring the critical importance of transparent and standardized allocation procedures for accurately assessing the environmental performance of NETs.

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1. Introduction

To combat climate change, the Paris Agreement aims to limit global warming to "well below 2 °C" compared to preindustrial levels[1]. The Intergovernmental Panel on Climate Change (IPCC) emphasizes the need for negative emission technologies (NETs) to permanently remove CO₂ from the atmosphere by 2100 [2]. Estimates suggest that a total of 800 Gt of CO₂ removal is required by 2100, making the rapid scaling of NETs essential [3, 4].

Various NETs exist, including pyrolysis carbon capture and storage (PyCCS; also known as Biochar Carbon Removal, BCR), bioenergy with carbon capture and storage (BECCS), and direct air capture and storage (DACCS). Each

technology has different technical readiness levels, environmental impacts, and CO₂ emissions that affect carbon capture [3, 5–8].

PyCCS involves pyrolyzing biomass to produce biochar, pyrogas, and bio-oil, with biochar enabling long-term carbon storage in soil [6, 9–11]. Previous studies have found carbon footprints of -274×10^{-3} kg CO_{2-eq} per 1 kg of woodchip input[12] and -378×10^{-3} kg CO_{2-eq} per 1 kg of biochar produced [13].

BECCS removes CO₂ through photosynthesis during biomass growth. The CO₂ capture can be achieved through a post-combustion process (e.g. amine scrubbing), a precombustion process (e.g. separation of CO₂ from a syngas prior to combustion of the hydrogen), and by oxy-fuel

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combustion process (e.g. combustion with pure oxygen instead of air to achieve high CO₂ content in the exhaust) [7]. For CO₂ sequestered in the purification of biomethane, carbon footprints of -647 kg CO_{2-eq} MWh⁻¹ to -1131 kg CO_{2-eq} MWh⁻¹ have been calculated [14–16].

DAC directly removes CO₂ from the air using sorbents or solvents [7, 17] that need to be regenerated through significant energy inputs [6, 17, 18]. Carbon footprints range between 0.06 and 0.30 kg CO_{2-eq} kg⁻¹ of CO₂ captured and stored [19, 20]

A comparative LCA of various NETs including BECCS and DACCS was conducted for the North American context, using 1 ton of CO₂ sequestered as the FU, found that the global warming potential (GWP) for BECCS ranged from 51 to 201 kg CO_{2-eq} t⁻¹ CO₂ removed, while DACCS exhibited a GWP ranging from 205 to 964 kg CO_{2-eq} t⁻¹ CO₂ removed [21, 22]. However, they did not include PyCCS, which is currently the most commonly applied NET in Europe [23].

In this study a comparative LCA is conducted for the following NETs: PyCCS, BECCS, and DACCS within the context of Germany, using a common FU of 1 ton of CO₂ captured and stored. The technologies are already in use today and relevant life cycle inventory (LCI) data is available and accessible [20, 24, 25]. Further, through the application of multiple allocation procedures, this study aims to provide insights into the influence that different allocation procedures have on the individual results.

2. Methods

To assess and compare the environmental impacts of the three different NETs, a comparative life cycle assessment (LCA) study is conducted, following the ISO standards 14040 and 14044 [26, 27]. The environmental modeling is conducted in SimaPro version 9.4, coupled with the ecoinvent database version 3.8. [25], and the EF3.0 methodology [28]. Further, the influence of different common allocation principles on the respective result is quantified.

2.1. Goal and scope definition

The study is conducted on PyCCS, BECCS and DACCS using the same FU and comparable system boundaries. In the context of this study - and NETs in general - three major concepts need to be distinguished. First, the amount of CO2 that is captured (CO_{2 captured}) is the amount CO₂ that is removed from the atmosphere through (1) photosynthesis and bound within the biomass or (2) the direct air capture unit. In this study, one ton of CO2 captured is used as the functional unit. To capture and store CO₂, the different technologies cause different amounts of greenhouse gas (GHG) emissions, due to the process itself, but also due to supply chain and infrastructure impacts. These emissions are accounted for in this study as CO2 emitted or climate change impacts (per functional unit). From these two values, the net CDR (CDR_{net}), which is the difference between the amount of CO₂ removed from atmosphere and the amount of CO₂

emitted during NET's operation is calculated according to equation 1 [29]:

$$CDR_{net} = CO_{2 \ captured} - CO_{2 \ emitted}$$
 (1)

The net CDR quantifies the actual benefit (or burden) of the respective technology regarding climate change impacts.

The study is geographically centered in Germany. Consequently, data selection from the ecoinvent database prioritizes the regional production mix if available. The cut-off system model is applied in this study, exclusively considering impacts originating from the analyzed life cycle, while disregarding previous or subsequent cycles.

2.2. System boundaries and Life Cycle Inventory (LCI)

To conduct a meaningful comparison, the system boundaries for all three investigated technologies must be consistent. The following sub-chapters give a detailed insight into the investigated system boundaries of the respective NET.

PyCCS: The LCA is conducted on a pyrolysis unit, which is designed to process nearly 2500 tons of dry biomass annually [24]. The biomass input is assumed to be forest wood from forests with demonstrably greater regrowth than removal and is therefore considered carbon-neutral input [30]. The PyCCS system includes the biomass production, the operation of the pyrolysis plant, and the biochar use (see Figure 1). "Biomass production" includes the harvesting and transportation of biomass[12, 30, 31]. The growth of the biomass is not within the system boundaries. "Operation of the pyrolysis plant" includes the chipping and drying of the delivered wood to improve their lower heating value and the efficiency for the thermal conversion [12], before it is pyrolyzed in the reactor. The products from the pyrolysis (biochar and the pyrogas) are separated through a filter. Water is added to cool the biochar and prevent the risk of

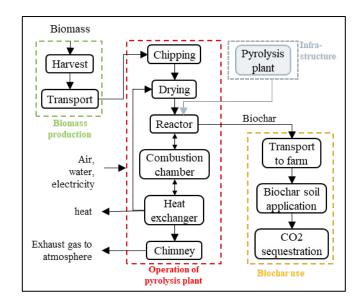


Figure 1: The system boundaries of the investigated PyCCS system.

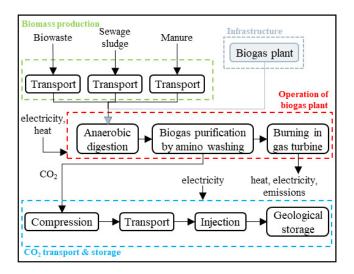


Figure 2: The system boundaries of the investigated BECCS system.

fire, resulting in a biochar water content of 20%. The pyrogas is burned in the combustion chamber, which provides heat for the pyrolysis, and the drying of the woodchips via a heat exchanger. Further, heat is re-distributed back from the heat exchanger to the combustion chamber, and any remaining excess heat is fed into a district heating grid. The combustion products of the pyrogas are emitted, including waste heat. "Biochar use" includes its transportation, application to soil, and the CO₂ sequestration [12]. The carbon fraction of biochar dry matter is 81.5% [24]. The carbon stable factor (CSF) is the fraction of carbon that is permanently stored in the soil as biochar and is assumed to be 75% of the biochar carbon content [30, 32] in this study. This means that only 75% of the carbon content in biochar is counted as captured carbon, while the remaining 25% are expected to gradually degrade over a period of 50 to 100 years [32, 33].

LCI data for the pyrolysis plant construction are sourced from [12, 13], while background data is taken from the ecoinvent database [25].

BECCS: The LCA on BECCS is conducted including the biomass production, the biogas plant operation, and the CO₂ sequestration (Figure 2). The biomass for this process is a mixture of manure and biowaste which are transported to a biogas plant with an annual production capacity of 350'000 m³ [25]. Through anaerobic digestion, biogas is produced. The biogas upgrading is done through amine washing, producing biomethane and CO₂. The CO₂ is captured and compressed to be transported via former natural gas pipelines and injected into the geological storage [20]. The biomethane is burned in a gas turbine, producing heat and electricity which are fed into a district heating network and the electricity grid, respectively, as well as exhaust gases (including CO₂ from biomethane oxidation) that are emitted through the chimney [25]. The LCI for the BECCS system is mostly adapted from the ecoinvent database [25] in consultation with technology experts. The CO₂ transport and sequestration are adapted from the DACCS model.

DACCS: The DACCS system is assumed to be a commercial temperature swing adsorption DAC unit [20]. This unit relies on the low-temperature solid amine-based sorbent category and temperature swing adsorption for

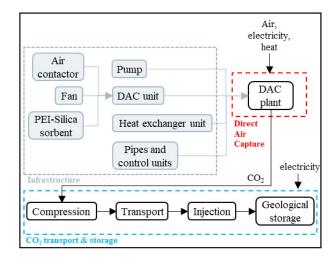


Figure 3: The system boundaries of the investigated DACCS system. source: own graphic adapted from [20].

sorbent regeneration. It has a CO₂ capture capacity of 50 tons per year, sustained over a 20-year period. The core process within the investigated system boundary of the DACCS system include direct air capture (DAC), and the CO₂ compression, transport, and storage, as illustrated in Figure 3. The DAC process entails the CO₂ binding to a sorbent, the subsequent desorption into a concentrated stream, and the regeneration of the sorbent for repeated cycles. The captured CO₂ is compressed, transported, and stored within a geological sink [20]. The foreground data for the DACCS system are sourced from [20].

2.3. Allocation procedures

To account for the additional benefits (useful heat and electricity) from PyCCS and BECCS, some form of allocation is necessary. Ideally, allocation should be avoided by dividing into sub-processes or expanding the product system [26]. However, division is not feasible here since outputs cannot be clearly attributed to inputs, and system expansion is not possible due to varying co-products among NETs.

If allocation cannot be avoided, ISO 14044 recommends using physical relationships, either mass-based or energybased. All allocation procedures simply redistribute impacts among the produced co-products of a process. There is no accounting for impact credits due to the avoided production of any of those co-products from other sources. For the production of biochar and heat (in the form of pyrogas) in the pyrolysis, an allocation based on the energy content is possible. This leads to a separation of flows at this point, with the pyrogas leaving the system boundaries, as it does not directly contribute to the sequestration of CO₂ in this system setup. Consequently, the pyrogas combustion with the heat production and resulting emissions are then outside the system boundaries. Only the further processing of the biochar is considered. In BECCS a mass-based allocation between the CO₂ and its co-product methane following the amin washing is feasible. However, the heat and electricity generation resulting from the combustion of the methane are omitted. When physical allocation is unsuitable, economic allocation can be applied, allowing allocations between coproducts not measured in the same unit. This method allocates more impacts to the product generating the most revenue. However, it connects the environmental footprint of CO₂ capture to market prices for heat and electricity. To investigate this dependency, heat and electricity prices are varied in the calculation of allocation shares.

3. Results and discussion

Three NETs are analyzed through a comparative LCA, with a special focus on the influence of the allocation procedures. As of now, there is no clear recommendation on how to allocate impacts related to NETs, which is why there is a lack of compatibility among those results. First, the results are presented without allocation (chapter 3.1). This means, that all impacts are allocated to the CO2 capture and storage. Benefits due to the co-productions of heat and electricity are not considered at this point. The co-products are assumed to leave the system boundaries burden-free. Second, to account for co-benefits, allocation procedures are applied, assessed, and evaluated following recommendation by the DIN EN ISO 14040, namely physical (chapter 3.2) as well as economic allocation (chapter 3.3). It should be noted that the change of allocation procedure only affects PyCCS and BECCS, as they generate heat and electricity to feed into a grid. The investigated DACCS technology does not produce useful co-products, therefore no allocation is required.

3.1. Net CDR without allocation

Within the defined system specifications for PyCCS, a biomass (dried forestry wood) input of 1.85 t with a total energy content of 9.35 MWh is required for the capture and storage of 1 t of CO₂ (CDR). Emissions of 220 kg CO_{2-eq} t⁻¹ CO₂ captured occur within the system boundaries of the PyCCS system (Figure 4), resulting in a net CDR for PyCCS of -780 kg CO_{2-eq} t⁻¹ CO₂ captured (see equation 1). This negative value shows that this technology is in fact a net negative emission technology. These GHG emissions are mainly attributed to the electricity consumption (44%, due to the fossil fuel dependency of the German electricity mix), transportation and pre-treatment processes (44%) and infrastructure components such as the storage bunker and pyrolysis system (about 2% each – due to the energyintensive production of steel). Direct process emissions emitted through the chimney contribute an additional 1.2%. It should be noted that CO, CO2 and CH4 emissions that originate from biomass are considered biogenic emissions and are therefore not or only partly counted towards climate change impacts (including the unstable fraction of carbon in the biochar).

In the BECCS model, the biomass input is calculated at 13.7 t biomass t⁻¹ CO₂ captured and stored with an energy content of 19.8 MWh. The considered biomass consists of a mixture of manure, biowaste, and sewage sludge. The carbon

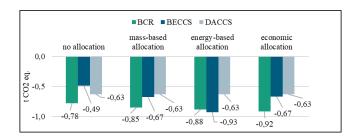


Figure 4: Comparison of net CDR of the investigated NETs without allocation and with allocation based on physical properties as well as economic value. FU = 1 t of CO_2 captured. Assessment method: EF3.0.

footprint is calculated at 513 kg $\rm CO_{2\text{-eq}}$ t⁻¹ $\rm CO_2$ captured, resulting in a net CDR of -487 kg $\rm CO_{2\text{-eq}}$ t⁻¹ $\rm CO_2$ captured. The main contributors are the heat and electricity consumption with 63 % and 21 %, respectively, while emissions from the anaerobic digestion process contribute about 5 % to the carbon footprint.

For DACCS, the GWP is 367 kg CO_{2-eq} t⁻¹ CO₂ captured, resulting in a net CDR of -633 kg CO_{2-eq} t⁻¹ CO₂ captured. Nearly 90 % of the carbon footprint originate from the energy consumption in the form of heat and electricity.

When comparing these results among the technologies, PyCCS shows the lowest carbon footprint in the process of carbon sequestration and storage, followed by DACCS and BECCS which both entail higher carbon emissions.

3.2. Physical allocation

In the BECCS process, both heat and electricity are produced which leave the system as they are fed into a grid. In the PyCCS, useful heat is generated. DACCS does not produce useful co-products. To account for the useful energy production, an allocation procedure must be implemented. As mentioned in chapter 2.3, whenever allocation cannot be avoided, physical properties should be used to allocate impacts among co-products of a process. Two common allocation procedures are mass-based and energy-based allocation. A comparison of the allocated impacts per NET and impact category are displayed in Figure 4 and elaborated in the following two sub-chapters.

Mass-based allocation: The application of mass-based allocation alters the system boundaries of both the PyCCS and the BECCS system. In the PyCCS process chain, the mass-based allocation is applied to allocate emissions between the produced biochar (77%) and the pyrogas (23%). The pyrogas then leaves the system boundaries, while the biochar is applied to soil and CO₂ is captured.

In BECCS, the allocation is made between the CO_2 (59%) and the CH_4 (41%) after the amine washing process. The CH_4 then leaves the system boundaries, while the CO_2 is collected, compressed, and stored underground.

Due to the fact, that following this approach, impacts are allocated to co-products and combustion processes are omitted, the impacts allocated to the CO₂ captured and stored are lower than in the reference case of no allocation.

Energy-based allocation: Another possibility of using physical relationships for allocation is the energy-based

allocation. In PyCCS, this is, again, applied in the pyrolysis process, producing biochar (40%) and pyrogas (60%). In the BECCS system, the allocation is applied among the CO₂ and CH₄ produced in the amine washing process. Since CO₂ has no energy content, all impacts are allocated to the CH₄ leaving the system boundaries. The remaining impacts that are displayed in Figure 4 result from the pressing and storing of the sequestered CO₂.

Both, the mass- and the energy-based allocation procedures alter the system boundaries of the PyCCS and BECCS system so that certain processes are omitted entirely in the results. This may lead to an underestimation of impacts, as the emissions attributed to the products leaving the system boundaries are not further quantified.

3.3. Economic allocation

Another possibility for allocation is the economic allocation. Here, the basic idea is that any emissions and impacts are allocated to the respective products of a process, depending on the revenue they generate. In this analysis, this allocation procedure can be applied to account for the heat and electricity co-production of the BECCS and PyCCS system. In PyCCS, the heat is generated from the pyrogas. The assumed economic value for the pyrogas is calculated by multiplying its energy content with a heat price of 0.1 €/kWh. Biochar is assumed to be valued at 81€/t. This assumption is based on the current German CO2 price and in coherence with the BECCS system. This results in allocation shares of 7% and 93% for the biochar and the pyrogas, respectively. In BECCS, the economic allocation is made between the captured CO₂, heat and electricity. The electricity price is taken from last year's average with 0.1€/kWh. The resulting allocation shares are 11% for the CO₂, 35% for the electricity and 54% for the heat.

While this approach allows for allocation of impacts among co-products that cannot be connected through physical relationships, it relies on market values that entail uncertainties and fluctuations. Emissions are allocated based on market prices. This means that the allocation in this case is not only dependent on the carbon pricing but also on heat and electricity prices. With an increasing heat price, more revenue can be generated by the heat supply, and the share of impacts allocated to the heat increases. This in turn reduces the impacts that are allocated to the capturing and storage of CO₂. The influence of a fluctuating heat price is much higher for PyCCS than for BECCS. This is because in the assessed PyCCS process, heat is the only co-product next to the biochar. For BECCS this dependency is not as significant, since also electricity is produced. Similar trends can be seen, when assessing the other impact categories, as well as the dependency on the electricity price in BECCS.

It is important to note that allocation does not reduce emissions but re-distributes them according to certain predefined procedures. The overall emissions that occur are not reduced, even though less are allocated to the main product. Without allocation all emissions in relation to a product or service are attributed to the functional unit, which is in this case the capture and storage of CO₂. Co-generated heat and electricity are assumed to be emission-free to avoid double counting. With the introduction of allocation, the emissions and impacts are simply redistributed, not reduced. To still ensure a complete picture of the process-impacts, it is relevant to also have a look at the emissions that are allocated to the co-products, namely heat and electricity, depending on their market price.

3.4. Limitations

Despite careful selection and application of data and methods, this study has limitations. First, the level of detail in the LCI data varies among the NETs. PyCCS is based on primary LCI data, while BECCS and DACCS rely on literature sources. Although system boundaries are harmonized, some impacts may still be underestimated due to lower levels of detail.

Second, due to a lack of reliable data on BECCS, a 90% $\rm CO_2$ capture rate from the amine washing process is assumed. Since $\rm CO_2$ concentration in the gas mix of the BECCS system is higher than in ambient air, the sequestration rate may be underestimated. Additionally, there is no CCS step to capture $\rm CO_2$ out of the exhaust gas that results from the biomethane combustion. First estimations suggest, that through the capture of $\rm CO_2$ from the exhaust gas of the biomethane combustion, the climate change impacts per ton of $\rm CO_2$ captured may be lowered by up to 170 kg, resulting in a net CDR of -657 kg $\rm CO_{2\text{-eq}}\,t^{-1}\,CO_2$ captured (without allocation).

Thirdly, as mentioned, emissions originating from the biomass directly are classified as "biogenic" and are therefore not counted towards the carbon footprint. While this is currently common practice in LCA, it is a controversial issue and requires some attention. A conducted sensitivity analysis on this indicates that if emissions from biomass are no longer regarded as "biogenic", they can significantly influence the overall carbon footprint. This highlights the importance of using sustainable biomass sources as well as waste materials as inputs.

Another sensitivity analyses indicates, that especially the use of renewable electricity influences the carbon footprint of the respective technologies, resulting in emission reductions between 20% and 40%.

Lastly, the different allocation procedures underly specific assumptions. Especially the results by economic allocation entail high uncertainties as energy prices can fluctuate quite significantly. Likewise, the economic values of sequestered and stored CO₂ and biochar underly uncertainties.

4. Conclusion

The findings emphasize PyCCS as the technology with the lowest climate change impacts when compared to the other NETs within the defined system boundaries when no allocation procedure is implemented. The introduction of allocation procedures lowers the climate change impacts of

captured CO₂, as impacts are shared among the products biochar and heat. Through physical allocation (energy-based) a reduction of nearly 50% of the impacts allocated to the captured CO₂ is reached. The allocated impacts when applying economic allocation are even lower (-60%). This highlights the significant influence and importance of the introduction of allocation procedures to ensure fair comparisons. At the same time, it underscores the importance of transparency regarding allocation choices, as this influences the results and hence the comparability of the different NETs.

Similar results are seen for BECCS in this analysis. A major difference being here, the co-production of not only heat but also electricity. In the economic allocation, the share of impacts that is allocated to the captured CO₂ is not only dependent on the heat price, but also on the electricity price. Climate change impacts allocated to the captured CO₂ are reduced by more than 35%. As mentioned above, the impacts are likely to be overestimated, as no additional capture is assumed after the combustion of methane.

In contrast, DACCS is not impacted by the introduction of allocation procedures, as it does not produce additional useful outputs. This affects the overall comparison. Whenever an allocation procedure is introduced, the impacts associated to the captured CO_2 are highest for DACCS.

Moving forward, future research should prioritize the comparison of NETs in terms of their environmental impacts. This includes assessing different configurations and the economics of various NETs. The collection of precise data remains crucial to ensure comprehensive comparisons. The importance of carefully selecting an allocation procedure has been demonstrated. Still, further research is needed to develop reliable allocation strategy for NETs.

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Data Availability Statement

Additional information on Life Cycle Inventory data can be made available upon request.

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