Life cycle assessment-based guidance for development of new energy technologies

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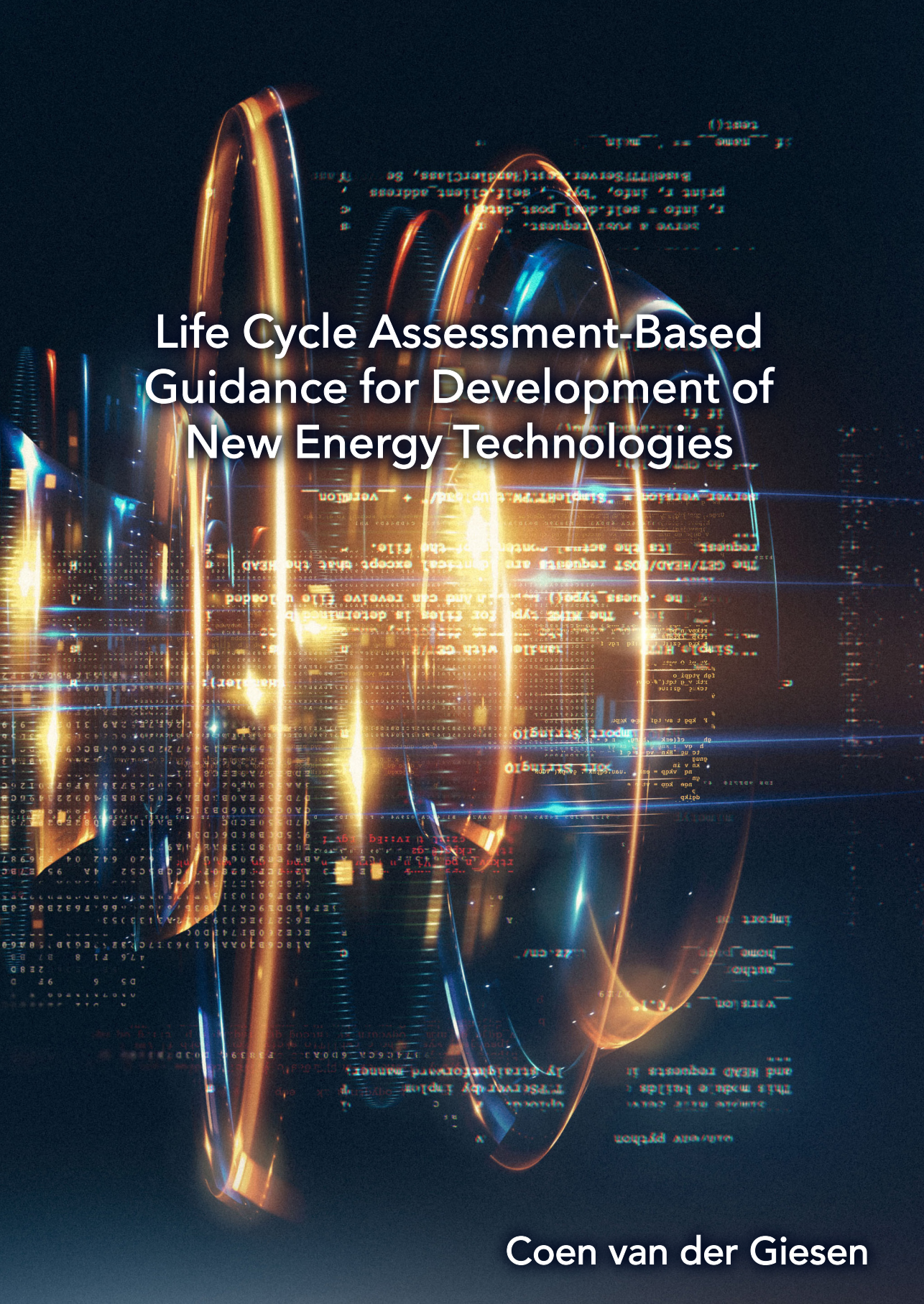

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Coen van der Giesen

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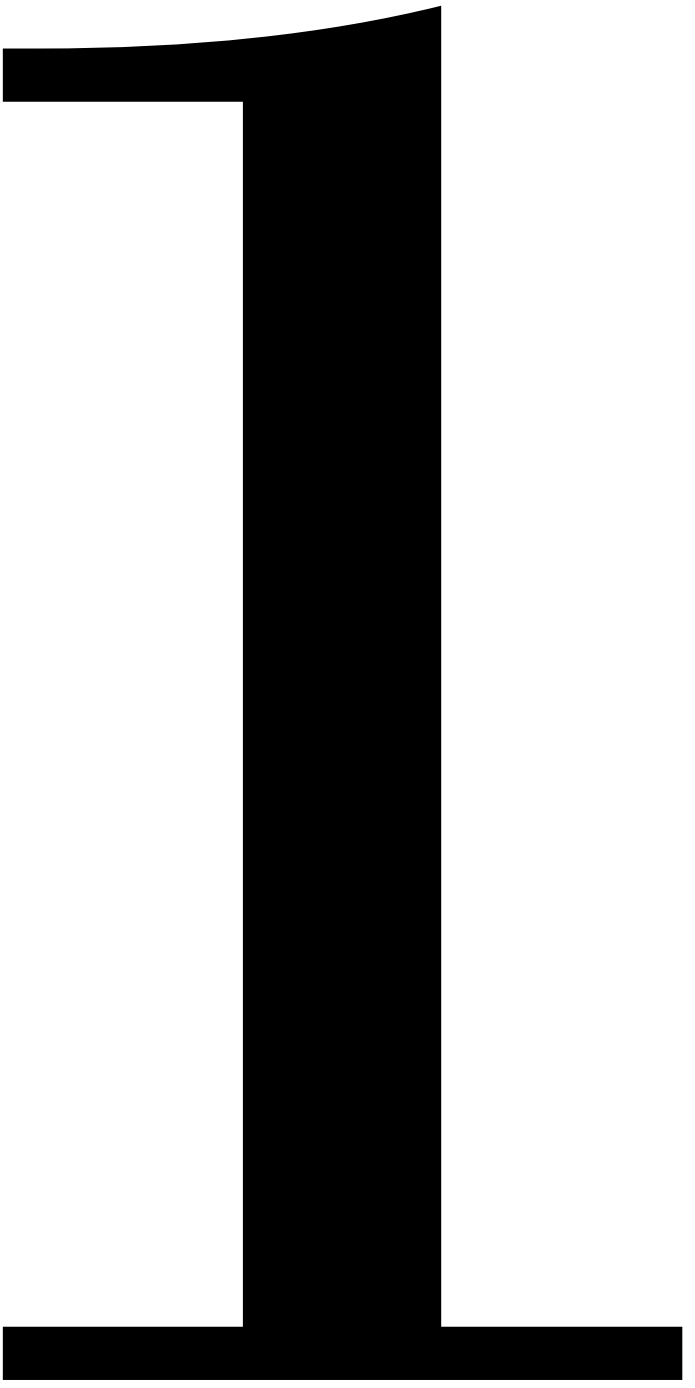


Chapter 1

Introduction
-
Toward Sustainability Through Technology

1. Introduction – Toward Sustainability Through Technology

Research, development, deployment and widespread diffusion of new environmentally sound technologies is a major route towards achieving sustainability (United Nations 2017; Giovannini et al. 2015). In the Emissions Gap Report 2018 by the UNEP it is made clear that current efforts to reduce GHG emissions to keep global warming below 1,5 degrees Celsius by 2030 are not sufficient. By 2030 GHG emissions should be 25% lower than in 2017 to stay below a 2 degree temperature rise and even 55% lower to keep this rise below 1,5 degrees Celsius. According to UNEP, the key component to reach climate targets is to be found in the acceleration of innovation and increased investments in new technologies (Olhoff and Christensen 2018).

The relation between technology and sustainability has some paradoxical features. Technology can be seen as both the cause and the remedy for detrimental environmental change, but it also allows for monitoring this environmental change and can be seen as liberating humanity from environmental constraints (Grubler 2003).

In the scientific field of industrial ecology, technology is regarded as the most important factor for reducing environmental impacts of anthropogenic action. Industrial ecology's "master equation" (Graedel and Allenby 2010), which is derived from the IPAT equation (Ehrlich and Holdren 1971, 1972), is conceptually used to show the connection between environmental Impact (I) and the product of Population (P), Affluence (A) and technology (T).

$$\underset{\text{[impact]}}{(I)mpact} = \underset{\text{[capita]}}{(P)roduction} \times \underset{\text{[consumption / capita]}}{(A)fluence} \times \underset{\text{[impact / unit of consumption]}}{(T)echnology}$$

The world population is expected to increase to around 8 billion people in 2040 (Randers 2012) and peak at 10,9 billion in 2100 (UN DESA 2019). This increase in population is hard to stop or redirect, although an improvement in education and healthcare in countries with lower income levels will limit the amount of children being born and will slow down population increase in the coming decades (Rosling et al. 2019).

Also an increase in affluence can be expected. Affluence is often expressed in GDP and GDP growth is the focus of most national policies. Virtually all authoritative economic forecasts used for economic policy assume GDP growth in the next decades. In addition, one of the Sustainable Development Goals of the United Nations is reducing poverty and inequality in the world which will make that people in poor and lower income countries will increasingly become more wealthy. This increase in affluence will show a trade off in increased environmental impacts (Scherer et al. 2018).

When Population and Affluence will increase and have positively connection to Impact, it is up to influencing the factor technology to reduce the impacts per unit of consumption to decrease the environmental impacts (Scherer et al. 2019; Chertow 2001, 13). We need to keep in mind here that, in the equation, Technology has a negative connection Impact. Meaning, that an increase in (improvement of) Technology decreases impacts per unit consumed.

This reasoning can also be found in the "Global Materials Resources Outlook to 2060" by the OECD. It reports that an increase in population and affluence will drive an increase in demand for goods and services that also involve higher material intensities(OECD 2018). It further states that "more than half of all GHG emissions are related to material management activities and the fossil fuel use in these activities also leads to larger energy related emissions from material and resource use" (OECD 2018). The International Resource panel of the UN adds that materials do not only have a large influence on GHG emissions, but also contribute to over 90 per cent of biodiversity loss and water stress" (Oberle et al. 2019). Both organizations point to technological improvements to slow the growth in future material demand and reduce global materials intensity to reduce environmental impacts (Oberle et al. 2019, 9; OECD 2018).

1.1 The challenge of technology development

1.1.1. S-curves and the technology life cycle

When hopes are put on developing and improving technology to reduce global environmental impacts, we need to know how technology development works and how we can develop technologies that actually address the environmental challenges we are facing. The development of technology is often described by the use of s-curves in which technology performance is plotted against time (Figure 1.1). It shows that technology development is not linear and unlimited, but rather goes through several stages in which the rate of development changes over time.

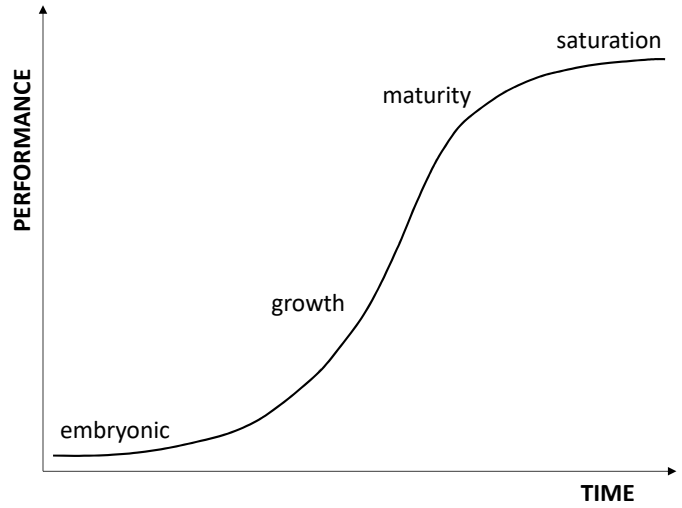


Figure 1.1: Technology development s-curve, based on Grubler (2003).

Using s-curves in combination with biological analogies can help to explain how technology develops through different phases in its life cycle (Grubler 2003). At the start we see a slow embryonic phase of exploration, research and development. In this phase production volumes and market shares are low because the focus is on demonstrating viability. Because of learning effects for both producers and consumers the applicability of a technology becomes clearer, its viability becomes more certain, and innovation becomes less risky. This is when a technology enters a period of growth in which performance improve faster. A technology enters a phase of maturity as costs and prices start decreasing. In this phase competition is driven by cost reduction rather than improvement of performance (Grubler 2003; Hirooka 2006, 128). Finally the pace of performance slows even further down as the market becomes saturated, and costs for further improvement the technologies' performance become too high.

1.1.2. Understanding the technology life cycle via innovation trajectories

Where s-curves help in providing a general picture of the life cycle of technologies, they do not provide details on what exactly happens in the different life cycle phases and how this might be influenced. More detail can be found in the work of Hirooka (2006) who uses empirical data to explain what happens in different development phases of technology development. He introduces the concept of *innovation paradigm* in which three consecutive trajectories for innovation and technology development can be discerned: the technology trajectory, the development trajectory and the diffusion trajectory. These consecutive (slightly overlapping) trajectories also follow the shape of an s-curve (see figure 2).

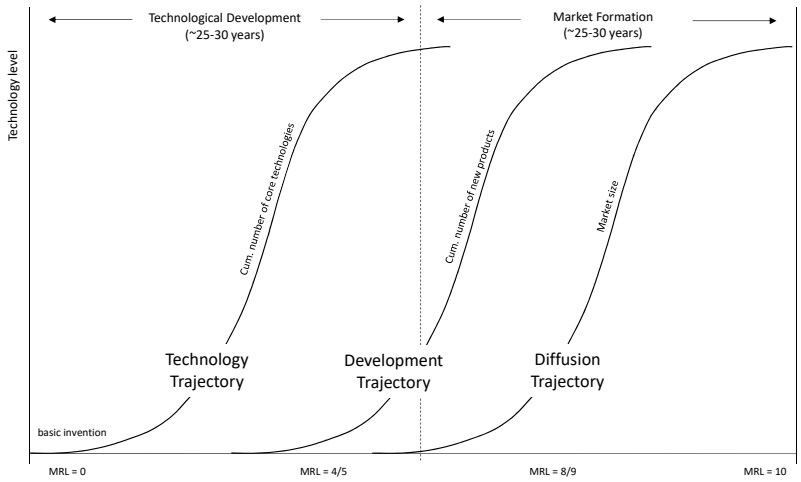


Figure 1.2: Innovation Paradigm: three consecutive trajectories in technology development, based on Hirooka (2006).

Hirooka (2006) explains that technology develops from idea or basic invention through development of core technologies to products that gain market share. He discerns two consecutive phases (left and right side in Figure 1.2) called *technological development* and *market formation*. The phase of market formation is the main focus in the scientific field of technology dynamics and has been extensively described by e.g. Geels (2002) and Hekkert (2007). Insights in the phase of technology development are however less commonly available.

The more tangible output of technology development can be found in the form of patents and is applied in new products. Hirooka defines technology development as "the phenomenon of knowledge transfer from one person to another in the field of human society" (Hirooka 2006, 130). Within technology development one can discern two trajectories. First, the technology trajectory, in which academic and industrial research institutes convert ideas into core technologies and patents. Second, the development trajectory in which these core technologies and patents are deployed into useful and marketable products.

The main insights from Hirooka's work for this thesis are the timeframes that he assigns to technology development. The time required for technology development is around 25-30 years, after which another period of 25-30 years is needed for full market formation. Using these time frames in combination with the description of different MRL phases (see box 1) we can construct a general technology development timeline that includes the different innovation trajectories and MRL scores (figure 1.2).

Box 1 - Connecting innovation trajectories to Technology (TRL) and Manufacturing Readiness Levels (MRL)

Technology Readiness Level (TRL) are used to indicate the level of development of a technology. From its original use, TRLs also found their way to other organisations like the U.S. Department of Energy (U.S. Department of Energy 2011). TRL was originally developed and introduced by the National Aeronautics and Space Administration (NASA) to assess the maturity of a technology and its fitness to be used in space.

The concept of TRL has been expanded into Manufacturing Readiness Levels (MRL). MRLs do not assess "just the maturity of a technology but also that of components and subsystems from a manufacturing perspective" (Gavankar et al. 2014). For the assessment of newt technologies it is important view these in their (intended) systems of use. This thesis will therefor use MRL as indicator to indicate the level of development of the technologies discussed.

Innovation Trajectory	**Technology Development stage**		**Technology Readiness Level (TRL)**	**Manufacturing Readiness Level (MRL)**
Technology trajectory	Conceptual development	1	Basic principles observed	Implications identified
		2	Formulation of concept	Basics identified
		3	Proof of concept	Proof of concept
		4	Validation in Laboratory	Laboratory sample
Development trajectory	Technology development	5	Components in representative (simulated) environment	Prototype components in simulated environment
		6	Prototype in representative (simulated) environment	Prototype system in simulated environment
	Engineering development	7	Prototype in operational environment	Prototype in production environment
	Small scale production	8	System qualification	Ready for small scale production
Diffusion trajectory		9	Technology ready	Transition to full scale production
	Mass production	10	n/a	Lean mass production

1.1.3 Technology development in practice

To come up with a practical approach for the environmental assessment of new technology we need more insight and further understanding of what happens in the technology development phase in a more practical sense. In other words, we need to know and understand what technology developers do and how they approach research and development so that the outcomes and insights of an assessment can be applied in the development of technology that will indeed provide an improvement for reducing environmental impacts.

Brian Arthur (2009) explains that technologies can be defined at different levels (see Figure 1.3). At the basis of all technology lies *science*, which allows us to fundamentally understand natural phenomena, e.g. the photovoltaic effect. If a natural phenomenon is 'captured' and can be 'steered in some sort of way', this is done by an *individual technology*, e.g. a PV cell. According to Arthur technology ultimately provides some sort societal functionality by "capturing natural phenomena that are orchestrated under human agency" (Arthur 2009). We will need to involve more technologies in tandem put a natural phenomenon is to good use. This groups of technologies we need for this are called *bodies of technologies*, e.g. PV technology or solar technology to provide us with renewable and clean energy. PV technology consists of PV cell but also of aluminum mountings and electronic convertors. When we talk about our reliance on technology for a sustainable future we are actually talking about *technology as a whole* without being specific.

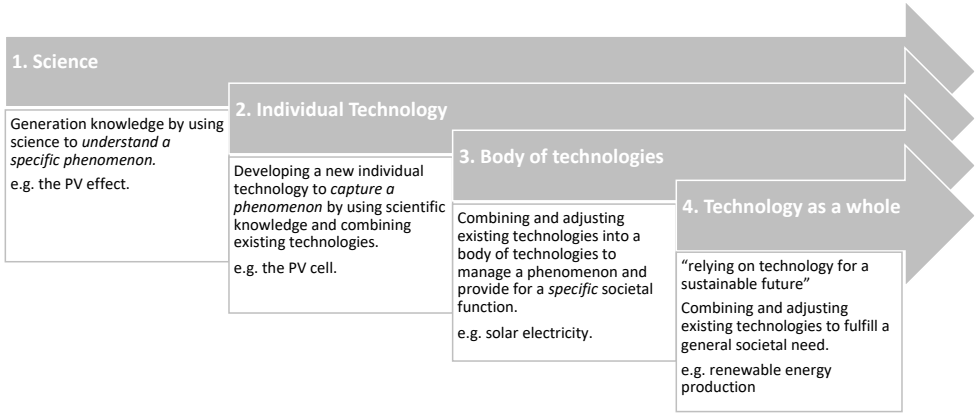


Figure 1.3: Different definition levels for technology, after Arthur (2009).

It is unavoidable not to mix up these different levels when discussing the development of technology. The different definition levels however also illustrate the process of technological evolution from idea to application and product. In practice existing technology is often used to develop new technologies that are better in managing natural phenomena or capturing new ones or put them to better use. Technology developers use the available stock of technologies and combine technologies from different levels in order to construct a new technology. This is a recursive process called *combinatorial evolution.* In this process suitable parts and functionalities of existing technologies from different levels are combined. "The initial version of a technology has a functional physical form and can only be pushed so far before some parts of its system run into a barrier that restrains it" (Arthur 2009). To overcomes these barriers technology developers apply other existing technologies that are taken from the total stock of technologies. This process of improving technology in small steps by introducing solutions to overcome internal design is called *structural deepening*. In essence technology is created out of itself and the larger the amount of available building blocks in the total stock of technology the bigger the chance for success and the faster development goes.

The processes of combinatorial evolution and structural deepening are daily practice for R&D engineers. When developing better technologies that have lower impacts on the environment it is important to understand that it is not only about getting the technology to work, but getting the technology to work in an environmentally sound manner. Different decisions to overcome design barriers will have different impacts on the technical performance and on the environmental consequences of these technologies. The possibility to assess pending decisions concerning combinatorial evolution and structural deepening during the R&D process is important so that a balanced choice can be made in the design of new technologies.

1.2 Technology development and time constraints

While the urgency in new technology development to face climate challenges is eminent it is just as important to realize that development and implementation of new technologies takes a considerable amount of time, especially when this technology has to replace old (unsustainable) technologies and has to fulfill a significant share of a functional and societal demand. "For global environmental transformations such as climate change, the long time-scale of technological change complicates decision making. The time-scale for pervasive technology approximates the time-scale of global environmental change. If climate change proves extremely serious, and we wait to act, it will be too late to implement the massive economic and technological changes required to reduce impacts." (Grubler 2003, 354)

Research on technology development time-frames mainly has mainly been done for the market formation phase. Kramer and Haigh (2009) have shown that it takes about 30 years for energy technologies to develop from introduction to the market to occupying about 1% of the world's energy mix. This 1% might be considerable when taking an economic look at global market shares but might only be a drop in the ocean for averting climate change for which established technologies with much higher market shares need to be improved or replace.

Hirooka (2006) has shown that developing a new technology from scratch until it finds its way in a product which provides for considerate market share can take 50 to 60 years. Fortunately not all technologies have to be developed from scratch although and when climate goals are set for 2030 we see that completely new ideas and technologies will not be able to contribute to those goals.

R&D of technology is subject to a phenomenon known as the Collingridge dilemma, which says; "when change is easy, the need for it cannot be foreseen; when the need for change is apparent, change has become expensive, difficult and time consuming" (Collingridge 1980). So in early phases of development is easy to make changes in design but we do not know the implications of a new technology until it has put to use. This is indeed a dilemma, when do we make the necessary changes? The inherent nature of a dilemma is that a way out is not possible using logic reasoning but is simply driven on personal choices. Assistance in making such choices might be found in early assessment of a technology's potential impacts by investigating different design choices, while keeping in mind that making the perfect decision is impossible.

Developing greener, cleaner and more efficient technologies (using fewer resources or using them more efficiently) has been and will be the focus of many research projects. It forms the backbone of many EU sustainability policies and research projects. To support the R&D of such environmentally sustainable technologies, international research frameworks, such as the European Horizon 2020 program (European Commission 2017), demand the application of life cycle assessment (LCA). Some examples of these research projects are as follows: Carbon4PUR (carbon4pur.eu) is a project in which it is investigated if CO_2 can be used as a resource for building insulation and coatings; The Sitasol project (sitasol.com) investigates new technologies to produce tandem solar cells; The Nano Tandem project (nano-tandem.ftf.lth.se) investigates the use of nano-technology in the production of tandem solar cells and pursues a balanced view on the environmental advantages and disadvantages of using nano-materials. All these projects use LCA to guide technology development. The research presented in this thesis was initiated in the Dutch, NWO funded, Biosolarcells project. This project focused on

fundamental research on artificial photosynthesis and included an early stage assessment of environmental value propositions of solar fuels (Biosolarcells 2011).

1.3 Technology assessment and LCA

The scientific field of technology assessment (TA) develops methods to assess the potential implications of the introduction of a new technology in society. The aim of this field is TA is to "broaden and positively influence technology development processes by addressing potential innovation obstacles or impacts as early as possible rather than assessing ex-post the impacts of more-or-less finalized products."(Baumann 2017, 38). It tries "to reduce the human costs of trial and error learning in society's handling of new technologies, and to do so by anticipating potential impacts and feeding these insights back into decision making, and into actors' strategies." (Schot and Rip 1996). It also provides a context-relevant overview of potential practical consequences of the introduction of a new technology that serves as a base for communication about this technology and strategies for technology implementation" (Fleischer and Grunwald 2008).

TA usually applies qualitative narrative methods to present prospective sociotechnical scenarios informed via face to face interviews with stakeholders (Baumann 2017, 40). Overall technology assessment can be defined as a form of policy research that examines short- and long-term consequences (e.g. societal, economic, ethical and legal) of the application of technology (Banta 2009).

TA is mainly informative for policy makers by providing insights in potential concerns and opportunities of a new technology in a desired sustainable society (Rip 1998). It is a qualitative approach that focusses on informing society at large about potential socio-economic impacts and risks. It is therefore limited in informing technology developers or in technology design because it tends to be qualitative and lacks practical quantitative assessment of potential impacts of design choices. Life Cycle Assessment (LCA) in principle is a more suitable method for such comparative quantitative environmental assessments. However experience with performing forward looking assessment is still limited in the field of LCA.

The standard approach to LCA is described in box 2 below. The main strength of LCA is that it takes a systems approach which allows for identifying *problem shifts*; the shift of environmental impacts to different life cycle phases or to different environmental impacts as the result of a change in practice. E.g. the introduction of the electric car shifts CO_2 emissions from the car to the power plant. Or by reducing CO_2 emissions

shift the problem to increasing demand for rare earth metals, needed in batteries and electric engines.

Conventionally, LCA is mostly used to evaluate technologies that already exist, are mature and are used in established markets (Fleischer and Grunwald 2008). LCA therefor uses general data that are based on the past performance of technologies. LCA until now did do not incorporate information of the future ahead and has therefore been kept outside the field of prospective TA (Fleischer and Grunwald 2008). One objective for contemporary TA to better address the sustainability challenge of technology development is to include and "develop LCA methodologies which allow the integration of (uncertain and incomplete) prospective knowledge" of new technologies under development (Fleischer and Grunwald 2008).

Table 1.2 lists some differences and characteristics of current approaches of TA and traditional LCA the required characteristics for forward looking LCA. TA and LCA should however not be considered competitors for technology assessment but should be seen as complementary. LCA should be tailored to include a forward looking perspective so that it can provide valuable insights for technology development. Developing such a forward-looking LCA method for the assessment of potential environmental impacts of new technologies is the main ambition of this thesis.

Table 1.2: Comparison of TA and LCA for early technology assessment.

Characteristic	**Technology assessment**	**Traditional LCA**	**Needed for forward looking LCA**
Research method	Qualitative	Quantitative	Quantitative
Research focus	Socio-economic impacts	Environmental impacts	Environmental impacts
		Life cycle or systems perspective	Life cycle or systems perspective
Research scope	Prospective	Retrospective	Prospective
Level of abstraction	High level of abstraction long cause effect chain	Low level of abstraction short cause effect chain	Low level of abstraction short cause effect chain
…	-	Comparative	Comparative
Goal	Policy support	Policy support	Technology design support
Research basis	Scenarios and probabilities	Empiric data	Expectations
Uncertainty	High but manageable uncertainty	Manageable uncertainty	High uncertainty

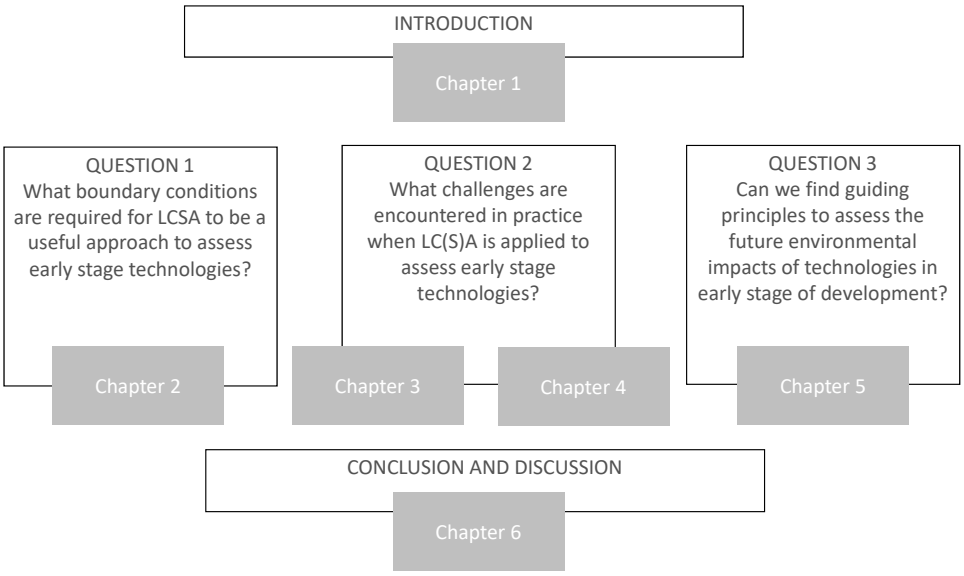


Figure 1.4: Thesis outline.

1.4 Aim, research questions and thesis outline

We identified a need to assess the potential future environmental impacts of technologies in the early stage of development. The aim of this thesis is to develop a forward-looking assessment method based on LCA that can be used to integrate early insights in potential environmental impacts in R&D, with a specific focus on new energy technologies.

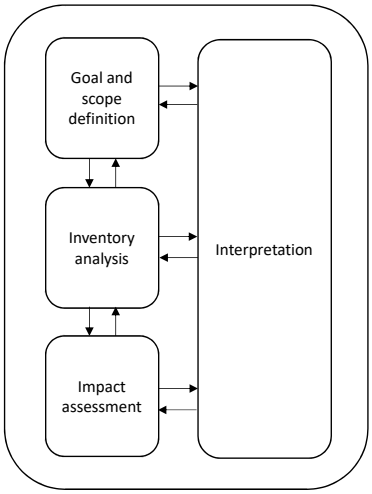
The research performed to propose a forward-looking LCA method to assess new technologies on their potential environmental impacts was guided by three research questions (see Figure 1.4).

At the start of this thesis trajectory, life cycle sustainability assessment (LCSA) was the most comprehensive form of LCA available. A logical point of departure for this thesis was therefore to start with investigating the applicability and usefulness of LCSA for the assessment of new energy technologies. Chapter 2 presents the research to answer question 1: "*What boundary conditions are required for LCSA to be a useful approach to assess early stage technologies?*" Since shortcomings of the LCSA approach were found, the decision was made perform two LCA case studies concerning two technologies in early stages of development: the production of fuels from CO_2 (chapter 3) and the use of two CO_2 capture technologies to reduce CO_2 emissions from coal powered electricity generation (Chapter 4). Experience and insights from these case studies help to answer

question 2: "*Which challenges are encountered in practice when LC(S)A is applied to assess early stage technologies?*" Putting the insights from these case studies side by side with a broader literature review of existing studies performing forward looking LCA forms the basis for answering question 3: "*Can we find generalizing principles to assess the future environmental impacts of technologies in early stage of development?*" Finally, chapter 6 presents the answers to the research questions and adds broader insights and recommendations for the future.

Box 2 - Principles of Life Cycle Assessment (LCA)

LCA is used to compile all physical inputs (for example, energy and resources) and outputs (e.g. CO_2 emissions) and calculates the connected environmental impacts of a product system throughout its life cycle. There is a standardized procedure to perform an LCA, which can be found in the ISO 14040 - 14044 series issued by the International Organization for Standardization (ISO).



LCA comprises four iterative phases as presented in the figure on the right.

Goal and scope (GSD)

In the first phase of LCA the practitioner determines the goal and scope; why is this study done, how will the results be used (goal)? And what is the study about? (scope)

Life cycle inventory phase (LCI)

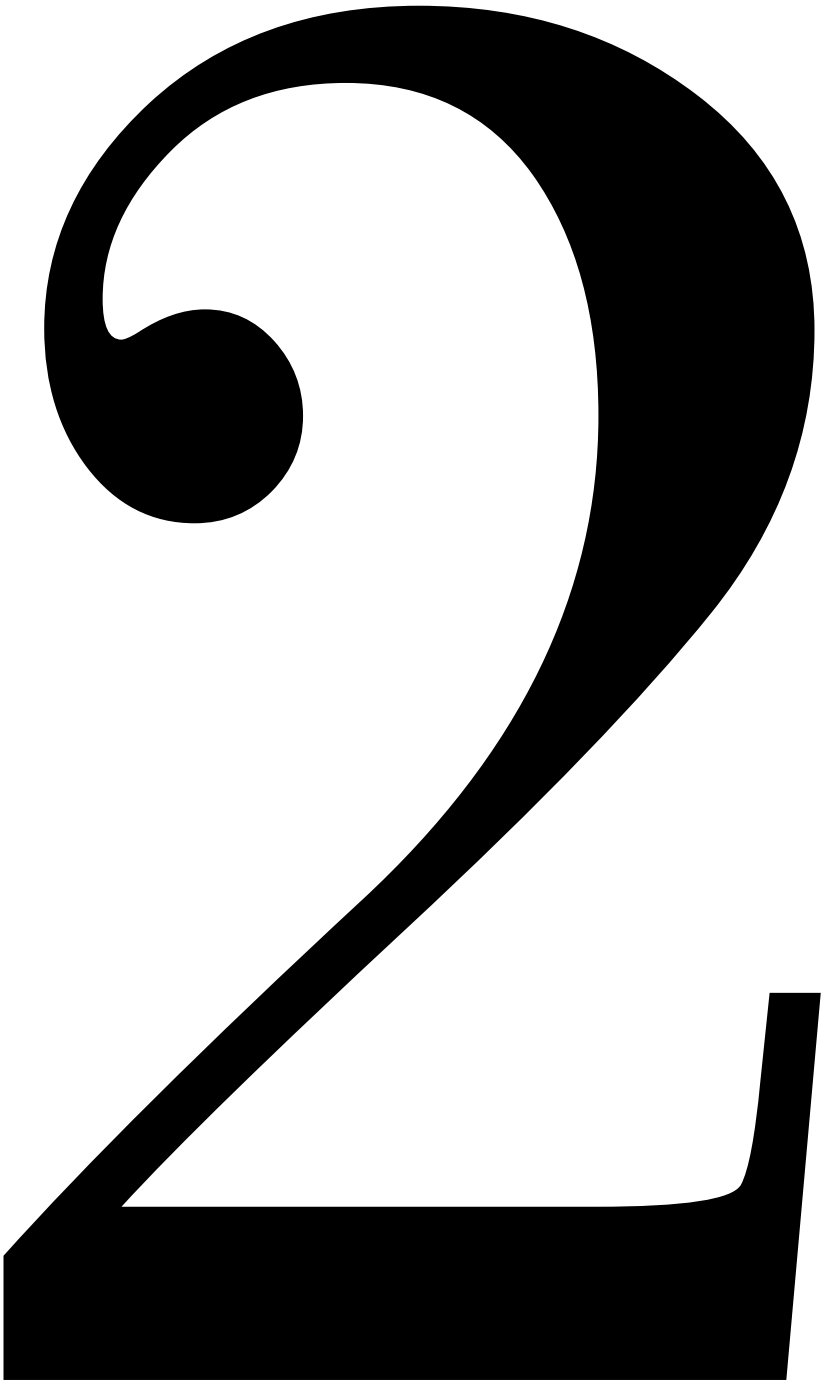
In the second phase of an LCA the practitioner makes a flowchart of the system in which all relevant processes are displayed. Then all extractions from and emissions to the environment are collected and quantified. The result of the LCI phase is an inventory table in which all exchanges between the system under assessment and the natural environment are shown.

Life cycle impact assessment (LCIA)

In the third phase of an LCA different predefined methods are used to group and aggregate the inventory data into environmental impact categories, e.g. greenhouse gas emissions like CO_2 and CH4 contribute to climate change. Other impact categories can be toxicity or resource use.

Interpretation

In the last phase of an LCA the outcomes from the previous phases are interpreted. Here it is possible to identify which processes contribute most to specific impacts, which is called a hotspot analysis. The practitioner also checks the potential influence of methodological choices, data-quality and uncertainty on the outcomes. These should also be reported to make sure that the work performed is in line with the goal and scope of the study.



Chapter 2

Towards Application of Life Cycle Sustainability Analysis

Coen van der Giesen, René Kleijn, Gert Jan Kramer, Jeroen Guinée

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Abstract

There is an increasing need for expanding the scope of traditional life cycle studies to answer system-wide sustainability questions. This has resulted in a framework for Life Cycle Sustainability Analysis (LCSA). Since the framework was first published in 2009, as one of the outcomes of the CALCAS project, several views and considerations concerning the methodological approach have been published. However, until now practical experience with LCSA is very limited.

This paper reports first efforts and experiences in bringing the LCSA framework into practice by assessing the sustainability of solar fuels. Starting point of the project is the hypotheses that new technologies can only be practically implemented if they fit into a socially, economically and ecologically sustainable context. The analysis therefore aims at identifying performance criteria which a new technology needs to fulfil in order to compete in the existing market.

It is argued that a LCSA study should be initiated with a broad but relevant system description as the first of in total five steps Of this five step approach, the first – system description – is discussed here. The system description identifies and describes the technological description, the intended application of a technology, which share of specific demand for service will be met, which other technologies contribute in meeting this demand, the (relevant indicators for addressing)sustainable impacts of meeting the demand and developments over time. By doing so it provides a solid basis for further steps in the LCSA study.

2.1 Introduction

2.1.1. Towards BioSolar Cells

The Dutch Towards Bio Solar Cells (TBSC) program aims at developing new technologies that make better use of solar energy by artificially increasing the efficiency of the photosynthesis process. These technologies range from developing biodiesel producing algae, modifying the photosynthesis process in plants to improve yields and the development of "artificial leafs" with which fuels can be produced directly from sunlight. The implementation of a new technology can only be successful if -once technological feasible- it is economically viable, environmentally beneficial and socially acceptable. The TBSC program therefore not only aims to develop new technologies, but will simultaneously investigate the initial response of society to find out elements that are critical for acceptance of biosolar technologies. For this, societal debates will be organized and ex ante comparative sustainability assessment of the new technologies are performed to identify sustainable opportunities, constraints and necessary improvements for further development and successful implementation.

2.1.2. A new approach to sustainability assessment

For the ex-ante assessment of biosolar cell technologies to produce solar fuels a novel approach, labelled "*Life Cycle Sustainability Analysis*" (LCSA) will be used. The LCSA framework, used in this paper, was first presented as an outcome of the CAL-CAS project . In the past decades the focus of sustainability analyses has been on the environmental impacts of products or services providing a specific function. An in-creasing need to take a broader view on the possible impact on sustainability of decisions and developments has been recognized (Kloepffer 2008; Finkbeiner et al. 2010; Guinée et al. 2011; Valdivia and Sonnemann G. 2011). A first step towards a broader view introduces the analysis of economic and social impacts of product systems in addition to its environmental impacts. A second step expands from product level assessment towards assessing the consequences of introducing changes in product systems on meso (company, regional) and economy-wide (nation, global) levels. Consequences can be expected, for example, in resource availability for other products or in consumption patterns (Zamagni 2010). It is clear that assessment of new technologies in the TBSC project requires the broadest view on sustainability as is provided by the LCSA approach.

2.1.3. This paper

Since the range of technologies developed in the TBSC project is rather wide, the project of which this paper is a first exploration, focusses on the application of biosolar cell technologies to produce solar fuels. Since the biosolar technologies are still in their early infancy, the primary focus is currently on the production of solar fuels through existing technologies as is proposed by Haije (2011). The aim of this paper is to provide

a practical approach for performing an LCSA study focused on solar fuels. First a concise overview of the LCSA framework will be presented, which will be supplemented with insights and experiences from a limited set of recent publications considering LCSA. Insights gained from this overview are then translated in a practical approach for a LCSA concerning biosolar cell technologies to produce solar fuels.

2.2 Current status of life cycle sustainability analysis

2.2.1. Life cycle sustainability analysis (LCSA) framework

One of the main results of the CALCAS project (Zamagni et al. 2009; Guinée et al. 2009) is a high level methodological framework for LCSA (see Figure 2.1). The framework suggests using LCA practice as a steppingstone towards conducting life cycle sustainability assessments and consists of three phases; goal & scope definition, modelling and interpretation.

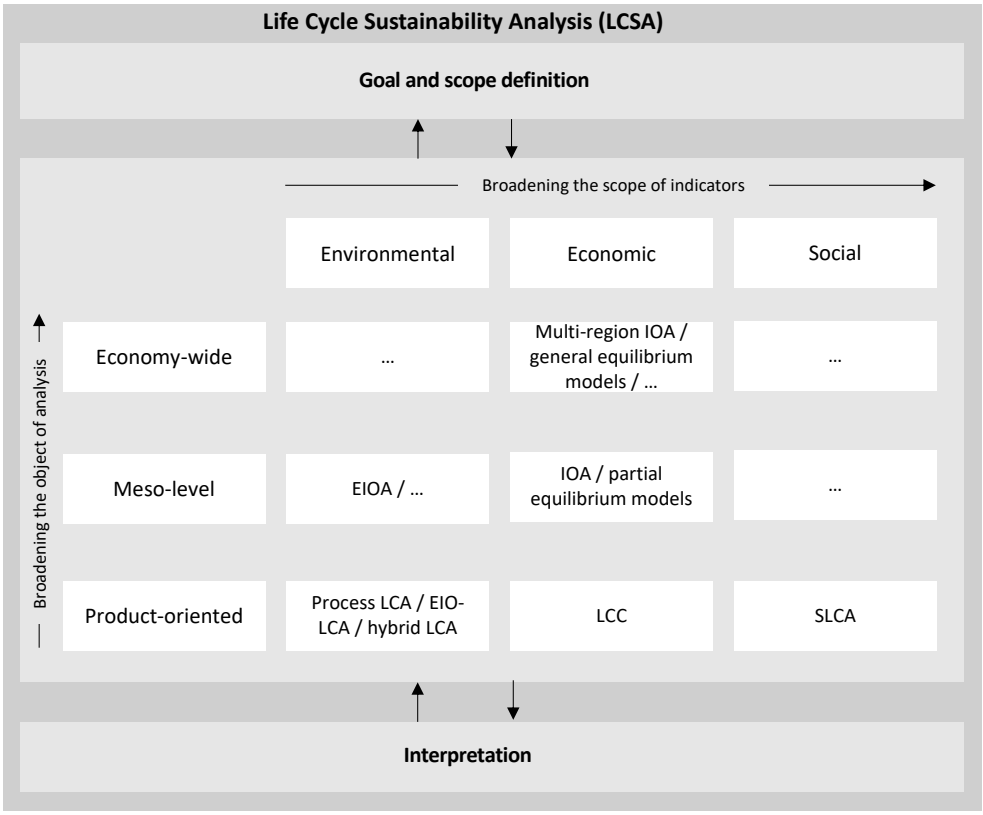

Figure 2.1: Transdisciplinary integration framework for LCSA taken from Guinée et al. (2011).

Goal and scope definition in LCSA

The LCSA framework is not a fixed guide, Guinee describes it as a transdisciplinary framework of different models rather than a model in itself (Guinée et al. 2011). It is intended to guide practitioners in selecting the most appropriate life-cycle based models for a given life-cycle based question (Guinée and Heijungs 2011). In classic LCA studies the goal of a study is formulated in terms of the exact question, target audience and intended application. The scope is defined in terms of temporal, geo-graphic and technological coverage and the level of sophistication of the study in relation to its goal. Finally the object of analysis is described in terms of function, functional unit and reference flows (Guinée et al. 2002). Of course in LCSA studies these issues need to be defined as well, however because of the complexity of LCSA studies, increasing emphasis should be put in particular on framing the right question (Zamagni 2010). A well-defined question will help to choose the right approach and tools for solving this question and thus should be framed in close relation to the relevant (sustainability) criteria shaping decisions at hand (Zamagni 2010). It is important to involve the stakeholders when investigating what the problem is, what the system concerned does, what impacts an intended decision might have, if the system analysed simply replaces other systems on a small scale, or if the technology used is expected play a role on a larger scale (Zamagni et al. 2009; Zamagni 2010).

Modelling

The modelling block in figure 2.1 shows an overview of models, methodologies and tools that can be used for calculating specific sustainability impacts. Selecting and applying the proper model to answer a specific sustainability question however is the main challenge (Guinée et al. 2011) but is guided by three modelling considerations that will be described shortly below.

A broadened scope of indicators

LCSA incorporates not only environmental impacts but also economic and social impacts to cover all dimensions of sustainability, what is presented as "broadening the scope of indicators". This is in line with a general accepted view on sustainability assessments that should not only involve environmental impacts or consequences of decisions but also social and economic impacts (Kloepffer 2008; Dobon et al. 2011). A broadened scope calls for a different approach for analysis. In the classic inventory analysis (LCIA), physical flows that connect processes are defined and in the impact assessment specific environmental impacts are calculated from the properties of these flows. Where it is possible to connect a price to a physical flow, most other economic and social impacts are not connected to a system by physical flows but by economic, political, behavioral or cultural relations (Guinée and Heijungs 2011). This makes it impossible to take the conventional scientific reductionist approach used in LCA to model economic and

social impacts (Halog and Manik 2011). The proper way of assessing these impacts depends on the specific study at hand. Guidance can be sought in experiences with Life Cycle Costing (LCC) and Social Life Cycle Assessment (SLCA), where attention needs to be given to the fact that these are assessment tools appropriate for product level assessments.

A broadened object of analysis

Where classic LCA focusses on product level assessments, contemporary sustainability questions go beyond products and services and involve questions concerning technologies, product groups or industries (meso-level) and even state economies or geographical or political entities (economy-wide). This is presented as "broadening the object of analysis". Recent publications on LCSA mainly focus on the product or service level for their assessment (Dobon et al. 2011; Brandão et al. 2010; Gheewala et al. 2011; Achilleos et al. 2011; Schau et al. 2011). Kissinger (2010) stresses that increasing interdependencies of our world economy as a result of globalization also determine the need of assessing sustainability on the same (global) scale. Sustainability in a globalizing world demands that the scale of material flows and impact studies match the scale of human economic activities (Kissinger and Rees 2010). Schau (2011) identifies a possible conflict between the microeconomic aim of reducing cost, mainly concerned on a product level, and the higher level goal of economic growth. Which shows that decisions aimed at improving sustainability on a product level might have adverse effects on a higher level when practiced on a larger scale. Impacts related to choices made at the product level cannot simply be extrapolated to find the impacts on an economy wide level. Modelling of impacts beyond product level can benefit from experiences in economic modelling and environmentally extended input-output assessment (E-IOA).

Deepening

In the CALCAS framework the concept of deepening is introduced to emphasize that broadening the scope of indicators and object of analysis requires a deepened assessment approach. A specific impact cannot longer be attributed to a specific flow alone, but also depends on other impacts or other than physical relations that can be identified in the system studied. Sustainability impacts in LCSA are not only physically related to processes or flows of commodities but are related to a system or process via economic, social, political and cultural relations and mechanisms (Zamagni et al. 2009). The concept of deepening displays the rationale behind LCSA. One should keep in mind that the pursuit of only one-sided benefits of decisions may not achieve sustainability (Moriizumi et al. 2010). Meaning that impacts need to be assessed in relation to other impacts and the overall performance of a system. Finkbeiner (Finkbeiner et al. 2010) stresses that to

keep a sustainable balance, trade-offs or interrelations between the three dimensions of sustainability need to be addressed with utmost care. In other words, attention should be assigned to mechanisms like spill-over or rebound effects. Also Halog and Manik (2011) state that covering all possible impacts as separate entities by using models next to each other neglects trans and cross-boundary impacts or interrelationships between and among parts of the study. The added value of performing an integrated sustainability assessment lies in the fact that sustainability as a whole concerns more than the sum of its parts (Halog and Manik 2011).

Interpretation

The final phase of an LCSA study is the interpretation which is comparable to the final phase in LCA. It is the phase in which the results of the analysis and all choices and assumptions made during the course of the analysis are evaluated in terms of soundness and robustness, and overall conclusions are drawn. Results are evaluated and analysed by performing consistency and completeness checks and by performing contribution and perturbations analyses (Guinée et al. 2002). Drawing conclusions on favourable alternatives can be a challenge in LCA studies because an alternative might show a decrease in global warming potential but an increase in ecotoxicity. To base decisions on these varying outcomes in LCA weighing practices are used. It is easy to imagine that LCSA studies provide a wider pallet of changing and interrelated impacts, spreading over different dimensions of sustainability and different disciplines. By that even increasing the complexity of formulating and understanding solid conclusions that form a basis for decision making. Kloepffer (2008), Finkbeiner (2010), Heijungs (2010) and Halog (2011) stress the need for transparent and easy to understand results from an LCSA study. Suggestions are made on introducing multi criteria decision analysis (MCDA) for drawing conclusions (Halog and Manik 2011; Moriizumi et al. 2010).

2.3 Towards applicaton of LCSA

The LCSA framework provides a theoretical approach on how to perform an LCSA study. Next to that it provides the supporting concepts of broadening and deepening in modelling the increased complexity of LCSA. Most recent LCSA studies present a broadened scope of indicators and only Morizumi et al. (2010) report on their approach. Using insights from work reported by Zamagni (2009) and Moriizumi et al. (2010) the remainder of this article seeks to provide a systematic and practical approach for life cycle sustainability assessment of new (biosolar) technologies.

Since the experience with LCSA is still limited and there is variety of questions that can be answered by using the LCSA framework, it cannot be expected that there is one

general research approach that fits all LCSA assignments. Here it is suggested that the LCSA level of complexity demands an extensive primary phase in which the object and goal of the study is defined. A thorough system description will provide a solid foundation to frame the right research question and define further research steps, f.e. which impacts to focus on and which models to use to quantify these. In practice, the scope of an LCSA will have to be adapted to the resources available for the study (Zamagni et al. 2009). One should therefore aim for only addressing those issues that are relevant for the specific study instead of aiming for a complete system description.

2.3.1. Describing the system

Figure 2.2 gives a schematic overview of the system concerned in an LCSA study and general themes that need to be addressed. As a reference the system concerned in classic LCA is displayed on the left. Describing the system is an iterative process in itself and will not always be strictly separated from other phases of the LCSA. Insights developed in the process of describing the system will identify additional stakeholders that can be approached to further detail the system. All relevant stakeholders need to be questioned on their concerns, needs, problems, priorities and reasons to act regarding the technology under consideration (Moriizumi et al. 2010).

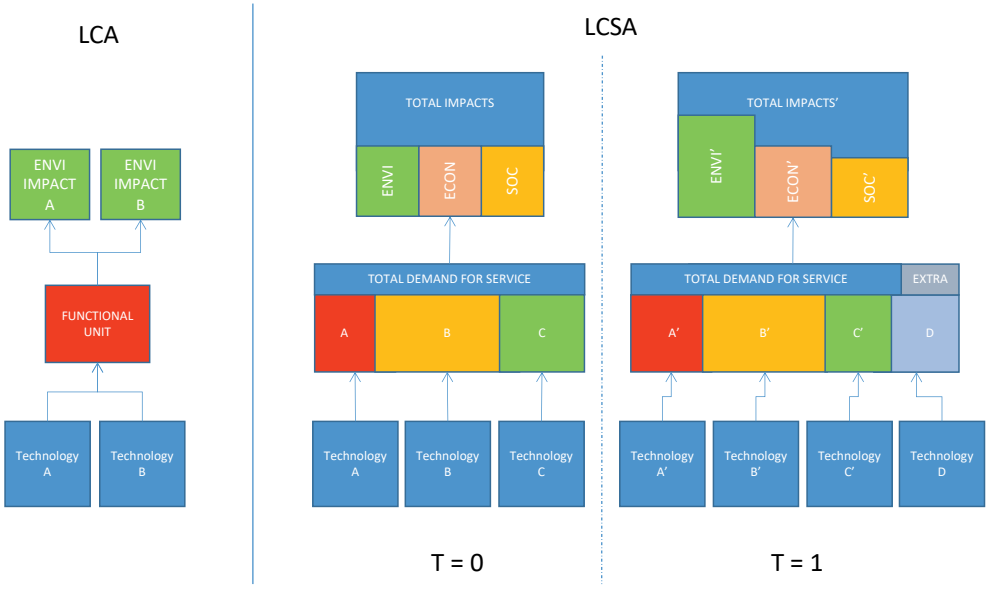

Figure 2.2: LCSA system.

In order to perform a LCSA of a technology one first needs a clear technological description and the application of the technology at hand. The application of a technology contributes to meeting a certain demand for service, for example national or global energy demand. By knowing its application it is possible to identify alternative technologies that also contribute to delivering the same total demand for service. Meeting the total service demand results in environmental, economic and social impacts. If a service demand is provided in a sustainable manner depends on the balance of these impacts in relation to the service provided. Which technologies are deployed and how big their share in meeting the total demand is, depends on the overall impact of the system. In other words if it is technological feasible, economically viable, environmentally beneficial and socially acceptable As an illustration we will give a hypothetical example for total electricity demand. Electricity prices are kept within reasonable limits by using fossil fuel power plants. In addition it is tried to cut CO_2 emissions by investing in more expensive renewable energy options. In the mean while fossil fuel power plants are still under construction because it gives a boost to local economies and renewable energy sources are currently not capable to meet increasing energy demand.

System complexity makes it impossible to identify the direction of causal relations be-tween technology, demand for service and impacts. For the sake of modelling it is assumed here that a certain demand for a service is fulfilled by applying certain technologies and that fulfilling this demand with these technologies results in certain impacts. These impacts might result in the improvement of existing technologies or the introduction of new ones. In the real world the introduction of a new technology in a system sets of a dynamic in which both total impacts and the division of the total demand for service over available technologies will change.

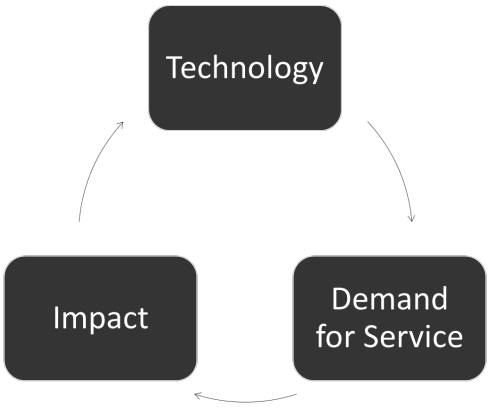

Figure 2.3: Causal relations in modelling LCSA.

Assessing a new technology to identify constraints for development and successful implementation demands for taking developments over time into consideration as well. For developing a technology, its deployment and successful implementation a typical timeframe of 30-40 years can be expected (Hirooka 2006). In this period of time the total demand for service and the (performance of) technologies supplying this service will have changed. A new technology needs to be compared in a future situation. Information on the expected future system especially the demand for service and its supply divided over alternative technologies is required (right side of Figure 2.2). Developing and using scenarios will therefore be one of the steps following the system description.

2.3.2. Proceeding from a system description to a full assessment

Describing the broad system provides a solid basis for approaching next phases that will result in a life cycle sustainability assessment of a technology. It provides insights to frame the right question and relevant stakeholders are identified. By discussing and describing alternative technologies and how these technologies contribute to supply the total service demand also relevant indicators in the three sustainability domains are identified. Information is gathered so far by taking a bottom up approach, by involving stakeholders. From an Industrial Ecologists point of view it is suggested to discuss the system description via a bottom up approach with actors not directly involved to identify possible impacts or points of attention that are probably not ad-dressed but are relevant in a broader view.

After describing the system additional steps need to be taken to finally present quantified sustainability indicators and resulting impacts. These steps consist of defining scenarios on possible developments in technology performance and service demand, finding the right tools to quantify the indicators identified, applying the these tools (including additional data gathering) and interpretation of results presented by the tools applied. All steps for an LCSA to be used to assess biosolar technologies are dis-played in Figure 2.4.

The outcomes of the study need to be used in to discuss what mix of technologies provides the best balance over all sustainable domains. And additionally which performance criteria, depending on sustainable impacts, new biosolar technologies need to fulfil to be added to the list of technologies that will provide the total demand for service.

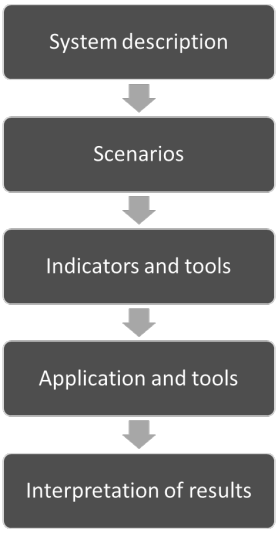

Figure 2.4: Five step approach for LCSA.

2.4 Conclusions and outlook

This paper investigates and proposes a practical approach for performing a life cycle sustainability assessment. It is found that, until now, limited attention is given to the issue of how a LCSA can be approached practically. The paper therefore provides an approach founded in a concise overview of the LCSA framework as presented in the CALCAS project supplemented with insight from recent scientific reporting on LCSA studies. It is argued that a LCSA should be initiated with describing a broad but relevant system description as the first of in total five steps (Figure 2.4). Of this five step approach, the first – system description – has been discussed. The system description identifies and describes the technological description, the intended application of a technology, which share of specific demand for service will be met, which other technologies contribute in meeting this demand, the (relevant indicators for addressing)sustainable impacts of meeting the demand and developments over time. The approach presented in this paper will be brought into practice by applying it in a LCSA study on solar fuels. The practical experience from the latter will provide inputs for fine tuning the approach as presented here.

Acknowledgements
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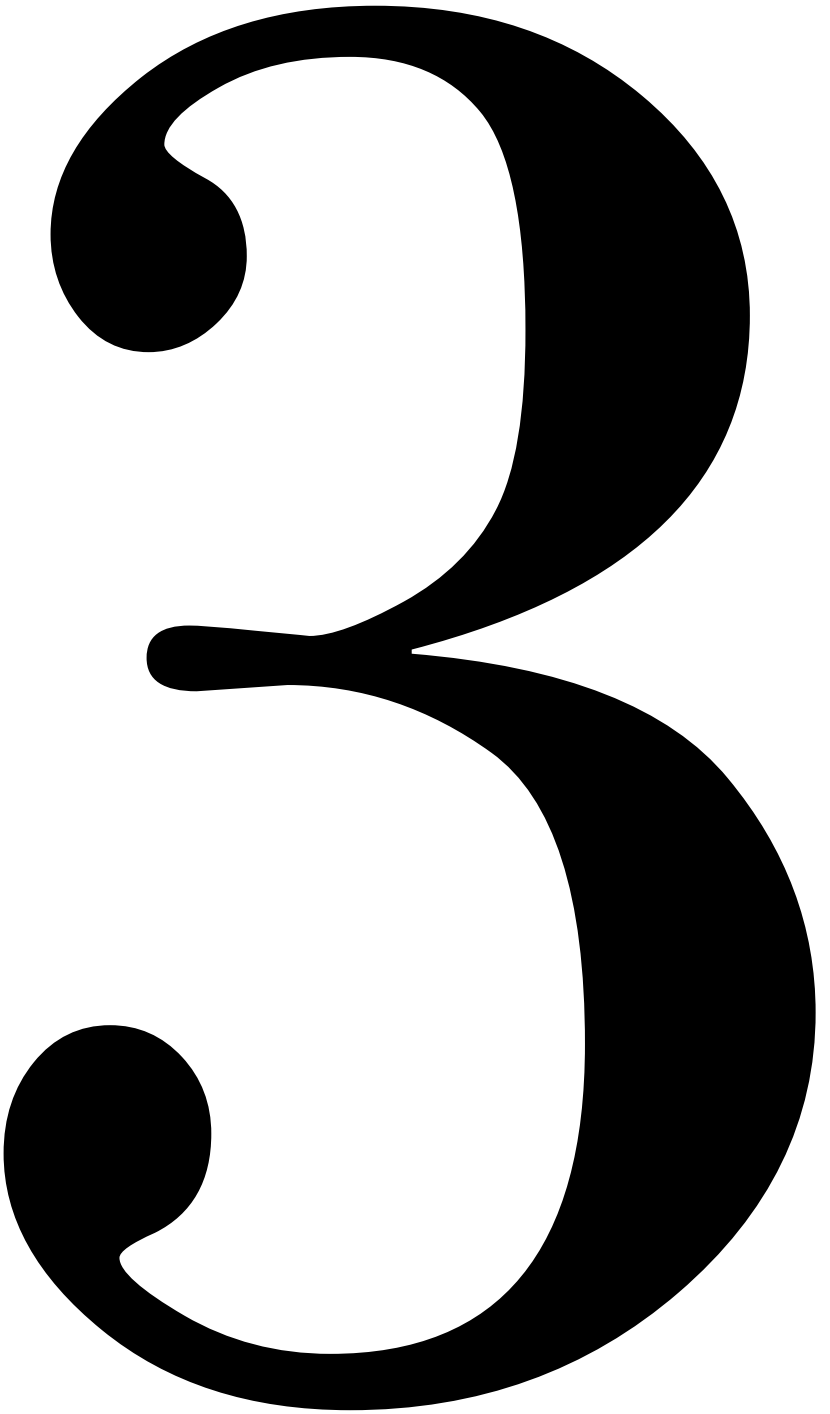

Chapter 3

Energy and Climate Impacts of Producing Synthetic Hydrocarbon Fuels from CO_2

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Abstract

Within the context of carbon dioxide (CO_2) utilization there is an increasing interest in using CO_2 as a resource to produce sustainable liquid hydrocarbon fuels. When these fuels are produced by solely using solar energy they are labelled as solar fuels. In the recent discourse on solar fuels intuitive arguments are used to support the prospects of these fuels. This paper takes a quantitative approach to investigate some of the claims made in this discussion. We analyse the life cycle performance of various classes of solar fuel processes using different primary energy and CO_2 sources. We compare their efficacy with respect to carbon mitigation with ubiquitous fossil-based fuels and conclude that producing liquid hydrocarbon fuels starting from CO_2 by using existing technologies requires much more energy than existing fuels. An improvement in life cycle CO_2 emissions is only found when solar energy and atmospheric CO_2 are used. Producing fuels from CO_2 is a very long term niche at best, not the panacea suggested in the recent public discourse.

3.1 Introduction

There is an increasing interest in the use of carbon dioxide (CO_2) as a resource. The recent introduction of the Journal of CO_2 Utilization and conferences like the Carbon Utilization Summit 2013 (Carbon Utilization Summit 2013 2013), the International Conference on CO_2 Utilization (ICCDU XII 2013 2013) and the 2nd Conference on CO_2 (Nova-Institut GmbH 2013) testify to this. These platforms make the case for (more) research on the conversion of CO_2 into synthetic fuels as means to utilize CO_2 and thereby mitigate its accumulation in the atmosphere. As such it is presented as a useful addition to, or alternative for geological sequestration, offering the prospects of 'productive use' and or 'recycling' as opposed to mere (unproductive, unprofitable) 'storage'. In this spirit, the Global CCS institute initiated a study to investigate possible uses of CO_2 to accelerate the development and commercial deployment of carbon capture and sequestration (CCS)(Global CCS institute and Parsons Brinckerhoff 2011). One of its conclusions is that, once technically mature, producing liquid hydrocarbon fuels from CO_2 might be a promising driver for the development of carbon capture.

In this paper we assess the environmental merits of synthetic liquid hydrocarbon fuels produced from CO_2 compared to alternative fuel production routes. When the required energy for the production of these fuels is taken from the sun, these fuels are also labelled as "solar fuels". This term is at present loosely defined and is applied to a wide variety of sustainable energy pathways, encompassing anything from solar power, solar hydrogen, algal fuels, next-generation biofuels to synthetic fuels from CO_2 using either conventional chemical processes or yet-to-be-invented 'artificial leaves' (Nocera 2012). This ambiguity in definition allows for the opportunistic use of sustainability arguments. It also suggests both a sweeping future scope of solar fuels as a class, as well as having both short-, medium- and long-term prospects while the implied breadth makes it difficult to quantify metrics of success. The following question has been neglected so far: what is success? Under what conditions is a 'solar fuel' better than (a combination of) existing processes? For instance: is recycling CO_2 from a coal power plant into a synthetic fuel with hydrogen generated from renewable energy sources preferable over the direct displacement (phase-out) of coal by that same renewable power and the (continued) use fossil-based liquid fuel?

For this purpose we define solar fuels as liquid hydrocarbon fuels produced from CO_2, water and solar energy. This is a narrower definition than others that might include solar hydrogen or even solar electricity. Since solar electricity or more broadly renewable electricity is an established and well-described branch of technology, we propose not to include it in a new fuzzy term, as long as we have not explicitly established the merit of so doing. The same goes for the production of hydrogen from renewable sources. Hydrogen

production – via natural gas steam reforming or via alkaline electrolysis – is established technology, and hydrogen has an existing brand-name of its own as a renewable fuel. This leaves us with 'solar fuels' as the umbrella term to describe hydrocarbon fuels produced from CO_2 with renewable energy inputs. A solar fuels process can thus be anything contained in a 'box' that has CO_2, water and renewable energy going in, and hydrocarbons coming out. The only ambiguity this leaves is with respect to biofuels. We go forward mindful of that and exclude conventional biofuels from the term (for the same reasons cited above for renewable electricity and hydrogen), but have the term cover (advanced) algal biofuels and other, even more advanced bio-routes, eventually leading to the elusive 'artificial leaf'.

Scientific research in the field of fuel production from CO_2 mainly focuses on specific techniques to convert CO_2 into hydrocarbon fuels. The most common of these techniques are thermochemical and electro-catalytic approaches and the development of new and improved catalysts to convert hydrogen and CO_2 into synthetic fuels (Romero and Steinfeld 2012; Centi and Perathoner 2010; Roy et al. 2010; Inglis et al. 2012; McDaniel et al. 2013; Woolerton et al. 2012; Tran et al. 2012; Haije and Geerlings 2011; Stechel and Miller 2013; Goeppert et al. 2012; Ampelli et al. 2010).Solar fuels can be produced with a solar-to-fuel efficiency of 10% by using existing technologies (Haije and Geerlings 2011). This efficiency is also suggested as a reasonable minimum goal for research and development for solar fuels (Stechel and Miller 2013).

To contextualize our analysis in the subsequent sections, we first present a structured overview of the arguments that we find in the current public discourse to substantiate the desirability or the need for 'solar fuels'. We identify four discrete arguments.

Firstly, solar fuels are considered to contribute to a sustainable energy future because they are renewable (hydrocarbon) fuels that can be used in our existing energy infrastructure. This is the same argument that drives the development of biofuels. There can be no doubt that hydrocarbons have properties that make them highly desirable and virtually irreplaceable in certain end-use sectors, notably heavy-duty transport and aviation. Solar fuels offer the promise of enhancing the scope of renewable fuels beyond biofuels. If biofuels now rely by necessity on photosynthesis, a solar fuels process (in our hypothetical 'box') would have the same functional use (CO_2, water and renewable energy in; fuel out), but would be driven by a process other than (natural) photosynthesis. This raises two questions: is the solar fuels process better than the incumbents? And: Are solar fuels easier, cheaper or otherwise more attractive than a shift away from hydrocarbon fuels, to hydrogen or electricity?

Secondly, – more intuitive in character – is the argument that solar fuels provide a synergistic solution to the problem of overabundance of CO_2 and the abundance of renewable energy on the one hand, and the scarcity of hydrocarbon fuels on the other. It follows that producing the latter from the former must be inherently attractive (Goeppert et al. 2012; Olah et al. 2011; Keith 2009; Lackner 2009). But is CO_2 really abundantly available? It is abundant only in diluted form, and it takes effort to make it available as a practical feedstock (Roy et al. 2010). And how does the release of CO_2 upon combustion of the solar fuel compromise its value proposition, especially if the CO_2 is of fossil origin? It delays rather than avoids the release of CO_2 into the atmosphere, which is why some – but by no means all – proponents stress that atmospheric CO_2 should be used to produce solar fuels (Stechel and Miller 2013; Goeppert et al. 2012; Olah et al. 2011; Wang 2012; Graves et al. 2011). This study investigates to what extent using CO_2 from a specific source makes a difference to solar fuels' claimed carbon neutrality and also takes into account additional system related GHG emissions.

Thirdly, solar fuels offer the promise of solar energy storage – arguably one of the biggest challenges in a world predominantly relying on renewables (Romero and Steinfeld 2012; Roy et al. 2010; Haije and Geerlings 2011; Gray 2009). The world needs scalable energy storage solutions. But for this solar fuels need to outperform other (existing) energy storage options such as batteries or hydrogen storage, of which the latter is excluded from our solar fuels definition. These options are equally suited for renewables matching, have no a priori limit to scale and are likely to have higher round-trip efficiencies than solar fuels. Moreover, for the production of solar fuels using existing technologies, it can be expected that a continuous process with a continuous resource supply provides the best means of production. A continuous supply of renewable energy is simply not possible, unless one of the previous mentioned energy storage technologies is used.

Finally there is the claim that the costs of carbon capture can be offset by producing valuable fuels or chemical products (CO_2 as a feedstock for chemicals is another strand of recent research activity. Many of the problems/issues are similar to those of solar fuels. We merely note this, while staying focused in this paper on fuel production) from CO_2 which is inherently more financially attractive than storing it underground (Global CCS institute and Parsons Brinckerhoff 2011). But is it? The aim of sequestering CO_2 underground is to prevent its accumulation in the atmosphere while all fuels are eventually combusted, releasing the painstakingly captured CO_2 once again into the atmosphere. Although a possible profit can be made by producing and selling fuels, this argument completely abandons the primary aim of carbon sequestration.

If we review these four broad claim areas, we observe that all of them have merit in principle, but all of them call forth immediate questions of utility. These questions often cannot be answered in abstracto but will require some form of (quantitative) life cycle assessment (LCA). Although the importance of a life cycle approach for these questions has been acknowledged (von der Assen et al. 2013, 2014), the authors are not aware of any existing LCA studies on CO_2 utilization. It has been the observation of the authors that – in the absence of such evidence – the participants in the public and scientific discourse on solar fuels use any or all of the four claims above to substantiate their argument. In the absence of quantitative evidence for or against, four intuitively persuasive arguments make a very strong case indeed. This is the state of affairs as we perceive it to be. It is the purpose of this paper to hold the claims up to quantitative scrutiny and review the general merit of the idea of solar fuels – subject to the definition above.

Here we question if the production of liquid hydrocarbon fuels from CO_2 using existing technologies leads to lower life cycle greenhouse gas emissions and cumulative energy demands than existing liquid fuel routes and aim to answer this question by performing a life cycle assessment (LCA). First the required technologies are identified and possible production routes are described based on the origin of the energy and CO_2 that is used. Then the required processes and technologies are described in such detail that the LCA model can be constructed. Third, the total greenhouse gas emissions and cumulative energy demand are assessed and compared to existing liquid fuel routes. We close by discussing the results and possible implications.

3.2 Research scope and approach

The scope of this study concerns a hypothetical fuel production system that converts CO_2 into liquid hydrocarbon fuels using proven technologies. In this study fuels produced in this way are labelled as synthetic fuels, where the route that uses solar energy is considered to produce solar fuels. Because the system is ultimately intended to be solar energy based it is assumed to be located in Spain, thereby specifying the use of life cycle inventory (LCI) data for this geographical area.

Figure 3.1 shows an overview of two possible system layouts to produce liquid hydrocarbon fuels from CO_2. Fischer-Tropsch (FT) technology is used to convert a combination of CO and hydrogen, called syngas, into synthetic liquid hydrocarbon fuels. CO can be obtained from CO_2 via the reversed water gas shift (RWGS) reaction and hydrogen is produced via water electrolysis. The required CO_2 is obtained from fossil fuelled power plants or directly from the atmosphere. When natural gas or biomass

are combusted, it is assumed that all CO_2 and energy from this process will be used to produce liquid hydrocarbon fuels. Additional electricity supply is required to produce sufficient hydrogen to convert all CO_2 into liquid fuel. In the case of using CO_2 from the atmosphere, all required electricity is generated by PV.

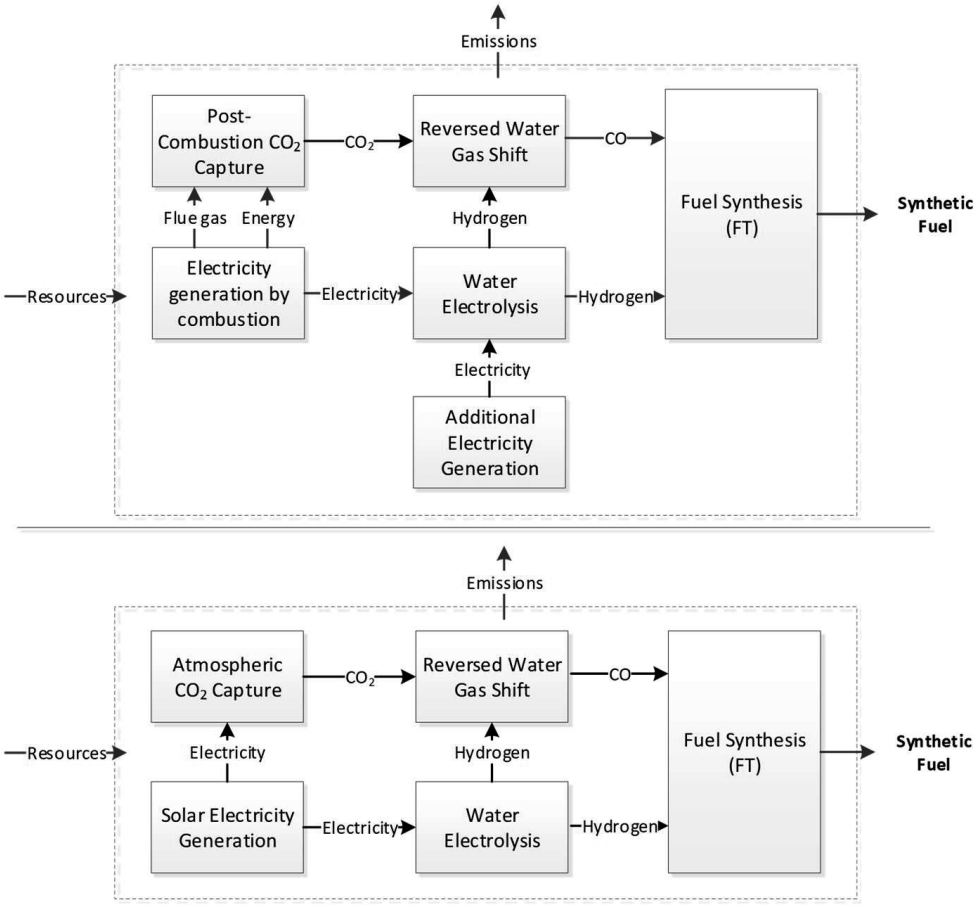


Figure 3.1: Fuel production from fuel combustion based energy and CO_2 (top) and from atmospheric CO_2 using solar electricity (bottom).

Based on three possible CO_2 sources and three energy sources, nine fuel production routes are defined of which five are assessed (see Table 3.1). The NGNG and BMBM alternative are implausible since they provide a very inefficient route to convert natural gas and biomass into liquid fuels. The preferred and currently applied route for this is gas to liquids (GTL) or biomass to liquids (BTL) technology, both of which are based on gasification. However, in this analysis we focus on the conversion of CO_2 to liquid fuels,

therefore we have chosen to include the NGNG and BMBM routes but we have also added GTL and BTL as reference technologies. The assessment of hybrid alternatives like NGBM and BMNG is excluded because these are not fundamentally different in system layout, causing the outcomes to be within the range of the outcomes for NGNG and BMPV. Combining direct air capture (DAC) with electricity from natural gas or biomass combustion (DACNG or DACBM) is also not fundamentally different from the alternatives already assessed. Where NGNG introduces extra CO_2 emissions connected to the use of additional electricity generation, a DACNG system will emit CO_2 emissions in the same range because only part of the total emissions related to the electricity generated will be captured to be converted into a liquid fuel.

The proposed fuel production routes are assessed through LCA with a main focus on the energy and material inputs. The assessment uses fuel production from source-to-fuel expressed in 1 MJ of liquid fuel as functional unit and since the function of a fuel is to deliver energy through combustion, the CO_2 emissions from combustion are also taken into account when calculating net GHG emissions. The outcomes are compared to the performance of existing hydrocarbon fuels like diesel, bio-ethanol from sugar cane, GTL and BTL diesel. For fair comparison pure ethanol is considered. Where possible the ecoinvent 2.2. database (Swiss Centre for Life Cycle Inventories 2010) was used to provide LCI data for the alternatives. Data for GTL and BTL diesel were taken from the GREET (Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation) model (Elgowainy et al. 2012). CMLCA software version 5.2 (Institute of Environmental Sciences) was used to model the different fuel production routes. The following paragraphs describe the main characteristics of all processes and related assumptions from fuel combustion to CO_2 capture (also see the supporting information).

Table 3.1: Different fuel production routes based on CO_2 and energy sources.

	Electricity from natural gas (NG) combustion	**Electricity from biomass (BM) combustion**	**Electricity from PV**
CO_2 from natural gas (NG) combustion	**NGNG** NG power plant with carbon capture supplies all required CO_2 and 65% of the energy required to generate sufficient H_2 to convert all CO_2. The additional 35% required energy is supplied by a NG power plant without carbon	**NGBM** Not assessed	**NGPV** NG power plant with carbon capture supplies all required CO_2 and 65% of the energy required to generate sufficient H_2 to convert all CO_2. The additional 35% required energy is supplied by PV electricity generation
CO_2 from biomass (BM) combustion	**BMNG** Not assessed	**BMBM** BM power plant with carbon capture supplies all required CO_2 and 10% of the energy required to generate sufficient H_2 to convert all CO_2. The additional 90% required energy is supplied by a BM power plant without carbon capture	**BMPV** BM power plant with carbon capture supplies all required CO_2 and 10% of the energy required to generate sufficient H_2 to convert all CO_2. The additional 90% required energy is supplied by PV electricity generation
CO_2 from direct air capture (DAC)	**NGDAC** Not assessed	**BMDAC** Not assessed	**DACPV** CO_2 is supplied via direct air capture and PV electricity (100%) is used to power this process and supply sufficient hydrogen

*acronyms are based on CO_2 source followed by electricity source. NG = natural gas combustion, BM = biomass combustion, PV = photovoltaic electricity, DAC = direct air capture.

Final fuel combustion

It is assumed that all produced fuels are transported and combusted in a highly comparable manner and that the environmental impacts connected to this part of the life cycle (from gate to end use) do not need to be taken into account. However, since carbon neutrality is an important claim connected to the sustainability of fuels produced from CO_2 this study calculates the GHG balance and takes the hypothetical CO_2 emissions from fuel combustion explicitly into account. It is assumed that GTL and BTL diesel as well as FT diesel have properties comparable to conventional diesel, that is a lower heating value of 43,3 MJ / kg and 0,0716 kg CO_2 emission per MJ fuel combusted (Jungbluth 2007; National Institute of Standards and Technology). For ethanol these numbers are 26,8 MJ / kg and 0,0714 kg CO_2 / MJ (Jungbluth, N., Chudacoff, M., Dauriat, A., Dinkel, F., Doka, G., Faist Emmenegger, M., Gnansounou, E., Kljun, N., Schleiss, K., Spielmann, M., Stettler, C., Sutter 2007).

Fischer - Tropsch process

Fischer-Tropsch (FT) technology is commercially deployed to convert syngas, a 2 : 1 molar ratio mixture of hydrogen and carbon monoxide into liquid hydrocarbon fuels (Haije and Geerlings 2011; Olah et al. 2011; Graves et al. 2011; Eilers et al. 1990). Currently Shell uses the FT technology in their gas-to-liquids (GTL) plant in Qatar where natural gas is converted into syngas (by gasification) which is subsequently converted into liquid fuel. We use the basic performance data of the Shell plant to model the synthesis step in the fuel production processes below, and assume that the straight-run product is equivalent to conventional diesel.

Based on common operating parameters (Eilers et al. 1990; Reinalda 2013) we state the FT reaction to take place at 25 bars (for which both CO_2 and hydrogen are supplied at 25 bar pressure) and a temperature of 230°C with an efficiency of 80%, meaning that 80% of the energy available in the syngas is transferred into fuel. The remaining 20% consists of hydrocarbon chains that are too short to convert into fuels but are sufficient to generate energy to run the GTL plant (Reinalda 2013). The production capacity of this plant is 140.000 barrels of GTL fuel per day, with an average heating value of 43,2 MJ / kg. The process uses 2,3 kg syngas to produce 1 kg of GTL fuel.

Reversed Water Gas Shift (RWGS) reaction

The RWGS reaction is used to produce carbon monoxide (CO) that is blended with hydrogen to form syngas. It is an endothermic equilibrium reaction that uses part of the excess heat produced in the Fischer-Tropsch process for sorbent regeneration (Haije and Geerlings 2011; Reinalda 2013; Haije 2013). Experimental results have shown that CO can be produced from CO_2 and H_2 in a fixed bed reactor which contains a mixture of a

WGS catalyst and a water sorbent. A large scale RWGS reactor has not been constructed until now but could be made available in the short term (Geerlings 2013). Here it is assumed that the RWGS reaction takes place at 230°C / 25 bar and uses 1,57 kg of Carbon dioxide and 0,07 kg of hydrogen to produce 1 kg of carbon monoxide.

Hydrogen production

Hydrogen is commonly produced from natural gas by steam methane reforming (SMR), a process that also emits CO_2. In order not to increase the total system CO_2 emissions further we consider the production of hydrogen through water electrolysis using the electricity from the power plant from which the CO_2 is captured. In the supplementary information an alternative route that uses hydrogen and CO_2 from SMR to produce liquid fuels is discussed. The results of that route are highly comparable to the natural gas based alternatives presented below and are there for not presented in the results section.

The most evolved and widely used water electrolyzing process uses an alkaline electrolyte, typically potassium hydroxide (KOH) which can be retrieved and reused in the process (Tributsch 2008; Pagliaro, M. Konstandopoulos et al. 2010; Kruse et al. 2002; Ivy 2004). Based on standard operating parameters (see section 3.5) the production of 1 kg of hydrogen needs 11 kg of water and uses 56.7 kWh of electricity. An additional amount of 0.66 kWh electricity is needed to compress the hydrogen to 25 bar, which is needed for the downstream processes (Rovers 2005; Sumner 2006).

CO_2 and electricity from fossil fuel combustion

Natural gas or biomass combustion in a power plant with post-combustion carbon capture using monoethanolamine (MEA) provides CO_2 and electricity. Typically only 90% of the CO_2 present in the flue gases is captured, the remaining 10% is emitted into the atmosphere. Because CO_2 and electricity from this power plant are completely used up in the production of FT fuels, there is no need to specifically allocate or assign environmental interventions to electricity or carbon dioxide both produced by the power plant. The required additional electricity to produce sufficient hydrogen to convert all CO_2 into liquid fuel is either generated by an additional power plant without carbon capture running on the same fuel or by PV electricity. PV electricity data is directly taken from the ecoinvent life cycle inventory database (Jungbluth et al. 2007).

Here we assume that a general coal fired power plant with carbon capture can be used for the combustion of biomass as well. When equipped with carbon capture, the energy output from power plants decreases as a result of the energy needed for the carbon capture and compressing the CO_2 to a pressure at which it can be transported. The

energy penalties are dealt with by decreasing the output of energy produced by the power plants, while using the same quantity of fuel input (and related CO_2 emissions). The energy penalties for natural gas and biomass in this study are set at 15% (Van der Giesen 2008) and 30% (International Energy Agency 2011) respectively. An overview of the relevant details for the modelled power plants with and without carbon capture and compression to 25 bars can be found in the supplementary information.

Direct air capture (DAC)

Capturing CO_2 from the atmosphere, also called direct air capture (DAC), is an industrial technology already used for decades in air revitalization systems on submarines and in space vehicles (Herzog 2003) and is considered as a future option for reducing global atmospheric CO_2 concentrations (Olah et al. 2011; Keith 2009; Zenz House et al. 2011). Until now, there has been no experience with large scale application of the technology (Keith 2009) ,and thus solid and transparent information of air capture technology used on larger scales is limited (Herzog 2003; Zenz House et al. 2011). Two routes are proposed for air capture (Stechel and Miller 2013; Olah et al. 2011). One route is using basic absorbents like calcium-, potassium or sodium hydroxides to bind the CO_2 from the atmosphere and regenerate the formed carbonates to release the CO_2 again. Because of the extreme high energy demands related to the regeneration of these absorbents, recent developments focus on the second and less energy intensive route. This route is based on the moisture swing process as developed by Klaus Lackner (2009). This process uses anionic resins that absorb CO_2 when dry and release it when exposed to moisture. It is demonstrated that simply exposing the resin to water (vapour) would release CO_2 from the resin and that a water temperature of 45°C is sufficient to drive most of the CO_2 off the resin and have it revert to the carbonate state that it can be re-used. The sorbent can be regenerated many times without degrading (Lackner 2009). For now we assume that the resin can be used infinitely and that life cycle environmental impacts are so low, that modelling of the resins is not necessary. Lackner proposes a CO_2 capture device that consists of a wind driven filter system, covered with resin, through which ambient air is led and a CO_2 compressor (67 bar) contained in a cargo container (12 m x 2,5m x 3 m). One of these units should be able to collect 1 ton of CO_2 per day. Additionally all the heat that is needed will be produced as an incidental by-product of the compressor. In total 50 kJ / mol CO_2 or 1,1 MJ (=0,31 kWh) electrical energy / kg CO_2 is needed (Lackner 2009).

3.3 Results and impact assessment

Different routes to produce liquid hydrocarbon fuels from CO_2 with existing technologies as defined in Table 1 were assessed on their net greenhouse gas (GHG) emissions and cumulative energy demand (CED) and compared with relevant existing liquid fuel routes. To calculate the net GHG emissions the characterization factors relating to GWP 100 from the CML 2001 impact methodology were used and extensions for biogenic CO_2 and biogenic CO (biogenic addresses emissions from biomass combustion) were added. Also the uptake of CO_2 from the air as a result of biomass growth or using DAC is accounted for. The energy taken into account in calculating the CED is the amount of energy withdrawn from nature (Frischknecht et al. 2007). This is defined as the total energy available in biomass after growth, the energy content of crude oil or the energy that is send from a PV panel to the inverter. The following paragraphs show the schematic system layouts for this routes and discuss their GHG and CED performance.

Producing synthetic fuels from fossil fuel based CO_2 and energy

The top part of figure 3.2 shows a system in which CO_2 and electricity from natural gas combustion are used to produce liquid synthetic fuels. To produce and combust 1 MJ of synthetic fuel this system emits 0,29 kg of CO_2-eq, of which 0,05 kg are emissions from background processes for example needed to build the required infrastructure. The main GHG emitting process is the generation of additional electricity. To generate and capture sufficient CO_2 to produce 1 MJ of liquid fuel 1,5 MJ of natural gas is combusted. The additional electricity is obtained by combusting another 2,9 MJ of natural gas without capturing the emitted carbon dioxide. Energy needed in background processes is 0,68 MJ. This means that in total 5 MJ of primary energy is required to produce 1 MJ of liquid fuel.

Existing alternatives to producing liquid hydrocarbon fuels from fossil resources are conventional diesel or GTL diesel. Both show an expected better performance than the NGNG route described above but do not start from CO_2 as a feedstock. If we apply ecoinvent data (von der Assen et al. 2013) on conventional diesel production in our LCA model including its combustion, the outcomes show a CED of 1,25 MJ / MJ and emits 0,08 kg CO_2-eq/MJ. A system for GTL diesel shows a CED of 1,7 MJ/MJ and emits 0,11 kg CO_2-eq / MJ.24

The biggest share of the total GHG emissions and CED is connected to the generation of additional electricity. A straightforward improvement can therefore be sought in adjusting this process, for example by using PV electricity to provide the additional electricity. The bottom part of figure 3.2 shows such a system. This system shows an

improved performance over the previous but still performs worse than conventional or GTL diesel. The weak performance of both natural gas-based systems is especially visible in their CED scores. These high scores are mainly caused by the inefficient use of natural gas through combustion in which carbon and hydrogen atoms are separated. It is more efficient to use existing CH bonds, present in fossil fuels, than to combine these atoms artificially.

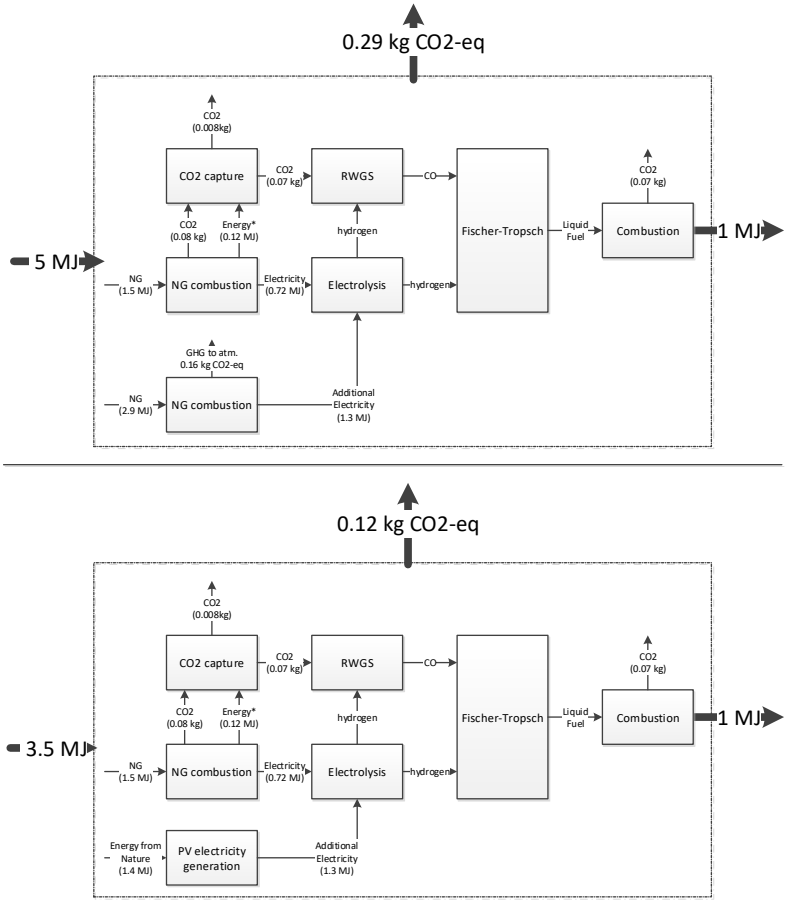


Figure 3.2: Liquid fuel production from natural gas based CO_2 and energy (NGNG) on the top and form natural gas based CO_2 and PV electricity on the bottom (NGPV).

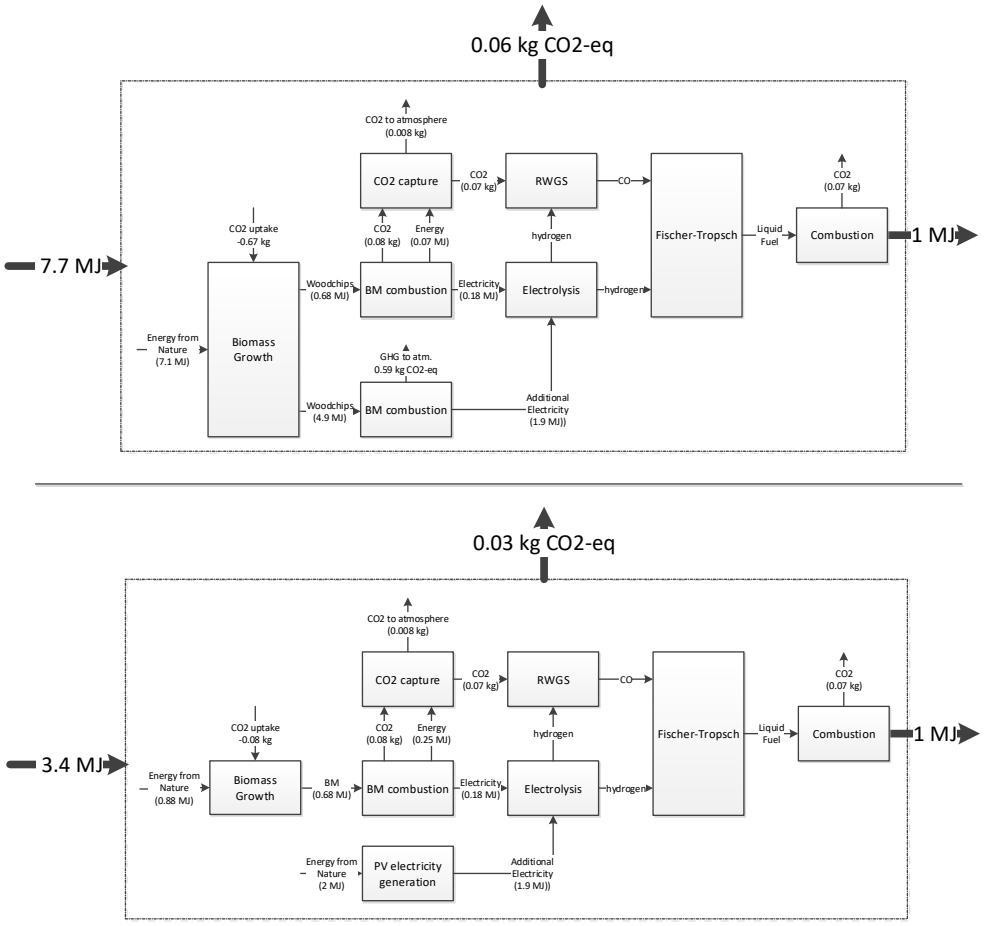


Figure 3.3: Liquid fuel production from biomass based CO_2 and energy (BMBM) on the top and from biomass based CO_2 and PV electricity on the bottom.

Producing synthetic fuels from biomass based CO_2 and energy CO_2 and energy can also be generated through biomass combustion. Because biomass has taken up the CO_2 shortly before it was harvested, the use of CO_2 from biomass combustion is intuitively assumed to be carbon neutral. Figure 3.3 shows a system layout to produce synthetic fuels from CO_2 and electricity from biomass combustion. The production and combustion of 1 MJ of synthetic fuel produced from biomass emits 0,06 kg of CO_2-eq which are all caused by background processes. Also in this system the main GHG emitting process is the generation of additional electricity, while the use of biomass causes a total uptake of 0,67 kg CO_2. To generate sufficient CO_2 and electricity to produce 1 MJ of synthetic liquid fuel, 0,68 MJ of biomass, here in the form of woodchips, needs to be combusted in a power plant equipped with carbon capture technology. For the additional electricity

another 4,9 MJ of woodchips is combusted without carbon capture. To generate this amount of woodchips in total 7,1 MJ of energy was taken up by biomass. In total 7,7 MJ of primary energy is required to produce 1 MJ of liquid fuel from biomass. In comparison with the previously discussed alternatives this system shows lower GHG emissions but much higher energy demand than producing synthetic fuel from natural gas, GTL diesel or conventional diesel. The main cause for this large energy demand is the energy that is lost in converting biomass into woodchips that can be used in a biomass fired power plant and the low efficiencies of biomass combustion.

Existing alternatives to produce liquid hydrocarbon fuels from biomass are ethanol from sugar cane or BTL diesel. Both show an improved performance. By using ecoinvent data in our LCA model it can be derived that ethanol has a CED of 2,9 MJ / MJ and emits 0,04 kg CO_2-eq/MJ.25 BTL diesel shows a CED of 2,1 MJ/MJ and emits 0.01 kg CO_2-eq/MJ (Elgowainy et al. 2012).

Also for the biomass alternative the largest contributing process to GHG emissions and CED is the generation of additional electricity. The bottom part of Figure 3.3 shows a production system in which the additional electricity produced from biomass is replaced by PV electricity. This system shows a better performance compared to biomass FT fuels but performs worse than BTL diesel and slightly better than bio-ethanol regarding GHG emissions. The CED score is worse than both alternatives which is mainly caused by the inefficient use of biomass through combustion and the energy needed to combine carbon and hydrogen atoms, as in the natural gas route.

Producing synthetic fuels from atmospheric CO_2 and PV electricity

Another source for CO_2 is the atmosphere. The energy needed to capture CO_2 from the atmosphere and to generate the required amount of hydrogen can be obtained from non-carbon energy sources, for example PV electricity. In Figure 3.4 a possible system layout is presented.

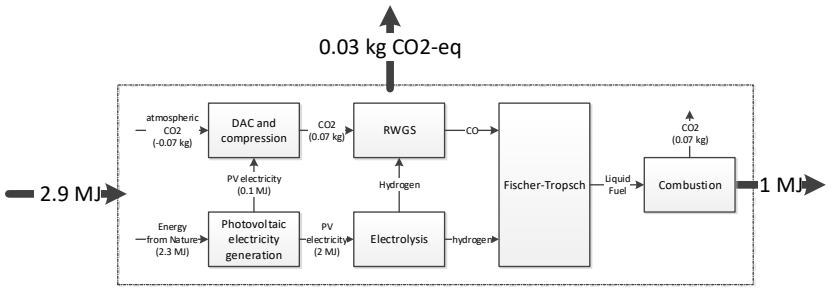
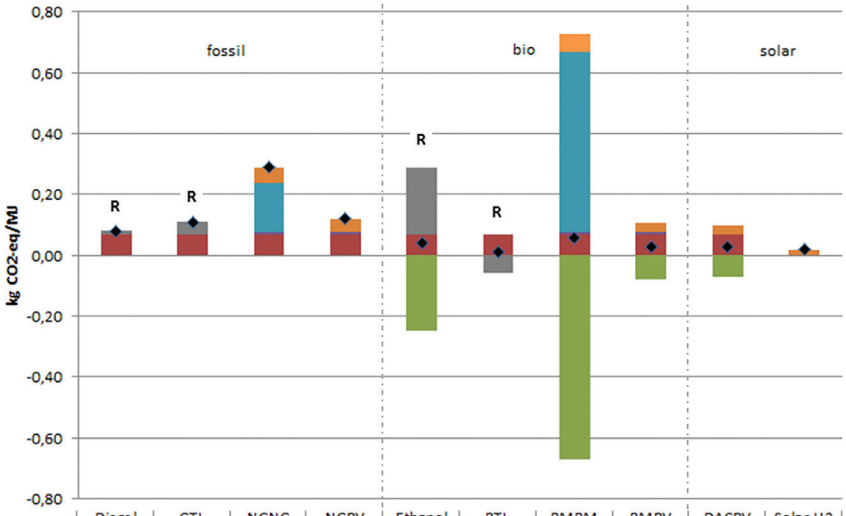


Figure 3.4: Liquid fuel production from atmospheric CO_2 and solar electricity (DACPV).

In total, the system emits 0,03 kg of CO_2-eq if 1 MJ of liquid fuel is produced and combusted. The net GHG emissions are the result of background processes. In total 2,9 MJ of energy is taken up from nature of which 2,3 MJ is transferred into electricity to capture CO_2 and mainly generate hydrogen. The remaining 0,6 MJ is taken up by background processes. Producing synthetic fuels through this system shows an improvement over existing liquid hydrocarbon fuels, like diesel and ethanol, production systems regarding total GHG emissions. This alternative has the best CED performance of all routes producing synthetic fuels from CO_2, but is still worse than existing fuel routes.

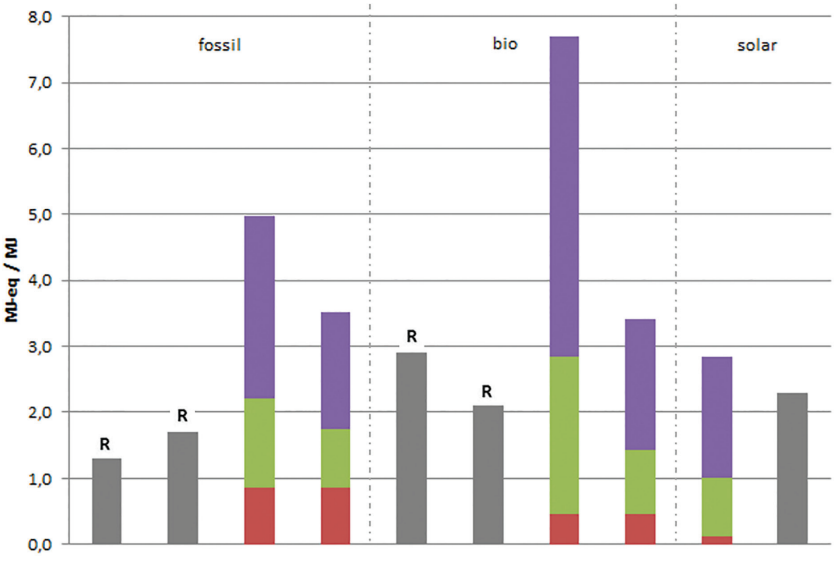
Figure 3.5 and figure 3.6 give an overview of the impact assessment results. In these figures, one extra alternative is added, being solar hydrogen. This alternative does not concern a hydrocarbon fuel, but is expected to be one of the promising transportation fuels in the shorter run, a possible energy storage medium and an intermediate product for producing hydrocarbon fuels from CO_2. It is therefore considered as an important competitor in fulfilling the claims presented in the introduction of this study. Although attention needs to be given to the fact that the required hydrogen infrastructure is not considered here. Solar hydrogen can be produced through water electrolysis with PV electricity and is compressed to around 300 bars to be used in existing hydrogen buses (Rovers 2005). Producing and using 1 MJ hydrogen at a pressure of 300 bars emits 0,02 kg CO_2-eq emissions and uses 2,3 MJ of energy.



	Diesel	GTL	NGNG	NGPV	Ethanol	BTL	BMBM	BMPV	DACPV	Solar-H2
Background	0,01 *	0,04 *	0,05	0,04	0,22 *	-0,06 *	0,06	0,03	0,03	0,02
Additional electricity			0,16	0,00			0,59	0,00	0,00	N/A*
CO2 capture loss			0,01	0,01			0,01	0,01	0,00	
CO2 uptake			0,00	0,00	-0,25		-0,67	-0,08	-0,07	
Fuel combustion	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,07	0,00
◆Total	0,08	0,11	0,29	0,12	0,04	0,01	0,06	0,03	0,03	0,02

Figure 3.5: GHG emissions of different liquid fuel production routes related to a functional unit of 1 MJ. (*The data for diesel, GTL, ethanol, BTL and solar hydrogen cannot be divided into the uptake of CO_2 and connected processes and are therefore aggregated. R indicates reference alternatives not explicitly modelled in this study.)

In Figure 3.5 the GHG emissions for all alternatives discussed are displayed. It is clearly displayed that fossil based production of synthetic fuels does not show any improvement over existing liquid hydrocarbon production routes based on fossil fuels. Considering bio-based fuel production routes, large quantities of carbon dioxide are taken up from the atmosphere, the related background emissions however show that synthetic fuel production is competitive but not a clear improvement in comparison with existing bio fuel routes. Producing liquid hydrocarbon fuels, completely based on solar energy show a competitive result to existing fuels, when looking at GHG emissions.



	Diesel	GTL	NGNG	NGPV	Ethanol	BTL	BMBM	BMPV	DACPV	Solar-H2
H2 to syngas	N/A*	N/A*	2,76	1,78	N/A*	N/A*	4,86	1,99	1,84	N/A*
H2 to RWGS			1,35	0,87			2,38	0,97	0,90	
CO2			0,9	0,9			0,5	0,5	0,1	
Total	1,30	1,70	5,00	3,50	2,90	2,10	7,70	3,40	2,90	2,30

Figure 3.6: Disaggregated CED scores of different liquid fuel production routes related to a functional unit of 1 MJ. (*CED data for diesel, GTL, ethanol, BTL and solar hydrogen are presented in aggregated form since separate production steps for hydrogen and CO_2 cannot be discerned. R indicates reference alternatives not explicitly modelled in this study.)

Figure 3.6 shows an overview of the CED scores for all alternatives. The NGNG and BMBM routes are the least efficient and all synthetic fuel alternatives have higher energy demand than the reference fuel routes. Most energy is not used for producing CO_2 but for providing sufficient hydrogen for both converting CO_2 into CO by RWGS and for producing the syngas that is converted into FT fuel.

The CED scores can also be divided into the use of renewable and non-renewable energy sources (Figure 3.7). The synthetic fuel systems described here all use more energy than existing hydrocarbon fuels, but this energy is for a large part renewable energy and therefore often regarded as freely available.

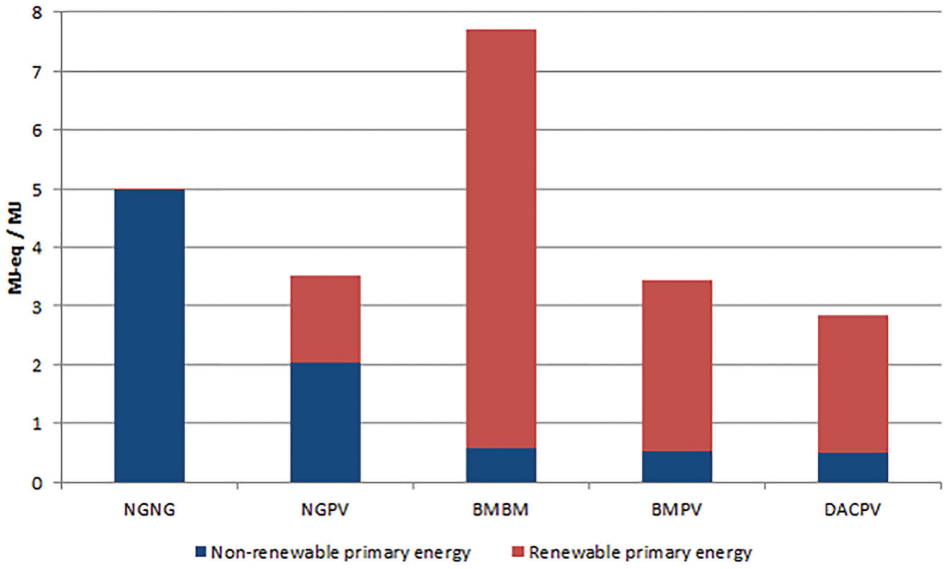


Figure 3.7: Contribution of non-renewable and renewable energy to cumulative energy demand related to a functional unit of 1 MJ.

3.4 Discussion

Synthetic liquid hydrocarbon fuels produced from CO_2 and solar energy have been proposed to contribute to a sustainable energy future. In this study we have investigated four arguments in favour of this claim from the recent public and scientific discourse. Our analysis is based on peer-reviewed data and the results should however be regarded as a first attempt to quantify the CO_2 emissions and energy demand of fuel production from CO_2. This is certainly the case for the DAC technology because this technology is still in the early stages of development and data for industrial scale operations is still lacking.

The claim that fuels produced from CO_2 are renewable is countered. It is shown that even the best performing fuel production route, which is DACPV, both now and in the near future depends on non-renewable fuels and large amounts of renewable electricity. Where the non-renewable energy demand comes from background processes, it can be argued that renewable energy can be deployed for reducing atmospheric CO_2 concentrations more efficiently by replacing fossil based electricity directly than by producing fuels from CO_2 (Graves et al. 2011).

Second, it was claimed that carbon neutral fuels can be produced when overabundant CO_2 is used. The present study clearly shows that using CO_2 from a fossil origin will

not provide a carbon neutral fuel. This supports an earlier claim by Dumont (2012) that "positive environmental effects of a CO_2 consuming reaction cannot be taken for granted" (Dumont et al. 2012). Using atmospheric CO_2 is the best option but does not provide a carbon neutral fuel because of the CO_2 emissions related to fossil energy demand in background processes. This implies that carbon neutral fuels might be produced from CO_2, but only in a future energy system completely based on renewable energy.

The third claim, that producing fuels from CO_2 presents a route to storing renewable energy cannot be completely supported or countered from the LCA results presented here. It is clear that producing fuels from CO_2 by using solar electricity converts solar energy into chemical - and therefore easy to store - energy. To assess if this is a preferable route to store energy, additional research is needed in which the production and use of fuels from CO_2 needs to be compared with battery and hydrogen storage and possibly others storage options, such as water reservoirs.

The fourth argument, that fuel production from CO_2 is preferable to CO_2 sequestration has already partly been countered in the introduction by explaining that producing a fuel from CO_2 neglects the primary goal of carbon sequestration. This counter argument is further supported by the fact that far more energy is needed to produce a fuel from CO_2 than it will deliver when combusted. It would be far more profitable to sell the required energy directly on the market than to lose a great deal of energy in the inefficient production of liquid fuels.

In conclusion, we state that the energy demand and climate impacts of using CO_2 to produce synthetic hydrocarbon fuels by using existing technologies are higher than the impacts of existing hydrocarbon fuels. The intuitive claims in the public discourse cannot be supported by our quantitative assessment which shows that producing liquid fuels from CO_2 is not the straightforward panacea as suggested. However, novel technologies (e.g. photocatalytic, solar thermal and enzyme based routes) might show a slightly improved system performance.

In a distant future, we foresee possible niches for hydro carbon fuels produced from CO_2. Since hydrocarbon fuels offer high energy to mass and volume ratios compared to alternatives like electricity and hydrogen, the application of fuels produced from CO_2 in applications such as airplanes and heavy duty transport may prove to be a valuable but future niche. Another application could be as a storage medium for so-called stranded renewables. Because renewable energy technologies produce difficult-to-store electricity and renewable electricity supply increasingly exceeds demand, research has begun in

levelling supply and demand using concepts such as smart grids. Even in smart grid systems it can be expected that some sort of energy storage will still be needed. For storing large amounts of renewable energy, the production of synthetic hydrocarbon fuels is one of the technologies that could be used. However, it would have to compete with other storage technologies such as batteries, hydrogen storage, water reservoirs or compressed air storage.

Acknowledgements
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3.5 Appendix

This appendix contains the supporting information for this case study and includes details on the modelled processes, supporting calculations.

3.5.1. Foreground process descriptions

Hydrogen generation and supply
Hydrogen is mostly produced from natural gas and, because of its fossil origin, cannot be considered as sustainable or renewable energy carrier. Therefor it needs to be produced by using solar energy as the energy source. According to Tributsch (2008): "the simplest and most elegant photovoltaic hydrogen generator consists of a solar cell and an electrochemical device, an electrolyser, which is liberating hydrogen from liquid water." From the different routes to electrolyze water the process which uses an alkaline electrolyte is the most evolved and widely used (Tributsch 2008) and will therefore be the process used in this exercise. Typically this electrolyte is potassium hydroxide (KOH) that is supplied via a 25 -30% aqueous solution (Kruse et al. 2002; Pagliaro, M. Konstandopoulos et al. 2010; Ivy 2004) and which can be retrieved and reused in the process (Pagliaro, M. Konstandopoulos et al. 2010). Here we consider the production of hydrogen via water electrolysis using photovoltaic (PV) electricity.

Background data for modelling the process of hydrogen production by electrolysis is taken from work performed by Rovers (2005) and Sumner (2006). Both have modelled the production of hydrogen by electrolysis. An overview of the foreground processes modelled in this study can be found in figure 3.8.

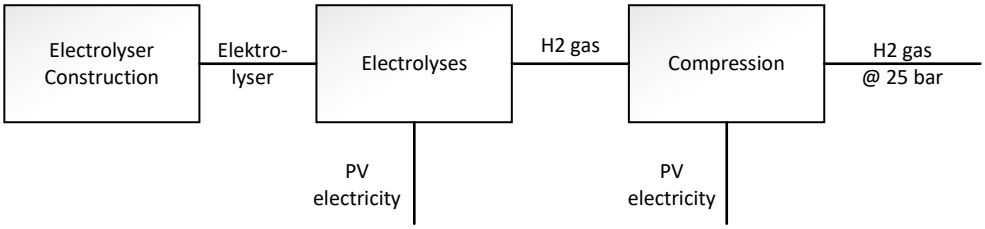


Figure 3.8: LCA scheme: Electrolysis of water for hydrogen production (foreground processes).

Electrolyser construction

Data for the Electrolyser (60 m^2 Hydrogen / hour) is based on Rovers (2005) in which background data from Ecoinvent 1.01 is used. To use this data with the Ecoinvent 2.2 database (Swiss Centre for Life Cycle Inventories 2010) a number of processes have been adjusted. First Rovers used an input of sodium hydroxide which has been changed to potassium hydroxide (KOH). Second, Rovers states that for purging the Electrolyser during maintenance, nitrogen and hydrogen are used and emitted. The updated process contains this amounts of nitrogen as well, the hydrogen can be considered as product emitted to the atmosphere (product loss) resulting from venting the system and is not modelled as an economic input but as an emission. In Table 3.2 all adjustments executed are stated.

Table 3.2: Electrolyser construction, updated from Rovers (2005).

Rovers (2005)	**Rovers (2005), updated to Ecoinvent 2.2 data**
Economic inputs	Economic inputs
Sodium Hydroxide, 50% in H_2O, production mix, at plant, (RER)	Potassium hydroxide, at regional storage (RER)
Transport, lorry 32 t (RER)	Transport, lorry 16 -32t, EURO 3 (RER)
	Added: Nitrogen, liquid at plant (RER)
Resources	*Resources*
Occupation, industrial area, built up (landscape)	Occupation, industrial area, built up (resource_land)
Emissions	*Emissions*
Nitrogen	Cut-off; no environmental impact is associated with nitrogen to air.
Hydrogen	Hydrogen (air_unspecified)

Electrolysis

Data for the electrolysis process itself is based on the process modelled by Sumner (Sumner 2006) and uses the electrolyser modelled above. Sumner's process is compared with existing research and publications on electrolysis (see Table 3.3) which shows no large variety among different data sources.

Table 3.3: Overview of literature on electrolysis; economic inputs per kg hydrogen (STP).

Reference	Inflows		Additional information
	Water	Energy	
Kruse et al. (2002)	N/A	50,1 kWh	η_{syst} = 80%
Pagliaro et al. (2010)	N/A	44,5 – 54,5 kWh	η_{syst} = 55 - 75%, 25-30% KOH aq. solution
Ivy (2004)	11.1 kg	46,7-48,7 kWh	η_{syst} = 80%
Sumner (2006)	11.1 kg	56,7 kWh	N/A
Levene et al. (2006)	N/A	39 kWh	1 bar / 25°C
Stojic et al. (2003)	N/A	50,1 – 55,6 kWh	-
IEA (2007)	N/A	48-60 kWh	1 – 25 bar / 70-90°C
NEL Hydrogen (2012)	10 kg	45,6-48,3 kWh	1 bar / 80 °C, η_{syst} = 80% , 25% KOH aq. sol.
Hydrogenics (2012)	16,7 -22,2 kg	54,5-60,1kWh	1 bar / 80 °C, 30% KOH aq. Solution, HySTAT electrolyser

*Values in this table are all recalculated to the production of 1 kg hydrogen, 1 kg of hydrogen outflow is considered as 11,1 Nm3.

Assuming a lifetime of 10 years an 8000 hours operation a year, one needs 2,31 x 10^{-6} electrolysers to produce 1 kg of hydrogen gas. Table 3.15 shows the economic inputs for the production of 1 kg of hydrogen gas.

Hydrogen compression

Hydrogen is produced at atmospheric pressure and temperature. For the other processes downstream in the FT fuel production chain, the hydrogen needs to be pressurized to 25 bar. For compressing hydrogen the process in this study is modelled by using information from Sumner (2006). Sumner modelled a compressor that compresses hydrogen to 85 bars, requiring 2,23 kWh / kg hydrogen. Because the present study assumes a pressure of 25 bars the energy requirements for compression are 25 / 85 * 2,23 = 0,66 kWh/kg.

Assuming a compressing capacity of 300m^3/hr, a 10 year lifetime and 8000 hours operation a year one needs 3,42 x 10^{-8} compressor to compress 1 kg hydrogen gas. In

Table 3.16 and Table 3.17 the economic inputs for compressing 1 kg of hydrogen to 25 bar can be found.

Syngas production via RWGS

Haije and Geerlings (2011) suggest to use the reverse water-gas shift reaction (RWGS) to produce carbon monoxide (CO) that will be blended with hydrogen to form syngas (see Figure 3.9). The data used in modelling this process is checked with the authors via personal communication.

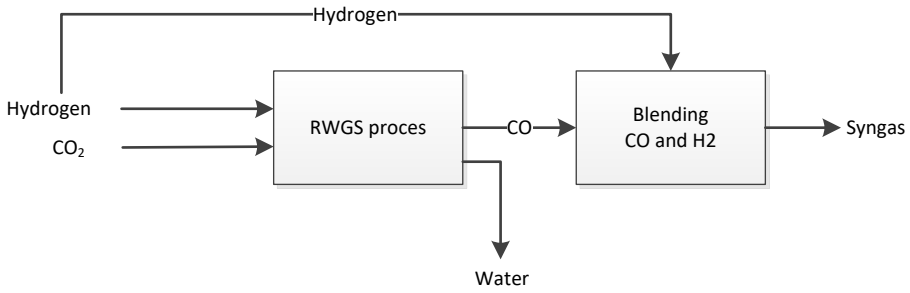


Figure 3.9: General overview of syngas production.

The RWGS reaction is an endothermic equilibrium reaction. At lower temperatures the conversion of CO_2 and H_2 into CO is 100% stoichiometric, regarding that all water that is produced in the reaction is selectively removed to prevent the back reaction, called water gas shift (Haije and Geerlings 2011). With a simple lab experiment it is shown that CO can be produced from CO_2 an H_2 in a fixed bed reactor which contains a mixture of a WGS catalyst and a water sorbent. The heat that is released by the sorbent reaction inside the reactor is considered to be sufficient to drive the RWGS reaction. Excess heat from the Fischer-Tropsch reaction downstream in the FT fuel production process can be used to regenerate the sorbent (Haije 2013). It is assumed that after a 30 minute cycle of CO generation another 30 minute cycle of sorbent regeneration is required per reactor. Using two reactors at the same time, one in adsorbing and the other in desorbing modus, makes a continuous process possible (see Figure 3.10).

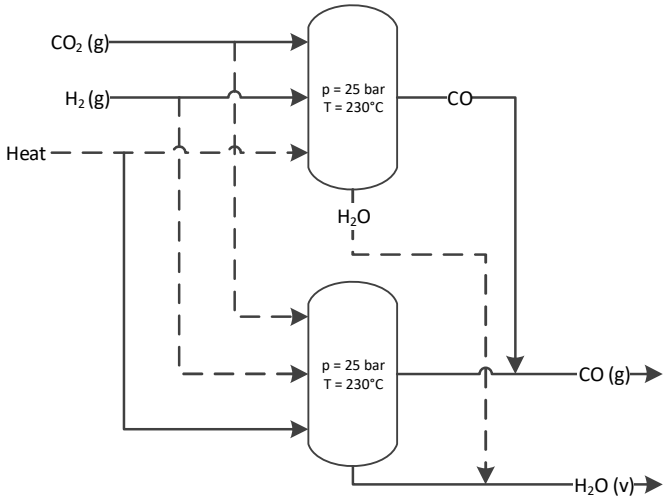


Figure 3.10: Hypothetical configuration RWGS process using two reactors.

For modelling the RWGS process in an LCA model the foreground processes as shown in Figure 3.11 are considered. In order to produce carbon monoxide via the RWGS process a fixed bed reactor and chemicals like a water sorbent and RWGS catalyst are needed. Below short descriptions of modelling these flows and processes are given.

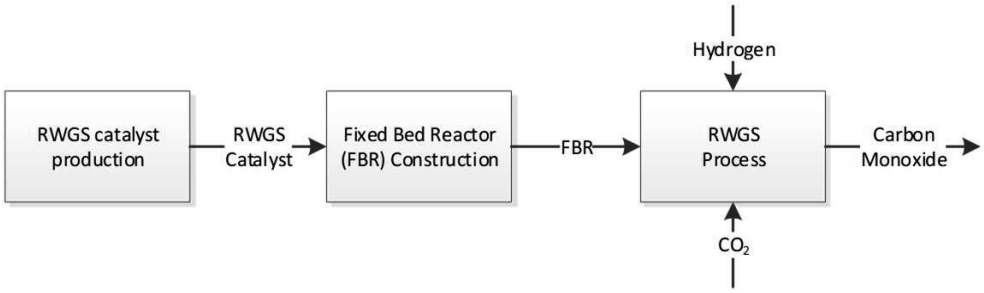


Figure 3.11: Hypothetical configuration RWGS process using two reactors.

RWGS catalyst and water sorbent

As RWGS catalyst Südchemie Shiftmax 240 is proposed. This catalyst consists of 57% CuO, 31% ZnO and 11% Al_2O_3. In the LCA model the RWGS catalyst is assumed to be composed as presented in Table 3.18.

As water sorbent zeolite (Merck) is proposed, which is modelled as zeolite powder provided by the Econinvent v2.2 database. Both chemicals are assumed to be regenerated infinitely and are therefore considered as part of the fixed bed reactor.

Fixed bed reactor construction
The fixed bed reactor is assumed a steel vessel as shown in Figure 3.12.

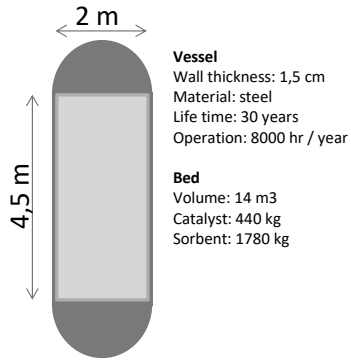


Figure 3.12: Hypothetical fixed bed reactor.

The reactor vessel consists of 4760 kg of steel (assuming 50% low alloyed steel and 50% chromium steel) and is filled with a mixture of 440 kg of RWGS catalyst and 1780 kg of sorbent, which need to be renewed every 4 years (Haije 2013). Over a reactor lifetime of 30 years therefore 30/4 x 440 kg = 3300 kg of catalyst and 30/4 x 1780 kg = 13350 kg of sorbent is needed. Since two identical reactors are use in parallel all numbers are doubled in the unit process representing the construction of the fixed bed reactor (see Table 3.19).

RWGS process
With regard to the Fischer-Tropsch (FT) reaction downstream for which the produced carbon monoxide is an inflow it is assumed that the RWGS reaction takes place at a pressure of 25 bars and a temperature of 230°C, being high enough for proper performance of the FT catalyst. It is assumed that the experimental outcomes (retrieved from a 1 bar reactor) can be extrapolated to the conditions above. All flows are related to the production of 1 kg of carbon monoxide gas @ 25bar / 230°C (Haije 2013).

By extrapolating Haije's experimental results, it can be assumed that 2900 kg of CO gas can be produced in one cycle of 30 minutes, or 1 kg CO gas / 0,6 seconds. Considering a reactor lifetime of 30 years and 8000 operating hours a year, the production of 1 kg of CO gas requires the input of $1{,}93 \times 10^{-9}$ fixed bed reactor.

For every kg of CO produced also 0,64 kg of water vapour is produced as a by-product. In this study this flow is not modelled (cut-off), but it can be considered to use this flow as a (partial) water input for other processes. Table 3.21 shows the in- and outflows of the RWGS process in this study.

Producing syngas by blending carbon monoxide and hydrogen

For obtaining syngas with the required H_2 : CO ratio of 2 : 1, hydrogen and carbon monoxide gas are blended. The syngas has a calculated net heating value of 23,9 MJ/kg and a calculated density of 0,470 kg/m^3 (@ 273 K / 1 bar) or 6,38 kg/m^3 (@ 503K / 25 bar). Producing 1 kg of syngas requires blending 0,125 kg hydrogen with 0,875 kg carbon monoxide.

Gas to liquids conversion (GTL)

For modelling the GTL process in the present study general numbers from public literature combined with public available data on the Pearl GTL plant in Qatar, operated by Shell are used and checked with Shell GTL specialists. In general producing liquid fuels from gas takes place as presented in Figure 3.13. First natural gas is transferred into syngas and liquefied natural gas products like LPG, Ethane and condensate. The syngas is then fed into the Fischer-Tropsch (FT) reactor where it is converted into syncrude that is subsequently cracked into a variety of GTL products like Naphta, Kerosene, Gasoil and Base-oil. In the Shell's Pearl GTL facility in Qatar daily 120.000 barrels of gas products are produced and 140.000 barrels of GTL products.

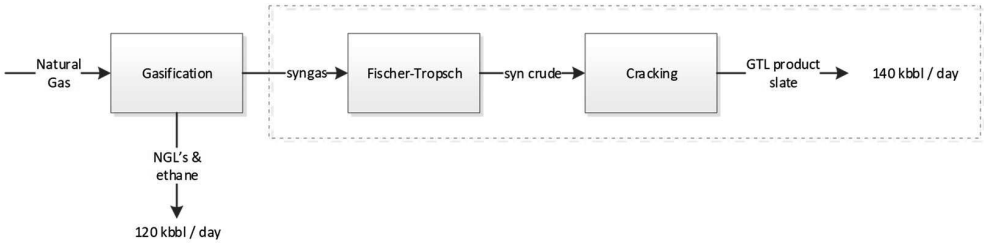


Figure 3.13: General GTL process lay-out.

For this study we will focus on the processes within the dotted line in Figure S6. The syngas used in the FT-process has a hydrogen : carbon monoxide ratio of 2 : 1 (Eilers et al. 1990; Reinalda 2013). The FT process takes place in a fixed bed reactor in which it is run over a metal based catalyst, in this study cobalt is assumed as being the main component, where the syngas is converted to syncrude. The FT process is an exothermic reaction, which takes place at pressures between 20 – 60 bars and temperatures 180 – 250°C (Tijmensen et al. 2002; Boerigter et al. 2004). Here we assume the FT reaction to take place at 25 bars and 230°C. FT efficiencies have been reported from 70% (Tijmensen et al. 2002) to 80% (Boerigter et al. 2004; Forman et al. 2011), meaning that 80% of the energy available in the syngas is transferred into syncrude. The remaining 20% consists of hydrocarbon chains that are too short to convert into fuels and which are mainly used to generate electricity to run the GTL plant and of heat that can be used

to produce steam. In this study it is assumed that the energy not transferred into fuels is completely used for internal energy and for regeneration of the RWGS catalyst and is not modelled separately. The efficiency for the hydrocracking process in which syncrude is transferred into different GTL products is assumed as 100% (Boerigter et al. 2004). The GTL process that will be modelled here, has a syngas to GTL product efficiency of 1 x 0,8 = 80% (see Figure 3.14).

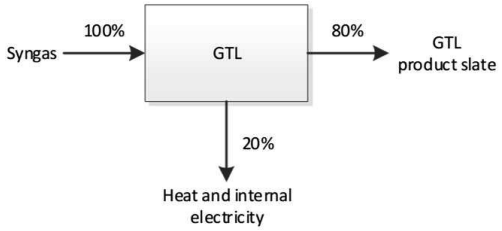


Figure 3.14: General energy balance GTL process.

The Pearl GTL plant in Qatar is designed for optimal diesel production. The GTL products are divided into naphtha, kerosene, gasoil and base oil (Gainsborough 2009). Using the efficiencies discussed above and this product slate the quantity of syngas required is calculated on the basis of energy content, using the syngas properties calculated before (see Table 3.4 for details). The FT fuel regarded in this study that is produced is a mix of all products produced. Its LHV value is also derived from the same table and is set at 43,2 MJ/kg.

Table 3.4: GTL in- and outflows energy balance.

	Density	**Production**			**Net caloric value**	**Energy content**	**Energy content**
Products OUT	[kg/m^3]	[kbbl/day]	[m^3/day]	[ton/day]	[MJ/kg]	[GJ]	[%]
Naphtha	768	35	5.565	4.274	44,5	190.189	19%
Kerosene	800	25	3.975	3.180	45,0	143.100	15%
Gasoil	832	50	7.950	6.614	43,3	286.404	29%
Base oil	860	30	4.770	4.102	40,2	164.908	17%
Total Product slate		140	22.260	18.171	43,2	784.601	**80%**
Heat	N/A	N/A	N/A	N/A	N/A	196.150	**20%**
Product IN							
Syngas	6,38	N/A	N/A	40.967	23,94	980.752	**100%**

*kbll = 1000 barrel = 1000 x 159 litre.

Based on the Shell GTL process as presented above a LCA unit process in constructed in Figure 3.16.

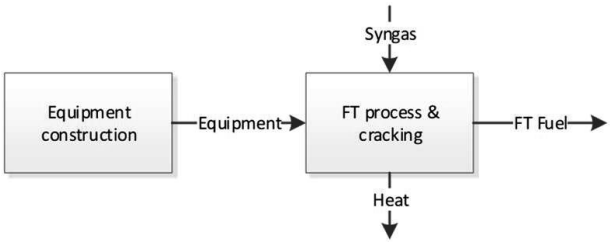


Figure 3.15: LCA unit process for GTL.

GTL Plant

The material demand of the required GTL plant is based on publicly available data and comprises the use of concrete, steel and the area occupied by the plant. An overview of required materials is given in Table 3.5.

Table 3.5: LoremMaterial requirements for GTL plant.

Resource	Value	Reference
Land area	1.4 x 1.6 km = 2.240.000 m^2	Green Car Congress (2006)
Steel use	24 steel reactors weighing 1200 ton each = 28.800 ton	Shell (2009a)
Concrete	690.000 m^3	Shell (2009b)

Since the gasification step is excluded from the process considered here, only about half of the plant materials are required. Resulting of an input of 1.120.000 m^2 of land, 14.400 ton of steel in an assumed equal division of reinforcing and low alloyed steel, 345.000 m^3 of concrete (Table 3.22).

GTL production

Based on energy contents of flows and an assumed GTL plant efficiency 80% the GTL plant converts 2,3 kg of syngas to 1 kg of GTL product with an average heating value of 43,2 MJ / kg. Concerning that the Shell Pearl GTL plant in Qatar has a production capacity of 140.000 barrels or around 18.000 ton (1000 kg) per day. Assuming 300 days operation per year and lifetime of 30 years for the reactor, per kg of solar fuel 1 / (300*30*18000*1000) = 6,7 x 10^{-12} reactor is needed.

Energy and carbon dioxide supply

To produce 1 kg of synthetic fuel using these technologies 3,16 kg CO_2 and 24,6 kWh of electricity is needed (see Figure 3.20). When we want to convert all CO_2 into solar

fuel a ratio between electricity and CO_2 can be calculated; being 24,6 kWh / 3.16 kg CO_2 = 7,78 kWh / kg CO_2.

We assume that electricity and CO_2 are supplied in a combined manner by combusting natural gas and biomass to generate electricity and capture the emitted CO_2 by post-combustion carbon capture. Electricity and CO_2 can also be generated independently via photovoltaic (PV) electricity and direct air capture (DAC). The combustion of fuels in this study provides us with two products from one process, which calls for allocation. However, because all the electricity and CO_2 produced in this process are completely used in the downstream processes allocation is not needed.

Natural gas and coal combustion with post-combustion carbon capture

An LCA study on the combustion of natural gas and pulverized coal with post-combustion capture has been done by Van der Giesen (2008). Data for the power plants and fuels supply was based on the Ecoinvent database while carbon capture processes were added. Here we assume that the coal fired power plant and carbon capture plant can be used for the combustion of biomass as well. The power plants used in this study are based in Spain.

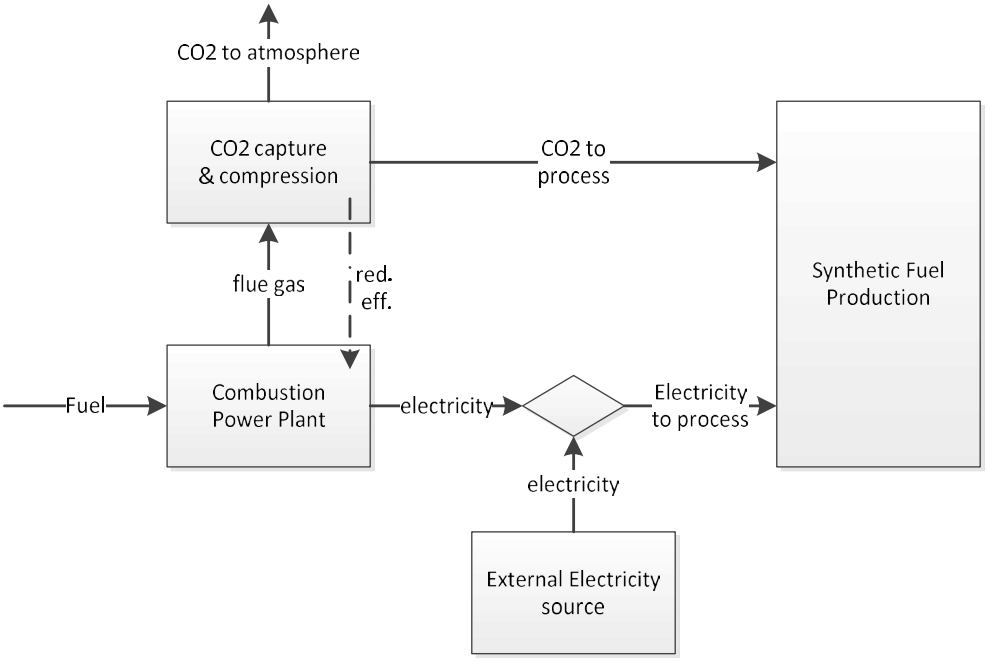


Figure 3.16: Fuel combustion with post combustion carbon capture for electricity and CO_2 generation.

When equipped with carbon capture the energy output from power plants decreases as a result from energy needed for the carbon capture like pumps and compressing the CO_2 to a pressure at which it can be transferred. The energy penalties are dealt with by decreasing the output of energy produced by the power plants while using the same quantity of fuel input (and related CO_2 emissions). The energy penalties for natural gas and biomass in this study are set at 15% (Van der Giesen 2008) and 30% (International Energy Agency 2011) respectively.

An overview of the relevant details for the modelled power plants with and without carbon capture and compression can be found in the table below. The main article excludes the use of coal because this alternative is worse than using natural gas and the exercise does not provide additional insights.

Table 3.6: Power plant properties.

	NGCC power plant	**BM power plant**	**Unit**
Power	400	400	MWe
Efficiency	57,70%	29,70%	%
Lifetime	180000	150000	hours
Operation	8000	8000	hours / year
Annual electricity production	3,20E+06	3,20E+06	MWh
Plant CO_2 emissions	0,35	1,14	kg CO_2 / kWh
Annual CO_2 emission	1,12	3,65	MT CO_2 / year
CO_2 capture data			
Energy penalty	15%	30%	%
Annual electricity prod, with capture	2,72E+06	2,24E+06	MWh
Annual CO_2 production (90%)	1,01	3,29	MT CO_2 / year
Annual CO_2 emissions to atmosphere	0,11	0,37	MT CO_2 / year
Synthetic fuel production requirements			
Electricity required per kg CO_2 for SF prod	7,78	7,78	kWh / kg CO_2
Electricity available per kg CO_2 ATP	2,70 (35%)	0,68 (9%)	kWh / kg CO_2
Additional electricity needed	5,08 (65%)	7,10 (91%)	kWh / kg CO_2

Biomass combustion and post-combustion carbon capture

For electricity generation by the combustion of wood with carbon capture to supply solar fuel production with energy and CO_2 a hypothetical power plant that uses 100% biomass (short rotation wood) is modelled. The power plant and CO_2 capture plant are assumed to be copies of the pulverized coal fired power plant and capture unit modelled by Van der Giesen (2008). Where this system might not be explicitly representative for a real world situation, it is expected to give valuable insights. Next to that the model is built in such a way that any adjustments that might need to be done in the future are easily implemented. Although the best proven technology for conversion from biomass into energy is combustion of wood, this is mainly done in small and medium sized units. For large scale power plants biomass is co-fired with coal because of limited regional biomass availability. As an overall electrical efficiency of bio electricity 30% is assumed (Nussbaumer 2003). The following paragraphs will explain the explicit modelling choices, assumptions and data sources for modelling biomass combustion with carbon capture, following Figure 3.17.

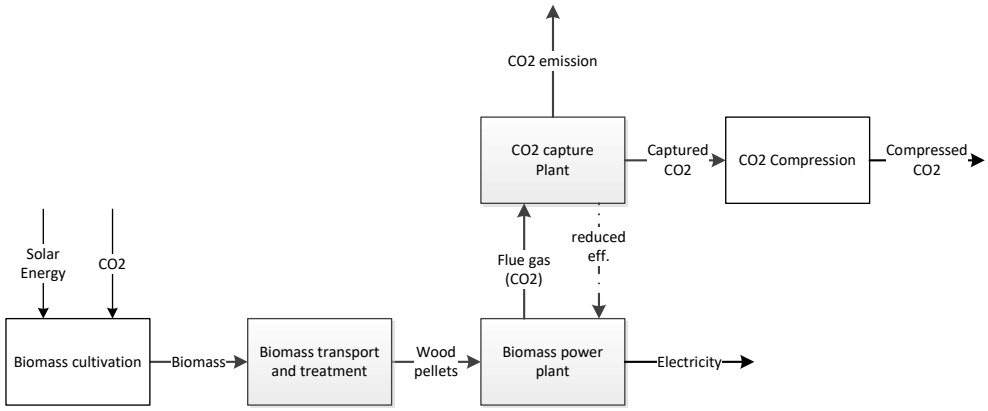


Figure 3.17: Electricity from biomass combustion with post-combustion carbon capture.

CO_2 uptake in biomass cultivation

Data for biomass cultivation are taken from Jungbluth (2008). Jungbluth's data also introduces values for land use and additional environmental extractions and emissions. The most important data provide by Jungbluth are the uptake of 1,78 kg CO_2 and 18,8 MJ of energy per kg of biomass (short rotation wood). These number are comparable to the values provided by Sannigrahi (2010).

Biomass treatment

It is assumed that the short rotation wood is converted into wood chips and is transported further to the power plant. This is process directly taken from the Ecoinvent 2.2 database

for which only the input of wood is changed into short rotation wood as described above. It is assumed that the wood pellets have an energy content of around 18,8 MJ / kg (Jungbluth et al. 2008). Next to that it is assumed that the biomass is taken from a radius of 500 km, transported over 250 km by rail and 250 km by lorry.

Wood pellet combustion and electricity generation

To liberate the energy and CO_2 that has been fixated by biomass (18,8 MJ / kg) it is combusted in a conventional biomass boiler with an efficiency of 78% which produces steam. The steam is subsequently used to generate electricity in steam turbine with an efficiency of 38%. The overall efficiency is therefore 29,6%. It is assumed that the electricity generation can be done in a conventional hard coal power plant as provided in the Ecoinvent database.

The combustion of 1 kg biomass delivers 0,78 x 18,8 MJ = 14,7 MJ of heat and emits 1,78 kg CO_2. To generate 1 MJ of heat we can calculate that 1 / 14,7 = 0,068 kg wood pellets are needed which emit 0,068 * 1,78 = 0,121 kg CO_2 / MJ.

Producing electricity from this heat with an efficiency of 38% shows that for the production of 1 kWh requires and input of 9,47 MJ of heat. Resulting in plant CO_2 emissions of 9,47 * 0,121 kg = 1,14 kg CO_2 / kWh electricity (without carbon capture).

Electricity generation from biomass with carbon dioxide capture

The flue gases containing CO_2 are guided to a post-combustion capture plant using MEA. This system captures 90% of the CO_2 in the flue gases, so that 10% is still emitted to the atmosphere. The capture plant uses energy generated in the power plant which results in a decreased power (electricity) output of 30% (International Energy Agency 2011). This is modelled as follows; a BM power plant without carbon capture uses 9,47 MJ of heat to generate 1 kWh of electricity and emits 1,14 kg CO_2. Connecting a CO_2 capture plant captures 90% of all CO_2 emissions; being 0,9 x 1,14 = 1,03 kg decreases the output of electricity to 0,7 kWh. So where 9,47 MJ generates 1 kWh now 9,47 / 0,7 = 13,53 MJ is required to generate 1 kWh of electricity.

Another important number is the combined generation of 0,7 kWh of electricity and the capture of 1,03 kg CO_2. This means that per kg CO_2, 0,7 / 1,03 = 0,68 kWh electricity is produced. The process data are copied from the carbon capture process connected to a 400 MW coal fired power plant, as modelled by Van der Giesen (2008). The exact size and resources needed for the process depend on the flue gas flow and composition, which for now is assumed to be comparable for burning coal or biomass.

Additional electricity requirements for SF production

As can be expected and seen in Table 3.7 (and in Figure 3.21) the electricity generated by the combustion power plants is not sufficient to convert all captured CO_2 into solar fuels and additional electricity is needed. This modelled by producing and electricity mix existing of electricity produced by the power plants which is supplemented by grid electricity or PV electricity via the ratios in the table below.

Table 3.7: Electricity mix compositions.

	Power plant	Additional	Electricity mix
NG mix	0,35 kWh	0,65 kWh	1 kWh
BM mix	0,09 kWh	0,91 kWh	1 kWh

Direct air capture (DAC)

Capturing CO_2 from the atmosphere, also named (direct) air capture (DAC), is an industrial technology already in use for decades in air revitalization systems on submarines and in space vehicles (Herzog 2003). Air capture is one of the considered options for reducing global atmospheric CO_2 concentrations as a route to manage global warming (Keith 2009; Jones 2011; Olah et al. 2011). While large scale application of the technology is still not feasible there is a variety of proposed routes for air capture (Olah et al. 2011; Goeppert et al. 2012). The best known and common way is by using basic absorbents like calcium-, potassium or sodium hydroxides to bind the CO_2 from the atmosphere and regenerate the formed carbonates to release the CO_2 again. Another option is to replace the hydroxides by carbonates. These bind more weakly to CO_2 making regeneration easier and less energy intensive. Absorption rates are however slower, requiring more time and a higher amount of sorbents to capture the same amount of CO_2 as achieved with hydroxides (Olah et al. 2011). More recent developments focus on using anionic resins that absorb CO_2 from the air when dry and releasing it when exposed to moisture (Lackner 2009). The latter is the least energy intensive and is therefore taken in the present study.

Restrictions for using NaOH CO_2 capture systems concern costs and energy demand. Lackner (2009) identified more reasons why the NaOH might not be the preferred technology to capture CO_2 from the air. Large binding energies between CO_2 and the sorbent requires high energy demands for regeneration, considerable energy demand is connected to moving the solvent around to prevent it from drying and serious concerns exist over the corrosiveness of NaOH solutions. His research shifted towards the use of a solid strong base ion-exchange resin. By using this resin CO_2 from a gas stream even more dilute than in ambient air can be captured and the required energy is far lower than needed in a process using NaOH (Lackner 2009).

Lackner (2009) proposes a specific approach based on a solid sorbent in the form of an anionic exchange resin, that absorbs carbon dioxide when dry and releases it when exposed to moisture, which is known as moisture swing. Lackner demonstrated that simply exposing the resin to water (vapour) would release CO_2 from the resin and that a water temperature of 45°C is sufficient to drive most of the CO_2 off the resin and have it revert to the carbonate state to be used again. Since the resin only comes in contact with water vapour and not the liquid water source, it is possible to use a salt brine as the source of water. The sorbent, which is comparable to Marathon A resin, produced by Dow Chemicals can be regenerated many times without degrading. For now we assume that the resin can be used infinitely and that life cycle impacts are so low, that modelling is not necessary (Lackner 2009).

Lackner (2009)also proposes a collector design. A CO_2 capture device, consisting of wind driven filter system, covered with resin, through which ambient air is led and a CO_2 compressor (to 67 bar) contained in a cargo container (12 m x 2,5m x 3 m) can collect 1 ton of CO_2 per day. Additionally all heat that is needed will be produced as an incidental by-product of the compression. In total 50 kJ / mol CO_2 or 1,1 MJ electrical energy / kg CO_2 is needed (1,1 MJ electrical energy = 0,31 kWh).[34]

When modelling this process in our LCA model we assume the collector itself to consist of steel for the container alone and that additional materials are not needed. The resin is not modelled since it is assumed that it can be used infinitely and will therefore have a very limited impact over the lifecycle of the collector. The air capture process itself only requires the collector and a certain amount of electricity. Water use is neglected since water quality is not an issue as described above.

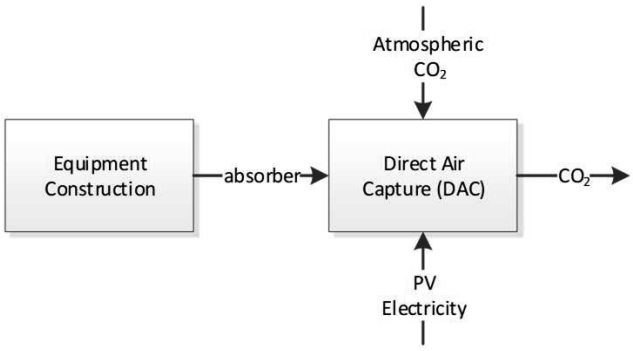


Figure 3.18: LCA unit process for direct air capture (DAC).

The energy supply is modelled in two ways. First the electricity is supplied via CSP, like also done for the NaOH DAC process. Second it is supplied via PV, which seems more logic, and is more efficient, since only electricity (and no heat) is required.

FT fuel combustion

In order to analyse the energy and carbon balances of solar fuel production it is assumed that the solar fuels produced are hypothetically combusted at the gate of the production plant. The outcomes will show how much energy is consumed to produce 1 MJ FT fuel and how many GHG emissions are emitted. The table below provides the relevant fuel properties and assumptions that are used for calculation in this study. For CO_2 emissions theoretical values are calculated based on stoichiometric chemical formulas for combustion. The LHV for FT fuels is an average value calculated from of the LHV values of the possible GTL product slate (Table 3.4).

Table 3.8: Selected (alternative) fuel properties.

Fuel	Formula	CO_2 emission from combustion		LHV		density
		kg CO_2 / kg Fuel	kg CO_2 / MJ fuel	MJ/kg	kg/MJ	kg/m3
Diesel	C13H18	3,11	0,0716	43,4	0,0230	751
Ethanol	C2H6O	1,91	0,0714	26,8	0,0373	789
FT Fuel*	C13H18	3,11	0,0716	43,2	0,0231	816

*Data taken from this study, CO_2 emission are assumed to be comparable to fossil diesel

For the GTL alternative that use fossil based energy and CO_2 (NG), the CO_2 emissions are defined as fossil based. For the bio-based alternatives, the CO_2 emissions are defined as biogenic.

Solar Hydrogen production

Figure 3.19 shows the unit process for generating hydrogen through electrolysis with PV electricity.

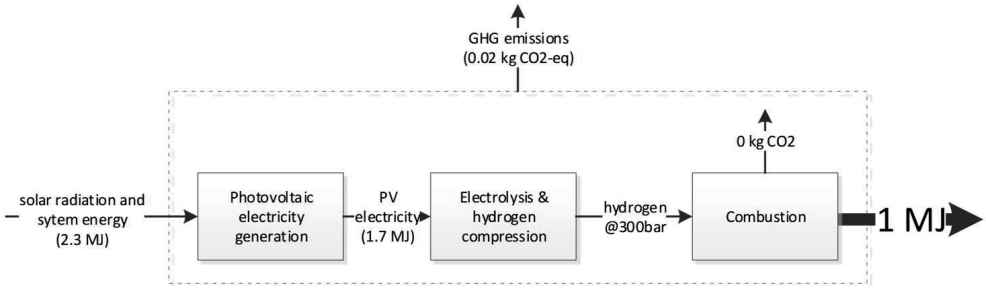


Figure 3.19: LCA unit process for solar hydrogen production and use (including LCA outcomes).

3.5.2. System calculations

Energy and carbon dioxide demand for FT fuel production.

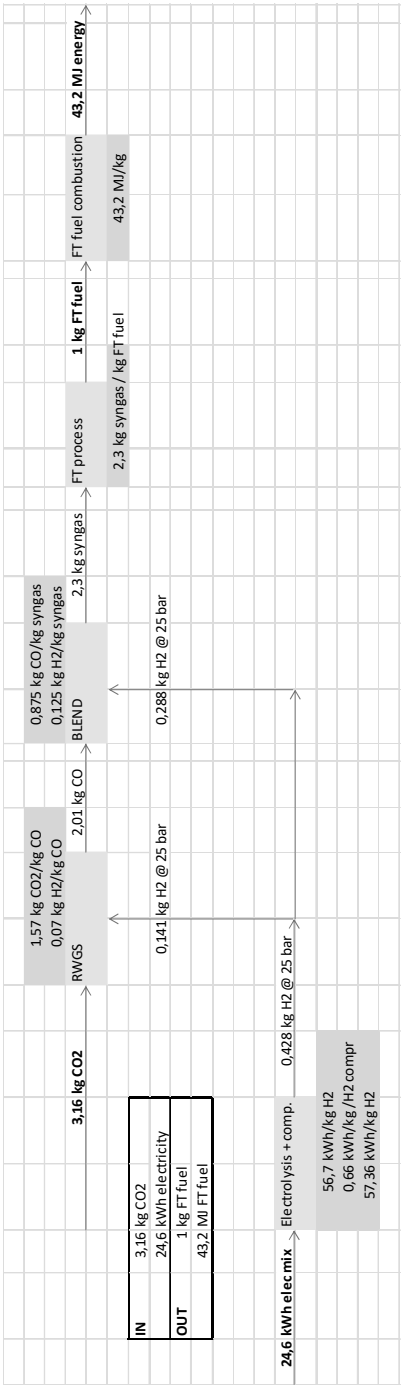


Figure 3.20: Carbon dioxide and electricity demand for FT fuel production.

Electricity mix calculation

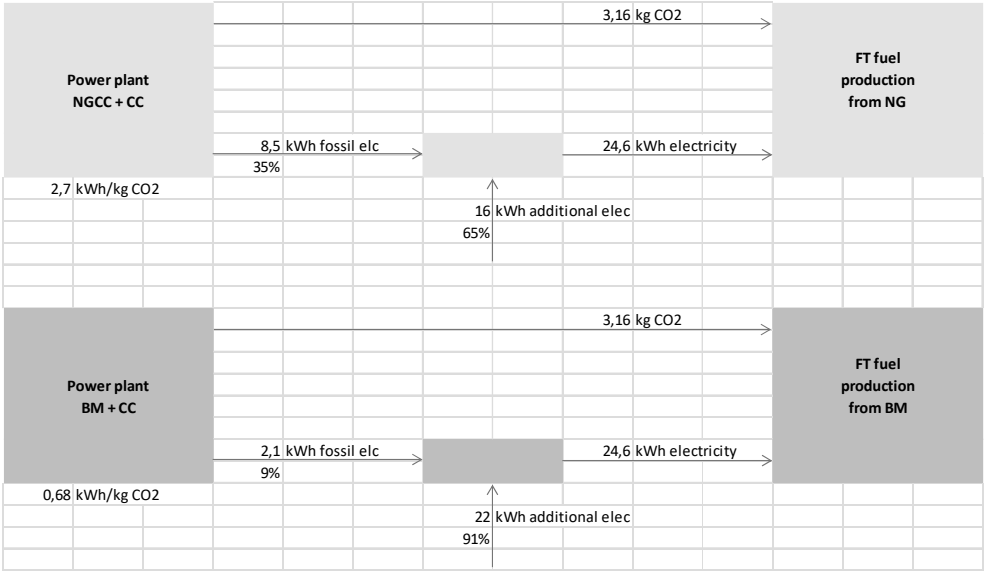


Figure 3.21: Electricity mix calculations (CO_2 demand is leading).

Syngas property calculations

Table 3.9: Blending H_2 and CO to syngas.

		M	mass	
Unit	**mol**	**g/mol**	**g**	**%**
H_2	2	2	4	12,5%
CO	1	28	28	87,5%
Syngas	3	10,7	32	100,0%

Table 3.10: Syngas density calculation.

	0°C/1bar	230°C/25bar
M [kg/mol]	0,0107	0,0107
p [N/m2]	100000	2500000
Z	1	1
R [Nm / (mol K)]	8,314	8,314
T [K]	273	503
density = (M x p) / (Z x R x T)	0,470	6,377

Table 3.11: Syngas LHV calculation.

	mass	LHV
	kg	MJ/kg
H2	0,004	120,1
CO	0,028	10,2
Syngas	0,032	23,9

Additional interpretation of LCA outcomes

Contribution of different greenhouse gases to total GHG emissions

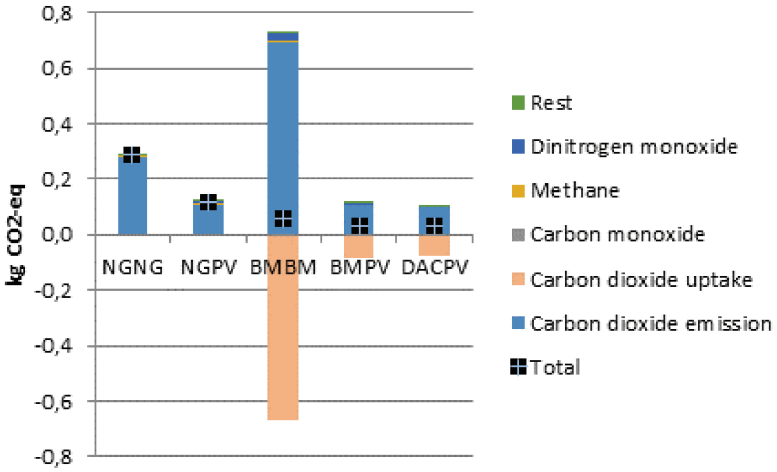


Figure 3.22: Contribution of different greenhouse gasses to net GHG emissions in kg CO_2-eq.

From Figure 3.22 it can be seen that the net GHG emissions mainly exist of carbon dioxide emissions. The uptake of carbon dioxide in the biomass alternative is higher than the uptake from the atmosphere because not all carbon dioxide that is taken up by the biomass is captured and converted into fuel again. Even the best route to produce fuels from carbon dioxide (DACPV) cannot be considered as carbon neutral, because of the emission connected to background processes.

Sensitivity analysis

Because the energy used for capturing carbon dioxide and water electrolysis is the highest contributor to the impacts a sensitivity analysis is performed to see how much a different amount of energy required for these two processes influences the end results. When the electricity demand of the electrolysis process is decreased with 10% from 56.7 kWh / kg to 51kWh / kg the impacts also decrease with 10%, making the energy demand of electrolysis an important flow (Table 3.12).

Table 3.12: Change in GWP and CED resulting from a 10% decrease in electrolysis electricity demand.

	NGNG		NGPV		BMBM		BMPV		DACPV	
	GWP	CED	GWP	CED	GWP	CED	GWP	CED	GWP	CED
Original	0,288	4,98	0,117	3,52	0,0567	7,72	0,0328	3,44	0,0296	2,85
Adjusted	0,26	4,49	0,114	3,25	0,0515	6,98	0,0302	3,16	0,0267	2,58
Change	-11%	-11%	-3%	-8%	-10%	-11%	-9%	-9%	-11%	-10%

Decreasing the CO_2 capture energy demand with 10% results in a change in electricity mix for the combustion alternatives, since more electricity per amount of carbon dioxide becomes available. Table 3.13 shows that the outcomes are hardly influenced, meaning that the energy demand for carbon dioxide capture is not the most important parameter.

Table 3.13: Change in GWP and CED resulting from a 10% decrease in carbon capture electricity demand.

	NGNG		NGPV		BMBM		BMPV		DACPV	
	GWP	CED	GWP	CED	GWP	CED	GWP	CED	GWP	CED
Original	0,288	4,98	0,117	3,52	0,0567	7,72	0,0328	3,44	0,0296	2,85
Adjusted	0,287	4,97	0,117	3,52	0,0552	7,71	0,0314	3,44	0,0294	2,84
Change	0%	0%	0%	0%	-3%	0%	-4%	0%	-1%	0%

Producing liquid fuels from H_2 and CO_2 from SMR

Although not starting directly from using (waste) CO2 as a feedstock, an additional alternative to the fuel production routes in the main article might be to use hydrogen produced by steam methane reforming (SMR). The SMR process is far more efficient than using water electrolysis with renewable electricity but it emits CO_2. It is possible to capture the CO_2 emissions from SMR by using MEA based carbon scrubbing.

SMR takes place in two steps (Sumner 2006). First natural gas (methane, CH4) and steam (water and heat) are fed to a reformer where syngas is formed (1).

$$CH_4 + H_2O + \text{heat} \Rightarrow 3H_2 + CO \qquad (1)$$

It is possible to process this syngas to a Fischer-Tropsch reactor to produce liquid fuels. However gasification of natural gas gives a more beneficial H2 : CO ratio in the syngas to be fed to the FT reactor. This is done in gas-to-liquids (GTL) being more efficient than making syngas through reforming and therefore the latter is not considered here.

The second step in SMR, is passes the resulting stream from the first reaction through a shift reaction where more steam is added and the CO and H_20 react to form more H_2 and CO_2 (2).

$$CO + H_2O \Rightarrow H_2 + CO_2 \qquad (2)$$

This CO_2 can be captured by flue gas scrubbing using MEA.

Sumner (2006) executed an LCA study on hydrogen production via steam methane reforming (SMR) with carbon capture using MEA. His system comprised of SMR unit that produces hydrogen and electricity from NG by methane reforming (see Figure 3.23).

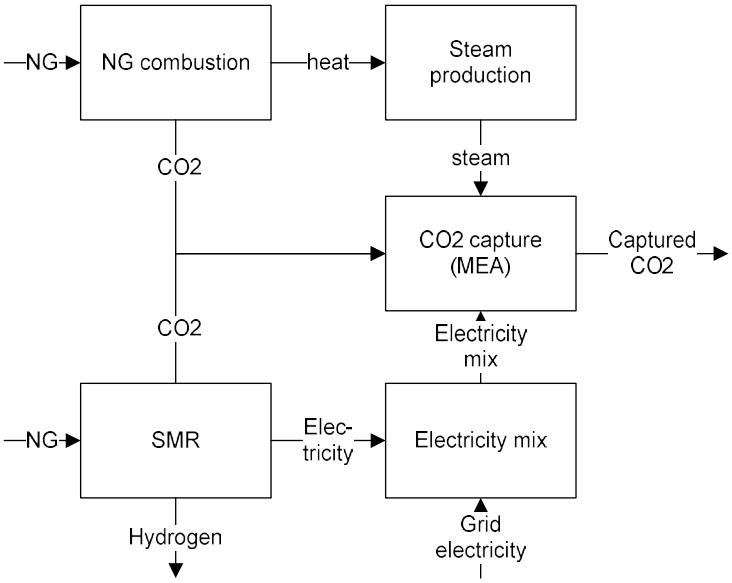


Figure 3.23: System for CO_2 and H_2 from SMR (Sumner, 2006).

Using the hydrogen and CO2 from SMR to produce liquid fuels in the proposed route using RWGS result in 0,14 kg CO_2-eq/ MJ and 3,28 MJ / MJ. Being in the range of all other natural gas based routes presented in the main article.

For detailed LCI information we point to Sumner (2006) and the appendix of this SI (Table 3.36 - Table 3.43).

3.5.3. Life cycle inventory data of processes

Table 3.14: Electrolyser construction.

Economic inflows	Value	Unit
reinforcing steel, at plant[RER]	4,00E+03	kg
disposal, building, fibre board, to final disposal[CH]	51	kg
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	8,00E+03	kg
chromium steel 18/8, at plant[RER]	8,00E+03	kg
transport, freight, rail[RER]	2,55E+03	tkm
nitrogen, liquid, at plant[RER]	85	kg
potassium hydroxide, at regional storage[RER]	30	kg
sodium silicate, spray powder 80%, at plant[RER]	200	kg
corrugated board, recycling fibre, double wall, at plant[RER]	51	kg
disposal, building, reinforcement steel, to sorting plant[CH]	4,00E+03	kg
transport, lorry 16-32t, EURO3[RER]	1,23E+03	tkm
Economic outflows		
Electrolyser for hydrogen production	1	unit
Environmental resources		
Occupation, industrial area, built up[resource_land]	29,8	m2a
Environmental emissions		
Hydrogen[air_unspecified]	5,36	kg

Table 3.15: Hydrogen production, electrolysis.

Economic inflows	Value	Unit
tap water, at user[RER]	11,1	kg
Electrolyser for hydrogen production	2,31E-06	unit
Electricity mix	56,7	kWh
Economic outflows		
Hydrogen gas from electrolysis (NG NG)	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.16: Hydrogen compressor construction.

Economic inflows	Value	Unit
steel, low-alloyed, at plant[RER]	3,00E+03	kg
disposal, building, concrete, not reinforced, to sorting plant[CH]	2,60E+04	kg
aluminium, production mix, at plant[RER]	500	kg
concrete, normal, at plant[CH]	12	m3
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	8,00E+03	kg
chromium steel 18/8, at plant[RER]	5,00E+03	kg
lubricating oil, at plant[RER]	150	kg
transport, freight, rail[RER]	1,79E+03	tkm
disposal, aluminium, 0% water, to municipal incineration[CH]	500	kg
transport, lorry 16-32t, EURO3[RER]	2,16E+03	tkm
Economic outflows		
Hydrogen compressor	1	unit
Environmental resources		
Occupation, industrial area, built up[resource_land]	16,2	m2a
Environmental emissions		
-		

Table 3.17: Hydrogen compression to 25 bar.

Economic inflows	Value	Unit
Hydrogen compressor	3,42E-08	unit
NGNG based electricity mix	0,66	kWh
Hydrogen gas from electrolysis	1	kg
Economic outflows		
Hydrogen gas @ 25 bar	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.18: RWGS catalyst production.

Economic inflows	Value	Unit
copper oxide, at plant[RER]	0,57	kg
aluminium oxide, at plant[RER]	0,11	kg
zinc oxide, at plant[RER]	0,31	kg
Economic outflows		
RWGS catalyst	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.19: Fixed bed reactor construction.

Economic inflows	Value	Unit
steel, low-alloyed, at plant[RER]	4,76E+03	kg
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	9,52E+03	kg
chromium steel 18/8, at plant[RER]	4,76E+03	kg
zeolite, powder, at plant[RER]	2,67E+04	kg
RWGS catalyst	6,60E+03	kg
Economic outflows		
Fixed bed reactor for RWGS	1	unit
Environmental resources		
Occupation, industrial area[resource_land]	3,14	m2a
Environmental emissions		
-		

Table 3.20: CO production via RWGS.

Economic inflows	Value	Unit
Fixed bed reactor for RWGS	1,93E-09	unit
CO2 from NGCC @25 bar	1,57	kg
Hydrogen gas @ 25 bar	0,07	kg
Economic outflows		
Carbon Monoxide from RWGS	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.21: Syngas production.

Economic inflows	**Value**	**Unit**
Hydrogen gas @ 25 bar	0,125	kg
Carbon Monoxide from RWGS	0,875	kg
Economic outflows		
Syngas	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.22: GTL plant construction.

Economic inflows	**Value**	**Unit**
concrete, sole plate and foundation, at plant[CH]	3,45E+05	m3
steel, low-alloyed, at plant[RER]	7,20E+06	kg
reinforcing steel, at plant[RER]	7,20E+06	kg
disposal, building, reinforced concrete, to final disposal[CH]	8,28E+08	kg
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	1,44E+07	kg
Economic outflows		
GTL plant	1	unit
Environmental resources		
Occupation, industrial area[resource_land]	1,12E+06	m2a
Environmental emissions		
x	x	x

Table 3.23: Syngas production.

Economic inflows	**Value**	**Unit**
Syngas	2,3	kg
GTL plant	6,70E-12	unit
Economic outflows		
GTL product	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.24: natural gas, burned in combined cycle plant, best technology, with carbon capture + compression.

Economic inflows	Value	Unit
water, decarbonised, at plant[RER]	0,5	kg
hydrochloric acid, 30% in H2O, at plant[RER]	2,50E-06	kg
sodium hydroxide, 50% in H2O, production mix, at plant[RER]	2,00E-06	kg
natural gas, high pressure, at consumer[RER]	1	MJ
disposal, residue from cooling tower, 30% water, to sanitary landfill[CH]	1,00E-06	kg
gas combined cycle power plant, 400MWe[RER]	2,20E-12	unit
Economic outflows		
natural gas, burned in combined cycle plant, with carbon capture[RER]	1	MJ
Carbon dioxide from NGCC before capture	0,056	kg
Environmental resources		
Heat, waste[air_high population density]	0,525	MJ
Nitrogen oxides[air_high population density]	2,55E-05	kg
Particulates, < 2.5 um[air_high population density]	5,00E-07	kg
Carbon monoxide, fossil[air_high population density]	2,20E-06	kg
Dinitrogen monoxide[air_high population density]	1,00E-06	kg
Acetic acid[air_high population density]	1,21E-07	kg
Methane, fossil[air_high population density]	1,00E-06	kg
Sulfur dioxide[air_high population density]	5,00E-07	kg
Toluene[air_high population density]	1,50E-09	kg
Acetaldehyde[air_high population density]	8,00E-10	kg
Propane[air_high population density]	7,05E-07	kg
Propionic acid[air_high population density]	1,60E-08	kg
Formaldehyde[air_high population density]	3,31E-08	kg
Benzene[air_high population density]	9,26E-10	kg
Benzo(a)pyrene[air_high population density]	5,29E-13	kg
Dioxins, measured as 2,3,7,8-tetrachlorodibenzo-p-dioxin [air_high population density]	2,90E-17	kg
Mercury[air_high population density]	3,00E-11	kg
PAH, polycyclic aromatic hydrocarbons[air_high population density]	8,00E-09	kg
Butane[air_high population density]	9,26E-07	kg
Pentane[air_high population density]	1,15E-06	kg
Ethane[air_high population density]	1,37E-06	kg
Hexane[air_high population density]	7,93E-07	kg
Acenaphthene[air_high population density]	7,93E-13	kg

Table 3.25: electricity, natural gas, at combined cycle plant, best technology, with carbon capture and compression.

Economic inflows	Value	Unit
natural gas, burned in combined cycle plant, with carbon capture	7,36	MJ
Economic outflows		
electricity, natural gas, at combined cycle plant, with carbon capture	1	kWh
Environmental resources		
-		
Environmental emissions		
-		

Table 3.26: Carbon capture + compression to 25 bar.

Economic inflows	Value	Unit
tap water, at user[RER]	216	kg
monoethanolamine, at plant[RER]	1,6	kg
Carbon dioxide from NGCC before capture	1,11E+03	kg
CO_2 capture + compression plant for NGCC power plant	3,30E-08	unit
Economic outflows		
CO_2 from NGCC @25 bar	1,00E+03	kg
Environmental resources		
-		
Environmental emissions		
Heat, waste[air_low population density]	855	MJ
Carbon dioxide, fossil[air_high population density]	111	kg
Heat, waste[water_unspecified]	3,41E+03	MJ

Table 3.27: CO_2 capture plant + compressor construction.

Economic inflows	Value	Unit
steel, low-alloyed, at plant[RER]	3,91E+06	kg
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	4,86E+06	kg
chromium steel 18/8, at plant[RER]	9,48E+05	kg
Economic outflows		
CO_2 capture + compression plant for NGCC power plant	1	unit
Environmental resources		
Transformation, from unknown[resource_land]	4,00E+04	m2
Occupation, industrial area[resource_land]	1,44E+06	m2a
Transformation, to industrial area[resource_land]	4,00E+04	m2
Environmental emissions		
-		

Table 3.28: wood chips, burned in power plant with carbon capture.

Economic inflows	Value	Unit
transport, lorry >16t, fleet average[RER]	0,00013	tkm
transport, freight, rail[RER]	0,000351	tkm
water, decarbonised, at plant[RER]	0,15	kg
chlorine, liquid, production mix, at plant[RER]	1,00E-05	kg
disposal, residue from cooling tower, 30% water, to sanitary landfill[CH]	5,00E-06	kg
light fuel oil, at regional storage[RER]	1,70E-05	kg
water, completely softened, at plant[RER]	0,006	kg
hard coal power plant[RER]	1,30E-12	unit
SOx retained, in hard coal flue gas desulphurisation[RER]	0,000118	kg
NOx retained, in SCR[GLO]	3,09E-05	kg
disposal, hard coal ash, 0% water, to residual material landfill[ES]	0,00651	kg
Wood chips from short-rotation wood	0,068	kg
Economic outflows		
wood chips, burned in power plant with carbon capture[ES]	1	MJ
Carbon dioxide from biomass combustion before capture	0,121	kg
Environmental resources		
Water, cooling, unspecified natural origin[resource_in water]	0,0035	m3
Environmental emissions		
Heat, waste[air_low population density]	0,548	MJ
Dinitrogen monoxide[air_low population density]	8,90E-07	kg
Nitrogen oxides[air_low population density]	0,000362	kg
Carbon monoxide, fossil[air_low population density]	8,00E-06	kg
Sulfur dioxide[air_low population density]	0,000731	kg
Methane, fossil[air_low population density]	1,00E-06	kg
Benzene[air_low population density]	2,17E-07	kg
Particulates, < 2.5 um[air_low population density]	4,85E-05	kg
Cadmium[air_low population density]	4,47E-10	kg
Chromium[air_low population density]	4,23E-09	kg
Copper[air_low population density]	6,07E-09	kg
Nickel[air_low population density]	1,19E-08	kg
Zinc[air_low population density]	4,13E-08	kg
Benzo(a)pyrene[air_low population density]	2,00E-13	kg
PAH, polycyclic aromatic hydrocarbons[air_low population density]	1,00E-09	kg
Selenium[air_low population density]	7,95E-09	kg
Lead[air_low population density]	2,64E-08	kg
Particulates, > 10 um[air_low population density]	7,85E-06	kg

Hydrogen fluoride[air_low population density]	4,23E-06	kg
Particulates, > 2.5 um, and < 10um[air_low population density]	5,70E-06	kg
Uranium-238[air_low population density]	1,57E-05	kBq
Thorium-228[air_low population density]	3,10E-06	kBq
Radium-226[air_low population density]	1,88E-05	kBq
Radon-222[air_low population density]	0,000582	kBq
Lead-210[air_low population density]	7,29E-05	kBq
Polonium-210[air_low population density]	0,000133	kBq
Potassium-40[air_low population density]	1,43E-05	kBq
Thorium-232[air_low population density]	4,87E-06	kBq
Heat, waste[water_river]	0,144	MJ
Antimony[air_low population density]	5,36E-10	kg
Arsenic[air_low population density]	7,63E-09	kg
Barium[air_low population density]	5,10E-08	kg
Boron[air_low population density]	6,51E-07	kg
Bromine[air_low population density]	2,94E-07	kg
Butane[air_low population density]	1,90E-08	kg
Chromium VI[air_low population density]	5,23E-10	kg
Cobalt[air_low population density]	2,18E-09	kg
Dioxins, measured as 2,3,7,8-tetrachlorodibenzo-p-dioxin[air_low population density]	7,00E-15	kg
Ethane[air_low population density]	4,10E-08	kg
Formaldehyde[air_low population density]	5,80E-08	kg
Hydrocarbons, aliphatic, alkanes, unspecified[air_low population density]	2,19E-07	kg
Hydrocarbons, aliphatic, unsaturated[air_low population density]	2,16E-07	kg
Hydrogen chloride[air_low population density]	1,31E-05	kg
Iodine[air_low population density]	1,35E-07	kg
Manganese[air_low population density]	1,33E-08	kg
Mercury[air_low population density]	5,16E-09	kg
Molybdenum[air_low population density]	8,72E-10	kg
Pentane[air_low population density]	1,47E-07	kg
Propane[air_low population density]	3,50E-08	kg
Propene[air_low population density]	1,60E-08	kg
Radium-228[air_low population density]	5,76E-06	kBq
Radon-220[air_low population density]	0,000327	kBq
Strontium[air_low population density]	7,04E-08	kg
Toluene[air_low population density]	1,09E-07	kg
Vanadium[air_low population density]	1,24E-08	kg
Xylene[air_low population density]	9,22E-07	kg

Table 3.29: electricity, wood pellets, power plant with carbon capture.

Economic inflows	Value	Unit
wood chips, burned in power plant with carbon capture[ES]	13,5	MJ
Economic outflows		
electricity, wood chips at power plant with carbon capture	1	kWh
Environmental resources		
-		
Environmental emissions		
-		

Table 3.30: CO_2 capture plant for biomass power plant.

Economic inflows	Value	Unit
steel, low-alloyed, at plant[RER]	6,11E+06	kg
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	7,58E+06	kg
chromium steel 18/8, at plant[RER]	1,47E+06	kg
Economic outflows		
CO_2 capture plant for Biomass power plant	1	unit
Environmental resources		
Transformation, from unknown[resource_land]	4,20E+04	m2
Occupation, industrial area[resource_land]	1,44E+06	m2a
Transformation, to industrial area[resource_land]	4,20E+04	m2
Environmental emissions		
-		

Table 3.31: Carbon capture + compression from biomass power plant.

Economic inflows	Value	Unit
tap water, at user[RER]	262	kg
monoethanolamine, at plant[RER]	1,6	kg
Carbon dioxide from biomass combustion before capture	1,11E+03	kg
CO_2 capture plant for Biomass power plant	1,24E-08	unit
Economic outflows		
CO_2 from BM @25 bar	1,00E+03	kg
Environmental resources		
-		
Environmental emissions		
Heat, waste[air_low population density]	744	MJ
Carbon dioxide, fossil[air_high population density]	111	kg
Heat, waste[water_unspecified]	4,25E+03	MJ

Table 3.32: Direct air capture (DAC) equipment.

Economic inflows	Value	Unit
steel, low-alloyed, at plant[RER]	3,80E+03	kg
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	3,80E+03	kg
Economic outflows		
DAC equipment (absorber)	1	unit
Environmental resources		
Occupation, industrial area[resource_land]	36	m2a
Environmental emissions		
-		

Table 3.33: Direct air capture.

Economic inflows	Value	Unit
electricity, production mix photovoltaic, at plant[ES]	0,31	kWh
DAC equipment (absorber)	9,13E-08	unit
Economic outflows		
CO_2 from direct air capture @25 bar	1	kg
Environmental resources		
Carbon dioxide, in air[resource_in air]	1	kg
Environmental emissions		
-		

Table 3.34: Electricity mix.

Economic inflows	Value	Unit
electricity, source A	0,65	kWh
electricity, source B	0,35	kWh
Economic outflows		
electricity mix	1	kWh
Environmental resources		
-		
Environmental emissions		
-		

Table 3.35: SMR construction.

Economic inflows	Value	Unit
steel, low-alloyed, at plant[RER]	3.01E6	kg
disposal, building, concrete, not reinforced, to sorting plant[CH]	9.42E6	kg
concrete, normal, at plant[CH]	4.28E3	m3
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	3.05E6	kg
transport, freight, rail[RER]	6.15E5	tkm
aluminium, primary, at plant[RER]	2.48E4	kg
pig iron, at plant[GLO]	3.68E4	kg
disposal, aluminium, 0% water, to municipal incineration[CH]	2.48E4	kg
transport, lorry >32t, EURO4[RER]	7.79E5	tkm
Economic outflows		
Steam methane reformer	1	unit
Environmental resources		
Occupation, industrial area[resource_land]	2.25E4	m2a
Environmental emissions		
-		

Table 3.36: Steam methane Reforming.

Economic inflows	Value	Unit
tap water, at user[RER]	10.9	kg
natural gas, high pressure, at consumer[RER]	183	MJ
Steam methane reformer	9.21E-10	unit
Economic outflows		
CO_2 to amine fluegas scrubbing	9.23	kg
Hydrogen gas from SMR + CC	1	kg
Electricity, med V, from SMR + CC	2.98	kWh
Environmental resources		
Water, cooling, unspecified natural origin[resource_in water]	0.0286	m3
Environmental emissions		
Carbon dioxide, fossil[air_low population density]	1.02	kg
Nitrogen oxides[air_low population density]	0.00475	kg
Particulates, < 2.5 um[air_unspecified]	2.2E-5	kg
Carbon monoxide, biogenic[air_low population density]	7.98E-5	kg

Table 3.37: Natural gas, burned in industrial furnace >100kW, with carbon capture.

Economic inflows	Value	Unit
electricity, low voltage, production UCTE, at grid[UCTE]	0.00111	kWh
natural gas, high pressure, at consumer[RER]	1	MJ
industrial furnace, natural gas[RER]	2.8E-9	unit
Economic outflows		
CO_2 to amine fluegas scrubbing	0.0504	kg
Natural gas, burned in industrial furnace >100kW, with carbon capture[RER]	1	MJ
Environmental resources		
-		
Environmental emissions		
Carbon dioxide, fossil[air_low population density]	1.02	kg
Nitrogen oxides[air_low population density]	0.00475	kg
Heat, waste[air_high population density]	1.1	MJ
Nitrogen oxides[air_high population density]	1.79E-5	kg
Particulates, < 2.5 um[air_high population density]	2,00E-07	kg
Carbon monoxide, fossil[air_high population density]	2.1E-6	kg
Carbon dioxide, fossil[air_high population density]	0.0056	kg
Dinitrogen monoxide[air_high population density]	1,00E-07	kg
Acetic acid[air_high population density]	1.5E-7	kg
Methane, fossil[air_high population density]	2,00E-06	kg
Sulfur dioxide[air_high population density]	5.5E-7	kg
Toluene[air_high population density]	2,00E-07	kg
Acetaldehyde[air_high population density]	1,00E-09	kg
Propane[air_high population density]	2,00E-07	kg
Propionic acid[air_high population density]	2,00E-08	kg
Formaldehyde[air_high population density]	1,00E-07	kg
Benzene[air_high population density]	4,00E-07	kg
Benzo(a)pyrene[air_high population density]	1,00E-11	kg
Dioxins, measured as 2,3,7,8-tetrachlorodibenzo-p-dioxin[air_high population density]	3,00E-17	kg
Mercury[air_high population density]	3,00E-11	kg
PAH, polycyclic aromatic hydrocarbons[air_high population density]	1,00E-08	kg
Butane[air_high population density]	7,00E-07	kg
Pentane[air_high population density]	1.2E-6	kg

Table 3.38: Steam production from NG for carbon capture.

Economic inflows	**Value**	**Unit**
tap water, at user[RER]	1.17	kg
Natural gas, burned in industrial furnace >100kW, with carbon capture[RER]	3.23	MJ
Economic outflows		
Steam for carbon capture	1	kg
Environmental resources		
-		
Environmental emissions		
-		

Table 3.39: Steam production from NG for carbon capture.

Economic inflows	**Value**	**Unit**
steel, low-alloyed, at plant[RER]	9.02E8	kg
bitumen, at refinery[CH]	1.1E6	kg
disposal, building, concrete, not reinforced, to sorting plant[CH]	5.96E6	kg
concrete, normal, at plant[CH]	2.71E3	m3
disposal, building, bulk iron (excluding reinforcement), to sorting plant[CH]	9.18E5	kg
disposal, bitumen, 1.4% water, to sanitary landfill[CH]	1.1E6	kg
transport, freight, rail[RER]	8.44E5	tkm
transport, lorry >32t, EURO4[RER]	5,00E+05	tkm
Economic outflows		
Carbon scrubber	1	unit
Environmental resources		
-		
Environmental emissions		
-		

Table 3.40: Carbon scrubbing from SMR and steam generation.

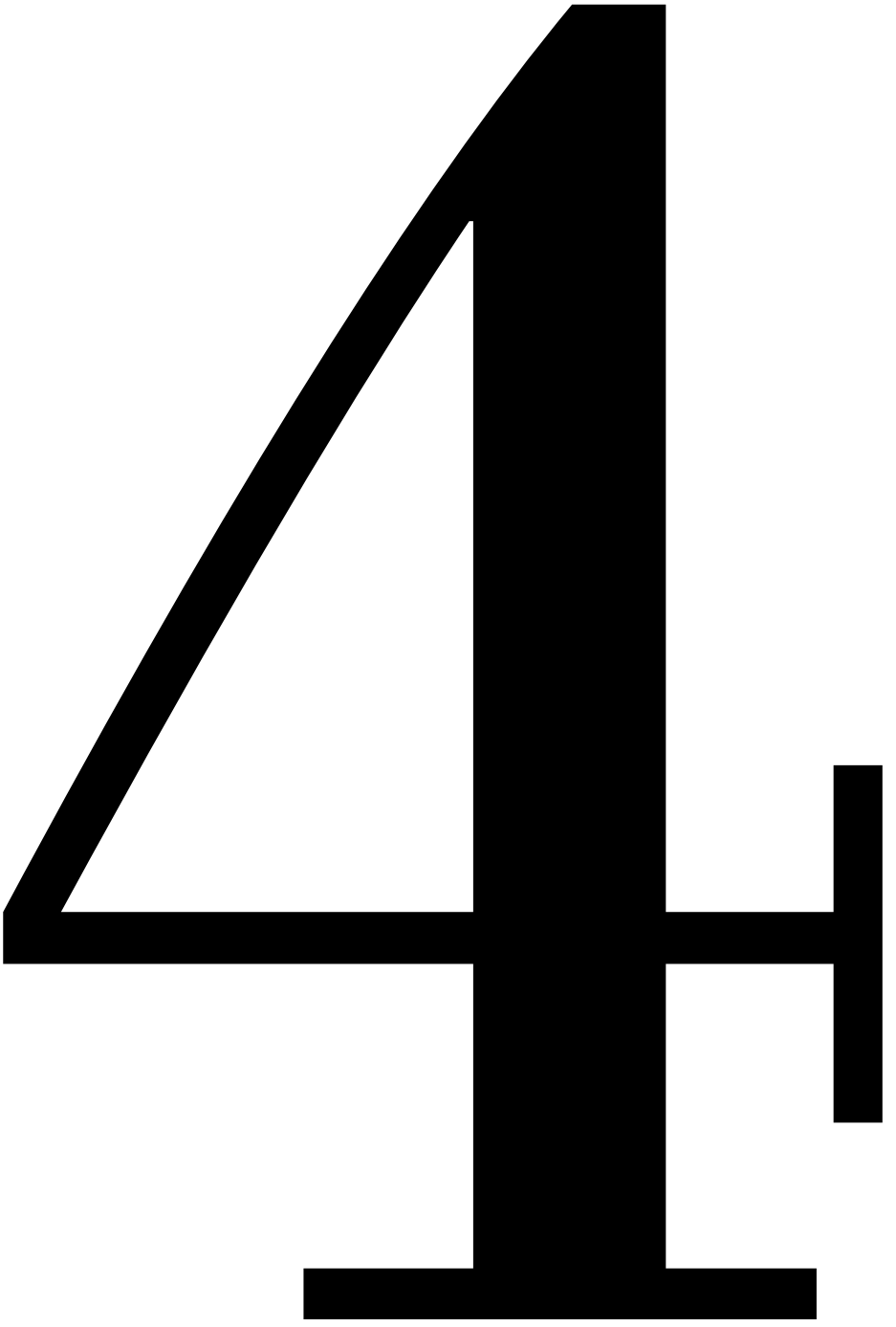
Economic inflows	Value	Unit
tap water, at user[RER]	2.49	kg
monoethanolamine, at plant[RER]	0.002	kg
CO2 to amine fluegas scrubbing	1	kg
Steam for carbon capture	1.39	kg
Hydrogen compressor (James)	4.23E-7	unit
Carbon scrubber	1.16E-10	unit
mixed electricity at SMR plant	0.132	kWh
Economic outflows		
CO2 from SMR and Steam gen	1	kg
Environmental resources		
-		
Environmental emissions		
Monoethanolamine[air_high population density]	0.001	kg

Table 3.41: Electricity mixing for Carbon scrubbing (SMR).

Economic inflows	Value	Unit
electricity, medium voltage, production UCTE, at grid[UCTE]	0.525	kWh
Electricity, med V, from SMR + CC	0.475	kWh
Economic outflows		
mixed electricity at SMR plant	1	kWh
Environmental resources		
-		
Environmental emissions		
-		

Table 3.42: Hydrogen (SMR +CC) compression.

Economic inflows	Value	Unit
Hydrogen gas from SMR + CC	1	kg
Hydrogen compressor (James)	4.23E-7	unit
mixed electricity at SMR plant	0.223	kWh
Economic outflows		
Hydrogen (SMR+CC) @ 25 bar	1	kg
Environmental resources		
-		
Environmental emissions		
-		



Chapter 4

A Life Cycle Assessment Case Study of Coal-fired Electricity Generation with Humidity Swing Direct Air Capture of CO_2 versus MEA-based Post-combustion Capture

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Abstract

Most carbon capture and storage (CCS) envisions capturing CO_2 from flue gas. Direct air capture (DAC) of CO_2 has hitherto been deemed unviable because of the higher energy associated with capture at low atmospheric concentrations. We present a Life Cycle Assessment of coal-fired electricity generation that compares mono-ethanolamine (MEA) based post-combustion-capture (PCC) of CO_2 with distributed, humidity-swing-based DAC (HS-DAC). Given suitable temperature, humidity, wind, and water availability, HS-DAC can be largely passive. Comparing energy requirements of HS-DAC and MEA-PCC, we find the parasitic load of HS-DAC is less than twice that of MEA-PCC (60-72 kJ/mol versus 33-46 kJ/mol, respectively). We also compare other environmental impacts as a function of net greenhouse gas (GHG) mitigation: To achieve the same 73% mitigation as MEA-PCC, HS-DAC would increase 9 other environmental impacts by on average 38%, whereas MEA-PCC would increase them by 31%. Powering distributed HS-DAC with photovoltaics (instead of coal) while including re-capture of all background GHG, reduces this increase to 18%, hypothetically enabling coal-based electricity with net-zero life-cycle GHG. We conclude that, in suitable geographies, HS-DAC can complement MEA-PCC to enable CO_2 capture independent of time and location of emissions and re-capture background GHG from fossil-based electricity beyond flue stack emissions.

4.1 Introduction

To keep global warming below 1.5-2°C targets, a balanced mix of greenhouse gas (GHG) mitigation measures is needed. Among these, carbon capture and storage (CCS) is projected to play an important role, as are increasing energy efficiency and deployment of renewable and nuclear power.(International Energy Agency 2012, 2008; Fuss et al. 2014) Besides traditional carbon capture from point sources, additional research into a wider portfolio of carbon dioxide removal (CDR) technologies, including distributed solutions, is seen as more and more important.(National Research Council 2015; Williamson 2016; Lackner et al. 2012) We henceforth refer to all such capture and storage options as CCS.

Relative (dis-) advantages of different CCS technologies

CCS is not tied to a specific technology. For coal-fired electricity generation, arguably the most mature technology is post combustion capture (PCC) using mono-ethanolamine (MEA), which scrubs CO_2 directly from the flue gas of fossil fuel power plants.(Rochelle et al. 2011; Abu Zahra 2009; Leung et al. 2014) It is often argued that there are no technological obstacles to large-scale implementation of PCC; rather that its main hurdle is a lack of policy or market mechanisms to incentivize adopting PCC amidst several challenges:(International Energy Agency 2012; Leung et al. 2014) The power plant must be located in sufficient vicinity of a suitable storage site, or else CO_2 transport from the plant to such a site will become prohibitively expensive. PCC results in reducing operating efficiency compared to a conventional power plant. It can best be implemented in modern coal-fired power plants, i.e. fewer than 29% of currently installed coal-fired power plants globally.(International Energy Agency 2012; Leung et al. 2014) Further, as an optimum design choice, MEA-PCC typically captures 85 - 90% of the flue stack CO_2 emissions (Reiter and Lindorfer 2015) but none of the so-called background emissions (e.g., from mining and transporting coal, or building and decommissioning the plant itself). MEA-PCC thus typically eliminates only ~70% of total life cycle greenhouse gas (GHG) emissions of coal-based electricity (Corsten et al. 2013; Sathre et al. 2012). Finally, many power plants are located near population centres, a fact that can be problematic for transport and storage of CO_2. The IEA notes that "examples in Germany and The Netherlands illustrate that under-appreciation of public concerns over CO_2 storage can easily be fatal for CCS".(International Energy Agency 2012)

CO_2 does not necessarily have to be captured at the point of combustion. A CCS alternative to PCC is Direct Air Capture (DAC), a form of CDR that captures CO_2 directly from the atmosphere.(Tavoni and Socolow 2013; Lackner et al. 2012; Keith 2009) Relative dis-advantages and advantages of DAC versus PCC have been previously discussed (American Physical Society 2011; Reiter and Lindorfer 2015). One possible

advantage of DAC is that its location and schedule of operation is not necessarily tied to the emitting power plant itself. This flexibility imbues the technology with benefits (Boot-Handford et al. 2014; American Physical Society 2011; Wang et al. 2011) that may partially alleviate above limitations of PCC-based CCS: With regard to location, DAC plants can be built at or near the final CO_2 storage site, so that CO_2 transport infrastructure can be largely avoided. This could alleviate social opposition arising from CO_2 transport through populated areas (The Royal Society 2009). And because its unit size and operating schedule is not tied to the power plant itself, DAC could be scaled to capture not 90% but 100% of CO_2 emissions emitted from a plant and in addition compensate for background GHG emissions, thus allowing for true, net-zero GHG electricity from coal. Similarly, DAC could compensate for emissions from non-stationary GHG sources in sectors where costs or technological feasibility may prohibit phasing out fossil fuels (e.g. aviation). The Royal Society notes that 'there is surprising value in the economic freedom to build a capture plant where it is cheapest to do so and near the best sequestration site' (The Royal Society 2009).

Above theoretical advantages notwithstanding, capturing low concentrations of CO_2 from air simply is thermodynamically more challenging than from concentrated point sources (American Physical Society 2011). In a recent, comprehensive review of DAC and other CDR, DAC is expected to consume up to 45GJ per ton carbon equivalent (540kJ/mol CO_2) – levels that would arguably render DAC economically unviable (Smith et al. 2016). Whether as part of a portfolio of climate responses (National Research Council 2015) or as a last-resort backstop technology to reduce atmospheric CO_2 levels via negative emissions (Pielke 2009), improved understanding of DAC's energy requirement and environmental impacts is therefore crucial.

Humidity-swing DAC (HS-DAC)

Previous research on DAC has focused on gas scrubbing technologies using sorbents such as $CaOH_2$, KOH and NaOH (American Physical Society 2011; Zenz House et al. 2011; Baciocchi et al. 2006). The binding potential of these is relatively high, and therefore large amounts of high-exergy energy (electricity and/or high grade heat) are required to release the CO_2 during sorbent regeneration (Zenz House et al. 2011; Goeppert et al. 2012). In contrast, HS-DAC utilizes ambient-temperature heat for the sorbent regeneration: It is realized through water drying off the sorbent, i.e. spontaneously evaporating at ambient conditions (driven by the surrounding air and, if available, sun shine).(Lackner 2013) This is akin to other DAC technologies, such as temperature-swing DAC, which can use low grade heat for cycling the sorbent material and whose energy balance analysis thus also shows limited requirement for mechanical (or electrical) energy (Wurzbacher et al. 2011, 2012). However, for HS-

DAC, the relatively low energy consumption of the overall capture process is in part achieved through reliance on ambient wind conditions to drive air through filters and later dry them for regeneration. This causes one of HS-DAC's main disadvantages: Its performance becomes weather/geography dependent (Goldberg et al. 2013). In this study, we quantify this dependence via sensitivity analyses.

Previously, relative (dis-)advantages of HS-DAC have been analysed, (Lackner et al. 2012) current understanding of its molecular process (Wang et al. 2013) and thermodynamics (Lackner 2013) described, and its energy consumption has been estimated at 50kJ/mol (Lackner 2009). Briefly, a non-toxic anionic exchange resin such as the commercially available MarathonA adsorbs CO_2 when dry and releases it when exposed to 100% humidity. Driven by ~2m/sec or higher ambient wind, air flows through filters in which the resin adsorbs some CO_2, reducing the air's CO_2 concentration from 400ppm to ~360 ppm (design-dependent). Using an electrically powered conveyor system, the filters enter a desorption chamber. After evacuating to 0,01 bar the sorbent is sprayed with water, causing the sorbent to release CO_2. The resulting low-pressure gas mix in the desorption chamber (CO_2 and H_2O, <=1% N2 and O2) is pumped through a cold trap (operating at variable temperature down to -10°C; see SI) where the H_2O in the gas mix condenses and is removed. The CO_2 is compressed to 100bar for underground injection. The filters are moved from the desorption chamber back outside where the remaining water dries off, thus regenerating filters for a new cycle.

Note that HS-DAC is unique primarily in the mechanism that cycles the sorbent's equilibrium binding characteristics back and forth between favouring adsorption and desorption. Following desorption into a vacuum, HS-DAC is similar to temperature-swing DAC. Both technologies have to include a step to remove the water vapour that comes off the sorbent along with the CO_2 from the desorption stream. Either, water vapour in the desorption stream is condensed out via cooling (Wurzbacher et al. 2011), followed by compression of CO_2 to the desired final pressure (typically 100 bar). Or the water vapour and the CO_2 are compressed concurrently such that the water gradually condenses as pressure increases, leaving mostly CO_2 (Lackner 2009). Different implementations of this CO_2 purification step are possible, including having a CO_2-enriched humid stream of air from a DAC machine purified via a conventional PCC-type, e.g. amine-based process. Different implementations will result in different water and energy consumptions. In this work, we assume a simple evacuation and drying via cooling implementation (Wurzbacher et al. 2011), which avoids the need for any specialized or costly hardware components. While not necessarily the most energy-efficient, the chosen implementation offers the advantage that its energy requirement can be robustly quantified as previously described (Lackner 2009). Here, we refine these

previous calculations based on more recent laboratory experiments on the sorbent's temperature and humidity dependence of the binding characteristics (Wang et al. 2011) as well its binding kinetics (Wang et al. 2016). This allows us to adjust the yield of the sorbent in response to varying weather conditions and thus quantify HS-DAC's sensitivity to suitable climate/geographies (see section 4.3).

Energy balance and life cycle environmental impacts

The goal of any CCS technology is to reduce anthropogenic CO_2 emissions. However, it is important to ensure that no prohibitive problem shifting occurs, because any form of CCS will cause increased energy and material use, with their own associated environmental impacts beyond GHG. A key aspect here is the previously identified inherent disadvantage of DAC over PCC which results from the more dilute CO_2 source and thus a priori higher parasitic energy losses: This underlines the importance of detailed energy balance studies and their system level effects on energy use per net GHG avoided (rather than per CO_2 captured) (American Physical Society 2011). While HS-DAC differs from PCC in other aspects as well, including hardware, consumable materials, and logistics of CO_2 transport, such background processes have much less impacts on the life cycle carbon balance and other environmental impacts (Results).

While detailed lifecycle assessments are available for PCC (Corsten et al. 2013) and environmental risks and constraints of various CDR technologies have been widely discussed (Williamson 2016; Smith et al. 2016; Reiter and Lindorfer 2015; von der Assen et al. 2016), a detailed life cycle assessment of HS-DAC has not been available to date. Thus, assessments of HS-DAC's relative performance vis-a-vis PCC are incomplete. In this work, we therefore perform a life cycle assessment (LCA) on HS-DAC to answer the following question: How do the lifecycle environmental impacts of HS-DAC compare with those of MEA-PCC when applied to CCS for coal-fired power generation?

4.2 Method and approach

LCA of emerging technologies

We used LCA to calculate environmental impacts, using CMLCA software (Institute of Environmental Sciences) with the ecoinvent 2.2 database (Swiss Centre for Life Cycle Inventories 2010) and the 10 CML-IA version 4.5 (baseline) impact characterization factors (CML-Department of Industrial Ecology 2015) recommended by the Handbook of LCA (Guinée et al. 2002) and commonly used in LCA of electricity generation (Koornneef et al. 2008; Corsten et al. 2013). The LCA thus covers climate change (measured as CO_2-eq on a 100a horizon, henceforth "GHG emissions"), eutrophication, resource depletion, acidification, summer smog, terrestrial ecotoxicity, marine

ecotoxicity, freshwater ecotoxicity, ozone depletion, and human toxicity. Additionally, "water depletion" was assessed via the RECIPE method (Goedkoop et al. 2009).

Performing an LCA on emerging technologies such as HS-DAC is challenging since empirical data other than laboratory implementations of the technology are lacking and therefore uncertainty is introduced where assumptions and predictions of real-world implementations need to be used (Frischknecht et al. 2009; Hetherington et al. 2014; Hospido et al. 2010; Miller and Keoleian 2015; Wender et al. 2014b). However, since energy demand is often responsible for the largest share of the environmental impacts in LCA studies (Huijbregts et al. 2010; Sugiyama et al. 2008; Patel et al. 2012), energy demand is considered an important focal point in LCAs of new technologies(Patel et al. 2012; von der Assen et al. 2016) and a key factor for the costs and environmental impacts of CO_2 capture (von der Assen et al. 2016).

In this study, HS-DAC's and MEA-PCC's energy demand dominate their respective LCAs as well (see section 4.3). We therefore place particular emphasis on the robustness of the energy demand in the inventory data: Energy demand per amount of CO_2 captured is based on technical engineering calculations parameterized by previously published laboratory experiments (HS-DAC) and by pilot capture installations (MEA-PCC). Other life cycle inventory (LCI) data such as sorbent and hardware consumption and estimated machine size are based on laboratory measurements such as sorbent yield per cycle and lifetime and calculations of required hardware components such as pumps and compressors. Sensitivity analyses of LCA results to these input data are provided.

In contrast to MEA-PCC, no large-scale pilot of HS-DAC is yet available. We therefore report its energy and hardware requirements to allow for critical review and insights into HS-DAC's technical feasibility. This also allows a methodological foundation for future studies that will have the benefit of more detailed engineering designs. Our results include sensitivity analyses to gauge the effect of uncertainties in all PCC and HS-DAC inventory data on their respective LCAs.

System boundaries and functional unit

The LCA covers impacts of all materials and processes in the power generation and CCS system up to providing 100 bar CO_2 at injection sites. This includes mining, transporting, and combusting coal, building the power plant, transmission, and the CO_2 capture and compression units, photovoltaic (PV) panels, as well as hardware decommissioning and material disposal. Only preparing the injection sites themselves is not included because the infrastructure requirements are the same for PCC and DAC and, in any case, have been shown to be of minor impact in CCS LCA studies (Koornneef et al. 2008; Modahl et al. 2012; Singh et al. 2011).

We devised two alternative CCS systems that both provide low-carbon electricity from coal to the grid (functional unit = 1 kWh for end-use (at desired amount of net GHG emissions), i.e. not counting electricity output used for capture). In the MEA-PCC scenarios (Figure 4.1) the coal-fired power plant provides the energy for capturing CO_2 using amines, compressing to 130 bar for pipeline transport over a typical 200 km and arrival at injection sites at ~100 bar (Torp and Brown 2005; Maldal and Tappel 2004; BAKLID et al. 1996; Wildbolz 2007). Larger transport distances might be considered but the LCA model shows very minor sensitivity to changes in this distance (see section 4.5), indicating this in not a crucial parameter. In addition, the larger the CO_2 transport distance, the less likely the implementation of CCS using PCC, because costs for pipeline construction will increase with distance.

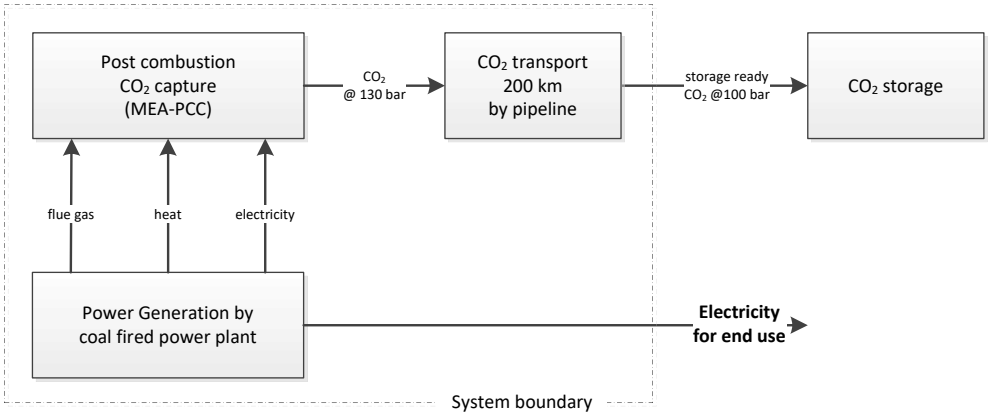

Figure 4.1: System boundaries of post combustion CCS (scenario "MEA-PCC$_{coal}$").

The HS-DAC system (Figure 4.2) is modelled as powered by either coal fired power plants or by a locally installed PV system; multicrystalline-Si, panel efficiency = 13.2%, average annual yield 820 kWh/kW$_{peak}$ (Jungbluth et al. 2007).

Our base case is electricity produced from coal with no carbon mitigation technologies yet in place. The respective inventory data are derived from the ecoinvent (v 2.2) database. The coal-fired power plant in this database (ID868) has an average efficiency of 35%. But newer power plants can have efficiencies up to 45% (World Coal Association 2014), thus reducing their CO_2 emissions in comparison with currently installed power plants. Since it can be expected that PCC will be installed on newly built plants, this study assumes a modern coal fired power plant with 45% efficiency (World Coal Association 2014). This is modelled by adjusting the primary energy demand required per kWh electricity generated (ID859) from 10.2 MJ to 8 MJ which also results in a corresponding reduction of upstream environmental emissions. The plant's CO_2 emissions, without capture, are 0.744 kg CO_2/kWh, or 78% of the emissions from the 35% efficient coal

fired power plant in said database. We base our LCA on an average 500 MW (peak) power plant operating for 4,000 hours per year at full capacity (Dones et al. 2007). Such plant thus generates 500 MW·4000 h = 2 TWh annually and emits $2 \cdot 10^9$ kWh·0,744 kg CO_2 / kWh = $1{,}49 \cdot 10^6$ tonnes CO_2 per year (1 tonne = 1,000 kg).

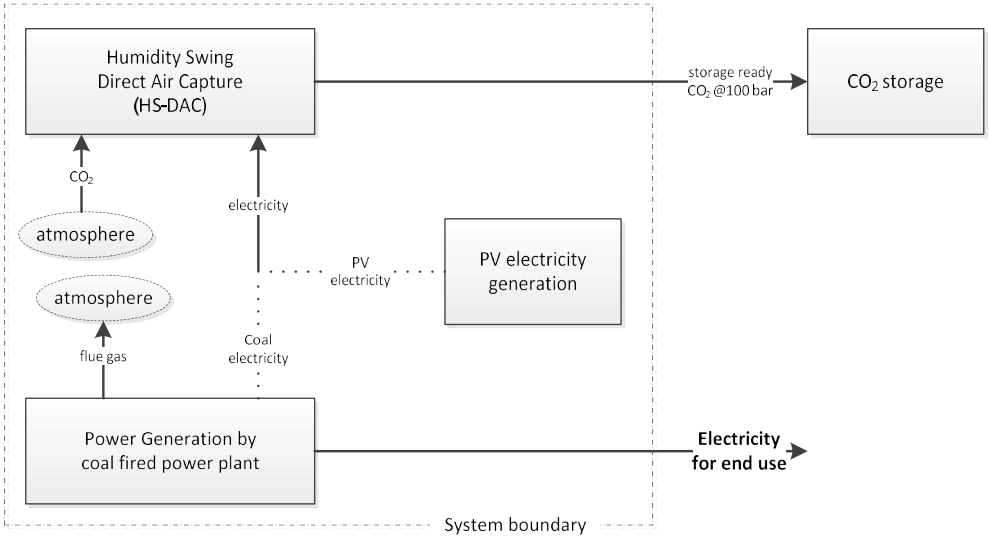

Figure 4.2: System boundaries of CCS using HS-DAC. HS-DAC is powered either by PV-based electricity (scenarios "HS-DAC$_{PV}$(Zero GHG)" and "MEA-PCC$_{coal}$+HS-DAC$_{PV}$(Zero GHG)") or by coal-based electricity (scenarios "HS-DAC$_{coal}$" and " HS-DAC$_{coal}$(Zero GHG)". HS-DAC re-captures the CO2 emissions in flue gas (scenario "HS-DAC"), and, in addition, life cycle GHG from mining and transporting coal, and manufacturing and operating infrastructure ("Zero GHG" scenarios).

We compare this base case to six scenarios, which add various CCS technologies to the base case in order to generate coal-fired electricity with reduced or zero net GHG per kWh for end-use.

- MEA-PCC$_{coal}$: Coal-fired electricity generation with typical MEA-based PCC that captures 90% of stack CO_2 emissions from combusting coal in the plant. The electricity output of the base case coal-plant (2 TWh) is reduced by the energy penalty (below). The LCA and our choice of functional unit quantifies life cycle impacts per kWh of the electricity remaining for end use. Considering the system beyond the power plant itself, this implicitly assumes that the plant's electricity shortfall for end users will be compensated by more MEA-PCC equipped coal plants of the same type (whose LCA per kWh would be the same).
- HS-DAC$_{coal}$: Coal-fired electricity generation combined with HS-DAC, set to capture an amount of CO_2 such that total net lifecycle GHG emissions per

kWh electricity available for end-use are the same as in the previous scenario. The electricity to power HS-DAC is assumed to come from the base case coal plant (or an equivalent plant in another geography).

- MEA-PCC$_{PV}$: Same as the MEA-PCCcoal scenario but the reduction of usable electricity output from the coal plant is compensated by PV instead of by coal plants. This scenario thus assumes a mixed coal-fired and PV generation per functional unit of electricity for end use. The LCA does not include possible need for energy storage to enable use of the intermittent PV electricity on the grid.
- HS-DAC$_{coal}$(zeroGHG): Same as HS-DAC$_{coal}$, but with increased HS-DAC capacity such that net lifecycle GHG emissions are further reduced to zero (i.e., compensating for 100% stack emissions as well as all background emissions).
- HS-DAC$_{PV}$(zeroGHG): Same as HS-DAC$_{coal}$(zeroGHG), but with electricity for HS-DAC provided by PV.
- MEA-PCC$_{coal}$+HS-DAC$_{PV}$(zeroGHG): Combination of previous two scenarios, such that MEA-PCCcoal captures 90% of flue gas CO_2 emissions and HS-DACPV all remaining life cycle GHG.

Life cycle inventory data

As motivated in the introduction, we focus on the energy requirements for both CO_2 capture technologies. Other inventory data (LCI) used in this study (base case and scenarios) are reported in detail in the supporting information (see section 4.5), including sensitivity analyses. Briefly, for MEA-PCC, the LCI data is derived from recent peer-reviewed publications. For HS-DAC, sorbent and hardware consumptions are based on extrapolations from laboratory prototypes and expected sorbent lifetimes. Per tonne CO_2 captured, neither energy nor other consumptions inherently depend on scaling laws such as surface-to-volume or structural performance and are thus largely independent of the HS-DAC's unit envisioned daily capture capacity (here 1 tonne CO_2/day; Discussion). Therefore, LCI data of a real life HS-DAC installation will likely not differ significantly from the hypothetical installation considered here.

Energy requirements for MEA-PCC

Energy requirements for PCC are typically expressed as an "energy penalty" (or equivalent work) that quantifies the drop in electricity output (per tonne CO_2 captured) of a PCC-enabled versus a non-capturing plant (Boot-Handford et al. 2014). Considerable uncertainty exists with regards to this performance metric (Zapp et al. 2012; Jenni et al. 2013): A recent review by Boot-Handford et al. (2014) cites a wide range for PCC in coal-fired power plants, 450kWh/ton to 200kWh/ton, for separation and compression to pipeline pressure to 100 bar or above (Boot-Handford et al. 2014). Specifically for MEA-based PCC (coal), van der Assen et al. (2016) reviewed 10 studies ranging from

473kWh/ton to 211kWh/ton (von der Assen et al. 2016). Importantly, both reviews show a trend towards lower energy penalties seen in more recent studies, indicating potential for lower energy penalties via current and future process improvements/ learning (Rochedo and Szklo 2013).

To account for both the wide range and the longitudinal trend, we therefore carried out LCAs for two energy penalties for MEA-PCC, (i) current/likely case; and (ii) future/ best case. These were determined as explained below and summarized in SI. For ease of comparison with related studies on PCC, we report energies in kWh/tonne captured (1 kJ/mol equals 6,31 kWh/tonne; 1 tonne = 1,000 kg). Further benchmarks and relevant equations to convert between other related performance metrics are given in section 4.5.

For the current/likely case, we determined the energy penalty based on specifications by engineering contractors and process licensors for the International Energy Agency (Davison 2007). For an implementation that reflects the standard studied here (MEA-based PCC for a new pulverized coal plant with 43% gross thermal efficiency), these find an energy penalty of (using parameters from "Fluor" scenario in Tables 1 and 2 from Davison (2007)):

$$\frac{(\eta_{\text{gross}} - \eta_{\text{net}})/\eta_{\text{gross}}}{90\% \cdot \text{PP}_{\text{CO2}}} = \frac{290\text{kWh}}{\text{tonne CO}_2}, \qquad \{\text{Eq1}\}$$

Where η_{gross} denotes the gross thermal power plant efficiency without capture, η_{net} the net thermal power plant efficiency with capture, and "PP_{CO2}" the amount of CO_2 emitted (tonne CO_2 per kWh generated) by the base case power plant.

We consider 290kWh/tonne (46 kJ/mol) to be at the low/optimistic end of the ranges cited in available studies and therefore representative of a current/likely case. For example, 290kWh/tonne closely matches the average energy penalty of the four most recent MEA-based studies reviewed in Boot-Handford et al. (2014); ~280kWh/tonne. Further, 290kWh/tonne is 15% lower than the average of 10 studies (coal, PCC with MEA) reported by van der Assen (2016) ; 1,22 GJ/tonne or 339kWh/tonne. 290kWh/ tonne is 21% lower than the energy penalty used in the landmark 2011 APS study (American Physical Society 2011) which was based on the DoE/NETL's engineering study of MEA-based PCC coal-fired plants (368kWh/tonne) (Netl- 2007).

For the future/best case, we selected one of the lowest energy penalties reported in recent literature (211kWh/tonne = 33kJ/mol), which was determined via simulation for

a MEA-based (9 molar) optimized PCC design (interheated columns).(Van Wagener and Rochelle 2011) 211kWh is in line with the 0.8GJ/tonne (222kWh) predicted by Rochedo et al. (2013, Figure 6) for future energy penalties expected by 2025 (Rochedo and Szklo 2013). 211kWh is also consistent with predictions by NEEDS for 2025 and 2050 (see details in section 4.5 and Bauer et al. (2008). Smaller simulated energy penalties have been reported for novel solvents such as aqueous piperazine (Boot-Handford et al. 2014), as well as solid or membrane sorbents (Goto et al. 2013). These sorbents are not the focus of this study.

Energy and water demand for HS-DAC

To calculate HS-DAC's energy and water consumption, we followed similar considerations as set forth in Lackner et al.(2009). However, we refined these based on recent laboratory studies and common adsorption-on-surface theory (Wang et al. 2011, 2013). We discern four steps of electricity consumption (Figure 4.3). Our model calculates the consumptions as a function of ambient temperature and relative humidity. Below we summarize the calculation steps for 20°C and 30% relative humidity. Further details for each calculation are provided in section 4.5. For ease of comparison with related studies on DAC, we report energies in kJ/mol (1 kJ/mol equals 6,31 kWh/tonne; 1 tonne = 1000 kg).

First, after adsorbing CO_2 from ambient air, sorbent filters are moved to a desorption chamber which is subsequently evacuated (0.01bar). Depending on the chosen filter geometry (i.e., sorbent versus air volume), the capture of 1 mol CO_2 requires evacuating 27 litres of air. Accounting for pump inefficiency (assume 70% electrical-to-mechanical conversion) (Wurzbacher et al. 2011), this evacuation requires 3.8 kJ/mol (24 kWh/tonne) electricity. 0,01 bar is chosen to remove enough of the residual air from the chamber such that the subsequently desorbing CO_2 is ~99% pure. This is followed by spraying the sorbent with water (see auxiliary electricity, below) which prompts the desorption process.

Second, CO_2 drying is achieved through condensation of water from the desorbing CO_2-H_2O gas mixture. Using previously measured sorbent characteristics (Wang et al. 2011, 2013) we calculate that on average 2.89 mol of H_2O have to be condensed per mol CO_2 captured. The condensation is triggered in a cold trap operated by an electrically powered heat pump with a coefficient of performance (COP_r) of 5 (section 4.5). Using 40,65 kJ/mol condensation heat for H_2O, this process requires 23 kJ/mol (145kWh/tonne) CO_2 electricity.

Third, compres

sing CO_2 from its varying desorption pressure (0,0044 bar – 0,0338 bar) to storage-ready 100 bar: We use Aspen HYSYS v8.6® to calculate the electricity input for a 15-stage, intercooled adiabatic compression train, followed for pressures above 57,3 bar (liquid) by a pump (compression ratio 1,88 to 1.64 per stage; Peng-Robertson properties; Schultz polytrophic method; adiabatic efficiency 75%(Mccollum and Ogden 2006)). To adjust for the gradually lower inlet pressure and mass flow rate as the sorbent depletes during each cycle, the train uses centrifugal compressors with controlled inlet guide vanes (or throttle valves) and variable speed (Boyce 2003; Biliyok et al. 2015). The electricity requirement for this step is 31 kJ/mol (196kWh/tonne).

Fourth, we estimate auxiliary electricity, for pumping water (40 molH_2O/mol CO_2), for powering the conveyor to move sorbent filters in and out of the chambers, and for overhead operations & controls (e.g., sensors, actuators, electronics) at ~3% of total electricity required for steps 1-3, or 2.0 kJ/mol (12.6kWh/tonne).

For HS-DAC, the regeneration of the sorbent occurs passively, via the sorbent drying at ambient conditions (see Introduction). Other than the electricity to propel the conveyor system (above), this drying does not require electricity or fuel input. Given a sorbent mass yield of 0,66%, the 37mol water per mol CO_2 captured correspond to ~100 gram of water evaporated per kg of sorbent regenerated. This water-to-material ratio is akin to drying damp sheets of paper on a clothesline. The air flowing through the sorbent filters will gain humidity and cool by 2.1 K (Section 4.5) without any other impact on the environment.

Therefore, for this step, only the water consumption itself, but no other energy or life cycle impact is included in the LCA.

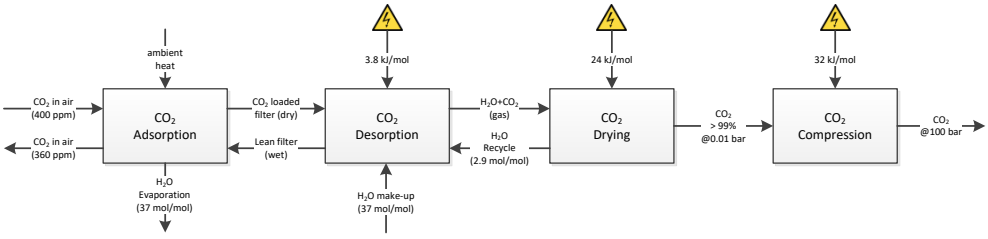

Figure 4.3: Schematic of HS-DAC process with electricity and water consumption as derived in Methods for ambient conditions of 20°C, 30% relative humidity, and 1 bar (details: SI). Not shown is the estimated 2.0 kJ/mol for auxiliary equipment and controls. Except for the temporary cooling during the drying and the multi-stage compression, all units and streams are at ambient temperature. Pressures for H_2O vapour and CO_2 in the desorption and drying steps vary as specified in Methods.

Comparison of energy balances and effect of CO_2 dilution

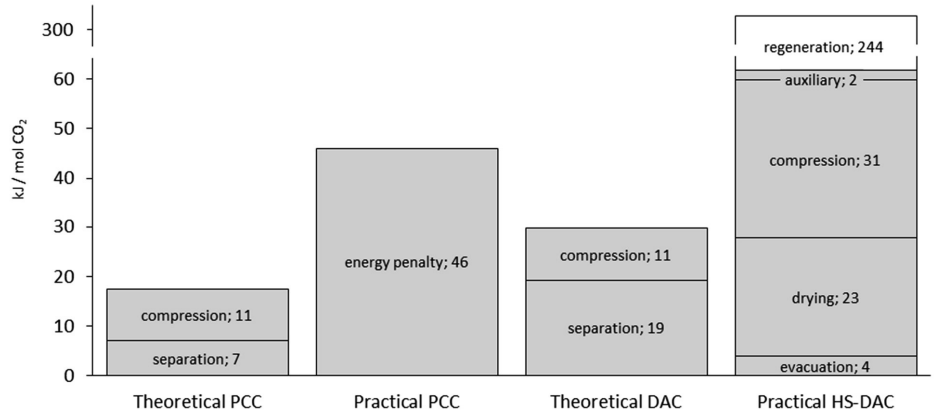

Figure 4.4: Theoretical (thermodynamic minimum) and practical energy breakdown for capturing CO_2 from a 12% concentration (PCC, 90% reduction, 20°C) versus 0.04% concentration (HS-DAC, 400ppm to 360ppm reduction, 20°C) and delivering this CO_2 at 100 bar (130bar for 'Practical PCC'). Required sorbent regeneration energy for HS-DAC (244 kJ/mol) was estimated based on experiments indicating that approximately 6 mol H_2O are swapped for any mol CO_2 on the anionic exchange membrane.(Wang et al. 2013) The required 40.65kJ/mol H_2O evaporation energy is drawn from ambient conditions and therefore not included in the LCI inventory. However, the actual water consumption of the process (37mol H_2O per mol CO_2) is higher, and this higher water consumption is reflected in the LCA.

Based on thermodynamic entropy considerations, separating CO_2 from air via DAC is expected to require ~3 times more energy than from flue-gas via PCC, because DAC captures CO_2 from a ~300 times more dilute source (Figure 4.4) (American Physical Society 2011; Goeppert et al. 2012; Lackner 2013). However, considering practical processes instead of thermodynamic minima, addressing the heat versus electricity portions of the total energy demand in HS-DAC versus PCC separately, and considering 100bar instead of 1bar, gives a more nuanced result: HS-DAC requires 60 kJ/mol CO_2 captured (20°C, 30% humidity case) versus 46 kJ/mol for MEA-PCC (current/likely case). This shows that while the higher dilution of CO_2 has a factor ~3 effect when analysing thermodynamic minimum conditions and 1 bar, this does not translate into equally different electricity requirements of the two processes for practical implementations. This feature of the energy balance is crucial for the comparative LCA of the two processes.

4.3 Results and impact assessment

Life cycle GHG balance

The life cycle GHG balance of the base case power generation and the six scenarios are presented in Figure 4.5. In line with previous LCA studies (Corsten et al. 2013), we find that PCC (at 90% capture rate) reduces the system-wide lifecycle GHG intensity of coal-based electricity by 73%, from 0,85 to 0,23 kg CO_2-eq/kWh.

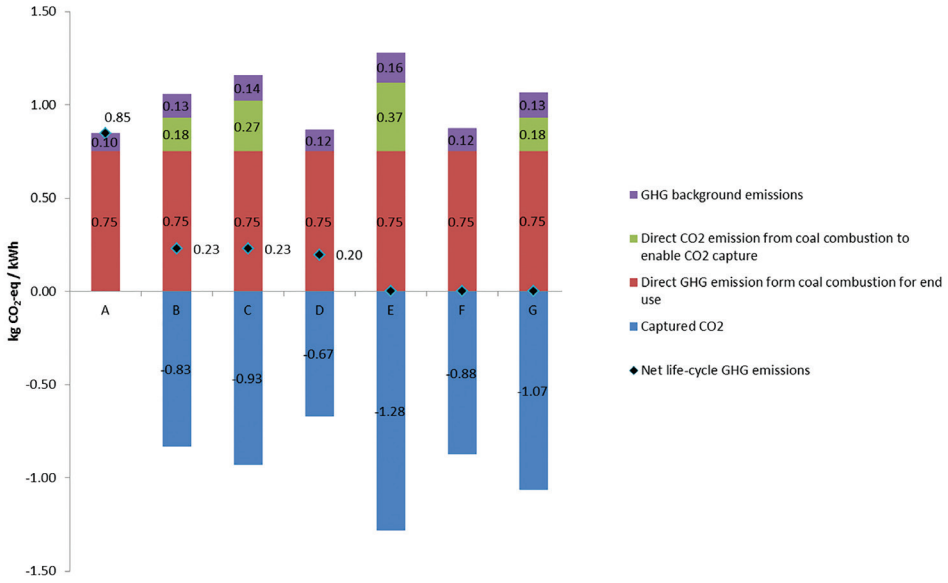

Figure 4.5: Net GHG emissions and breakdown per scenario. Background emissions refer to CO_2 and non-CO_2 GHG emissions that are not the result of coal combustion in the plant itself. Background emissions also include emissions related to the use of PV electricity in scenarios D, F and G. (A=Base case, B=MEA-PCC$_{coal}$, C=HS-DAC$_{coal}$, D=MEA-PCC$_{PV}$, E=HS-DAC$_{coal}$(ZeroGHG), F=HS-DAC$_{PV}$(ZeroGHG), G=MEA-PCC$_{coal}$+HS-DAC$_{PV}$).

When HS-DAC is sized to achieve the same net life-cycle GHG emissions as MEA-PCC (i.e, 0,23 kg CO_2-eq per functional unit), 0,93 kg CO_2/kWh need to be captured, compared to 0,83 kg CO_2/kWh for MEA-PCC. This is because of the higher (coal-based) electricity demand of HS-DAC per amount CO_2 captured. To show the potential of HS-DAC to be proportionally scaled to capture more CO_2 (simply by adding more of the same HS-DAC units), we also modelled two alternatives with zero net GHG emissions. When HS-DAC is powered by electricity from coal, the net zero scenario must capture 1,28 kg CO_2/kWh (0.88 kg CO_2/kWh when powered by PV). In addition, Figure 5 shows the GHG balance for supplementing coal-powered MEA-PCC with PV-powered HS-DAC and for employing PV to make up for the energy penalty of MEA-PCC (see Methods for details of all 6 scenarios).

Non-GHG environmental impacts, problem shift, and role of water

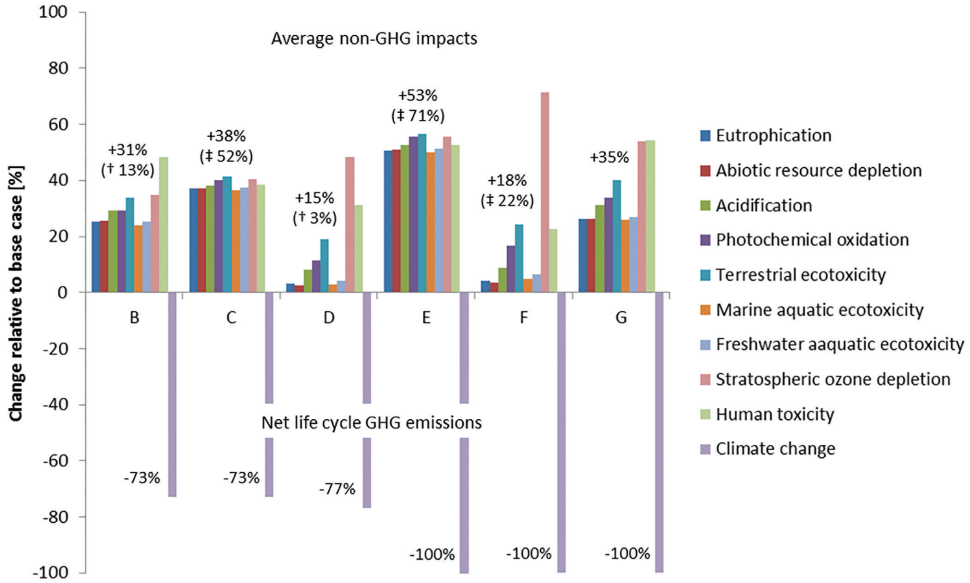

Figure 4.6: Relative change in environmental impacts relative to base case (500 MW coal fired power without CCS). The analysis assumes the current/likely energy penalty for PCC and 20°C/30% ambient conditions for HS-DAC. †Shows average non-GHG impacts (except water) for future/best case energy penalty for PCC. ‡Shows average non-GHG impacts for 10°C/60% ambient conditions for HS-DAC. (B=MEA-PCC$_{coal}$, C=HS-DAC$_{coal}$, D=MEA-PCC$_{PV}$, E=HS-DAC$_{coal}$(ZeroGHG), F=HS-DAC$_{PV}$(ZeroGHG), G=MEA-PCC$_{coal}$+HS-DAC$_{PV}$).

While MEA-PCC and HS-DAC scenarios reduce GHG emissions and thus climate change impacts of coal-based electricity, the increased background processes such as manufacturing additional hardware and mobilizing more materials such as coal (except in the PV scenarios), sorbent, and water increase other environmental impacts per functional unit (Figure 4.6). Consistent with previous LCA studies (Koornneef et al. 2008; Singh et al. 2011), we find that MEA-PCC increases the other 9 impacts tracked in our study (except water) by 31% on average versus the base case (see Figure 4.7 for water depletion). Employing HS-DAC to achieve the same GHG mitigation increases this "problem shift" from 31% to 38%, reflecting the higher electricity demand of HS-DAC per amount CO_2 captured. However, the LCA also shows that with HS-DAC powered by PV (intermittently), a complete removal of GHG per functional unit can be achieved with a smaller problem shift of 18% (Discussion). The increase in stratospheric ozone depletion in this scenario is due to upstream emissions in the production of the specific PV cells and is noticeable in all scenarios that use PV. For HS-DACcoal, the average penalty is 53% (all impacts except GHG and water). If CCS implementation does not aim for net zero GHG – but rather for the capture technology with the smallest

problem shift per GHG avoided – then the preferred scenario is employing MEA-PCC while compensating the energy penalty with PV (smallest problem shift 15%). Corresponding metrics of all impact categories expressed per kg net GHG avoided are provided in section 4.5.

HS-DAC requires fresh water in order to prompt the humidity swing (low quality, non-potable water is sufficient, however salt water would likely lead to prohibitive contamination of the sorbent) (Lackner et al. 2012). Direct water demand of HS-DAC is 15,2 m^3 per tonne CO_2 captured compared with 0,8 m^3 / tonne for MEA-PCC. Therefore, life cycle water depletion in scenarios involving HS-DAC is larger than in the two PCC scenarios (Figure 4.7). Note that the coal-burning power plant assumed in our base case does not feature coal transport via slurry pipeline or wet tower cooling, two technologies that would lead to higher water withdrawal even in the uncaptured base case (up to 4m3 and 6m3 water per tonne CO_2 emitted, respectively) (Fthenakis and Kim 2010).

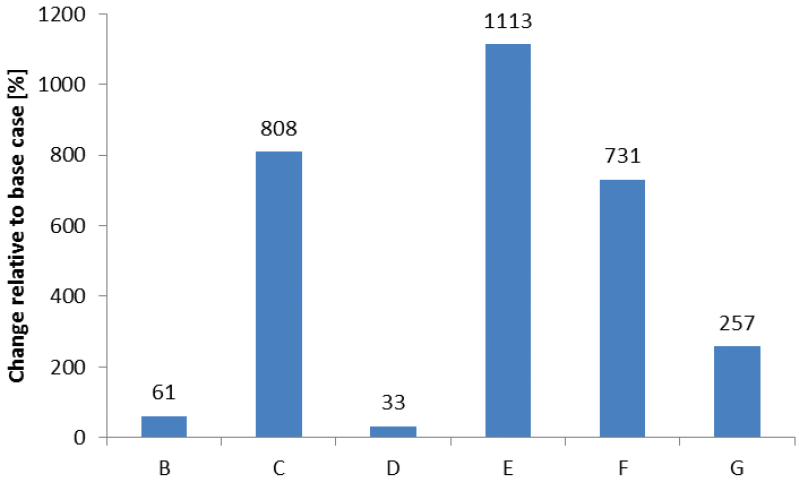

Figure 4.7: Water depletion for all scenarios (20°C, 30% relative humidity case for HS-DAC and current/likely case for MEA-PCC) in % relative to base case. (B=MEA-PCC_{coal}, C=HS-DAC_{coal}, D=MEA-PCC_{PV}, E=HS-DAC_{coal}(ZeroGHG), F=HS-DAC_{PV}(ZeroGHG), G=MEA-PCC_{coal}+HS-DAC_{PV}).

Uncertainty and sensitivity to ambient conditions

The dominant driver of the LCA results for both HS-DAC and MEA-PCC (except PV scenarios) is the additional combustion of coal (per functional unit) to enable the capture processes, rather than the additional materials and infrastructure (Figure 4.5). This is driven by the capture process' electricity requirement which could be robustly quantified based on previously published laboratory measurements (HS-DAC) and engineering studies (PCC) (Methods). In contrast, sorbent and hardware consumptions

are estimated, implying higher uncertainty. However, these parameters are less material to the LCA (sensitivity analyses on sorbent and hardware consumptions are included in section 4.5).

Crucially, the operation of a HS-DAC unit is sensitive to ambient conditions, particularly temperature and humidity, as these affect the sorbent's loading characteristics and thus HS-DAC's electricity and water consumption. We therefore ran sensitivity analyses to quantify this effect on electricity and water requirement and corresponding LCA results. For example, changing the 20°C and 30% humidity in our model to reflect a colder, more humid (10°C|60%) climate (or equivalent variations throughout the day) will lower the sorbent's initial CO_2 load when it enters the desorption chamber. The resulting detrimental effect on sorbent yield and thus long term sorbent consumption per tonne CO_2 captured can be partially counteracted by desorbing to lower pressures in a cycle. Such operation at 10°C|60% achieves 0.49% yield (versus 0.66% for the 20°C|30% operation; yield expressed as mass CO_2 captured per mass sorbent per cycle). But, this causes more direct electricity (+21%) and water consumption (+33%) relative to the base case. Therefore, under such cold/humid operation, the problem shift would increase from 38% to 52%, 53% to 71%, 18% to 22% and from 35% to 37% for the HS-DAC_{coal}, HS-DAC_{coal}(ZeroGHG), HS-DAC_{PV}(ZeroGHG) and MEA-PCC_{coal}+HS-DAC_{PV}(ZeroGHG) scenarios respectively (Figure 4.6). See section 4.5 for all impacts including water.

In contrast, due to the sorbent loading responses around these conditions, warmer and dryer conditions (30°C|10%) would only marginally increase the sorbent's initial CO_2 load. If desorption pressure is adjusted to maintain the same mass yield 0,66%, this would have only very small effects on electricity and water consumption (<1%) and likewise result in nearly the same average non-GHG LCA impacts and water demand as those for the 20°C|30% operation shown in Figure 4.6. Note however that such warmer and dryer conditions would likely accelerate sorbent drying (and vice versa). This would improve capacity utilization of a given machine and thus economics. Similarly, actual sorbent yields during continuous cycling will be somewhat lower than the equilibrium sorbent yields calculated here, again affecting economics but not energy balance (details on sorbent kinetics: section 4.5).

4.4 Discussion

In this paper we have analysed the comparative merit of flue gas capture and direct air capture, by comparing the environmental impacts of two CO_2 capture processes: the traditional one where CO_2 is captured from coal flue gas using MEA-based PCC; the other with CO_2 captured from air with HS-DAC. Previous studies stressed that HS-

DAC may be a promising technology but that more research on DAC is needed and that R&D and commercialization of DAC at larger scales will provide the necessary experience for further development (Williamson 2016; Smith et al. 2016). In particular, DAC has been viewed as being too energy-intensive (American Physical Society 2011), citing up to 45GJ per tonne carbon equivalent (540kJ/mol CO_2) (Smith et al. 2016). The research presented here contributes to the necessary broadened knowledge base for HS-DAC.

We conclude that - based on the currently available data - by virtue of its limited high-exergy energy requirement and corresponding environmental penalties, HS-DAC could expand the scope of CCS, including the realization of net-zero GHG electricity from coal. This could unlock potential advantages of DAC, including fewer social obstacles related to CO_2 transport and storage near population centres, the option to compensate for GHG emissions from sectors with diffuse emissions (e.g., aviation), the possibility of re-capturing 100% of the GHG emissions along the entire life cycle, and the possibility of CCS being used as a last resort back-stop technology to reduce GHG in the atmosphere (negative emissions) (National Academy of Sciences 2015). Still, PCC is clearly the preferred short-term option to capture CO_2 from flue gases, particularly when the goal is reducing GHG emissions (with minimal problem shift to other environmental impacts) rather than full net-zero-GHG electricity.

The higher electricity demand of HS-DAC versus MEA-PCC, other chemical and hardware requirements, and the separation of combustion and capture site could conceivably lead to a problem shift, i.e. prohibitive increases in environmental issues other than climate change. From our research we find that this fear is probably unfounded: Comparing MEA-PCC to HS-DAC (at equivalent GHG emissions), the shift to other environmental impacts does increase (from 31% to 38%, Figure 4.6) but is still lower than the factor 3 that might have been expected based on the increased minimum energy demand for separating CO_2 from a gas mixture. If HS-DAC were used to reduce lifecycle GHG emissions of coal-based power to net-zero (but is itself driven by PV), other impacts are on average lower than with MEA-PCC.

However, our analysis also shows HS-DAC's higher water consumption and sensitivity to climate (temperature, wind speed, humidity). Resulting crucial disadvantages of HS-DAC are therefore that it cannot be universally sited but rather requires access to water and minimal ambient wind speeds and temperature / humidity conditions outside which HS-DAC becomes less efficient. Quantifying sorbent drying times and how these can be optimized will be an important area of further study. As an extreme example, a complete standstill of the unit while in rainy, wind-still weather would raise capital

costs, albeit not energy requirement per tonne CO_2 captured. This makes HS-DAC somewhat akin to other distributed technologies in the sustainable energy realm such as wind farms or PV: its largely passive operation offers advantages for average energy balance; but exposure to the elements leads to intermittent operation and geographical limitations.

Decoupling emissions from capture

Zero-GHG DAC leverages two unique features of distributed CDR. First, to achieve net-zero GHG, HS-DAC is scaled up proportionally, without incurring the non-linear energy penalties caused by scrubbing flue gas to >90% captured from stacks. Also, PCC could not clean up any background emissions. Second, running HS-DAC intermittently on renewables (while still generating grid electricity via coal to serve society's continuous demand) leverages HS-DAC's ability to operate in remote geographies, possibly on continents other than the one on which the coal-fired power plant itself is located, where CO_2 may be easier to store and intermittent renewable electricity may be plentiful; meanwhile the atmosphere (and its coupling with ocean systems) acts as a temporal buffer for the emitted and re-captured GHG (Eisaman et al. 2012).

Implementation considerations

In addition to its reliance on suitable ambient temperature, wind, and relative humidity, HS-DAC's large water consumption means that its implementation will be limited to geographies where such amounts of water are available – even though this water is not polluted or contaminated but simply evaporates and thus returns to the natural water cycle.

While other practical considerations were not the immediate focus of this study, our process data on MEA-PCC and HS-DAC also allows us to evaluate the relative requirements of HS-DAC in terms of land use, installation size, and operating cost.

For MEA-PCC, the actual land requirement of the capture installation (excluding CO_2 transport infrastructure) can be estimated to be of roughly the same size as the power plant from which it captures (IEA Greenhouse Gas R&D programme 2005). For reference, the 500 MW power plant used in this study occupies an area of 125,000 m^2 (Dones et al. 2007). For HS-DAC, the land requirement will be dominated by and can thus be approximated as its required vertical wind facing cross section, using ambient wind speeds (2m/s, 20°C), and CO_2 uptake from air per pass through the DAC unit (40ppm or 10%). We further assume an idle time of 12 hours per day, for lack of sufficient wind or suitable weather. To capture 1 tonne CO_2 per day, an HS-DAC unit would thus require a wind facing cross section of 160m^2. This cross section must be

occupied by an array of sorbent filters that are in the adsorbing stage of the humidity swing cycle. Behind these filters, there are additional filters (in the drying phase) and a container-like structure on the ground where filters undergo desorption (as well as pumps, cold traps, compressors, and infrastructure for the captured CO_2). Capturing 90% of the 1.49×106 tonnes CO_2 / year emitted by the coal plant would thus require 3674 such HS-DAC units or 587.835 m^2 of land (4,7 times that of MEA-PCC). To allow CO_2 concentration and wind speed to recover in-between units, the units would be scattered over a larger land area. However, this area would be available for other use (akin to the space in-between individual windmills on large windfarms).

Regarding costs, consumption of electricity (20°C|30% case, 378 kWh/tonne CO_2 at \$0,07/kWh=\$26,5/tonne CO_2) (American Physical Society 2011), water (15,2m^3/tonne CO_2 at \$0,40 per m^3=\$6,1/tonne CO_2) and make-up sorbent filters using Marathon A (1,52kg/tonne CO_2 at \$6/kg=\$9,1/tonne CO_2) would yield direct operational costs of \$42 per tonne CO_2. To facilitate comparison to previous studies, we use the same electricity cost as in the APS study (American Physical Society 2011). Other costs are conservative estimates for non-potable water and customized, extruded resin filter elements. Benchmarking HS-DAC costs against a lower bound of \$60 per tonne CO_2 for MEA-PCC (American Physical Society 2011), this would leave \$18/tonne CO_2 for HS-DAC capital charge and O&M costs, or ~\$43.000 capital layout for a 1 tonne CO_2/ day HS-DAC unit (assuming capital charge of 10% of capital layout and annual O&M costs of 5% of capital layout). A more conservative CO_2 price of \$100/tonne would allow ~\$142.000 per unit. While it is difficult to predict the capital layout of HS-DAC units – as with any future technology –, we conclude that HS-DAC does not require difficult process conditions such as high temperatures, nor does it need rare or expensive materials or equipment. Furthermore, the economic benefit from the freedom to locate the HS-DAC units and injection at optimal sites independent of coal power plants and transport concerns is difficult to assess without specific case studies, but it may be significant (The Royal Society 2009). A detailed analysis of the total costs of HS-DAC should be the focus of future research.

Acknowledgements
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4.5 Appendix

This appendix contains the supporting information for this case study and includes details on the modelled processes, supporting calculations.

4.5.1. Energy analysis

HS-DAC energy consumption

The energy consumption for humidity swing based direct air capture (HS-DAC) is broken down into 5 steps, 4 for electricity, and 1 for ambient heat, addressed one by one below. The 5 steps deliver >99% pure CO_2 at 100 bar pressure. For ease of comparison with related studies, we report energies in kJ per mol CO_2 captured (1 kJ/mol equals 6,31 kWh / tonne; 1 tonne = 1000 kg).

Overview: Determining equilibrium loading of CO_2 for sorbent yield

To obtain the electricity consumption in the DAC process, we follow similar considerations and calculations as set forth in Lackner et al.(2009) However, we refined these calculations based on recent laboratory studies and common adsorption-on-surface theory.(Wang et al. 2011, 2013) Together, these let us quantify the equilibrium loading of the sorbent with CO_2 as a function of partial CO_2 pressure (P_{CO2}), relative humidity (H), and temperature (T) to which the sorbent is exposed. Following Langmuir adsorption theory, we write:

$$RL(P_{CO2}, H, T) = \frac{P_{CO2}}{K(H,T)\cdot\left(1+\frac{P_{CO2}}{K(H,T)}\right)} \quad (1)$$

where K (in atm) is the partial pressure at which the relative loading RL is 50%. RL is thus a dimensionless number between 0 and 1 that indicates the fraction of total available CO_2 adsorption capacity that is utilized (in equilibrium) at any specific PCO_2. The absolute loading AL (in mol CO_2 per kg of sorbent) can be found based on the carbon chemistry of the particular sorbent (Lackner 2009):

$$AL(P_{CO2}, H, T) = RL(P_{CO2}, H, T) \cdot \frac{1.78}{2} mol/kg \quad (2)$$

$K^*(T, H)$, in which * indicates dimensionless pressures expressed as fractions of the standard condition 1 atm, can be rewritten as a function of the standard molar Gibbs free energy of adsorption dG:

$$K^*(T, H) = e^{\frac{dG(T,H)}{RT}} \quad (3)$$

where R is the ideal gas constant. Laboratory measurements showed that *dG(T, H)* is well described by the following fit function (Table 2, 3rd of 4 parameter sets, in Wang et al. (2013).

$$dG(T,H) = -32.20\frac{kJ}{mol} + 15.45\frac{kJ}{mol}\cdot H + 0.02564\cdot(T-288.15)\frac{kJ}{mol\cdot K} \qquad (4)$$

Using above *AL*, we can now determine the sorbent yield Y_{CO2} (in mol CO_2 per kg of sorbent) per any one cycle, where a cycle is characterized by *H* and P_{CO2} during adsorption (here: 30% and 400 ppm of 1 bar, respectively) and *H* and P_{CO2} during desorption. At desorption, *H* is 100% per definition of the process and PCO_2 can be controlled by the flow-rate of the compression pump. At 30% and 400 ppm of 1 bar, the sorbent adsorbs to RL_{Start} = 0,97. Once in the desorption chamber, evacuated, and exposed to *H* = 100%, it desorbs gradually from *RL* = 0,97 (corresponding to P_{CO2} = 0,0340 bar) to RL_{End} = 0,80 (corresponding to P_{CO2} = 0,0044 bar, which is the lowest pressure from which CO_2 must be subsequently compressed). Based on above *AL* as function of *RL*, resulting YCO_2 is 0,15 mol CO_2 per kg of sorbent (0,66% mass yield).

Step 1: Evacuation and desorption (electricity)

The energy required for desorption of the sorbent consists of electricity to pre-evacuate ambient air from the desorption chambers. The DAC process studied herein uses sorbent elements constructed such that, in the desorption chamber, each kg sorbent is surrounded by 4 litres of air (i.e., ~20% volume ratio given that the sorbent's density is close to 1 kg/litre). Given above YCO_2, capture of every 1 mol CO_2 therefore requires evacuating 27 litre (design dependent). At 100 kJ per m^3 (1 bar, 20˚C) and further accounting for pump inefficiency (assume 70% electrical to mechanical conversion),(Wurzbacher et al. 2011) this gives 3,8 kJ/mol. Note this calculation uses a simple P·V view to estimate the mechanical work required to evacuate a given volume against a constant outside pressure. Overall, the contribution of the desorption step to the total electricity consumption (steps 1-4) is small (<10%). Therefore, we leave the calculation at this level of detail.

Step 2: CO_2 drying (electricity)

During desorption, the sorbent gives off a mixed stream of CO_2 and H_2O. While P_{CO2} in this stream drops gradually as loading decreases from RL_{Start} to RL_{End} (see explanation above pressure provided in main manuscript), the partial pressure of H_2O in the stream stays constant (given by the vapour pressure at 293 K: 0,024 bar). This means that the CO_2:H_2O mol ratio of the stream, which is proportional to the ratio of partial pressures in the stream, decreases during desorption of one cycle. The average mol-ratio for a full cycle can be found by numerically integrating equations (1) and (2) using standard

software. We find that on average 2,89 mol of H_2O have to be removed for every mol of CO_2 retrieved. The removal is achieved by condensation via a cold trap cooled by a heat pump with a coefficient of performance (COP_r) of 5. Using 40,65 kJ/mol condensation heat for H_2O, this yields 23 kJ per mol CO_2.

Efficiency of cold trap:

COP_r is derived as follows: Cooling the desorption stream by 30°C lowers the H_2O vapour pressure to 0,0032 bar (Thangavel et al. 2013). The theoretical maximum COP_r in an ideal Carnot process, assuming 20°C=293,15 Kelvin ambient, is thus (293,15K-30K)/30K = 8,77. With a further drop of 43% due to inefficiencies in a real system, we arrive at COP_r of 5,0 which is achievable with real refrigeration systems (Thangavel et al. 2013).

Phase changes of H_2O in cold trap:

In order to yield a sufficiently pure CO_2 stream (if near zero residual water is desired before entering the multi-stage compression), the partial pressure of water vapour in the desorption stream must be sufficiently reduced. At 20°C ambient, the cold trap therefore has to be cooled to temperatures below 0°C. In this regime, the water vapour in the desorption stream will undergo a phase change directly to a solid. After each cycle, the cold trap will refill with ambient air and warm up; the ice will melt and can thus be removed from the system by gravity. The HS-DAC installation envisioned in our study has a capacity of 1 metric tonne CO_2 per day and during its 12h/per day operation undergoes 30 cycles. About 39kg of ice have to be removed per cycle. However, this amount of ice is an upper bound. In a real implementation, the cold trap does not have to be at a single uniform temperature < 0°C. Rather, the water removal process will be spread out through a series of interconnected pipes: The desorption stream which exits the sorbent at 20°C with 0.024bar vapour pressure (above) will first be cooled to temperatures still above 0°C, such that a portion of the water already condenses out as a liquid. The lower temperature drop also increases COP_r in this portion of the piping system (because of the higher Carnot efficiency), thus reducing the electricity requirement for this portion of condensed water. The temperature of the desorption stream will then gradually be dropped further as the desorption stream is pumped through the piping system. Only the last portion, before entering the multi-stage compression train, has to be cooled to temperatures sufficiently low to reduce any remaining water vapour (by forming ice) in the desorption stream to less than 1/100 of the partial CO2 pressure (~99% purity).

Step 3: CO_2 compression (electricity)
Energy required to compress the produced CO_2 consists of electricity to drive a CO_2 compressor. As explained for the drying step above, different portions of CO_2 desorb at different pressures (to which they fill the desorption chamber). Therefore, each such portion requires different compression work to yield 100 bar CO_2. The calculation (Aspen) is explained in the main text.

Step 4: Auxiliary processes (electricity)
The steps above cover all energy-intensive processes of the proposed HS-DAC. Remaining are some electricity to move sorbent (in and out of the desorption chamber) and water, and furthermore some overhead for operations & controls (such as sensors, actuators, monitoring electronics).

This electricity was estimated bottom-up, using an envisioned machine size of 1 tonne CO_2 captured per day (which runs 24h-12h=12h each day). 2,0kJ/mol (or 13kWh per tonne) which amounts to 3,3% of the total electricity consumption (60kJ/mol) comprises the following elements (all other electricity requirements are accounted for in other steps (see Figure 4.3):

- 0,7kWh to move the filters on a horizontal conveyor-belt like structure (for drying and adsorption outside the regenerator as well as for shuttling them through the regenerator where CO_2 desorbs): Except for friction and efficiency losses of the electric motor powering the conveyor, this process does not consume energy. To estimate the electricity consumption of the real system (i.e., with friction), the conveyor motor (and appropriate gear system) is envisioned at a power to overcome 1% incline at 1m/s speed, i.e. 0,01W per kg mass conveyed. To move the total mass of filter elements in the envisioned machine (5,56 tonne) this accounts for 0,67kWh over 12h operation per day. This is only about 5% of the total auxiliary electricity requirement, and therefore a more accurate calculation is deemed unnecessary.
- 1,3kWh to spray water onto the sorbent filters in the evacuated regenerator: Per tonne CO_2, 16,4m^3 water needs to be sprayed (of which 1,2m^3 is later condensed out and thus recovered rather than consumed; see main manuscript). This is envisioned by compression of the water from 1 to 3 bar by a 70% efficient electric pump (Wurzbacher et al. 2011).
- 11kWh to power electronic controls, sensor, and actuators for water and air valves: We estimate this conservatively (i.e., high) to require 100W of continuous power per tonne of total machine weight. The total machine weight including filter elements is 8.9 tonnes. This accounts for 11kWh over 12h operation per day.

Step 5: Sorbent regeneration (ambient heat)

During regeneration the sorbent is returned to its thermodynamic base state (i.e., mostly dry, and low CO_2 loading). As shown previously and explained in the main manuscript (Wang et al. 2011, 2013), the required energy for this sorbent regeneration is driven predominantly by ambient heat (provided by the 30% humidity/20°C air) that causes the sorbent to dry. However, this heat does not have any associated life cycle impacts and is therefore not counted as part of the inventory in the environmental assessment.

MEA-PCC energy consumption

Energy consumption of MEA-PCC is treated via the concept of energy penalty (see the main text and Table 4.1 below).

Table 4.1 Energy penalties for PCC used in this study and literature benchmarks. All figures for 90% capture rate, MEA-based PCC in coal-fired plants, and inclusive of compression to ~130bar.

Table 4.1: Energy penalties for PCC used in this study and literature benchmarks. All figures for 90% capture rate, MEA-based PCC in coal-fired plants, and inclusive of compression to ~130bar.

Overall	Typical range reported in literature (Table S1 in Von der Assen et al. (2016), including older, not state-of-the-art implementations	211-473 kWh/tonne
Current/likely case used in this study: 290 kWh/tonne	Average of four most recent MEA-based studies reviewed in figure 3 in Boot-Handford et al. (2014)	280 kWh/tonne
	Specifications by engineering contractors and process licensors for the International Energy Agency (Davison 2007)	290 kWh/tonne
	Average of 10 studies reported in table 1 in Von der Assen (2016)	339 kWh/tonne
	American Physical Society (2011) and DoE/NETL's engineering study by Davison (2007)	368 kWh/tonne
Future/best case used in this study: 211 kWh/tonne	Future scenarios predicted for 2025 and 2050 horizons by NEEDS (2008) and Bauer et al. (2008) (Table 4.18, details see SI)	~200 kWh/tonne (6% efficiency penalty)
	Lowest recent simulation result reported in literature by Wagener and Rochelle (2011)	211 kWh/tonne
	Past PCC learning curve; figure 6 in Rochedo et al. (2013) extrapolated to 2025	222 kWh/tonne

Here we provide further benchmarks. In addition to energy penalty, which is specified in electric energy per mass CO2, three other often published performance metrics exist in literature (Vasudevan et al. 2016). As % figures, these are a function not only of the actual energy penalty (in kWh/tonne) but further of the plant's CO2 emissions and

thermal efficiency, and therefore a less direct metric when comparing different studies. However, we include these here as a further point of comparison for the energy penalties used in our calculations reported in the main article (current/likely and future/best case). We use the following definition for the three metrics (e.g., Vasudevan et al.(2016)

$$\textbf{\textit{Parasitic load}} = \frac{\eta_{gross} - \eta_{net}}{\eta_{gross}} \qquad \{\text{Eq S1}\}$$

Where η_{gross} denotes the gross thermal efficiency of the power plant without capture and η_{net} denotes the net thermal power plant efficiency with capture. Note that in some

$$\textbf{\textit{Increase in fuel use}} = \frac{1}{(1 - \%\ Parasitic\ load)} - 1 \qquad \{\text{Eq S2}\}$$

$$\textbf{\textit{Efficiency penalty}} = \eta_{gross} - \eta_{net} \qquad \{\text{Eq S3}\}$$

(Note this is sometimes referred to as energy penalty.)

Substituting EqS1 in Eq1 (main text) and using 744 gram CO_2/kWh from our base case, we find that the 290kWh/tonne for the current/likely case corresponds to a *Parasitic load* of 19,4% (or an *Increase in fuel use* of 24,1%). Further using the 45% thermal efficiency of the un-capturing plant in our base case, we infer from EqS1 and EqS3 that the *Efficiency penalty* corresponding to 290kWh/ton is 8,7%. 8,7% is at the low end of those reported in Zapp et al.(2012) and close to the 9% for PCC in pulverized coal power plants used by Reiter et al. (2015), table 2. The *Parasitic load* corresponding to our future/best case (211kWh/ton) is 14.1%, in line with the lower end (~15%) of the wide range predicted for 2025 by a group of 12 experts surveyed by Jenni et al.(2013), Figure 1. The *Efficiency penalty* corresponding to 211kWh/ton is 6.4%, consistent with the 6% reported for optimistic, future looking scenarios as those specified for 2025 and 2050 horizons by NEEDS (2008) Bauer et al. (2008), table 4.18.

Temperature drop in air as it aids water evaporation

The sorbent is regenerated and rendered back into its adsorbing state once all originally applied water is evaporated off the sorbent during the drying phase. We can therefore calculate the ambient heat drawn from the (30%humidity/20°C) ambient air per ton CO_2 captured and infer the resulting temperature drop in the airflow through the filters.

At a capture rate of 10% (400 ppm to 360 ppm, design-dependent), $13{,}9 \times 10^6$ m^3 (1bar, 20°C) need to pass through the array of sorbent filters for any metric ton of CO_2 captured. This amount of air will also pass through the second set of sorbent filters in the drying phase of the HS-DAC cycle (main manuscript). The heat to evaporate 37mol

H_2O per mol CO_2, at 40,7 kJ/mol (evaporation heat of water), will be drawn from this air. Given an isobaric volumetric heat capacity of air of 0,00121 J/(mol·K), this will cool the air by 2,1 Kelvin.

Note this calculation assumes that the airflow speed through the second set of filters (i.e., the ones in the drying state likely positioned at some distance behind the ones in the adsorbing state) is the same as that through the first set (and not slower). This assumption likely overestimates the air volume flowing through the second set and thus underestimates the temperature drop. At the same time, however, this calculation disregards the effect of evaporation heat supplied by sunlight, thus overestimating the temperature drop.

4.5.2. Environmental analysis

Defining the scenarios

A base case and six scenarios for comparison have been defined. Here we describe these scenarios as modelled in the LCA model, as well as one additional 7th scenario (scenario H=PV electricity) that is included here only for the purpose of reference and comparison.

Table 4.2: Scenarios for environmental assessment.

A. Base case	Providing electricity to the grid with a 45% efficient capture ready Europe based 500 MW coal fired power plant. Life cycle environmental impacts are based on this system delivering 1 kWh of electricity to the grid.
B. MEA-PCC_{coal}	Coal-fired electricity generation with typical MEA-based PCC that captures 90% of stack CO_2 emissions from combusting coal in the plant.
C. HS-DAC_{coal}	Coal-fired electricity generation combined with HS-DAC, set to capture an amount of CO_2 such that net lifecycle GHG emissions per kWh electricity available for end-use are the same as in the previous scenario (B). The electricity to power HS-DAC is assumed to come from the base case coal plant (or an equivalent plant in another geography). N.B. in the LCA model the amount of CO_2 captured is increased (to 0,935 kg) such that the end-to-end climate change impacts of both scenario B and C are the same (0,229 kg CO_2-eq).
D. MEA-PCC_{PV}	Same as the MEA-PCC scenario (B) but the reduction of usable electricity output from the coal plant, caused by post combustion capture, is compensated by PV. This scenario thus assumes a mixed coal-fired and PV generation per functional unit of electricity for end use. The LCA does not include possible need for energy storage to enable use of the intermittent PV electricity on the grid.
E. HS-DAC_{coal} (ZeroGHG)	Same as the HS-DAC_{coal} scenario (C), but with increased HS-DAC capacity such that net lifecycle GHG emissions are further reduced to zero (i.e., compensating for 100% stack emissions as well as all background emissions). N.B. to demonstrate the full potential of HS-DAC the amount of CO_2 captured is increased (to 1,27 kg) such that the end-to-end climate change impact is netted to zero (0 kg CO_2-eq).

F. HS-DAC$_{PV}$ (ZeroGHG)	Same as the previous scenario (E), but the electricity demand for HS-DAC is provided by PV. N.B. to demonstrate the full potential of HS-DAC the amount of CO_2 captured is increased (to 0,875 kg) such that the end-to-end climate change impact is netted to zero (0 kg CO_2-eq).
G. MEA-PCC$_{coal}$+ DAC $_{PV}$ (ZeroGHG)	Combination of previous two scenarios (E and F), such that MEA-PCC captures 90% of flue gas CO_2 emissions and HS-DAC all remaining GHG emissions such that the end-to-end climate change impact is netted to zero (0 kg CO_2-eq). N.B. capturing 0,235 kg CO_2 is used to bring the net-system GHG emission back to zero.
H. PV electricity	Delivering 1 kWh of electricity generated by PV (without any capture, see SI chapter 0).

Life cycle inventory (LCI) data

Capture ready power plant

It has been reported that an additional reduction of SO_2, NO_x and particulate matter (PM) emissions of respectively 95%, 2,5% and 50% can be expected when adding post combustion carbon capture to a power plant (without CO_2 capture). Installing additional flue gas cleaning will prevent the carbon capture equipment from being affected (Volkart et al. 2013; Giannoulakis et al. 2014) by SO_2, NO_x and particulate matter (PM). Therefore, in the LCA model the coal combustion process is adjusted to include the additional reduction of NO_x, SO_x and PM emissions. For NO_x and SO_x this results in larger quantities retained and less emitted, while for PM only a reduction in emissions is modelled. The PM that is retained is not accounted for and cut-off, due to lack of information for the electrostatic precipitator (ESP). The additional equipment need is not modelled since all scenarios use the same capture ready power plant and this will not influence the relative results. The changes in environmental emissions are displayed in Table 4.3.

Table 4.3: Difference in selected environmental emissions between basic and capture ready power plant.

	Non capturing	**Capture ready**
SO_x retained, in hard coal flue gas desulphurization	5.11 x 10^{-4} kg	5.69 x 10^{-4} kg
NO_x retained, in SCR	2.11 x 10^{-4} kg	2.13 x 10^{-4} kg
Nitrogen oxides	7.86 x 10^{-5} kg	7.66 x 10^{-5} kg
Sulfur dioxide	6.07 x 10^{-4} kg	3.04 x 10^{-6} kg
Particulates, < 2.5 um	1.82 x 10^{-6} kg	9.1 x 10^{-7} kg
Particulates, > 10 um	5.11 x 10^{-6} kg	2.56 x 10^{-6} kg
Particulates, > 2.5 um, and < 10um	2.14 x 10^{-7} kg	1.07 x 10^{-7} kg

Post combustion CO_2 capture (PCC)

Figure 4.8 shows the unit processes for the construction of the PCC plant and the capture process itself.

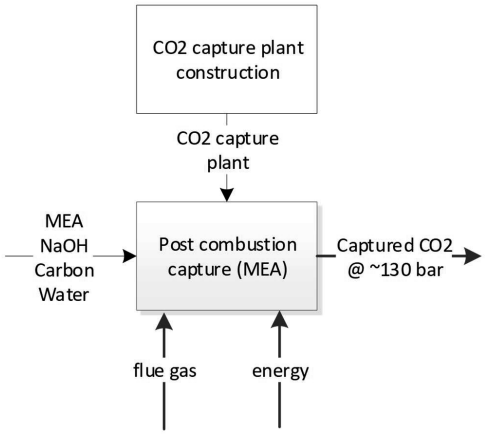


Figure 4.8: Post combustion CO_2 capture process (the 130 bar pressure of the captured CO_2 is based on the pressure needed for subsequent CO_2 transport over 200 km.

Construction of the PCC plant

The size of the capture installation and the amount of required resources depend on the amount and the concentration of CO_2 in the flue gas from which CO_2 is captured. Van der Giesen(Van der Giesen 2008) estimated the steel demand for a PCC installation that captures ~3MT CO_2/year from a PC power plant. The PCC installation is modelled on its expected steel demand, namely 5.560 tonne low –alloyed steel and 1.470 tonne stainless steel. For this study the steel demand is scaled down proportionally to represent a PCC plant that emits 1,5 MT a year. In this study a capture rate of 90% is assumed, hence an annual amount of 0,9 x 1,5 MT CO_2 = 1,35 MT CO_2 is captured. With a life time of 30 years it needs 1 / (1,35 x 10^6 x 30) = 2,46 x 10^{-8} units of PCC installation per tonne CO_2 captured.

The PCC installation does not solely consist of steel but also of other materials such as concrete and other building and installation equipment. In addition, energy has to be used to construct the PCC installation. It can be assumed that the PCC plant is rather similar in size and material mix to the power plant from which the CO_2 is captured. (Van der Giesen 2008) Therefore, in addition to the above derived steel demand, the appropriate LCI data from the coal fired power plant are used to represent the PCC installation. These numbers represent estimates. However, a sensitivity analysis, presented below, shows that these inputs only marginally contribute to the overall LCA impacts of the system.

Construction of the CO_2 compressor

Specific Life cycle inventory data for a CO_2 compressor capable of compressing 1,35 MTonnes per year to a pressure of 130 bar is not available. We roughly based the CO_2 compressor in our model on a MAN 25 MW RG80 compressor(MAN 2014) that is capable of compressing CO_2 from a pressure of 1 bar to 200 bar, with a displacement of ~50 kg /s and weight of 60 tonnes.(MAN Diesel & Turbo) To include the complete life cycle of the compressor we scale all inputs to an available 300 kW air compressor, weighing 4600 kg. A detailed LCA inventory is available for such a compressor in the ecoinvent database(Steiner and Frischknecht 2007). Using the weights of the equipment needed, we multiply the inputs and emissions of the air compressor with a factor 60,000/4,600 = 13.

As a lifetime for the compressor unit we assume 15 years. With a total displacement of 50 kg/s or 50x 60x60x4000=$7.2x10^5$ tonne CO_2/year or 1.1 x 10^7 tonne CO_2 over a life time, the PCC installation thus needs $1/1.1x10^7 = 9.26x10^{-8}$ compressor units / tonne CO_2 for compressing the CO_2. A sensitivity analysis (0) shows that these inputs only marginally contribute to the overall LCA impacts of the system.

Monoethanolamine (MEA) consumption and related emissions

To capture CO_2 from the flue gases, a 30% MEA solution is used (American Physical Society 2011; Volkart et al. 2013; Koiwanit et al. 2013; Singh et al. 2012; Fadeyi et al. 2013; Rochelle et al. 2011). Because of thermal degradation and oxidation of the MEA in the capture process, MEA is consumed and needs to be supplemented (Rochelle 2009; Rochelle et al. 2011). In literature MEA consumption is reported over a range from 0,5 – 3,0 kg MEA / tonne CO_2 (American Physical Society 2011; Volkart et al. 2013; Koiwanit et al. 2013; Singh et al. 2011; Fadeyi et al. 2013; Van der Giesen 2008; Koornneef et al. 2008). We therefore use 1,5 kg MEA / tonne CO_2 captured and carry out a sensitivity analysis presented below.

Due to the degradation of MEA, ammonia forms and is emitted to the atmosphere. Ammonia emissions of 0,136 kg NH_3 / tonne CO_2 (Koiwanit et al. 2013) and 0,035 kg NH_3 / tonne CO_2 have been reported (Fadeyi et al. 2013; Singh et al. 2011). Here we choose to use 0,035 kg NH_3 / tonne CO_2. In addition, MEA itself is emitted to the atmosphere for which values of 0,014 – 0,063 kg MEA / tonne CO_2 have been reported (Corsten et al. 2013). Here we estimate that 0,0385 kg MEA / tonne CO_2 is emitted.

Other PCC related requirements and emissions

Table 4.4 gives an overview of other reported material flows related to PCC that are also taken into account in this study.

Table 4.4: Additional, minor material flows of post-combustion capture (Koiwanit et al. 2013; Singh et al. 2011; Fadeyi et al. 2013).

IN: Sodium hydroxide	0.13 kg NaOH/tonne CO_2 captured
IN: Activated carbon	0.075 kg activated carbon / tonne CO_2 captured
IN: Water	0.8 m3 / tonne CO_2 captured
OUT: Formaldehyde	0.262 g / tonne CO_2 captured
OUT: Acetaldehyde	0.167 g / tonne CO_2 captured

CO_2 pipeline (PCC)

The LCA model is used to obtain the total CO_2 quantity that is captured from the flue gases per year. This number determines the size of the required pipeline. To dimension the pipeline we calculate transport of 1,35 MT CO_2/ year over 200 km. This is an estimated, typical distance for CCS, but a sensitivity analysis in which the distance is doubled shows a very limited change in outcomes of the model. Longer distances will increase the environmental impacts of the CO_2 transport because of larger material demands and energy use. Using McCoy's model (Mccoy 2008; Mccoy and Rubin 2008) the transport of this amount of CO_2 over a distance of 200 km requires a NPS 12 line pipe. His model shows an NPS line pipe with an outer diameter of 0,324 m and an inner diameter of 0,310 m, leaving a wall thickness of 0,007 m. According to the American Petroleum Institute (API) a pipeline with this specifications weighs 109,2 kg/m or 109,2 tonne / km (American Petroleum Institute (API) 2004). This number is used to adjust the ecoinvent process that "pipeline, natural gas, high pressure distribution network" [EI ID1339] that produces pipelines with an assumed lifetime of 40 years. Only the amount of reinforced steel per km pipeline is adjusted according to the numbers provided by APS. With an expected lifetime of 40 years and a transportation quantity of 1,35 MT / year, the transport of 1 tonne CO_2 over 200km needs 200 / ($1{,}35 \times 10^6 \times 40$) = $3{,}7 \times 10^{-6}$ km pipeline / tonne CO_2 transported.

Humidity swing-based direct air capture (HS-DAC)

Inventory data (consumption of water, hardware, and sorbent per CO_2 captured) in this study is derived from previously published laboratory experiments and respective publications (Lackner 2009; Wang et al. 2011, 2013; Lackner et al. 2012; Lackner 2013; Wang et al. 2016). An overview is presented in Table 4.5, the unit process is shown in Figure 4.9, and details are explained and derived in subsequent sections.

Table 4.5: LCI data for HS-DAC [resource demand for CO2 captured and delivered @ 100 bar].

	Cold	Standard	Warm
[RH = relative humidity]	10°C/60%RH	20°C/30%RH	30°C/10%RH
Electricity demand [kWh/tonne CO_2] (see section 1.1)	456	378	382
Sorbent consumption [kg/tonne CO_2]	2.02	1.52	1.52
Water consumption [m^3/tonne CO_2]	20.2	15.2	15.2
Hardware consumption [kg/tonne CO_2]	0.701	0.526	0.527
DAC equipment weight [tonne/1-tonne-a-day unit]	5.12	3.84	3.85
Land use [m^2/1-tonne-a-day unit]	155	160	166

N.B. All calculations, modelling results and graphs in the main document are based on "standard" conditions. Where appropriate, climate differences are taken into account, using the values for "cold" and "warm".

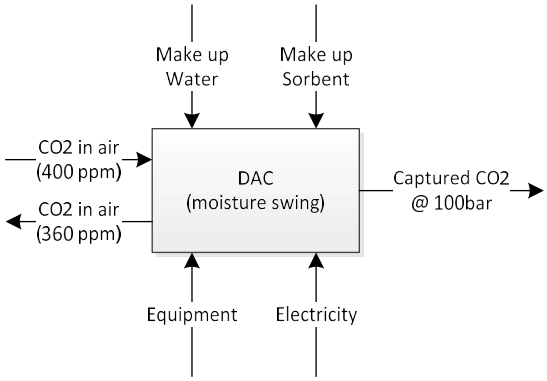


Figure 4.9: Unit process of DAC.

HS-DAC sorbent consumption

The sorbent used is a heterogeneous, quaternary amine, anionic exchange membrane. This sorbent is an approximate 50:50 blend (by mass) of polypropylene plastic and a quaternary anionic exchange resin like Marathon-A as produced by Dow Chemical. Marathon A is a non-hazardous chemical (Avantor Performance Materials 2011) that consists of 50% water and 50% Benzenemethanamium (CAS60177-39-1). Inventory data for Benzenemethanamium chemical is not available and since it can be considered to only have minor hazards (Avantor Performance Materials 2011) it is not taken into account in the LCA (cut-off). Thus, only the polypropylene portion of the HS-DAC sorbent is taken into account. However, a sensitivity analysis using MEA as a proxy for the Benzenemethanamium is available below. The sorbent is re-used and only a small fraction that reaches the end of its lifetime needs to be supplemented. Extrapolating from observations in the laboratory over 10+ years, we estimate a lifetime of the sorbent of 100.000 cycles. At a mass yield of 0.66% per cycle this is equivalent to 1,52 kg sorbent consumed per tonne CO_2 captured.

HS-DAC water consumption

At normal, not further optimized operation (Lackner 2009; Wang et al. 2011, 2013), 0,1kg water is applied per kg sorbent per cycle (to change sorbent kinetics to the 100% humidity regime). This water is subsequently lost to the environment via evaporation. At a mass yield of 0,659% per cycle this is equivalent to 15,2m^3 water per tonne CO_2 captured (= 15,2 tonne of water). Because of the nature of the sorbent, this water does not have to be high-purity. The only restriction is that the water must not have high concentrations of anions such as chloride, sulfates or nitrates because anions would contaminate the resin, reducing its yield over time (Lackner et al. 2012). To remain conservative, in our model we use tap water (higher LCA impacts).

Equipment and materials for HS-DAC

Based on laboratory scale prototypes and tests, a hypothetical industrial scale sized HS-DAC installation is defined here. This installation is capable of capturing 1 tonne of (> =99% pure) CO_2 per day @ 100 bar (see main text for sensitivity analysis to weather conditions). The installation has an assumed lifetime of 20 years (365 days/year). Over its lifetime a HS-DAC installation will capture 20 x 365 x 1 = 7300 tonne CO_2. For 1 tonne CO_2 in the DAC process we need 1 / 7300 = 1,37 x 10^{-4} installations. For capturing larger amounts of CO_2, the installation can be scaled up proportionally. Neither energy nor other consumptions (per tonne CO_2) inherently depend on scaling laws such as surface-to-volume or structural performance and are thus largely independent of a DAC's unit envisioned daily capacity.

The types of materials used and the complexity of manufacturing the HS-DAC installation are assumed similar to that of a passenger vehicle (i.e., steel, advanced plastics, pumps and compressors, electronics, actuators, electric motors, etc.). Here we use inventory data of a passenger vehicle as a proxy for the DAC installation. A 1 tonne-a-day installation weighs 3,84 tonnes (see Table 4.6) and assuming the material composition and distribution of a passenger car we can model the DAC installation (scaled proportionally by weight). In the ecoinvent 2.2 database a Golf A4 passenger car is modelled (EI-ID 1936), this car weighs around 1200 kg (1,2 tonne) (Volkswagen 2014). For modelling the DAC installation we need therefore 3,84/1,2=3,2 of these passenger cars. (5,12/1,2=4,3 for cold weather conditions, 3,85/1,2=3,2 for warm weather conditions).

Table 4.6: DAC installation weight (standard climate 20ºC, 30% relative humidity).

Total sorbent mass in a '1 tonne / day' HS-DAC unit	5.06	tonne
Sorbent enclosures (mass calculated as 10% of sorbent mass) to form filter elements	0.50	tonne
Filter elements (sorbent and enclosures)	**5.56**	**tonne**
Conveyor (calculated as 10% of filter elements)	0.56	tonne
Desorption chamber (calculated as 100% of the filter elements inside desorption chamber), but only 50% of all elements in desorption mode at any given time: 5.56 tonne·50%	2.78	tonne
Total hardware of '1 tonne/day' HS-DAC unit (excluding sorbent)	**3.84**	**tonne**

PV electricity

In this study PV electricity is used to power DAC (DAC PV scenario) and to make up for the electricity lost when applying PCC to a coal fired power plant (PCC PV scenario). This PV electricity is generated by multicrystaline-Si solar cells (panel efficiency = 13,2%, average annual yield = 820 kWh/kW_{peak}) for which the data is taken from the Ecoinvent 2.2. database. Details on this system can be found in ecoinvent report 6-XII (Jungbluth et al. 2007).

Environmental impact assessment

Baseline categories for the CML 2001 impact assessment method(CML-Department of Industrial Ecology 2015) are taken into account in this study. The category scores are displayed in Table 4.7. The impact category climate change is adjusted to include CO_2 captured from the atmosphere, which is usually excluded in the CML2001 impact assessment method. The impact category water depletion is taken from the RECIPE impact assessment method (Goedkoop et al. 2009).

Table 4.7: Impact assessment scores per kWh of electricity delivered to the grid.

Impact Category	Base Case	MEA-PCC$_{coal}$	HS-DAC$_{coal}$	MEA-PCC$_{PV}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DACPV (ZeroGHG)	MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)	PV electricity	Unit
Eutrophication	1.97E-03	2.47E-03	2.70E-03	2.03E-03	2.97E-03	2.05E-03	2.49E-03	1.85E-04	kg PO4-Eq
Abiotic resource depletion	6.48E-03	8.13E-03	8.88E-03	6.64E-03	9.78E-03	6.70E-03	8.19E-03	4.53E-04	kg antimony - Eq
Acidification	1.60E-03	2.07E-03	2.21E-03	1.73E-03	2.44E-03	1.74E-03	2.10E-03	2.98E-04	kg SO_2-Eq
Photochemical oxidation	4.89E-05	6.32E-05	6.85E-05	5.45E-05	7.60E-05	5.71E-05	6.54E-05	1.86E-05	kg ethylene-Eq
Terrestrial ecotoxicity	7.85E-04	1.05E-03	1.11E-03	9.34E-04	1.23E-03	9.76E-04	1.10E-03	4.48E-04	kg 1.4-DCB-Eq
Marine aquatic ecotoxicity	1.04E+03	1.29E+03	1.42E+03	1.07E+03	1.56E+03	1.09E+03	1.31E+03	1.46E+02	kg 1.4-DCB-Eq
Freshwater aaquatic ecotoxicity	2.97E-01	3.72E-01	4.08E-01	3.09E-01	4.49E-01	3.16E-01	3.77E-01	4.56E-02	kg 1.4-DCB-Eq
Stratospheric ozone depletion	6.88E-09	9.28E-09	9.67E-09	1.02E-08	1.07E-08	1.18E-08	1.06E-08	1.38E-08	kg CFC-11-Eq
Human toxicity	2.40E-01	3.56E-01	3.32E-01	3.15E-01	3.66E-01	2.94E-01	3.70E-01	1.46E-01	kg 1.4-DCB-Eq
Climate change	8.48E-01	2.29E-01	2.29E-01	1.97E-01	~0	~0	~0	6.20E-02	kg CO2-Eq
Water depletion	2.07E-03	3.34E-03	1.88E-02	2.75E-03	2.51E-02	1.72E-02	7.40E-03	3.03E-04	m^3

Baseline categories for the CML 2001 impact assessment method(CML-Department of Industrial Ecology 2015) are taken into account in this study. The category scores are displayed in Table 4.7. The impact category climate change is adjusted to include CO_2 captured from the atmosphere, which is usually excluded in the CML2001 impact assessment method. The impact category water depletion is taken from the RECIPE impact assessment method (Goedkoop et al. 2009).

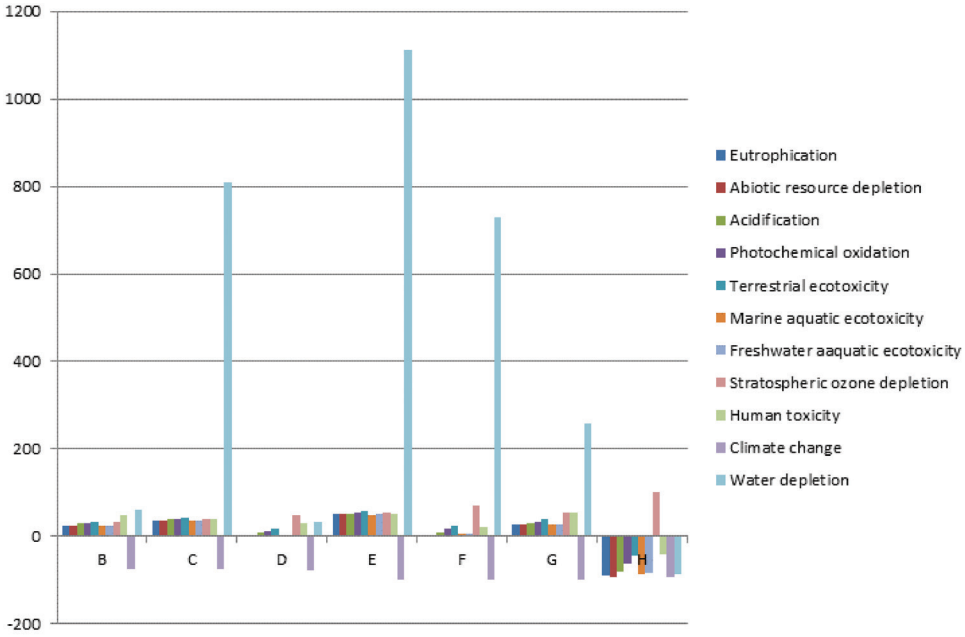


Figure 4.10: LCA results: %Change of impact categories across all scenarios relative to the base case. (B=MEA-PCC$_{coal}$, C=HS-DAC$_{coal}$, D=MEA-PCC$_{PV}$, E=HS-DAC$_{coal}$(ZeroGHG), F=HS-DAC$_{PV}$ (ZeroGHG), G=MEA-PCC$_{c\ al}$+HS-DAC$_{PV}$, H= PV electricity).

Figure 4.10 shows an overview of all impacts in relation to the base case and allows for direct comparison of the average increase in non-GHG impacts with the net life cycle GHG emissions. The numbers for Figure 4.10 LCA results: %Change of impact categories across all scenarios relative to the base case.

Table 4.8: LCA results: %Change of impact categories across all scenarios relative to the base case.

Impact Category	Base Case	MEA-PCC$_{coal}$	HS-DAC$_{coal}$	MEA-PCC$_{PV}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)	PV electricity
Eutrophication	0	25	37	3	51	4	26	-91
Abiotic resource depletion	0	25	37	2	51	3	26	-93
Acidification	0	29	38	8	53	9	31	-81
Photochemical oxidation	0	29	40	11	55	17	34	-62
Terrestrial ecotoxicity	0	34	41	19	57	24	40	-43
Marine aquatic ecotoxicity	0	24	37	3	50	5	26	-86
Freshwater aquatic ecotoxicity	0	25	37	4	51	6	27	-85
Stratospheric ozone depletion	0	35	41	48	56	72	54	101
Human toxicity	0	48	38	31	53	23	54	-39
Climate change	0	-73	-73	-77	-100	-100	-100	-93
Water depletion	0	61	808	33	1113	731	257	-85

Table 4.9: Change in Impact assessment scores per net kg CO_2-eq avoided (base case equals zero)
Impact score per kg CO_2 avoided = ($Impact_{scenario}$ – $Impact_{basecase}$)/($GHG_{basecase}$-$GHG_{scenario}$).

Impact Category	MEA-PCC$_{coal}$	HS-DAC$_{coal}$	MEA-PCC$_{PV}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)	PV electricity	
Eutrophication	8.08E-04	1.18E-03	9.22E-05	1.18E-03	9.43E-05	6.13E-04	-2.27E-03	kg PO4-Eq / kg CO_2-eq avoided
Abiotic resource	2.67E-03	3.88E-03	2.46E-04	3.89E-03	2.59E-04	2.02E-03	-7.67E-03	kg antimony-Eq/kg CO_2-eq avoided
Acidification	7.59E-04	9.85E-04	2.00E-04	9.91E-04	1.65E-04	5.90E-04	-1.66E-03	kg SO2-Eq/kg CO_2-eq avoided
Photochemical oxidation	2.31E-05	3.17E-05	8.60E-06	3.20E-05	9.67E-06	1.95E-05	-3.85E-05	kg ethylene-Eq/kg CO_2-eq avoided
Terrestrial ecotoxicity	4.28E-04	5.25E-04	2.29E-04	5.25E-04	2.25E-04	3.71E-04	-4.29E-04	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Marine aquatic ecotoxicity	4.04E+02	6.14E+02	4.61E+01	6.13E+02	5.90E+01	3.18E+02	-1.14E+03	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Freshwater aquatic ecotoxicity	1.21E-01	1.79E-01	1.84E-02	1.79E-01	2.24E-02	9.43E-02	-3.20E-01	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Stratospheric ozone	3.88E-09	4.51E-09	5.10E-09	4.50E-09	5.80E-09	4.39E-09	8.80E-09	kg CFC-11-Eq/kg CO_2-eq avoided
Human toxicity	1.87E-01	1.49E-01	1.15E-01	1.49E-01	6.37E-02	1.53E-01	-1.20E-01	kg 1,4-DCB-Eq/kg CO_2-eq avoided
Climate change	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	-1.00E+00	kg CO_2-Eq/kg CO_2-eq avoided
Water depletion	2.05E-03	2.70E-02	1.04E-03	2.72E-02	1.78E-02	6.29E-03	-2.25E-03	m3/kg CO_2-eq avoided

Sensitivity analyses

In addition to the sensitivity analyses reported above (MEA-PCC: current/likely versus future/best case; HS-DAC: effect of temperature and relative humidity) a series of further sensitivity analyses were performed to assess the effect of any uncertainties in our inventory data on the LCA results and the robustness of our conclusions. We found that our conclusions remain valid even when varying inventory data within their respective uncertainties based on ranges reported in the literature. Details are given below.

CO_2 compressor use in post combustion capture

To examine the sensitivity of the model to the CO_2 compressor hardware used in the PCC installation we doubled the inputs of this compressor to the PCC process from $9{,}26 \times 10^{-8}$ compressor / ton CO_2 captured and compressed to $1{,}85 \times 10^{-7}$ compressor / ton CO_2 captured and compressed. This change in inputs shows no noteworthy change (<0,1%) in the outcome of our model (Table 4.10).

Table 4.10: Change in impact scores for sensitivity to CO_2 compressor hardware.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00247	0.00203	100.0%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00813	0.00664	100.0%	100.0%
Acidification	0.00207	0.00173	0.00207	0.00173	100.0%	100.0%
Photochemical oxidation	6.32E-05	5.45E-05	6.32E-05	5.45E-05	100.0%	100.0%
Terrestrial ecotoxicity	0.00105	0.000934	0.00105	0.000935	100.0%	100.1%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.29E+03	1.07E+03	100.0%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.372	0.309	100.0%	100.0%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.28E-09	1.02E-08	100.0%	100.0%
Human toxicity	0.356	0.315	0.356	0.315	100.0%	100.0%
Climate change	0.229	0.197	0.229	0.197	100.0%	100.0%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

Physical size of the PCC plant

The capture of 1 tonne of CO_2 with PCC requires $2{,}46 \times 10^{-8}$ PCC plants. This number is based on estimations of the materials required. Here we check the sensitivity of our LCA model with regard to this parameter by increasing its value by 10% to $1{,}1 \times 2{,}46 \times 10^{-8} = 2{,}71 \times 10^{-8}$ and checking how much the outcomes of the model will change. The model does not show sensitivity to changes in this parameter.

To examine the sensitivity of the model to the PCC plant CO_2 we doubled the inputs of the PCC plant to the PCC process from $2{,}46 \times 10^{-8}$ PCC installation/ton CO_2 captured to $4{,}92 \times 10^{-8}$. This change in inputs results in an only very minor change in the

outcome of our model (Table 4.11).

Table 4.11: Change in impact scores for sensitivity to PCC installation.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00248	0.00203	100.4%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00815	0.00665	100.2%	100.2%
Acidification	0.00207	0.00173	0.00208	0.00173	100.5%	100.0%
Photochemical oxidation	6.32E-05	5.45E-05	6.42E-05	5.53E-05	101.6%	101.5%
Terrestrial ecotoxicity	0.00105	0.000934	0.00108	0.000961	102.9%	102.9%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.30E+03	1.07E+03	100.8%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.374	0.31	100.5%	100.3%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.49E-09	1.03E-08	102.3%	101.0%
Human toxicity	0.356	0.315	0.36	0.318	101.1%	101.0%
Climate change	0.229	0.197	0.232	0.199	101.3%	101.0%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

CO_2 pipeline transport distance

With an expected lifetime of 40 years and a transportation quantity of 1,35 MT / year. The transport of 1 tonne CO_2 over 200km needs 200 / (1,35 x10^6 x 40) = 3,7 x 10^{-6} km pipeline / tonne CO_2 transported. To examine the sensitivity of our model we doubled the input of this parameter to 7,4 x 10^{-6} km pipeline / tonne CO_2 transported. This change in inputs shows no noteworthy change in the outcome of our model (Table 4.12).

Table 4.12: Change in impact scores for sensitivity to CO_2 pipeline transportation distance.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00247	0.00203	100.0%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00813	0.00664	100.0%	100.0%
Acidification	0.00207	0.00173	0.00207	0.00173	100.0%	100.0%
Photochemical oxidation	6.32E-05	5.45E-05	6.35E-05	5.48E-05	100.5%	100.6%
Terrestrial ecotoxicity	0.00105	0.000934	0.00106	0.000942	101.0%	100.9%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.29E+03	1.07E+03	100.0%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.372	0.309	100.0%	100.0%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.32E-09	1.02E-08	100.4%	100.0%
Human toxicity	0.356	0.315	0.356	0.315	100.0%	100.0%
Climate change	0.229	0.197	0.23	0.197	100.4%	100.0%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

MEA consumption

In literature MEA consumptions in the range of 0,5-3 kg / tonne CO_2 have been reported (see above). The MEA consumption in the PCC scenarios is set to 1,5 kg MEA / tonne CO_2. To examine the sensitivity of our model we doubled the input of this parameter to 3 kg MEA / tonne CO_2. This change in inputs results in only a negligible change in the outcome of our model (Table 4.13).

Table 4.13: Change in impact scores for sensitivity to MEA consumption.

	Original scores		New scores		New/Old	
	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}	MEA-PCC_{coal}	MEA-PCC_{PV}
Eutrophication	0.00247	0.00203	0.00248	0.00203	100.4%	100.0%
Abiotic resource depletion	0.00813	0.00664	0.00818	0.00668	100.6%	100.6%
Acidification	0.00207	0.00173	0.00209	0.00174	101.0%	100.6%
Photochemical oxidation	6.32E-05	5.45E-05	6.41E-05	5.53E-05	101.4%	101.5%
Terrestrial ecotoxicity	0.00105	0.000934	0.00108	0.000958	102.9%	102.6%
Marine aquatic ecotoxicity	1.29E+03	1.07E+03	1.30E+03	1.07E+03	100.8%	100.0%
Freshwater aquatic ecotoxicity	0.372	0.309	0.373	0.309	100.3%	100.0%
Stratospheric ozone depletion	9.28E-09	1.02E-08	9.69E-09	1.05E-08	104.4%	102.9%
Human toxicity	0.356	0.315	0.408	0.357	114.6%	113.3%
Climate change	0.229	0.197	0.234	0.2	102.2%	101.5%
Water depletion	0.00334	0.00275	0.00334	0.00275	100.0%	100.0%

Hardware requirements of a HS-DAC unit

The actual equipment weight of DAC units has considerable uncertainty because it has been estimated based on laboratory prototypes. The inventory of a passenger car is used as a proxy to model the DAC installation and based on the weight of equipment needed assumptions are made. For the capture of 1 tonne CO_2 from the atmosphere we need 1,37 x 10^{-4} DAC installation. To examine the sensitivity of our model we doubled the input of this parameter to 2,74 x 10^{-4}.

This change in inputs results in no noteworthy change in the outcome of our model (Table 4.14).

HS-DAC sorbent composition

To examine the sensitivity of our model to the cut-off assumption for Benzenemethanamium (see above) we used MEA as a proxy for Benzenemethanamium (1:1 mass ratio).

This change in inputs results in no noteworthy change in the outcome of our model (Table 4.15).

HS-DAC sorbent amount consumed

As average consumption 1,5 kg HS-DAC sorbent per tonne CO_2 captured is modelled. To examine the sensitivity of our model we doubled the input of this parameter to 3 kg sorbent per tonne CO_2 captured.

This change in inputs results in no noteworthy change in the outcome of our model (Table 4.16)

Water use of HS-DAC

The HS-DAC processes consumes 15,2 m^3 water / tonne CO_2 captured. As shown in the impact assessment the water depletion of DAC is a factor 9 higher than for PCC. Water demand therefore seems an important parameter in our model. To examine the sensitivity of our model we doubled the input of this parameter to 30,4 m^3 water per tonne CO_2 captured.

As expected, this change in inputs results in a large change in the outcome of our model for water depletion (but only minimal change for all other environmental impacts). When the input is doubled, the life cycle water depletion changes by 50 – 90% (Table 4.17).

Table 4.14: Change in impact scores for sensitivity to DAC installation size.

	Original scores				New scores				New / Old			
Impact Category	**$HS\text{-}DAC_{coal}$**	**$HS\text{-}DAC_{coal}$ (ZeroGHG)**	**$HS\text{-}DAC_{PV}$ (ZeroGHG)**	**$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)**	**$HS\text{-}DAC_{coal}$**	**$HS\text{-}DAC_{coal}$ (ZeroGHG)**	**$HS\text{-}DAC_{PV}$ (ZeroGHG)**	**$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)**	**$HS\text{-}DAC_{coal}$**	**$HS\text{-}DAC_{coal}$ (ZeroGHG)**	**$HS\text{-}DAC_{PV}$ (ZeroGHG)**	**$MEA\text{-}PCC_{coal}+DAC_{PV}$ (ZeroGHG)**
Eutrophication	2.70E-03	2.97E-03	2.05E-03	2.49E-03	2.70E-03	2.97E-03	2.05E-03	2.49E-03	100.0%	100.0%	100.0%	100.0%
Abiotic resource depletion	8.88E-03	9.78E-03	6.70E-03	8.19E-03	8.89E-03	9.80E-03	6.71E-03	8.19E-03	100.1%	100.2%	100.1%	100.0%
Acidification	2.21E-03	2.44E-03	1.74E-03	2.10E-03	2.23E-03	2.46E-03	1.75E-03	2.11E-03	100.9%	100.8%	100.6%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.93E-05	7.70E-05	5.78E-05	6.56E-05	101.2%	101.3%	101.2%	100.3%
Terrestrial ecotoxicity	1.11E-03	1.23E-03	9.76E-04	1.10E-03	1.13E-03	1.26E-03	9.94E-04	1.11E-03	101.8%	102.4%	101.8%	100.9%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.57E+03	1.10E+03	1.31E+03	100.0%	100.6%	100.9%	100.0%
Freshwater aquatic	4.08E-01	4.49E-01	3.16E-01	3.77E-01	4.09E-01	4.51E-01	3.17E-01	3.77E-01	100.2%	100.4%	100.3%	100.0%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.77E-09	1.09E-08	1.18E-08	1.06E-08	101.0%	101.9%	100.0%	100.0%
Human toxicity	3.32E-01	3.66E-01	2.94E-01	3.70E-01	3.35E-01	3.70E-01	2.96E-01	3.71E-01	100.9%	101.1%	100.7%	100.3%
Climate change	2.29E-01	~0	~0	~0	2.30E-01	~0	~0	~0	100.4%	x	x	x
Water depletion	1.88E-02	2.51E-02	1.72E-02	7.40E-03	1.88E-02	2.51E-02	1.72E-02	7.41E-03	100.0%	100.0%	100.0%	100.1%

Table 4.15: Change in impact scores for sensitivity to DAC sorbent composition.

Impact Category	Original scores				New scores				New / Old			
	HS-DAC_{coal}	HS-DAC_{coal} (ZeroGHG)	HS-DAC_{PV} (ZeroGHG)	MEA-PCC_{coal}+DAC_{PV} (ZeroGHG)	HS-DAC_{coal}	HS-DAC_{coal} (ZeroGHG)	HS-DAC_{PV} (ZeroGHG)	MEA-PCC_{coal}+DAC_{PV} (ZeroGHG)	HS-DAC_{coal}	HS-DAC_{coal} (ZeroGHG)	HS-DAC_{PV} (ZeroGHG)	MEA-PCC_{coal}+DAC_{PV} (ZeroGHG)
Eutrophication	0.0027	0.00297	0.00205	0.00249	0.0027	0.00297	0.00205	0.00249	100.0%	100.0%	100.0%	100.0%
Abiotic resource depletion	0.00888	0.00978	0.0067	0.00819	0.00889	0.0098	0.00671	0.00819	100.1%	100.2%	100.1%	100.0%
Acidification	0.00221	0.00244	0.00174	0.0021	0.00222	0.00245	0.00174	0.00211	100.5%	100.4%	100.0%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.88E-05	7.63E-05	5.73E-05	6.54E-05	100.4%	100.4%	100.4%	100.0%
Terrestrial ecotoxicity	0.00111	0.00123	0.000976	0.0011	0.00112	0.00125	0.000983	0.00111	100.9%	101.6%	100.7%	100.9%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.56E+03	1.10E+03	1.31E+03	100.0%	100.0%	100.9%	100.0%
Freshwater aquatic	0.408	0.449	0.316	0.377	0.408	0.45	0.316	0.377	100.0%	100.2%	100.0%	100.0%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.79E-09	1.09E-08	1.19E-08	1.06E-08	101.2%	101.9%	100.8%	100.0%
Human toxicity	0.332	0.366	0.294	0.37	0.347	0.387	0.308	0.374	104.5%	105.7%	104.8%	101.1%
Climate change	0.229	~0	~0	~0	0.23	~0	~0	~0	100.4%	x	x	x
Water depletion	0.0188	0.0251	0.0172	0.0074	0.0188	0.0251	0.0172	0.0074	100.0%	100.0%	100.0%	100.0%

Table 4.16: Change in impact scores for sensitivity to DAC sorbent demand.

Impact Category	Original scores				New scores				New / Old			
	HS-DAC$_{coal}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$+DAC$_{PV}$ (ZeroGHG)	HS-DAC$_{coal}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$+DAC$_{PV}$ (ZeroGHG)	HS-DAC$_{coal}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$+DAC$_{PV}$ (ZeroGHG)
Eutrophication	0.0027	0.00297	0.00205	0.00249	0.0027	0.00297	0.00205	0.00249	100.0%	100.0%	100.0%	100.0%
Abiotic resource depletion	0.00888	0.00978	0.0067	0.00819	0.0089	0.00981	0.00672	0.00819	100.2%	100.3%	100.3%	100.0%
Acidification	0.00221	0.00244	0.00174	0.0021	0.00222	0.00245	0.00174	0.00211	100.5%	100.4%	100.0%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.88E-05	7.64E-05	5.73E-05	6.55E-05	100.4%	100.5%	100.4%	100.2%
Terrestrial ecotoxicity	0.00111	0.00123	0.000976	0.0011	0.00111	0.00123	0.000976	0.0011	100.0%	100.0%	100.0%	100.0%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.56E+03	1.09E+03	1.31E+03	100.0%	100.0%	100.0%	100.0%
Freshwater aquatic	0.408	0.449	0.316	0.377	0.408	0.449	0.316	0.377	100.0%	100.0%	100.0%	100.0%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.67E-09	1.07E-08	1.18E-08	1.06E-08	100.0%	100.0%	100.0%	100.0%
Human toxicity	0.332	0.366	0.294	0.37	0.332	0.366	0.294	0.37	100.0%	100.0%	100.0%	100.0%
Climate change	0.229	~0	~0	~0	0.23	~0	~0	~0	100.4%	x	x	x
Water depletion	0.0188	0.0251	0.0172	0.0074	0.0188	0.0251	0.0172	0.0074	100.0%	100.0%	100.0%	100.0%

Table 4.17: Change in impact scores for sensitivity to DAC water demand.

Impact Category	Original scores				New scores				New / Old			
	HS-DAC$_{coal}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$+DAC$_{PV}$ (ZeroGHG)	HS-DAC$_{coal}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$+DAC$_{PV}$ (ZeroGHG)	HS-DAC$_{coal}$	HS-DAC$_{coal}$ (ZeroGHG)	HS-DAC$_{PV}$ (ZeroGHG)	MEA-PCC$_{coal}$+DAC$_{PV}$ (ZeroGHG)
Eutrophication	0.0027	0.00297	0.00205	0.00249	0.00271	0.00299	0.00206	0.0025	100.4%	100.7%	100.5%	100.4%
Abiotic resource depletion	0.00888	0.00978	0.0067	0.00819	0.00891	0.00982	0.00673	0.00819	100.3%	100.4%	100.4%	100.0%
Acidification	0.00221	0.00244	0.00174	0.0021	0.00223	0.00247	0.00175	0.00211	100.9%	101.2%	100.6%	100.5%
Photochemical oxidation	6.85E-05	7.60E-05	5.71E-05	6.54E-05	6.96E-05	7.74E-05	5.81E-05	6.57E-05	101.6%	101.8%	101.8%	100.5%
Terrestrial ecotoxicity	0.00111	0.00123	0.000976	0.0011	0.00114	0.00127	0.001	0.00111	102.7%	103.3%	102.5%	100.9%
Marine aquatic ecotoxicity	1.42E+03	1.56E+03	1.09E+03	1.31E+03	1.42E+03	1.57E+03	1.10E+03	1.31E+03	100.0%	100.6%	100.9%	100.0%
Freshwater aquatic	0.408	0.449	0.316	0.377	0.41	0.453	0.318	0.378	100.5%	100.9%	100.6%	100.3%
Stratospheric ozone depletion	9.67E-09	1.07E-08	1.18E-08	1.06E-08	9.90E-09	1.10E-08	1.20E-08	1.06E-08	102.4%	102.8%	101.7%	100.0%
Human toxicity	0.332	0.366	0.294	0.37	0.334	0.37	0.296	0.371	100.6%	101.1%	100.7%	100.3%
Climate change	0.229	~0	~0	~0	0.233	~0	~0	~0	101.7%	x	x	x
Water depletion	0.0188	0.0251	0.0172	0.0074	0.0348	0.0471	0.0323	0.0114	185.1%	187.6%	187.8%	154.1%

Impact of weather conditions on HS-DAC performance

Table 4.18 presents operational parameters for HS-DAC in different weather conditions. Running the LCA model with these parameters shows that under cold and humid (CH) conditions HS-DAC is likely to perform worse than under standard (S) or warm and dry (WD) conditions. There difference between standard and warm dry conditions is minimal (see Table 4.19).

Table 4.18: Operational parameters of HS-DAC under different weather conditions (RH=relative humidity).

Operation mode	**CH**	**S**	**WD**
	Cold / Humid (10°C/60% RH)	**Standard (20°C/30% RH)**	**Warm / Dry (30°C/10% RH)**
Max. sorbent loading (determined by ambient conditions. see section 0)	0.876	0.968	0.985
Min. sorbent loading (tunable)	0.750	0.800	0.817
--> Sorbent mass yield [CO_2 per sorbent]	0.49%	0.66%	0.66%
Sorbent consumption [per tonne CO_2]	2.02 kg	1.52 kg	1.52 kg
Electricity consumption [per tonne CO_2]	456 kWh	378 kWh	382 kWh
Water consumption [per tonne CO_2]	20.2 m^3	15.2 m^3	15.2 m^3
Hardware excl. sorbent [per "1tonne a day" machine] [8]	5.12 tonnes	3.84 tonnes	3.85 tonnes
Hardware consumption [per tonne CO_2]	0.701 kg	0.526 kg	0.527 kg
Land-use [per "1tonne a day" machine]	155 m^2	160 m^2	166 m^2

Table 4.19: Impacts under different weather conditions (Cold. Standard and Warm) for different alternatives.

	MEA-PCC$_{coal}$	**HS-DAC$_{coal}$**	**MEA-PCC$_{PV}$**	**HS-DAC$_{coal}$ (ZeroGHG)**	**HS-DAC$_{PV}$ (ZeroGHG)**	**MEA-PCC$_{coal}$ +DAC$_{PV}$ (ZeroGHG)**	**PV electricity**
COLD							
Eutrophication	25	50	3	68	5	27	-91
Abiotic resource depletion	25	50	2	68	4	27	-93
Acidification	29	52	8	71	11	32	-81
Photochemical oxidation	29	55	11	75	21	35	-62
Terrestrial ecotoxicity	34	57	19	78	30	43	-43
Marine aquatic ecotoxicity	24	49	3	67	7	26	-86
Freshwater aquatic ecotoxicity	25	51	4	69	8	27	-85
Stratospheric ozone depletion	35	56	48	76	86	58	101
Human toxicity	48	52	31	71	28	55	-39
Climate change	-73	-73	-77	-100	-100	-100	-93
Water depletion	61	1195	33	1634	977	322	-85

STANDARD							
Eutrophication	25	37	3	51	4	26	-91
Abiotic resource depletion	25	37	2	51	3	26	-93
Acidification	29	38	8	53	9	31	-81
Photochemical oxidation	29	40	11	55	17	34	-62
Terrestrial ecotoxicity	34	41	19	57	24	40	-43
Marine aquatic ecotoxicity	24	37	3	50	5	26	-86
Freshwater aquatic ecotoxicity	25	37	4	51	6	27	-85
Stratospheric ozone depletion	35	41	48	56	72	54	101
Human toxicity	48	38	31	53	23	54	-39
Climate change	-73	-73	-77	-100	-100	-100	-93
Water depletion	61	808	33	1113	731	257	-85
WARM							
Eutrophication	25	38	3	51	4	26	-91
Abiotic resource depletion	25	38	2	51	3	26	-93
Acidification	29	39	8	53	9	32	-81
Photochemical oxidation	29	41	11	55	17	34	-62
Terrestrial ecotoxicity	34	43	19	58	25	40	-43
Marine aquatic ecotoxicity	24	37	3	50	6	26	-86
Freshwater aquatic ecotoxicity	25	38	4	52	6	27	-85
Stratospheric ozone depletion	35	41	48	56	72	54	101
Human toxicity	48	39	31	53	23	54	-39
Climate change	-73	-73	-77	-99	-101	-100	-93
Water depletion	61	813	33	1103	736	257	-85

In the different alternatives the following values change due to differing weather conditions (see also Table 4.2).

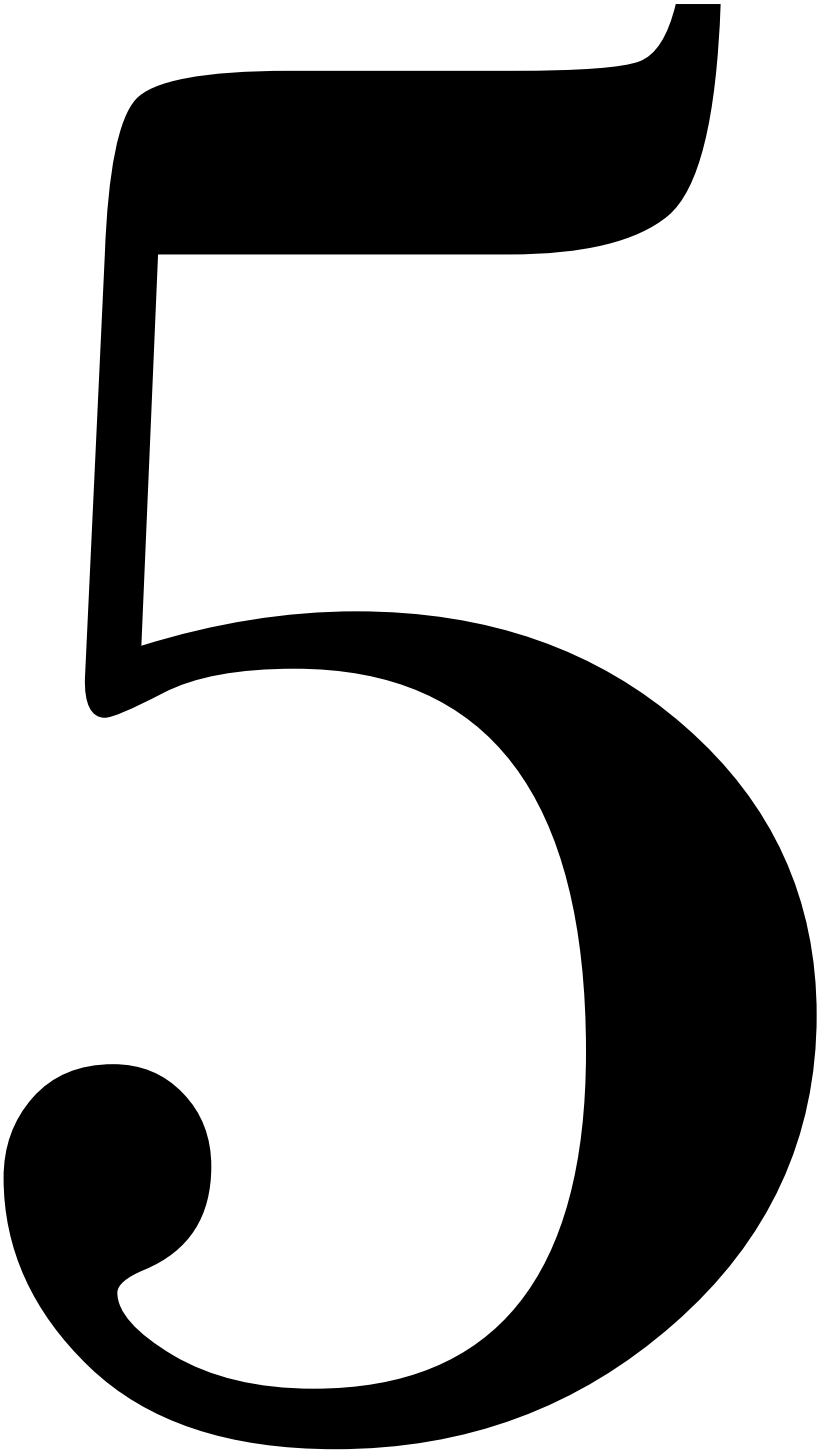
Table 4.20: LCA model lay-out for different scenarios.

	Cold weather scenario	**Warm weather scenario**
HS-DAC_{coal}	captures 1.039 kg CO_2 to have same climate change (CC) impacts as PCC	captures 0.935 kg CO_2 to have same CC impacts as PCC
HS-DACcoal (ZeroGHG)	captures 1.42 kg CO_2 to have no net CC impacts	captures 1.27 kg CO_2 to have no net CC impacts
HS-DAC_{PV} (ZeroGHG)	captures 0.88 kg CO_2 to have no net CC impacts	captures 0.875 kg CO_2 to have no net CC impacts
MEA-PCC_{coal}+DAC_{PV} (ZeroGHG)	captures 0.235 kg CO_2 to have no net CC impacts	captures 0.235 kg CO_2 to have no net CC impacts

4.5.3. Alternative scenario: Changing coal based to PV electricity

While outside the scope of our work, a logical question to ask with respect to the study is what the environmental impact would be if coal-based electricity generation were replaced 1:1 by PV electricity (without any capture). Based on ecoinvent data(Jungbluth et al. 2007) representing multi-crystalline photovoltaic electricity generation we find that replacing coal-based electricity with PV electricity on average reduces non-GHG emissions by over 50% while the GHG-emissions are reduced by over 90%.

It needs to be taken into account here that this calculation assumes only the PV installations. Any required electricity storage to ensure non-intermittent supply is not taken into account.



Chapter 5

A Critical View on the Current Application of LCA for new Technologies and Recommendations for Improved Practice

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Abstract

LCA is a well-known assessment tool that identifies and provides insights on the environmental impacts of products and services over their lifecycle. The guidance provided by the existing manuals typically applies to modelling and assessing environmental impacts ex-post, meaning that information is available from empirical experience after products have been commercially in use for extended periods of time. This information is not available if LCA is applied in an ex-ante manner before a technology is commercially deployed at scale. We identify the major challenges of applying LCA in an ex-ante manner and propose a route forward in dealing with these challenges that combines intuitions from other disciplinary fields. The first challenge is how to model consistent future foreground systems for the incumbent and new technology systems. Learning curves and scenario approaches are the way forward. The second challenge is how to model future background systems. Here a solution is to transform existing LCI databases towards future contexts, informed by the Integrated Assessment Models (IAMs) that provide scenarios in line with the Shared Socioeconomic Pathways (SSPs). Finally, uncertainty in ex-ante LCA is of a different nature as in ex-post LCAs. The main difference with conventional LCA studies is the highly uncertain information for the future. To acknowledge this, considerable attention should be attributed to the discussion on these uncertainties, both in the design of the assessment and the data used. Responsive evaluation can play a supportive role here. This will increase the transparency and efficacy of the results because the relevant stakeholders and experts are involved. In this way technology designers and other stakeholders derive insights on the influence of design choices or contextual factors (that are important, but hard to influence) on the potential environmental impacts of their foreseen technology.

5.1 Introduction

Research, development, deployment and widespread diffusion of new environmentally sound technologies is a major route towards achieving sustainability (United Nations 2017). To support the research and development of claimed environmentally sustainable technologies, international research frameworks such as the European Horizon 2020 program (European Commission 2017) demand the application of quantitative methods, such as life cycle assessment (LCA) (Wender et al. 2014b).

LCA is a well-known policy support tool that identifies the environmental impacts of products and services over their lifecycle (Hellweg and Milà i Canals 2014). The consolidated practice of conducting LCA studies for innumerable product systems for over three decades also makes LCA the preferred option for assessing the potential environmental impacts of new technologies.

The execution of conventional LCAs is guided by the ISO 14040-14044 standards (International Organization for Standardization (ISO) 1997) for which practical guidance is given in various LCA manuals (European Commission - Joint Research Centre - Institute for Environment and Sustainability 2010; Guinée et al. 2002; Curran 2012; Baumann and Tillman 2004). The guidance provided by the existing manuals has been typically applied to modelling and assessing environmental impacts ex-post, meaning after products have been commercially in use for extended periods of time and information and data are available from empirical experience. The ISO standard could and should also be used when LCA is applied in an ex-ante manner. Ex-ante is defined as before a product or technology is commercially deployed at scale (Tecchio et al. 2016) and information and insights on the topic under assessment are not (yet) readily available. Using LCA in an ex-ante manner should allow anticipating potential avoidable environmental impacts as well as avoid environmental lock-ins. It promises the assessment of the potential environmental impacts of a technology at an early stage of its development curve when little information is available and greater and less costly opportunities to change can still be gauged. Guided by the results of LCA studies, technology developers can take appropriate action at an early stage of the development of a technology to prevent investments in those technologies that will eventually prove to have a higher environmental impact.

Performing the ex-ante exercise introduces additional challenges. These include the lack of representative information for the product systems under study, the lack of a clear vision into the future of the technological landscape in which the technology will operate, and the lack of direct access to representative data for lab-scale processes. Some of these challenges have been identified long ago. Frischknecht et al. (2009) questioned the fitness of standard LCA to assess the potential impacts of future technologies already

in 2009,. More recent reviews of the challenges of using LCA to inform early research decisions are also available now (Hetherington et al. 2014; Arvidsson et al. 2018; Cucurachi et al. 2018).

Despite the existing efforts in the literature to categorize studies, there are no structured guidelines to perform ex-ante LCA which link the ex-ante challenges to the classical ISO framework for LCA (International Organization for Standardization (ISO) 1997). While an update of methods and concepts is necessary, the framework and the standard phases of LCA (i.e. goal and scope definition, inventory, impact assessment, interpretation) can be enriched with additional guidance for doing ex-ante LCA. Our work complements the framework of Buyle et al. (Buyle et al. 2019), which reviewed ex-ante LCA studies with a focus on technology development, technological learning and technology diffusion.

Hence, with this paper we aim to integrate the challenges and recommendations for ex-ante LCA identified in the literature into the ISO LCA structure in order to provide guidance for the execution of ex-ante LCA studies. This research is built up around the following research questions:

- How can ex-ante LCA be defined and positioned in other forward-looking approaches to LCA (see section 5.2)?
- Which challenges in ex-ante LCA can we identify in case studies and by deductive reasoning that are different from ex-post LCA practices (see section 5.3)?
- What solutions can be thought of to deal with these challenges (see section 5.4)?
- And concluding: what additional guidance can be provided on this basis when performing an ex-ante LCA study according to the ISO guidelines (see section 5.5)?

5.2 Defining ex-ante LCA

A growing number of studies apply LCA to new, emerging or future technologies and product systems. A variety of approaches and modes of conducting forward-looking LCA that can be applied for these assessments is reported in the literature. An overview and summary of these approaches can be found in Table 5.1.

Table 5.1: Selected literature on forward-looking LCA, adapted from Cucurachi et al. (2018).

Type of assessment	Definitions or descriptions given in literature
Consequential LCA	Consequential LCA provides understanding on "the potential effects of policies on market responses to support environmental decision making" (Kätelhön et al. 2016). "[T]o provide information on the environmental burdens that occur directly or indirectly as a consequence of a decision (usually represented by changes in demand for a product)" (Guinée et al. 2018).
Lifecycle sustainability analysis (LCSA)	Life cycle sustainability analysis proposes a transdisciplinary framework of methods and in principle broadens the assessment to include social and economic impacts, and deepens to include more than just technological relations e.g. by scaling up the technology to society wide implementation. This may imply making future scenarios, but not necessarily looking into innovative technologies (Guinée et al. 2011; Van Der Giesen et al. 2013; Hu et al. 2013).
Dynamic LCA	"The analysis of individual technologies must consider the extremely dynamic development. This concerns the development of products and their production processes as well as their technical performance and the development of background systems." (Pehnt 2006) This approach focusses on including the dynamics of parameters that are expected to change over time and to compare different development pathways (Alfaro et al. 2010).
Anticipatory LCA	Takes a forward-looking (not retrospective assessment) and engages stakeholders to inform critical modelling decisions and increase credibility and relevance of results. Anticipatory LCA can be defined as "non-predictive and inclusive of uncertainty, which can be used to explore both reasonable and extreme-case scenarios of future environmental burdens associated with an emerging technology." The aim is to identify the most relevant uncertainties and engaging research and development decision makers to guide research and development and innovation. (Wender et al. 2014a).
Prospective LCA	"An LCA is prospective when the (emerging) technology studied is in an early phase of development (e.g. small-scale production), but the technology is modelled at a future, more-developed phase (e.g. large-scale production)" (Arvidsson et al. 2018). Prospective LCA integrates forecasting methods in its approach (Hummen and Kastner 2014). See also (Mendoza Beltran et al. 2018; Miller and Keoleian 2015)
Ex-Ante LCA	In ex-ante LCA an environmental analysis of a technology that is typically still in its R&D phase is performed (Schrijvers et al. 2014; Hesser et al. 2017b; Tecchio et al. 2016) to guide R&D. Villares et al. (2016, 2017) stress the ex-ante application of LCA, meaning before (ex-ante) market introduction (Roes and Patel 2011)

Consequential LCA's main focus is on quantifying the potential environmental impacts that accompany a change in policy. LCSA tries to include all dimensions of sustainability since next to environmental impacts, the introduction of a new technology will also have a considerate impact on the economy and society. Dynamic LCA tries to take into account that the world is dynamic and development will most likely choose a different path than one can anticipate at the start of development. Anticipatory LCA stresses that conventional LCA is always performed with hindsight and is not easily attributed to assess future developments, and in addition states that stakeholders should be included in the process to obtain more valuable results. Prospective LCA and ex-ante LCA seem to be similar labels for the same exercise and might be seen as umbrella terms under which practices from other approaches could be used.

Table 5.1 illustrates that it is not easy to discern between different modes of LCA used for future technology assessment. And following the notion by Suh and Yang (2014) that "no model is perfect and the question is whether it provides useful insights (..) given questions and available data" we do not aim to identify a single best term. A discussion or categorization of best approaches has little merit since all modes have their own, often overlapping, strengths and weaknesses depending on the analysis for which they are applied.

For the purpose of this paper we prefer to define and use the term ex-ante LCA because it gives a clear focus on the analysis at hand. Using the term "ex-ante" makes clear that the assessment is performed before market introduction of a technology, where for example a prospective LCA can also be performed on an established technology to see its environmental impacts in a defined future. Hence, leaving aside epistemological and semantic differences, in this paper we use the term ex-ante LCA to refer to performing an environmental life cycle assessment of a new technology before it is commercially implemented in order to guide R&D decisions to make this new technology environmentally competitive as compared to the incumbent technology mix.

5.3 Challenges for performing ex-ante LCA

Using deductive reasoning based on a review of the relevant literature, this section analyses which typical challenges ex-ante LCA faces. We conducted a literature search in Web of science (Thomson Reuters) using the following keywords[1]: "dynamic LCA", "prospective LCA", "ex ante LCA" and "anticipatory LCA". The combination of keywords delivered 112 hits. The titles, keywords and abstracts of

1 "dynamic LCA" OR "prospective LCA" OR "ex ante LCA" OR "anticipatory LCA" OR "dynamic life cycle assessment" OR "prospective life cycle assessment" OR "ex ante life cycle assessment" OR "anticipatory life cycle assessment"

these papers were screened on the question if an LCA for technology development was performed, which led to a reduction of the list to 54 papers. After this, the abstracts of these papers were analysed in more detail to see if the paper would discuss methodological challenges. In this way, we ended with 26 papers listed in Table 5.3.

Interestingly, publications that, in our view, provide crucial insights in this field, such as Hetherington et al. (2014) and Cucurachi et al. (2018) did not surface in this search despite the structured approach we applied. It seems that the novelty of this field and the multitude of terms used still inhibits a proper, structured meta-analysis for ex-ante LCA. After having reviewed about half of the selected papers we noted however, that reviewing additional papers did not lead to identification of additional challenges and solutions in ex-ante LCA. We therefore feel confident that our selection of papers provides a good overview of issues in this field.

The selected studies were analysed on their discussion of challenges, difficulties and bottlenecks in performing the assessment in practice. We review such challenges below following the LCA phases discerned in the ISO 14040 standard. Some of the challenges found can also be relevant for ex-post LCA, but have more prominence when an ex-ante LCA is performed. We will eventually see that similar challenges occur across different LCA phases. Section 5.4 discusses potential remedies for the challenges encountered. Most remedies provided by the selected studies make (implicit) use of scientific disciplines and concepts that are often not necessarily the strengths of the LCA practitioner (e.g. scenario development).

Goal and scope definition (GSD)

Methodological choices in any form of assessment or research are determined by the goal of the study and the research questions asked (Zijp et al. 2015; Hetherington et al. 2014; Miller and Keoleian 2015). In order to understand the merit of any assessment, choices and research questions should be clearly defined. The ISO standard prescribes to define the goal and scope of the study as a first step (International Organization for Standardization (ISO) 1997). When using scenarios in LCA, the GSD is the place where these scenarios need to be framed (Pesonen et al. 2000).

Goal (and research question)

The starting premise for ex-ante LCA studies is that new technologies are developed to improve the status quo situation by implementing a new technology in the technology mix. These new technologies will likely compete with well-established mature technologies (Frischknecht et al. 2009) and a comparison with these incumbent technologies should be part of the assessment. The inherent goal of any ex-ante LCA is therefore to compare

the future potential environmental performance of the new technology, vis-a-vis one or multiple incumbent technologies (Hetherington et al. 2014) in order to gain insights on the further developments of these not-yet-introduced technologies and guide upcoming efforts in research and development of the new technology. Therefore, we see technology developers as the main audience for these studies although they could also be insightful for policy makers.

Scope (temporal, geographical, functional unit and alternatives, choice for impact categories)

Making a balanced comparison of technologies requires a consistent modelling framework for all lifecycle phases and boundaries conditions (Bauer et al. 2015). Specific attention points in ex-ante LCA include:

Temporal and geographical scope. An obvious implication of assessing technologies before market implementation is that ex-ante LCA studies should consider a hypothetical future commercial state (e.g. technological performance, market situation) of the technology under assessment (Frischknecht et al. 2009). One has to be particularly explicit in defining at what moment in the future what level of maturity and what level of market penetration the novel technology may reach. One might even consider how the introduction of the new technology might influence the existing market. Defining a specific moment in time is an essential step to provide a balanced comparison between new and incumbent technologies. As a result, an explicit temporal scope will also have implications on how the competing incumbent technology and background systems are modelled.

Functional unit and alternatives. It is often a challenge to comprehensively define its foreseen function of new technologies and, based on that, the incumbent technology (Hetherington et al. 2014). Although one of the strengths of LCA is that assessments are based on functionality (service) and not on a specific product. Defining this function and related functional unit is however, a big challenge in ex-ante LCA (Hesser 2015; Aldaco et al. 2019; Peters and Weil 2017). Such a challenge is complicated by the necessity to identify the alternative technologies that compete in providing the same function or functions (Arvidsson et al. 2018). An example of such challenge is reported in van der Giesen et al. (2014), in which a total of four functions were defined for solar fuels produced from CO_2. In that study, it is clearly shown that an unambiguous function (or reason) to produce solar fuels is hard to define. Hence, the choice for the functional unit needs a more careful deliberation than is common practice in classical ex-post LCA. Moreover it might even happen that a technology identified as "the alternative" might not be the future incumbent (Arvidsson et al. 2018).

Choice for impact categories. Ex-ante LCA usually aims to analyse if the new technology can have (expected or projected) environmental gains over the incumbent technology. However, since the properties of the new technology are not yet well known and experience in practice is absent or low, full insight in all the relevant impact categories may not yet exist. Challenges with impact categories for new technologies and materials are identified and will be discussed under Life Cycle Impact Assessment below.

In sum, in the goal and scope definition in ex-ante LCA has the following additional challenges compared to regular, ex-post LCA:

- At what moment in time is the new technology to be expected to be operational at which level of maturity?
- How does the functional unit need to be defined so that the new and the incumbent technology provide the same (similar) functionality?
- What is the incumbent technology?

Inventory (LCI)

Unavailability of life cycle inventory data is a common hurdle encountered in ex-post LCA, which is often overcome by making use of secondary data in combination with a sensitivity analysis. To limit the time and resources of the LCA practitioner a distinction is made between foreground data, data that is gathered by the LCA practitioner and background data, data that is taken from existing LCI databases. For ex-ante LCA studies, we expect that foreground data is collected to describe and model the new technology and the incumbent. Background data is required to represent the context of the study, usually consisting of data for upstream supply chains necessary for the emerging and incumbent technologies to perform the selected functions, which can also include socio-economic considerations. While a technology developer typically has limited influence on the background system, manipulation in the face of consistent modelling might be needed.

Foreground data, is usually not readily available for new technologies (Frischknecht et al. 2009). The available data and knowledge is specific for the case and context at hand (Gavankar et al. 2014; Hospido et al. 2010) and is highly subjective to a variety of factors that cannot be controlled (Miller and Keoleian 2015). Existing LCI databases are based on historic data and the new technology under study is not available therein (Kunnari et al. 2009; Wender et al. 2014b). The data that are available most likely originate in lab experiments or pilot projects and are therefore not representative of operational scales (Arvidsson et al. 2014; Gifford et al. 2016; Hesser et al. 2017a; Schulze et al. 2018). One should be aware that a comparison between the new technology with the incumbent,

for which data at industrial scale is available, should not be done without first making apparent the assumptions and scenarios about the future of these technologies (Piccinno et al. 2015; Tecchio et al. 2016; Hetherington et al. 2014).

Inventory data for the new technology can be collected from scientific articles, patents, collected via expert interviews, or can be found in unpublished lab results or through process simulation (Arvidsson et al. 2018). These data however, mainly provide proof of principle for a new technology and are far from representative for future commercial scale operation. It might be necessary to manipulate whatever data is available to hypothetically represent the future situation. In this context, using assumptions (e.g. based on learning curves) is unavoidable (Villares et al. 2016). Data representing commercial scale operation is most likely to be available for the incumbent technology given the more advanced level of development of the incumbent over time. Further optimization of performance for such technologies can be expected in the future. An additional difficulty here is that the available data for both systems most probably does not automatically allow comparable systems boundaries. This demands for a hypothetical expansion of the system boundaries of one of the systems by the practitioner (Hetherington et al. 2014) and will obviously also contribute to significant uncertainties about how such a new technology will perform in the future.

Background data, should represent the future situation for when the new technology is defined to be commercially operational. This is highly relevant, since background processes usually make up to 99% of all unit processes in a product system and only occasionally fall below 95% (Wernet et al. 2016). This means that the impacts caused by background processes are of considerable influence on the outcomes of an LCA study. Several authors have stressed that changes over time in background systems (e.g. in the competing technology as well as in the technological landscape) should also be taken into account (Arvidsson 2013; Sanden et al. 2005; Frischknecht et al. 2009). A temporal mismatch between back- and foreground systems is to be avoided (Arvidsson et al. 2018), which can only be facilitated by a clear definition of the temporal scope in the GSD of the study. The data used for modelling the background system should represent the future operational playing field and be consistent for the same scope defined for the technology and incumbent(s) under study. A challenge here is that available background data, like the ecoinvent database (Swiss Centre for Life Cycle Inventories 2010), are commonly assumed to be representative for the current situation while data is clearly already outdated.

In some cases, forward-looking LCA studies do not explicitly compare a specific new technology with an incumbent technology as such, but want to assess the environmental

impacts of a proposed sustainable future. A good example of this is the work of Hertwich et al. (2015) and Berrill et.al (2016) who perform 'integrated life cycle assessment of long-term, wide-scale implementation of electricity generation from renewable sources' (Hertwich et al. 2015) using IEA scenarios. Where this approach is different from our definition of ex-ante LCA (the goal is not to guide R&D of a specific new technology), it provides very useful insights on how to deal with future changes for the incumbent and background system. At the same time, these studies show the importance of assessing new technologies on wider economic scales and go beyond the limitations of the functional unit to assess economy-wide implications. Estimating the potential market share of new technologies over time may be relevant in ex-ante LCA and should at least be considered when making claims on the future performance of new technologies.

In sum, the Inventory phase in ex-ante LCA has the following additional challenges compared to regular, ex-post LCA:

- How will new technological systems develop into the future, perform under the scope defined and is representative LCI data available?
- How will incumbent technological systems develop into the future, perform under the scope defined and is representative LCI data available?
- How do background systems and their performance develop over time, perform under the scope defined and is representative LCI data available?
- What is the potential market share of the new technology in the future?

Impact assessment

In ex-ante LCA studies it is important to realize that potential environmental impacts of new technologies are not automatically covered by the existing impact categories commonly used in ex-post LCA studies described in the LCA handbook (Guinée et al. 2002) or in the ReCiPe life cycle impact assessment method (Huijbregts et al. 2017). Recent ex-ante LCA studies of new technologies and new materials stress the great limitation of the lack of characterization factors at the life cycle impact assessment (LCIA) phase of LCA studies (McKone et al. 2011; Tufvesson et al. 2012; Deng et al. 2016; Wender et al. 2014b). The absence of suitable impact categories and specific characterization factors may mask the true environmental performance of a new technology compared to the incumbent technology, as many biosphere flows with a potential environmental impact are left unclassified due to a lack of adequate models and data. Hetherington (2014) stresses that environmental impact assessment methodologies will lag behind the formation of new materials with potential impacts in the environment. A good example of this is the upcoming interest in nano-technology. It has been recognized that properties of nano-technology are not yet well known and

that considerate efforts should be put in developing methods to assess the environmental impacts on the environment (Guineé et al. 2017).

In sum, the Impact Assessment phase in ex-ante LCA has the following additional challenges compared to regular, ex-post LCA:

- Can the new technological systems display unexpected new impacts not yet covered in LCIA?
- Can characterization factors relevant for the incumbent and new technical systems change over time?

Interpretation

The inherent necessity of making assumptions and high uncertainty surrounding modelling choices that have to be made makes that the outcomes of an ex-ante LCA study should not be regarded as a final result, but rather as a possible implication a technology can have under a specific set of assumptions (Villares et al. 2017). It is even said that ex-ante LCA provides a set of answers and not the answer, it rather provides a structure for debating and guiding research and development with all necessary stakeholders involved (Frischknecht et al. 2009). The execution of an ex ante LCA should thus be regarded as a process and not a product in itself (Wender et al. 2014b). Absolute outcomes should be regarded as indicative (Kunnari et al. 2009) or preliminary (Pehnt 2003), and should be used to inform decision making with the warning that decision-makers should use their wisdom and experience when using the results (McKone et al. 2011). So how can we interpret the results we get from performing an ex-ante LCA?

One issue that plays up more with new technologies in early development stages is that classical uncertainty analysis used in LCA that focuses on the 'known unknowns' may be insufficient. We simply do not know what the future will look like. Classical uncertainty analyses (in LCA) assume complete information on the (LCI) systems under assessment. Further, the impact categories and characterization factors are known and uncertainties can be measured. Quantifying uncertainties for future situations enters another dimension of uncertainty (see section 0), and we should be careful not to "add a surrogate quantified layer to the already evidently ambiguous generated information related to the scenarios and development paths considered" (Villares et al. 2016). For ex-ante LCA, it is a challenge to deal with this advanced dimension of uncertainty.

Connected to the issue of increased uncertainty is that new technologies more likely exhibit 'unknown unknowns' than mature technologies. An example of this issue is the introduction of biofuels and the use of micro-plastics. Before being commercially

available, these technologies were seen as a huge step forward in terms of potential and innovation, only to find out their negative and pervasive impacts years later. The question is if we could not have been more considerate during research and development of these technologies, a question that goes far beyond the focus of ex-ante LCA, but at least deserves consideration.

In sum, the Interpretation phase in ex-ante LCA has the following additional challenges compared to regular, ex-post LCA:

- How can the increased uncertainty around modelling new technologies in the future be dealt with?
- How can the possibility that new technologies will display unknown unknowns be dealt with?

5.4 Methods, techniques and approaches supportive to performing ex-ante LCA

While section 3 identified challenges by phase in the LCA process, it is clear that certain challenges come back in different stages of the LCA. For instance, the GSD phase needs to specify at which moment in time the new technology is operational, while in the Inventory phase an estimate of future performance needs to be made. This all relates to the issue of modelling consistent future foreground systems. To identify methods, techniques and approaches supportive to performing ex-ante LCA, we cluster the challenges into three main focus points that methods for ex-ante LCA need to address specifically and discuss them in the subsequent sections (see Table 5.2 for a full explanation of relations):

- Modelling consistent future foreground systems
- Modelling the incumbent technology into the future (future LCI data);
- Modelling the new technology into the future (future LCI data and potential market share).
- Modelling consistent future background systems
- Dealing with the uncertainty in ex-ante LCA
- Covering uncertainty around the future incumbent technologies;
- Covering uncertainty around the future of new technologies;
- Dealing with potential 'unknown unknowns' with regard to e.g. impact categories.

Modelling consistent future foreground systems

When a new technology is benchmarked against the incumbent it is important that modelling choices consistently represent the compared systems, including the background system at a time that the new technology is supposed or expected to be

commercially operational (Bauer et al. 2015). The LCA practitioner needs to be aware and take into account that these three parts of the model might be at different levels of development. Knowledge and data on these three parts are most likely only available on different scales of operation and, in addition, depend on the level of development of the technologies themselves.

At the time of execution of an ex-ante LCA, the new technologies are typically in their technology or development trajectory (see Figure 5.1). It will likely take a considerable amount of time to get from patent to market introduction. Only at this point does the new technology start to compete with the incumbent. Based on historic data, this period can be defined at up to 25 years, although this time frame seems to decrease for newer technologies (Hirooka 2006). After market penetration it will take a similar amount of time for the technology to become mature (Kramer 2009). When comparing new technologies to incumbent technologies, it is very important to take into account their level of development and that considerable time is required before technologies operate at similar (comparable) scales. To account for this in ex-ante LCA we build on work done by Gavankar et. al (2014), who discuss the role of technology maturity in LCA and introduced the concept of Technology Readiness Levels (TRL) and Manufacturing Readiness Levels (MRL) to the field of LCA. They stress that the outcomes of LCA studies on emerging technologies should be represented in relation to their scale of operation because the environmental impact at lower levels (kW scales) is most likely not linearly scalable to higher levels (GW scales), while the new technologies proposed are intended to perform at those higher scales. We stick to the concept of MRL because technology readiness only implies that a technology is feasible, but does not provide information to whether the technology is ready for large-scale manufacturing or operation. It is easy to understand that technologies with a low MRL (~5) encounter larger uncertainties when modelled to be at their hypothetical full scale and that modelling incumbent technologies with a typical MRL of 10 have much lower uncertainties.

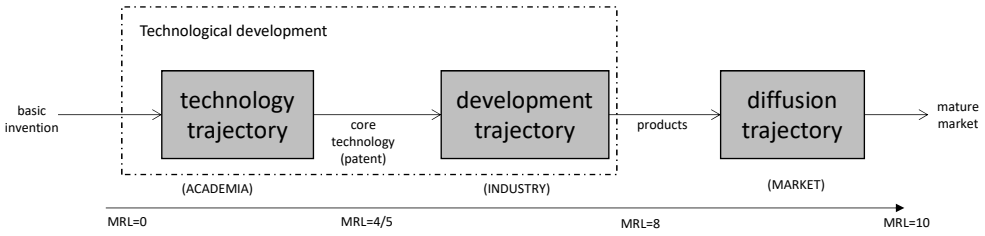

Figure 5.1: Innovation trajectories and development over time based on Hirooka (2006).

Ex- ante LCA studies are by definition based on a hypothesized state of operation (Hospido et al., 2010), or even on more than one stylized or extreme state that allows the illustration of differences between specific technological options (Arvidsson, 2013; Sanden et al., 2005). The main challenge is defining the states of the compared technologies consistently by making similar choices on the anticipated operation and scale-up of the technology from the lab to its industrial state. The subjective choices, individual preferences and perceptions of the practitioners and of the technology developers are an integral part of the modelling process and are bound to be affected by the inherent changes in basic socio-economic conditions over time (Frischknecht et al., 2009).

Selecting and modelling the incumbent technology into the future

Selecting the incumbent technology should be based on (similar) functionality as is common in LCA. However, new technologies often have multiple functionalities that often are not found simultaneously in one existing (incumbent) technology (mix). It is therefore important to be very clear on the intended application of a new technology, although this is often an uncertainty. Keeping this implicit and qualitatively discussing multiple intuitive functions makes it very hard if not impossible to perform a fair assessment. Moreover, this approach does not allow giving insights in the potential implications of certain design choices. Choosing the potential application of a technology could be guided by the public discourse, but could also be determined by available data for the insights desired. At least a clear statement on this needs to be presented. For example, Van der Giesen et al. (2014) explicitly choose to compare fuels produced form CO_2 to existing diesel fuels where, based on other possible functionalities discussed in the paper, also a choice for carbon capture and sequestration could have been made. It is clear that a single best incumbent is hard to define, however, organizing stakeholder discussions to identify the incumbent technology for the assessment enables making the assessment as insightful as possible.

Even after a product is at MRL 9 or 10 it is still possible that minor advances and developments take place. It might be possible that the incumbent technology, typically operational at an MRL of 10, will have developed further at the time that the new technology will start entering the market. In their future scenario, work Hertwich et. al (2015) used a combination of industry roadmaps, technology learning curves and expert consultation to arrive at substantiated performance data for technologies that are already on the market. These sources provide a first solid base to integrate potential further development of the incumbent technology to be aligned with the scope defined for the ex-ante LCA study.

Modelling a new technology at scale into the future

The big question for modelling a new technology at the same scale as the incumbent is how to base a full-scale model of the technology on the available knowledge from the lab and the technology trajectory. The use of technology learning curves that describe technology progress in terms of "decreasing costs as a function of accumulating experience with that technology" (McDonald and Schrattenholzer 2002) or "as a function of cumulative production" (Bergesen and Suh 2016) unfortunately seems not directly applicable for new technologies since market-based experience with these new technologies simply does not exist. However, experience with similar technologies might provide an idea for further technology development in combination with basic laws of physics and critical expert input. One should, however, be informed that a learning curve "hardly represent[s] a physical law, but rather describe[s] a persistent empirical phenomenon with still significant uncertainties surrounding both the estimation of specific learning rates and their extrapolation in scenarios" (McDonald and Schrattenholzer 2002).

Another way is to combine learning curve insights with knowledge and experience on 'upscaling' from the field of e.g. chemical engineering, for which considerate expertise is required. Piccinno et. al (2016a) provide a framework with which LCA practitioners with limited chemical engineering knowledge can obtain a first estimate about the impacts a chemical produced at an industrial scale when only laboratory scale data is available. The authors stress that the framework is only applicable to existing technologies and not for new technologies in the future. Another contribution to including upscaling in LCA was provided by Caduff (2014) who investigated the use of scaling factors from cost engineering and power-law relationships and applies this to energy technology. Also here the scaling factors are based on empirical experience with existing technologies. Thus, it is hard to say if this approach is applicable to new technologies in early stages of development.

Parvatker and Eckelman (2019) reviewed eight different upscaling methods that are used in chemical engineering to fill up data gaps in LCA. In addition, these methods do not take into account any future developments. Although these approaches are not equipped to look into the future, they might provide a starting point for defining scenarios. Including scenarios analysis is found to be crucial in up-scaling exercises since it makes the influences of assumptions and uncertainty transparent (Piccinno et al. 2016b). It even seems that the methods ranked by Parvatker and Eckelman (2019) are indicative of application at certain MRL levels and are also in line with levels of uncertainty encountered (see Figure 5.2).

Concluding, we can say that extensive guidance for modelling new technologies in the future exists (in e.g. the field of chemical engineering). A remaining question is how we can perform some form of upscaling in other fields like nano-, energy or food technology. For now, the only option seems to be to organize a structured discussion with experts on the future expectations and potential of new technologies. The practitioner should not expect the perfect and correct answer, but gather enough information to investigate different hypothetical routes of development using scenarios.

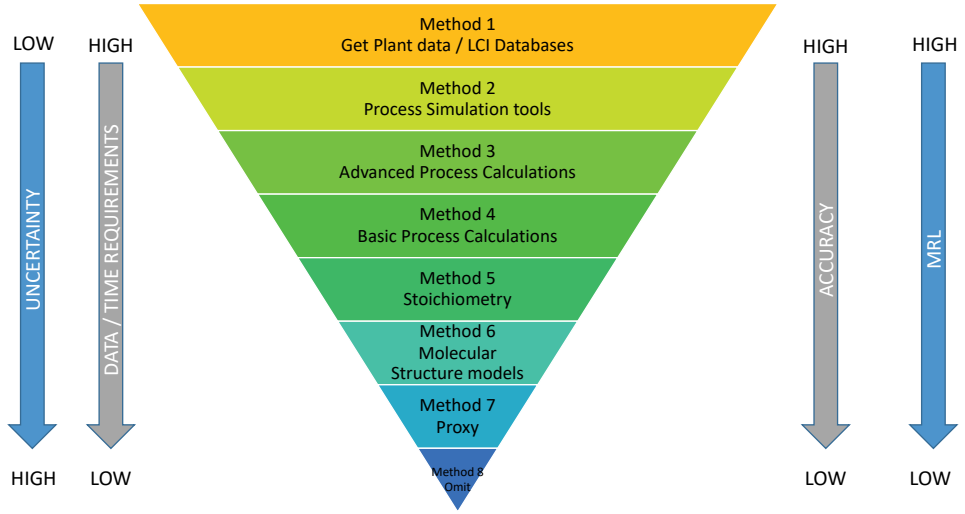

Figure 5.2: hierarchy of methods in LCI data generation adapted from Parvatker and Eckelman (2019).

Modelling future background systems

It has already been identified that existing ex-ante LCA studies do not necessarily take into account developments in the background system, while it is important that "a temporal mismatch between back- and foreground systems in prospective LCA studies should be avoided" (Arvidsson et al. 2018). We already showed the importance of choices in background systems for the final outcomes of the study under *Inventory LCI*. In practice, the foreground system is modelled at a future state while for the background system, one assumes the current (even outdated) temporal state (Mendoza Beltran et al. 2018). The question is how to take time into consideration so that fore- and background systems cover the same temporal scope?

A common and fruitful approach for dealing with time and future systems in LCA is to make use of scenarios (Pesonen et al. 2000). Several studies attempt to integrate time to account for potential future developments in the fore- and background systems

(Nordelöf et al. 2014; Krishna Manda et al. 2015; Spielmann et al. 2005). Even though these studies show that it is essential to integrate a time dimension in the assessment, most existing projects attempting to do so encounter challenges. The scenarios used are often inconsistent and lack transparency, technology maturity is often not accounted for and reproducibility of these scenarios is difficult because of large amounts of data needed and tracing assumptions made during scenario generation (Mendoza Beltran 2019). To overcome these, Mendoza et al. (2018) propose to explicitly discern between scenario generation and scenario assessment (following Fukushima and Hirao (2002)). For scenario generation they build on the Shared Socio-economic Pathways (SSPs) developed by the climate change research community and constructed with Integrated Assessment Models (IAMs) (van Vuuren et al. 2014). For the evaluation of these scenarios they combine the market shares for e.g. energy technologies as predicted by IAMs with LCA data from databases like ecoinvent 3 (Wernet et al. 2016).

Mendoza et. al (2019) show that it is feasible to implement technology scenarios calculated with IAMs into LCI databases for the electricity sector. The challenge remains to develop similar future background LCI databases for other sectors that are closely linked to future sustainable technology development. The approach taken is new and therefore not yet common practice. It would be very beneficial to develop or expand on existing LCI databases like ecoinvent to represent potential future situations via clearly defined scenarios. Integrating these scenarios is a project in itself, so externally developing scenarios that address critical future parameters should be a point of focus for further development of ex-ante LCA practices.

As long as IAM based LCI background data are not available, practitioners should at least attempt to be transparent about potential temporal mismatches between back- and foreground system and the implications of that on the outcomes of the study. As is commonly done, one can assume that the current background data is also representative for a defined future, or one can assume that the background system does not change over time and is constant. This might however, result in huge errors as shown by Cox et al. (2018). Therefore, a thorough discussion on the impact of such modelling decisions on the outcomes should be included. Another option is to use the NEEDS database (NEEDS), which has its focus on energy. This consistent database is outdated because it is based on ecoinvent 1.3 and does not use the scenario approach as proposed by Mendoza et. al (2019). However, taking all limitations in mind and providing a proper discussion on the use of this database could allow for an ex-ante LCA study in which the temporal mismatch between back- and foreground data is covered.

Dealing with uncertainty in ex-ante LCA

It is obvious that studies assessing technologies in the early stage of their development deal with higher levels of uncertainty than technologies that are already implemented. This includes issues that have been termed 'unknown unknowns' and that cannot easily be covered by traditional, quantitative uncertainty assessments that are now applied in state of the art LCAs. The field of technology assessment therefore came up with concepts such as post-normal science (Funtowicz and Ravetz 1993) and indeterminacy (Wynne 1992) to capture a broader view on uncertainty. Mendoza Beltran (2018) used the typology of Wynne (1992) to position her PhD thesis as being mainly focused on quantifiable risks and uncertainties. We use Wynne's typology pragmatically, being a rather straightforward typology discerning a) risk (system parameters and probabilities are known), b) uncertainty (system parameters are known, but not the probability distributions), c) ignorance (neither system parameters nor probabilities are known) and d) indeterminacy (the future development is inherently undetermined).

The four types of uncertainty can respectively be assigned to high development and early development stages of the technology under assessment (see Figure 5.3). Conventional ex-post LCA studies and incumbent technologies in ex-ante LCA typically deal with technologies at a high MRL level. If the behaviour of the technology is well known, the chance of ignorance or indeterminacy is low and the more traditional fields of risk and uncertainty have to be covered. The LCA community has developed state of the art methodologies that can be applied here. These are Monte-Carlo simulations to deal with parameter uncertainty, or scenarios to deal with uncertainties related to methodological choices, see e.g. Mendoza Beltran (2018). In principle, such approaches can also be used to address e.g. changes in characterisation factors due to e.g. changing background concentrations as discussed under *Impact Assessment* above.

Ex-ante LCA must somehow deal with the problems of ignorance and indeterminacy for the assessment of new technologies. This is a field where the LCA community has a limited track record, in part since it requires a somewhat different view on the role of science. A so-called positivist view on science assumes that the world can be fully known, which fits well with the quantitative approaches including the assessments of uncertainty that the LCA community is so familiar with. A more constructivist view on science assumes that the world cannot be fully known, leading to the acknowledgment that overarching views exist, using different parts of available knowledge, combined with certain, often implicit, differences in interpretation.

To give the subjects of ignorance and indeterminacy a sound place in decision-making, various strands of science have developed the following approaches. Authors such

as Hamarat et al. (2013), Kwakkel and Pruyt (2013) and Maier (2016) have coined the concept of 'deep uncertainty', which is defined as "uncertainty for which experts do not agree on models to describe interactions among a system's components, and subsequently do not agree upon (exact causal structures) corresponding probabilities and possible outcomes" (Tegeltija et al. 2018). The remedy is to "think about the future in terms of multiple plausible futures rather than probability distributions" (Maier et al. 2016). In essence this approach is still somewhat fitting in the positivist tradition in the sense that it is deemed possible to define and quantify specific scenarios capturing the elements of ignorance and indeterminacy. Policy and social scientists however, seek the solution in embarking on more participatory approaches that deliberately encourage uncovering different, but plausible perspectives or framings of the possible risks and impacts of the new technology. An illustration of this is the 'Fourth Generation Evaluation" proposed by Guba and Lincoln (1989) who propose to bring this about in a hermeneutic dialectic process which is done in a responsive evaluation in 4 steps:

- Identify stakeholders and collect their claims (any assertion favourable to the evaluand), concerns (any assertions unfavourable to the evaluand) and issues (any state of affairs about which reasonable persons may disagree) on the evaluand;
- Claims, concerns and issues are presented to other stakeholder groups for response and discussion;
- Unresolved claims, concerns and issues steer additional needs for information that need to be collected by the evaluator;
- Negotiations using information added in step 3 to arrive at a consensus ("as each group copes with the construction of all the others, their own constructions alter by virtue of becoming better informed and more sophisticated" (Guba and Lincoln 1989)).

To some extent, this reflects the classical peer-review in LCA, but it is also obvious that in this case the panel will not only consist of technical specialists. The panel will consist of stakeholders that are likely to have (highly) opposing views and therefore allow the identification of potential ignorance and indeterminacy. This will help to counter the (usually positive) biases of the developers of the new technology as well as bring potential drawbacks of a new technology to light at an early stage when it is still possible to adjust the technology for the better. In the end, it is not about being right, but about aiding the design of new sustainable technologies in the best possible way.

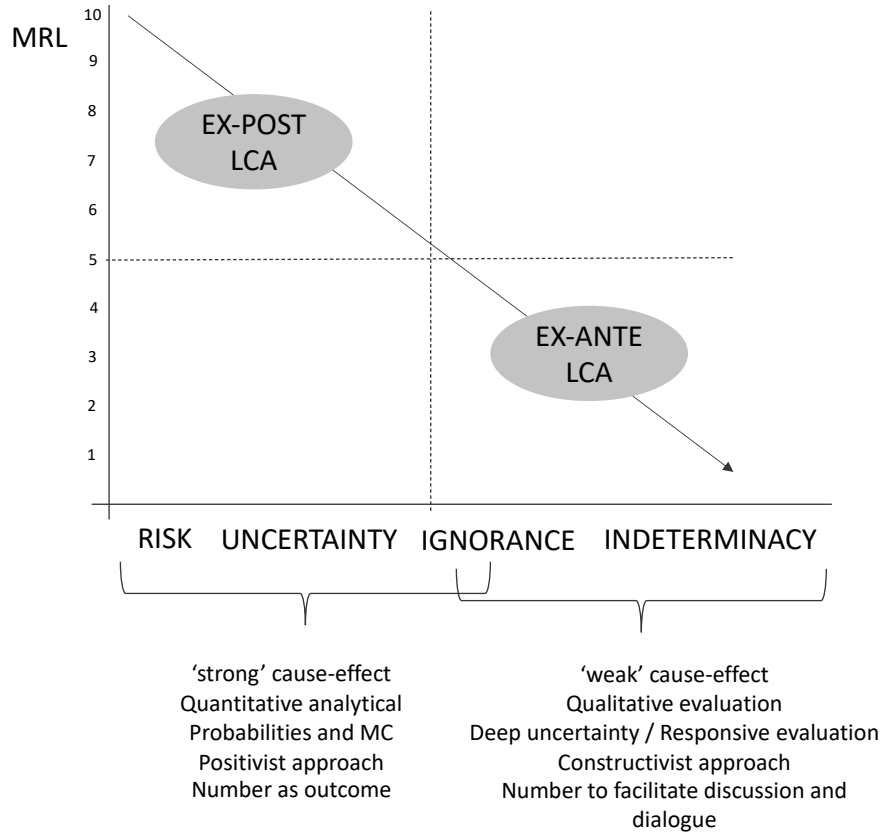

Figure 5.3: Relation between MRL, uncertainty and type of LCA assessment.

5.5 Discussion and conclusions

In this paper, we investigated how to perform an environmental assessment of potential new technologies that are still in their R&D phase. We identified the main challenges in comparison with conventional ex-post LCA practices and proposed practical remedies for coping with these challenges. We chose to use the term ex-ante LCA and defined this as performing an environmental life cycle assessment of a new technology before it is commercially implemented in order to guide R&D decisions to make this new technology environmentally competitive with the incumbent technology mix.

Having started from this definition, we conclude that a fixed way of performing an ex-ante LCA does not exist and that the challenges defined here as well as suggestions proposed are also applicable to other modes of LCA like consequential and prospective LCA, which are discussed in section 5.2. Choosing a method depends on the question at hand. We found different questions in literature that are assessed by different forward-

looking LCA approaches using different terminology. In this paper we specifically looked at a method to environmentally assess a new technology in order to guide research and development. Where such an assessment can provide important insights for R&D, one should be careful not to reach a verdict on a new technology before it has been implemented.

The challenges and proposed potential remedies for ex-ante LCA based on the former sections are summarized in Table 5.2. Modelling future foreground systems for the incumbent and new technology is a first challenge, which includes e.g. estimates of learning curves and scenarios for future market penetration. Although not the direct focus of this paper, a further investigation on the relationship between incumbent and marginal technology, the latter being common practice in consequential LCA approaches, could bring up interesting practices for defining the incumbent technology. Future background systems could be modelled by imposing futures as predicted by Integrated Assessment Models on Life Cycle Inventory databases, but this is not yet widely done. The main difference with conventional LCA studies however, is the highly uncertain information for the future. To acknowledge this considerable attention should be paid to the discussion on these uncertainties both in the design of the assessment as for the data used. This can be done in the form of responsive or third generation evaluation, which resembles the traditional LCA peer review procedure, but is different since it involves a much wider set of actors, and a more specific assessment of their claims and concerns on potential future impacts. This will increase the transparency and efficacy of the assessment because the relevant stakeholders and experts are involved. In this way, technology designers and other stakeholders derive insights on the influence of design choices or contextual factors (that are important, but hard to influence) on the potential environmental impacts of their foreseen technology.

The main recommendation for ex-ante LCA that we can give is of a scientific philosophical nature. In order to perform an ex-ante LCA the assessment requires a shift from a positivist to a more constructivist approach. Ex-ante LCA studies encounter such high uncertainties and require considerate assumptions that need to be based on debate rather than on fixed statements that are gratuitously presented as correct. In this way ex-ante, LCA finds a perfect application in guiding the discourse along the R&D path by providing structure following the ISO norm and quantifications that enable a proper discussion on design choices and potential future scenarios of new technologies. Practical experience in taking this more constructivist approach is still limited, it needs to be further developed and can only be obtained by application of the suggested solutions in real case studies. This is the aim for our ongoing case study work and we are looking forward to seeing the experience and insights of other research in this developing field.

Performing an ex-ante LCA study should follow the ISO norm, but would also require departing from the standard by increasingly involving stakeholders (Tsoy et al. 2019) as well as disciplines and skills[2] for which the conventional LCA practitioner is not typically equipped. It should however, not be the goal that the LCA practitioner masters these disciplines, rather that the discourse between the different disciplines is properly managed. Instead of performing a critical review after the LCA study one should invite the proper critical scientists on all modelling choices that need to be made, by incorporating for example the practice of responsive evaluation. Moreover, there is a great need for background databases that can be used in modelling future technologies. Research on integrating IAMs and LCI databases, which was initiated by Mendoza (2019), should definitely be followed up. Lastly, practices to deal with 'higher' less defined uncertainties (ignorance and indeterminacy) should be investigated, for which a good starting point is the concept of deep uncertainty.

2 Responsive evaluation / Technology expert involvement / Scenarios / Upscaling techniques / Technology learning curves / technology roadmaps / Combining LCI databases and IAMs / Deep uncertainty / general morphological analysis

Table 5.2: Challenges and suggested remedies for ex-ante LCA.

LCA phase	Challenge	Potential Remedy	Explanation
Goal and scope definition	At what moment in time is the new technology to be expected to be operational at which level of maturity?	Responsive evaluation Expert involvement	It is unclear when and where a new technology will be applied in the future or how it will exactly be designed. Defining a clear scope however, sets the base for many modelling choices that need to be made for the assessment.
	How does the functional unit need to be defined so that the new and the incumbent technology provide the same (similar) functionality?	Responsive evaluation expert involvement scenarios	The application of new technologies is a question for the future and can only be proposed with high uncertainty, sometimes multiple functions need to be defined.
	How can the incumbent technology be defined?	Responsive evaluation expert involvement	The application of new technologies is a question for the future and can only be proposed with high uncertainty. Existence of multiple functions makes defining the right incumbent uncertain.
Life cycle inventory	How will new technological systems develop into the future, perform under the scope defined and is representative LCI data available?	Upscaling techniques, Responsive evaluation expert involvement scenarios	Data is often only available at lab scale for the current moment, How this will play out in the future is uncertain.
	How will incumbent technological systems develop into the future, perform under the scope defined and is representative LCI data available?	Technology learning curves technology road maps Responsive evaluation expert involvement scenarios	Data is often available at full scale, but incumbent technologies might slightly improve or change over time.
	How will background systems and their performance develop over time, perform under the scope defined and is representative LCI data available?	Combining LCI databases with IAM scenario Responsive evaluation expert involvement scenarios	Background data is available for a different scope and not always include newer technologies or socio-economic change.
	What is the potential market share of the new technology in the future?	Responsive evaluation expert involvement scenarios (IAMs, IEA, IPCC)	Which market share a new technology will occupy in the future is highly uncertain.

Impact assessment	Can the new technological systems display unexpected new impacts not yet covered in LCIA?	Responsive evaluation expert involvement scenarios	Existing (base-line) impact categories might be irrelevant for the future or different, yet undefined, impacts will become relevant.
	Can characterisation factors relevant for the incumbent and new technical systems change over time?	Responsive evaluation expert involvement scenarios	Characterisation factors for impact categories defined above might change based on new insights and research.
Interpretation	How can the increased uncertainty around modelling new technologies in the future be dealt with?	deep uncertainty responsive evaluation scenario modelling expert involvement	In conventional (ex post) LCA uncertainties that go beyond risks and uncertainty are not acknowledged systematically, although doing so might provide valuable information for R&D efforts.
	How can the possibility that new technologies will display unknown unknowns be dealt with?	Responsive evaluation expert involvement	Not acknowledged systematically in conventional (ex post) LCA, but relevant for new technologies intended to contribute to a sustainable future.

Table 5.3: Selected papers used for the identification of practical challenges in ex-ante LCA.

Author (year)	Title
Aldaco et al. (2019)	Bringing value to the chemical industry from capture, storage and use of CO2: A dynamic LCA of formic acid production
Arvidsson et al. (Arvidsson et al. 2018)	Environmental Assessment of Emerging Technologies: Recommendations for Prospective LCA
Arvidsson and Molander (2016)	Prospective Life Cycle Assessment of Epitaxial Graphene Production at Different Manufacturing Scales and Maturity
Arvidsson et al. (2014)	Prospective Life Cycle Assessment of Graphene Production by Ultrasonication and Chemical Reduction
Aryapratama and Janssen (2017)	Prospective life cycle assessment of bio-based adipic acid production from forest residues
Cardellini et al. (2018)	Temporalis, a generic method and tool for dynamic Life Cycle Assessment
Gifford et al. (2016)	Reducing environmental impacts of metal (hydr)oxide nanoparticle embedded anion exchange resins using anticipatory life cycle assessment
Hesser et al. (2017a)	Injection moulding unit process for LCA: Energy intensity of manufacturing different materials at different scales
Hesser et al. (2017b)	Integration of LCA in R&D by applying the concept of payback period: case study of a modified multilayer wood parquet
Joyce et al. (2018)	Identifying hotspots of environmental impact in the development of novel inorganic polymer paving blocks from bauxite residue
Krishna Manda et al. (2015)	Prospective life cycle assessment of an antibacterial T-shirt and supporting business decisions to create value
Miller and Keoleian (2015)	Framework for Analyzing Transformative Technologies in Life Cycle Assessment
Pehnt (2006)	Dynamic life cycle assessment (LCA) of renewable energy technologies
Peters and Weil (2017)	Aqueous hybrid ion batteries - An environmentally friendly alternative for stationary energy storage?
Piccinno et al. (2016b)	From laboratory to industrial scale: a scale-up framework for chemical processes in life cycle assessment studies
Ravikumar et al. (2018)	Novel Method of Sensitivity Analysis Improves the Prioritization of Research in Anticipatory Life Cycle Assessment of Emerging Technologies
Schulze et al. (2018)	An Ex-ante LCA Study of Rare Earth Extraction from NdFeB Magnet Scrap Using Molten Salt Electrolysis
Spielmann et al. (2005)	Scenario modelling in prospective LCA of transport systems - Application of formative scenario analysis
Tan et al. (2018)	Combining ex-ante LCA and EHS screening to assist green design: A case study of cellulose nanocrystal foam
Tecchio et al. (2016)	Ex-ante Life Cycle Assessment approach developed for a case study on bio-based polybutylene succinate
Tsang et al. (2016)	Anticipatory life-cycle assessment of supercritical fluid synthesis of barium strontium titanate nanoparticles

Tsoy et al. (2019)	Anticipatory Life Cycle Assessment of sol-gel derived anti-reflective coating for greenhouse glass
Villares et al. (2016)	Applying an ex-ante life cycle perspective to metal recovery from e-waste using bioleaching
Villares et al. (2017)	Does ex ante application enhance the usefulness of LCA? A case study on an emerging technology for metal recovery from e-waste
Wender et al. (2014a)	Anticipatory life-cycle assessment for responsible research and innovation
Wender et al. (2014b)	Illustrating Anticipatory Life Cycle Assessment for Emerging Photovoltaic Technologies

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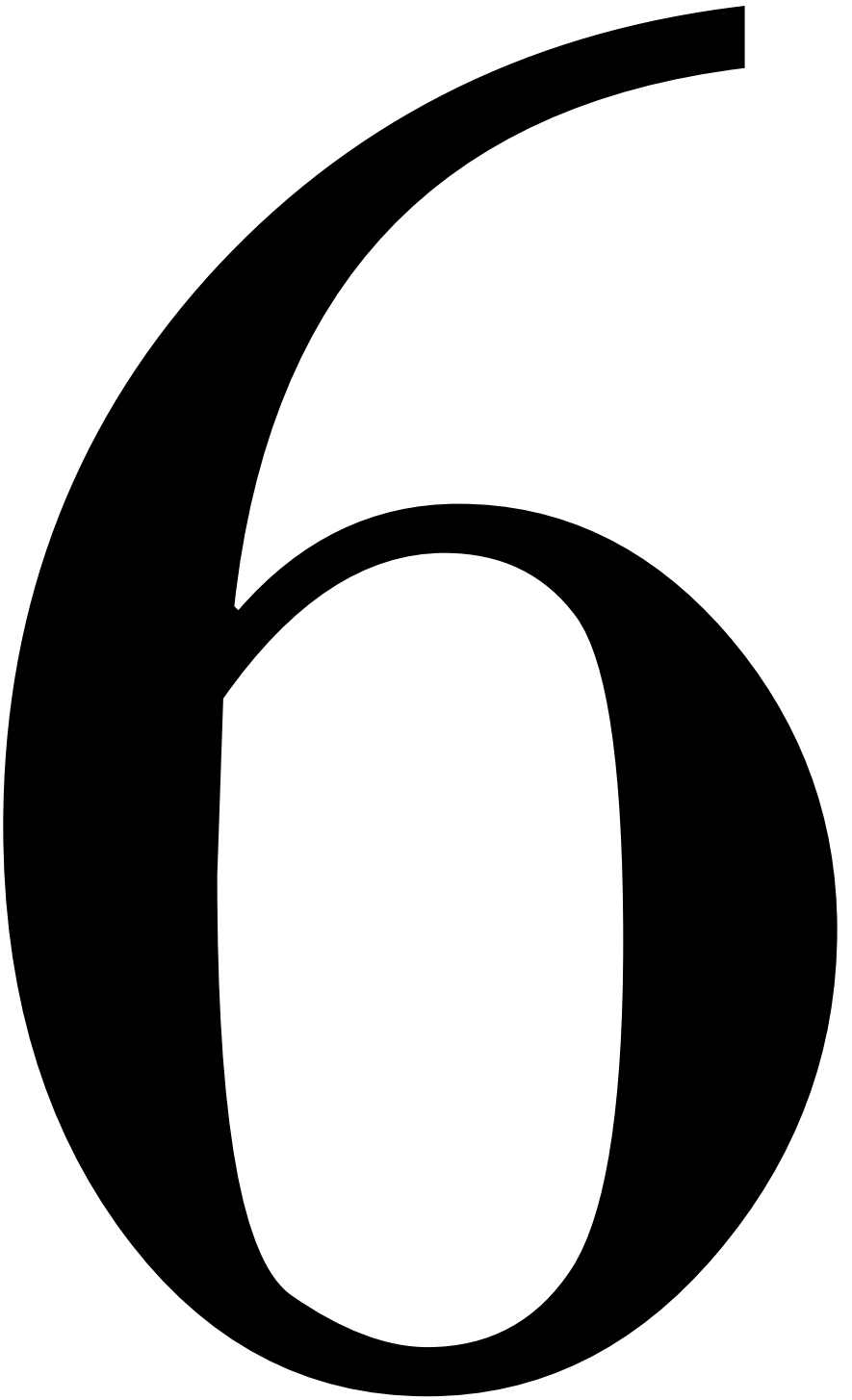

Chapter 6

Conclusions and General Discussion

6. Conclusions and General Discussion

The aim of this thesis was to find a method that facilitates the integration of potential future environmental impacts in the development of new technologies, with an emphasis on energy technologies. The idea is that having these insights early in the development process will help researchers and developers in making better informed design decisions and prevent that these new technologies show new problems when they are eventually put to use.

In the field of Industrial Ecology, questions on environmental impacts are approached by taking a systems approach. This is also relevant for assessing new technologies because these are not developed or used in isolation, but make up part of a larger technical-economic system. Extensive experience on how to take an analytical systems approach is found in different LCA-based methods. LCA therefore forms the starting point for the method called ex-ante LCA, which is developed in this thesis. The main difference between the conventional LCA-practice is that LCA is normally applied *ex-post*, after a technology is implemented. Here we apply the use of LCA in an ex-ante manner, before a technology is implemented and widespread experience and data is available.

This thesis started by investigating the usefulness of the life cycle sustainability (LCSA) framework, at the time the most comprehensive LCA-based framework available for the assessment of new energy technologies (chapter 2). Since shortcomings in the LCSA approach were found when applied to novel technologies, the following approach was taken in a next step. Two separate LCA case studies assessing new energy technologies with future applications were performed to identify the challenges encountered when LCA is applied ex-ante (chapters 3 and 4). As a final step, recommendations and guiding principles for the further practice of ex-ante LCA were then provided. These were based on learnings from the case studies and a critical review of the current practice of LCA for the assessment of new technologies (chapter 5). This concluding chapter will provide a set of answers to the research questions posed in the introduction, based on the findings reported in chapters 2 through 5.

Guiding research questions covered in this thesis

1. What boundary conditions are required for LCSA to be a useful approach to assess early stage technologies? (chapter 2)
2. Which challenges are encountered in practice when LC(S)A is applied to assess early stage technologies? (chapter 3 and 4)
3. Can we find generalizing principles to assess the future environmental impacts of technologies in early stage of development? (chapter 5)

Further thoughts for future practice and research will be presented in section 6.4. These thoughts are based on experiences in disseminating the knowledge obtained including review processes encountered when getting the research published as well as based on informal discussions with peers at conferences and project meetings. While these insights and thoughts might be merely based on observations and may sometimes be more anecdotal than factual scientific observations, they do provide interesting insights in the daily practice of R&D. Ultimately they are used to outline a set of questions for further developing methods to guide R&D of technologies that are believed to provide a piece of the puzzle in shaping a sustainable future.

6.1 Question 1 - What boundary conditions are required for LCSA to be a useful approach to assess early stage technologies?

There was an ongoing lively discourse on the framework for Life Cycle Sustainability Analysis (LCSA) at the start of this research project,. LCSA was presented as a multidisciplinary framework capable of answering broad sustainability questions related to the life cycle impacts of products and technologies. Chapter 2 investigated if the LCSA approach could also be used to do a sustainability assessment of new technologies and what the boundary conditions are for such an assessment to be useful.

LCSA is intended as a broad framework of different methods with which different questions at different levels of analysis can be answered. The person performing the assessment should choose the method(s) applicable to answer the question posed in the specific case. Most studies applying LCSA lack a clear goal for the assessment and often miss a specific scope, which hinders setting up a clear research approach. Chapter 2 recommends that when used for the purpose of performing an ex-ante LCA, a first step in LCSA should be the definition of a thorough system description. This system description should cover at least the following elements: a technological description, the intended application or function of the technology, the expected market share (technology share in the mix), alternative and/or competing technologies providing the same function and which environmental impacts should be covered.

A missing thorough system description leaves ample room to manoeuvre in discussing the efficacy of a new technology, but hampers the execution of a strict assessment. This makes it difficult to give developers of new technologies precise guidance on how the environmental impacts of such new technologies can be minimized. This was also the conclusion of a more recent review of sustainability assessment approaches based on life cycles which states that "the presented holistic approaches for LCSA are either too broad or too narrow for scientific guidance. Therefore, many questions concerning LCSA are still open, e.g., regarding definition of sustainability dimensions and the desire or

need for multi-criteria decision-analysis“ (Wulf et al. 2019). Starting the assessment with a thorough system description enables answering questions on a new technology's potential impacts in a concrete manner that provides insights for further R&D of these technologies. The overall conclusion is thus that LCSA as a holistic, open method is not specific enough for performing a dedicated ex-ante LCA of novel technologies.

6.2 Question 2 - Which challenges are encountered in practice when LC(S)A is applied to assess early stage technologies?

Two case studies to assess the environmental impacts of two early stage technologies were performed with the insights from chapter 2 in mind. With these case studies, hands-on experience with the challenges that arise when applying LCA to new technologies was obtained. These insights were used later to formulate guidelines for ex-ante LCA in chapter 5.

6.2.1. Case study 1: Energy and climate impacts of producing fuels from CO_2

An LCA case study that assesses the energy and climate impacts of producing liquid hydrocarbon fuels from CO_2 is reported in chapter 3. In this case study the required technologies in themselves are not specifically new. The different technologies in the system assessed, are however, at different stages of development. For example, using the Reversed Water Gas Shift (RWGS) reaction to produce CO is still in the experimental phase (applying the reaction in the other direction, known as the water gas shift reaction is common practice in steam reforming of fossil fuels). In comparison, post combustion capture of CO_2 has been occasionally applied at an industrial scale and there is abundant experience with the Fischer Tropsch reaction. The combination of the technologies to produce hydrocarbon fuels has not been applied at an industrial scale nor are these fuel types yet available on the market.

The literature review in chapter 3 made clear that this fuel production route is believed to provide a sustainable fuel for the future. The aim of the case study was to scrutinize the broad and abstract claims used to support this belief in a quantitative manner. The claims were quantified by applying conventional LCA, basic physics and chemistry were used to quantify data not readily available in existing LCA databases. The study set up was designed and reported in a transparent manner so that assumptions and the numbers used are easily accessible and the outcomes could support the discussion on the efficacy of these fuels. The main conclusion was that producing liquid hydrocarbon fuels starting from CO_2 by using existing technologies requires much more energy than existing fuels. An improvement in life cycle CO_2 emissions is only found when solar energy and atmospheric CO_2 are used. Therefore, when looking at currently available technologies, the production of fuels from CO_2 is a very long term niche at best and not

the panacea suggested in the recent public discourse (section 3.4 in this thesis).

Since the case study research presented in chapter 3 has been performed, science has progressed and much more research has been done on the environmental gains of using CO_2 as a feedstock for chemicals and fuels. Here we make some additional remarks to put the research performed in context.

To start, in chapter 3 we assumed that in the alternative biofuel production route, the growth of biomass extracts CO_2 from the atmosphere. More recent studies (i.e. Daioglou et al. (2017)) make clear that large quantities of CO_2 are emitted into the atmosphere caused by the initial removal of vegetation when setting up biomass plantations. Although this was not the main focus of the research presented in chapter 3, it should be stressed here that, when taking these more recent insights into account the environmental impact for biofuel production would even be less favourable than presented in chapter 3.

Second, Liu et al. (2020) performed a similar study to ours but uses a hydroxide based process instead of humidity swing DAC process to supply the CO_2. The data for the study performed by Liu et al. (2020) is more elaborate and comes from a real time operating DAC plant, but still arrives as similar conclusions as presented in chapter 5. In addition Liu et al. (2020) also stress the importance of not neglecting the intentional motivation to produce fuels from CO_2, which is obtaining lower GHG emission to the atmosphere. This is an important remark and it was also experienced during the research performed in this thesis. In this context it is important to report that in practice there seems to be no favourable combination of environmental and cost performance (Fernández-Dacosta et al. 2019).

Third, in connection to the functionality discussion presented in chapter 3, other research (Wang et al. 2019) has been focussing on the most efficient use of excess renewable energy (in chapter 5 producing fuels from CO_2 with stranded renewables was one of the potential functions discussed). Wang et al. (2019) prioritize the application of excess renewable energy for useful purposes and find that using it to produce hydrogen as transportation fuel is the most efficient application. Wang et al. (2019) and Liu et al. (2020) contribute to an important discussion here which also shows that is crucial to be clear on the reason why you would produce fuels from CO_2. This discussion is even further investigated in the work performed by Tanzer et al. (2019; 2020). They also stress the need to keep in mind the initial motivation to develop negative emissions technologies or use CO_2 in new products as a GHG mitigation option. It is very important, as encountered in chapter 3 and 4 of this thesis, to clearly define

systems boundaries (Tanzer et al. 2020; Tanzer and Ramírez 2019; Fernández-Dacosta et al. 2019) to allow assessing the carbon balance and all proposed properties of new technologies.

6.2.2. Case study 2: Comparing the environmental impacts of HS-DAC and PCC

An LCA case study comparing two CO_2 capture technologies: humidity swing direct art capture (HS-DAC) and post combustion capture (PCC) was reported in Chapter 4. HS-DAC is a new technology still in its development phase that allows the capture of CO_2 from the atmosphere and is not yet applied at an industrial scale. Although different in system setup, its performance is often intuitively compared to PPC which allows the capture of CO_2 at point sources. PCC is much further in development, and although it seems a promising technology for averting CO_2 emissions, it has only been limitedly applied on larger scales, mainly because of political and economic reasons.

In the case study a comparison was made between two systems in which electricity is produced from coal. Either PCC or HS-DAC was used to capture the CO_2 emitted. Defining similar functionalities for both technologies allowed for a fair and structured analysis. The main outcome of the study was that, depending on the right water conditions and the availability of fresh water, HS-DAC seems to be a good complement to PCC. DAC complements PCC in that where PCC allows for capture of only around 70% of life cycle CO_2 emissions of coal fired electricity generation, by that increasing environmental impacts of coal fired electricity generation on average by 30% compared to no capture at all. DAC allows to capture 100% of life cycle CO_2 emissions by increasing environmental impacts by 50% if DAC is driven by coal electricity and just 20% when DAC is driven by PV electricity.

Existing comparisons between PCC and DAC generally provide the concern that DAC's energy requirements are much higher than those of PCC. It will therefore have a far higher cost per amount of CO_2 captured and is therefore too expensive to apply for a sustainable future. What is easily forgotten, however, is that DAC has other favourable properties, such as the possibility to capture CO_2 at its potential storage location, by this eliminating the need for CO_2 transportation via pipelines. In addition, HS-DAC can also be applied for capturing background CO_2 emissions and even making up for non-CO_2 greenhouse gas emissions. These properties would even allow overcoming the restraining political challenges that PCC faces. This case study provides a quantification of some (for now) hypothetical scenarios to investigate the possibilities and restrictions of HS-DAC. It thus facilitates a more balanced discussion on the application and development of HS-DAC that goes beyond process efficiencies CO_2 capture yields per dollar spent.

Looking at research outcomes published after the work done in chapter 4 we now find that humidity swing DAC seems to be a more exotic form of DAC than currently focused on. For direct air capture the more mature technologies at this moment concern the use of hydroxide sorbents or amine minerals (Realmonte et al. 2019). Shortly after publishing the research from chapter 4, David Keith published the first highly detailed description of a hydroxide based DAC paper (Keith et al. 2018). The details in that paper were drawn from commercial experience, where the study performed in chapter 4 was based on basic physics-based calculations and a very first lab scale prototype.

Recent studies on the performance of DAC technology are based on more mature technologies but arrive at similar general conclusions saying that DAC has a definite role in GHG mitigation efforts and will be most contributing when developed and applied alongside other negative emissions technologies (Realmonte et al. 2019; Fasihi et al. 2019; de Jonge et al. 2019).

6.2.3. Insights from the case studies

The main insight from doing these two case studies is that even more effort and resources should be attributed to defining the function or future application of a new technology when performing an ex-ante LCA. The basis of any LCA model lies in defining system functionality and the functional unit. A simple rule of thumb in the field of LCA says that the function of a system should be chosen as close as possible to the "end use". In practice, this might require expanding the system boundaries or even change from a cradle to gate (e.g. capturing CO_2) to a cradle to grave (e.g. producing CO_2 free electricity from coal) assessment to define a system in which all (or most) attributes of the technology under assessment are taken into account. This requirement of expanding system boundaries causes an ex-ante LCA assessment to automatically look beyond the technology under assessment alone and also identifies auxiliary technologies that are required for making a technology perform its function.

The process of defining the technologies' application provides insights in its context consisting of competing technologies, but also (auxiliary) technologies required to make the new technology function as defined. Although the future application of a new technology might be uncertain, providing a strict definition to start with provides the foundation to investigate this uncertainty further, gives insights in the technological system and navigates technology developers in taking a systems perspective.

Taking a systems perspective is important to not lose sight of the reason why the development of a new technology is initiated in the first place. We need to remember that technology is a means to an end and not a means on itself. For example, technologies

that capture CO_2 enable the reduction of CO_2 emissions to the atmosphere caused by fossil fuel use. This reduction is needed to keep temperature rise below 2°C with the ultimate goal to reduce climate change. In the end these technologies are not developed to capture as much CO_2 as possible, which sometimes seems to be the main focus of people involved in developing these kind of technologies.

The insightful challenge of defining the function of a technology is well illustrated in the case study on producing (solar) fuels from CO_2 presented in chapter 3. In the discourse around solar fuels, a combination of intuitive arguments is used to substantiate general claims on the sustainability of these fuels. However, when attempting to quantify these arguments with an LCA, more specific questions concerning the technologies context need to be answered in order to set a running model that makes sense. The separate unquantified arguments to support the case for producing fuels from CO_2 are repeated below, while immediate questions from the LCA practitioner are added.

1. Argument: Solar fuels can be used in the existing energy infrastructure. Immediate LCA questions: But do solar fuels have better environmental performance than incumbent fuel technologies that also use the existing infrastructure? How do we represent the incumbent? How do solar fuels relate to other fuel technologies such as hydrogen or electricity?
2. Argument: Abundant CO_2 and renewable energy can be used to provide scarce hydrocarbon fuels. Immediate LCA questions: Is CO_2 abundantly available for the production of fuels? Which technology do we need to supply sufficient CO_2 of good quality to produce these fuels?
3. Argument: Solar fuels are a way of storing solar energy. Immediate LCA question: Which other energy storage systems exist and how do these perform compared to producing fuels from CO_2?
4. Argument: By producing solar fuels one can offset costs of CCS by making valuable fuels and underground storage of CO_2 only costs money. Immediate LCA question: Is the main goal of CCS not to separate CO_2 from the atmosphere? Are we talking about two different functionalities, making money vs preventing CO_2 from entering the atmosphere, that cannot be compared vis a vis?

Asking these questions is not intended to disqualify the idea of making fuels from CO_2, but is vital in investigating if the production of solar fuels might indeed be a good way to go. Answering these questions would at least identify the conditions under which solar fuels might make a contribution to a sustainable future. Performing an ex-ante LCA with the cooperation of technology developers provides insights by discussing possible applications of foreseen technologies and brings to light the conditions under which a

technology is more likely to improve the status quo. It seems, however, that technology developers are not always comfortable with looking beyond the isolated efficacy of their technology alone, while it might be beneficial to at least be aware of other issues that do influence the overall environmental performance of a new technology in the end.

The case study in chapter 4 illustrates the need for attributing sufficient resources to defining a technologies application in its intended system as well. Rejecting HS-DAC to PCC based on costs or energy consumed per unit of CO_2 captured is like rejecting a car with high fuel consumption that seats six people and has big storage space over a smaller car with lower fuel consumption that only seats two people and has only limited storage space. That might be justified when the car's functionality is defined as driving in a city environment. However, when going on a family holiday the higher fuel consumption suddenly becomes less of an issue. It all depends on how you use a specific technology; without making a comparison in functionality, the comparison easily becomes arbitrary and unfair.

Another interesting insight from performing the casestudies is based on various critiques on the case studies during the journal review processes and informal discussions at conferences. It was often brought forward that the case study results report only support the obvious (e.g. that making fuels from CO_2 indeed demand more energy input than conventional fuels). Another recurring remark was that the analysis was performed on a supposedly wrong or suboptimal system of technologies for the production of solar fuels, or for using direct air capture to capture CO_2 from the atmosphere. Instead of providing insights based on the knowledge available, some saw the studies as being biased. These critiques seem to be founded in common engineering practices, in which the conventional stand is about getting a technical system to work. This automatically leads to optimizing specific process parameters such as process efficiencies and process production yields. These are then translated as having the lowest economic cost per ton CO_2 captured. It could be argued that a lower process efficiency also provides a lower systems efficiency which is necessary to improve the status quo situation. This can however only be investigated by taking a systems perspective. As a consequence of this experience it is recommended to put more effort in expanding the classical engineering approach and practice to include taking a systems view.

6.3 Question 3 - Can we find generalizing principles to assess the future environmental impacts of technologies in early stage of development?

Experiences from the case studies described in chapters 3 and 4, supplemented by reviewing existing studies that use LCA to asses new technologies provide valuable insights in the practice of ex-ante LCA. The main challenges for ex-ante LCA and

recommendations to overcome these challenges are reported in chapter 5. In this section these findings are generalized with the aim to provide guidance for upcoming ex-ante LCA studies.

The first challenge for ex-ante LCA is modelling consistent future foreground systems for both the incumbent and the new technology. A second challenge is to model a future background systems that is consistent with the foreground system. The final challenge is dealing with highly uncertain information that is inherently connected to forward-looking assessments.

A general recommendation for dealing with these challenges is of a more philosophical nature. In order to perform an ex-ante LCA the assessment requires a shift from a positivist approach - in which one assumes that the world can be fully known by observation- to a more constructivist approach in which one assumes that the world cannot be fully known because of different sources of knowledge that cause differences in interpretation of observations. Ex-ante LCA studies encounter such high uncertainties and require considerate assumptions that it is more informative to base the study performed on debate rather than on fixed statements that are gratuitously presented as correct. The ongoing debate uses and provides information from the LCA model constructed.

It is recommended to follow the existing conventional LCA framework when performing an ex-ante LCA. The following questions should, however, be covered and preferably answered explicitly per LCA phase.

1. Goal and scope phase
    - At what moment in time is the new technology expected to be operational at which level of maturity?
    - How does the functional unit need to be defined so that the new and the incumbent technology provide the same (similar) functionality?
    - How can the incumbent technology be defined?
2. LCI phase
    - How will new technological systems develop into the future, perform under the scope defined and is representative LCI data available?
    - How will incumbent technological systems develop into the future, perform under the scope defined and is representative LCI data available?
    - How will background systems and their performance develop over time, perform under the scope defined and is representative LCI data available?
    - What is the potential market share of the new technology in the future?

3. Impact assessment phase
    - Can the new technological systems display unexpected new impacts not yet covered in LCIA?
    - Can characterisation factors relevant to the incumbent and new technical systems change over time?
4. Interpretation phase
    - How can the increased uncertainty around modelling new technologies in the future be dealt with?
    - How can the possibility that new technologies will display unknown unknowns be dealt with?

Answering these questions might not always be the direct forte of an LCA practitioner and it involves branching out towards other scientific disciplines and practices such as social sciences, responsive evaluation, scenario building, integrated assessment modelling and more advanced techniques to deal with increased uncertainties like the concept of deep uncertainty. Consequently, this means that in an ex-ante LCA project more effort should be attributed to involving the right stakeholders and experts on the additional methods proposed. Different disciplines provide different insights that are all valuable in answering the bigger questions of sustainability. While doing ex-ante LCA it is preferable to investigate these differences and integrate the outcomes as part of an ex-ante LCA study.

Interest in performing ex-ante LCA studies to guide emerging technology has recently increased and a variety of recommendations to perform such studies have been published (Thonemann et al. 2020; Buyle et al. 2019; van der Hulst et al. 2020; Moni et al. 2020). It seems that a lot of research on the same topic has been performed in parallel, showing the need for guidance in performing ex-ante LCA. In general they all come to similar conclusions regarding the challenges of performing ex-ante LCA, which are *comparability* (choosing the functional unit and system boundaries), *data availability* (no data for the future) and uncertainty. Where Moni et al. (2020) and Thonemann et al. (2020) focus on presenting a framework on how to perform an ex-LCA study. Like is done in chapter 5 of this thesis Van der Hulst et al. (2020) and Buyle et al. (2019) focus more on the practical engineering approach development of new technologies and aim for providing knowledge to the technology development process than setting up the perfectly performed LCA study,. They do this by introducing using problem solutions spaces and participatory approaches (Buyle et al. 2019) or by focussing on the knowledge needed and available in the different phases of technology development (van der Hulst et al. 2020). As a final comment all emphasize that there is a need for a database to provide data on upscaling from lab to industrial scale technologies and on

new materials that are developed and used for new technologies.

6.4 Reflections and suggestions for future research

This thesis illustrates that using LCA in the R&D stage of new technologies, provides valuable insights in potential environmental impacts of design choices. This thesis shows that methodological challenges that are already common in conventional LCA are emphasized even more in ex-ante LCA. Applying LCA to new technologies without giving these challenges proper attention will not provide any insights to the development of better technology.

The use of LCA for the identification of potential environmental impacts of new technologies is a very new field. Fortunately, more and more methodological experiences are being shared even though a general method for ex-ante LCA does not yet exist. This thesis aimed to provide recommendations on performing such ex-ante LCA by reporting experiences from the assessment of new energy technologies. The set up presented in chapter 5 is a solid starting point for application and further development of this method.

A critical step back is taken below to identify limitations and potential bottlenecks that need to be the focus of further research in this field.

Ex-ante LCA is about the process not the numerical outcomes per se

When reflecting on the relevance of the work presented in this thesis it seems that it is not so much the numerical outcome of the assessment that are important, but rather the insights one gains during the assessment. As stated by Mc Kone et al. (2011) in their LCA research on biofuels "LCA is a process and not a product," a statement that is well known by practitioners inside the LCA community which should also be communicated more explicitly to commissioners of (ex-ante) LCA research and technology developers.

The view on technology development coming from the work performed in this thesis is that the essence of technology development seems to be in developing technical solutions for big problems. The focus is often on getting the technology to work at the lowest possible costs. Presented environmental gains seem to be mainly based on intuition or taken for granted. For example, assuming that using solar energy or CO_2 in a technology is by definition sustainable might be the start of a hypothesis but does not prove this hypothesis to be correct. To investigate a hypothesis like this you need analytical tools that take a systems approach, e.g. LCA. However, if LCA is applied in technology development, it seems to be used to provide a stamp of approval for the technology being developed, not as a critical tool that helps to gain insights.

This can be illustrated using an anecdotal example from the field of artificial photosynthesis. In this technical scientific field, the aim is to develop technologies that produce hydrogen from solar energy and water. In a discussion on the use of iron or platinum as a catalyst to split water into hydrogen and oxygen, the proposal to investigate this in a broader perspective system was ignored because the focus was on obtaining the highest water-splitting efficiency and using indium would provide this high efficiency.

Taking a systems perspective one could have investigated the hypothesis that using iron, although providing lower efficiencies, would be less costly and less disadvantageous for the environment because it is more abundantly available than platinum. It would have been very insightful to investigate if the lower water-splitting efficiency based on using iron catalyst could be compensated by using more production facilities based on iron that in the end could provide comparable quantities of solar hydrogen at potentially lower environmental costs.

One side remark is in place here; technology developers working on new technologies with an MRL < 5 are still focusing on getting the technology to work and the fundamental science involved. Taking a systems perspective to investigate broader implications so early in research probably distracts efforts and resources from this fundamental step. However, using claims about future applications for new technologies are often required to obtain grants to do this research. We should be careful that these claims are not empty and based on intuition alone. It should be acknowledged that when claiming sustainability of a new technology that "for any specific technology, there is no set of design characteristics that would make it sustainable. Rather sustainability of a technology can only be determined through a socio-political process, as the process has to deal with various human capabilities and preferences. Engineers may be uncomfortable with this truth, but we do, both individually and collectively, have to humbly learn to live with this" (Mulder et al. 2011).

Applying ex-ante LCA in technology development almost automatically forces technology developers and all involved to look beyond the pure technological performance of a technology. For the development of technologies intended to make our future more sustainable it is important to take a systems perspective for which ex-ante LCA could be an option. Inviting the use of ex-ante LCA in the R&D process might indeed feel uncomfortable because it does not give one unambiguous answer on which design choices to favour. It could even show that primary intuitive claims on environmental gains turn out to be less favourable when quantified, moreover ex-ante LCA will even provide more questions to answer which hampers quick development of a new technology. However, ex-ante LCA does provide quantification of potential environmental claims and insights

in the dependence on the context of a technology. These numbers and insights support making better-informed design decisions. The challenge in practicing ex-ante LCA is to support technology development by investigating solutions for potential barriers, while R&D engineers should allow the process to actually take place by acknowledging the importance for a systems perspective in R&D.

When to apply ex-ante LCA?

Throughout this thesis the question of when and where in the R&D process ex-ante LCA should be deployed has been lingering. The Collingridge dilemma still seems to apply here. If ex-ante LCA is applied too early, it interferes with the fundamental science needed to develop new technologies, if we do this too late, required changes cannot be initiated because of technical lock-inn.

A practical way of answering this question is to make use of the MRL of a technology. A critical note is in place here. In reality, it is not straightforward to assign an MRL level to a technology because in reality technology development does not start from scratch (MRL 1). It starts by combining different technologies at different development levels into a technology for which the application will become clearer when its MRL increases. When the technology up for assessment can be defined as "a body of technologies" (Figure 1.3) using MRL levels seem to be informative and allow for giving practical recommendations.

Ex-ante LCA is best applied to technologies at MRLs between ~3 and ~7. Technologies at an MRL below 3 are mainly subject to fundamental science for understanding basic (natural) principles. In this phase R&D is still far from managing these principles and it is therefore very hard to identify possible applications of the knowledge developed. Since it is fundamental to define the potential functionality of a new technology as a first step in the analysis, performing ex-ante LCA on technologies with MRL < 3 is not recommended. Qualitative discussions on potential application of a technology might, however, provide fruitful insights for shaping expectations. Data is still too uncertain and submerged in the realm of indeterminacy (see section 5.4) in this phase for quantification of environmental claims, and therefore ways to deal with these high uncertainties in ex-ante LCA need to be found.

Knowledge on technologies at an MRL between 3 -7 are becoming more concrete. It is easier to identify potential applications and come up with possible scenarios at these levels also there is still considerable freedom to change specific parts of the design of the technology under development. When MRLs rise above 7, changes in design might have become too difficult.

Future improvements in the application of ex-ante LCA

Having arrived at the end of this thesis, the concluding question is: how do we proceed from here? As presented in chapter 5, there are three practical areas for further research and development of the ex-ante LCA method. First, there is a need for experience in using scenarios that allow representation of new energy technologies and incumbents in a foreseen future. Second, to obtain representative future background data there is a need for further integrating the use of Integrated Assessment Modelling and LCI databases. Third, future assessments encounter increased levels of uncertainty. It should be investigated how to deal with these, for which a good starting point seems to be integrating *responsive evaluation* in the process of ex-ante LCA and investigating the possibilities of using the concept of *deep uncertainty*.

Overall, it is important to understand that ex-ante LCA is a process in which relevant questions for technology development are dealt with. Answering these questions demands the involvement of the relevant stakeholders and facilitates the necessary discourse that leads to understanding the system in which these technologies are developed. When we understand that system it is also possible to identify the boundary conditions for new technologies to contribute to a sustainable system. Technology developers can influence part of these conditions (choices in materials and supporting technologies), by making changes in design. Other conditions are the subject of governance, in which it might be possible to alter the context of technology development, deployment and use. An ongoing discourse between technology developers and policymakers should therefore always been pursued and LCA can provide valuable input via quantification of certain options under discussion.

A last challenge is to determine who should be responsible for using and guiding the ex-ante LCA process. A good start seems to be LCA practitioners with a multidisciplinary background such as Industrial Ecology. In the end however, main insights come from experiences. To build experience in doing ex-ante LCA technology development projects should be encouraged to use ex-ante LCA and report their experience. This might even help to make engineers more comfortable with taking a systems perspective that is utterly important for developing new technologies that contribute to reaching climate goals.

Chapter 7

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Summary

The development of new environmentally sound technologies is seen as a key route towards achieving sustainability. Also technology is regarded as the most important factor in the scientific field of industrial ecology for reducing environmental impacts of anthropogenic action. Developing greener, cleaner and more efficient technologies (using fewer resources or using them more efficiently) has therefore increasingly become the focus of many research projects that include the need to assess the potential future environmental impacts of technologies in the early development stages. The aim of this thesis was to develop a forward-looking assessment method based on LCA that can be used to integrate early insights in potential environmental impacts in R&D, with a specific focus on new energy technologies.

The research performed was structured around three complementary and successive research questions:

1. "What boundary conditions are required for LCSA to be a useful approach to assess early stage technologies?" (chapter 2)
2. "Which challenges are encountered in practice when LC(S)A is applied to assess early stage technologies?" (chapter 3 and 4)
3. "Can we find generalizing principles to assess the future environmental impacts of technologies in early stage of development?" (chapter 5)

At the start of the thesis research trajectory, life cycle sustainability assessment (LCSA) was the most comprehensive form of LCA available. A logical point of departure for this thesis was to therefore start with investigating the applicability and usefulness of LCSA for the assessment of new technologies (question 1). Since shortcomings of the LCSA approach were found, the decision was made to perform two LCA case studies concerning two technologies in early stages of development to investigate the challenges that are encountered in practice when performing an LC(S)A study (question 2). Finally, all insights were combined in formulating a general approach to guide upcoming ex-ante LCA studies (question 3).

Chapter 2 answers question 1 and explores the boundary conditions for the LCSA framework to be useful in the early assessment of new technologies. The conclusion of chapter 2 is that LCSA as a holistic, open method is not specific enough for performing a dedicated ex-ante LCA of novel technologies. An LCSA study should be initiated with a broad, but relevant system description. A missing thorough system description leaves ample room to manoeuvre in discussing the efficacy of a new technology, but

hampers the execution of a strict assessment. This makes it difficult to give developers of new technologies precise guidance on pathways with which the environmental impacts of such new technologies can be minimized. Starting the assessment with a thorough system description enables answering questions on a new technology's potential impact in a concrete manner that provides insights for further R&D of these technologies.

Chapter 3 reports an LCA case study on the production of fuels from CO_2. There is an increasing interest in using CO_2 as a resource to produce a new form of liquid hydrocarbon fuels. When these fuels are produced by solely using solar energy, they are labelled as solar fuels and claimed to be sustainable. Chapter 3 takes a quantitative approach to investigate some of the sustainability claims concerning solar fuels. The life cycle performance of various classes of solar fuel processes using different primary energy and CO_2 sources were analysed. It was concluded that producing liquid hydrocarbon fuels starting from CO_2 by using existing technologies requires much more energy than existing fuels. An improvement in life cycle CO_2 emissions is only found when solar energy and atmospheric CO_2 are used.

Chapter 4 reports an LCA case study in which mono-ethanolamine (MEA) based post-combustion-capture (PCC) of CO_2 with a new technology called humidity-swing-based DAC (HS-DAC). Most carbon capture and storage (CCS) envisions capturing CO_2 from flue gas by using post combustion capture where DAC is deemed unviable because of the higher energy associated with CO_2 capture at low atmospheric concentrations. Chapter 4 looks beyond the capture process alone and shows that given suitable temperature, humidity, wind and water availability, HS-DAC can be largely passive and therefore HS-DAC can complement MEA-PCC in suitable geographies. In addition HS-DAC also enables CO_2 capture independent of time and location of emissions and van re-capture background GHG emissions from fossil-based electricity beyond flue stack emissions.

The main insights from executing the case studies in **chapters 3 and 4 answer question 2**. First, for setting up an ex-ante LCA study, more effort and resources should be attributed to defining the function or future application of a new technology when performing an ex-ante LCA. The basis of any LCA model lies in defining system functionality and the functional unit. A simple rule of thumb in the field of LCA says that the function of a system should be chosen as close as possible to the "end use". In practice, this might require expanding the system boundaries to define a system in which all (or most) attributes of the technology under assessment are taken into account. The process of defining the technologies' application provides insights in its context consisting of competing technologies, but also (auxiliary) technologies required

to make the new technology function as defined. Although the future application of a new technology might be uncertain, providing a strict definition to start with provides the foundation to investigate this uncertainty further, give insights in the technological system and navigate the technology developers to taking a systems perspective.

Second, applying a systems view to assess potential impacts of different alternatives is often not straightforward in technology development. This insight is based on various critiques on the case studies during journal review processes and informal discussions at conferences. It was often brought forward that the case study results reported only support the obvious (e.g. that making fuels from CO_2 indeed demands more energy input than conventional fuels). Another recurring remark was that the analysis was performed on a supposedly wrong or suboptimal system of technologies for the production of solar fuels, or for using direct air capture to capture CO_2 from the atmosphere. Instead of providing insights based on the knowledge available, some saw the studies as being biased. These critiques seem to be founded in common engineering practices in which the conventional stand is about getting a technical system to work. This automatically leads to optimizing specific process parameters such as process efficiencies and process production yields. These are then translated as having the lowest economic cost per ton CO_2 captured. It could be argued - or at least should be investigated from a systems perspective- whether a lower process efficiency also provides a lower systems efficiency which is necessary to improve the status quo situation. As a consequence of this experience it is recommended to put more effort in expanding the classical engineering approach and practice to include taking a systems view.

Chapter 5 answers question 3 by listing methodological challenges and providing potential remedies or a route forward for dealing with these challenges. The first challenge is how to model consistent future foreground systems for the incumbent and new technology systems. Learning curves and scenario approaches are the way forward. The second challenge is how to model future background systems. A solution here to transform existing LCI databases towards future contexts informed by the Integrated Assessment Models (IAMs) that provide scenarios in line with the Shared Socioeconomic Pathways (SSPs). Finally, uncertainty in ex-ante LCA is of a different nature as in ex-post LCAs. The main difference with conventional LCA studies is the highly uncertain information for the future. To acknowledge this, considerable attention should be attributed to the discussion on these uncertainties, both in the design of the assessment and the data used. Responsive evaluation can play a supportive role here. This will increase the transparency and efficacy of the results because the relevant stakeholders and experts are involved. In this way, technology designers and other stakeholders derive insights on the influence of design choices or contextual factors (that are important, but

hard to influence) on the potential environmental impacts of their foreseen technology.

Chapter 6 concludes that methodological challenges that are already common in conventional LCA are emphasized even more in ex-ante LCA and that ex-ante LCA is about the process not the numerical outcomes per se. It is the experience gained during the assessment that provides the insights, while the numbers used and numerical outcomes of the assessment help in explaining these insights. The experience gained from this research provides the following recommendations to improve the existing practice of ex-ante LCA. First, there is a need for experience in using scenarios that allow representation of new energy technologies and incumbents in a foreseen future. Second, to obtain representative future background data there is a need for further integrating the use of Integrated Assessment Modelling and LCI databases. Third, future assessments encounter increased levels of uncertainty. It should be investigated how to deal with these, for which a good starting point seems to be integrating responsive evaluation in the process of ex-ante LCA and investigating the possibilities of using the concept of deep uncertainty.

Samenvatting

De ontwikkeling van nieuwe milieuvriendelijke technologieën wordt gezien als een belangrijke stap naar een duurzame wereld. In het wetenschappelijke veld van de industriële ecologie wordt technologie beschouwd als de belangrijkste factor voor het verminderen van de milieueffecten van menselijk handelen. Het ontwikkelen van groenere, schonere en efficiëntere technologieën (bijvoorbeeld door minder of efficiënter gebruik van grondstoffen) is daarom in toenemende mate de focus van veel onderzoeksprojecten. In deze projecten speelt het onderzoeken van potentiële toekomstige milieueffecten van nieuwe technologieën een steeds grotere rol. Het doel van dit proefschrift was het ontwikkelen van een toekomstgerichte beoordelingsmethode gebaseerd op levenscyclus analyse (LCA). Deze methode kan worden gebruikt om in een vroeg ontwikkelstadium van nieuwe technologie de potentiële milieueffecten te onderzoeken en deze inzichten mee te nemen in de ontwikkeling en het ontwerp van nieuwe energietechnologieën.
Het uitgevoerde onderzoek is opgebouwd rond drie complementaire en opeenvolgende onderzoeksvragen:

1. "Aan welk randvoorwaarden moet de LCSA methode voldoen, wil dit een nuttige methode zijn om nieuwe technologieën in een vroeg stadium van ontwikkeling te beoordelen?" (Hoofdstuk 2)
2. "Welke uitdagingen komen in de praktijk voor wanneer LC(S)A wordt toegepast om technologieën in een vroeg stadium van ontwikkeling te beoordelen?" (hoofdstuk 3 en 4)
3. "Kunnen we algemene principes vinden om de toekomstige milieueffecten van technologieën in een vroeg ontwikkelingsstadium te beoordelen?" (hoofdstuk 5)

Bij de start van dit promotie onderzoek was *Life Cycle Sustainabiity Analysis* (LCSA) de meest uitgebreide vorm van LCA beschikbaar. Een logisch uitgangspunt voor dit proefschrift was dan ook om te beginnen met het onderzoeken van de toepasbaarheid en bruikbaarheid van LCSA voor de beoordeling van nieuwe technologieën (vraag 1). Omdat tekortkomingen in de LCSA-aanpak werden gevonden, werd besloten om twee LCA-casestudy's uit te voeren met betrekking tot twee technologieën in vroege stadia van ontwikkeling. Door uitvoering van deze casestudies kon worden onderzocht welke praktische uitdagingen zich voordoen als LCS(S)A wordt toegepast in een vroeg ontwikkelstadium van een technologie (vraag 2). Ten slotte zijn alle inzichten gecombineerd in het formuleren van een algemene benadering voor toekomstige ex-ante LCA-onderzoeken (vraag 3).

Hoofdstuk 2 beantwoordt vraag 1 en verkent aan welke randvoorwaarden voor het LCSA-kader moet voldoen om nuttig te zijn voor de beoordeling van nieuwe technologieën. De conclusie uit hoofdstuk 2 is dat LCSA als holistische methode niet specifiek genoeg is voor het uitvoeren van een ex-ante LCA van nieuwe technologieën. Een LCSA-onderzoek dient te starten met een brede maar relevante systeembeschrijving. Het ontbreken van zo'n systeembeschrijving geeft dan wel voldoende speelruimte bij de kwalitatieve bespreking van de mogelijke milieu impact van een nieuwe technologie, maar belemmert het krijgen van kwantitatief inzicht van de oorzaken van mogelijke milieu impact. Dit maakt het moeilijk om ontwikkelaars van nieuwe technologieën specifieke handvatten te geven over ontwikkelkeuzes waarmee de milieueffecten van dergelijke nieuwe technologieën kunnen worden geminimaliseerd. Door de beoordeling te starten met een brede systeembeschrijving, kunnen vragen over de potentiële impact van een nieuwe technologie concreet worden gemaakt en specifiek worden beantwoord, wat inzicht biedt voor verdere research & development van deze technologieën.

Hoofdstuk 3 rapporteert een LCA-casestudy over de productie van brandstoffen uit CO_2. Er is toenemende belangstelling voor het gebruik van CO_2 als grondstof in alternatieve routes voor de productie van vloeibare fossiele brandstoffen. Brandstoffen die worden geproduceerd uit CO_2 en door gebruik te maken van zonne-energie, staan ook wel bekend als *solar fuels,* waarvan wordt beweerd dat ze duurzaam zijn. Hoofdstuk 3 gebruikt LCA om enkele duurzaamheidsclaims met betrekking tot *solar fuels* te onderzoeken. De conclusie is dat het produceren van vloeibare fossiele brandstoffen uit CO_2 met bestaande technologieën veel meer energie kost dan conventionele productie routes. Een verbetering van de totale CO_2 uitstoot over de gehele levenscyclus wordt alleen gevonden als gebruik wordt gemaakt van zonne-energie en CO_2 die wordt afgevangen uit de atmosfeer.

Hoofdstuk 4 rapporteert een LCA-casestudy waarin twee manieren om CO_2 af te vangen worden vergeleken. Het gaat om *post combustion capture* (PCC) waarbij met behulp van mono-ethanolamine (MEA) CO_2 wordt afgevangen uit de rookgassen van een elektriciteitscentrale en een nieuwe technologie genaamd *humidity swing direct air capture* (HS-DAC) waarmee CO_2 rechtstreeks uit de atmosfeer wordt afgevangen.

Het meest voorgestelde systeem voor CO_2-afvang en -opslag (CCS) maakt gebruik van de afvang van CO_2 uit rookgassen. De technologie voor het afvangen van CO_2 uit de atmosfeer wordt als niet levensvatbaar geacht vanwege de hogere energiebehoefte die gepaard met de lage CO_2 concentratie in de buitenlucht. Hoofdstuk 4 kijkt echter verder dan het CO_2 afvangproces alleen en laat zien dat HS-DAC, afhankelijke van de geschikte weersomstandigheden (temperatuur, luchtvochtigheid, wind) en

waterbeschikbaarheid, grotendeels passief kan opereren, waardoor de energie behoefte laag is. HS-DAC is daarom in specifieke geografische zeer geschikt als aanvulling op de MEA-PCC technologie. HS-DAC kan CO_2-afvang onafhankelijk van tijd en emissie-locatie mogelijke maken en tevens ook compenseren voor andere broeikasgassen dan CO_2.

De belangrijkste inzichten uit de casestudy's in **hoofdstukken 3 en 4 beantwoorden vraag 2**. Ten eerste kost het opzetten van een ex-ante LCA-studie meer inspanning en middelen. Bijvoorbeeld in het definiëren van de functie of toekomstige toepassing van een nieuwe technologie. De basis van elk LCA-model ligt in het definiëren van systeem functionaliteit en de functionele eenheid. Een simpele vuistregel op het gebied van LCA zegt dat de functie van een systeem zo dicht mogelijk bij het 'eindgebruik' moet worden gekozen. In de praktijk kan dit betekenen dat de systeemgrenzen moeten worden uitgebreid om een systeem te definiëren waarin rekening wordt gehouden met alle (of de meeste) kenmerken van de te beoordelen technologie. Het proces waarin de mogelijke toepassing van een nieuwe technologie wordt gedefinieerd biedt ook inzicht in de relevante context. Deze bestaat uit concurrerende technologieën maar ook (hulp) technologieën die nodig zijn om de nieuwe technologie te laten functioneren zoals gedefinieerd. Hoewel de toekomstige toepassing van een nieuwe technologie misschien onzeker is, biedt het geven van een strikte definitie om de analyse mee te beginnen de basis om deze onzekerheid verder te onderzoeken. Daarnaast geeft deze inzicht in het totale technologische systeem en technologie-ontwikkelaars inzicht in de milieueffecten op systeemniveau in plaats van proces niveau

Ten tweede blijkt dat het toepassen en gebruiken van een systeem benadering om potentiële effecten van verschillende alternatieven te beoordelen niet gebruikelijk is in de praktijk van technologieontwikkeling. Dit inzicht is gebaseerd op verschillende kritieken op de casestudy's. Vaak werd naar voren gebracht dat de gerapporteerde case study-resultaten alleen het voor de hand liggende ondersteunen (bijv. dat het maken van brandstoffen uit CO_2 inderdaad meer energie vereist dan conventionele brandstoffen). Een andere terugkerende opmerking was dat de analyse werd uitgevoerd op een zogenaamd verkeerd of suboptimaal systeem. In plaats van inzichten te verschaffen op basis van de beschikbare kennis, zagen sommige technologie ontwikkelaars de uitgevoerde analyses als vooringenomen. Deze kritiek lijkt te zijn gebaseerd op de gangbare technische praktijk waarbij de focus primair ligt op technische werkzaamheid van een technisch systeem terwijl men lijkt te vergeten dat het argument om deze technologie te ontwikkelen is om de milieu impact te verminderen. Dit leidt automatisch tot het optimaliseren van specifieke procesparameters zoals efficiëntie en opbrengst. Deze worden vervolgens vertaald naar de laagste economische kosten per afgevangen ton

CO_2. Vanuit systeemperspectief kan worden beargumenteerd of in ieder geval worden onderzocht of een lagere proceseffici‌ëntie ook een lagere systeemeffici‌ëntie oplevert die nodig is om de status quo-situatie te verbeteren. Als gevolg van deze ervaring wordt aanbevolen om meer moeite te doen om de klassieke technische benadering en praktijk uit te breiden met systeem analyses.

Hoofdstuk 5 beantwoordt vraag 3 door methodologische uitdagingen voor ex-ante LCA te identificeren en mogelijke oplossingen te bieden voor deze uitdagingen. De eerste uitdaging is het modelleren van consistente toekomstige voorgrondsystemen voor de reeds bestaande en de nieuw voorgestelde technologiesystemen. Het gebruik van leercurven en scenario's bieden hier uitkomst. De tweede uitdaging is het modelleren van toekomstige achtergrondsystemen. Een oplossing is hier om bestaande LCI-databases te transformeren naar een mogelijke toekomst door deze te combineren met informatie uit *Integrated Assessment Models* (IAM's). Ten slotte is de onzekerheid in ex-ante LCA's van een andere orde dan de onzekerheid in conventionele (ex-post) LCA studies . Het belangrijkste verschil met conventionele LCA-onderzoeken is de zeer onzekere informatie voor de toekomst. Om dit te erkennen, moet veel aandacht worden besteed aan de discussie over deze onzekerheden, zowel bij de opzet van de studie als bij de gebruikte gegevens. Benaderingen uit de sociale wetenschap zoals *responsieve evaluation* kunnen hierbij een ondersteunende rol spelen. Deze benadering vergroot de transparantie en bruikbaarheid van de resultaten doordat de relevante stakeholders en experts worden betrokken. Op deze manier ontlenen technologie ontwikkelaars en andere belanghebbenden inzicht in de invloed van ontwerpkeuzes en contextuele factoren (die belangrijk, maar moeilijk te beïnvloeden zijn) op de potentiële milieueffecten van de door hun voorziene technologie.

Hoofdstuk 6 concludeert dat de methodologische uitdagingen die reeds aanwezig zijn in conventionele LCA's nog uitdagender zijn in ex-ante LCA. Daarnaast blijkt dat ex-ante LCA inzichten verschaft door de uitvoering van de studie zelf en dat de numerieke resultaten op zich minder relevant zijn hoewel deze wel de discussie over de inzichten faciliteren. De ervaring die met dit onderzoek is opgedaan, biedt de volgende aanbevelingen om de bestaande praktijk van ex-ante LCA te verbeteren. Ten eerste, er is grote behoefte aan ervaring met het gebruik van toekomstscenario's op het gebied van nieuwe energietechnologieën als wel van reeds gevestigde technologieën. Ten tweede is er, om representatieve toekomstige achtergrondgegevens te verkrijgen, behoefte aan verdere integratie van het gebruik van Integrated Assessment Modeling en LCI-databases. Ten derde zijn ex-ante LCA studies onderhevig aan toenemende onzekerheid. Er moet worden onderzocht hoe met deze onzekerheden om te gaan. Een goed uitgangspunt hiervoor lijkt het toepassen van *responsive evaluation* en de nu nog conceptuele benadering van hoge onzekerheid door middel van *deep uncertainty*.

Curriculum vitae

Coen van der Giesen was born on March 10, 1976 in Nieuwegein in The Netherlands. In 1995 he completed secondary school (VWO) at the Da Vinci College in Leiden. In the same year he started training as a Maritime Officer at the Maritime Institute de Ruyter (Hogeschool Zeeland) in Vlissingen, where he graduated in 1999. After graduation he worked as a maritime officer on several ships.

Curious about the system behind rules and regulations that he had experienced on board, he started studying Public Administration at Leiden University in 2002. He obtained her bachelor's degree in 2005. After this he worked as a policy researcher on the themes of working conditions and safety at Bureau KLB in The Hague.

In 2006 he started studying Industrial Ecology program at Leiden University, from which he obtained his master's degree in 2008. He performed his master thesis research within Shell for which he used the life cycle analysis (LCA) method to analyse the material requirements for CO_2 capture and storage.

From 2009 he worked as a consultant on energy, sustainability and environment at MWH (later Stantec) in Delft. In this capacity he advised several companies on the topics of energy efficiency and energy saving as well as quality systems, working conditions and environment.

At the end of 2011 he started as a PhD candidate at the Center for Environmental Sciences (CML) of Leiden University. After his first year, this position was expanded with various teaching tasks in the master's program Industrial Ecology and in the minor Sustainable Development. He developed, coordinated and taught courses on the various analytical methods, especially LCA, in the scientific field of Industrial Ecology field. In addition, he guided students on their graduation.

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