THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Towards Sustainable Shipping: Climate change and other environmental perspectives on carbon-based marine electrofuels and onboard carbon capture

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To the women in my life.

Abstract

The global, local, and regional environment is under pressure from human activity. Shipping is a human activity causing emissions to air, water, and soil, which has direct and indirect effects on the environment. New fuels and propulsion technologies are required to lower the emissions from the shipping sector and reduce the impact on the environment. Fuels produced through electricity, water, and carbon dioxide, so-called carbon-based electrofuels, are one group of fuels suggested to reduce the environmental impact of shipping. Another proposed solution is onboard carbon capture. The aim of this thesis is to promote further discussion on how to assess future marine fuels and propulsion technologies by establishing the environmental impacts of these emerging technologies.

A mixed methods approach to environmental assessment is used, combining thematic analysis, literature reviews, and life cycle assessment. Through case study applications, the environmental performance of electromethanol, electromethane, and onboard carbon capture are investigated when applied in the maritime sector in northern Europe. Thematic analysis is used to investigate what is hindering low-emission fuels from being further utilized in maritime cargo transportation.

The results show that if renewable energy is used in fuel production and CO_2 is captured from a source not acting as a driver of fossil fuel extraction, climate change impact can be reduced by using carbon-based electrofuels instead of fossil fuel options. Potential trade-offs were identified as carbon-based electrofuels can lead to higher pressure on human health impacts than today's conventional fuels. The extent of the trade-offs is uncertain and affected by limitations in the methodological approach. Suggestions on how to address these uncertainties are introduced and analyzed. Assessment of future scenarios for large-scale marine electromethane production in Sweden reveals that combined biofuel and electrofuel production likely results in the lowest environmental impacts. Onboard carbon capture can lower the climate change impact if combined with electrofuel production or carbon capture and storage. The environmental impacts at large depend on the bunkered fuel and the choice of carbon capture technology.

The results underscore the importance of integrating life cycle assessment with other scientific methodologies. The environmental impacts of capital goods should be included in life cycle assessments of future marine fuels, and scenario-based assessments are preferable over single-vessel evaluations.

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My friends. You know what you mean to me. I am sorry for having disappeared from the face of the planet in the past years. My reasons are enclosed in this book. I hope you accept my apology.

In addition to the wonderful people mentioned above, I want to express my appreciation to my family and the life skills you've passed on to me. For my mother's thoughtfulness and dedication. For my father's ingenuity and argumentative skills. For my sister unwavering trust in the good of man. I love you.

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LIST OF APPENDED PAPERS

PAPER A

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M. G.: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Visualization (Chapters 1, 2, 4, 6 and 7); Writing - Review & Editing, Supervision, Funding acquisition (entire paper), E. M.: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Visualization (Chapters 5 and 6); Writing - Review & Editing (entire paper), A. D. K.: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Visualization (Chapters 3 and 4); Writing - Review & Editing (entire paper), J. E. A.: Writing - Review & Editing (entire paper), M. T.: Software, Formal analysis, Investigation, Writing - Original Draft, Visualization, (Chapter 4); Writing - Review & Editing (entire paper), S. B.: Conceptualization, Writing - Review & Editing, Supervision (entire paper), J. H.: Writing - Review & Editing, Supervision (entire paper), I. R. S.: Writing - Review & Editing, Supervision (entire paper), T. J. W.: Writing - Review & Editing, Funding acquisition (entire paper)

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PAPER D

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LIST OF OTHER RELEVANT PUBLICATIONS

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Brynolf, S., M. Grahn, J. Hansson, A.D. Korberg, and **E. Malmgren**, Chapter 9 - Sustainable fuels for shipping, in Sustainable Energy Systems on Ships, F. Baldi, A. Coraddu, and M.E. Mondejar, Editors. 2022, Elsevier. p. 403-428.

Brynolf, S., J. Hansson, F.M. Kanchiralla, **E. Malmgren**, E. Fridell, H. Stripple, and P. Nojpanya, Nordic Roadmap Publication No.1-C/1/2022. 2022.

Kanchiralla, F.M., S. Brynolf, **E. Malmgren**, J. Hansson, and M. Grahn, Life-Cycle Assessment and Costing of Fuels and Propulsion Systems in Future Fossil-Free Shipping. *Environmental Science & Technology*, 2022. 56(17): p. 12517-12531.

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Malmgren, E., S. Brynolf, M. Grahn, J. Hansson, and K. Holmgren, The feasibility of alternative fuels and propulsion concepts for various shipping segments in Sweden, in 29th Conference of the International Association of Maritime Economists. 2021: Rotterdam.

Malmgren, E., Brynolf, S., Borgh, M., Ellis, J., Grahn, M., and N. Wermuth. The HyMethShip Concept: an investigation of system design choices and vessel operation characteristics influence on life cycle performance. Conference paper at Transport Research Arena Conference (TRA) 2020, conference canceled due to COVID-19 regulations. Peer reviewed.

TABLE OF CONTENTS

ABBREVIATIONS, ACRONYMS AND TERMINOLOGY

The nomenclature and terminology chapter outlines the definitions and abbreviations as used in this thesis. The exact usage varies within the research community, and as such this list should be viewed as definitions as they are used here.

Abbreviations

AD	Anaerobic digestion
Bio-e-fuels	Bio-electrofuels representing combined e-fuel and biofuel production using excess CO_2 from biofuel production
CBG	Compressed biogas
CCS	Carbon capture and storage
CCU	Carbon capture and utilization
C-H_2	Compressed hydrogen
CH_4	methane
CI	Compression ignition
CI-HPDI	Compression ignition high pressure direct injection
CNG	Compressed natural gas
CO	Carbon monoxide
CO_2	Carbon dioxide
DAC	Direct air capture of carbon
DME	Dimethyl ether
ELCD	European reference Life Cycle Database
eMeOH	Electromethanol
e-RLMG	Electromethane
FT	Fischer-Tropsch
GHG	Greenhouse gases
GJ	Gigajoule
GWP	Global warming potential
GWP100	Global warming potential over 100 years
H_2	Hydrogen
HFO	Heavy fuel oils
HyMethShip	The Hydrogen-Methanol Ship propulsion system using onboard pre-combustion carbon capture
ICEs	Internal combustion engines
IMO	International Maritime Organization
km	kilometer
LBG	Biogas

LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LMG	Liquified methane gas
LNG	Liquified natural gas
LPG	Liquefied petroleum gas
MEA	Monoethanolamide
MGO	Marine gas oil
MRV	Monitoring, reporting, and verifying
N_2	Nitrogen
N_2O	Nitrous oxide
NOx	Nitrogen oxides
OMEs	Oxymethylene ethers
PEM	Polymer electrolyte membrane/ Proton exchange membrane
PM	Particulate matter
PN	Particulate number
PSA	Pressure swing adsorption
RED	Renewable Energy Directive
RLMG	Liquified renewable methane gas
Ro-pax	Roll-on/Roll-off passenger vessel
SOx	Sulphur oxides
TA	Thematic analysis
TRL	Technology readiness level

Terminology

Allocation — The distribution of flows between multiple units.

Allocation problems — Allocation problems occur in an LCA when several products (or functions) share the same processes and the environmental loads of these processes need to be expressed in terms of a single product. Allocation can be achieved using, for example, a physical relationship or the monetary value of the products. Allocation is described here as one method for solving allocation problems. Thus, allocation methods include both allocation (also called partitioning) and system expansion.

Alternative fuels — Alternative fuels are fuels not commonly used in the shipping sector today i.e., fuels which take up a small proportion of the current market, are not available commercially in the harbors, or are only used on singular vessels.

Attributional LCA — An attributional LCA is one that strives to be as complete as possible by accounting for all environmental impacts of a product. This type addresses such questions as “What would be

	the overall environmental impact of marine transportation using Fuel A?”
Anthropocene	The period during which human activity has been the dominant influence on climate and the environment.
Boil-off gas	The gas created by the surrounding heat input (while maintaining constant pressure during storage of a cryogenic liquid such as liquefied natural gas) is called boil-off gas. Boil-off gas is inherent to the storage of a cryogenic gas due to the heat input from the surroundings.
Business-as-usual	The reference points informed by historical norms, i.e., no disruptive change occurs.
Characterization factors	Characterization factors are factors derived from a characterization model which are applied to convert an assigned life cycle inventory analysis result to the common unit of the category indicator. This is done to assess the total impact on the category. There are characterization factors both at midpoints and endpoints.
Consequential LCA	A consequential LCA is one that compares the environmental consequences of alternative causes of actions and evaluates the effects of change on a surrounding system. This type addresses such questions as “What would be the environmental consequence of using Fuel A instead of Fuel B?”
Downstream emission/s	Emissions generated by product and waste streams after they have exited the foreground system.
Elemental flows	Elemental flows are the flows between the environment and the technical system associated with each process in the system.
Endpoint	The endpoint is a point of interpretation of the aggregated emission flows. It represents the end in a cause-effect chain and may be of direct relevance to society’s understanding of the final effect, such as measures of biodiversity change.
Energy carriers	Energy carriers acts as transmitters of energy between the initial primary energy source and the end-use application. Examples include solid, liquid and gaseous fuels.
Eutrophication	Eutrophication is the increased availability of one or more limiting growth factors needed for photosynthesis leading to excessive plant and algal growth. Nitrogen and phosphorus are the most common growth-limiting nutrients.
Functional unit	A functional unit is a quantitative unit representing the function of the system. The use of a functional unit enables comparisons of various products that fulfil the same function.

Goal and scope	The goal and scope is the first step in an LCA. It describes the system under study and the purpose of the study. The goal should include, for example, the intended application and reasons for the study.
Human health	Human health is an area of protection. Damage to human health is measured by mortality and morbidity over space and time.
HyMethShip	The Hydrogen-Methanol Ship propulsion system using onboard pre-combustion carbon capture project.
Impact assessment	Impact assessment is the third step in an LCA. It includes classification of the elemental flows into various impact categories and the characterization of these flows, e.g., the calculated relative contributions of the emissions and resource consumptions to the impact categories.
Inventory analysis	Inventory analysis is the second step in an LCA. It consists of three parts: the construction of a flow model based on the system boundaries, the data collection and the calculation of resource use and emissions of the system in relation to the functional unit.
Life cycle inventory	The phase of LCA which involves the compilation and analysis quantification of inputs and outputs for a product throughout its life cycle.
Methane slip	Methane slip is the leakage of methane from marine engines.
Midpoint	Midpoints are links in the cause-effect chain (environmental mechanism) of an impact category. Common examples of midpoint characterization factors include ozone depletion potential and global warming potentials.
Natural environment	The natural environment is an area of protection. The impact on the natural environment is measured by the loss or disappearance of species and the loss of biotic productivity.
Natural resources	Natural resources are an area of protection. The natural resources can be divided into the following subcategories: atmospheric resources, land resources, water resources, mineral resources, metal ores, nuclear energy, fossil fuels and renewable resources.
Photochemical ozone	Photochemical ozone is an impact category that accounts for the formation of ozone at the ground level of the troposphere. Ozone formation is complex and depends on several factors, e.g., the concentrations of NO, NO_2 and VOC and on the level of ultraviolet radiation.
Prospective	This term, meaning forward looking, is used to denote LCAs looking at future systems.

Renewable fuels	Renewable fuels are fuels produced from renewable energy sources, where renewable energy sources refer to energy which is generated from natural processes and are constantly regenerated.
Retrospective	This term, meaning backward looking, is used to denote historic perspectives on LCA.
Ro-pax ferry	A ro-pax ferry is a roll-on/roll-off ship with high freight capacity and limited passenger facilities.
Swedish shipping context	Consists of the maritime activities taking place on Swedish territorial water, between Swedish maritime stakeholders or within the sphere of influence of the Swedish maritime authorities.
System	Connected objects, concepts, functions, etc. how they interact, and their purpose, goal, or effects make up a system.
System expansion	System expansion is an allocation model in an LCA. It implies the expansion of the system to include affected processes outside the cradle-to-grave system, or to include multiple functions into the system boundary.
Tank-to-propeller	In this study, this term is used for the part of a marine fuel's life cycle beginning when the fuel is delivered to the vessel's onboard tank and ending when it is combusted for transportation of goods and/or passengers.
Tail pipe emissions	Emissions in exhaust gases discharged from an internal combustion engine, i.e., emissions generated at the tail pipe.
Upstream emission/s	Emissions generated by a product or resource flow before they enter the foreground system.
Well-to-propeller	Used to describe the part of a marine fuel's life cycle from the acquisition of the raw material to when the fuel is combusted for transportation of goods and/or passengers.
Well-to-tank	Used to describe for the part of a marine fuel's life cycle from the acquisition of the raw material to the delivery to the vessel's tank.
Well-to-wheel	Well-to-wheel is a term commonly used in LCAs of road fuels. These studies usually consider only energy use and climate impact.

"It was the secrets of heaven and earth that I desired to learn."

– Victor Frankenstein, Frankenstein
by Mary Wollstonecraft Shelly

1 INTRODUCTION

Maritime transport is an essential part of the global economy which allows for raw materials, energy, and goods to be available across economies [1]. If we are to maintain large scale productions and globalization, shipping is likely essential, and energy to provide for this activity is therefore needed. However, emissions from the fuel use in maritime transport negatively impacts the oceans (e.g. [2]), human health (e.g. [3]), the climate (e.g. [4]), biodiversity, ecosystems and more. The shipping sector is currently contributing significantly to climate change (~3% of global anthropogenic greenhouse gas emissions [5]). Legislations, as well as regulations, have also been put on Sulphur and nitrogen oxides (NO_x) emissions from the tail pipe of the vessels, as well as energy efficiency requirements for newbuilt vessels, and most recently greenhouse gas (GHG) emissions. With new legislation now being introduced and an increased public interest in environmental issues, ways to incorporate environmental aspects into the maritime decision-making processes are increasingly relevant. Concepts and content for sustainable shipping is not yet fully established, and further research on environmental aspects of the maritime transport is required to further define the field [6].

1.1 Addressing environmental problems

From introduction of farming, which meant adaption of the surrounding environment for production, to emission of freons to the atmosphere damaging the ozone layer, humans have affected their surroundings [7]. In the ongoing anthropogenic age, the scale of these effects is increasing [8], with the current climate change being an example of human's interaction with the environment on a global scale [9]. Environmental problems can be defined as harmful effects on the biophysical environment developed because of human interference or mistreatment of the planet. They can range from local issues, such as water shortage due to over-usage [10], regional issues, such as eutrophication and acidification, and global issues, such as climate change [11]. The problems are often caused by conflicting interests regarding utilization or extraction of natural resources and the preservation of the environment [12].

Since the start of the modern environmental movement, environmental issues have become better understood, but most issues are not yet solved. The best action to take or decision to make for us humans and/or other agents are not always apparent[13], as for example reduction of tail pipe emissions in a car by increasing the fuel conversion efficiency might lead to lower costs for driving and by extension increased use of the vehicle, a so called rebound effect. Specific knowledge, of the environmental problem and which activities cause it, is therefore required when discussing environmental problems and the possible solutions [11].

It has been predicted that the fuel demand from the shipping industry will keep growing for the next decades to come, despite expectations on more energy efficient vessels (e.g. larger ships, improved hull forms and propellers and more fuel-efficient engines) [4, 14]. Transitioning the sector to other fuels is therefore one potential route to meet the implemented legislations, targets and reduce the pressure from the sector on the environment [4, 15, 16]. The environmental concerns regarding marine transportation and new regulations are driving forces behind the introduction of new marine fuels. However, choosing alternative marine fuels is a difficult task as many alternatives to fossil fuel oils have low technical maturity, have high costs, and/or are not available at scale [17]. An alternative fuel does also not necessarily perform better from an environmental perspective. To be an attractive alternative fuel from an environmental perspective, it must also be a low-emission fuel, associated with low emissions of all types of damaging emissions over its entire life cycle. To facilitate how to understand the term low-emitting fuel, see Figure 1 for illustration of where emissions occur in the life cycle. This thesis avoids using the wording “alternative fuel” as any fuel can be viewed as alternative as long as it is not conventionally used, and as such it does not give clarity to the aim of the thesis.

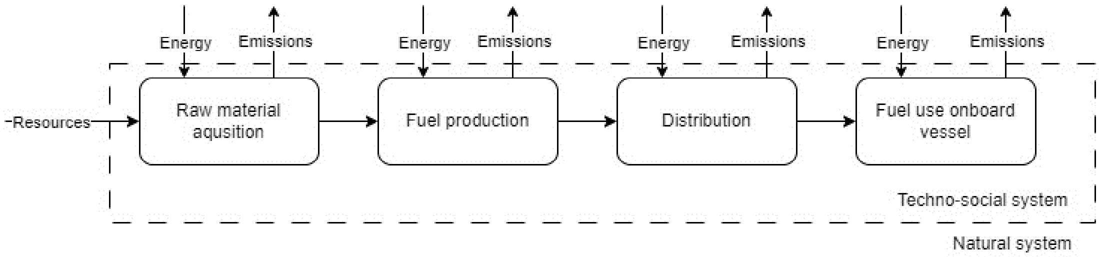

Figure 1 Simplified illustration of the life cycle of marine fuels and how emissions occur throughout the life cycle. The term emissions include all components which are emitted from the processes.

Understanding the climate impacts of carbon flows are essential in mitigating climate change. The strive to reduce climate change have led to two main strategies: reduce the carbon flows from society to the environment and to circulate the carbon flows.

One solution proposed to mitigate climate impact is carbon capture (CC) [18], combined with further utilization (CCU) or permanent storage (CCS). The maritime sector has two direct routes to utilize this technology: onboard the vessels in the form of onboard carbon capture (i.e., carbon previously emitted from the engine system is captured) or by shifting to synthetic variants of carbon-based fuels, often called electrofuels, e-fuels, or power-to-x [19-21]. These electrofuels can then be used in existing propulsion technologies but be produced through different feedstocks. But what are the environmental impacts of using these forms of CC in marine transport? And what is hindering the industry from moving to low-emission fuels today?

1.2 The research gap

In this thesis, carbon-based electrofuels are defined as liquid or gaseous energy carriers produced from water, electricity, and carbon dioxide (CO_2). This limits the definition of carbon-based fuels produced through synthetic processes. However, the definitions vary slightly between the appended papers and term electrofuels include also electrolytically produced hydrogen and ammonia in one appended paper (Paper B). Other research includes broader or more narrow definitions [21]. Two main resources are required for production of electrofuels as defined in the thesis: hydrogen (H_2) and CO_2. These two resources are then combined through fuel synthesis processes and treatments, which detailed set-up depends on the final fuel product.

Environmental impact assessments of marine fuels have been conducted for a multitude of different fuels, from competing fossil fuel options [22-30], to biofuel alternatives, [27, 31-33] and future options such as hydrogen [27, 34-39], or electricity [19-21]. Most of the papers are limited to climate change impact, but some look at a wider scope of impacts [23, 24, 27, 33, 40-43]. Carbon-based marine electrofuels are less investigated. The assessments of electrofuels environmental performance are acknowledged to be in the early method development stage [19, 44-48], but researchers have investigated the environmental effects of using electrofuels in other segments of the transport sector [49] as well as for energy storage [50, 51]. The environmental performance of carbon-based electrofuels have been investigated both in the form of electromethanol [22-30], electromethane [27, 31-33], and longer carbon chains.

Maritime vessels travel long distances, have life spans of 20-50 years, bunker large amounts of fuels with high energy density, and the maritime sector acts in a different decision-making context compared to the other transport sectors. To further investigate the case of carbon-based marine electrofuels is therefore of interest. Assessments of the environmental performance of onboard carbon capture are fewer in the literature. Through the work of this thesis, 46 papers [16, 52-97] investigating onboard carbon capture were identified of which two [82, 91] consider a broader scope of environmental impacts beyond greenhouse gas emissions.

1.3 Aim and research questions

The aim of this thesis is to establish under which conditions carbon-capture could have a role to play in an environmentally sustainable maritime sector, both when applied directly onboard and when utilized to produce carbon-based marine electrofuels. The work is motivated by the need to have a holistic picture of the potential environmental impacts from using the technologies, already before the technologies are implemented at scale. The need to identify potential future fuel production pathways is evident, and the thesis investigates if these emerging technologies could be viable options from an environmental perspective. The maritime cargo transport stakeholder’s role in the decision of marine fuel is studied to identify important fuel attributes.

This thesis will contribute to providing decision-makers with insights and tools to compare the environmental performance of carbon-based marine electrofuels with other alternatives and identify strategies for improving the involved sectors’ sustainability. The research questions are structured according to how they relate to the specific field, with maritime stakeholders in focus initially and general applications presented later. Specifically, this thesis addresses the following research questions:

RQ1) How do stakeholders on the maritime cargo market view their possibilities to choose a low-emission fuel today?

The uptake of emerging fuels will depend on how they are received by stakeholders on the maritime market. It is therefore interesting to assess how they view their possibilities to adopt low-emission fuels, and consequently relate these perspectives to carbon-based electrofuels.

RQ2) How can life cycle assessments of marine fuels be further developed to include carbon-based electrofuels?

Marine fuels are associated with environmental impacts over the entire fuel life cycle, and life cycle assessment is proven to be a useful tool to assess fossil fuels and biofuels for marine applications. However, the circular carbon flows associated with carbon-based electrofuels are not compatible with the previously linear LCA framework, and previous assessments have focused on less stringent environmental impact reduction targets. The marine fuel LCA is therefore reviewed, and proposals brought forward on how to further develop the methodology to account for the electrofuel life cycle.

RQ3) What are the environmental impacts of carbon-based marine electrofuels: under which conditions can they be defined as low-emission fuels?

It is important to be able to quantify the environmental impacts of carbon-based electrofuels to be able to consider the environmental aspects in the choice of marine fuels. The requirements for the environmental performance of marine fuels have become increasingly stringent, and therefore the conditions under which they achieve low-emission profiles are of particular interest to hopefully inform regulation and stakeholder decisions.

RQ4) Under which conditions could onboard carbon capture mitigate environmental impacts?

The increased interest in onboard carbon capture is primarily driven from the possibility to directly mitigate CO_2 emissions, but the overall environmental performance will depend on the full life cycle. To determine whether carbon capture would be an interesting option from an environmental perspective, it is important to understand which factors and circumstances that contribute to the overall environmental impacts.

RQ5) How can CO_2 be modeled in life cycle assessment of carbon capture and utilization (CCU) technologies?

Carbon-based electrofuels uses carbon as a resource, whereas it previously has been considered an emission. How the removal of carbon from the atmosphere should be treated in environmental assessments has not been fully established [19, 47], and there is therefore a need for a framework to compare biofuel, fossil fuels and carbon capture based fuel systems with a special attention to the different carbon flows. Figure 2 illustrates how the research questions relate to the appended papers.

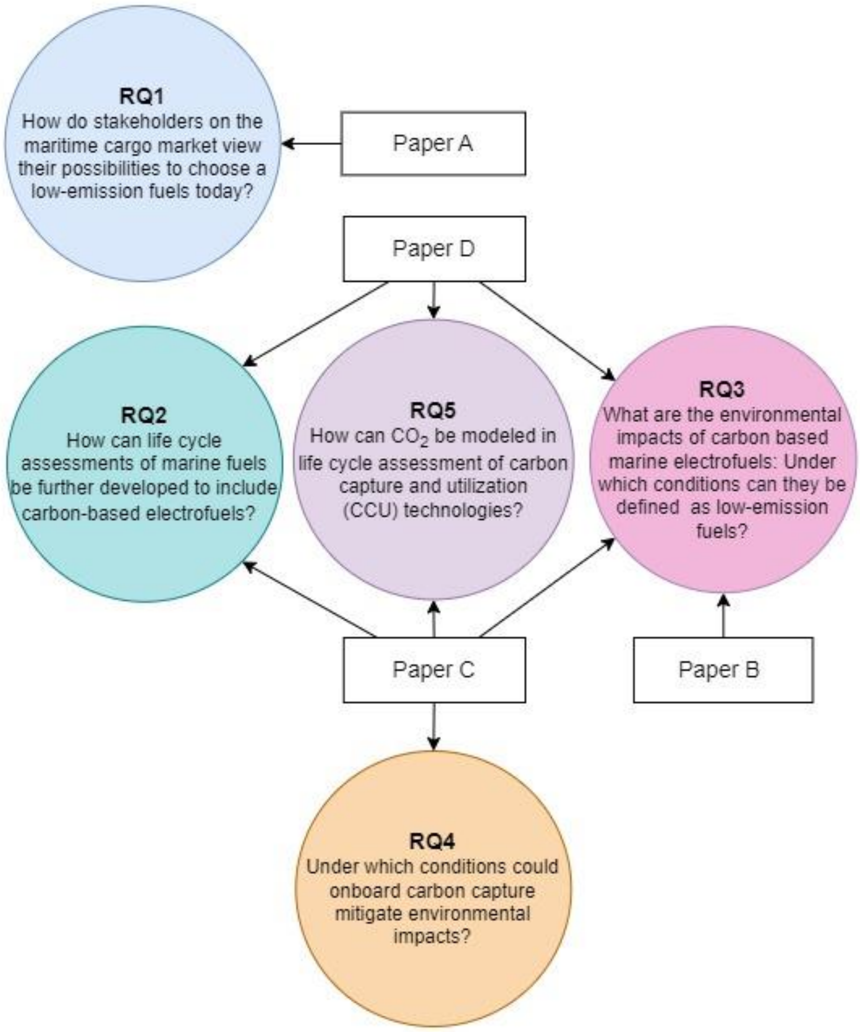

Figure 2 Research questions and their connection to appended papers.

1.4 The research context of this thesis

The research fields addressed in this thesis have grown significantly in recent years. This can be represented by 150 papers around sustainability of marine fuels in 2018 and 374 in 2023 in Scopus, 8 papers related to maritime decarbonization in 2018 and 295 in 2023, and 1 311 on electrofuels in 2018 and 3 435 in 2023. By the time Paper C was sent for review, only four papers had been published on onboard carbon capture: Luo and Wang [57], George et al. [53], Peilin and Haibin [54], Haibin et al. [56], Luo and Wang [57] and Wang et al. [58]. Today, 53 papers have been identified which study carbon capture onboard vessels. The first assessment (besides Paper C) studying the system from a life cycle perspective was published in 2022 [82], followed by [91] in 2023.

It is imperative to recognize and appreciate the broader context in which this thesis resides, wherein countless scholars and researchers have furthered the frontiers of knowledge in various disciplines. Papers B and C were created when there was a greater lack of data on carbon capture and utilization in scientific literature, and the papers contributed to the knowledge gap of those periods. The later papers reflect a later discourse where there is a higher knowledge utilization among the researchers as well as potentially the public.

1.5 Funding

The research presented in this thesis would not have been possible without the funding provided by several institutions since 2018. The work carried out during 2018-2021 was primarily funded through the Hydrogen-Methanol Ship propulsion system using onboard pre-combustion carbon capture (HyMethShip*)* project. The project was funded by the European Union's Horizon 2020 research and innovation program under grant agreement no. 768945. Funding was also provided from the European Union through the Interreg Baltic Sea Region program 2021-2027 for the HyTruck project, project number #C031.

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1.6 Delimitations and limitations

Delimitations are characteristics defined by the researcher to limit the scope and define the boundaries of the study [98]. The delimitations of this work include (in addition to the delimitations implied in Chapter 1.3):

- The field of environmental science is multifaceted and can be studied from a variety of different perspectives and research frameworks. This thesis takes an anthropogenic perspective, where the best for humans is in center of the assessment. This thesis examines the environmental impact of marine fuels from the perspective of the products' technical life cycles. By taking a life cycle approach a holistic assessment of the product is possible, but the pressures on individual ecosystems are not assessed.
- The work of this thesis is limited to assessing electrofuels in the context of today's conventional fuel options. The results are limited to investigating the environmental performance of carbon-based marine electrofuels and do not reflect assessment of which are the preferred future low-emission technologies.
- The context of this work is maritime transport, which differs significantly from other transport research domains. Although certain elements of the results can be applied to other sectors, the research has been designed for maritime research.
- The work presented in this thesis was developed with focus on the geographical area of Northern Europe, with a focus on Sweden. Most of the respondents were

from Northern Europe, predominantly Sweden. The results can therefore only be extrapolated with caution beyond northern Europe.
- The types of environmental impacts investigated are the main categories commonly used in environmental LCA and do not include any assessments focusing on noise pollution, or social aspects.

The limitations are factors within a study that are beyond the researcher's control [98]. The work presented is carried out during the time-period of 2019-2023. The known limitations of this thesis include:

- The work primarily employs life cycle assessments, which is a well-researched methodology with several known limitations. The models used for assessment, such as life cycle impact models, are limited to assessment at a specific timepoint and do not account fully for secondary and third-degree feedback-loops.
- This work is limited by the lack of available and high-quality data on technical performance, emission profiles and material demand for the assessed emerging technologies.
- The qualitative data was gathered through semi-structured interviews over video-conferences which may not capture the full context of the respondents' experiences.

2 BACKGROUND

2.1 Thinking in systems during the Anthropocene

The scientific principles applied in this thesis begin with systems thinking. To assess the impacts on the environment several methods and frameworks have been developed over the past 50 years. The start of looking at environmental impacts from a systems perspective is commonly credited [99], where large scale models were constructed to investigate the future impact of economic growth, but the scientific field has grown and developed rapidly.

Systems thinking, also called systems theory, has its roots in general systems theory developed by [100] among others, and has over the past 70 years developed into a wide research field with multiple applications. Systems thinking is the act of describing how different objects, concepts, functions, etc. interact and what purpose, goal, or effects they have. A system consists of several components and the interactions between them. Together, these components and interactions form a whole. When objects, factors, and their relationships are dependent on how they interact, and that interaction affects further consequences, it is not always given how the system looks in a given moment [99]. The relationship between the individual objects will influence each other, and knowledge of the relationships is therefore required. In his 2008 lecture on "Why model?", Epstein [101] talks about how a model is a way of structuring the world. He does not distinguish between conceptual models, systems models, or simple mathematical facts. A model is anything consisting of different parts and their interactions, portrayed in a way that creates an image for the beholder. It can be a physical image, an equation, or a story.

Drivers and dampeners, positive and negative feedback, creates a need for systems thinking [102]. Understanding these links and collecting knowledge on how they interact might not lead to explicit solutions, but systems thinking will make it possible to acknowledge trade-offs and connections between different linked components. The system's boundaries set the system's limits to the rest of the world, called the surroundings or environment. Interactions with the surroundings occur through input to or output from the system. A system can usually be divided into sub-systems. The sub-systems are considered part of the larger system but do themselves involve several objects and the interactions between them. There are thus many systems levels and the viewpoint from which you look at a system is central to the applied research questions.

There are various system types, including machinery systems, biological systems, social systems, socio-technical systems, and nature-society-technology systems [103]. This thesis focuses on the interaction between technology and nature but involves society; thus, socio-techno-environmental systems exist in this thesis. The natural, or environmental, systems may be understood in an ecological sense as the set of interactions between the elements of the biosphere. A technology system may be understood as the interactions between elements of technical components, or the full technological system.

Interactions between technical systems constructed by humans and the environment have occurred for centuries, but to understand these more complex systems interactions modelling and analysis is required. Technologies does not only interact with the environment in one point, but throughout the entire life cycle links occur between a technology, the user, and the environment through energy extraction, material use and emissions [13]. When determining how the Technosphere affects the global, regional, and local ecosystems how material and energy flows to and from the environment becomes essential.

2.1.1 Planning the future

The real world in which we all interact consists of endless combinations of objects, factors, and relationships, where the effect of different decisions is not always given. Since experimental research in a real-world system is not feasible, models are often needed when investigating complex decision-making situations with direct applications in the real world. A model is thereby something that strives to be an image of reality and then attempts to reflect the impact of a shift in either guiding principles, behaviors within the systems, or influence from outside of it. Scientific modeling, in general, is a scientific activity to make part of the world, or a feature in it, easier to understand or analyze by describing an observed phenomenon.

The construction of models depicting reality is therefore inherit simplifications of more complex or even wicked scenarios. Where wickedness means problems which are incomplete, contradictory, ever changing, or ungraspable. When first introducing wickedness [104] argued wickedness was a new challenge facing decision makers, where now that the basic needs of the people had been met the more complex, less clear, issues were raised. In his review “Wicked problems revisited” [105] did a retake on this assumption and instead states that it is not the wicked that is abnormal but the formal rules and calculations. Wickedness is the most common thing there is. As soon as you have the possibility of a diverse group of decision makers (diverse as in different values, mindset, or opinions) you have the potential of a wicked problem.

An essential part of the research presented in this thesis is the undefined goal of the main system of “lower environmental impact” or “better environmental performance”. This goal does not mean the same thing for all, if any, individuals. Environmental problems are often wicked, which in a shipping-related environmental context could appear when competing interests are an inherent part of the cause of the issue [106, 107]. The results of this thesis, therefore, need to be viewed from a wicked problem viewpoint, which entails acknowledging the limitations of the results. The scale at which humans interact with the natural world is at a scale never reached in history. The socio-technical system puts pressure on the natural world at a scale where we trigger a response and change the equilibrium. We have entered the Anthropocene.

The work performed in the thesis is linked to the field of future studies, as defined by among others [108]. Despite not yet being fully integrated with its frameworks, the work asks questions about the future and makes quantitative assumptions on how technologies and maritime transport will evolve [109]. This thesis takes the view of the future as malleable; the course of future events is not predictable, but it is also not chaotic [110], meaning that human actions can influence its development although not shape it.

2.1.2 Scenario development

This thesis acts within the scope of transition research and directly and indirectly is based on future scenarios. Since the environmental performance of carbon-based marine electrofuels (and electrofuels at large) are modelled at a future point in time, they are dependent on future scenarios [111]. The different ways the world could develop (different future scenarios) creates the solutions space for how the environmental impact might vary [110]. Thus, if an LCA study assesses a future point in time, it uses scenarios, either explicit or implicit, and thereby shows a part of the solution space, also referred to as the "spread of the scenario funnel" [110].

Creating scenarios often involves considering various factors, uncertainties, and variables that could affect the outcome. By exploring different scenarios, individuals and organizations can better prepare for the future and make more informed decisions [112]. Researchers always make assumptions on the context when discussing research results beyond physical and mathematical laws where the conditions can be described in full. If transformative changes are the focus, the implications are also most likely not in line with historical data [112]. The environmental effects from technologies are dependent on first, secondary and third feedback loops both in the ecosystems, material demands, technology development and society [11]. Therefore, the limitations and coherences of the future scope in which the environmental performance and impacts of the carbon-based marine electrofuels must be developed consciously. To create a transparent LCA, the scenario in which the assessment is conducted needs to be stated clearly and not limited by assumptions based in current socio-techno-environmental reality. However, to do this in practice is challenging as will be developed further in Chapter 1.7 of this thesis.

2.2 Life cycle assessment

This thesis addresses the impact from flows of emissions from technical systems to the surrounding environment on ecosystems as well as human health. Exposure to emissions of pollutants is known to cause negative health effects such as respiratory deceases [113, 114], and along with local, regional, and global environmental problems, investigation is needed to identify trade-offs and quantify the impact in terms which can be incorporated into decision making.

Previous research in the maritime sector has assessed the environmental performance of marine fuels from a life cycle perspective and described their life cycle, leading to improved theoretical developments in environmental assessment [23, 24, 27, 33, 40-42]. Fuel does affect the surrounding environment when the vessel is used at sea, but so do the human activities connected to producing the fuel and propulsion equipment. These processes cause emissions that would not have occurred if the fuel was not required. Therefore, to be able to assess the environmental impacts of electrofuels, knowledge of the future production path, use characteristics, emissions to the environment, and efficiencies are needed. Life cycle assessment (LCA) is a primarily quantitative methodology that looks at energy and material flows in a technical system and which flows of emissions to the natural environment they cause to quantify the environmental impacts of human activities [115].

LCA often aim at informing a decision maker, for example a consumer, product manufacturer, business owner, policy maker or the public, and is used both directly in regulation as a quantification method of environmental impact and by researchers as a modelling tool. It stems from an engineering tradition, where the problem and product are in focus. LCA has moved from being used in waste management issues (something acknowledged to be a wicked problem), to database creation, to standardization, to the current methodology discussion [116, 117]. The method is useful when trying to avoid shifting problem from e.g., one phase of the life-cycle to another, from one region to another, or from one environmental problem to another [13], for the systems investigated. In the scope of an LCA study the time-period investigated is stated, but it is difficult to predict the future and an LCA study itself often does not investigate that change. Instead, it makes assumptions about how the socio-technical system will look like, which, if unsuitable, might affect the results greatly.

An LCA considers the environmental impacts from a products or technology's full life cycle: from the extraction of resources, through production, use, and recycling, up to the disposal of remaining waste (see Figure 3). The methodology includes four main iterative steps according to the ISO 14040 standard: goal and scope definition, inventory analysis, impact assessment, and interpretation [118].

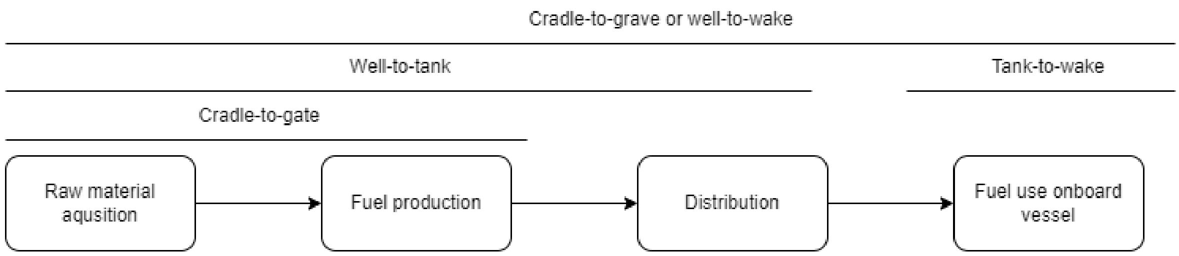

Figure 3 Illustration of a life cycle with nomenclature commonly used to describe the system boundary studied.

First, goal and scope are identified, which sets the stage for the assessment. Here what to compare is established, both in terms of which unit that is directly compared and the systems surrounding it [119]. To compare different options a quantitative unit called the functional unit is defined in detail [120]. This unit represents the function of the system i.e., what specifically that is compared, and this unit should be deemed as being the same throughout all the different options which are compared, so for the context of this thesis the functional unit is the energy used in the engine or the transport work conducted.

Life cycle inventory data collection is the process of gathering, analyzing and summarizing data. All emissions, energy, and material required are added to a life cycle inventory. This life cycle inventory acts as the model of the investigated unit and the surrounding. Here resources and emissions related to the functional unit are calculated, a model for materials and energy flows within and over the system boundaries are mapped and data is collected.

A detailed LCA requires large amounts of data which is not always available nor practical to use, thus simplified or average data is commonly applied. There are databases and models developed which provide environmental performance data and/or LCI data. For papers on the environmental performance of marine fuels, the American database and modelling system GREET developed by Argonne energy laboratory is also common.

Lastly, the environmental impacts are calculated from the environmental loads quantified in the inventory analysis phase [120]. These impacts are categorized based on what they are affecting in the environment and vary depending on which type of LCA methodology is used. However, the basic principle is the same: everything crossing the system boundary (emissions, energy, materials) is added together based on how much they affect a specific category of impact compared to a reference emission/substance [13]. In this way the results are the total amount of the reference emission/substance, which can be compared between different technology options etc.

$$IR_C = \sum_i CF_i \times m_i \qquad (1)$$

The number converting emission from the system to reference emission/substance is called the characterization factor. The total environmental impact results (IR) for different categories (C) can be calculated from the characterization factor (CF) of the substance (i) and the amount of substance (m_i) emitted to the environment using Equation 1.

These three steps are done iteratively, going back and forth, while interpreting the results to make sure that everything is coherent and to create depth to the study. When conducted, an LCA includes many assumptions of the context investigated and the technologies used (see Chapters 2.2.2 and .). The results can appear as a singular number for a specific impact category, but the model as such contains more information.

2.2.1 Life cycle assessment types

LCA is a heavily standardized methodology used outside the scientific context. However, the discussions on terminology and methods within LCA are still active. This thesis will not account for all ongoing discussions but will be limited to some of the methodological choices and frameworks.

Today's life cycle assessment manuals in general applies to modeling and assessment of environmental impacts ex-post (i.e. "after the event" which refers to when information is available from empirical experience) [121], whereas this thesis makes an attempt at transcending this application and look at modelling and assessment ex-ante (i.e. before the event). There have been various efforts in the literature to categorize and develop ways the LCA methodology can be used to look at systems not yet established [121, 122]. Table 1 shows an overview of some of the categorization/methods proposed. There is still discussion on how various types should be used [123-125] and what their differences are. For example, Cucurachi et al. [122] proposed consequential LCA to be a subset of ex-ante LCA [122, 126]. Some of the types presented in Table 1 are more mature than others and there is a continuous development, but as the different examples differ in methodology approach and scope they do share the life cycle perspective [127]. Scenario development in combination with LCA has been used earlier in the field [128] with various fundamentally different key elements [129].

Table 1: Post-ante and ex-ante LCAs used in various literature.

Type and method subsets of LCA	**Description or approach used.**
Attributional LCA	Models the share of environmental impact contributes to a product in a steady state [119].
Conventional LCA	Assesses the environmental impact from existing products [111].
Consequential LCA	This type assesses the consequences caused by changes in the technological landscape, such as the introduction of a new technology or changes in policies [130].
Dynamic LCA	This method emphasizes incorporating the dynamics of parameters that are anticipated to change over time and comparing various development pathways over time [121].
Spatially differential LCA	This method uses special differential impact assessment (can therefore be used together with other types on this list) to account for regional variations in environmental sensitivity [131-133].
Risk-based LCA	This method incorporated risk assessment with the goal and scope definition [134].
environmental input-output based LCA (EIO-LCA)	This method emphasizes the economic dynamics by using aggregate sector-level data to quantify the amount of environmental impact directly attributed to each sector of the economy and as such how much each sector purchases from other sectors in producing its output.
Hybrid LCA	This method is closely linked to EIO-LCA (sometimes considered the same) [135] and addresses LCI design.
Anticipatory LCA	This method focuses on the most relevant uncertainties, exploring both reasonable and extreme scenarios of future environmental burdens, including the values of decision-makers in the analysis to guide research, and development, and innovation [121].
Prospective LCA	This type is used to assess emerging technologies in their early stages of development (experimental stage, small-scale production etc.), but the technology is being modeled at a later, more advanced stage (e.g., large-scale employment) [111].
Ex-Ante LCA	An assessment is done on a new technology before it is used commercially to guide R&D decisions so that this technology is more environmentally friendly than the incumbent technology [121].
Absolute LCA	This approach relates environmental impacts of a product in relation to environmental boundaries [136, 137].

Two emerging types of LCA increasingly applied in literature reviewed through the work of this thesis are prospective and Absolute LCA. Prospective LCA is used to model emerging technologies, i.e., technologies which are in their early stages of development, at a state when they are at large scale production. LCA can then be used to inform technical design choices in the early design process. Absolute LCA is a term originating in the concept of assessing environmental impact in relation to the absolute limits enforced by the planetary boundaries [136]. Its application is driven by the need to assess if the technology or product contributes to a world where we stay within its limits or if we will cause the ecosystem to shift.

2.2.2 Attributional vs consequential modelling

The distinction between attributional and consequential modelling of the LCI has a rich history (see [138] for an outline) and the exact definitions varies between papers and guidelines [119, 139]. This thesis takes the view that consequential and attributional data can be used in both forward-looking and backwards-looking LCA models, in line with the view of Finnveden et al. [124]. Consequential is viewed as a model where the consequence of a choice is in center: what would be the consequences of using Fuel A. In theory, all changes in the surrounding systems (as earlier described as interactions within objects of the systems) should here be modelled to show the environmental consequences of the choice. This leads to scope issues such as how to include first, second and third levels of feedback loops [111]. Attributional[1] aims instead to investigate what is the share if environmental impact from a product or technology [124]. Attributional LCAs are designed to account for all emission, material, and energy data for one technical system, to make a design as complete as possible. They are designed to answer questions such as "What would be the environmental impact of using fuel A for maritime transportation?".

The distinction between attributional and consequential data modelling often boils down to the use of average and marginal data [119]. The general idea among the scholars is that average data should only be used for a retrospective accounting of environmental impact (attributional) and not for future scenarios. Ekvall et al. [119] argues that average data of the new scenario should only be used when the scenario results in a "complete elimination or change of a production system". However, what constitutes a complete change of production system is left unsaid. In practice, LCAs often require a mix of average and marginal data [140] either due to restricted access to specific data, uncertainties in time scales, or due to investigation of subset questions. The issue becomes further complicated when assessments use technology specific data.

Another aspect of the attributional vs consequential LCI distinction is how to solve for multifunctionality [119, 124]. Multifunctionality occurs when several functions (products, technologies etc.) share the same process and the environmental burden (also called load or pressure) must be expressed by only one function. The main tools to address this issue is "system expansion", i.e. where the model is expanded to include the full set of functions in the foreground system, and "allocation", i.e. where the input or outputs are divided between functions or processes (model flows) [13, 120]. The ISO standard 14044 states that allocation should be avoided by either refining the system or expanding it if possible.

A common methods used in system expansion when a system has multiple functions (several products) is to use crediting. A credit is given to the primary assess system for the emissions which should have been released to the atmosphere from the secondary/by product. By doing this, a system change is shown where market mechanisms are included. Closed-loop allocation is one method used to solve allocation issues for waste material, where the recycled material made available at the end of life is used as a material input in the same product system, a closed material loop is formed.

2.2.3 Sensitivity and uncertainty

Uncertainty and sensitivity must be clearly addressed to validate the results of an LCA study [141, 142]. Sensitivity and uncertainty analyses can quantify data uncertainties and technology development, and scenario analysis can assess epistemic uncertainties from modelling choices and future developments [124].

In the context of LCA, the aim of a sensitivity analysis is to determine the robustness of the assessment and identify assumptions which may change the results of the study [120].

[1] Sometimes referred to as accounting LCA.

LCA is strongly dependent on the background system. To be able to generalize results outside of a set study goal and scope, it is therefore crucial to investigate how sensitive the results are to variations in assumptions on background systems. One approach to investigate this sensitivity is by implementing the LCI model into several potential future systems, to indicate which emissions and processes have a large impact on the natural environment. Different technologies are assessed in several potential future systems to indicate which emissions, processes and over all circumstances (local regulations, electric mix, or carbon taxes for instance) has a large impact on which technologies that are competitive and their environmental impacts. Furthermore, they can also help quantify 'known unknowns' with statistical methods.

Uncertainties are of major concern when dealing with emerging technologies [111]. Several methods have been developed to address these concerns in LCA methodology, but no consensus has yet been reached on a general approach [17]. One method proposed to address and display uncertainty of LCA results is Monte-Carlo analysis. Monte Carlo provides a range of possible outcomes and probabilities to allow for analysis of the likelihood of different outcomes [143]. This is done by randomized input variables given an uncertainty range for all/any factor which has an inherent uncertainty [143]. The method is widely used in the LCA community and monte-Carlo analysis has been assessed as a more accurate method to display uncertainty of LCA results than the approaches suggested by standards [142].

Electrofuels are still at an early development stage, where a lot of uncertainties are still inherent to the system. In development processes, decisions are consciously taken on how to design the technology [102]. Initially few choices have been made and a lot of design freedom remains, however as the technology matures, fewer decisions can be changed without stepping back in the design process [144]. This duality creates a scenario where information required to perform a full LCA is available at later stages in the design process, while decisions made early could be optimized for higher environmental performance if information on what would affect this were available [111]. The practical need to proactively assess emerging technologies has been widely acknowledged in LCA at large [111] and for CCU in particular [44, 145-147].

The following chapter outlines modelling methods and frameworks to address uncertainty and sensitivity concerns.

2.2.4 LCI data and model procedures

The data requirement for an LCA is high [13] and strategies to filling gaps of data with information is an essential part of creating a coherent LCI [120]. There are various challenges when creating the data inventory for ex-ante LCAs [45, 111, 121, 123, 130], including for direct general uses for all LCAs such as when available background data is outdated [121].

Lack of inventory data becomes apparent when emerging technologies are investigates, as they are not mature systems [121]. Parvatker and Eckelman [148] investigated data estimation models and their advantages and disadvantages in post-ante LCA (see Figure 4), where more of the technical system is known. Tsoy et al. [149] discussed the data estimation models for ex-ante LCAs in relation to post-ante and found the characteristics to be similar, making Figure 4 valid also for ex-ante assessments.

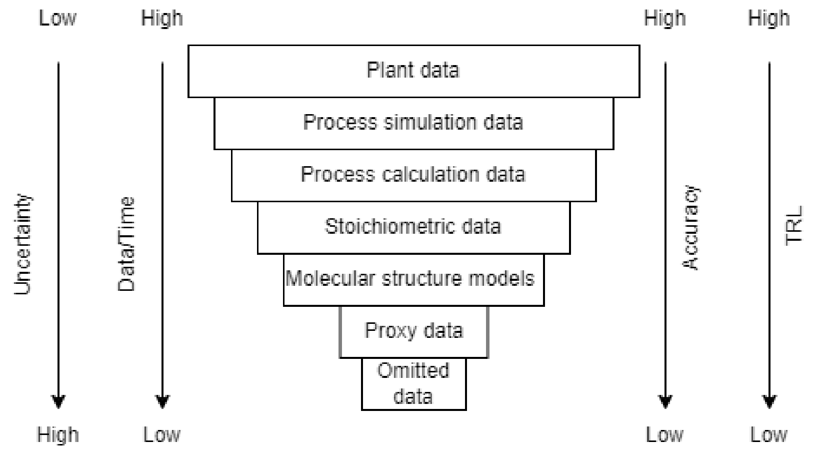

Figure 4 Hierarchy of data estimation models in LCI data generation adapted from van der Giesen et al. [121] and Parvatker and Eckelman [148]. Acronyms used: TRL= Technology Readiness Level.

Three approaches to fill data gaps were proposed by Finnveden et al. [124], the "scientific way", the "social way" and the "statistical way". The scientific way includes further developing the scientific approach by, for example, identifying better data and developing better models. The social way limits uncertainties through discussions with stakeholders, with the aim to reach a consensus on the methodology choices and data used. The statistical way looks at ways to incorporate uncertainties into the analysis.

Wynne [150] broadens the traditional view of uncertainty to distinguish between four types i) risk (system parameters and probabilities are known), ii) uncertainty (system parameters are known, but not the probability distributions), iii) ignorance (neither system parameters nor probabilities are known) and iv) indeterminacy (the future development is inherently undetermined). The four types can correspondingly be assigned to high development (high TRL levels) and early development stages (low TRL levels) of the technology under assessment [121]. To scale up the technology, [149] propose utilizing technological experts to hypothesize the projected future technology scenarios. Once defined, the scenarios could be translated into LCA flow charts by LCA experts, and the unit process data (including energy, material, and elementary flows) could be estimated to the desired scales [149].

van der Giesen et al. [121] point out that risk and uncertainty by these definitions have well-developed methods within LCA to deal with (for example Monte-Carlo analysis is used for this in the appended papers here), but that ex-ante LCA must also deal with ignorance and indeterminacy. Tsoy et al. [149] used review of ex-ante LCA studies and meta-analysis to develop a framework for the upscaling steps of a technology for an ex-ante study. The framework includes three main steps: 1. Projected technology scenario definition, 2. Preparation of a projected LCA flowchart and 3. Projected data estimation. The steps differ in terms of expertise, decisions, choices, and assumptions made in each.

It is of importance to not only consider the TRL level of the technology as it sits in society at the time of the study, which still is important for the underlying assumption of use scenario etc., but also to consider the TRL level of the technology when the data set was developed. Upscaling of emerging technologies with varying maturity levels would require a tap into the plausible future scenarios of their industrial-scale implementation [122]. The technical maturity of fossil, biogenic and electrofuel production differs significantly, and assumptions on future development therefore must be made to assess the technologies at scale. Berglund and Borjesson [151] and Lindorfer [152], among others, established that different biofuel pathways have different environmental impact and that the choice of which technologies to include affects the overall LCA results. Artz et al. [46] established the same relationship for electrofuels. The selection of technology within the life cycle when several options are available therefore becomes significant for the assessment.

2.2.5 Life cycle assessments of marine fuels and carbon capture systems

Electrofuels are an interesting case study for LCA theory, as it is resting between being an energy product, material recycling, a potential carbon sink, as well as an energy storage function. In 2020, a guideline was published by Müller et al. [146], outlining general principles for the life cycle assessment of products from carbon capture utilization. The guide includes principles on how to set the system boundaries and choose the functional unit for the analysis. Principles on how to allocate burdens/costs between different end-products produced from the same processes (i.e., solving for multifunctionality) are also discussed, but no principles are proposed. One of the first meta-reviews of LCAs of carbon capture and utilization products von der Assen et al. [153] included method analysis for some potential pitfalls when assessing electrofuels, as:

i) intuitively interpreting utilized CO_2 as a negative greenhouse gas emission
ii) allocating environmental burdens wrong over multiple functions (multifunctionality)
iii) Overestimating the temporary storage aspect

The use of LCA studies to assess marine fuel has increased in the last decade to approach alternative fuels from a holistic perspective. LCA of marine fuels have been conducted for a multitude of different fuels with varying goals and scope. From competing fossil-fuel options [22-30], to biofuel alternatives, [27, 31-33, 154, 155] and future options such as hydrogen [27, 34-39, 156], or electricity [157-159]. Specific papers studying use of LNG or Methanol from an environmental perspective include: [28, 160-174] . Most studies are limited to investigation of GWP from the marine fuel use and only a few studies has been found which considers environmental impacts of carbon-based marine electrofuels [170, 175, 176]. Only two LCAs of onboard carbon capture have been identified [82, 91].

2.3 Environmental impact categories

This thesis views impacts/effects on the environment in terms of damage and areas of protection. There are three areas of protection used as the foundation in this thesis: Human health, natural environment, and natural resources [177]. There are other ways to construct the framing of environmental impacts, but this way is generally accepted in the LCA community [124]. However, there are other distinctions emerging. The areas of protection are ways to view the impact caused by the technological system on the surrounding natural systems. Adverse effects on human health are measured by mortality and morbidity through space and time. The natural environment is measured by the loss or disappearance of species and the loss of biotic productivity. Natural resources are measured through resource depletion [178].

The different categories for environmental concerns are referred to as “impact categories”. An indicator for an impact category can be chosen anywhere along the impact pathway, where the impact pathway is the chain from emissions and resource use to the final impact on the areas of protection, also sometimes referred to as the cause-effect chain [179]. The impacts are categorized based on what they are affecting in the environment and vary depending on which type of methodology is used. Which environmental problems to consider is not an arbitrary choice. The consensus process in LCA has applied the following principle: what can be assessed with the current available knowledge? What is relevant for the function/system investigated? However, there are also inherit values in the characterization models.

Emissions are added together based on how much they affect a specific category of impact compared to a reference emission/substance [13]. The number converting emission from the system to reference emission/substance is called the characterization factor. Fate factors, exposure factors, and effect factors are combined to characterize the impact. The fate factor will be determined by factors assumption on context, time perspectives, knowledge on movement patterns etc. and exposure factors are sometimes dependent on social aspects such as socioeconomic status among a population, among other things. Impact categories related to environmental issues, such as climate change and acidification, are called midpoint impact categories, whereas endpoint impact categories are associated directly with the areas of protection [120].

The development of impact categories and the methodologies to assess them have been uniformed through consensus models, where experts and other stakeholders discuss and analyze existing LCIA methods to determine what is considered the state-of-the-art (for example [180], [177] and [181]). The goals are to identify the best among existing characterization models and provide recommendations to the LCA practitioner. Research purposes are also included in the discussions. Today, there are several consistent LCIA frameworks, some examples are IMPACT World+ [132], IMPACT 2002+, ILCD[2] [179], ReCiPe [133, 182], CML and European Union Environmental Footprint (EU-EF). There are many environmental impacts occurring in the natural system which are integrated to the LCIA frameworks to varying degrees. The methods are data quality sensitive and often limited in scope. ReCiPe lacks impact pathways to the marine environment, such as, marine acidification, marine eutrophication, invasive species, and plastic debris [133].

2.3.1 Climate change

Climate change impacts both the natural environment and human health through a broad variety of environmental mechanisms. Climate change, i.e., global warming, is currently one of the main discussed environmental issues [183, 184]. The natural environment is affected by the loss of species due to temperature increases, by the changes in oceans and

[2] collection of recommended methods rather than specific framework

seas and by the impacts of extreme weather. Human health, for example, is affected directly by heat waves and indirectly by infectious diseases and malnutrition.

The length of the period considered for impacts on climate change influences how different emissions contribute to the phenomenon, due to that different greenhouse gas emission are not actively impacting the climate for the same length of time [185]. Methane emissions, for example, impact the climate over a shorter timeframe compared to the reference substance carbon dioxide, meaning that with a longer time frame for the impact category methane emissions will matter less in the total result. The midpoint characterization factors used in this thesis for climate change is the widely used global warming potential (GWP), which quantifies the integrated infrared radiative forcing increase of a greenhouse gas (GHG), expressed in kg CO_2-eq. [184].

The ILCD impact categories only consider GWP100 for climate change. To show the influence of different time horizons the impact category climate change GWP20 is also considered in this thesis. In relation to other impact assessment methods, the knowledge on compounds effect on the climate is well established with the latest scientific consensus being presented in the sixth assessment report of the Intergovernmental Panel on Climate Change (IPCC) for calculating GWP20 and GWP100 [184]. Increased concentrations of greenhouse gases in the atmosphere are the triggering effect for climate change, which puts the flow of greenhouse gases to and from the atmosphere as a central keystone to deal with this issue [186].

Table 2 Overview of different metrics used for estimating the direct contribution to climate change [187] of some non-CO_2 greenhouse gases.

	g CO_2-eq./g fossil CH_4	CO_2-eq./g non fossil CH_4	CO_2-eq./g N_2O
IPCC Sixth Assessment Report, GWP100	29.8 ± 11	27.0 ± 11	273 ± 130
IPCC Sixth Assessment Report, GWP20	82.5 ±	79.7 ±	273 ±
IPCC Fifth Assessment Report, GWP100 without climate-carbon feedbacks	28	30	265
IPCC Fifth Assessment Report, GWP20 without climate-carbon feedbacks	84	86	264
RED II	25	25	298

There are emissions other than greenhouse gases addressed by the IPCC that have a secondary impact on radiative forcing [184]. These are emissions that contribute to the formation of ozone, aerosols, and cloud formation, such as NO_X, SO_2 and black carbon.

2.3.2 Acidification

Acidification as an impact category is calculated in terms of acidification potential. The acidification potential addresses the impact generated by emissions of airborne acidifying pollutants, of which the main are SO_2, NO_x and NH_3. The pollutants affect soil, groundwater, surface waters, biological organisms, ecosystems, and materials. The pollutants travel across regions and the acidification response is dependent on the environmental status in the receiving natural system.

In the ILCD recommended method the acidification potential is defined as the number of H^+ions emitted per kg. In LCIA, atmospheric fate factors, soil exposure factors, and effect factors are combined to characterize potential impacts of acidifying substances in terrestrial environments [188]. The methodology is limited to terrestrial and freshwater acidification. Acidification potential is spatially differential, but generic methods are used. Characterization of terrestrial acidification in LCIA is continuously improved, for example in [188] where effect factors for spatial distribution (biome and ecoregion dependent) for Brazil were developed.

Fleet emission of international trade is believed to highly impact acidification, since ships are among the world's highest polluting combustion sources per quantity of fuel consumed. Deposition of sulfuric and nitric acids from maritime transport occurs on the ocean surface water [189] and while averaged across the world's oceans, ship emissions may have a relatively minor role in total ocean acidification, the intense activity of ships in ports and in shipping lanes may have significant impacts [190]. Because of ocean acidification, marine life faces a two-fold challenge: decreased carbonate availability and increased acidity. Marine fuels impact on ocean acidification should therefore be included in an LCA if possible. However, consensus ocean acidification characterization models are currently lacking, but models are in development [191].

2.3.3 Eutrophication

Eutrophication is an example of an environmental problem which occurs only if specific regional and/or local criteria are met. The problem occurs when nutrients limiting growth are supplied in abundance and as such the primary production is enhanced resulting in accumulation of particulate organic matter, which encourages microbial activity and the consumption of dissolved oxygen in bottom waters [192]. This is an environmental problem considered highly important in northern Europe, as the Baltic Sea is directly affected by eutrophication [193]. There is no commonly agreed definition of eutrophication, but there is a conceptual understanding of what the consequences of nutrient enrichment are [194].

Separate impact categories have been developed for terrestrial and aquatic eutrophication. The ILCD recommended using the accumulated exceedance method for terrestrial eutrophication and the ReCiPe impact assessment methods for marine and freshwater eutrophication.

2.3.4 Particulate matter formation

Particulate matter (PM) has both natural and anthropogenic origin and is typically classified as PM_{10} or $PM_{2.5}$ (particulate matter less than 10 and 2.5 micrometer in diameter, respectively). Secondary particles are formed in the air from emissions of SO_2, NH_3, NO_x and others. The main contribution to particulate emissions globally is combustion processes, and the characteristics of these processes affect the number of particulate emissions released. In European coastal areas, shipping emissions contribute to 1–7% of ambient air PM_{10} levels, 1–14% of $PM_{2.5}$, and at least 11% of PM_1.

The characterization factors for PM include the environmental fate, exposure, the dose-response of the pollutant midpoint factors and the severity of the endpoint factors. The fate and exposure can be combined into an intake fraction, and the dose-response and severity can be combined into an effect factor. The RiskPoll model Rabl and Spadaro [195] is the recommended method ILCD, which assesses PM based on global characterization. The main effects of PM on health are on humans' respiratory systems, and as such spatially emission patterns are relevant.

2.3.5 Toxicity

Human health and the natural environment can be affected by toxicity. Toxicity is the degree to which a substance can cause damage to an organism, such as a plant, an animal, or a cell. The impact categories are related to the environmental persistence (fate), accumulation (exposure) and toxicity (impact) of the toxic substances [196]. The model for human toxicity effects represents a full model-based description of chemical fate, exposure, effect and optionally severity. The most common impact categories associated with toxicity are human toxicity and eco-toxicity. There are several proposed methods for implementing toxicity assessment to LCA.

The human toxicity impacts were modelled using the United Nations Environment Programme/Society for Environmental Toxicology and Chemistry (UNEP/SETAC) scientific consensus model USEtox for characterizing toxicity impacts [197]. The fate and effects of chemical emissions expressed in kg 1,4-dichlorobenzene-equivalents (1,4DCB-eq) are used in USEtox as characterization factor at the midpoint level for human toxicity, freshwater ecotoxicity, marine ecotoxicity and terrestrial ecotoxicity. The method is continuously updated based to fill knowledge gasps [198] and the scope of the toxicity impact assessment method has been recently updated [199].

2.4 Energy sources and fuels

Fuel is a broad term used for a material which can be converted to various usable forms of energy, such as thermal, mechanical, or electric energy. A fuel is primarily an energy carrier, where the fuel is used to carry energy from the extraction source to the use point. There are many possible energy carriers for the shipping sector, with various characteristics suitable for different use cases. Energy carriers in solid[3], liquid[4], and gaseous[5] states are all thinkable. Some energy carriers discussed in shipping today are heavy fuel oil (HFO), marine gas oil (MGO), diesel (and diesel variants), methane, methanol, ammonia, hydrogen, and electricity. Some energy carriers, for example methane, can be liquified under high pressure and low temperature to be a liquid energy carrier despite being gaseous at ambient temperature and pressure.

There are many potential pathways to produce energy carriers, and various types of energy sources can be used (Figure 5). The energy carriers in turn can be used in various propulsion systems and transport modes. Figure 5 shows an outline of plausible

[3] For example, uranium, coal, wood
[4] For example, alcohols, diesel quality fuels, liquified gases
[5] For example, methane, dimethyl ether

connections between the four stages. The role of carbon-based electrofuels can be viewed as a pathway to bridge electricity sources to liquid and gaseous fuels (pink boxes), which enables electricity as an energy source in a larger part of the transport system.

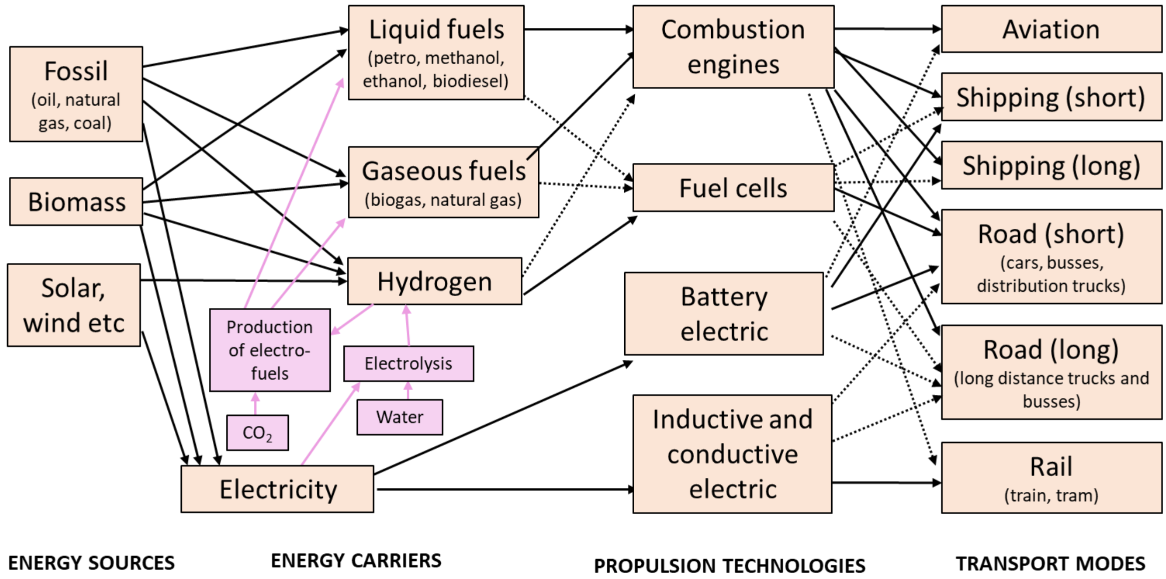

Figure 5 Simplified schematics over the main fuels and propulsion technology options in different transport modes. Adapted from Wismans et al. [200] and through continuous work within the research group at Maritime Environmental Sciences led by M. Grahn at Chalmers University of Technology. Pink boxes indicate the place of carbon-based electrofuels within the system. Dotted lines are pathways used today but to a lesser extent.

Some fuels are produced solely through natural processes without human interaction, others range from cultivation of natural resources to synthetic production. The energy source has proven to be crucial in sustainability assessments of marine fuels [201], but there are various definitions categorizing energy sources in different categories in relation to sustainability [202]. The exact definition of energy sources as renewable or fossil is not crucial for the assessment in this thesis as all emissions to the environment are considered independently of definition.

The definition of renewable energy is largely coherent today, with the International Energy Agency (IEA) defining renewable energy as "energy derived from natural processes that are replenished at a faster rate than they are consumed". However, the term "renewables" history is complex [202], and the definition is not consistent with the concepts of fossil and biogenic often used in the fuel discourse. For example, fuels from biomass have varied regeneration rates from less than a year (e.g., many agricultural crops) to thousands of years (e.g., peat) and as such can be either renewable or not. Figure 6 shows one possible outline (of many) of how energy sources can relate to different historical terminology. Harjanne and Korhonen [202] criticizes the use of the term renewable and suggests a two-dimensional definition of energy sources instead, where the distinction is between carbon content and if the energy carrier is reliant on combustion processes.

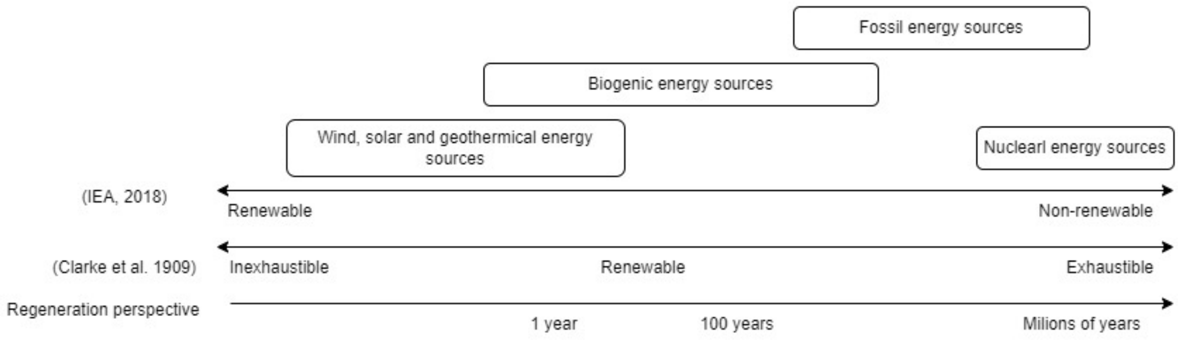

Figure 6 Potential view comparison between some scales used historically to distinguish between energy resources regenerability [203].

The European Union includes wind, solar, hydro, and tidal power, geothermal energy, biofuels, and the renewable part of waste as renewable energy in its statistical accounting [204]. However, renewable does not mean lacks environmental impact nor is it necessarily sustainable.

2.4.1 Fossil fuels

The energy sources primarily used in the shipping sector today are non-renewable and extracted from the Earth. A fossil resource is removed from the ground, treated, and eventually oxidized carbon accumulate in the atmosphere [205]. The types of fossil fuels possible to utilize in shipping are broad, ranging from heavy fuel oil (HFO) and marine gas oil (MGO) of different degrees of purity to gases such as liquified natural gas (LNG). Fossil fuels make up 99.9% of the global maritime bunker. Less than 6% of the vessel fleet is using any form of alternative fuels [206], with the majority being fossil liquified natural gas (LNG), and only approximately 10% of the vessels in the global ship-building order books are adopting alternative fuels.

2.4.2 Biogenic fuels

Biogenic fuels, or biofuels, are fuels which energy source is biomass. Biomass was the primary energy source used by humans before the nineteenth century and is considered a potential renewable energy source for fuel production (Figure 6). Biogenic fuels have the potential to be climate neutral, but supply is limited due to lack of sustainable biomass extraction [207]. Biomass is biological material gathered from agricultural crops, forest products, aquatic plants, crop residue, animal manure and, depending on definition, waste. Estimates of the global supply potential varies based on perspectives e.g., its theoretical potential, technical potential, market potential, and sustainable potential [208]. Different studies present vast differences on global biomass supply potential, e.g., in the range 10-245 EJ/yr [209] as well as in the range 1135-1550 EJ/yr [210]. Many of the authors claim up to 100 EJ/yr of bioenergy can be produced in a sustainable way and that 300–500 EJ/yr may be technically possible, but that such expansion might challenge sustainability criteria. As such, the global biomass supply potential is limited and harvesting large fractions of the available biomass would result in severe adverse impacts on the natural environment.

Biomass, as it does regenerate, may be considered carbon neutral on a long enough (often decadal) timescale if new growth absorbs enough carbon dioxide. They are, however, not necessarily without carbon content. For example, biomethanol, which contains carbon in its molecule and therefore emits carbon at tail pipe. To consider the type of biomass source therefore is essential to evaluate the climate performance as well as other environmental impacts. Biofuels has known environmental concerns, primarily in relation to land use requirements, competition with food cultivation, and destruction of biodiversity.

2.4.3 Electricity as an energy carrier

Electricity cannot be generally categorized as renewable or non-renewable. Its qualities depend on the energy source used during the energy extraction. Wind power, solar power, hydro power are examples of energy sources commonly considered renewable. Electrofuels are synthetic fuels produced through human activities from the energy source (merged with carbon) to the energy carrier. The life cycle emissions to the environment from electricity production varies between electric power sources, in terms of GHG emissions as well as emissions affecting health and other impact categories [211]. The resource demand such as land occupation rate and water demand also differ [212].

The intergovernmental climate panel's (IPCC) synthesis report (AR5) [185] contains a compilation of GHG emissions for electricity production from different energy sources (see Table 3). The large variations within electric power types depend, among other things, on how the fuel is extracted, the construction of the power plant and how efficient the power plant is [213]. The emission intensity of renewable energy production is dependent on both their efficiency and utilization degree, where the utilization is the factor suggested to vary the most between power production sites.

Table 3 GHG emissions for different electricity power sources for some data sets

Electricity power source	IPCC (g CO_2e/kWh)	Scarlat et al. [214] (g CO_2e/kWh)	Ecoinvent (g CO_2e/kWh)
Hydro power	~4	~19	~12
Wind power	~11	~11	~9
Nuclear	~12	~23	~13
Solar power	~41	~41	~42
Natural gas	290-930	~430	398-915 [215]
Oil	510-1170	~780	~800
Coal	740-1689	~970	814-1710 [215]

2.5 Propulsion systems and the maritime use scenario

The global maritime cargo fleet is growing, with a deadweight tonnage (dwt) increase of 63 million during 2022 according to UNCTAD. The current vessel fleet consists of over 100 000 vessels equipped with different engine types. Commercial marine engines range in size from 1 MW with the largest main engines reaching above 20 MW. Two engine types stands for the majority of global fuel consumption: Slow-speed diesel (SSD) 2-stroke engines (70%) Medium-speed diesel (MSD) four-stroke engines (17%) [175]. The vessel age affects the emission level and the load dependence of the emissions [175].

2.6 The regulatory landscape of shipping

The policy landscape of shipping is under rapid development. Emissions from international shipping cannot be attributed to any national economy due to its global nature and complex operation, which has led to few regulations applied to this sector historically [14]. However, legislation aimed at emission reduction is being introduced at the international level and within the European Union.

Concerns about emissions to land, air, and water from ships were brought up to international levels in 1968, and the primary international convention regulating pollution from shipping, MARPOL 73/78[6], was introduced by the International Maritime Organization (IMO)[7] in 1973. However, most regulations influencing marine fuel choices were introduced much later. In 1997, the first direct international regulation on marine fuels was adopted through MARPOL Annex VI "Prevention of Air Pollution from Ships". Figure 7 shows the timeline for implementation of measures on Sulphur content in marine fuel and NOx emissions from marine engines outlined in MARPOL Annex VI and air emissions from Swedish maritime transport (defined as emissions from shipping traveling between, to, and from Swedish harbors). MARPOL has since its introduction been amended continuously, and today also regulate Volatile organic compounds (VOC) (only for tankers and some gas carriers), Black carbon emissions in the artic region[8], and GHG emission by energy efficiency regulations.

IMO regulation related to climate change impact mainly covers two topics: increased energy efficiency of the vessels and reduced GHG emissions. In July 2011 the Energy Efficiency Design Index (EEDI) (for new ships) and the Ship Energy Efficiency Management Plan (SEEMP) (for all ships) were added to MARPOL Annex VI. The EEDI sets specific energy demand requirements for design of new ships, formulated as g CO_2/tonne mile.

[6] The International Convention for the Prevention of Pollution from Ships, 1973, as modified by the Protocol of 1978 (MARPOL 73/78)
[7] Then called the Inter-Governmental Maritime Consultative Organization (IMCO)
[8] MEPC.342(77)

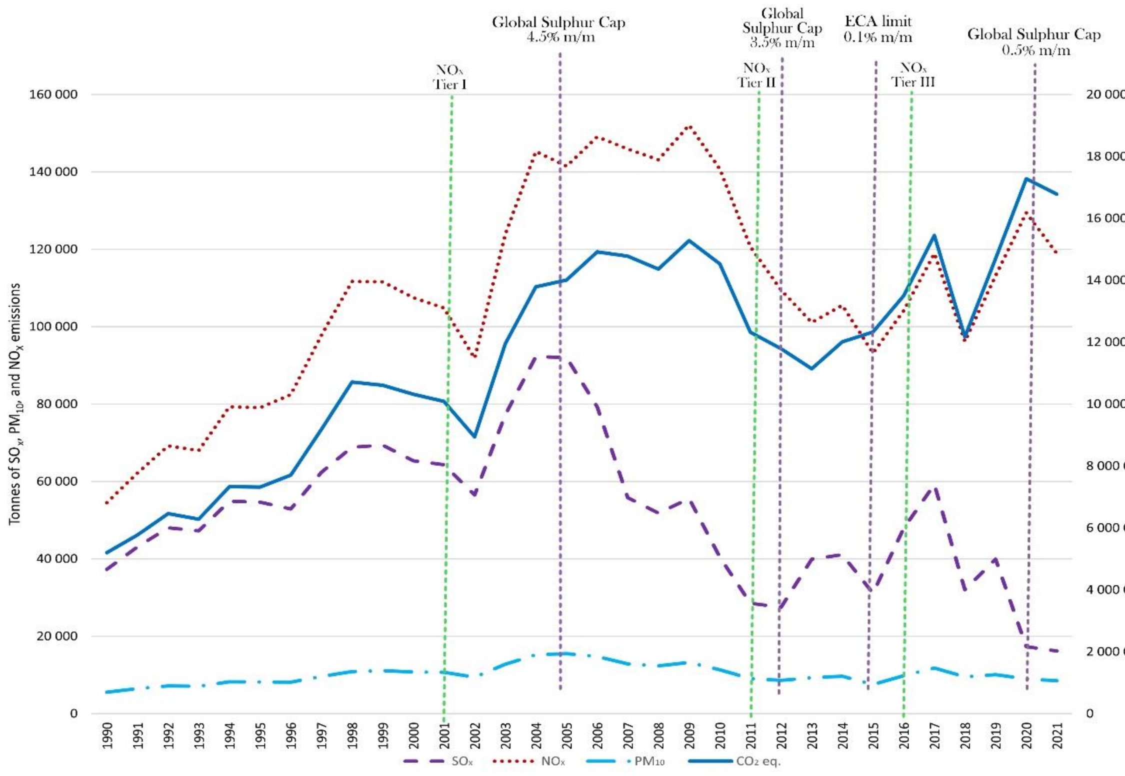

Figure 7 The yearly emissions of Greenhouse gases (in CO_2 equivalents), Sulphur (in SOx), Nitrogen oxide (in NOx), and Particulate matter (in PM10), from 1990 until 2021 in Swedish domestic shipping and Swedish international shipping [216]. Information on legislation is gathered from The Marine Environment Protection Committee [217] (adapted from Paper A).

The first resolution at the IMO on CO_2 emissions from ships was adopted in 1997 [218], which initiated the Marine Environment Protection Committee's (MEPC) work with climate change related emissions. In 2000, the first IMO study on GHG emissions from shipping was published which quantified the total emissions of GHG gases[9] from international shipping. The initial study has been followed by three [4, 14, 219] additional IMO GHG studies, with the latest publication in 2020 [4].

In 2018 the Marine Environment Protection Committee (MEPC)[10] adopted an initial strategy for the reduction of GHG from ships called *Initial IMO Strategy on reduction of GHG emissions from ships*. The initial strategy defined the continued work process for handling climate changes issues in shipping, including a plan for revision of the document in 2023. In July of 2023, the initial strategy was replaced with the *2023 IMO Strategy on Reduction of GHG Emissions from Ships*, which includes a broad range of measures and supportive actions. Of specific interest to this thesis is the introduction of life cycle assessment to the strategy:

"The levels of ambition and indicative checkpoints should take into account the well-to-wake GHG emissions of marine fuels as addressed in the Guidelines on life cycle GHG intensity of marine fuels (LCA guidelines) developed by the Organization with the overall objective of reducing GHG emissions within the boundaries of the energy system of

[9] Primarily CO_2, but the scope has expanded to include CH_4 and N_2O

[10] The MEPC consists of all Member States and consider any matter within the scope of IMO related to prevention and control of pollution from ships.

international shipping and preventing a shift of emissions to other sectors". (MEPC 80/WP.12 Annex 1, Para. 3.2)

The *Guidelines on life cycle GHG intensity of marine fuels* [220] are limited to emissions of CO_2, CH_4, and N_2O. The characterization factors used are the GWP100 factors as given by IPCC AR5 [185]. The LCA approach is stated as attributional LCA (in reference to data modeling), with the comparative well-to-wake GHG emissions of fossil MGO noted as 94 gCO2 eq./MJ fuel. The specific approach is designed to avoid double accounting across sectors. The development of guidelines for life cycle GHG emissions of marine fuels is ongoing, as well as a revision of the full 2023 strategy planned for 2028. The *2023 IMO Strategy on Reduction of GHG Emissions from Ships* is to be reviewed with a focus on i) default emissions, ii) sustainability criteria, iii) fuel certification, and iv) handling of onboard carbon capture.

The European Union has recently introduced legislation aimed at reducing the carbon footprint of the maritime sector in the EU as a part of the EU Fit for 55 package [221]. The European Union Emissions Trading System (EU ETS) will be expanded to include maritime transport for vessels above 5000 GT entering EU ports. The EU ETS reporting will be based on data reported according to the monitoring, reporting, and verifying (MRV) reporting scheme, cover CO2 emissions (starting in 2024), and will cover CH4 and N2O emissions from 2026. The FuelEU maritime initiative aims to increase the demand for and consistent use of renewable and low-carbon fuels. Most notable for the scope of this thesis is the inclusion of electrofuels in the legislation. The FuelEU maritime initiative directly includes specific incentive regimes to support the uptake of "renewable fuels of non-biological origin" which includes carbon-based electrofuels if the energy content is from renewable sources other than biomass. Methods to calculate life cycle GHG emissions from fuels are given for example in Council directive 2015/652.

2.7 Quantifying emission from maritime transport

To assess the impact of emissions on the environment, the emissions must first be quantified. A rough assessment of the current and future emissions from shipping could be described as:

$$Total\ emission_i = GDP \times \frac{Transport\ work}{GDP} \times \frac{Energy}{Transport\ work} \times \frac{Emissions_i}{Energy} \qquad (2)$$

Where:

- i is the emission type.
- Gross domestic product (GDP) is used as an indicator for the size of the economy.
- The second term is the transport intensity of the economy.
- The third is the energy required to perform the transport work.
- The fourth is the emission intensity of the energy.

Currently the amount of maritime transportation activities has been directly coupled with the global economy [1], and an increase in economic activities has historically led to a direct proportional increase in transport work. Energy efficiency (i.e., the energy used per transport work) has improved at several points both historically and today due to technology implementation, behavioral changes, and introduction of legislation. Research on improved energy efficiency in shipping is extensive and active, and directly tied to reducing the total emissions from the sector [222]. The emission intensity of the energy does not only relate to emissions from the fuel production nor only the onboard activities. Instead, it should be viewed as the total emissions over the life cycle for the energy used, which does not only relate to direct emissions to air but also average emissions from accidents etc. Different models have been used to quantify emissions from maritime transport. A short summary of some of the model which has inspired the work of this thesis is presented in Table 4.

Table 4 Models used to assess emissions from maritime transport. The list is not comprehensive but shows a range of different approaches and examples.

Model name	Institution	Type of model	Aim of the model	Context	Examples of studies	Method reference
GREET (The Greenhouse Gases, Regulated Emissions, and Energy Use in Technologies Model)	Argonne national laboratory	Life cycle assessment model	Evaluation of energy and environmental effects of vehicle technologies and energy and product systems	Northern America	[223], [224]	[225]
GET (Global Energy Transition)	Chalmers University of Technology	Energy systems model – linear optimization	Cost optimization model focusing on the transport sector	Global 10 regions	[226], [227]	[228]
the MariTEAM Model	NTNU, SINTEF	Bottom-up analysis of	Assess global shipping emissions from a Well-to-Wake Perspective	Global	[229]	[229]
STEAM (the Ship Traffic Emission Assessment Model)	FMI	Bottom-up analysis	Evaluation of the exhaust emissions of marine traffic	Flexible (based on AIS data)	[3]	[230]
Shipair	SMHI	Bottom-up analysis	Assess the emissions the shipping fleet in an area	Sweden	[231]	[231]
IMO4 bottom-up vessel based	IMO	Bottom-up analysis	Assess the total amount of GHG emissions of shipping (international, domestic and fishing)	Global	[4]	[4]
IMO4 bottom-up voyage based	IMO	Bottom-up analysis	Assess the total amount of GHG emissions of shipping (international, domestic and fishing)	Global	[4]	[4]
IMO4 top-down	IMO	Top-down analysis	Assess the total amount of GHG emissions of shipping (international, domestic and fishing)	Global	[4]	[4]

2.8 Carbon Capture

Carbon capture is the process by which CO_2 is isolated from dilute mixtures. Several different types of carbon capture technologies have been developed and are at different levels of technical maturity [232]. Examples of different technologies are shown in Table 5. The technologies for carbon capture are still under development, but include options such as membrane carbon capture, direct air capture, and flue stack cleaning [233-236], and carbon can be captured from various sources [237]. What is considered alternatives for the carbon supply varies between paper and research questions [19, 45].

Table 5 Summary of carbon capture technologies.

Capture stage	Description	Examples of technologies	Related papers on onboard carbon capture
Post-combustion	Capture takes place after a combustion process, for example from flue gas	Chemical solvents	[71-75, 81, 82, 85, 88, 90, 96]
		Solid sorbent	[16, 238]
		Membrane	[79]
		Cryogenic	
		Solidification	[54]

Pre-combustion,	Capture takes place before a combustion (or another use-phase) is completed	Membrane	Paper C, [56, 80]
		Chemical solvents	
		Chemical sorbent	
		Solid solvents	
		Solid sorbent	
Oxy-combustion (also called oxyfuel)	The combustion takes place in an atmosphere enriched with oxygen		[56, 95]
Chemical looping	Uses circulation of metal (oxidation and reduction reactors) to facilitates the combustion of fossil fuels		
Ambient capture	Direct air capture	Chemical solvents	N/A
		Solid sorbent	N/A
	Direct Ocean Capture	Chemical solvents	
		Solid sorbent	

2.8.1 The literature on onboard carbon capture

Carbon capture technology is mainly applied to thermal power generation, cement and steel production, and other industries [239]. The motivation for using onboard carbon capture primarily brought forward in the literature is to mitigate greenhouse gases. [240] is simply stated that:

> *"If the application of CCS technology can be actively promoted on ships, it will be of great significance for the shipping industry's energy conservation and emission reduction."*

Of the identified papers on onboard carbon capture their potential to meet EEDI standard, and as such only tail pipe emission reductions, is the motivation and investigative scope for most studies [58, 73-75, 79, 81, 85, 90, 96]. Only two papers (besides Paper C) of onboard carbon capture which considers a broader scope of environmental impacts where found: Negri et al. [82] and Negri et al. [91]

The first study on onboard carbon capture was published in 2014 and reviews the possibility to use carbon capture and storage onboard vessels [52]. The first technical paper looking at a more detailed system set up was published in 2017 and looked at a solvent based system [57]. The study found the carbon capture level to be limited to 73% when the existing ship energy system is integrated with the carbon capture process due to limited heat and electricity supply for CCS. With additional heating systems the capture rate at the tail pipe could be raised to 90% but the system energy efficiency drops, and the total fuel requirement is increased by 21.5%. The 2021 study Long et al. [71] also noted this relationship and concluded that there is a need to develop a CO2 capture process with a higher capture rate or lower energy use. Law et al. [88] shows a 90% capture rate for solvent based capture, a level also reached in Feenstra et al. [64]. Ji [241] concludes the capture rate at the flue gas point varies depending on engine technology and conditions between 30% and 98%. Stec et al. [75] includes spatial system aspects on the onboard carbon capture and concludes capture in tropical conditions to be higher due to a lower heating demand, supporting the claims of Luo and Wang [57]. Awoyomi et al. [242] performed a process evaluation on a solvent-based post-combustion capture process for the energy system onboard a CO_2 carrier. The simulation was based on capture on the flue gas stream from using LNG in a DF marine diesel engine.

Güler and Ergin [73] looked at carbon capture onboard as simply a carbon mitigation option and compares it to LNG and speed reduction measures as a carbon mitigation option. The study is clearly driven by a tail pipe emission perspective and does not employ any life cycle thinking. The capture rate varies between 46 and 49%. Lee et al. [74] also looks at an Energy Efficiency Design Index (EEDI) perspective and designed the study to assess a scenario where CCS is used to meet 70% reduction for 2050. Oh et al. [79] evaluates a membrane onboard capture system designed to meet the same reduction target. The goal with the studies was primarily to assess the technical feasibility of the technology and no discussion on environmental impacts of life cycle carbon performance was included.

The TRL level varies between assessments primarily due to different assumptions on how different a marine carbon capture system must be compared to an onshore system. Long et al. [71] investigated an onboard carbon capture system concept and stated it as having a TRL below 3. This was also the case for the HyMethShip system. From the assumption point that maritime applications will have significant differences from onshore applications, onboard carbon capture should be treated as an emerging technology still.

3 METHODS, METHODOLOGY, AND ANALYTICAL FRAMEWORK

This chapter provides an overview of the procedures and theoretical context adopted to achieve the results and conclusions presented in this thesis. Details of the methodology for each study can be found in the appended papers. The terminology presents how the terms are used and viewed in this thesis and is not fully inclusive of all current uses of terms in literature.

3.1 Philosophical position

The papers vary in methods, methodologies and analytical frameworks used, but they all fit under one research paradigm: pragmatism. The choice of paradigm is driven by the assumptions of the nature of science which includes elements such as the nature of reality, knowledge, value/s, and methodology [243]. In terms of ontology (nature of reality) and epistemology (nature of knowledge), pragmatism is not committed to any single system of philosophy and reality [244]. Reality is actively created as individuals act in the world, and it is thus ever changing, based on human experience, and oriented toward solving practical problems.

Choosing marine fuels is viewed as a practical problem containing both qualitative and quantitative aspects where a range of research approaches should be applied to find solutions and increase understanding. Throughout this thesis, the marine fuel choice is viewed as the problem of choosing the most reasonable technologies. The problem is heavily discussed and rather than taking a philosophical position where the fundamental truth of science is in focus, pragmatism allows the research to be focused on informing the research question at hand. In the words of William James from his second lecture on pragmatism at Harvard University 1906:

"Pragmatism is willing to take anything, follow either logic or the sense, and to count the humblest and most personal experience. She will count mythical experiences if they have practical consequences" [244].

The research of this thesis is designed to inform if there is any new evidence to include in the scientific understanding of the marine fuel choice, a reasoning in line with the pragmatic approach. Pragmatism aims to approach research from a practical point of view, where knowledge is not fixed, but instead is constantly questioned and interpreted [243, 245]. The philosophy is also compatible with systems thinking, where even in what's considered one of the first lectures on Pragmatism, the interactions between all things in the world and all its parts was discussed as "systems of influence or non-influence" [246].

3.2 Research approach

This thesis' research questions have been informed by the technical development of new technologies and a need to fill an identified knowledge gap of the potential application effects of these technologies. The research process could be described as occurring between the space of society and science. The research has been conducted within larger research projects, and in collaboration with a range of stakeholders with societal questions giving input to the research process. Figure 8 shows an outline of the broad societal questions (green) initiating the research conducted in the appended papers and how these then relate to the research questions of this thesis. For an overview of the different methodological approaches included in this thesis, see Table 6.

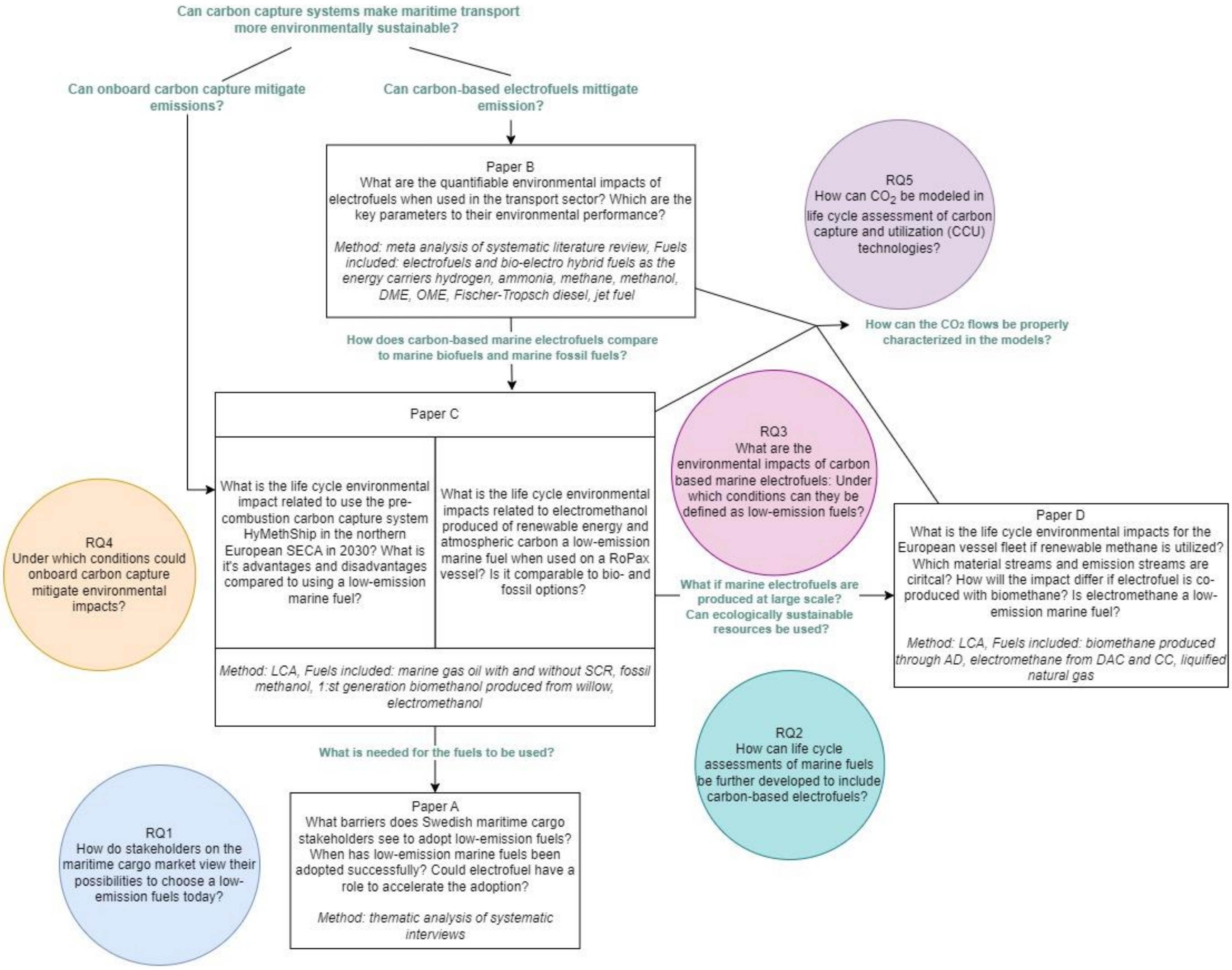

Figure 8 Outline of the appended papers, the questions linking them through the research work process, and how they relate to the research questions of this thesis. Acronyms used: AD=anaerobic digestion, DAC= direct air capture, DME=dimethyl ether, CC=carbon capture, SCR= selective catalytic reduction, LCA=life cycle assessment, OME=oxymethylene ether

Table 6 Summary of the methodological approach for each paper included in the thesis.

Paper	Approach	Research design	Methodology[11]	Methodological tools[12]	Analysis method[13]	Study object
A	Qualitative	Case study	Case study	Systematic literature search Coding framework Semi-structured interviews	Thematic analysis	Swedish shipping stakeholders
B	Quantitative	Literature study	Traditional literature review[14]	Data extraction forms Systematic literature search	Meta-analysis	-
C	Quantitative	Case Study	Life cycle assessment	Inventory data collection Data quantification and normalization Data characterization	Life cycle impact assessment Monte Carlo analysis	RoPax vessel travelling from Gothenburg to Kiel
D	Quantitative	Case study	Life cycle assessment	Scenario development Systematic literature search Inventory data collection Data quantification and normalization Data characterization	Life cycle impact assessment Monte Carlo analysis	Swedish bio-methane production and the European LNG propelled vessel fleet

[11] Methodology is defined as a broad approach to data collection.
[12] Methodological tools (or methods) are the tools used for data extraction.
[13] Analysis methods are the exact tools used for analyzing the data.
[14] More about the different types of literature reviews can be found in text.

3.3 Case Study

Case studies are used to describe, compare, evaluate, and understand different aspects of a research problem or application scenario. Case study can be defined as an exhaustive study of a person, a group of people or a unit, with the aim to produce knowledge which can be generalized over several people, contexts, or units [247]. The study object (see Table 6) is investigated through an in-depth analysis to gain insights into a particular phenomenon [248]. The methodology often focuses on understanding the context, dynamics, and complexities of the case under investigation. The specific case can be described, compared, evaluated, and understood within its context. This thesis uses case study design as described by Yin [248]. Paper A was conducted as an explorative case study [248], whereas Papers C and D were conducted as descriptive case studies. For details on the case study context view the appended papers.

3.4 Literature study and review

A literature review is a survey of scholarly sources on a specific topic. The aim is to establish an overview of the state-of-the-art knowledge of the subject and, through this process, identify relevant theories, methods, gaps, controversies, and developments in the field [249, 250]. The systematic literature review approach is a useful research method that supports qualitative, quantitative, or mixed methodology and can assist in reducing researcher's bias while enabling a precise scope of review [251]. Literature reviews can be categorized as a methodology or method depending on the extent to which it is applied/conducted [250]. The literature reviewed within this thesis has contributed to the development of the research questions, theoretical framework, methods, analysis, data collection and future research directions. For one of the appended papers (Paper B) literature review was the primary methodology (Table 6).

3.4.1 Traditional literature review as conducted in Paper B

The primary methodology for Paper B was a literature review. The approach used was in line with a traditional literature review. A traditional literature review is a quantitative approach where quantitative data from various sources is gathered and analyzed through **meta-analysis**. The study presents data gathered through a **systematic literature search** in databases and snowballing methods (i.e., identify additional relevant publications from reading publications' reference lists). To identify relevant publications search term groups with constraints were used to exclude publications not analyzing fuels produced from carbon or nitrogen. The study covers general aspects, costs, and environmental aspects of electrofuels. However, the content first and foremost included in this thesis is related to environmental impacts (Chapter 5 of Paper B).

The literature search string used (noted as AB in Paper B) resulted in 196 publications relevant for the review of environmental aspects. Additional relevant publications have been added through snowballing. The data extraction was carried out systematically using data extraction forms where each selected source was examined in detail. Key information, such as authorship, publication year, research methodologies, sample characteristics, and key findings, was recorded to facilitate a structured analysis of the literature. The information extracted to specifically review the environmental performance was:

- Specific technology description
- LCIA method/other quantification methods used.
- System boundaries (as described and as implied from the study design)
- Main environmental impact hotspots
    - Assumptions related to the hotspots (for example system boundary or background emission sources).
- Impact categories investigated.
- Primary emissions included.
- Energy demand

A generic model of the life cycles of electrofuels was created to establish the context for the various quantitative and qualitative conclusions presented in the literature. A **meta-analysis** was then performed based on the type of fuels investigated, scope, type of assessment, investigated environmental impacts, main identified hot spots, and quantitative impact results.

3.5 Thematic analysis

Thematic analysis (TA) is a qualitative research method used to analyze and interpret data in a systematic and comprehensive way [252]. In TA, the researcher generates patterns of shared meaning (so-called themes) to show how different data points relate to a central concept or idea [253]. Thematic analysis techniques identify patterns, common topics/ideas, meaning, and divergent viewpoints across datasets. The themes structure the coded data and tell a story. The method can be used to explore a wide range of research questions and can be applied to data collected from various sources, including social media, literature, and personal narratives.

The thematic analysis explores complex phenomena and experiences, including those related to market behaviors [254]. The marine fuel choice is an inherited market-related choice, as the value comes from the transport service rather than the fuel [1]. However, the fuel choice is still made by humans and, therefore, inherently holds qualitative decision-making qualities. By exploring the qualitative aspects of market behaviors, such as attitudes, beliefs, and motivations, thematic analysis can provide a more in-depth understanding of why certain behaviors occur and what factors contribute to them [252]. More quantitative methods are easier to validate repeatability and full reporting, but the simplifications required to perform a quantitative study limit their value. TA can be and has been, used in a wide spectrum of theoretical frameworks [252]. As qualitative inquiry gains prominence in applied science, it is critical to take advantage of qualitative methods' diversity and flexibility.

3.5.1 Thematic analysis as conducted in Paper A

Paper A collected primary data through **semi-structured interviews**. 17 interviews were conducted for a total duration of 23 hours and 45 minutes. The results in Paper A reflect the measurement period (Nov 2022- Feb 2023). The interviews were then transcribed verbatim. The transcribed interviews were analyzed using reflective TA, as presented by Braun and Clarke [253] and further developed in Braun and Clarke [254] and Braun and Clarke [252]. A **coding framework** was developed iteratively throughout the analysis. The initial codes/labels were identified after the first six interviews were transcribed and reviewed. The codes were then refined, and additional codes added as they were identified through data reviews and revisions of data. Each interview transcript was revisited at least twice through the coding process. The codes were then grouped into themes which depict the meaning of the underlying data. The research process followed the guidelines of Braun and Clarke [253] for good thematic analysis, including transcription, coding, analysis, and reporting.

3.6 Life cycle assessment

In this thesis, LCA is used as a modelling tool to understand the potential environmental impact from a technology or decision. Model systems with large uncertainties are generally used to investigate multiple parameters, rather than models assessing singular environmental flows with a large degree of certainty (see Chapters 3.6.1 and 3.6.2). The goal of the modelling in this thesis is to identify what drives the environmental impact, how it shifts depending on the future use case, and where current knowledge on the technology's environmental impact must be expanded. This follows the pragmatic viewpoint established earlier in this thesis – the marine fuels' environmental impact is a parameter which decision makers must take into consideration and researchers can support the framing of the knowledge base.

In this thesis primarily two types of LCA methodology frameworks are considered as method foundations (in addition to the broader ex-ante umbrella term): prospective LCA and the scenario-based Ex-Ante LCA. The appended papers might be categorized differently by other authors, but these future oriented LCA approaches give a foundation for the academic context. The LCAs assessed in Paper B cover a large range of LCA types, but the meta-analysis primarily focused on dynamics within the LCA results.

A challenge when conducting this thesis was the availability of data both in the fuel production and in the use phase. To address the data gaps in the inventory, the data gathering in the appended papers have followed the hierarchy in Figure 4 dependent on what has been the highest available data model at the given time. This leads to inventory with fuller data, but also that no-known parameters have been omitted as more mature technologies are compared to emerging. This creates the need to project the LCI data to be at similar levels of maturity, which has been forecasted using expert assessments and literature data on future performance. Buyle et al. [255] provides an outline of possible data estimating methods focused on ex-ante LCA. The LCI models used in this thesis are primarily built using technology specific data or average data mixes from future scenarios. In Paper C, the LCA model results were directly discussed with technical designers during the design process.

Table 7 Characterization methods used in the life cycle assessments. Based on the International Reference Life Cycle Data System (ILCD)[118] recommended characterization methods.

	METHOD USED IN PAPER C				METHOD USED IN PAPER D			
	Impact category	Reference substance/unit	Impact assessment method	Method reference	Impact category	Reference substance/unit	Impact assessment method	Method reference
ACIDIFICATION	Acidification	molc H+ eq.	ReCiPe	[256]	Acidification	molc H+ eq.	ReCiPe	[256]
CLIMATE CHANGE	GWP20	kg Co2 eq.	IPCC	[183]	GWP20	kg Co2 eq.	IPCC	[184]
CLIMATE CHANGE	GWP100	kg Co2 eq.	IPCC	[183]	GWP100	kg Co2 eq.	IPCC	[184]
EUTROPHICATION	Freshwater eutrophication	kg P eq	ReCiPe	[256]	Eutrophication, aquatic freshwater	kg P eq	EUTREND model (as implemented in ReCiPe)	[257]
EUTROPHICATION	Marine eutrophication	kg N eq.	ReCiPe	[256]	Eutrophication, Aquatic marine	kg N eq	EUTREND model (as implemented in ReCiPe)	[257]
EUTROPHICATION	Terrestrial eutrophication	molc N eq	Accumulated Exceedance	[256]	Eutrophication, terrestrial	molc N eq	Accumulated Exceedance	[256]
IONIZING RADIATION	Ionizing radiation	kBq U235 eq.	Human health effect model	[258]	Ionizing radiation, human health	kBq U^{235}	Human health effect model	[258]
IONIZING RADIATION	Ionizing radiation E (interim)	CTUe	Human health effect model	[259]				
LAND USE	Land use	kg C deficit	Soil organic matter loss	[260]	Land use	Soil quality index (dimension less)	LANCA	[261]
OZONE DEPLETION	Ozone depletion	kg CFC-11 eq	Based on 1999 WMO assessment	[262]	Ozone depletion	Ozone depletion potential	Steady-state OP	[263]
PARTICULATE MATTER	Particulate matter	kg PM2.5 eq	RiskPoll model	[195]	Particulate matter	Disease incidences	UNEP	[264]
PHOTOCHEMICAL OZONE FORMATION	Photochemical ozone formation	kg NMVOC eq	ReCiPe	[265]	Photochemical ozone formation	kg NMVOC eq	ReCiPe	[265]
RESOURCES	Mineral, fossil and ren resource depletion	kg Sb eq.	CML 2002	[266]	Resource use, minerals, and metals	kg Sb eq.	CML 2002	[266]
RESOURCES					Resource use, energy carriers	MJ	CLM 2002	[266]
RESOURCES	Water resource depletion	m3 water eq.	scarcity adjusted of water	[267]	Water scarcity	kg water eq. Deprived	AWARE	[268]
TOXICITY	Freshwater ecotoxicity	CTUe	USEtox	[197]	Freshwater ecotoxicity	CTUe	USEtox	[197]
TOXICITY	Human toxicity, cancer effects	CTUh	USEtox	[197]	Human toxicity, cancer effects	CTUh	USEtox	[197]
TOXICITY	Human toxicity, non-cancer effects	CTUh	USEtox	[197]	Human toxicity, non-cancer effects	CTUh	USEtox	[197]

The impact categories and methods used in this thesis are presented in Table 7 and in more detail below. This thesis uses global default characterization factors and the International Reference Life Cycle Data System (ILCD) [118] recommended characterization methods. The ILCD set of impact assessment methods is commonly used in LCAs performed in a European setting. Various impact categories and characterization factors were tested and evaluated throughout the thesis to investigate whether the choice of various characterization factors for the same impact category would alter the results. This investigation was performed iteratively (as new characterization facts were introduced to the modelling tool OpenLCA) as the characterization factor models are developed continuously. The same approach was taken for background databases when accessible to the research group.

Some of the technologies included in this thesis are under rapid development or are expected to be optimized further in the coming years. To easier interpret the results presented in this thesis, the uncertainties and technology development, carried out in already published papers (Papers C and D), are analyzed further in the related chapters.

3.6.1 Life cycle assessment model and approach in Paper C

Paper C presents an LCA of two different propulsion systems that use electromethanol: electromethanol use in a so-called RoPax ferry, and one where electromethanol is used in combination with a CO_2 capture system onboard. The life cycle assessment in Paper C was modelled with a clear distinction between foreground and background systems, as well as with separate computational models for the seven fuel concepts assessed. An outline of the HyMethShip processes and the electromethanol processes are shown in Figure 9. The HyMethShip onboard carbon capture system applies a novel more efficient technology for carbon capture (membrane reformer) [269], which has been proposed for use onshore, but is currently not used at scale.

The onboard systems are designed based on primary data from project partners in the HyMethShip project, where system models and emission measurements were combined with scientific models to create a unit-based flow model. This included onboard components as well as vessel power demand. The fuel production pathways (from energy input to bunkering) were modelled using data gathered from peer-reviewed and grey literature (industry documents, consultant reports etc.) which was converted into LCI grade data. The model was built in the computer software openLCA using primarily NEEDs [270] and European reference Life Cycle Database (ELCD) as background databases. The model as it was designed for the academic paper is available in full online (see Paper C). A closed loop approach was used for the system to solve multifunctionality (see Chapter 2).

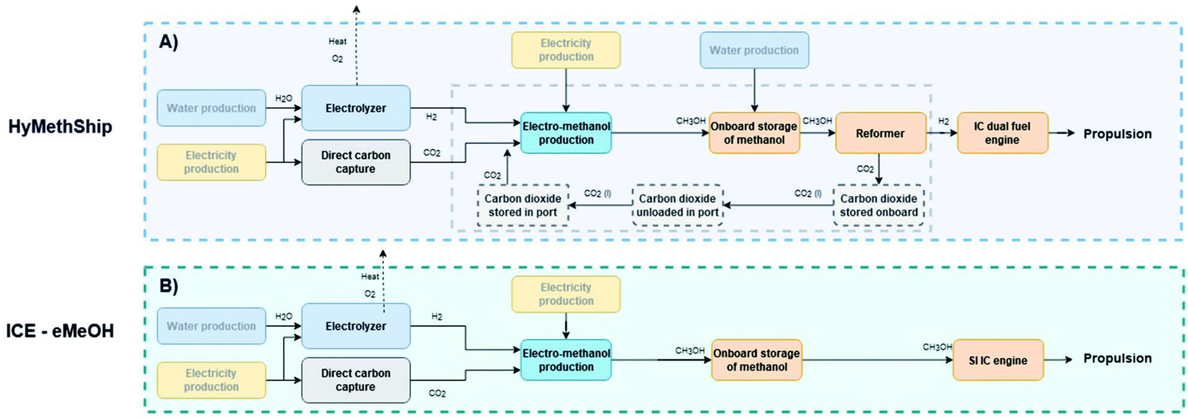

Figure 9 Outline of the life cycle model for the two concepts related to carbon-based marine electrofuels assessed in Paper C. Acronyms used: HyMethShip= The Hydrogen-Methanol Ship propulsion system using onboard pre-combustion carbon capture, H2=hydrogen, CO_2=carbon dioxide, CO_2(l)= liquefied carbon dioxide, CH_3OH=methanol, O_2=oxygen, H2O=water, ICE=internal combustion engine, eMeOH=electromethanol, SI=spark ignited, IC=internal combustion.

The influence of the uncertainties around the potential future technology development in the main life cycle was investigated using Monte-Carlo analysis. The uncertainty ranges were estimated by experts within the project, as well as through literature data. Several fuel and propulsion concepts were investigated to identify when the electrofuel pathways are the main driver of environmental impact and how different concept set-ups compare. The normalization used in Paper C simply gives an indication of the comparison between the alternatives and does not say anything about the size of the individual impact in relation to the environmental effects they cause in the natural systems.

3.6.2 Life cycle assessment model and approach in Paper D

The life cycle assessment in Paper D was modelled based on the amount of available biomass waste streams in Sweden rather than from the start point of a technology's use. It takes a more absolute approach in its design and investigates if a production of this scale would be beneficial from the viewpoint of the maritime stakeholders. The argumentation for this approach, and the general design, is in line with the proposal of an absolute LCA based in scenario development as proposed by Hauschild et al. [136] among others. The model was built in the computer software openLCA for LCA simulations and Excel for quantitative estimates in scenario development. NEEDs, Ecoinvent, and ELCD were used as background databases. An outline of the foreground system processes is shown in Figure 10. The functional unit is the total mechanical energy to the propeller shaft from using 21.95 TWh/year of liquified renewable methane gas produced in Sweden, 2045.

Paper D intends to inform on if the total environmental impact from the investigated product system (renewable methane production in Sweden in different scenarios) are within the environmental space which is available for the activity. The assumptions on engine models, biogas upgrading technology, electricity production, and electrolyzers are therefore based on assumptions on technology mixes and the sensitivity of these assumptions assessed through sensitivity analysis. The model includes prospective parts as it models emerging technologies but combines these aspects with theory around absolute LCA.

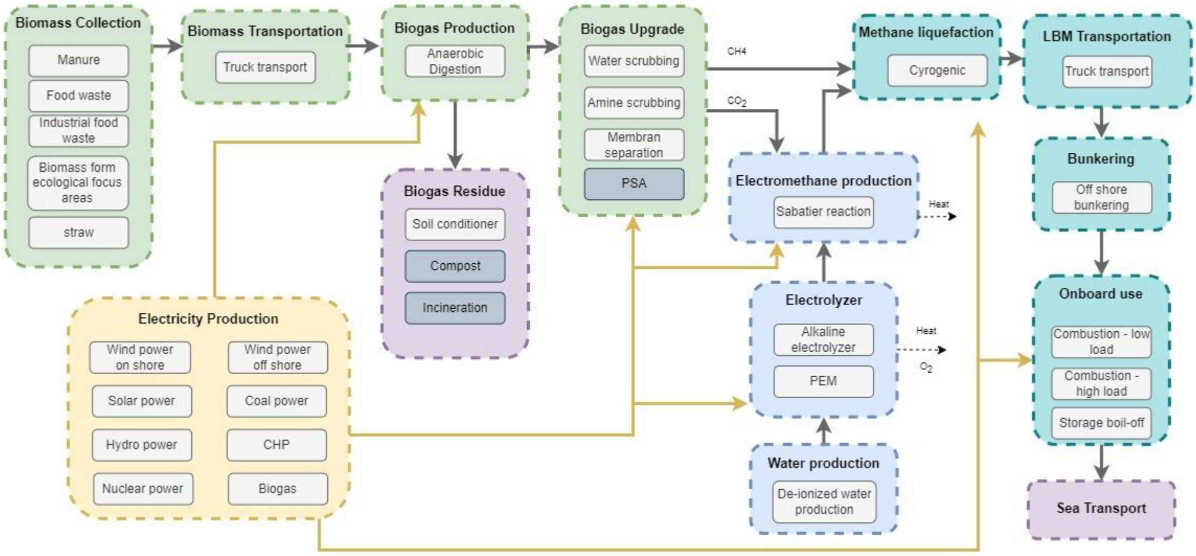

Figure 10 Outline of the processes included in the modelling of the carbon-based marine electrofuel concepts assessed in Paper D. Green boxes are processes tied to the biogas production. Blue boxes are processes related to electro-methane production. Purple boxes are products produced by the system. The yellow box shows the energy production processes. Teal boxes show processes common for the electro- and bio-pathways. Grey processes have been considered in the technology selection but excluded in the final assessment. Acronyms used: PSA= Pressure swing adsorption, CGP=combined heat and power, PEM=Proton exchange membrane.

4 THE LIFE CYCLE DATA INVENTORY MODEL

Life cycle inventory data has been developed throughout the research process of this thesis. This chapter gives a short overview of the tank-to-wake emissions used in Papers C and D. Data for included processes can be found in the electronic supplementary information appended to each Paper. The use phase (i.e., tank-to-wake) emissions for different marine fuels investigated in the thesis are presented in Table 8.

Table 8 Tank-to-wake emissions used for assessment in Paper C and Paper D. Note that they are not developed for exact comparison. For comparative assessment figures, see Brynolf et al. [271].

Fuel and engine	2-stroke LMG		4-stroke LMG		2-stroke MGO		Otto Methanol		4 stroke Methanol retrofit		HyMethShip		
Load factor	75-80%	15-20%	75-80%	15-20%	75-80%	15-20%	75-80%	15-20%	75-80%	15-20%	75-80%	15-20%	Back-up using MeOH
Emission in g/kWh	Paper D (notable value range)	Paper D (notable value range)	Paper D (notable value range)	Paper D (notable value range)	Paper C	Paper C	Paper C	Paper C	Paper C	Paper C	Paper C	Paper C	Paper C
CO_2	403	403	402	436,0	677	728	583	630	597 [173]	640 [173]	0	0	697
Black carbon (BC)	3.6E-6	3.6E-6	3.6E-6	1.08E-5	0.053 [272]	0.057 [272]	0	0	0.016 [173]	0.033 [173]	0	0	0
CO	1.9	1.9	1.9	6.15	1.1[3]	2.2 [3]	4.8 [273]	3.5 [273]	3.7 [173]	6.6 [173]	0.128	0.0037	10.69
N_2O	0.01	0.01	0.01	0.01									
CH_4	0.2 (no verifying measurements found)	0.2 (no verifying measurements found)	3.5 (1.5-6.3 [274])	20 (6.7-100 [274])	0.004 [3]	0.008 [3]	0	0	0.02 [173]	0.04 [173]	0	0	0
NO_x	3.4	3.4	2.1	2.7	13.2 [275]	13.2 [275]	2.6 [273]	3.8 [273]	6.5 [173]	12.3 [173]	0.078	1.6	1.78
NMVOC	0	0	0.5	4	0.2 [275]	0.4 [275]	0	0	1.295 [173]	1.46 [173]	0.0028	0	0.843
PM_{10}	0.04	0.04	0.04	0.04	0.38 [3]	0.43 [3]	0.141 [273]	0.153 [273]	0.093 [173]	0.01 [173]	0.0213	0.0126	0.17
SO_x	0	0	0	0	0.408	0.439	0	0	0.05 [173]	0.074 [173]	0	0	0
NH_3	0.05	0.05	0	0									
Formaldehyde	0	0	0	0	0	0	0.225 [273]	0.242 [273]	0.00049 [173]	0.00053 [173]	0.0141	0.0119	0.27
SFC	136.8	136.8	147	172	204	219.8	414	446	365	370	361	389	501
Pilot fuel consumption	8.16	8.16	3	9	N/A	N/A	N/A	N/A	25	37	N/A	N/A	N/A

5 THE MARINE FUEL CHOICE

The results of Paper A are directed towards answering RQ1 (see Figure 2 and Figure 8). The results are presented as Themes, which act as qualitative descriptors of how maritime cargo stakeholders view barriers and drivers to the marine fuel choice. Table 9 presents the eight themes of how the stakeholders views barriers and paths forward for the marine fuel choice. The barriers identified in Paper A are interlinked and interdependent, indicating a complex relationship where more than one countermeasure will be required to overcome the barriers. The marine fuel choice is governed by several goals which will not be optimized at the same point (for example cost and sustainability), creating a complex issue which requires further attention (Paper A).

The higher fuel price of alternative fuels (a term primarily used by the respondents) is a central barrier. However, the stakeholders indicate that under stringent environmental targets or with clear customer demands the fuel price barrier will be overcome. The respondents indicate that more knowledge of the environmental performance of marine fuels, their production pathways, and how they might be assessed in coming legislation to be key issues for alternative fuels to be used more. The fuel supply and uncertainty of which fuels will be competitive still require further attention, with several respondents highlighting a need to secure their specific fuel supply pathways. The introduction of efficient and fair legislation was viewed as positive by all interviewees. however, there are identified measures that can be taken by the cargo owners and shipping companies regardless of legislative measures: communicate demands and wishes clearly between parties, participate in collaborations, create sustainable business models, track emissions through the value chain, and actively support legislation.

Paper A reflects a broader perspective on marine fuels, whereas Paper B, C, and D focus on electrofuels and their environmental impacts. How the themes of Paper A relate to carbon-based marine electrofuels are shown in Table 9. The stakeholder's views of mitigating environmental impacts from maritime transport are not all coherent with the scientific theory presented in this thesis, as they represent each individual's perspective on sustainable shipping. However, the results provide insights into how the stakeholders view the challenge of marine fuel choice, as well as direct method choices such as the function to investigate, the environmental impacts to consider, and the background system. The final function provided by a fuel, i.e., the reason for its use, depends directly on the system boundary of the viewpoint of the user. The results from the LCAs in Paper C and D are dependent on the use case and if the electrofuels could have a role in the marine fuel choice (where example of uncertainties for an increased use are high costs, non-feasibility, or limited environmental impact mitigation).

Table 9 Themes developed from 17 stakeholder interviews of shipping companies, cargo owners, cargo brokers, freight forwarders, and ports (Paper A) and how they relate to the specific subject of carbon-based marine electrofuels.

Themes	Summary	Type	Connection to marine electrofuels
Communication is challenging	It is challenging for stakeholders to aligning their language, definitions, and perspectives to effectively communicate, collaborate, and implement cohesive strategies and initiatives	Barrier	Overall aim of this thesis
If someone pays, we can choose better options	There is a lack of agreement of whom should pay for the transition of the maritime industry	Barrier	Frames the conversation on the cost of electrofuels
There is risk in choosing a low-emission fuel today	There is a tendency to apply nirvana fallacy and continue to look for a silver bullet rather than to make a choice	Barrier	Electrofuels could act as a complimentary fuel production pathway to secure future fuel supply
We lack knowledge and data	There is a lack of relevant knowledge and data regarding low-emission fuel options, technologies, and their implementation as well as uncertainties of how to weigh trade-offs between options	Barrier	RQ 2-4
We want to do this, but we need support	There is an agreement among the stakeholders that low-emission fuels are required, but external pressure is needed as there are prohibitive costs and too many uncertainties	Barrier	RQ3-4
Collaboration creates stability	Involving multiple stakeholders foster communication and innovation, and supports development of comprehensive solutions	Path forward	-
You adapt when you must	Change is often driven by necessity rather than convenience or preference. External drivers are needed to successfully navigate towards low-emission maritime transport	Path forward	Will electrofuels be promoted in the legislation?
Business models exist	There are cases where the barriers are overcome today by singular stakeholders and by groups	Path forward	-

The choice of marine fuel is complex as each option has specific characteristics and different technical, economic, environmental, and social performance (Paper A). As such, the criteria to consider when choosing fuels are varied and with new social expectations and demands new aspects must be considered. However, whether to adopt a low-emission marine fuel is not just a choice between fuel options. Figure 11 shows a summary of the environmental pressure mitigation technologies which can be applied in maritime transport, where combinations of measures are possible to use to reach reduction targets. The marine fuel choice must be understood within this context as well as the socio-technical decision context, meaning that the LCA results from Papers C and D must be understood in the context of the decision-making process.

There are cases where low-emission fuels have been implemented at scale in other transport sectors and cases where implementation measures have failed. An example where the use of an alternative fuel was increased is the ethanol program in Brazil [276], where direct government mandates and economic tools were used to increase domestic ethanol production and establish an ethanol demand in cars. In contrast, ethanol promotion as a transport fuel in the USA, as well as in Europe, did not lead to a lasting market although despite government interventions which generated an initial market growth. See for example Sprei [277] describing how sale shares of new sold ethanol flex-fuel cars increased, reaching in 2008 almost 25% of the Swedish market, but then dropped to 5% in 2011.

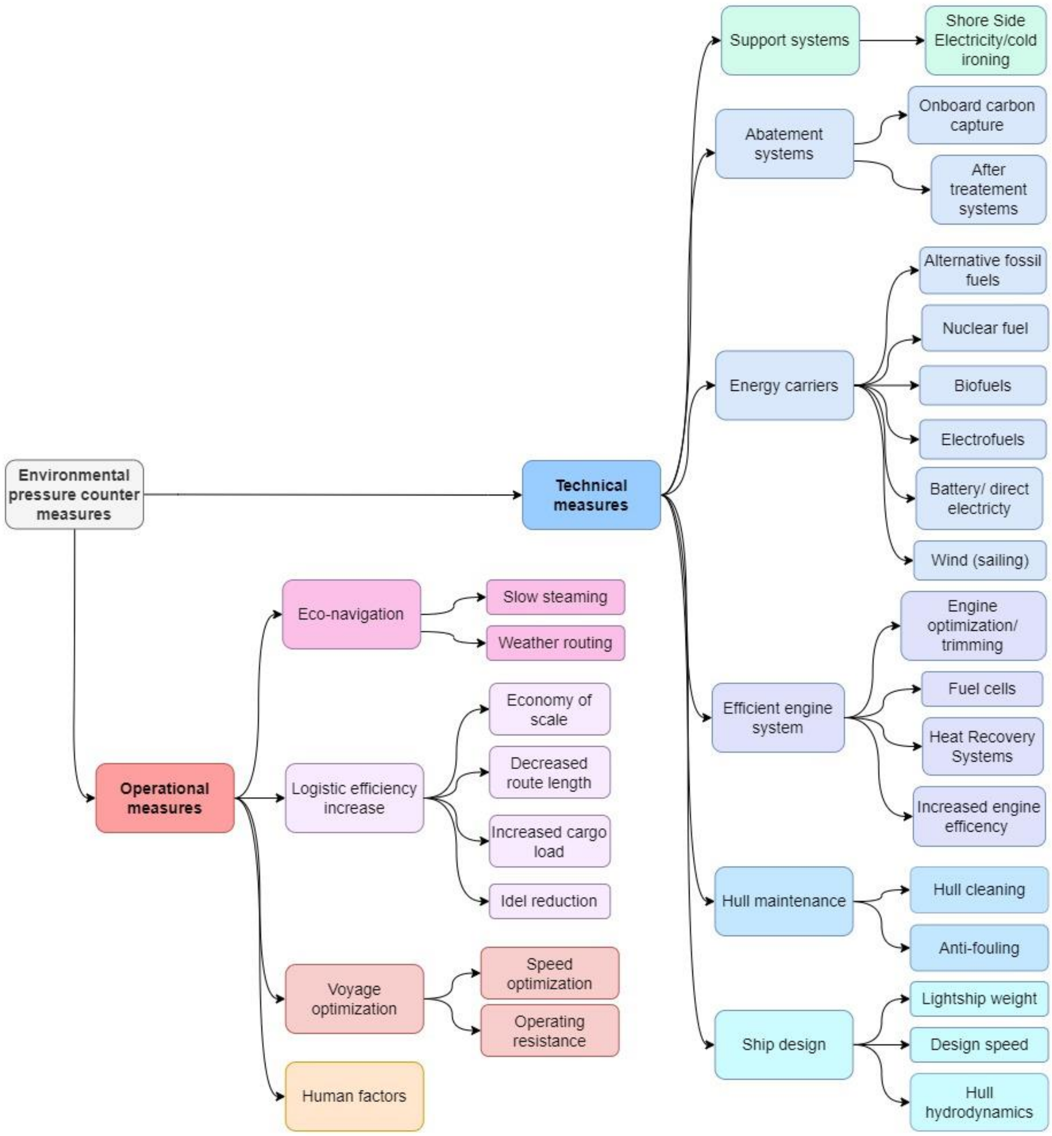

Figure 11 Environmental pressure mitigation technologies categorized based on measure types. Adaptation from Xing et al. [278] and Andersson et al. [15] based on interview material in Paper A. Measures presented with direct impact on environmental performance the vessel level.

6 ENVIRONMENTAL IMPACTS OF CARBON-BASED MARINE ELECTROFUELS

Paper B identified several papers that study the environmental performance of electrofuels, but no full life cycle assessment was found investigating applications in the maritime transport sector. Figure 12 presents an outline of the generic electrofuel life cycle, including system boundary set-ups from the literature review in Paper B and the system boundaries used in Papers C and D (see Boundaries I and K). The environmental impacts of marine electrofuels were modelled in Papers C and D. The focus of Paper C is the use of electromethanol in a RoPax vessel and Paper D focuses on use of electromethane at fleet level. Several studies conclude this variability in system boundary and functional units as being a negative aspect that should be removed through standardization, primarily due to the difficulties in comparing the results (Paper B). However, as shown in Paper B and through this thesis, this variability allows us to show what should be considered general driving factors for electrofuels' environmental impact.

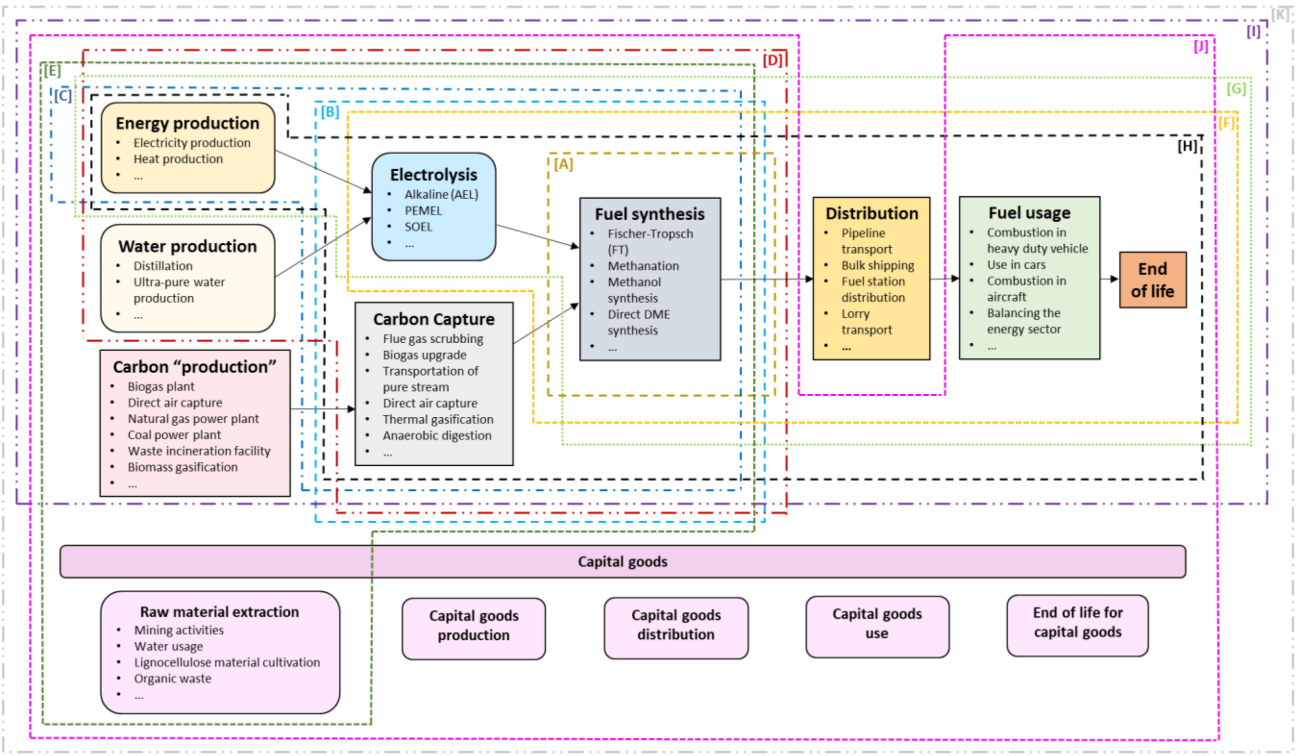

Figure 12 . Simplified illustration of the electrofuel life cycle from cradle to grave as presented in Paper B. Dotted lines mark system boundaries used in reviewed environmental assessments. Within each box different production alternatives for the same process step in the life cycle are listed (A:[279], B:[46, 280-284], C:[46, 285, 286] (however this only applies for one of the cases in [285]), D:[287], E:[281], F:[288], G:[282, 289], H:[290-293], I:[294] and Paper C, J:[284, 295] K: Paper D).

6.1 Life cycle assessment of electromethanol

Paper C shows the potential environmental performance of using electromethanol onboard vessels. The assessment suggests electromethanol to have low environmental impacts in most assessed categories in the conditions assumed in the study (Figure 13). Compared to marine gas oil, electromethanol shows a reduction in climate impact, particle emissions, eutrophication effects and acidification (Paper C). The results are at a level where utilization in the sector could lead to lower impacts compared to conventional fuels for most investigated impact categories. However, the assessment is limited to the environmental effects of vessel trip and some potential trade-offs are identified as human health impacts appears to increase (for results and further discussion on toxicity see Chapter 6.3). The environmental impacts of electromethanol are in the same range as those of using conventionally produced biogenic methanol, but this biomethanol show slightly higher impacts in all categories expect toxicity (when electromethanol is produced using wind power) (Paper C). The energy source used in fuel production is influential for the system performance of electromethanol for all environmental impacts (Paper C). This correlation is discussed further in the following chapters.

The methanol synthesis process for electromethanol significantly influences the impact on non-cancerous human toxicity (Paper C). This influence primarily stems from the adverse health consequences linked to methanol emissions during fuel production. Unintended releases of methanol in various stages of the production process will play a role in magnifying this effect. Consequently, it becomes crucial for producers, users, and designers to work towards minimizing fuel leakages across the entire lifecycle independent on production pathway.

The results of Paper C demonstrate that it is possible to provide live and timely feedback to maritime technology developers and identify intervention points and potential solutions for the optimization of the technology. Methanol is regardless of production pathway only used on singular vessels today (Paper A) but is an increasingly common fuel in the ship orderbook [296]. The data used for CI ICE calculations in Paper C was gathered from measurement of onboard operation of methanol, moving the input data to the highest level in the hierarchy of data estimation.

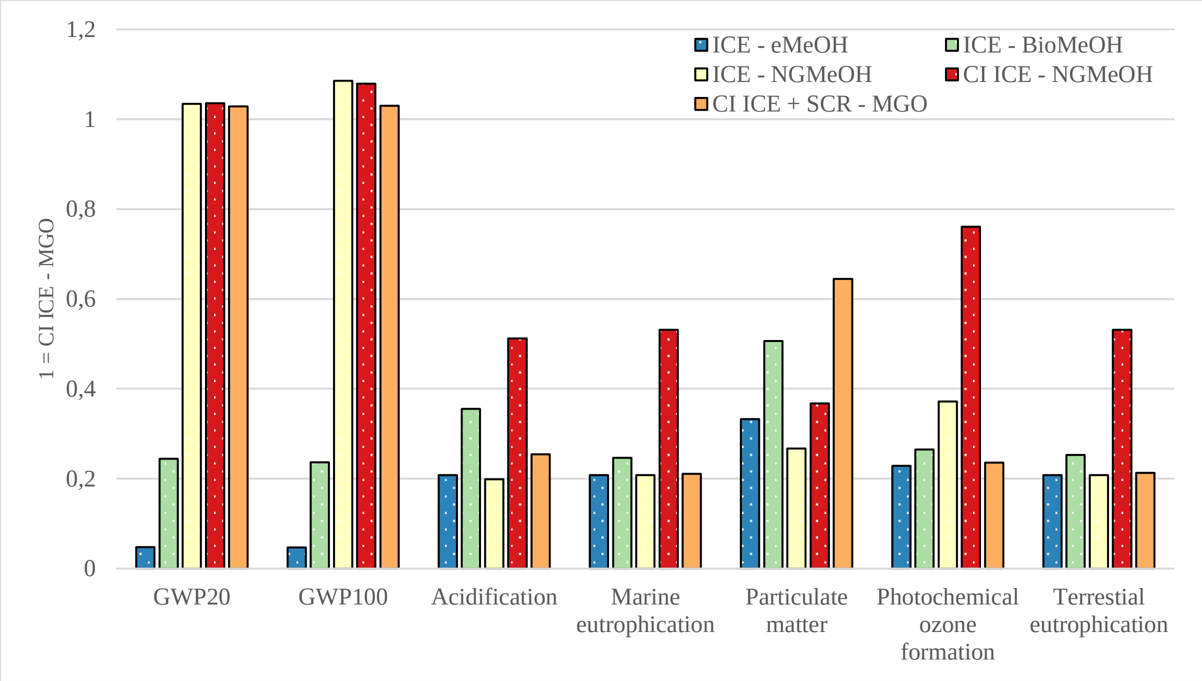

Figure 13 Life cycle assessment results from Paper C regarding methanol assessment cases. Impact categories where biomass and electrofuel were found to have lower impact than MGO are presented: GWP, Acidification, Marine eutrophication, Particulate matter, Photochemical ozone formation and Terrestrial eutrophication. The assessed concepts are: SI ICE using electro-methanol (ICE - eMeOH), SI ICE using biomethanol (ICE - BioMeOH), SI ICE using fossil methanol (ICE - NGMeOH), CI ICE using fossil methanol and pilot diesel (CI ICE - NGMeOH) , CI ICE using marine gas oil (CI ICE - MGO), and CI ICE using MGO and Selective Catalytic Reduction (CI ICE+SCR - MGO). Results normalized per CI ICE using MGO and presented per round trip between Gothenburg and Kiel on a RoPax vessel. The y-axis indicates the same values for both sides of the graph, where 1= CI ICE – MGO. It should be noted that the engines assumed for "ICE" cases are spark-ignited engines and as such do not fully represent the engines commonly used in maritime transport. CI ICE shows an engine system used today (retrofitted system). Acronyms used: GWP= global warming potential, SI= spark ignited, ICE= internal combustion engine, eMeOH= electromethanol, NGMeOH= natural gas-based methanol, CI= compressed ignited, MGO= marine gasoil.

6.2 Life cycle assessment of electromethane

Given that the maritime industry has already transitioned to methane gas (LNG) as its main alternative fuel (see the introduction of this thesis), exploring resource-efficient waste management for a renewable option (biogas) could be crucial in mitigating challenges faced by one of the hardest-to-abate sectors worldwide.

The of Paper D results show reduced climate impact for the shipping sector with increased production levels of renewable methane gas (RLMG) in Sweden, both when produced through anaerobic digestion (AD) and synthetically through electrolytically pathways (Figure 14). However, only one scenario archives reductions of 65% and notably scenarios for electromethane produced using DAC and flue gas capture does not reach a total reduction of more than 50% compared. The results are strongly linked to the domestic power sector and its environmental impact (Paper D). Domestic GHG emissions in Sweden will increase as the RLMG production is expanded beyond municipal waste and manure-based production. When produced at scale, the combined production of ad-RLMG and e-$RLMG_{as}$ has the lowest impact in most categories (Paper D). This is due to (1) the increased resource efficiency (a by-product/waste stream of AD is further utilized at no environmental cost) and (2) that environmental hotspots are divided across categories.

Examining trade-offs and environmental pressures related to scaling up Swedish biogas production from anaerobic digestion and using the waste stream of CO_2 to produce electromethane has led to insights on how to assess a complex system at scale. The goal is to comprehensively assess the resources required and the impact on climate, eutrophication, acidification, and human health.

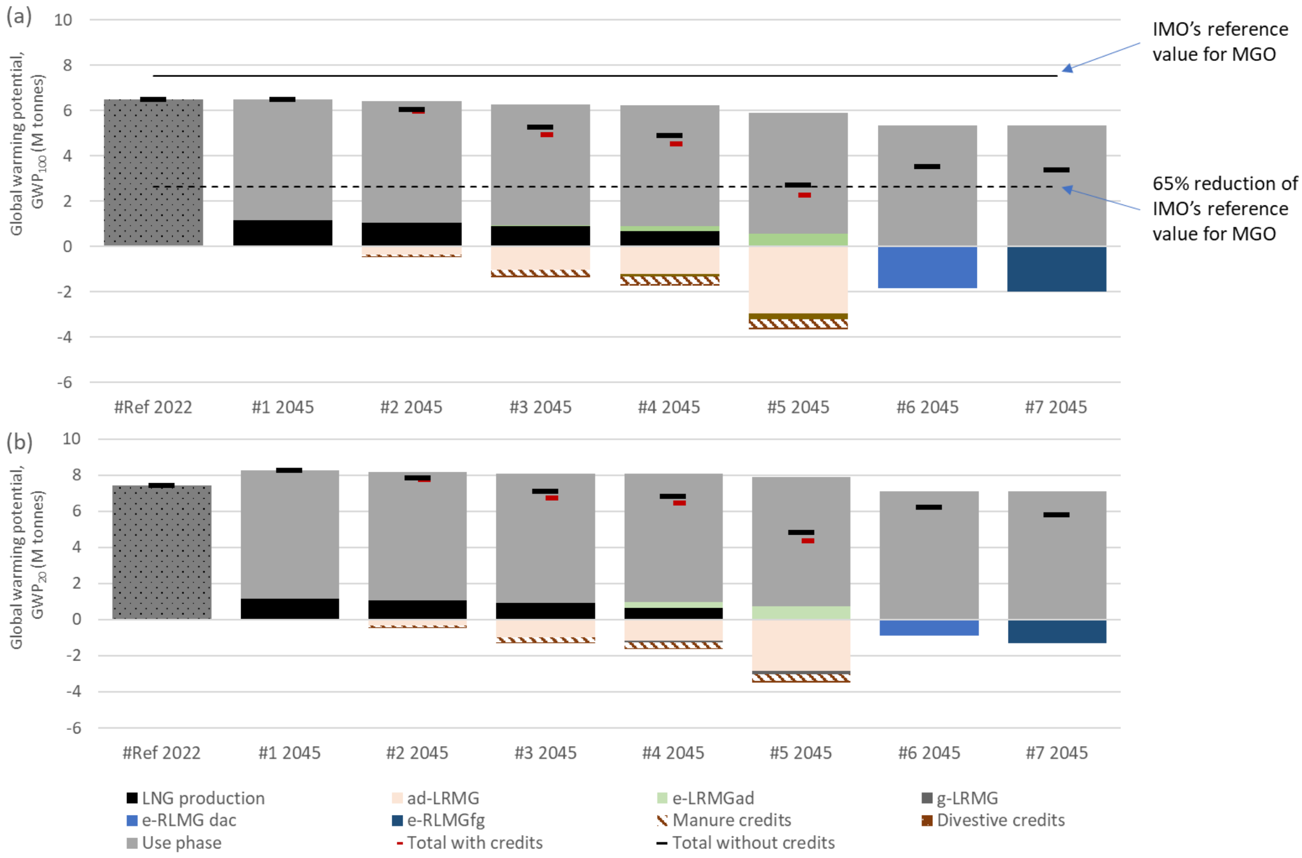

Figure 14 Results for GWP100 and GWP20 for renewable liquid methane (RLMG) production in Sweden in 2045 (Paper D). Acronyms used: GWP= global warming potential, LNG= liquified natural gas, e-RLMGdac= electromethane with carbon from direct air capture as feedstock, ad-RLMG= liquified biogas produced using anaerobic digestion, e-RLMGfg= electromethane with carbon from flue gas as feedstock, e-RLMGad= electromethane with carbon from anaerobic digestion as feedstock, g-RLMG= liquified methane produced through gasification of biomass.

One of the main influential factors on the climate impact which remains uncertain for engines running on methane is the amount of CH_4 emitted in the engine (i.e., methane slips or methane leakage). Methane when used as a fuel, whether it is produced from renewable sources or biogenic, will have a higher climate impact if not combusted [184]. The methane leakage is impacted by the engine operation, both in terms of load factor used, engine type, maintenance level and drift issues (Paper D). The difference between estimating the climate impact based on emission factors from optimal driving conditions and the actual climate impact from using the fuel might therefore be large.

In Paper D, leakage of methane from the engine had a significant effect on the LCA results, corresponding to approximately 10–30% of the total climate impact (GWP100), depending on the engine technology and fuel production route. The implications of methane leakage in the life cycles of methane-based fuels have been heavily discussed in maritime community. The scientific literature on the climate performance of LNG shows a broad range, with some articles concluding it to be critical [162, 297], others find that GHG emissions from LNG is lower than conventional fossil fuels [28, 298-300], and some stating that the benefits depend [29, 155, 301]. The range of the quantitative results in the above-mentioned studies does not appear to be the reason for this difference in conclusions, as they all show results in a similar range on both sides of the argument (from no reduction up to 40% reduction), but instead is related to what level of reduction is deemed enough to meet the climate targets.

There are several possible engine technologies to use together with the energy carrier methane. IMO's fourth GHG study [302] presents general emission factors for five methane propulsion technologies, with leakages in the main operational phase (travelling at speed, or where the load factor is around 70-85%) from 0.2 g CH_4/kWh engine power and 5.5 g CH_4/kWh engine power. The emission factor for lower loads is significantly higher for all engines [271, 303-307]. Published measurements for 4-stroke LNG engines show a wide range of methane slip (Table 8) and there is currently no standardized measurement method for methane emission from marine engines and a lack of measurements from independent researchers. No publicly available measurements of engines achieving 0.2 g CH_4/kWh engine power exists[15]. The actual emission rate to the environment is therefore not known and as such also the GWP of LNG and RLMG. Ongoing sniffer remote measurements of methane emissions from LNG vessels in operation in the Baltic region show higher methane slip per engine type than reported by IMO[16], and although the slip is lower for newer engines it remains high.

Further investigation is therefore required to establish the potential upper range of the future climate change impact of RLMG. Figure 15 shows the climate impact from using methane in an ICE from identified leakage values for a 4-stroke engine (high pressure and low pressure combined). The results are from a Monte Carlo analysis (10,000 iterations) of the net global warming potential (in g CO_2-eq.) from well-to-wake in a 20-year and 100-year time perspective for using e-$RLMG_{DAC}$ in Nordic shipping presented per 1 kWh propeller output. The methane leakage and engine efficiency for using e-LMG in 4-stroke engines as well as the share of maneuvering was varied uniformly. The methane emissions varied between 2 g CH_4/kWh to 5.5 g CH_4/kWh when cruising and between 3 g CH_4/kWh to 42 g CH_4/kWh when operating at lower speeds for the 2030 scenario, and 3 g CH_4/kWh to 20 g CH_4/kWh for the 2050 scenario. The share of maneuvering/low-speed operation performed by the vessel varied between 2% and 10%. The red dots show the results in the base assessment for the report [271], which uses a similar set-up for the same OpenLCA model as presented in Paper D (coupled with different assumptions on background

[15] Personal communication with MAN Energy solutions, August 31, 2023
[16] Ongoing work by J. Mellqvist et. al, personal communication through K. Salo. Primary data reviewed by the author.

systems to fit the Nordic context and where e-LMG refers to e-$RLMG_{DAC\ with}$ waste heat integration).

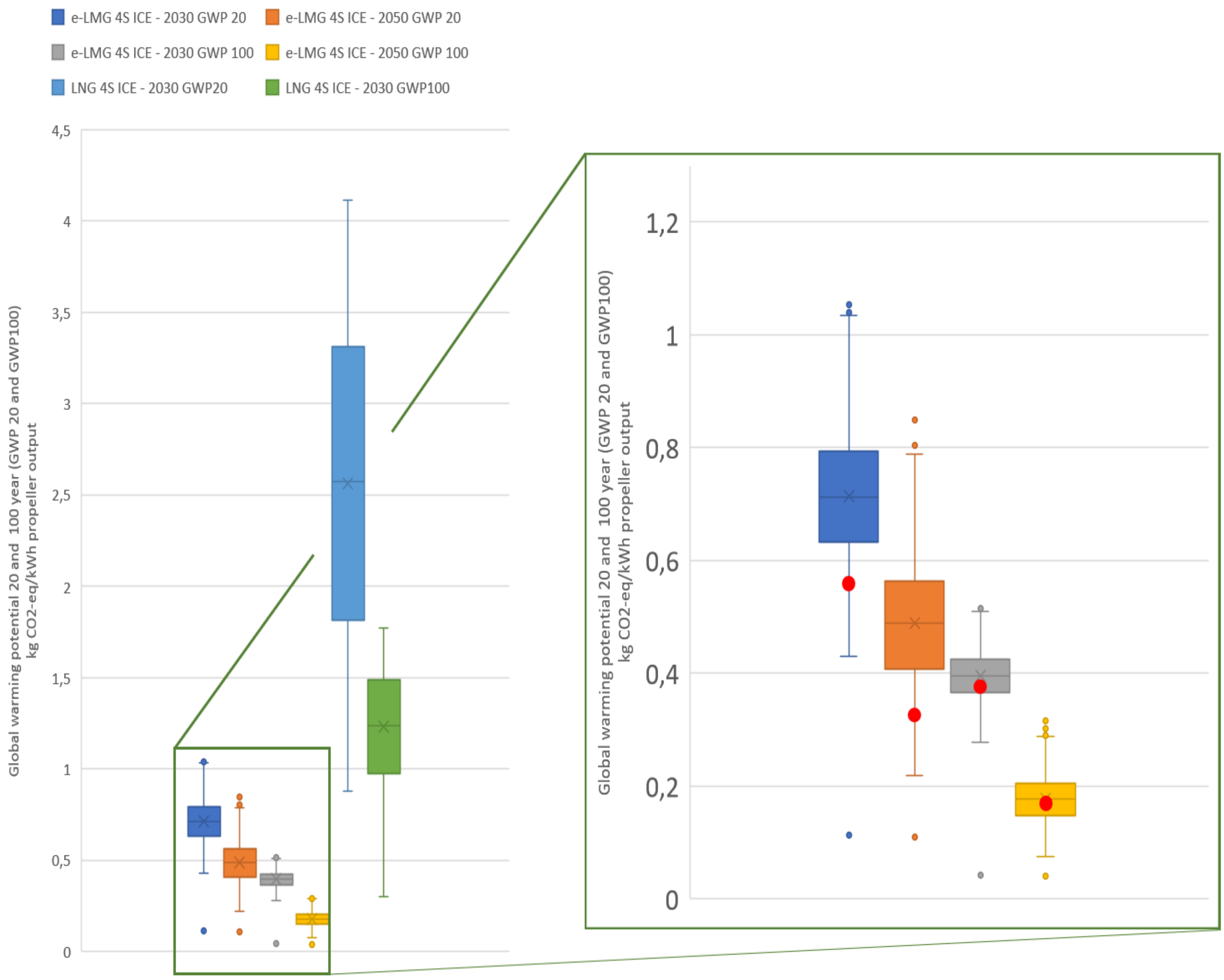

Figure 15 The results from a Monte Carlo analysis of the net global warming potential (in g CO_2-eq.) from well-to-wake in a 20-year and 100-year time perspective for using electromethane in Nordic shipping presented per 1 kWh propeller output. The red dots show the results in the base case. The box in the plot shows the 25th and 75th percentile of probability and the outliers in the simulated data are marked by singular points. The data is presented as boxplots of the probability of the different outcomes when all parameters are varied. Acronyms used: e-LMG= electromethane, LNG= liquefied natural gas, 4S ICE= four-stroke internal combustion engines.

The difference between the best-case scenario and the worst-case scenario for 2050 is large and indicates the importance of maintaining low methane emissions from the combustion process. For most values, even under remarkably high technological development scenarios, the maximum reduction possible compared to MGO is 80%, with the plausible scenario to be no more than 60%. This is data presented without additional leakages or accidents such as the 2022 Nord2 pipeline methane leak [308]. Methane leakage occurs regardless of origin and, as such, puts a lower bound on the possible climate impact of methane as fuel, regardless of the fuel production pathways. The significance of the methane slip on climate performance has led to further attention to potential technological solutions, and the development of catalysts to reduce methane slip is ongoing. However, the catalysts require heat to oxidize the CH_4 [309], and due to their high light-off temperature, heat produced during low-load operation is likely not sufficient [310]. Since the methane slip is higher at low loads (see Table 8), the use of climate impact mitigation of the catalysts is limited.

Creating a full inventory of methane emissions is challenging not only for the engine operation but also in other parts of the value chain, specifically for natural gas pathways. Emissions and leakage from pipelines are rarely monitored [311], natural gas fields emits more than often reported [312, 313], and methane feeder vessels travels nonrelated to goods traffic [314]. The climate impact assessments of LNG in academic literature must therefore be further informed with driving pattern data and emission monitoring. Fugitive emissions of methane are also occurring in biogenic processes, including biogas production [315], and have been showed to be higher than previously estimated.

6.3 Regarding the trade off with human toxicity

Increased toxicity impacts (i.e., human toxicity (cancer and non-cancer effects) and freshwater ecotoxicity)) is indicated when carbon-based marine electrofuels are used instead of fossil or biogenic fuels (Paper C, Paper D). Figure 16 shows the results for methanol presented in Paper C. The increased toxicity impacts found in Papers C and D are driven by emissions from the production of renewable energy. The same conclusions were observed in Paper B for electrofuels (see for example [283, 285, 293]) and have further been highlighted in several other review papers [19, 44, 45]. The assessment method USEtox, used in all these assessments, is known to have high uncertainties [199].

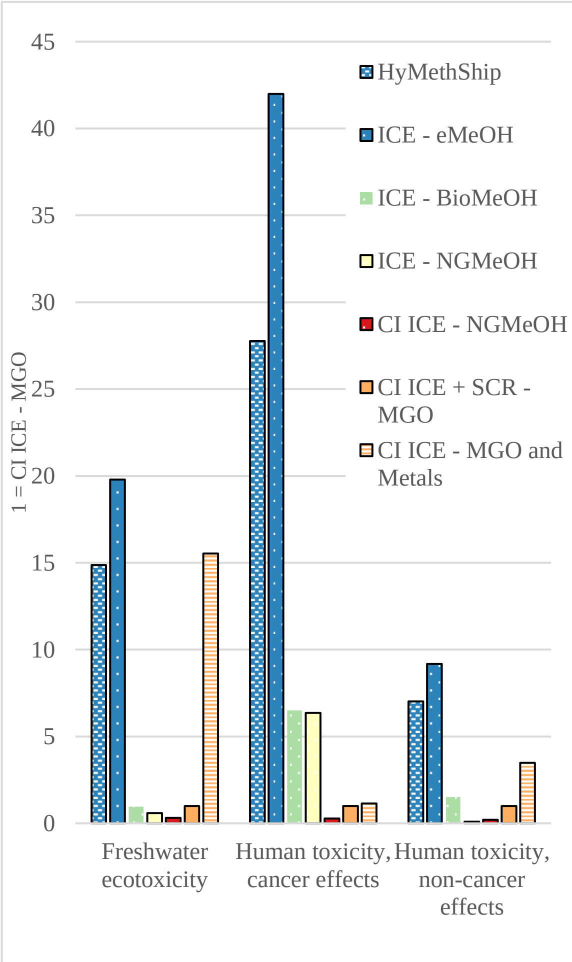

Figure 16 Toxicity impacts relative to ICE - MGO found in Paper C. Electricity input is offshore wind power in Denmark.

The relationship between renewable electricity production and toxicity impacts has been extensively investigated in the academic literature [211, 316, 317]. Laurent et al. [211] studied databased inventory data to investigate how the carbon intensity of power generation technologies correlated to other environmental impact categories. They concluded that per kWh of electricity the toxicity impact increased with lower climate intensity across all power generation technologies except hydropower and hard coal. Wind power and solar power are the two renewable technologies currently expanding the fastest on the European energy market [318], as well as often discussed as electricity sources in electrofuel production (Paper B). Wind power have requirements of rare earth elements (neodymium, praseodymium, terbium, dysprosium) and solar power often requires metals (indium, gallium, selenium, cadmium, tellurium) [270, 318]. Mining of these metals and rare earth elements causes emissions of several toxic compounds, including hexavalent chromium (chromium VI).

If the mining practices are not radically shifted these environmental impacts will increase with increased use of rare earth elements and metals [319], which would be required for new builds of wind and solar power plants [318]. Savvidou and Johnsson [317] analyzed the material requirements for large scale wind power expansion in Sweden by 2045 and pointed to the environmental pressure from mining of rare earth elements as a considerable challenge. Considering toxicity impacts is therefore highly relevant for electrofuels environmental impact if upstream emissions from electricity production can be allocated to the fuel.

For comparative analysis, we must also consider if the life cycle investigated fully informs the impact category. For the LCIA to reflect a proper assessment, the LCI must contain all relevant emissions. The electrofuels contain little to no trace elements (the synthesis process creates a pure product stream, but the distribution could cause impurities) and, as such, no emissions not already accounted for in the inventory relevant to toxicity was identified. However, the work of conducted through this thesis used previously published marine fuel LCAs as a starting point for the emission inventory and has not expanded the primary emissions to include emissions in the fossil fuel life cycle not traditionally included [201].

The appendix of Paper C expands on some of the known emissions with toxic properties (metals) found in marine fuels from fossil sources (crude oil). Exhaust gas emissions from fuel oil combustion are known to contain metal emissions [320]. Figure 16 include this assessment (CI ICE – MGO and Metals) and the resulting toxicity impacts increase significantly for non-cancerous health and ecotoxicity. It is thus shown that combustion emissions of metals from MGO have direct relevance for the toxicity impact category. However, electricity production is also shown to affect the toxicity impact category and it could be discussed under which conditions electrofuels can be considered low-emission fuels. If this definite hotspot leads to electrofuels having a higher toxicity impact, compared to MGO can, however, not be fully assessed by the LCA models used in this thesis. It should be noted that the USEtox characterization factors have a large uncertainty range depending both on lack of toxicity data and uncertainty in ecosystem fate assessment. The high uncertainty of data on toxic emissions to the environment speaks against further expansions of life cycle assessments since uncertainty in data will generate uncertainties in the results.

Propulsion systems can also directly require precious minerals/metals [321], such as some components in the HyMethShip system (Paper C). For some future fuels and propulsion system options, such as fuel cells [156] and batteries [322-325], material demands, which is tied to mining issues, have created hot spots of environmental impacts. The need for mined resources is linked to more than the material used in electricity production, but renewable electricity use is the main contributing process in all reviewed papers [322-325] as well as in Papers C and D. Mining practices also impact other environmental problems, such as biodiversity. These aspects have not yet been included in marine fuel LCAs.

6.4 Particulate matter impacts and electrofuels

Particulate matter formation and impacts from smokestack emissions on human health have been a primary concern, directly and indirectly, in the Swedish shipping context over the past decades. In Paper A, the impact of NOx and SOx, as well as the introduction of legislation, was brought up by the stakeholders as clear examples of the maritime transport impact on the environment and of concerns which should have a higher priority in the decision-making context.

With combustion processes acting as a driver for particulate matter formation (see background) and the indirect effects of SO_2, NH_3, and NO_x emissions, fuels used in ICEs are expected to have impacts on particulate matter. Paper B indicated combustion occurring in the life cycle as the primary contributor to particulate matter in the reviewed electrofuel studies, and combustion processes (primarily onboard ships) are also the driving factors in Papers C and D. The range of LCA results shown in Paper B depends on the actual operational pattern and it is therefore essential to consider values close to the actual operation pattern in the assessments. This argument holds true for particulate matter emissions, as well as NOx emissions. The amount of particulate matter formation is directly dependent on the fuel properties.

Methane as fuel has comparatively low emissions of particles when used in an ICE [326], which has been pointed out as an advantage of LNG as an alternative to fuel oils [23]. The same energy carriers (molecules) have similar emission profile regardless of the fuel production pathway. However, they differ between energy carriers and the electrofuel production pathways produce fuels which lack contaminants such as Sulphur, avoiding for example SO_X formation (Papers B and A).

In this thesis, Fischer-Tropsch (FT) fuels as well as the biodiesel hydrotreated vegetable oils (HVO) have been left out. Although they are relatively expensive, and the production capacities may be limited, especially the bio-oils needed as feedstock for HVO production is limited, they may still be part of the future fuel market. They are both suitable as drop-in replacements for fossil fuels in "hard-to-electrify-sectors" such as the aviation, off-road and marine sectors. Some new types of energy carriers with new chemical structures could also be possible to produce as electrofuels, such as oxymethylene dimethyl ether (OME). The additional oxygen in the molecular structure of OME appears to lead to lower particulate emissions in the combustion stage [294]. OME has been shown to reduce the emissions of NO_x and soot by 43% and 75%, respectively compared to diesel when used in lightweight vehicles (Paper B). The life cycle results are, however, dependent on the electricity source when producing the OME. Thermal power stations, which burn a fuel to produce electricity, e.g., coal power plants, can increase NO_x [294] and other particles emissions over the fuel's entire life cycle, depending on the combustion characteristics, and the electricity demand in the electrolysis process.

6.5 Acidification, eutrophication, and ozone formation

Terrestrial acidification, marine eutrophication, and ozone depletion are indicated to be lower for electrofuels than conventional fuel options in Papers B and C. However, these impacts appear to be driven by upstream emissions. The assessments in Papers B and C have primarily been limited to emissions from the direct production and use of the fuels rather than materials required in those processes. Paper D includes a larger amount of capital goods and shows higher impacts from the electrofuel investigated (methane) than earlier studies. Regarding the energy source used for electric power generation there is no direct correlation between carbon intensity and acidification nor eutrophication impacts [211], and the electricity input could therefore potentially be optimized to achive both low greenhouse gas emissions and low impact on acidification, eutrophication, and ozone formation.

The impact assessment methods used for assessing acidification and eutrophication have considerable limitations. Eutrophication potential is strictly dependent on the spatial (i.e., geographical) distribution of emissions, which is currently not considered in the ReCiPe methodology for marine and freshwater methodology (see Chapter 2.3). For a further discussion on spatial impact see Chapter 9.1.

6.6 Comparing electrofuels to biofuels

The sustainability challenges with biofuels are primarily tied to biomass scarcity, cultivation practices (water consumption, land use, and land use change), food competition, and biodiversity. The analysis in Paper D addresses the comparison between biomethane and electromethane and is limited to biomass flows available in Sweden which are deemed to not infringe on sustainability concerns tied to sustainability (see Börjesson [327] for calculations on potential). However, a reflection on the relationship between the environmental impacts from electrofuels and biofuels is of interest.

Water is one of the consumables in the electrofuel life cycle (see Figure 12) and is required in the electrolyzer as well as in some carbon capture processes and fuel synthesis related purposes (Paper B). Analysis of material in- and outflows show that the water consumption in electrofuels production is driven by the electrolyzer (Papers B and D). When producing H_2 from electricity all articles found in Paper B looked at production through water electrolysis. This technology has been available for the past 100 years but has not been used on a large commercial scale. The type of electrolyzer will influence the water demand of the fuel production [328]. The models in Papers C and D include water consumption of 11-15 l H_2O/kg H_2. Most LCA papers in the literature assume stoichiometric consumption at around 9 liters of water per kg H_2 or does not include water consumption as an input nor output of their analysis (Paper B). The environmental impacts of water demand from electrofuels have not yet been assessed in detail in the literature but might be of direct importance (Paper B). Sweden has low to no water scarcity issues, however, fresh water is a scarce resource and increased water use are critical in some geographical regions and the planetary boundary for freshwater change has been transgressed [329](see Chapter 9.1 for further discussion on spatial LCA). However, the quality of water required does have significance over the life cycle as the purification level (purified to deionized depending on type of electrolyzer) does have a higher production cost as well as energy requirement which impact the environmental performance (Paper B).

Land use and indirect land use change has not been in focus for electrofuel LCAs yet (see Paper B). The land use intensity per kWh of electricity is 30-100 times lower than for biofuels [202], but the energy required to produce one MJ of electrofuel is higher than that of one MJ biofuel of a similar composition (Paper B). If electrofuels are produced to satisfy the global energy need for the transport sector the electricity production sites would likely occupy a significant part of Earth's available surface [330]. Rennuit-Mortensen et al [330] have estimated that transforming the global energy sector from fossil fuels to electrofuels would be technically achievable but would require 1.1–13% of the Earth's land surface. This is however lower than for a fully biomass-based system according to their calculations. Schmidt and Weindorf [331] support this further, however, further investigations, which include for example land use connected to carbon capture and indirect land use change (including the quality of the occupied land), is needed.

6.7 The climate change impact of electrofuels

The results for climate change impacts varies in the literature (Paper B, see Figure 18) with some assessments stating negative GHG emissions while other show results five times higher than values presented for fossil fuels. Chapter 7.2 discusses the negative results in more detail, whereas here we will look at the absolute GHG emissions identified in the assessments. In Paper C, most of the climate change impact is attributed to electricity production. Figure 17 presents how the carbon intensity of the electricity impacts the overall results. The reliance on electricity input is expected as the LCA model for Paper C is designed by using open loop allocation and through assumptions of using electric support systems such as heating. No major sources of GHG emissions were identified from the system itself.

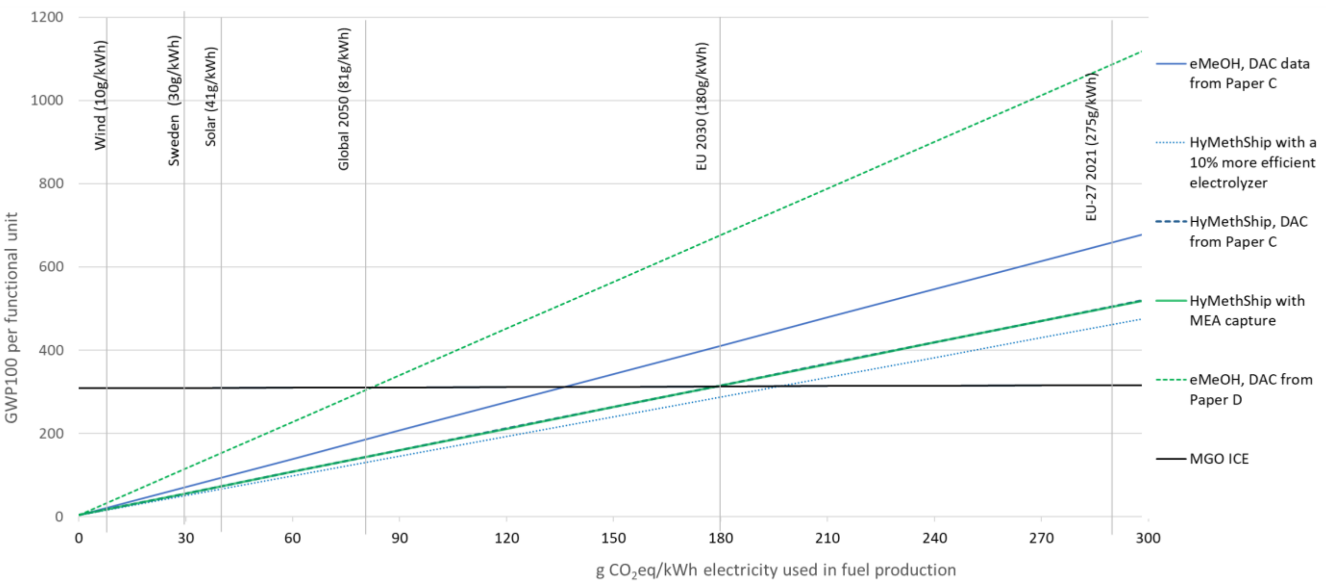

Figure 17 The results for GWP100 impact of electromethanol (using the HyMethShip concept and without) when the carbon intensity of the electricity grid mix is varied. The graph shows the systems configured in Paper C as well as updated pathways where carbon capture data presented in Paper D is used.

The literature review in Paper B indicates electricity to be the main driver in most assessments. Most papers present emissions from electricity production and/or carbon supply (carbon source and capture process) as the primary contributor to climate change

(Paper B), but electrofuels appears to offer greenhouse gas emission reductions if produced from low-carbon renewable electricity. The type of climate impacts included also varies between papers, with some including indirect land use changes leading to secondary effects, indirect emissions etc.

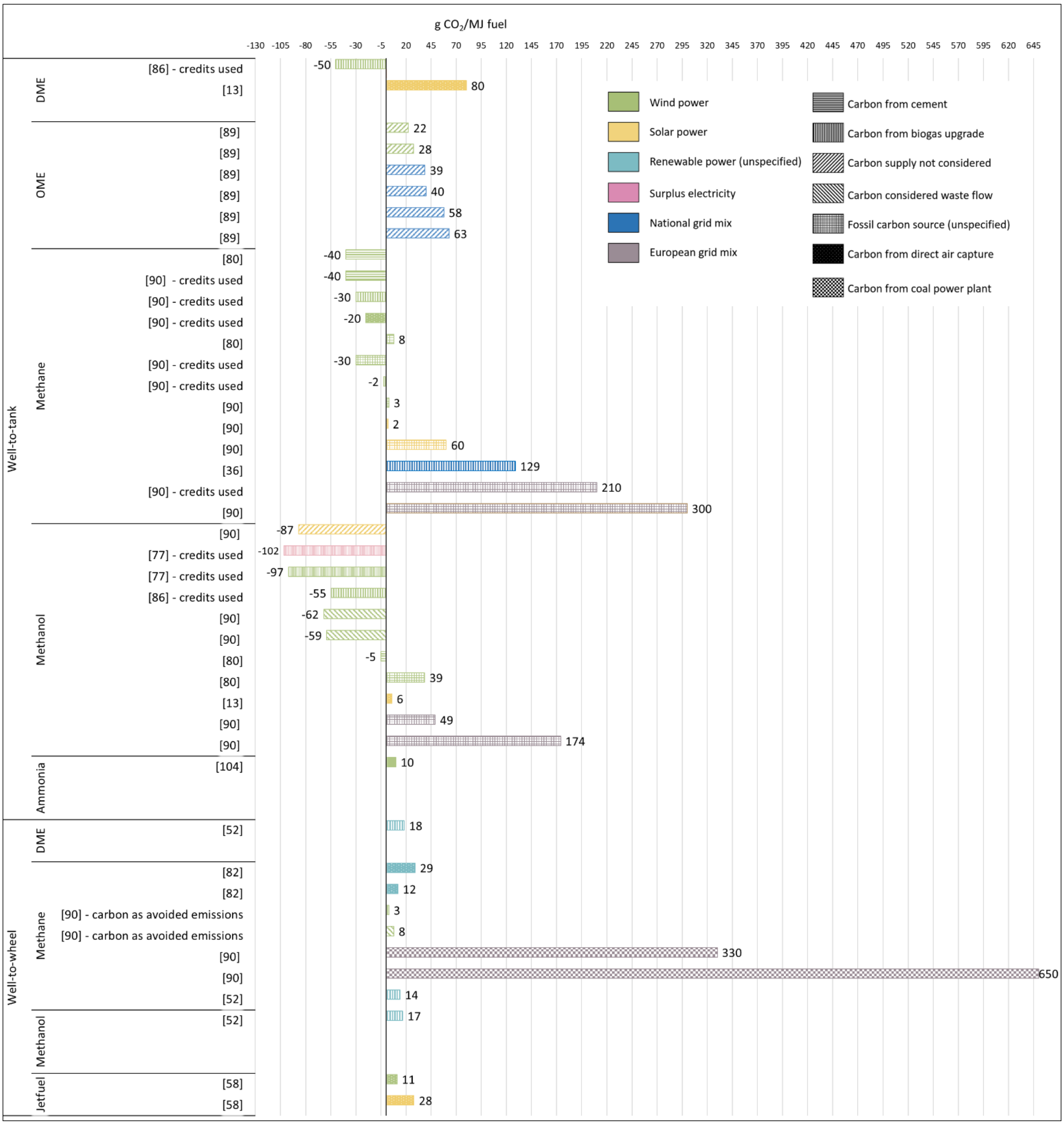

Figure 18 Summary of GHG emissions from the production of electrofuels identified in the reviewed assessments separated between well-to-tank and well-to-wheel studies. The results are not directly comparable as the methods for calculating the environmental impact from environmental streams vary. Note that the first jet fuel pathway represents methanol to jet and the second pathway uses Fischer-Tropsch. The list includes results from articles presenting values in emissions/kg, emissions/km, or emissions/MJ which have been converted to the same unit using lower heating value (LHV) of 22.5 MJ/kg for ammonia conversion, 20.1 MJ/kg for methanol, 28.8 MJ/kg for DME, and 50 MJ/kg for methane. The presented results in gCO_2/MJ_{fuel} can be converted into $kgCO_2/MWh_{fuel}$ and gCO_2/liter diesel equivalent, if multiplied with 3.6 or 36, respectively. For details, see Table S3 in Supplementary Material of Paper B. Acronyms used: OME= oxymethylene dimethyl ether, DME= dimethyl ether.

The carbon intensity of the electricity is necessary to consider when assessing the climate impact of electrofuels (Papers B, C and D), as a small change in this input alters the results significantly and can put the options as worse than their fossil counterparts. If the electricity is produced using fossil energy sources, GHG emissions are likely higher compared to fossil fuels.

Paper B concludes emissions from electricity production or carbon supply (carbon source and capture process) as the primary contributor to climate change according to the literature with two exceptions, (1) when the emissions from electricity production and carbon supply is assumed to be zero due to methodology related arguments, and (2) where heating required in the life cycle is provided by natural gas.

The heat needed for electrofuel production varies between fuels and assessment studies. However, most full-scale industrial plant requires some form of thermal energy at a system level (Paper B) and all identified plant simulations does show a need for heating, including assessments of electromethane which is created through an exothermic chemical reaction and as such produces some heat as well (Paper D.).

The heating requirement is higher for more complex energy carriers (Paper B). In the review by Artz et al [46] include two studies looking at Fischer-Tropsch liquids were harmonized, and the results indicate that the source for heat supply is of importance to reach low global warming impact for this type of electrofuels. Natural gas is already established as a fuel for domestic and commercial heating and large-scale electricity generation, and as such it could be a reasonable assumption for the heating supply in an LCA. However, for a low-emission fuel to be produced, the heating should be considered. Some studies integrate heat produced in the electrolyzer with fuel production.

Some of the life cycle GHG emission inventories presented for electromethane and electromethanol use in ships in recently published literature differ significantly from the results in this thesis. Aakko-Saksa et al. [175] report secondary well-to-wake values from Schuller et al. [170] of 6 g CO_2 eq./MJ fuel, and while the influence of electricity mixes on the overall emissions are commented on later in the text these GHG emission levels are not presented in the data tables. Lindstad et al. [176] claims to make a LCA of among other electromethane and electromethanol in shipping, but only include engine methane slip in the e-RLMG life cycle (total result of 12 g CO_2 eq./kWh engine power (0.2 g CH4 slip) and 80 g CO_2 eq./kWh engine power (5.5 g CH4 slip)) and 7 g CO2 eq./kWh engine power for electromethanol (only tank-to-wake). In a recent publication investigating marginal abatement cost [332], the well-to-tank emissions for all electrofuel pathways were set to zero, despite emissions from biopathways being considered. The tank-to-wake emissions of biomethane and electromethane also assumed a very low slip of methane from the engine (0.2 g/kWh fuel). If Lagouvardou et al. [332] used the values concluded by this thesis, the results would differ significantly.

7 ENVIRONMENTAL IMPACT OF ONBOARD CARBON CAPTURE

While the post-combustion carbon capture systems are in use and have been studied for onshore facilities, the maritime carbon capture system is still in an early stage mainly due to its unique characteristics and design limitations. Carbon capture technologies as an emerging technology was not discussed by more than one respondent in Paper A[17], and then as an option to maybe use when travelling on trade routes where options to fossil fuels are not yet available for the shipping company.

7.1 Life cycle assessment of an onboard carbon capture system

The HyMethShip concept, which is a pre-combustion carbon capture system (Figure 19), could have a lower impact on acidification, climate change, marine eutrophication, particulate matter, photochemical ozone formation, and terrestrial eutrophication compared to internal combustion engines run on either marine gas oil (0.1% Sulphur content), biogenic methanol, fossil methanol, or electromethanol (Paper C). However, this is due to remarkably high system efficiency.

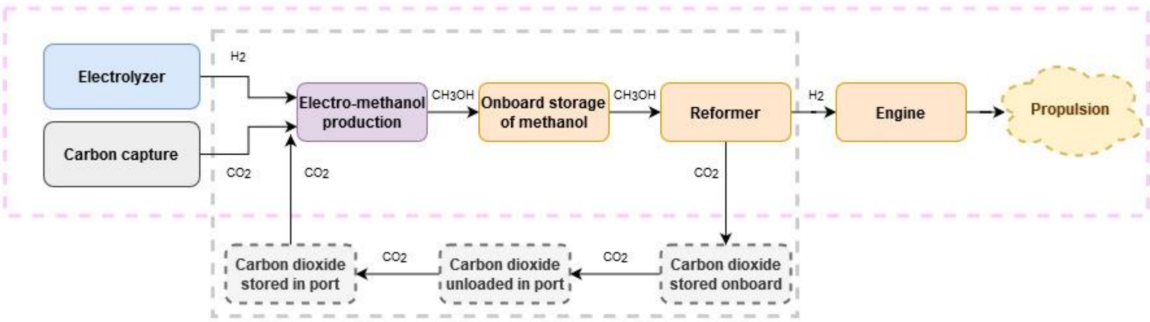

Figure 19 Outline of the HyMethShip concept. The HyMethShip concept combines a reformer, storage systems for CO_2 and methanol, as well as an ICE into one system (www.hymethship.com). Hydrogen produced from an electrolyzer is used together with captured carbon to produce methanol. The produced electromethanol is used as a hydrogen carrier stored on the vessel until required for propulsion. A reformer splits the methanol into hydrogen, which is used to propel the vessel, and carbon dioxide, which is captured and liquified, brought to shore and utilized in the electromethanol production. The orange-colored processes occur onboard the vessel. The pink system boundary shows the main processes included in this work and the grey system boundary marks the re-circulated CO_2. Acronyms used: CH3OH= methanol

Two different types of processes account for the main impact in most impact categories: (1) combustion processes (travelling at speed and maneuvering with hydrogen/methanol) for the propulsion of the vessel and (2) the electricity production processes (Paper C). To achieve the positive results of Paper C, electricity with low climate and environmental impact is required, and low NO_x emissions from combustion processes need to be maintained in the engine combustion. As HyMethShip is a pre-combustion process (Paper C), the combustion process is a hydrogen combustion process and has the combustion properties of a hydrogen propelled engine system. The engine used in the assessment is a

[17] raw data examination, not published in the main body of the paper.

hydrogen propelled ICE engine, and as testbed data on hydrogen ICE does show comparatively high values on NO_x emission. The NO_x emissions must be considered moving forward.

The same potential trade-off with toxicity impacts as with electromethanol in ICE is also seen with electromethanol in ICE with onboard carbon capture. This is expected as this is connected to electromethanol production and specifically the high electricity demand. This is also true when biomethanol and fossil methanol is used for the initial carbon input rather than the direct air capture, as the electrolyzer is the primary electricity consumer (87% of total electricity demand in the electromethanol production phase).

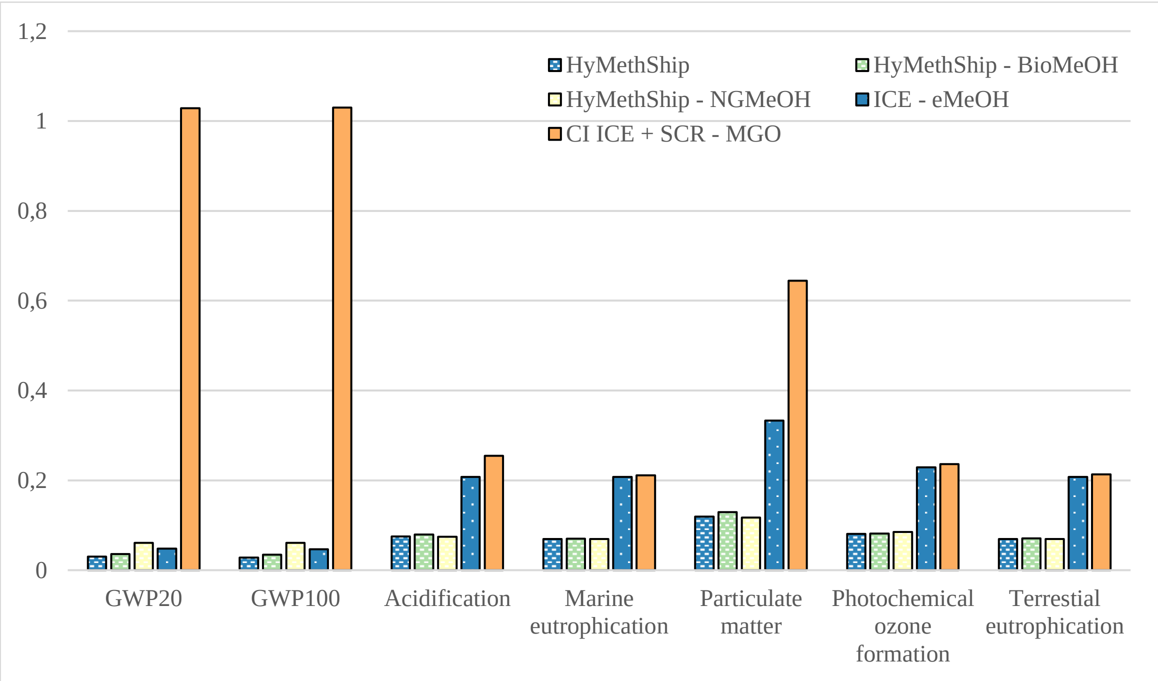

Figure 20 Life cycle assessment results from Paper C regarding onboard carbon assessment cases. Impact categories where biomass and electrofuel were found to have lower impact than MGO are presented: GWP, Acidification, Marine eutrophication, Particulate matter, Photochemical ozone formation and Terrestrial eutrophication. The assessed concepts are: HyMethShip using electromethanol from DAC and wind power (HyMethShip), HyMethShip using biomethanol (HyMethShip - BioMeOH), HyMethShip using fossil methanol (HyMethShip - NGMeOH), SI ICE using electromethanol (ICE - eMeOH), and CI ICE using MGO and Selective Catalytic Reduction (CI ICE+SCR - MGO). Results normalized per CI ICE using MGO and presented per round trip between Gothenburg and Kiel on a RoPax vessel. The y-axis indicates the same values for both sides of the graph, where 1= CI ICE – MGO. Acronyms used: MGO= marine gasoil, GWP= global warming potential, DAC= direct air capture, BioMeOH= biomass-based methanol, NGMeOH= natural gas-based methanol, SI= spark ignited, CI= compressed ignited, ICE= internal combustion engine.

One parameter not fully considered in Paper C which could impact the results is the lost cargo space due to the weight and/or the space needed for additional fuel tanks and system requirements. The technical investigations performed later in the project suggested this to not have a significant effect for the investigated case study vessel, however, this assessment was done for a RoPax vessel which is not fully optimized to carry cargo but also moves passengers and vehicles. Lee et al. [74] estimated the cargo loss, from capturing carbon from higher carbon dense fuels than methanol, to be between 2.9% and 5.3% depending on the vessel. The assessment considered three case study vessels. Law et al.

[88] assessed a similar cargo loss at less than 3% and Feenstra et al. [64] concluded the system to fit onboard a 3000 kW cargo vessel. An additional function requirement of 3% would not increase the environmental impact significantly if the relationship is directly correlated, but if the carbon capture system is associated with high costs the lost cargo spaced could be an important parameter. In Paper A, cost is identified as an important factor when the maritime sector considers fuels and propulsion technologies.

Review of the literature on onboard carbon capture indicates that the mitigation of onboard carbon emissions is with MEA capture capped around 70%, with higher emission reductions requiring increased energy use [60, 64, 69, 75, 96]. This increase of energy demand results in lower emissions of CO_2 at the tailpipe, but more requires more fuel and thereby results in additional GHG emissions being emitted cradle-to-tank [82, 91]. As demonstrated in Paper C, the mitigation potential is further limited by losses of carbon dioxide through the life cycle, such as in the off-bunkering process and during long-term storage, as well as climate emissions from energy and material required to minimize these losses and perform the storage activity.

7.2 Negative GHG emissions and the carbon source relevance for the environmental performance

Negative amounts of GHG emissions are shown as the results in several LCA studies of electrofuels (Paper B, Figure 18). Negative GHG emission results can occur in an LCA model due to the application of two different approaches: (1) GHG emissions have been removed from the atmosphere through some form of capture process, e.g., DAC or biomass cultivation, and not yet been released back to the atmosphere. For example, cradle-to-gate assessment does not include combustion of fuels and therefore can have negative results. (2) Some studies investigate how the system changes of an action, such as introduction of electrofuels, where the electrofuel are being credited benefits from replacing a product on the market.

As the captured CO_2 emissions are released when the electrofuels are used, these results should be viewed as comparative results. When the CO_2 supply is viewed as a by-product/waste where the environmental burden/cost has already been allocated to a main product [285] the environmental assessments show low climate change effects. However, when DAC is used to supply the carbon the impact on the climate is higher due to the additional electricity needed to capture the CO_2 (recall that also renewable electricity emits GHG over its life cycle) [287]. The model used in Paper C accounts for this by assuming several pathways of carbon into the system, i.e., both biogenic methanol and fossil methanol, and thereby showing the sensitivity of these assumptions. It was, however, found that the pathway of carbon had no direct influence over the closed loop system.

The literature is not consistent regarding the environmental impact from different CO_2 sources. There is a shortage of assessments focusing on the environmental impact from different CO_2 sources and the methodology for the assessments varies, however in the results presented in Figure 18, all pathways which include CO_2 from non-fossil origin in combination with electricity of low carbon intensity show low global warming impact.

There is a need for additional studies assessing the environmental impact of different CO_2 sources for different scenarios.

Paper B concludes that a central concept to the sustainability of electrofuels is the sustainability of the carbon source. The LCI models developed in Papers C and D explicitly model the carbon flows throughout the product's life cycle. This means that carbon dioxide entering the system boundary through a capture process has been considered as negative emissions. This allows for full transparency of the inventory data in the results. From a climate impact assessment perspective, the key method issue is that carbon emitted to the natural environment and carbon captured from the natural environment must be accounted for in the LCI coherently. We know that removal of greenhouse gas emission with a direct effect on GWP in absolute terms will result in less climate impact according to the LCA framework. For a correct assessment of the greenhouse warming potential, we therefore must account for the removal of greenhouse gases from the natural system in some way. Figure 21 shows a generic outline of the carbon life cycle alternatives.

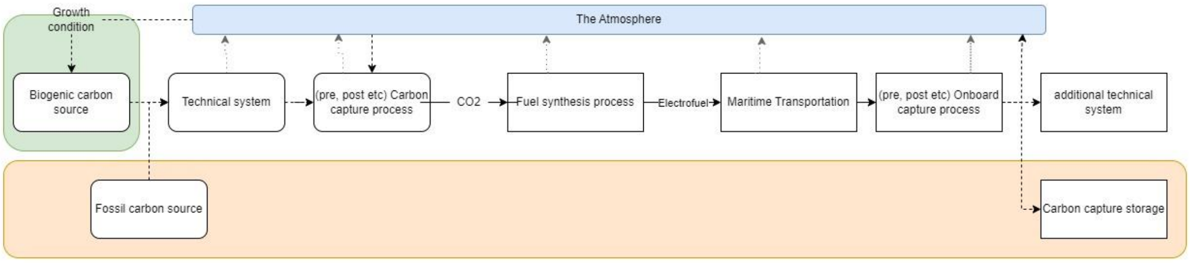

Figure 21 A generic picture of the carbon life cycle for a marine vessel using both carbon-based marine electrofuels and onboard carbon capture. The dashed lines symbolize points where the carbon flow can take different paths. The dotted lines show emissions to natural systems.

Two main ways of modelling GHG flows were identified in the literature. In the first way, established by e.g., the global CO_2 initiative publishing guidelines for techno-economic assessments and life cycle assessment for electrofuels and other forms of CO_2 utilization [333], all emissions and feedstocks were traced upstream in the supply chain and the impact calculated based on the overall sum of flows to and from the electrofuel production processes and the surrounding environment, see for example [80]. By using this method, it is possible to treat all CO_2 flows equally independent of origin. Negative when removed from the atmosphere (e.g., by DAC or biomass growth) and positive when emitted to the atmosphere at the combustion phase. In the second way, only CO_2 flows of fossil origin is considered while all CO_2 from non-fossil origin is treated as having zero climate impact and disregarded, see for example [50]. The choice of carbon capture technology in the fuel production, the assumed system boundary, and how emissions of CO_2 are treated in the calculations explain why some studies present negative impacts from the electrofuel technology (Paper B).

True negative GHG emission results mean that GHG emissions have been removed from the atmosphere through some form of capture process, i.e., DAC or biomass cultivation, and not yet been released back to the atmosphere. To assess if a technology could be able to be considered to have negative climate impact over the full life cycle a list of minimum criteria was set up in Tanzer and Ramirez [186]. The list includes the above-stated goal of physical greenhouse gas emissions to be removed from the atmosphere, as well as a permanent storage sink, including upstream and downstream processes, and that the full life cycle results conclude in a larger removal than additional release. The concept of net carbon emissions is central to the performance of carbon capture technologies, and a life cycle perspective is essential in assessing if a technology leads to actual negative emissions.

The methodological problem of how to differentiate between anthropogenic emissions form fossil fuel extraction and carbon originating in "short term storage" (i.e., decennial rather than millennia) was discussed in the LCA community regarding biogenic emissions. Concluded recommendation in ISO 14044 [334] was that despite carbon emissions technically entering the system boundary when the biomass grows, we assume that the cultivated biomass will regrow and therefore we can negate the carbon entering the system with the carbon exiting the system: all are considered zero and thereby not considered. This approach (here called "the biogenic approach") has led to difficulties when assessing different types of bioproducts, for example the carbon uptake is dependent on soil quality, crop rotation, atmospheric qualities, and therefore is not the same even after a full growth cycle. The shift over time in land use and indirect effects on land use change has also further complicated the calculation. Wiloso et al. [335] showed that treating all input-output flows in the same way (including biogenic carbon as well as fossil carbon as it travels across the system boundary) will in many cases provide different results compared to the same assessment excluding biogenic carbon, thus clearly challenging the assumption that biogenic emissions can be disregarded. Also for bioenergy this neutrality assumption of biogenic carbon has been challenges and several authors argue for a complete LCA inventory including also biogenic CO_2 [335]. However, the neutrality assumption and exclusion of carbon flow is still very common in LCA this is for example shown in a recent review of bio-based platform chemicals [336]. Variations in GWP results for biomass-based products depends on how the product's carbon life cycle is viewed, and different frameworks have been confirmed to lead to considerable variations in the results [337]. The biogenic approach also limits data transparency, as the carbon balance is often not accounted for as it is set to zero.

For conducting a LCI model of the electrofuel technical system where the flows of carbon to and from the natural environment is accounted for, we therefore should expand the system boundaries to include the upstream technical system and account for the flows to and from the environment as they happen (on a reasonable time scale). This would mean to move away from the biomass consensus and account for the "capture carbon" in the cultivation process and treat this and direct air capture the same.

The discussion on how to consider CO_2 in LCA also highlights how electrofuels and carbon capture interacts with the surrounding technical and natural systems – will their use lead to lower GWP emission? The carbon source has a direct influence on the climate change impact of electrofuels in LCAs (Paper B), and system expansion is preferred. A generalization of the possible characteristics of different pathways as presented in Table 10.

Table 10 The relationship between carbon sources and the technologies assessed in this thesis.

CO_2 source		Electrofuels	Onboard carbon capture combined with carbon storage	Circular onboard carbon capture concepts
Fossil fuels	Coal Oil Natural gas	Continued dependency on fossil resources but can reduce demand, all fossil carbon is released	Continued dependency on fossil resources, but can reduce emission intensity	Continued dependency on fossil resources, but can reduce emission intensity
Fossil carbon	Cement (limestone)	Continued dependency on fossil resources but can reduce demand, all fossil carbon is released	Continued dependency on fossil resources,	Continued dependency on fossil resources, but can reduce emission intensity
Biobased	Cultivated biomass	Circular economy if the biomass is regenerated	Negative GHG emissions if the biomass is regenerated	Circular economy, if the biomass is regenerated
	Biogas upgrade Biomass waste streams, for example pulp and paper industry Biomass not competing with food production	Circular economy	Negative GHG emissions	Circular economy
Waste flow	"No upstream emissions"	Directly depends on the waste "producer"	Directly depends on the waste "producer"	Directly depends on the waste "producer"
Ambient	Air Water	Circular economy	Negative GHG emissions	Circular economy

8 FUTURE MARINE FUELS AND PROPULSION SYSTEMS

The decision to use or not to use carbon-based marine electrofuels depends on their feasibility, cost, environmental performance, and other factors, but it also depends on which alternative options that are available. The future possible marine fuels and propulsion systems go beyond those assessed in this thesis. Fuel cells are discussed as an option to ICEs, energy carriers without carbon (i.e., primarily hydrogen and ammonia) are proposed instead of carbon-based fuels, direct use of electricity through shore power and battery electric propulsion is promoted, advanced biofuels are discussed, and energy efficiency measures are crucial in vessel design (Figure 11). The environmental performance of the maritime shipping fleet at large will depend on how all these possible measures are considered, as well as the total performed transport work. The further discussion of the results of this thesis will however be focused on other potential fuel options.

Carbon-based electrofuels of "diesel like" character, i.e., Fischer-Tropsch diesel etc., could be interesting marine fuel options as they could act as drop-in fuels to the current shipping fleet [338]. However, this thesis has shown that the energy efficiency over the life cycle drives the environmental impacts of carbon-based electrofuels (by increasing the demand for electricity and resources) and the energy conversion losses over the life cycle are higher for diesel-like fuels than for methane and methanol.

8.1 Hydrogen and ammonia

Two fuels gaining interest in the maritime transport community are hydrogen and ammonia. As the primary motivation for a fuel shift currently is climate change impacts, a discussion on how these carbon-free fuels relate to the climate change impact is warranted. There are some prospective LCAs available which investigating the environmental performance of renewable hydrogen and ammonia [156, 339], indicating that they could have a positive climate change impact under positive technical development conditions. Both fuels are at earlier technological development stages than methane and methanol, with few ships currently travelling on either fuel. Onboard emission measurements are therefore not available, and the emission estimates are not based on observational data from ships operating in real-world conditions. However, hydrogen and ammonia have relatively well-known production pathways (Paper B).

Model assessments have also shown that hydrogen has a high GWP with an estimated indirect GWP100 of 11 ± 5 [340]. This thesis discussed how the leakage amounts for the gaseous fuel methane is a concern in Chapter 6.2, and the hydrogen leakage potential through the life cycle is at similar levels (1-2%) [339] or higher [339, 341]. The climate mitigation impact of hydrogen is therefore potentially higher than for the electromethanol assessed in this thesis. The indirect climate change impacts of hydrogen have not to this authors knowledge yet been included in assessment of hydrogen as a marine fuel. Efficient distribution of hydrogen and larger volumetric onboard storage requirements are further concerns, whereas the compatibility with fuel cell propulsion speaks for increased utilization of hydrogen as a marine fuel.

Two potential show-stoppers for ammonia are: N_2O emissions generated from the combustion in ICEs and increased emissions of nitrogen to the natural system [342]. The amount of N_2O emissions is not yet fully identified, but as N_2O has a GWP100 of 273 g CO_2 eq./kg even small amounts emitted will have a significant climate change impact. Trade-offs between engine emissions of N_2O, NO and ammonia slip have been identified [343]. Ammonia contains nitrogen, and the amount of nitrogen possible to emit to the natural systems while staying within planetary boundary for the human disturbance to the nitrogen cycle has already been far exceeded [342]. Use of ammonia on a large scale in maritime transport could therefore amplify this trend. Consequently, it is imperative to assess the potential impact of such a shift and explore strategies to mitigate it effectively. Leakage of Ammonia throughout the life cycle is expected to be limited without environmental regulation, as ammonia is highly toxic for humans.

8.2 Cost, technical feasibility, and market preferences

The future use of carbon-based marine electrofuels will depend on their environmental performance, but also technical feasibility, cost, societal preferences, historical patterns, legislation etc. (Paper A). The technical feasibility of carbon-based electrofuels is confirmed, with production sites being in operation and vessels operating on methane and methanol, whereas hydrogen and ammonia as marine fuels are still being developed. Neither electromethane nor electromethanol has direct safety concerns (Paper A), in contrast to for example hydrogen and ammonia [344].

Paper A concludes higher fuel cost to only be one of several barriers for the maritime cargo stakeholders, and states that fair distribution of increased costs between stakeholders is considered more important than mitigating the cost. However, economic sustainability is crucial for a technology's successful implementation among options. The production cost of electrofuels will remain higher for electrofuel than fossil fuels (Paper B), with potential long-term production costs of 90-160 €/MWh, compared to 20-70 €/MWh for fossil fuels assuming oil prices in the range of 30-100 USD/barrel. Cost of electrolyzers and electricity are dominant factors, with the lowest production cost found in regions with good conditions for renewable electricity. As renewable electricity is a primary driver of environmental impacts, low production costs could be coupled with production of low-emission electrofuels. The fuel supply pathway links not only different production pathways with different emission patterns, but also the shipping sectors fuel consumption with other economic sectors energy usage as they compete for feedstocks and fuels.

Using surplus electricity for electrofuel production, which is coupled to low electricity prices, has been proposed as a potential strategy to create cost-competitive production. However, in Paper B as well as in other recent studies [338, 345, 346] it is shown that there are several uncertainties connected to that strategy. For example, it is uncertain if there will be any surplus electricity in future or if electricity users will adapt to varying electricity prices and with different demand side management strategies can even out the price variations. Also, low-cost electricity and low emission electricity are currently not sufficiently coupled on some markets. Furthermore, the relatively high investment cost for electrolyzers indicates that they need to be in operation with a higher capacity factor than what surplus electricity can offer. In Paper B, the lowest production costs generally appear in the interval 45-65% capacity factors, however it should be noted that the research design of Paper B does not consider that the lifetime of electrolyzers may depend on the capacity factor, leading to less strong results. One insight is, thus, that there are still several uncertainties around if producing electrofuels using surplus electricity are a cost-competitive strategy or not.

8.2.1 Carbon capture utilization or carbon capture and storage?

The differences in environmental impact if carbon capture utilization is used instead of carbon capture and storage has not been investigated in the appended papers. Results from the Global Energy Transition (GET) model [227] show how cost optimization seems to lead to carbon capture and storage being preferred over carbon-based electrofuels in the transport sector also under stringent climate targets. This is if large scale carbon capture and storage will be used in the future, which is depended of several factors[347], for example its feasibility and if it will be accepted by the public [348, 349].

The question of immediate feasibility under Nordic conditions has largely been put to rest with the start of Norway's large-scale carbon capture and storage project "Longship". However, after several delays, the final permanent CO_2 storage facility (called Northern Lights) is not yet in operation and full verification of leakages over time from the permanent storage has for example not been verified [350]. According to the National Energy Technology Laboratory [351], globally nine carbon storage site projects (not including enhanced oil recovery sites) are currently active or have been active as of January 2023.

The public acceptance of CCS [352] is less known, and a lack of public acceptance has historically been proven to be a key aspect for onshore carbon capture projects [347]. Culture influences the public perception of the risks and benefits of CCS [353], as well as familiarity with the technology, trust, and socio-economic parameters [352]. Some studies suggest the Nordic public perception to be more positive to CCS than in other regions [354]. A 2016 study of 1830 German citizens [355]concluded that the public perception in Germany on CCS was strongly related to the view of the respective CO_2 source. Literature on CCS has been focused on keeping fossil fuels within the economy [356], but bioenergy combined with carbon capture and storage (BECCS) and other combinations with biocarbon emissions are discussed. This thesis provides a theoretical outline of how different combinations of carbon sources, carbon capture technologies, and storage can be assessed from a life cycle perspective. As long as the future feasibility of large-scale CCS is highly uncertain, there is likely a use case for CCU, and thus an interest in producing carbon-based electrofuels. An early review of CCS and CCU technologies by Cuéllar-Franca and Azapagic [235] indicated that CCS has lower GWP but higher impact on other environmental impact categories.

8.2.2 Regulatory considerations

This thesis has highlighted that carbon-based electrofuels has environmental impacts of relevant scales which should be considered in decision-making processes both at shipping companies and cargo owners as well as in regulation. The influence of emissions from background systems on the overall impact is significant, and regulation must therefore create conditions which secure sustainable production practices. How the IMO regulations and EU legislations signed this year will play out in the maritime landscape is not yet known. However, the regulations and legislation do not rule out the use of electrofuels. The EU fuelMaritime directive goes as far as to promote the use (see Background) and the IMO *Guidelines on life cycle GHG intensity of marine fuels* [220] includes assessment pathways for electrofuels. The guideline includes emission factors calculated using the LCI model presented in Paper D but concludes that the numbers presented varies too much between researchers for default emissions factors to be adapted at this stage. However, the application of values and system boundaries differ depending on the perceived goal of the change from moving to a new fuel and it is thereby likely not a result of discrepancies in the scientific discussion.

The results of this thesis suggest that onboard carbon capture should be handled differently for fossil and renewable fuels. No significant shift occurs between environmental impacts related to fuel production, and bunkered fuel could therefore be treated the same as for vessels without carbon capture systems. The greenhouse gas mitigation of using onboard carbon capture which does not change the combustion characteristics will be the amount of CO_2 captured minus the emissions required for carbon capture and storage, as long as increased energy use onboard is considered. For a system like HyMethShip where hydrogen propulsion is utilized, differences in engine emissions factors must also be considered. The regulatory considerations have the potential to determine what future marine fuels and propulsion systems will be used.

9 METHOD DISCUSSION

The relationship between humans and the natural environment, as well as the decision-making that leads to a marine fuel choice, have been shown to be complex, maybe even wicked. Understanding complex phenomena requires approaching the problem from various perspectives. The research included in this thesis has been conducted at the interface of society and science, and the combined approach of reflective thematic analysis and life cycle assessment has been chosen to fit the explorative nature of the RQs (Table 6). The research design aims to collect new evidence to improve the scientific understanding of maritime fuel choices and has employed quantitative and qualitative data to do so. For both quantitative and qualitative research, it is important to consider the quality of the data analysis to gain meaningful results [357].

This thesis acts within the scope of transition research and directly and indirectly is based on future scenarios. To answer the research questions in this thesis, assumptions on a world where technology is used (regardless of scale) have been made and it is important to acknowledge that such a world is different from today's reality. The pragmatic approach taken in this thesis stipulates research where future scenarios are primarily considered through uncertainty, relationships between parameters, and by describing the context of the conducted studies. Since the environmental performance of carbon-based marine electrofuels (and electrofuels at large) are modeled at a future point in time, they are dependent on future scenarios [111]. The different ways the world could develop (different future scenarios) create the solution space for how the environmental impact might vary [110]. Thus, if an LCA study investigates a future point in time, it uses scenarios (either explicit or implicit) and thereby shows a part of the solution space, also referred to as the "spread of the scenario funnel" [110]. However, capturing too large part of the uncertainty spread can lead to the results becoming difficult for the reader to interpret. The results of this thesis show the connections between different parameters of the model/s and the results to create an understanding of how the results would shift in different plausible futures.

van Leeuwen and Monio [358] argue scholars and policy makers should work on a broader spectra and not only evaluate singular technologies' relationship with today's conventional fossil fuels. As it is known that the fossil option must be phased out, we must evaluate scenarios which are clearly aimed at reaching the goal of zero emissions by 2050. Paper D aims at closing this knowledge gap and showing how emissions relate to the overall goal of expanding the use of renewable methane in maritime cargo transport. However, for a transition of the entire industry, refitting or rebuilding more than 50,000 maritime cargo vessels would be required. Such adjustment of the entire industry will have environmental impacts outside the scope of Paper D.

The discussions on when and if abandoning fossil fuels and if it is feasible to carry out a transition the entire maritime sector is complex in the scientific discussions. Extrapolating life cycle data with a specific goal and scope to assess the climate impact from a full energy transition using singular fuels or fuel combinations are common in the literature, for example [229, 359-361] or assessments performed in the IMO reports [4, 14, 219]. However, they do not consider second or third feedback loops consequences of changing the energy sector, nor limitations to fuel supply. These methods are valid within the current socio-technical system but not if fuels are creating radically different systems. Paper D explores these connections to some extent by coupling LCA with energy system modeling. Hasselman and Erickson [362] discuss this dilemma in an North American context, highlighting the problem with assuming the wrong benchmark system and considering faulty consequential substitution (in their analysis referred to as the "no-action scenario" or "business-as-usual" scenario). The identified problem was that the conventional fuel production pathway was used as a baseline for "avoided emissions" and as such the fossil fuel assessed showed only its comparative performance and not its actual impacts. The argument for this approach was found to be both that this production is what is substituted on the market (an argument also brought forward for electrofuels) and that the fossil fuel would be used any ways so the comparison should be limited to this as a benchmark case. I suggest separating the LCI and LCIA from benchmark systems by avoiding crediting co-products and clearly separating the result with and without crediting when this cannot be avoided. Harjanne and Korhonen [202] conclude that the environmental impact assessments should not be performed in comparison with the current status quo, but instead by a more attributional approach (as used in Paper C) or by the technology's potential in relation to the desired future.

The LCA methodological classification of Papers C and D is not absolute, as ex-ante LCAs are continuously developed. Both Papers show aspects of prospective LCA but could also be classified as both attributional and consequential depending on the viewpoint of the researcher. However, my pragmatic stand is that the exact classification of the methodology is of lesser importance, as the research applicability to reality and to inform decision-making is of larger interest for the aim of the thesis.

Characterization factors are quantitative estimates of the impacts derived from emissions. The choice of characterization methods applied in this thesis has been outlined in the background and methodology chapters, but some potentially interesting impacts are not captured in the results in the appended papers. For example, [363] shows how the estimates of GWP presented in the sixth IPCC report might underestimate the cooling affects from aerosols emissions in shipping lanes. I recommend to always provide the data for the emissions of specific greenhouse gases as a complement to their combined global warming potential. This makes it possible to update the global warming potential when better data is available or to test the impact connected to different climate metrics.

The work in this thesis includes development of LCI both aimed at LCA practitioners and maritime stakeholders. Onboard measurements of ship emissions are challenging and limited by technical feasibility, and monitoring of environmental emissions is limited by costs and resources. Meeting the data requirement for conducting a full life cycle assessment which includes all emissions to and from the environment is not yet feasible. In part due to the large amount of information to be collected, and in part to knowledge not yet being generated. However, the LCA allows for comparison between the emissions from different technologies and to account for how changes in one part of the life cycle will affect emissions levels overall, while using limited data. The conclusions presented in this thesis are, thus, sensitive to changes in assumptions.

The technical data used within this thesis was gathered through literature review and in collaboration with technical experts. The selection of participants [18]can influence the quality of the research. Only Paper A is directly reliant on information from participants, but also the life cycle data inventory work performed in Paper C includes participants in the form of technology experts. The sampling of participants followed a purposive sampling approach [364] to limit selection bias. However, a consideration for future research in similar studies would be to include more questions about the participants history, experience, interest, and familiarization with sustainability, It is possible that the participants attitude in general towards sustainability influence their acceptance of introduction of countermeasures.

This thesis used reflective thematic analysis to analyze the qualitative data (Table 6), and followed the six step process defined by Braun and Clarke [253] (Familiarization of data, developing initial codes, searching for themes, reviewing themes, defining and naming themes, and writing the analysis). The review of the themes was performed systematically in accordance with the criteria set by Byrne [365]. The coherency of the themes was also reviewed by researchers external to the project. This systematic approach was used to validate that the identified themes could be interpreted from the data and reflected the important themes within the data set. The qualitative analysis was considered complete when thematic saturation[19] was met and the code saturation was continuously below 4%[366]. Qualitative research does not necessarily aim to achieve replicability, instead the focus is on transferability of the results [367] which is only possible when the description of the context is rich enough [368]. The case study approach to gather qualitative data was taken to increase the transferability of the results, as this further describes the research context [248].

9.1 The spatial distribution within electrofuel assessment

At first glance, low emission fuels may seem to be fuels with the least quantity of emissions emitted from their use. For emissions assessed with linear impact models and equal characterization value this would be the case; the emissions released to the air, water, or soil equal the pressure on the environment. However, where, when, and how an emission occurs are also highly relevant to the impact characterization. A low-emission fuel therefore becomes a fuel with emissions which are deemed through the impact assessment process to have relatively low impact. The quantification of the emissions is essential, but also their characterization.

The choice of the impacts and what is included in the respective impact, has both inherit values and explicit values, but there is rarely an ethical discussion around the implications. Human health is for instance often included when choosing environmental impacts to analyze in an LCA, whereas biodiversity and species extinction is not. Intra-generational and inter-generational justice are both reflected in the choices of impact

[18] Both technical experts and interview respondents
[19] No new themes were identified when additional interviews were analyzed

categories [369]. As many LCA researchers try to present aggerated results, the geographical distribution of emissions is not presented separately. Instead, all impacts of one type of emissions are aggregated into the same number. This is not a major issue for substances with a global impact, such as greenhouse gas emission's effect on climate change, but Sulphur emissions will affect acidification differently depending on the local environment where they are released and where these emissions occur will also affect to what extent people are affected. Inter-generational justice becomes part of the LCA discourse when we talk about how long-time span into the future that is considered.

If we look at the electrofuel cradle-to-gate system, assuming the spatial use distribution remains the same and that the distribution is environmentally beneficial, the environmental impacts primarily affected by spatial distribution are (see Chapter 2): acidification, eutrophication, particulate matter, land use, and water use. Two of these categories (eutrophication and land use) have minor impacts related to the fuel production processes (identified in Papers C and D) and are instead primarily impacted by energy production Bulle et al. [132] investigated the spatial distribution's influence on toxicity emissions and concluded that, despite exposure being a central part of the cause-effect chain for toxicity, the maximum spatial variability between continents is significantly lower than the total variability in the assessment. The results of this thesis and Bulle et al. [132] thereby indicate that spatial distribution of toxic emission might therefore not be a primary concern for the LCA results. Instead efforts should be put to increase information about the chemical composition and the exact quantities of toxic emissions [132].

The results of Papers C and D primarily have a Swedish context and with use-patterns that remains constant. The result from the papers therefore stands, even when regarding the spatial aspects. However, the impact characterization pathways are uncertain in absolute terms [197] and further research is required to assess the impacts if the marine fuels are used in specific regions, such as the Baltic Sea.

10 CONCLUSION

The work presented in this thesis is aimed at investigating the conditions under which carbon-capture could have a role to play in an environmentally sustainable maritime sector, both when applied directly onboard and when utilized to produce carbon-based marine electrofuels. The thesis provides emission, resource, and energy data on processes necessary to produce carbon-based electrofuels. More specifically the following conclusion can be drawn from the research made:

10.1 How do stakeholders on the maritime cargo market view their possibilities to choose a low-emission fuel today?

Analysis of Swedish maritime cargo sector shows a stakeholder landscape that, despite being of various types, has a coherent viewpoint on the lack of possibilities to choose a low-emission marine fuel today. Further adoption of low-emission fuels in Sweden is hindered by fuel cost concerns, information and communications gaps, diverse goals, and uncertainty. Despite these challenges, the stakeholders express a collective desire to transform the industry.

The higher fuel price keeps stakeholders from moving directly to low-emission fuels today. However, the increased cost can be overcome when environmental performance requirements (on for example emission levels) are put on the transportation service. The risk of not having access to fuel in port is viewed as more urgent as the ships cannot operate if no fuel is available. All the identified barriers are interlinked and interdependent, where quantitative assessments can only capture some of the aspects slowing down adoption. Electrofuels, produced using sustainable feedstocks, could address one of these concerns by securing future fuel supply but will likely increase fuel costs.

The stakeholders view introduction of efficient and fair regulation as positive, and stronger external drivers as necessary for low-emission marine fuel adoption. The introduction of new legislation will, therefore, likely increase the stakeholders' incentives to choose a low-emission fuel in the future. However, increased collaboration and more contextual information on environmental impacts will also contribute positively to the shift. By exploring the barriers and potential solutions, this work concludes that fuel price might not be the largest barrier to fuel transition, instead accessibility to low-emission fuel is considered significant.

10.2 How can marine fuel life cycle assessment be further developed to include carbon-based electrofuels?

Simplified well-to-wake assessment which only considers climate change impact is insufficient to accurately assess environmental impacts of marine fuels. LCA is a good tool which have considers a broad range of impact categories, however, the focus of published LCAs on marine fuels has been on GHG emissions and other emissions to air. Still research is moving forward with more comprehensive assessments, including, for example, how emissions to seawater impact the environment. Specifically, the following five areas must be developed in forthcoming marine fuel LCAs to better understand the environmental impacts of future marine fuels:

- Impact from material demands are no longer negligible, and capital goods must therefore be included in the system boundary.
- Toxicity impact assessment must be further improved, for human health impacts to be sufficiently assessed.
- The inventory should include metal emissions from the assessed technical systems should be included (preferably as primary emissions)
- Emissions to sea water should be considered, as the pressure on the marine environment increases due to emission from the fuel life cycle.
- Land use, as this impact category can be significant for both biofuels and electrofuels.

The results of this thesis clearly show how the hot spots for environmental impacts shift phases in the life cycle between LCAs of fossil marine fuels and marine electrofuels. The primary emission source shifts from fuel combustion to fuel production leading to impacts from background data becoming more critical. Traditional life cycle assessment therefore must be developed to include the background systems in more detail.

Papers C and D clearly show the dependency on background data, and while uncertainties remain, Paper D limits the uncertainty of which background system to assume by applying scenario based LCA modelling. As opposed to regional issues, such as water scarcity and eutrophication, which must be assessed at a more granular level. To conclude, the results show that LCA can be used to address questions related to the life cycle environmental performance of future marine fuels, but on its own its ability to support decision-makers is currently limited and traditional life cycle assessment tools would benefit from being developed according to the bullet list above. Maritime research should prioritize comprehensive environmental assessments over analysis against regulatory requirements.

10.3 What are the environmental impacts of carbon-based marine electrofuels: Under which conditions can they be defined as low-emission fuels?

This thesis shows that two carbon-based marine electrofuels, (i) electromethanol and (ii) electromethane, can be defined as low-emission fuels compared to fossil fuels under certain production conditions. First, the electricity and carbon inputs in fuel production must come from low-emissions sources, where, e.g., current Swedish electricity grid mix and biogenic carbon sources are sufficient conditions to achieve low GHG emissions. Secondly, fugitive emissions of methane and methanol must be limited throughout the life cycle. Thirdly, land use, water scarcity, and human toxicity impacts should not be adversely affected. At a system level, the environmental performance of carbon-based marine electrofuels will depend on the development of electricity production in the energy sector and access to sustainable carbon sources.

As climate impact, toxicity, land use and acidification of electrofuels are driven primarily by the energy source, a low-emission (low environmental impact) energy system becomes essential for the fuel to be viewed as low-emission. The amount of electricity required for electrofuels, and, as such, the materials required per kWh of energy eventually used onboard the vessel, becomes influential for the overall consideration of these impact categories. The system energy efficiency of carbon-based electrofuels is low compared to fossil fuel production pathways or direct electricity. Therefore, emissions that can be considered small per kWh of electricity get accumulated because of the system losses and become significant over the fuel's life cycle.

Further investigations are required to establish if future carbon-based marine electrofuels are low-emission fuels. Reduction targets, feasibility of other fuel options, technology development, and which environmental concerns that are of primary interest will affect the future landscape of marine fuels. However, the results show that electromethanol could potentially be a low-emission fuel also under stringent demands on climate performance, whereas the mitigation potential for electromethane is limited by emissions of methane throughout the life cycle and will not lead to more than an 80% reduction in GHG emissions compared to MGO.

10.4 Under which conditions could onboard carbon capture mitigate environmental impacts?

This thesis provides insights into carbon capture's potential role in maritime transport. The applied life cycle assessment shows that if a pre-combustion carbon capture system with high efficiency (>90% capture rate, <5% additional energy demand) is combined with electromethanol production close to the port, the system could reach low impacts in several impact categories (70-97% compared to MGO). The combustion characteristics onboard the vessel directly influence the acidification and eutrophication impacts, and limiting emissions of NOx is key to achieving impact reductions in these categories.

Onboard carbon capture must be combined with CCU or CCS for a reduction in global warming potential to occur. The carbon capture rate of the onboard carbon capture system is a central factor in realizing a reduction in climate change impacts. Lower system efficiency leads to higher impacts on all criteria. The onboard carbon capture systems' environmental performance generally shows mitigation of GHG emissions if the losses of carbon throughout the system remain low and the additional energy required to reach the reduction target is not significant for the overall system performance.

10.5 How can CO_2 be modeled in life cycle assessment of carbon capture and utilization (CCU) technologies?

The circular nature of CCU requires a structured approach to modeling the carbon life cycle. Papers C and D show that it is possible to conduct a full LCA of carbon-based marine electrofuels while accounting for carbon flows throughout the life cycle. CO_2 should be clearly accounted for in the life cycle inventory, regardless of if it is of fossil origin or not. From a climate impact standpoint, the main data which needs to be calculated through the life cycle inventory is the amount of GHG which is emitted from the technical system. To present well-to-tank results and tank-to-wake results separately is not compatible with the LCA approach taken in this thesis, where carbon dioxide is modelled as entering the system boundary when "captured" from the air by either carbon capture or cultivated crops and therefore show negative emissions in the initial step.

Use of fossil CO_2 only delays the release of fossil CO_2 to the atmosphere and lowers the emission intensity of the products. Systems dependent on fossil CO_2 will remain dependent. For carbon to be captured and not contribute to climate change requires that carbon stocks remain sequestered in the Technosphere on decadal or centennial timescales. The result is nonetheless that fossil CO_2 and non-fossil sources of CO_2 must be viewed in different ways. CO_2 should be considered negative when removed from the atmosphere and positive when released into the atmosphere. If the carbon is captured from another technical system, it is not removed from the atmosphere.

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