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A prospective Life Cycle Assessment

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NOVEL BROWN SEAWEED-BASED BIOPLASTIC

A PROSPECTIVE LIFE CYCLE ASSESSMENT

**BY
MADDALEN AYALA CEREZO**

DISSERTATION SUBMITTED 2023



AALBORG UNIVERSITY
DENMARK

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by

Maddalen Ayala Cerezo



AALBORG UNIVERSITY
DENMARK

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CV

Maddalen completed her Bachelor's degree in Environmental Science at the University of the Basque Country in 2015, gaining a holistic knowledge in various scientific disciplines related to the environment. Seeking to further her expertise and practical skills, she pursued a Master's degree in Environmental Management and Sustainability Science at Aalborg University from 2016 to 2018, where she focused on circular economy. She has international experience, both in academia and the private sector, in Germany, the Netherlands, Portugal, Spain and Denmark. Prior to the PhD she worked as a project manager. During this time, she developed a keen interest in LCA. In 2020 she started her PhD journey LCA for the blue bioeconomy.

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ENGLISH SUMMARY

The PhD thesis focuses on assessing the environmental impacts of bioplastic production from brown seaweed within the context of the emerging field of Blue Bioeconomy as a consequence of an increasing interest in biomaterials. The research is conducted in parallel with the PlastiSea project, which aims to develop bioplastics using alginate extracted from brown seaweed. The main objective of the PhD is to evaluate the environmental consequences of producing bioplastics from brown seaweed through a consequential Life Cycle Assessment (LCA) perspective.

The thesis is a compilation of scientific articles and the research is structured into four parts, each addressing a specific aspect of the study and corresponding to an article. The first part involves identifying marginal suppliers of brown seaweed, analysing the supply chain and determining key suppliers in the market. The second part focuses on evaluating the environmental impacts of the pilot-scale seaweed-based bioplastic production system and the potential of re-circulating seaweed co-products and different end-of-life pathways, while the third part examines the impacts of an industrial-scale system. The fourth part takes a broader perspective by considering seaweed-based bioplastic production from a supply chain standpoint.

The thesis presents relevant results in different areas. In terms of marginal suppliers, China emerged as the primary supplier and is projected to maintain its significance. Additionally, suppliers from northern Europe and America are anticipated to witness a moderate increase in their market share, but in smaller proportions.

The LCA at the pilot scale identifies the potential for recovering mannitol from seaweed, which could be utilized in the subsequent seaweed-based bioplastic production step. This approach exhibits a slight advantage compared to the base scenario; however, the observed difference is not deemed significant. The assessment of the end-of-life scenarios shows a significant advantage of composting end-of-life compared to incineration.

Moreover, the prospective LCA, which involves upscaling the processes to larger scales, reveals a noteworthy trend of decreasing environmental impacts. As the processes increase in volume, the impacts associated with their operation lower, emphasizing the potential benefits of scaling up production in terms of reducing environmental burdens. The inclusion or exclusion of biogenic carbon shows a substantial impact on the results, showcasing the importance of accounting for this factor in the assessment. The disparities in the outcomes highlighted the significance of considering the biogenic carbon component in LCA analyses.

The findings of the PhD reveal promising potential for brown seaweed-based bioplastics, particularly in terms of greenhouse gas emissions, which show lower impacts compared to conventional fossil-based plastics at larger scales. Moreover, seaweed-based bioplastic exhibits significant advantages in other impact categories, such as marine and freshwater eutrophication. The results collectively also provide valuable insights into the dynamics of marginal suppliers, the potential recovery of seaweed co-products and the assessment of different end-of-life pathways, the influence of biogenic carbon accounting, and the environmental implications of upscaling processes. However, some constraints need to be considered, including competition with other uses of the seaweed biomass and the specific properties and limited applications of the seaweed-based bioplastic, such as its high moisture absorption.

In conclusion, the thesis highlights the challenges and opportunities associated with the production of bioplastics from brown seaweed. It emphasizes the need for further research to address the identified constraints and explore potential solutions. The discussion and conclusion provide a comprehensive overview of the research outcomes, implications, and future directions for the development and utilization of brown seaweed-based bioplastics from an environmental perspective.

DANSK RESUME

Denne ph.d.-afhandling omhandler en vurdering af miljøpåvirkningerne ved produktion af bioplastik fra brunalger inden for rammerne af den fremvoksende industri, blå bioøkonomi som en konsekvens af stigende interesse for biomaterialer. Forskningen udføres parallelt med PlastiSea-projektet, der sigter mod at udvikle bioplastik ved hjælp af alginat udvundet fra brunalger. Hovedformålet med ph.d.'en er at evaluere de miljømæssige konsekvenser ved produktion af bioplastik fra brunalger gennem perspektivet af en konsekvent livscyklusvurdering (LCA).

Afhandlingen er en samling af videnskabelige artikler, og forskningen er struktureret i fire dele, der hver beskæftiger sig med en specifik del af studiet og svarer til en artikel. Den første del omhandler identifikation af marginale leverandører af brunalger, analyse af forsyningskæden samt identifikation af nøgleleverandører på markedet. Den anden del fokuserer på vurdering af de miljømæssige konsekvenser ved pilot-skala produktionssystemet for bioplastik baseret på brunalger og potentialet for genanvendelse af brunalge-bi-produkter og forskellige metoder til *end-of-life*, mens den tredje del undersøger konsekvenserne af et industriel-skala system. Den fjerde del tager et bredere perspektiv ved at betragte produktionen af bioplastik baseret på brunalger ud fra et forsyningskædeperspektiv.

Afhandlingen præsenterer relevante resultater inden for forskellige områder. Med hensyn til marginale leverandører fremstod Kina som den dominerende leverandør og forventes at fastholde sin position. Derudover forventedes leverandører fra Nordeuropa og Amerika at opleve en moderat stigning i deres markedsandel, men i mindre omfang.

LCA'en på pilot-skala projektet identificerede potentialet for at udvinde mannitol fra brunalger, som kunne anvendes i den efterfølgende produktion af bioplastik baseret på brunalger. Denne tilgang viste en lille fordel i forhold til basisscenariet; dog blev den observerede forskel ikke anset for signifikant. Vurderingen af scenarierne for produktets levetid viste en betydelig fordel ved kompostering i sammenlignet med forbrænding.

Desuden afslørede den fremadrettede LCA, der involverede en opskalering af processerne til større skalaer, en bemærkelsesværdig tendens til mindskede miljøpåvirkninger. Når processerne blev udvidet, reducerede disse påvirkninger, der var forbundet med deres drift, hvilket understreger de potentielle fordele ved opskalering af produktionen i form af formindskede miljøbelastninger. Inkluderingen eller udeladelsen af biogent kulstof havde en væsentlig indvirkning på resultaterne, hvilket viser vigtigheden af at tage højde for denne faktor i vurderingen. Forskellene

i resultaterne understregede betydningen af at overveje komponenten af biogent kulstof i LCA-analyser.

Ph.d.'ens resultater påviser lovende potentiale for bioplastik baseret på brunalger, især med hensyn til drivhusgasemissioner, der fremviser lavere miljøpåvirkninger sammenlignet med konventionelle fossilbaserede plastik produktion på større skalaer. Derudover har bioplastik betydelige fordele inden for andre påvirkningskategorier, såsom eutrofiering af marine- og ferskvandsmiljøer. Resultaterne giver også værdifulde indsigter i dynamikken hos marginale leverandører, potentialet for genanvendelse af brunalge-bi-produkter samt vurderingen af forskellige metoder til *end-of-life*, indflydelsen af med regning af biogent kulstof samt de miljømæssige konsekvenser ved opskalering af processer. Der skal dog tages hensyn til visse begrænsninger, herunder konkurrence med andre anvendelser af brunalgebiomassen og de specifikke egenskaber og begrænsede anvendelsesmuligheder for bioplastik baseret på brunalger, såsom dens høje fugtabsorption.

Afslutningsvis fremhæver afhandlingen udfordringerne samt mulighederne forbundet med produktion af bioplastik fra brunalger. Den understreger behovet for yderligere forskning for at håndtere de identificerede begrænsninger og udforske potentielle løsninger. Diskussionen og konklusionen giver en omfattende oversigt over forskningsresultaterne, implikationerne og fremtidige retninger for udviklingen samt anvendelsen af bioplastik baseret på brunalger set fra et miljømæssigt perspektiv.

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Aalborg, June 2023
Maddalen

PREFACE

The PhD project was financed by the PlastiSea project (Grant no. 9082-00011), funded by ERA-NET Cofund BlueBio program and the Innovation fund. PlastiSea aims to develop novel bioplastic materials based on cultivated and wild species of brown algae. www.sintef.no/en/projects/2020/plastisea-novel-enhanced-bioplastics-from-sustainable-processing-of-seaweed/

The thesis follows the following structure. Chapter 1 introduces the concept of Blue Bioeconomy, the novel seaweed-based bioplastic and prospective LCA. Chapter 2 focuses on the research design, including the research questions and overall goals that the project seeks to achieve. In Chapter 3, the methods used to achieve the research objectives are described in general terms. Chapter 4 presents the results of the research. In Chapter 5 the results from the PhD are critically discussed, together with the strengths, limitations, challenges and opportunities of the seaweed-based bioplastic and its potential in addressing environmental issues. Chapter 6 provides the conclusions, a summary of the findings, answers the research questions and offers some final remarks. The research work is organised into four parts, each of them being reported in one journal article, which are found in Appendix A-D.

PUBLICATIONS

- Ayala, Thomsen, Pizzol (2023). *Using quantitative story telling to identify constraints in resource supply: The case of brown seaweed* (**accepted with minor revisions, June 2023**).
- Ayala, Thomsen, Pizzol (2023). *Life Cycle Assessment of pilot scale production of seaweed-based bioplastic* (**published**).
- Ayala, Goosen, Michalak, Thomsen, Pizzol (202X). *Prospective LCA of brown-seaweed-based bioplastic: Upscaling from pilot to industrial scale* (**submitted June 2023**).
- Ayala, Arlov, Nøklung-Eide, Sæther, Dore, Vidal, Zhou, Michalak, Kyvik, Wang, Jolain, Aubel, Strand Jacobsen, Pizzol (202X). *A supply-chain perspective on producing and upscaling bioplastic from brown seaweed* (**submitted June 2023**).

CONFERENCES

- *Identifying unconstrained suppliers systematically for emerging technologies: the case of brown seaweed production*. SETAC 31st Annual Meeting, May 2021 (**Presentation**).
- *Identifying unconstrained suppliers systematically for emerging technologies: the case of brown seaweed production*. SETAC Europe 32nd Annual Meeting, May 2022 (**Presentation**).
- *Using quantitative storytelling to identify constraints in resource supply: The case of brown seaweed for bio-based plastics*. 11th Nordic Seaweed Conference, October 2022 (**Poster**).
- *Life Cycle Assessment of pilot-scale production of seaweed-based plastic*. 11th Nordic Seaweed Conference, October 2022 (**Presentation**).
- *Upscaling the seaweed-based bioplastic: an LCA perspective*. Seaweed Applications - Opportunities and Challenges, March 2023 (**Presentation**).
- *Prospective LCA of brown-seaweed-based bioplastic: From pilot to industrial scale*. SETAC Europe 33rd Annual Meeting, May 2023 (**Presentation**).

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LIST OF ABBREVIATIONS

BBE Blue bioeconomy

FU Functional unit

EoL End of Life

LCA Life Cycle Assessment

LCI Life Cycle Inventory

LCIA Life Cycle Impact Assessment

PLA Polylactic acid

QST Quantitative story telling

RQ Research question

TRL Technology readiness level

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CHAPTER 1. INTRODUCTION

The introduction provides a brief overview of the blue bioeconomy, highlighting the significance of seaweed within this field. The topic of seaweed-based bioplastics is outlined, which serves as the focal point of the PhD research conducted within the PlastiSea project. Later, the products that have been developed as part of the PlastiSea project are presented, underscoring their formulation and potential applications. Finally, the Life Cycle Assessment is introduced as the primary method employed for assessing the environmental impacts of the seaweed-based bioplastic production process.

1.1 THE BLUE BIOECONOMY

The blue bioeconomy (BBE) refers to the utilization of a wide range of value chain activities of aquatic bioresources, including marine and freshwater environments, for economic growth and development. It involves a variety of sectors such as fisheries, aquaculture, marine biotechnology, coastal tourism, animal feeds, food additives, pharmaceuticals, cosmetics, and new materials. The BBE is gaining momentum in response to the increasing demand for natural resources and the need to mitigate environmental impacts while promoting development (European Commission. Directorate-General for Maritime Affairs and Fisheries., 2018). Given the novelty of many of the activities in the BBE context, it becomes important to assess the environmental performance of these activities, products, and technologies in the early stage (Cucurachi et al., 2022).

1.1.1 SEAWEED AS A FEEDSTOCK IN THE BLUE BIOECONOMY

Seaweed, also known as macroalgae, is a type of marine plant that grows in the ocean and other bodies of seawater. It is characterized by its leaf-like or branching structures and is classified as a type of algae. Seaweed can vary in size, colour, and shape, ranging from small and delicate strands to large and robust forms. It plays a vital role in marine ecosystems, providing habitat and food for numerous aquatic organisms (Mouritsen, 2013). Seaweed is a source of blue carbon and, as a renewable and fast-growing resource with diverse applications, was the focus of interest in the most recent BBE report (EUMOFA, 2022). Blue carbon is defined as the carbon sequestered by living organisms within coastal ecosystems and subsequently stored in biomass and sediments (Macreadie et al., 2021).

There is a wide variety of seaweed species classified into three primary categories: red seaweed (Rhodophyta), green seaweed (Chlorophyta) and brown seaweed (Phaeophyta). Red seaweed, mainly found in tropical waters, is characterized by its complex cellular structure, which includes additional organelles like phycobilisomes

for efficient light absorption, enabling its adaptation to deeper waters. Agar and carrageenan polysaccharides are only present on red seaweed. Green seaweed, on the other hand, exhibits a simpler cellular composition, with chloroplasts responsible for photosynthesis, giving it a distinct green colour. This simpler cellular structure allows green seaweed to grow rapidly and thrive in both marine and freshwater environments. Brown seaweed, including kelp, is commonly found in cold waters and is known for its large size and the presence of unique compounds such as alginates and fucoidans, both versatile polysaccharides (Bhuyar et al., 2021). Brown seaweed, more specifically *Saccharina latissima* and *Alaria esculenta* are the seaweeds used in this project. Figure 1-1: A picture of red (top left), green (top right) and brown (bottom) seaweed. displays the three types of seaweeds mentioned.



Figure 1-1: A picture of red (top left), green (top right) and brown (bottom) seaweed.

Seaweed cultivation is on the rise globally. Worldwide, the production of seaweed in 2019 amounted to 35.7 million tonnes of wet biomass, with brown seaweed accounting for 12.3 million tonnes of this total. The human consumption market serves as the primary target market (Cai et al., 2021).

The interest in the cultivation of brown seaweed started in China in the 1950s, as seaweed was considered a valuable food resource to meet the projected population growth. Similarly, Japan and Korea also started seaweed cultivation, although on a smaller scale than China. Overall, seaweed cultivation in Asia started because of the necessity to support the expected increase in population (Zhang, 2018a). Today China stands out as the largest producer of seaweed supplying around 20.3 million tonnes of seaweed, of which 11 million tonnes are brown seaweed, food being the main target market (FAO, 2021a; Hu et al., 2021). In recent decades, western countries have also recognized the value of brown seaweed for its diverse applications (Araújo et al., 2016).

While the supply of seaweed from wild harvesting is staying constant (FAO, 2021a), cultivated seaweed dominates the market, offering controlled and constant production with higher yields. Notably, aquaculture accounts for 96.5% of global brown seaweed production, with Europe relying more on wild harvesting (Stanley et al., 2019a). However, there is a noticeable shift towards seaweed aquaculture, particularly in Europe, mainly driven by increasing demand for seaweed for different applications (Araújo et al., 2021). The literature also highlights the prospects of expanding seaweed cultivation globally (Ferdouse et al., 2018), in Europe (Araújo et al., 2021; van den Burg et al., 2021a) and especially in Norway (Stévant and Rebours, 2017), reflecting the growing interest and future potential.

Compared to feedstocks derived from arable crops, seaweed cultivation stands out for its advantages as it removes the need for land, fertilizers, pesticides, and freshwater, leading to a reduction in environmental impact (Duarte et al., 2021; Duarte and Krause-Jensen, 2018; Giercksky and Doumeizel, 2020). Additionally, seaweed cultivation sequesters carbon, reduces eutrophication and contributes to nutrient cycling (Seghetta et al., 2016b; Zhang et al., 2022). Moreover, it is a versatile and highly renewable feedstock, with a high variety of applications (Ghadiryfar et al., 2016).

1.2 SEAWEED-BASED BIOPLASTICS

Seaweed-based bioplastic is one of the emerging applications and it is still in an early stage of research and development (Carina et al., 2021a; Lim et al., 2021; Rinaudo, 2014; Shrivya et al., 2021) as a potential solution to reduce fossil-based plastics. Bioplastics are classified into three generations (Jayakumar et al., 2023a) [Click or tap here to enter text.](#), with the third generation being unique in that it does not rely on arable land for its production (Melchor-Martínez et al., 2022).

The seaweed component used to produce the bioplastic is alginate, a polysaccharide derived from brown seaweed. Alginate can form gels with various structures, crosslink with certain ions, and is biocompatible (Song et al., 2022). Alginate is, therefore, uniquely suited for a range of applications including biomedical

applications, the food industry, the cosmetic industry, agriculture, and bioplastics (Zdiri et al., 2022).

The process used to produce the seaweed-based bioplastic within the PlastiSea project consists of three main steps: Seaweed cultivation, biorefinery and film fabrication. The process of seaweed cultivation begins with the hatchery stage, followed by three steps in the biorefinery: acid treatment, wash, and alkaline treatment. The last step, film fabrication, involves crosslinking, compounding, casting, and drying. The system boundaries in all the LCAs conducted throughout the PhD include all the mentioned processes and the End-of-Life (EoL) of the seaweed-based bioplastic. Figure 1-2 is a graphical representation of the value chain to produce the seaweed-based bioplastic.

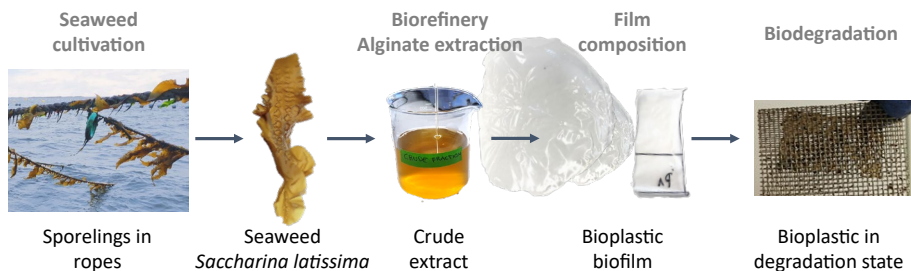


Figure 1-2: The entire value chain to produce seaweed-based bioplastics represented with figures.

Varios seaweed-based bioplastics were developed within the PlastiSea project. An overview of these novel materials is presented, focusing on their formulation, properties, and potential uses respectively.

1.2.1 SEAWEED-BASED BIOPLASTIC FILM

The seaweed-based bioplastic film was the main product developed within the PlatiSea project and its production process comprises three main steps (see Figure 1-3). First, the compounding step involves the thorough mixing of the mentioned components. This is followed by casting and crosslinking, during which CaCl_2 is sprayed onto the material. Lastly, the solution is dried on a flat surface, resulting in the final seaweed-based bioplastic film.

The formulation of the seaweed-based bioplastic used in this film follows a specific ratio of ingredients. It consists of 1 part alginate, 1.25 parts glycerol, and 0.05 parts cellulose, which are mixed together with water. This carefully designed formulation ensures the desired properties and performance of the seaweed-based bioplastic film.

The seaweed-based bioplastic film produced in the PlastiSea project possesses several distinct properties. It is a flexible and transparent/semi-transparent film, visually resembling low-density polyethylene (LDPE) plastic. Additionally, it has partial water solubility and holds the potential for water barrier enhancement. This seaweed-based bioplastic is also biodegradable. The inclusion of glycerol as a plasticizer allows for the adjustment of the seaweed-based bioplastic's mechanical properties by varying the glycerol content. Glycerol works as a plasticizer and varying the glycerol content the mechanical properties of the seaweed-based bioplastic change.

The seaweed-based bioplastic under consideration has a range of potential applications, primarily as a packaging material. It is suitable for use in food packaging, including greasy or dry food items, as well as packaging for fresh fruits and vegetables. Furthermore, it can be employed in packaging goods that require breathability. In addition to these intended uses, the seaweed-based bioplastic film could potentially be used as a packaging solution for the fashion industry, cosmetics or other dry goods.



Two samples of seaweed bioplastic film

Seaweed-based bioplastic roll

Figure 1-3: Seaweed-based bioplastic film.

1.2.2 SEAWEED-BASED BIOPLASTIC PELLETS

The process of manufacturing seaweed-based bioplastic pellets involves the use of an extruder compounder that mixes a combination of alginate, glycerol, and water. This mixture is then processed and transformed into seaweed-based bioplastic filaments using an extruder. The filaments produced in this process are long, continuous strands of seaweed-based bioplastic material, which are eventually cut into pellets. The extruded pellets can be melted and cast into other forms, such as glass or food trays. The cast extrusion machine is where the pellets are introduced and re-melted before being cast (Figure 1-4).

The formulation of the seaweed-based bioplastic pellets involves 70% alginate, 15% glycerol, and 15% water. This specific formulation ensures the desired properties and performance of the pellets keeping a high alginate content. For the food tray application, the formulation is modified to include a 50% mixture of the alginate and glycerol blend, combined with either Mater-Bi or Polybutylene Succinate (PBS). These additional components are bio-based and home-compostable thermoplastics.

The seaweed-based bioplastic formulation with its high alginate content offers potential applications for products that do not require direct contact with water. For instance, it can be utilized in the manufacturing of various items, such as non-water-based products or packaging solutions. In the case of the food tray, the incorporation of 50% Polybutylene Succinate (PBS) enhances its water resistance and durability, making it suitable for packaging food items that may come into contact with moisture.

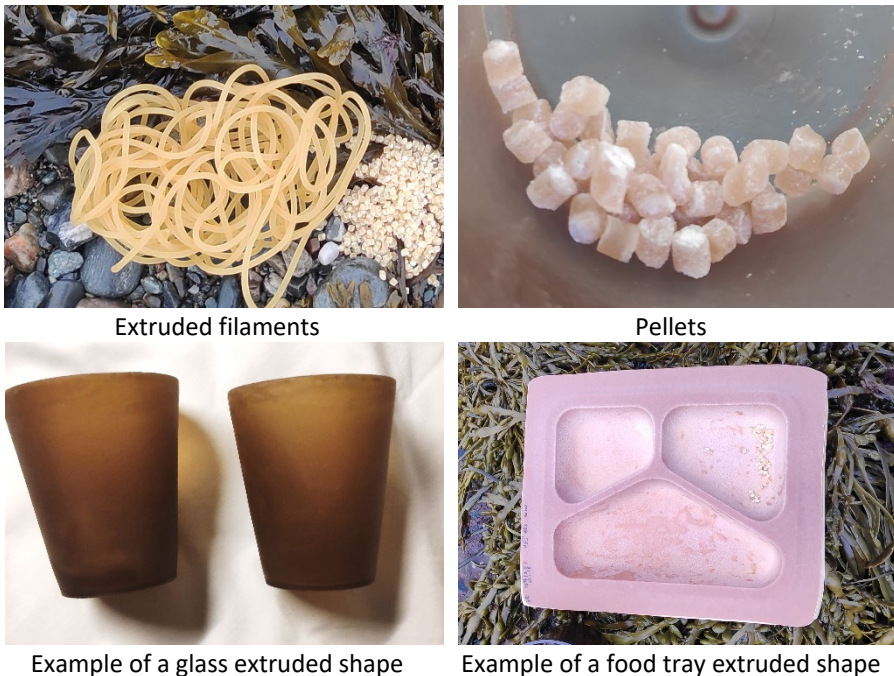


Figure 1-4: Seaweed-based bioplastic pellets and casted pellets into different shapes.

1.2.3 SUBSTITUTION OF PLA

This bioplastic is a co-product derived from the production of the seaweed-based bioplastic film. The seaweed residues obtained from the biorefinery are effectively utilized to replace polylactic acid (PLA) in the formulation. The seaweed-based

bioplastic can be employed in various forms, such as being extruded into desired shapes like a coffee pot or used as a printing filament for 3D printing applications, as demonstrated by the creation of the PlastiSea logo (Figure 1-5).

The formulation of this PLA bioplastic involves seaweed residues, ranging from 5% to 30% of the composition, along with polylactic acid (PLA), which makes up the remaining 95% to 70% of the formulation.

The potential uses of this bioplastic are extensive and include various applications similar to conventional PLA. It can be utilized in the production of items like plant pots, bottles, and biodegradable medical devices. Additionally, it is compatible with 3D printing technology. However, it is important to note that while a conceptual example of a coffee capsule is shown in Figure 1-5, the high temperatures involved in coffee preparation may pose a risk of damaging the bioplastic.

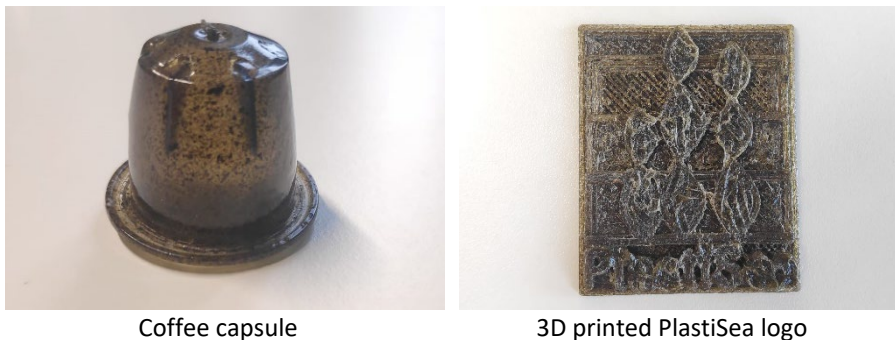


Figure 1-5: Casted PLA (left) and 3D printed PLA (right) with substituted seaweed pellets.

1.3 LIFE CYCLE ASSESSMENT OF SEAWEED PRODUCTS

Life Cycle Assessment (LCA) is a widely recognized method employed to quantify the environmental impacts associated with a process, product or service across its entire life cycle. The LCA is usually conducted according to the ISO 14040:2006 and 14044:2006 standards (ISO, 2006b; ISO, 2006a) and consists of four phases, starting with the establishment of the goal and scope of the assessment. This initial phase defines the boundaries and objectives of the LCA study. The second phase, Life Cycle Inventory (LCI), involves collecting data on material and energy inputs and outputs throughout the life cycle. The third phase, Life Cycle Impact Assessment (LCIA), consists of evaluating the potential environmental impacts of the product or process based on predetermined impact categories and indicators. Lastly, the Interpretation phase involves analysing and interpreting the LCA results to reach conclusions and identify areas for improvement (ISO, 2006b).

Prospective LCA is referred to the LCAs performed at a future time, usually to assess the environmental impacts of emerging technologies at the early stage of technological development (Arvidsson et al., (Euro. The goal of prospective LCA is to identify potential environmental consequences and trade-offs associated with the use of emerging technologies before they are implemented on a bigger scale. It aims to model a system that will exist at a future time and estimates the environmental impacts of that system based on the best currently available data and based on specific assumptions (Cucurachi et al., 2018).

From a prospective LCA point of view, the novel brown seaweed-based bioplastic is considered an emerging technology since it is in the early stage of technological development (Thonemann et al., 2020). There are several uncertainties associated with those emerging technologies given their immaturity. Uncertainties arise from various sources such as limited data availability and unpredictability in technological advancements (Blanco et al., 2020). These uncertainties pose challenges in accurately assessing the environmental performance of emerging technologies (van der Hulst et al., 2020). A comprehensive LCA approach that considers the uncertainties and other constraints associated the emerging brown seaweed-based bioplastic is therefore needed.

There are two main approaches to conducting LCA: attributional LCA and consequential LCA. Attributional LCA focuses on attributing a share of the potential global environmental impacts to a product's life cycle. It enables the assessment of environmental impacts that have occurred during the production of a product. This approach provides valuable insights into the environmental burdens associated with specific processes and activities. On the other hand, consequential LCA examines the environmental consequences of a decision or change in the demand for a product or service (Sonnemann and Vigon, 2011).

The consequential LCA approach is defined as follows by UNEP/SETAC (Sonnemann and Vigon, 2011):

The consequential LCA uses a system modelling approach in which activities in a product system are linked so that activities are included in the product system to the extent that they are expected to change as a consequence of a change in demand for the functional unit.

If the emerging technology is implemented in the future on a large scale, the demand for the new product will increase and the suppliers will need to respond by increasing their production capacity and adapting to meet the growing market needs (Ghose et al., 2017; Schmidt and Weidema, 2008). In the consequential LCA context, those suppliers increasing in the market are known as *marginal suppliers*. A marginal supplier refers to a supplier that provides an additional or incremental quantity of a product or resource to meet the growing demand or changes in market conditions

(Pizzol and Scotti, 2017; Sacchi, 2018; Weidema et al., 1999). Consequential LCA also offers a different way of handling co-products compared to the attributional approach. Instead of using allocation, which is commonly employed in attributional LCA, the study opted for substitution (Heijungs et al., 2021; Weidema et al., 2009).

In this PhD, the consequential LCA approach was chosen to evaluate the environmental consequences and implications of increasing the demand of the novel brown seaweed-based bioplastic. This decision was made considering the technology's immaturity and the need to assess the environmental impacts related to upscaling the production process to an industrial scale. In addition to that, the consequential LCA requires the identification of marginal suppliers, which was considered a more suitable approach in prospective LCA compared to the use of average suppliers in attributional LCA (Schaubroeck et al., 2021; Weidema et al., 2018). By identifying marginal suppliers, the study aimed to address environmental impacts associated with the increased demand for brown seaweed-based bioplastic.

The fact that the consequential approach employs the substitution method was deemed appropriate to address the utilization of seaweed residues from the biorefinery as a substitute for materials like PLA. By employing substitution, the study aimed to provide a more accurate assessment of the environmental consequences of using seaweed-based bioplastic, taking into account the potential benefits and drawbacks of this substitution.

In other words, in consequential LCA the focus is on identifying the changes and how that relate to the functional unit of the system. Using the consequential LCA approach, this study aims to identify the indirect effects and potential trade-offs that result from changes in the system under study and to assess the environmental implications of these changes.

Overall, the choice of consequential LCA in this study allowed for a comprehensive evaluation of the environmental consequences of the brown seaweed-based bioplastic, considering factors such as marginal suppliers, and co-product handling through substitution. Consequential LCA is particularly useful for prospective technology assessment, as it can assess the environmental impact associated with the increasing demand for emerging technologies.

CHAPTER 2. RESEARCH DESIGN

2.1 RESEARCH GAPS

Seaweed-based bioplastics is an emerging technology, and we are only now starting to understand the production process and its related impacts. There are therefore several notable research gaps in this context, that need further investigation. Firstly, there is a lack of LCA studies specifically conducted on seaweed-based bioplastic production, both at pilot and industrial scales. While there have been previous studies on the LCA of seaweed cultivation (Thomas et al., 2021), other seaweed-based products (Beckstrom et al., 2020) or other bioplastics not derived from seaweed (Bishop et al., 2021; García-Velásquez and van der Meer, 2022; Tonini et al., 2021), there is no research specifically focused on bioplastics derived from seaweed. There are also experimental research studies exploring the development of seaweed-based bioplastics, but they have not conducted LCAs (Albertos et al., 2019; Aragão, 2022; Helmes et al., 2018). Additionally, there have been reviews on the utilization of seaweed for bioplastics and packaging, providing insights into their potential benefits, but these reviews do not include a review of LCA results (Carina et al., 2021; Lim et al., 2021; Lomartire et al., 2022; Rinaudo, 2014; Shravya et al., 2021; Zanchetta et al., 2021).

Regarding the identification of marginal suppliers, existing methods used for that purpose rely on extensive data (Buyle et al., 2018; Sacchi, 2018), overlooking other uncertainties associated with emerging technologies and qualitative information. The current methods used to identify marginal suppliers for brown seaweed present a challenge due to the incomplete data sources on brown seaweed production (Araújo et al., 2021). This lack of accurate data poses a challenge for making future projections and identifying marginal suppliers in the brown seaweed industry. While quantitative information can be helpful in identifying potential marginal suppliers, it falls short of capturing the complex constraints that affect upscaling production, such as optimal growth conditions, regulatory shifts, and technological advancements. This highlights the need for alternative approaches that consider a broader range of qualitative aspects in identifying marginal suppliers. A method that allows for an in-depth examination of current market trends and the potential consequences of increasing demand for brown seaweed in bioplastic production.

Prospective LCA employs various methods to assess environmental impacts, including scenario analysis, uncertainty propagation, learning curves, and upscaling processes (Jouannais et al., 2022; Langkau et al., 2023; Sacchi et al., 2022; van der Hulst et al., 2020). Among these methods, process simulations are considered highly accurate for upscaling in prospective LCA (Parvatker and Eckelman, 2019; van der Giesen et al., 2020). Most of the current upscaling methods predominantly rely on

quantitative approaches, overlooking factors specific to seaweed cultivation, such as productivity enhancement techniques, raw material availability, and upscaling challenges. Existing process simulation tools lack standardized units to incorporate these factors, limiting their effectiveness. To bridge this research gap, it is recommended to complement quantitative methods with qualitative assessments based on expert judgment, to ensure a comprehensive evaluation of the upscaling process (Buyle et al., 2019). This integration of qualitative information provides deeper insights into emerging technology development in seaweed cultivation.

Lastly, it is worth noting that while there have been studies that delve into specific steps of the supply chain in seaweed-based bioplastic production, such as seaweed biomass sourcing (Araújo et al., 2021; van den Burg et al., 2021), alginate extraction methods (Nøklung-Eide et al., 2023), and casting and crosslinking techniques (Giz et al., 2020; Jost and Reinelt, 2018), there remains a significant research gap in terms of comprehensive overviews that encompass the entire value chain to produce seaweed-based bioplastics. Previous research has predominantly focused on examining individual steps in production, resulting in a lack of understanding of the complete picture and dynamics of the value chain for seaweed-based bioplastic production. These fragmented studies provide valuable insights into particular aspects of the process, but they fail to offer a holistic understanding of the interdependencies and interactions within the value chain. To effectively assess the environmental impacts and identify areas for improvement, a comprehensive overview that considers the entire seaweed-based bioplastic value chain would be needed.

Addressing all these research gaps is important for the advancement of seaweed-based bioplastics. This thesis contributes to the mentioned research gaps providing significant contributions to the knowledge and understanding of the environmental impact of the novel seaweed-based bioplastic. This research also enables a more holistic assessment and identification of key areas for improvement and optimization throughout the value chain.

2.2 MAIN GOAL OF THE PHD PROJECT

The primary objective of this PhD research was to evaluate the environmental impacts and feasibility of a novel seaweed-based bioplastic made from brown seaweed. To accomplish this, a prospective LCA approach was used to identify potential environmental implications across the product's entire life cycle. By employing prospective LCA, these impacts were anticipated at an early stage of technological development before the technology is implemented on a big scale.

In addition to LCA modelling, complementary methods such as conducting expert interviews, carbon balances and performing uncertainty analysis were integrated into the study. By combining these approaches, a more holistic analysis was

achieved, enabling a robust assessment of the environmental implications, benefits and constraints associated with bioplastic production from brown seaweed.

2.3 RESEARCH QUESTIONS

In alignment with the overall goal of the project, a main research question and four sub-questions were formulated. Each sub-question aligns with a specific research article, ensuring a comprehensive exploration of the topic.

Main research question:

What are the environmental consequences of the novel brown seaweed-based bioplastic from a life cycle perspective?

SUB-QUESTION 1

This novel brown seaweed-based bioplastic is considered an emerging technology requiring a prospective LCA. One important aspect in this assessment was the identification of the suppliers increasing in the market, referred to as marginal suppliers. Understanding the capabilities of these suppliers becomes necessary as the demand for brown seaweed rises with the potential growing interest of this seaweed-based bioplastic in the future.

Which are the brown seaweed suppliers that are growing in the market and can respond to an increase in future demand and what are the constraints?

This sub-research question included various areas of investigation related to the identification of brown seaweed marginal suppliers to achieve the so-called *marginal mix*. Firstly, an analysis was conducted to identify the current global suppliers of brown seaweed and to determine the projected growth of suppliers in the future. The study aimed to explain the underlying market trends and examine the potential consequences resulting from these trends. Some prospects for upscaling the brown seaweed supply were also investigated. Additionally, the research explored the policy and regulatory constraints that may influence the production and trade of brown seaweed. Lastly, the study sought to identify and analyse the market niche for bioplastics derived from brown seaweed. By understanding the specific market segment of these seaweed-based bioplastics, it becomes possible to assess the

market potential and explore opportunities for growth and market entry. Through an in-depth exploration of these research areas, Article 1 provided insights into the current and future landscape of brown seaweed-based bioplastics, including the global brown seaweed suppliers, upscaling challenges, policy constraints, the market niche of seaweed and the potentials of the novel seaweed-based bioplastic in the seaweed market. Sub-question 1 is answered throughout Article 1 (Appendix A).

SUB-QUESTION 2

After gathering empirical data at both the lab and pilot scale, a pilot LCA was performed which aimed at assessing the environmental hotspots on a small scale. The focus of this assessment was to explore the potential for recirculating seaweed co-products and evaluate various EoL scenarios.

What are the environmental hotspots at the pilot scale, the potentials to recirculate seaweed co-products and different EoL scenarios?

The objective of Article 2 was to investigate the environmental hotspots that arise at the pilot scale of brown seaweed-based bioplastic production. Additionally, the study aimed to explore the potential for recirculating seaweed co-products within the system and evaluate different EoL scenarios. This assessment involved scenario modelling and was further complemented by conducting a carbon balance analysis and an uncertainty analysis. Together, these analyses provided a comprehensive understanding of the environmental performance, carbon balance, and uncertainties associated with the production process of brown seaweed-based bioplastics, considering the recirculation of co-products. The purpose of these analyses was to determine the most optimal and realistic scenario for the upscaling phase, in Article 3. Sub-question 2 is answered in Article 2 (Appendix B).

SUB-QUESTION 3

While the pilot LCA served as a baseline for determining the most optimal and realistic scenario for the subsequent upscaling phase, assessing environmental impacts only based on laboratory-scale processes provides limited insight. Scaling up the processes was important to achieve a more precise estimation of the environmental impact at an industrial scale.

How can all the processes in the value chain be upscaled and what are the impacts of the novel seaweed-based bioplastic on an industrial scale?

The goal of Article 3 was to conduct a prospective LCA of the scaled-up production process of seaweed-based bioplastic from lab and pilot scale to industrial scale. The performed consequential LCA aimed to anticipate the impacts of increasing the demand the novel seaweed-based bioplastic. The study integrated qualitative and quantitative information across different life stages. This research represented a novel approach that combined upscaling methods and merged qualitative and quantitative data. This scaling-up enables better comparability between the new product and commercially available products in terms of their environmental performance. Sub-question is answered in Article 3 (Appendix C).

SUB-QUESTION 4

Finally, in order to get a broader perspective, the aim of Article 4 was to examine the challenges, opportunities, and lessons learned at each step of the value chain in the production of seaweed-based bioplastic. By analysing the entire process from seaweed cultivation to the final product, the study aimed to provide insights into the key factors that impact the overall effectiveness of the value chain.

What are the challenges, opportunities and lessons learnt at each step throughout the value chain and in overall?

Article 4 was a collaborative and multidisciplinary research, aiming to enhance the understanding and development of seaweed-based bioplastics by providing more details through the value chain. Through a comprehensive analysis of an existing seaweed-based bioplastic supply chain, the study contributed valuable insights, fostering progress in the field of sustainable plastic alternatives. Sub-question 4 is answered in Article 4 (Appendix D).

Figure 2-1 provides an overview of the research design, including the research questions, the methods employed to address these questions, and the corresponding results linked to their respective articles.

2.4 SUMMARY OF THE RESEARCH DESIGN

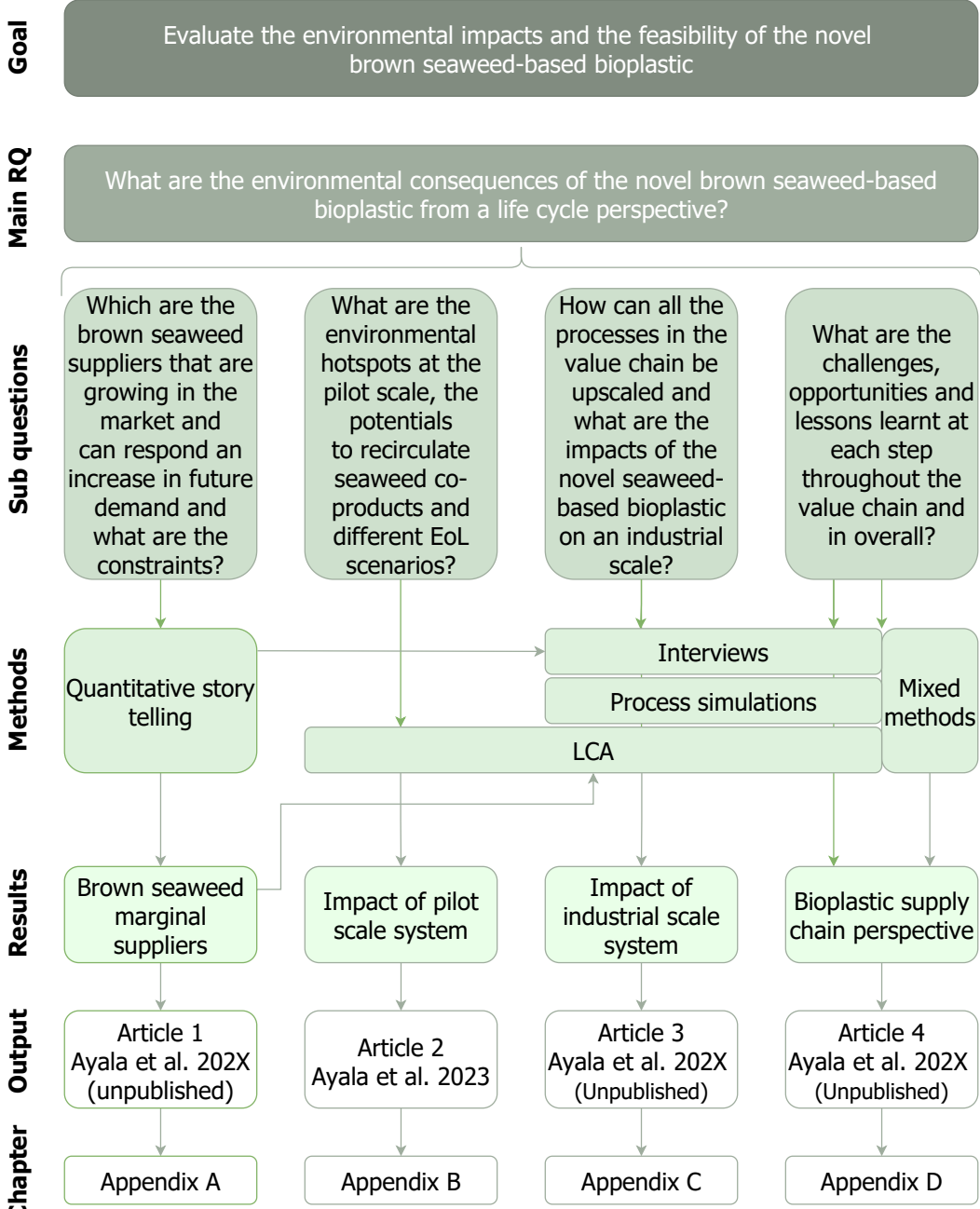


Figure 2-1: The research design schematically represented.

CHAPTER 3. METHODOLOGY

The main method used in this project was consequential LCA, from a prospective angle since the technology under assessment was an emerging technology. This approach was complemented by other qualitative and quantitative methods.

3.1 MIXED METHODS APPROACH

In this research, a mixed-methods approach was adopted by utilizing a combination of quantitative and qualitative methods. The quantitative aspect involved the use of models, simulation techniques, and LCA to assess and quantify various environmental impacts. Meanwhile, the qualitative aspect included interviews and surveys to gather valuable insights, perceptions and expert opinions. By employing this mixed-methods approach, the study was able to capture a broader and more comprehensive understanding of the subject matter, considering both the numerical data and the qualitative information.

In a prospective context, where quantitative data may be limited due to the early stages of the technology, the integration of mixed methods combining both qualitative and quantitative assessments was especially valuable. While quantitative data are typically preferred for their precision, relying solely on such data may be impractical or unfeasible in certain cases of emerging technologies. By incorporating qualitative information alongside quantitative analysis, a more comprehensive understanding of the technology's potential impacts can be achieved (Arvidsson et al., 2018). This combination of quantitative and qualitative methods facilitated a more robust analysis and provided a richer context for interpreting the research findings. This combination of mixed qualitative and quantitative methods is present in Articles 1, 3 and 4.

3.2 SUPPLIER ANALYSIS VIA QUANTITATIVE STORYTELLING

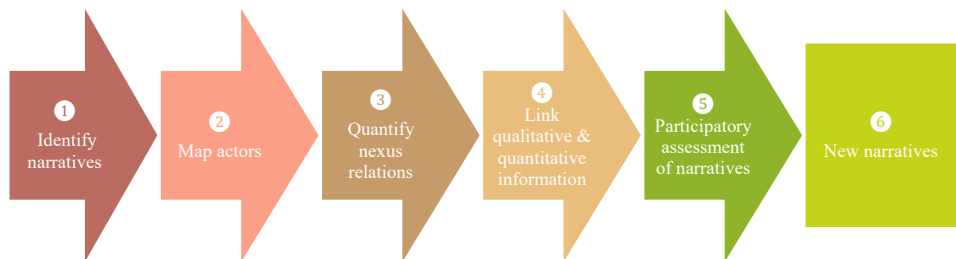
In the journey of identifying the marginal suppliers of brown seaweed, it became noticeable that current methods to identify marginal suppliers do not apply well to brown seaweed as they rely on quantitative information. Although there are two main statistical sources for seaweed production, the available data on brown seaweed is incomplete and countries have no legal requirements to publish precise data (Araújo et al., 2021). While quantitative information can serve as a starting point in identifying marginal suppliers, many constraints to upscale production can only be identified qualitatively, such as the current state of emerging trends in optimal growth conditions, including locations and cultivation designs, regulatory regime shifts, and technological developments.

A framework that combines quantitative and qualitative information was needed to identify marginal suppliers for emerging brown seaweed technologies. Quantitative story telling (QST) offers a solution to address the uncertainty and complexity of the brown seaweed production and seaweed-based bioplastic market. Originally used as a complementary method to evidence-based policy, explores multiple legitimate frames in a scientific study (Saltelli and Giampietro, 2017). In this research, the six-step procedure used in QST (Cabello et al., 2021) was adapted to identify brown seaweed marginal suppliers (see Figure 3-1).

As part of the framework, 11 brown seaweed experts with diverse backgrounds and nationalities were interviewed. The experts were selected based on their extensive knowledge of seaweed cultivation, industry experience, and academic background. The interviews aimed to gather qualitative information on global brown seaweed suppliers, biomass utilization, and the market niche for brown seaweed bioplastic.

The QST approach is elaborated on in Article 1 (Appendix A), providing a detailed understanding of its application. Part of the information gathered in the interviews was also used as a source of qualitative information in Article 3 (Appendix C) and Article 4 (Appendix D). On the other hand, the marginal mix resulting from Article 1 using the QST approach, was used as the marginal mix in the LCA in Article 3.

Steps in quantitative storytelling



Methodology in quantitative story telling

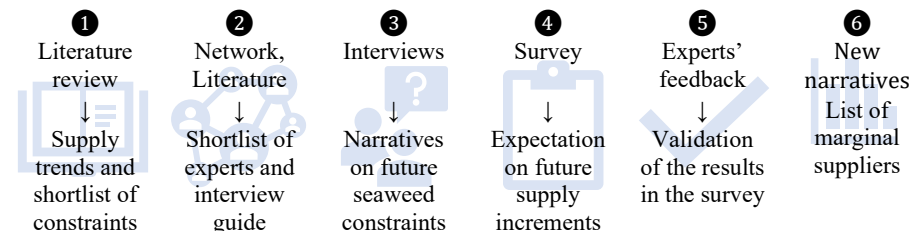


Figure 3-1: Steps in quantitative storytelling, methods applied. Inspired by Cabello et al. (Cabello et al., 2021) and adapted from (Ayala et al., 2023a).

3.3 PILOT-SCALE ANALYSIS VIA LCA

A pilot-scale LCA was conducted using the experimental primary data collected from the lab and pilot-scale processes within the PlastiSea project. The aim was to find the environmental hotspots, the potential of recirculating seaweed co-products and the most optional EoL scenario. The pilot LCA was complemented with a carbon balance and an uncertainty analysis.

Five main scenarios were modelled as part of this research. The first scenario, referred to as the base scenario, served as the reference point. Two additional scenarios involved the recirculation of co-products from the biorefinery step, which was later used in the film fabrication step. Another two scenarios focused on substituting seaweed residues for PLA (Figure 1-5). Moreover, each of these five scenarios was modelled with two different EoL options: incineration and biodegradation, resulting in ten scenarios in total. The ReCiPe midpoint (E) was used as the LCIA method.

The objective of conducting a carbon balance was to accurately monitor carbon flows and ensure a comprehensive carbon accounting throughout the entire life cycle of the process. The carbon content of the components in the seaweed, biorefinery and seaweed-based bioplastic was calculated based on their molecular weight, the number of carbon molecules per molecule and their proportions.

An uncertainty analysis was conducted to deal with the inherent uncertainties in data and assumptions of the system in the pilot scale. The uncertainty analysis was performed in the foreground using a pedigree matrix on impact scores, considering indicators *reliability*, *temporal correlation*, *geographical correlation*, *completeness* and *further technological correlation* (Ciroth et al., 2016). A stochastic approach was employed using a Monte Carlo simulation to propagate input uncertainties to the outputs through 1000 iterations.

The pilot-scale analysis following the mentioned steps was conducted in Article 2 (Appendix B).

3.4 UPSCALING ANALYSIS VIA PROCESS SIMULATION

Process simulations were used to upscale the biorefinery and film fabrications pilot-scale processes to an industrial scale. Two prospective scenarios were simulated: 4000 tonnes of seaweed-based bioplastic in 2030 and 2 million tonnes in 2035. The software Aspen Plus was used for this purpose, where the pilot-scale data was used as input data and the simulations result in mass and energy flows at the desired scales. When performing the LCA, the LCIA used was also ReCiPe. However, the ReCiPe method originally does not include biogenic carbon. Biogenic carbon refers

to carbon derived from renewable resources, such as seaweed. The carbon uptake of the seaweed was also modelled as biogenic carbon. The characterization factor for biogenic carbon was modified (from 0 to 1) and, thus, the biogenic carbon, including the carbon uptake in the seaweed cultivation, was accounted for.

Process simulations were part of the upscaling methods used in Article 3 (Appendix C). The process simulations were combined with expert opinions to upscale the seaweed cultivation and interviews to get a deeper understanding of the potential of upscaling and increasing the productivity of the seaweed cultivation.

The method of upscaling via process simulation and the prospective LCA performed on an industrial scale was used in Article 3 (Appendix C).

3.5 SUPPLY CHAIN ANALYSIS VIA MIXED METHODS

Article 4 (Appendix D) was a collaborative research among the PlastiSea partners. Different methods from different scientific domains were used to get the supply-chain overview.

This article employed a combination of different methods, with each step in the value chain being assessed using one or more specific methods. By utilizing multiple approaches, a comprehensive evaluation of the environmental impacts related to each stage of brown seaweed-based seaweed-based bioplastic production was obtained.

Each step involved different experiments and the use of a different research method, carried out by a different partner in the PlastiSea project, as outlined below:

- Seaweed cultivation: on-site experiments of seaweed growth in a real-world seaweed cultivation plant were conducted. This research focused on the identification of optimal parameters to cultivate seaweed. The experiments were performed by PlastiSea partner Seaweed Solutions.
- Seaweed processing or biorefinery: The method consists of an acid treatment, wash and alkaline extraction to extract the alginate. The objective was to identify the optimal method for extracting alginate while preserving its inherent properties. The experiments were performed by PlastiSea partner SINTEF.
- Seaweed-based bioplastic film at lab scale: The production of seaweed-based bioplastic films at the pilot scale consists of three methods - Solution preparation, casting, coating and drying. Aims to find the ultimate seaweed-based bioplastic formulation to achieve optimal properties. The experiments were performed by PlastiSea partner BZEOS.

- Seaweed-based cellulose nanofibers at lab scale: Preparation cationic cellulose nanofibers, producing composite microfibers using interfacial polyelectrolyte complexation drawing. The aim is to develop a technology for effectively utilizing cellulose nanofibers, investigating their cationic charge density and mechanical performance in composite microfibers. The experiments were performed by PlastiSea partner KTH.
- Film fabrication at pilot scale: Assessment of the bottleneck for scalability, specifically focusing on issues related to scalability and water evaporation. The aim was to identify strategies for achieving cost-efficient production of the seaweed-based bioplastic film. The experiments were performed by PlastiSea partner AITIIP.
- Analysis of the seaweed-based bioplastic properties: Assessment of moisture absorption and release, mechanical properties, and biodegradability. The experiments were performed by PlastiSea partner BZEOS.

These findings were combined with the findings from the analysis conducted in this PhD project using the methods presented in the previous sections: identification of marginal suppliers via QST, LCA and interviews to understand the prospect seaweed-based bioplastic market. An extended explanation of all the results is available in Article 4 (Appendix D).

CHAPTER 4. RESULTS

4.1 MARGINAL SUPPLIERS OF BROWN SEAWEED

In this section, the focus is placed on summarising the results of the identification of marginal suppliers, their marginal mix, and the constraints associated with future seaweed supply. Furthermore, a summary of the interview results with seaweed experts is provided. The full results are available in Article 1 (Appendix A).

Figure 4-1 presents the projected marginal mix for the years 2025, 2030, and 2035. It indicates that China is currently the main supplier and is also expected to be the primary supplier in the middle and long term. The other Asian suppliers, Korea and Japan, also have an important share in the future. Even if those suppliers are foreseen as the primary ones in the long term, they are expected to have a slightly smaller share in the long term. On the other hand, there are emerging suppliers that will play an important role in the future, Norway being an important one.

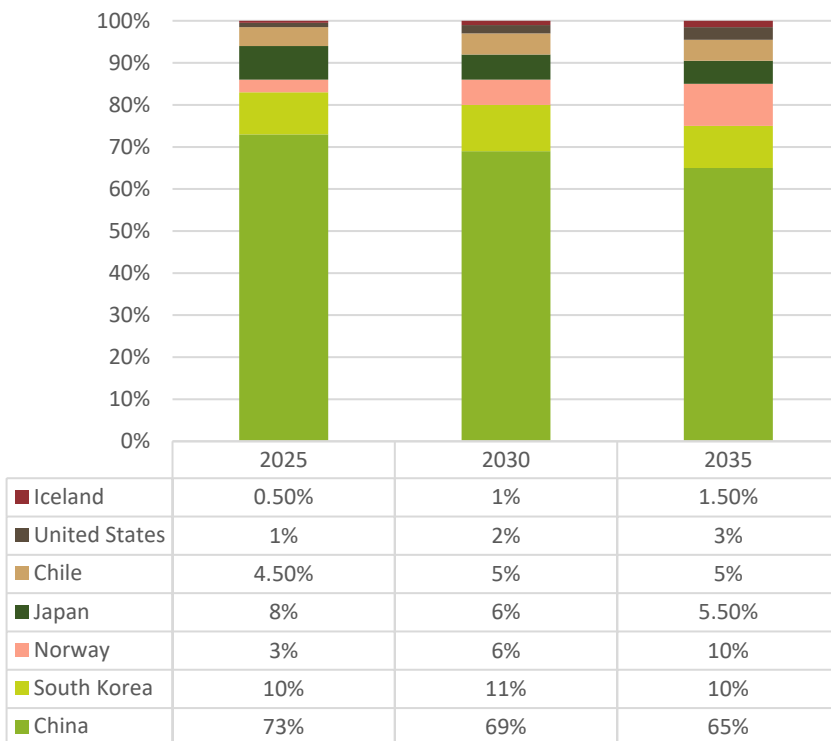


Figure 4-1: Marginal suppliers using the information from (Ayala et al., 2023a).

The identified constraints that can limit the seaweed supply were the geographical market delimitation, policies and regulations, technological development and natural constraints. The experts provided the following answers in the identified constraints:

Regarding geographical market delimitation and policies and regulations, the experts mentioned that seaweed is a global free market with no significant regulatory limitations, although quality regulations are necessary. Market preferences influence local seaweed purchasing, with Asian countries having high internal demand and European consumers prioritizing local sourcing. Regulatory constraints exist for seaweed cultivation permits, with more flexibility in Asian countries.

In terms of natural constraints and production capacity, the experts emphasized the importance of considering species productivity, local adaptation, and growth rates when upscaling brown seaweed production. Environmental factors such as water conditions, nutrients, light, and tidal variations play a crucial role in seaweed growth. Cultivation, selective breeding, and productivity-enhancing techniques were highlighted as essential for increasing production while preserving the marine environment.

With regard to technological development and upscaling, the experts mentioned emerging technologies such as offshore seaweed farms and shared infrastructure with wind farms to facilitate production. However, challenges such as high costs, environmental limitations, social acceptance, and market development need to be addressed. While Asian suppliers possess valuable knowledge, especially in breeding techniques, direct application in Europe is hindered by environmental concerns and labour costs. Automation was considered crucial in Europe due to the higher labour costs.

When it comes to the market for seaweed-based bioplastics, the experts indicated that seaweed-based bioplastics were perceived as a lower-ranked target compared to other applications such as food, feed, pharmaceuticals and cosmetics. However, as brown seaweed production scales up and production costs decrease, seaweed-based bioplastic production may become economically viable in the future. Utilizing lower-grade seaweed or exploring combined production methods were suggested as potential strategies. The importance of seaweed-based bioplastics in the transition from fossil fuels may increase if conventional plastic prices rise or with government incentives for biopackaging.

Extended results of this research are available in Article 1 (Appendix A).

4.2 LCA OF SEAWEED-BASED BIOPLASTIC PRODUCTION AT PILOT SCALE

In this section, the results of the pilot-scale LCA are presented. The potential recirculation of seaweed co-products, such as cellulose and mannitol, are assessed as well as the impact of different EoL choices, including composting and incineration. The results in this section are shown with and without accounting for biogenic carbon. However, the specific analysis of biogenic carbon is not included in Article 2 but it is addressed in this section. Additionally, the LCA is complemented with a carbon balance. The section is organized into two sub-sections based on the type of seaweed-based bioplastic under investigation. The extended results on the pilot LCA including the carbon balances and the results in the uncertainty analysis are presented in Article 2 (Appendix B).

All the presented results in this section employed the ReCiPe midpoint (E) LCIA method. It is important to note that the scenarios were modelled both with and without the inclusion of biogenic carbon, which includes the carbon uptake of the seaweed during its cultivation. In order to account for the biogenic carbon, a characterization factor of 1 was applied to the biogenic carbon component as ReCiPe originally does not include it.

It is also worth mentioning that the LCA analyses conducted in the articles focus on evaluating the environmental impact of the seaweed-based bioplastic film and PLA substitution. Therefore, the results on the seaweed-based bioplastic pellets are novel and exclusive to the thesis. All the results in this section show the contribution analysis at the pilot scale of the seaweed-based bioplastics (cf. Section 1.2) in the global warming impact category (CO₂-eq.) per kilogram of seaweed-based bioplastic (FU = 1 kg bioplastic).

The results in Article 2 contain however an error, which was identified after publication and corrected. See Corrigendum (Appendix B) for the details regarding the error and its correction. The corrected results are also reported in Figure 4-2, Figure 4-3 and Figure 4-5 (*PLA5* and *PLA30*). The results from Article 2 underestimated the impact of the production stage because the amount of crude extract needed in the seaweed-based bioplastic was not properly accounted for.

4.2.1 SEAWEED-BASED BIOPLASTIC FILM

Prior to the LCA, a carbon balance was performed to assess the carbon flows. Figure 4-2 illustrates the carbon balance in the mannitol (*MANN*) scenario where cellulose and mannitol from the seaweed are extracted in the *Biorefinery* and are recirculated and used in the *Film fabrication* step. This carbon balance was chosen to be attached

in the thesis as it is the most representative one to illustrate the recirculation of the co-products.

The carbon balance results in Figure 4-2 show the potential of recirculating the co-products derived from seaweed in the production of seaweed-based bioplastics. Despite the limited quantity of mannitol present in the seaweed, increasing its utilization slightly reduces the need for glycerol, resulting in a more efficient use of resources. Similarly, while the amount of cellulose in seaweed is relatively small, it is still sufficient for seaweed-based bioplastic production since only a small quantity of cellulose is required. In fact, there is an excess of cellulose available, indicating that the seaweed can meet the cellulose demands for seaweed-based bioplastic development. The C balance in the rest of the scenarios can be found in the corrigendum in Article 2 (Appendix B).

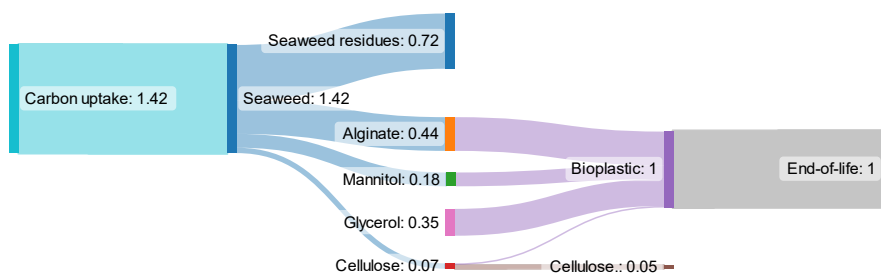


Figure 4-2: Sankey diagrams of the carbon balance in the cellulose and mannitol recirculation scenario MANN (3). Units: kg carbon. From Ayala et al. (2023b).

The LCA results in Figure 4-3 and Figure 4-4 show the contribution analysis at the pilot scale of the seaweed-based bioplastic film (cf. Figure 1-3) in the global warming impact category. The results include the seaweed-based bioplastic film in a pilot-scale production without recirculating seaweed co-product (*BASE*), recirculating the cellulose from the seaweed into the film fabrication step (*CELL*) and recirculating the cellulose and mannitol (*MANN*). Each scenario has two sub-scenarios: incineration (*inc*) and composting (*com*).

The highest impacts are observed in the Film fabrication stage across all scenarios, with values ranging from 1.64 to 2.37 kg CO₂-eq. The biggest contribution to the overall impacts varies depending on the scenario. In this study, assessing the recirculation of co-products played an important role. The scenario with the highest impact in terms of global warming is *CELL inc* (6.2 kg CO₂-eq.), which involves the recirculation of cellulose. On the other hand, the scenario with the lowest impact is *MANN com* (4 kg CO₂-eq.), which includes the recirculation of cellulose and mannitol.

The EoL stage of the seaweed-based bioplastic has different impacts depending on the chosen method, either incineration or composting. The impacts vary significantly across EoL scenarios. The EoL impact is 0.11 kg CO₂-eq. for all the composting scenarios, which is lower than the 1.27 kg CO₂-eq. attributed to the incineration scenario. Hence, the choice between incineration and composting in waste management practices influences the environmental performance of the seaweed-based bioplastic.

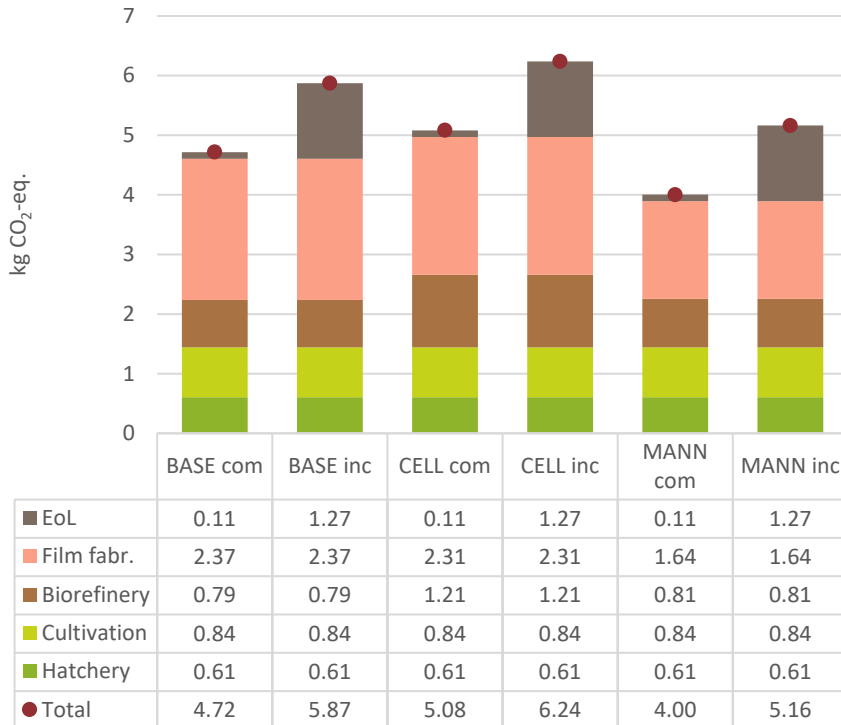


Figure 4-3: Impacts in the global warming category not accounting for the biogenic carbon based on (Ayala et al., 2023b), Corrigendum, excluding the PLA5 and PLA30 scenarios.

In Article 2, the exclusion of biogenic carbon from the analysis was due to the LCIA method (ReCiPe) used, where the characterization factor remained unchanged and did not incorporate the assessment of biogenic carbon. In this thesis, biogenic carbon was included in Figure 4-4 to compare the inclusion and exclusion of biogenic carbon.

The negative values in Figure 4-4 represent carbon uptake during seaweed cultivation, and accounting for biogenic carbon can result in lower overall CO₂ equivalent values, reflecting the positive contribution of seaweed cultivation to carbon sequestration. As expected, when comparing Figure 4-3 and Figure 4-4, the

difference between accounting and not accounting for biogenic carbon becomes noticeable, which significantly influences the results. For example, the highest impact without accounting for biogenic carbon in Figure 4-3 is 6.24 kg CO₂-eq. and the lowest impact is 4 kg CO₂-eq., while the highest impact decrease when accounting for the biogenic carbon is 4.21 kg CO₂-eq. in the *CELL inc* scenario and the lowest 3.06 in the *MANN com* scenario. The difference between accounting and excluding biogenic carbon is between 0.63 kg CO₂-eq. and 2.24 kg CO₂-eq., with a difference of 2.32 kg CO₂-eq. in the base scenario.

The inclusion or exclusion of biogenic carbon is also noticeable in the EoL stage. When accounting for biogenic carbon, the impact of incineration decreases to 0.52 kg CO₂-eq. with carbon uptake, compared to 1.27 kg CO₂-eq. without accounting for it. Contrarily, in the case of composting, the impact is slightly higher at 0.22 CO₂-eq. when accounting for biogenic carbon, compared to 0.11 kg CO₂-eq. without accounting for it. This difference can be attributed to the way background processes are considered and how ecoinvent handles the accounting of biogenic carbon.



Figure 4-4: Impacts in the global warming category accounting for the biogenic carbon based on the scenarios.

It can be argued that the decision to account for biogenic carbon depends on the chosen EoL process. If the EoL process is not included within the system boundaries, it is not recommended to include biogenic carbon. This is because only the carbon uptake would be considered, without accounting for the carbon release, overestimating the system's benefits. However, when considering composting as the EoL process, the decision regarding the inclusion or exclusion of biogenic carbon becomes less straightforward. In composting, carbon is stored and released gradually over a longer time frame. In this case, excluding biogenic carbon may prevent giving excessive credit to the system, as it does not account for the carbon released in the short term. On the other hand, accounting for the carbon uptake is relevant when modelling incineration as the EoL process, as all the carbon in the seaweed-based bioplastic is released during incineration. Therefore, the inclusion or exclusion of biogenic carbon should be carefully evaluated, considering the specific characteristics of the EoL process and its implications for accurately assessing the system's environmental impacts.

4.2.2 SEAWEED-BASED PELLETS AND SUBSTITUTION PLA

Figure 4-5 represents the global warming impact per kilogram of seaweed-based bioplastic in the extruded seaweed-based bioplastic pellets (cf. Figure 1-4) and the seaweed-based bioplastic film with PLA substitution (cf. Figure 1-5). As mentioned earlier in Section 1.2.3, the seaweed residues from the biorefinery process, which aims to extract alginate for seaweed-based bioplastic film, can be utilized as a substitute for PLA. The LCA results for PLA5 and PLA30 represent the environmental impacts of the seaweed-based bioplastic film, taking into account the incorporation of seaweed residues as a replacement for PLA. Notably, the findings from the four PLA substitution scenarios indicate that as the percentage of substituted PLA increases, the impact decreases.

When it comes to the environmental performance of the seaweed-based pellets (Pellets), the relevance of accounting for biogenic carbon becomes particularly evident. As outlined in Section 1.2.2, the pellet formulation consists of 70% alginate, requiring a significant amount of seaweed. Consequently, the carbon uptake of the seaweed becomes crucial. In the results pertaining to the pellets, the contribution of the film fabrication step is notably lower, primarily due to the reduced utilization of glycerol.

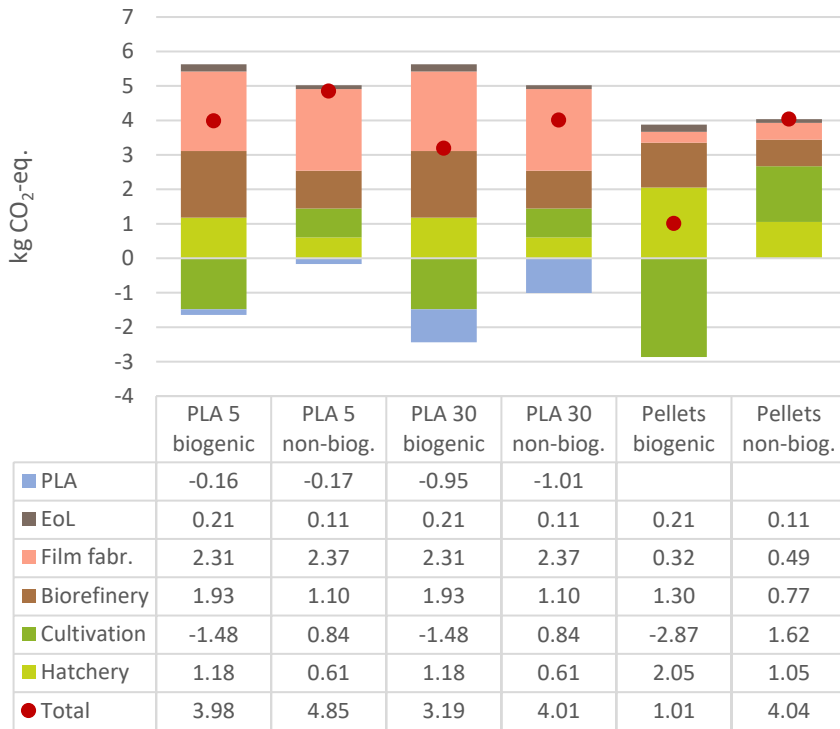


Figure 4-5: Impacts in the global warming impact category of seaweed-based bioplastic film with PLA substitution (5% and 30% substitution) and the pellets. Each product accounting and not accounting for biogenic carbon (non-biog.) (FU=1kg bioplastic).

The provided LCA results at the pilot scale serve as a summary of the findings in Article 2 (Appendix B) and a part of the results in Article 4 (Appendix D). Article 2 also provides an uncertainty analysis not included in this section.

4.3 PROCESS SIMULATION AND LCA OF THE UPSCALED SYSTEM

This section presents the findings of the pilot-scale modelling, focusing on two upscaling scenarios and considering both the inclusion and exclusion of biogenic carbon. The results provided here include the mass flow data obtained from the process simulation, as well as the corresponding LCA results. For more detailed and comprehensive results, refer to Article 3 (Appendix C).

In the assessment of the upscaled impacts, the choice was made to use composting as the most realistic EoL scenario for biodegradable seaweed-based bioplastic and the BASE scenario was selected as the reference case. While the results from Article 2 indicated a slight advantage for the MANN scenario (Figure 4-3 and Figure 4-4), this difference was not considered significant enough to prioritize that scenario in the prospective LCA at an industrial scale. Additionally, the use of mannitol as a substitute for glycerol had not been experimentally validated during the project. Therefore, in Article 3, both approaches of accounting and not accounting for biogenic carbon were employed, taking into consideration the upscaling of processes and conducting a prospective LCA at the industrial scale. Two prospective scenarios were modelled: 4000 tonnes of seaweed-based bioplastic in 2030 and 2 million tonnes in 2035.

The process simulation results provide valuable insights into material flows, revealing distinct resource requirements between the 4000t and 2Mt scenarios. In the 4000t scenario (Figure 4-6), the seaweed-based bioplastic flow constitutes 2.7% of the total material flow, whereas, in the 2Mt scenario (Figure 4-7), it accounts for 4.8% of the material flow, meaning that the flow is wider and less material is needed per outcome. For instance, the seaweed input flow decreases in the larger scenario. According to the process simulation results, in the 4000t scenario 17.12kg of seaweed is required per kilogram of seaweed-based bioplastic, whereas in the 2Mt scenario, the requirement decreases to 9.57kg of seaweed per kilogram of seaweed-based bioplastic.

These findings suggest that scaling up to 2Mt enables more efficient resource utilization, as it achieves the same seaweed-based bioplastic production with fewer resources compared to the 4000t scenario. Consequently, scaling up to 2Mt has the potential to enhance resource efficiency and reduce environmental impacts, emphasizing the significance of considering larger-scale production.

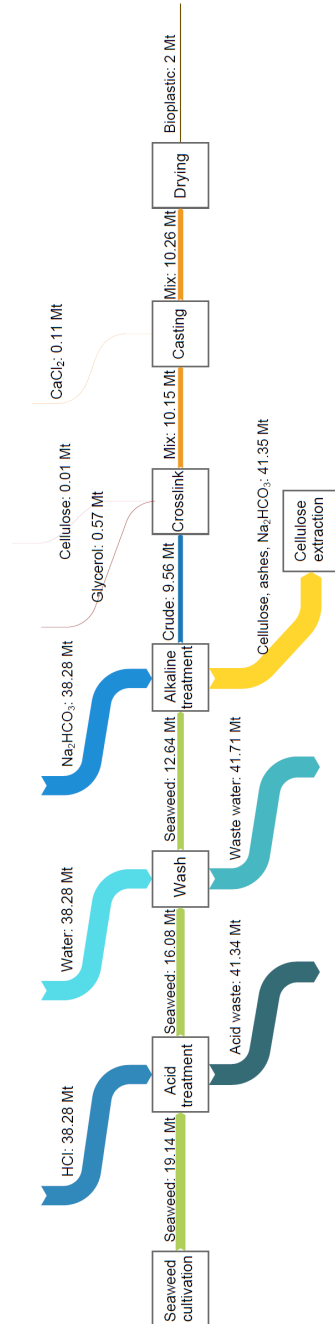
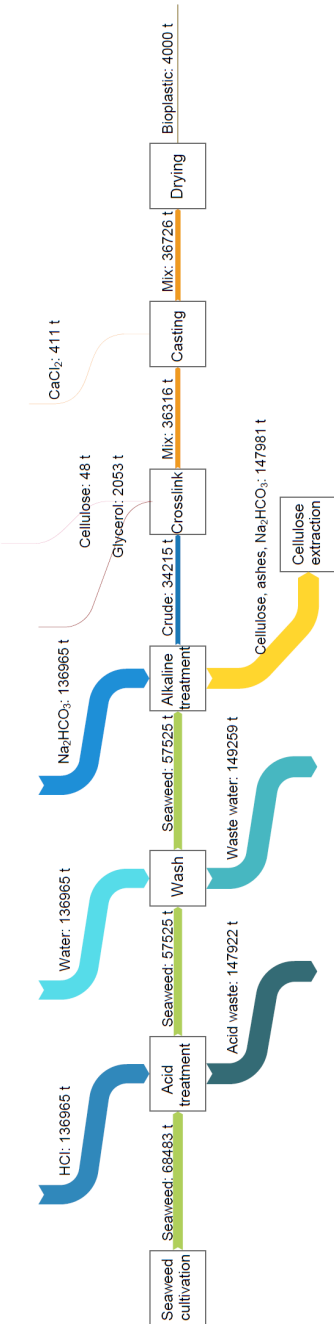


Figure 4-6: Mass flow in the 4000t scenario resulting from the process simulation illustrating the inputs and outputs within the system (Ayala et al., n.d.).

Figure 4-7: Mass flow in the 2Mt scenario resulting from the process simulation illustrating the inputs and outputs within the system (Ayala et al., n.d.).

The LCA results on the industrial-scale scenarios are based on the process simulation results on Aspen Plus. The energy and mass flows resulting from the process simulation were used as the LCI. The assessments were conducted by considering both the accounting and non-accounting of the biogenic carbon.

Figure 4-8 illustrates the trend of decreasing impacts as the production is scaled up. Notably, the pilot-scale scenario in Figure 4-3 and Figure 4-4 (*BASE com*) exhibits a higher environmental impact compared to the upscaled scenarios in Figure 4-8. Across all scenarios, the film fabrication step emerges as the primary contributor to the overall impacts. This is primarily attributed to the use of glycerol, which has substantial environmental consequences. The results of the upscaled system analysis demonstrate that the Film fabrication stage consistently exhibits the highest impacts in all scenarios, whether biogenic or non-biogenic and for both 4000t and 2Mt. This aligns with the findings of the pilot LCA, indicating that the use of glycerol still significantly contributes to the overall environmental impacts.

One of the main highlights of the results is the significant difference in impacts due to the presence of biogenic carbon, specifically attributed to carbon uptake during the cultivation stage. It has a negative impact in the biogenic carbon scenario, indicating a carbon sequestration effect. This is evident from the negative impact values in the cultivation stage for scenarios with biogenic carbon compared to the positive values in scenarios without biogenic carbon. The cultivation stage plays a crucial role in reducing the overall environmental impact by sequestering carbon. It highlights the importance of considering the biogenic carbon component and its impact on the environmental assessment of seaweed-based bioplastic production.

The scenario with the lowest overall impact is *2Mt biogenic* with an impact of 1.37 kg CO₂-eq. This scenario considers the use of 2 million tons of biogenic carbon. It shows relatively lower impacts across all stages compared to the 4000t scenario, with a carbon footprint of 1.75 kg CO₂-eq. accounting for biogenic carbon and 3.85 kg CO₂-eq. excluding it.

In summary, the film fabrication stage has the highest contribution in the global warming impact category, and the scenario with 2 million tons of biogenic carbon has the lowest impact. Including biogenic carbon in the analysis reduces the global warming potential, highlighting the importance of considering the carbon sequestration effect of biogenic materials in the evaluation of environmental impacts.

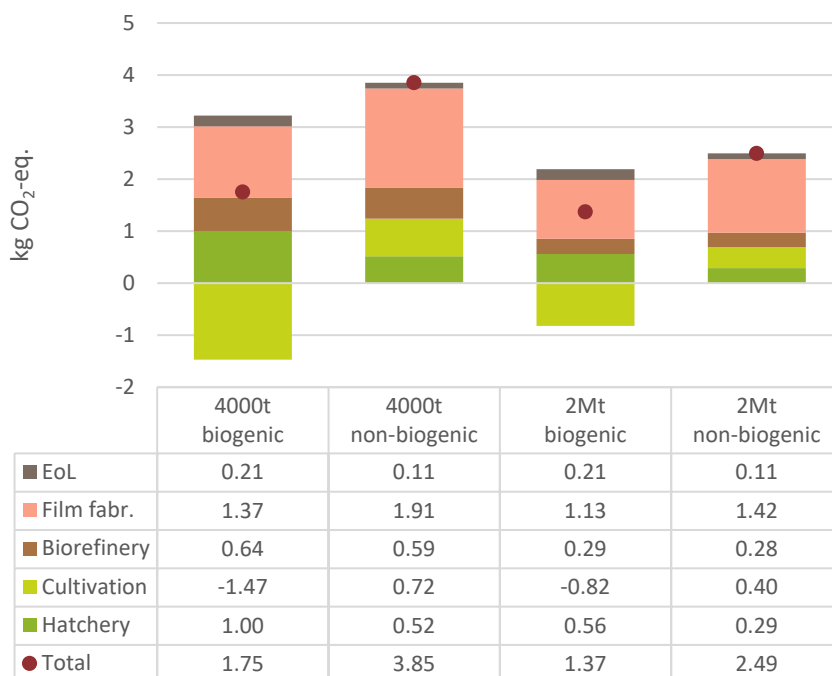


Figure 4-8: Impacts in the global warming category in the 4000 t in 2030 and 2 Mt in 2035 scenarios, accounting and not accounting for the biogenic carbon. Based on Fig5. 5 from Ayala et al. (202X.) (Article 3)

When assessing other impact categories, in the marine and freshwater eutrophication categories, the results show that seaweed cultivation has a significant impact on reducing nutrient levels. Specifically, in terms of marine eutrophication, the 4000 t scenario demonstrates a negative value of -0.017 kg N-eq., while the 2 Mt scenario has a value of -0.009 kg N-eq., seaweed cultivation being the main contributor to that negative value. Similarly, in freshwater eutrophication, the 4000 t scenario shows a negative value of -0.0037 kg P-eq., whereas the 2 Mt scenario has a value of -0.002 kg P-eq. These findings indicate that seaweed cultivation plays a crucial role in mitigating eutrophication and improving water quality in marine and freshwater environments.

The extended results on the upscaled scenario are available in Article 3 (Appendix C) and partly in Article 4 (Appendix D). The LCA result in Article 4 is a compilation of the results in Articles 2 and 3, showing the impacts on the pilot and upscaled scenarios modelled accounting for the biogenic carbon.

CHAPTER 5. DISCUSSION

The discussion section is structured into three distinct parts. Firstly, the methodological choices and the validity of the results are examined, including the limitations and uncertainties associated with expert-based assessments, the LCA model, and the assessment of upscaling. Secondly, there is a dedicated discussion on terminology, specifically exploring the meaning and implications of the terms *prospective* and *bioplastic*. Finally, the challenges and opportunities that arise in relation to seaweed-based bioplastics are discussed.

5.1 METHODOLOGICAL CHOICES AND VALIDITY OF RESULTS

5.1.1 ATTENTION POINTS USING EXPERT-BASED ASSESSMENTS

Expert-based studies have inherent limitations, including potential biases and subjectivity due to varying perspectives and experiences. Therefore, the limitations related to the expert-based assessment within this thesis can be discussed. In the case of Article 1, it can be argued that the relatively small sample of 11 experts can affect the representativeness of the findings. However, it is important to note that these experts were highly qualified and possessed diverse expertise, which enhanced the credibility of their insights. The validity of the assessment was strengthened by the variety of experts involved, from different backgrounds, providing a more comprehensive perspective. Additionally, data saturation was achieved during the interviews as similar answers were obtained after a few interviews, indicating that a sufficient amount of information was gathered. The results obtained from the interviews were further validated through triangulation with the questionnaires, providing additional robustness. The answers provided by the experts were systematically summarized, ensuring a comprehensive and reliable analysis. Consequently, the outcomes of this assessment can be considered reproducible to a good extent.

5.1.2 LIMITS OF THE LCA MODELS DEVELOPED IN THE PROJECT

The use of LCA also presents some limitations that should be considered. Firstly, LCA relies on data and assumptions, which can introduce uncertainty into the results. The accuracy and reliability of the assessment depend on the quality of the data and the completeness of the life cycle inventory. In this specific case, the quality of the data was high since it is based on empirical data. The boundaries and system boundaries of the assessment also need to be carefully defined to ensure a comprehensive evaluation of the environmental impacts, avoiding double counting.

Another important aspect is that the temporal dimension is not fully considered in LCA. Specifically, the current LCA framework may not adequately account for the release of carbon in the biowaste EoL scenario associated with seaweed-based bioplastics. As a result, the system may receive too much credit in terms of its environmental impact, as the carbon release may occur in the future rather than immediately. This temporal discrepancy calls for further research to develop more refined methodologies that incorporate the temporal aspects and provide a more comprehensive understanding of the dynamic environmental implications over time.

Additionally, the existing impact categories in LCA fail to capture the specific environmental impacts of microplastics derived from fossil plastics. As a result, some advantages of seaweed-based bioplastics over fossil plastics may not be adequately reflected in the current assessment framework. Furthermore, LCA may not capture all relevant environmental aspects, such as social or economic impacts, which are not directly related to the life cycle assessment of the product.

LCA has limitations when it comes to incorporating spatial considerations, particularly in relation to ecological, climate, water and biodiversity impacts. The lack of spatial resolution in LCA hinders a comprehensive understanding of where and how land-use change and its associated impacts will occur. Chaplin-Kramer et al., (2017) emphasize the importance of connecting supply chain decisions with sustainable raw material production by considering where increased demand will drive production, translating it into land use change, and assessing the meaningful impact of such changes. Incorporating spatial information into LCA enables a more informed understanding of environmental consequences and supports decision-making.

Another limitation of the methodology is the exclusion of biogenic carbon in commonly used LCIA methods, such as ReCiPe. These methods do not consider the carbon uptake associated with the growth of biogenic materials, including seaweed. As a result, the potential carbon sequestration and overall carbon balance of seaweed biomass are not accounted for in the assessment. Although it is possible to modify the characterization factor for biogenic carbon to include its carbon uptake, the current omission of this factor in existing methods challenges a comprehensive evaluation of the environmental impacts associated with biogenic materials.

5.1.3 UNCERTAINTIES IN THE ASSESSMENT OF UPSCALING

The choice of the upscaling scenarios, namely 4000t in 2030 and 2Mt in 2035, can be discussed since the 4000t scenario reflects a more realistic approach than the 2Mt scenario. The production volume of 2Mt can be considered too large to be achieved in a single plant, raising potential practical limitations and operational challenges. By focusing on the 4000t scenario, which represents a more feasible scale of production, the study maintains a more practical perspective that aligns with

industrial capacities. While the 2Mt scenario was included to test the upper limit and assess potential efficiency gains, it serves as a reference point rather than a practical implementation target. By prioritizing the 4000t scenario, the study maintains a realistic framework for evaluating resource utilization, environmental performance, and the overall viability of seaweed-based bioplastic production.

Another important factor to take into account is the inherent biases and constraints associated with the methodology utilized for upscaling. The accuracy of the results can be influenced by the validity of the assumptions made during the simulation. It should also be noted that Aspen software may not be optimal for modelling certain types of processes, particularly those involving biological systems. When scaling up biorefineries, difficulties may arise in finding suitable components within the Aspen database. It is important to consider these limitations while interpreting the findings.

5.2 TERMINOLOGY DISCUSSION

5.2.1 THE MEANING OF *PROSPECTIVE* IN LCA

The use of the terminology *prospective* in reference to conducting LCA for emerging technologies at low TRL could be discussed. In the literature, most of the time the terms *prospective*, *ex-ante* or even *anticipatory* are interchangeably used. However, according to (Arvidsson et al., 2018), and a recent update of the terms (Arvidsson et al., 2023), a subtle difference exists between those terms. *Prospective* LCA refers to assessing product systems at a future time. *Ex-ante* LCA also refers to assessing a product system at a future time, which is immature at the present but is expected to be mature in the future. In other words, according to that definition, *ex-ante* is a type of *prospective* LCA with a low TRL in the present and is expected to have a high TRL in the future.

Arvidsson also defines *Anticipatory* LCA based on (Wender et al., 2014). which is similar to *ex-ante* LCA but with considerable participatory stakeholder engagement. Stakeholder participation can also be present in *prospective* and *ex-ante* LCAs but the stakeholder involvement is more extensive in the case of *anticipatory* LCA. Even if there is some stakeholder involvement in this research, mainly seaweed cultivation stakeholders, is not extensive enough according to this definition. The term *anticipatory* was, therefore, discarded in this case study.

According to these definitions and considering that the technology under assessment is expected to be mature in the future, this LCA should be called *ex-ante*. However, the term *prospective* was chosen as the LCA is position in the future, regardless of its maturity. In that sense, it could be argued that the term *Prospective LCA of an emerging technology* would have the same meaning as *ex-ante*. Overall,

the terms are indistinctively used in the literature and the difference in the terminology was not considered relevant in this study.

5.2.2 WHAT DOES *BIOPLASTIC* MEAN?

In the thesis, the term *bioplastic* was used to refer to *bio-based plastic* since the seaweed-based bioplastic is also biodegradable. However, a few remarks regarding the chosen term are needed since limitations in the naming of bioplastics and their recyclability should be considered. The term *bioplastic* includes a wide range of plastics, some of which are biodegradable while others are not (Ansink et al., 2022). This can lead to confusion and misinterpretation. While a bioplastic may be derived from a bioresource, it does not necessarily mean it is fully biodegradable. In fact, some biodegradable bioplastics may still contain a small percentage of fossil-based components that do not degrade and can contribute to the generation of microplastics (Zanchetta et al., 2021).

Recycling poses another challenge, as the current recycling infrastructure is often not adequately equipped to handle bioplastics (Jayakumar et al., 2023b). It is important to note that the additives used in this particular bioplastic, such as glycerol and cellulose, are not derived from fossil fuels. Additionally, these additives are edible, which means they do not break down into harmful microplastics. However, the overall recyclability of bioplastics remains an area of concern and further research is needed to improve recycling processes and infrastructure for these materials.

5.3 CHALLENGES AND OPPORTUNITIES

5.3.1 SEAWEED PRESERVATION AND SEASONALITY

One of the challenges in working with seaweed is its preservation and the seasonality of the seaweed. The seaweed harvesting season in Norway goes from April to August (Handå et al., 2013) and most of the compounds present in seaweed degrade in the initial days after the harvest (Lytou et al., 2021). Seaweed preservation is, therefore, important. Research has been conducted on seaweed preservation methods such as drying, which requires high energy input, and fermentation, which necessitates large storage volumes. While these preservation methods help extend the shelf life of seaweed compounds, they often result in lower quality compared to fresh seaweed (Larsen et al., 2021). Additionally, transporting wet seaweed is expensive due to the high-water content and volume of the biomass.

An opportunity to address these challenges is to establish biorefineries near seaweed farms. This proximity would help preserve the quality of the seaweed components by minimizing transportation time. To mitigate the impact of

seasonality, diversifying the market and working with dry seaweed can be explored as an alternative.

Another solution to address seasonality could be to focus on red seaweed, which can be cultivated and harvested year-round and has a rapid growth rate. Investigating the potential use of compounds such as carrageenan, or other red seaweed compounds, to produce seaweed-based bioplastics could be a good opportunity (Lomartire et al., 2022). However, it is important to note that red seaweed is less readily available in Europe as it primarily grows in tropical areas (Msuya et al., 2022), which potentially presents social, including gender inequality (Msuya and Hurtado, 2017), and logistical challenges since they primarily grow in developing countries, Indonesia being the main producer (Ferdouse et al., 2018).

5.3.2 INVESTMENTS FOR UPSCALING THE PRODUCTION

The upscaling of seaweed-based bioplastic production presents several challenges and opportunities that need to be addressed for the successful development and implementation of this potential plastic alternative. One significant problem is the need for cost-effective biomass to produce bioplastics from seaweed, particularly to economically compete with other higher-value applications of the seaweed.

To address this challenge and avoid competing with other uses of seaweed, one potential solution is dedicating seaweed cultivation sites only to the production of seaweed-based bioplastic manufacturing. Alternatively, partnering with established seaweed producers who can supply the required biomass exclusively for seaweed-based bioplastic production is another viable option. By streamlining the cultivation process and focusing on the needs of seaweed-based bioplastic production, it becomes possible to optimize the supply chain and ensure a consistent and cost-effective supply of seaweed biomass.

Another opportunity would be collaborating with big players in the industry. Major companies or multinationals can play a significant role in driving the upscale of seaweed-based bioplastic production as they have market influence, financial resources, and manufacturing capabilities. In order to facilitate the upscaling process, one approach is for these larger companies to acquire patents or intellectual property rights from smaller or emerging companies that have developed innovative seaweed bioplastic technologies. Another approach related to big payers is that these companies may be willing to pay incentives to producers who utilise seaweed-based bioplastics, even if the cost is higher compared to conventional plastics. The benefits for these big companies would be to report lower emissions.

By exploring options such as exclusive cultivation, strategic partnerships, and collaboration with established companies, the challenges associated with sourcing cost-effective biomass for seaweed-based bioplastic production can be addressed.

5.3.3 POTENTIAL APPLICATIONS OF SEAWEED-BASED BIOPLASTIC

Another limitations of this seaweed-based bioplastic lies in its applications, primarily due to its water absorption characteristics. The propensity of the seaweed-based bioplastic to absorb water can restrict its usage in certain environments or applications where moisture resistance is crucial. This limitation poses challenges in industries where dryness or water resistance is essential, such as packaging materials for moisture-sensitive food products. Therefore, careful consideration is needed when selecting the appropriate applications for this seaweed-based bioplastic to ensure its performance and durability in the intended use cases.

One such application is in the packaging of dry goods such as cereals and snacks, where seaweed-based bioplastics can provide favourable mechanical strength and barrier properties to preserve the quality of the products. Additionally, the water and vapour permeability of these seaweed-based bioplastics makes them suitable for products that require breathability, such as cheese and other perishable goods. By carefully identifying suitable applications and leveraging the unique properties of seaweed-based bioplastics, opportunities can be found to integrate them into various industries.



Figure 5-1: Potential applications of the seaweed-based bioplastic. Credit: www.linkedin.com/company/bzeos/

CHAPTER 6. CONCLUSION

6.1 ANSWERS TO THE RESEARCH QUESTIONS

SUB-QUESTION 1

Which are the brown seaweed suppliers that are growing in the market and can respond to an increase in future demand?

Asian countries, China, Korea and Japan, was found to be the current main suppliers and is expected to be the main suppliers in the middle-long term. European and American suppliers are emerging and are expected to be important players in the future.

Given the lower price of seaweed for bioplastic production compared to other applications, the results in Article 1 indicated that a substantial market share for brown seaweed-based bioplastic in the short term would not be feasible. However, there were alternative avenues mentioned to expand the market presence of seaweed-based bioplastics. The findings suggested that integrating the production of brown seaweed bioplastics as a co-product of higher-value markets could be a promising strategy to enhance its market share. By positioning brown seaweed-based bioplastics as a complementary product to pricier target markets, the utilization of seaweed-based bioplastics could be maximized and its market potential can be effectively tapped into.

In this study, it was also concluded that current methods to identify marginal suppliers were not well-suited for addressing the challenges associated with the production of brown seaweed. Qualitative information was deemed necessary to understand most constraints associated with the QST and was spotted as a proper method to combine qualitative and quantitative information to identify marginal suppliers in this specific case. This sub-question aligns with the content discussed in Article 1, and for more detailed information, please refer to Appendix A.

SUB-QUESTION 2

What are the environmental hotspots at the pilot scale, the potentials to recirculate seaweed co-products and different EoL scenarios?

The base scenario had a carbon footprint of 4.7 kg CO₂-eq. in the composting EoL and 5.8 kg CO₂-eq. in the incineration EoL. The scenarios with the lowest impacts were mannitol recovery and the scenario with PLA 30% substitution, both with a global warming potential of 4 kg CO₂-eq. Both scenarios in with the composting EoL. The scenarios with the highest impacts were PLA 5% substitution and cellulose recirculation with incineration EoL, with an impact of 6.2 kg CO₂-eq. and 6 kg CO₂-eq. respectively, closely followed by the base scenario with incineration in its EoL (5.9 kg CO₂-eq.). The values presented here did not include biogenic carbon, as it was not taken into account during the modelling in Article 2.

The results showed that the hotspot in all the scenarios was the use of glycerol in the film fabrication step. There was a significant difference between the modelled EoL scenarios, with a reduction in CO₂-eq. emissions observed in the composting scenario compared to incineration. There was a slight decrease in global warming when recirculating seaweed co-products, but the difference was not significant.

In conclusion, the results suggested that recovering mannitol from the seaweed-based bioplastic material showed an advantage, but the base scenario, without mannitol recovery, also performed well. It is important to note that the composting EoL scenario appeared to be the most optimal EoL pathway considering the properties of the seaweed-based bioplastic. Article 2 corresponds to this specific sub-question, and further details can be explored in Appendix B.

SUB-QUESTION 3

How can all the processes in the value chain be upscaled and what are the impacts of the novel seaweed-based bioplastic on an industrial scale?

The processes were scaled up by integrating qualitative information from interviews with seaweed experts to expand seaweed production, and quantitative information from process simulations to scale up the biorefinery and film fabrication stages. The combination of both methods resulted appropriate to upscale all the processes needed to produce seaweed-based bioplastic.

In terms of global warming (CO₂-eq.), we see how that impact decreased with upscaling the production. The 4000T scenario in 2030 had a carbon footprint of 1.75 kg CO₂-eq. and 1.37 kg CO₂-eq. the 2MT scenario. In both cases accounting for the biogenic carbon in the seaweed cultivation. Not accounting for biogenic carbon led to an increase in the results within the global warming category: 3.85 kg CO₂-eq. in the 4000t scenario and 2.49 kg CO₂-eq. in the 2Mt scenario. The results, therefore,

highlighted the significance of including biogenic carbon in the assessment, where the impacts were significantly lower. It should, hence, be noted the importance of including the biogenic carbon and the carbon uptake. The focus of this sub-question relates to the subject matter covered in Article 3, and additional insights can be accessed in Appendix C.

SUB-QUESTION 4

What are the challenges, opportunities and lessons learnt at each step throughout the value chain and in overall?

This study provided a comprehensive evaluation of seaweed-based bioplastics, examining their supply chain and production processes. Key findings included the potential for extracting multiple co-products from seaweed, such as cellulose, which enhanced the economic viability of the biorefinery. The utilization of seaweed holdfast as a source of high-alginate content was found to be promising, as it avoided competition with other biomass uses and contributed to the valorization of underutilized seaweed resources.

The study demonstrated that both commercial and extracted alginate were suitable for producing high-quality films with desirable mechanical properties. The LCA indicated comparable global warming impacts between crude and dry alginate, as no significant difference was found between the two scenarios. The prospective scenarios showed potential for reducing environmental impact through upscaling and process efficiency improvements.

Seaweed-based bioplastics offers advantages over fossil-based plastics, including biodegradability, reduces reliance on fossil resources, and positive impacts associated with seaweed cultivation. The study highlighted the environmental benefits, the feasibility of the technology to produce the seaweed-based bioplastic and compatibility with existing industrial processes, paving the way for further research and development in sustainable seaweed-based bioplastics.

These findings highlighted the potential benefits of large-scale implementation, where higher production volumes resulted in reduced carbon emissions. For a comprehensive understanding of this sub-question, Article 4 provides relevant context, which can be accessed in Appendix D.

MAIN RESEARCH QUESTION

What are the environmental consequences of the novel brown seaweed-based bioplastic from a consequential life cycle perspective?

The main research question of this study focused on the environmental consequences of the novel brown seaweed-based bioplastic from a life cycle perspective. The findings of the study provided insights into the environmental impacts associated with the production and usage of this seaweed-based bioplastic throughout its life cycle. Through a comprehensive LCA, the environmental consequences of the brown seaweed-based bioplastic were evaluated. The LCA methodology, combined with qualitative research and process simulations, allowed for a holistic analysis, considering all stages of the bioplastic's life cycle, from seaweed cultivation to EoL disposal.

The results of the LCA presented valuable results into the environmental performance of the seaweed-based bioplastic at different scales. In the pilot-scale production, several scenarios were considered. In the scenario without the recirculation of co-products and in the composting end-of-life scenario, including the biogenic carbon, the seaweed-based bioplastic showed the following impacts in the respective categories: 3.55 kg CO₂-eq. in the global warming impact category, -0.003 kg P-eq. in freshwater eutrophication, and -0.017 kg N-eq. in marine eutrophication.

Furthermore, the LCA results showed that the environmental impacts of the seaweed-based bioplastic decreased when upscaling the production. Depending on the scale of upscaling, the impacts in global warming ranged from 1.75 kg CO₂-eq. to 1.37 kg CO₂-eq. This indicates that as the production of seaweed-based bioplastic increases, the overall environmental footprint decreases, suggesting potential improvements in resource efficiency and environmental performance.

However, it is important to note that the LCA results alone do not provide a complete picture. Several other aspects need to be taken into account to fully understand the potential of seaweed-based bioplastic production from brown seaweed. One such aspect was the identification of marginal suppliers, which played a crucial role in assessing the environmental impacts of the production process. Considering the specific characteristics of these suppliers and incorporating their practices into the analysis can provide a more accurate representation of the overall environmental performance.

Additionally, constraints related to the scaling up of seaweed-based bioplastic production should be carefully considered. Upscaling the production process may introduce new challenges and uncertainties, such as increased resource demand, infrastructure requirements, and market demand for the seaweed-based bioplastic. These factors can significantly impact the overall environmental performance and need to be addressed in decision-making processes.

6.2 OUTLOOK

6.2.1 THE PROSPECTS OF SEAWEED-BASED BIOPLASTICS

Seaweed-based bioplastics offer several benefits: they contribute to carbon capture by utilizing renewable resources and have the advantage of not competing with land resources, as they can be derived from fast-growing and regenerative sources. Additionally, seaweed-based bioplastics have the potential to be biodegradable, reducing their environmental impact and contributing to waste management efforts.

However, the prospects of seaweed-based bioplastics offer both challenges and opportunities in the quest for a novel seaweed-based bioplastic alternative. One of the major challenges is the upscaling of seaweed-based bioplastic production to meet a potential future growing demand of this novel product. As the demand for the new seaweed-based bioplastic increases, it becomes crucial to develop efficient and cost-effective seaweed biomass and production methods to produce seaweed-based bioplastics to achieve large-scale production. Another challenge lies in the identification and expansion of potential uses for seaweed-based bioplastics. The properties of seaweed-based bioplastics, such as permeability and water solubility, can limit their application in certain industries. Overcoming these limitations and expanding the range of viable applications is essential for the widespread adoption of seaweed-based bioplastic. Furthermore, competition with other uses of seaweed biomass poses a significant challenge for these seaweed-based bioplastics. Industries such as the food market and pharmaceuticals also rely on the same resources, creating competition for raw materials.

Despite these challenges, there are several opportunities to address them and unlock the full potential of seaweed-based bioplastics. The seaweed-based bioplastic industry can explore innovations and technological advancements to improve production efficiency and reduce costs. Additionally, collaborations and partnerships among stakeholders can facilitate knowledge sharing and accelerate progress in the field.

It is also important to note that while seaweed-based bioplastics offer advantages, they do not solve the entire plastic challenge. The primary goal should be to reduce

overall plastic consumption, promote the avoidance of single-use plastics, and encourage recycling and circular economy practices. Seaweed-based bioplastics serve as an improvement within the current system, providing a better-performing alternative in terms of environmental impact to conventional plastics.

In conclusion, while seaweed-based bioplastics present challenges and limitations, they offer opportunities to address the plastic challenge. Further research and development are needed to overcome current limitations, expand the range of applications, and enhance seaweed-based bioplastic production and usage.

6.2.2 BRIEF OUTLOOK ON PROSPECTIVE AND CONSEQUENTIAL LCA

Looking ahead, there is a need to expand the scope of prospective LCA by incorporating more qualitative research. This is particularly important considering the inherent uncertainties associated with prospective LCA. By integrating qualitative data alongside quantitative information, a more comprehensive and robust assessment can be achieved. Qualitative research methods, such as interviews and expert opinions, can provide valuable insights into future scenarios and help capture important factors that may not be quantifiable. This inclusion of qualitative research in prospective LCA would enhance the accuracy and reliability of the assessments, ultimately contributing to better decision-making.

The application of dynamic LCA, which considers the temporal aspects of environmental impacts, could be considered to further enhance the assessment of the environmental implications. Traditional LCA approaches often assume static conditions, not considering changes over time. By adopting dynamic LCA, the temporal dimension can be incorporated into the assessment, enabling a more accurate understanding of how environmental impacts evolve throughout a product's life cycle. This is particularly relevant in the context of seaweed-based bioplastics, where the decomposition and degradation rates can vary over time, impacting their environmental performance. Additionally, dynamic LCA including the time variable can help capture the long-term effects of carbon sequestration, such as when seaweed-based bioplastics are composted and carbon is stored in the soil.

On the other hand, the inclusion of biogenic carbon in LCIA methods is essential for a comprehensive assessment of the environmental impacts of seaweed-based bioplastics. Including biogenic carbon in LCIA methods ensures that the unique characteristics and benefits of seaweed-based bioplastics, such as carbon sequestration during growth or composting, are properly accounted for. By considering the biogenic carbon, the assessment can provide a more accurate representation of the overall carbon footprint and environmental performance of

seaweed-based bioplastics, taking into account their renewable nature and potential contributions to carbon mitigation.

In summary, these considerations allow for a more comprehensive evaluation that captures the temporal dynamics of impacts and recognizes the unique characteristics and benefits of seaweed-based bioplastics in terms of carbon storage and mitigation. By integrating these aspects into future LCA studies, a more accurate and holistic understanding of the environmental implications of seaweed-based bioplastics can be achieved.

6.2.3 RECOMMENDATIONS FOR FUTURE RESEARCH

In future research, it would be valuable to expand the scope of interviews beyond seaweed experts to include professionals from other industries and stakeholders involved in the upscaling of seaweed-based bioplastics. By engaging with a broader range of experts, including those with expertise in industrial processes, supply chain management, and market analysis, a more holistic understanding of the challenges and opportunities associated with scaling up production can be gained. These interviews can provide insights into the practical aspects of implementing large-scale production systems, address potential barriers, and identify strategies to enhance the feasibility and adoption of seaweed-based bioplastics in the industry. By incorporating perspectives from a diverse range of stakeholders, future research can contribute to the development of a robust roadmap for the successful upscaling of seaweed-based bioplastic production.

Another important progression in the context of future research would involve a more holistic assessment of the seaweed-based bioplastic's sustainability, considering not only its environmental impact but also the economic and social dimensions (Ali et al., 2023). By integrating the economic and social pillars alongside the environmental assessment, a more holistic understanding of the seaweed-based bioplastic's overall sustainability can be achieved. Conducting a techno-economic assessment would incorporate factors such as production costs, market potential, competitiveness, and economic feasibility. By evaluating the cost of seaweed cultivation and harvesting, the expenses associated with the biorefinery process, and the pricing dynamics of the final product, valuable insights can be gained into the economic viability of the technology. On the other hand, integrating social considerations would provide a comprehensive evaluation of the potential benefits and challenges associated with the technology. This comprehensive analysis would guide decision-makers and industry stakeholders, facilitating the transition to more sustainable and economically viable seaweed-based bioplastic production processes.

As part of future research, there is potential to assess the circular economy within the blue bioeconomy of the project. This objective would involve exploring circular

solutions for certain aspects of the blue bioeconomy and evaluating their environmental performance. Specifically, a comparison will be made between the circular economy principles and the production of seaweed-based bioplastics from seaweed, aiming to identify synergies and identify any missed connections from a critical perspective. This qualitative study would have the potential to complement the LCA results and provide valuable insights into the blue bioeconomy and its circularity.

Another aspect to consider in the future in line with the circular economy is the potential use of nitrogen and phosphorus components from seaweed as fertilizers (Seghetta et al., 2016b, 2016a). These valuable nutrients, if not utilized for seaweed-based bioplastic production, can be employed for agricultural purposes. By recirculating the N and P from the seaweed towards fertilizer applications, seaweed-based bioplastic production can contribute to sustainable agricultural practices and nutrient management. Conversely, if the N and P components are incorporated into the seaweed-based bioplastic material, it presents an opportunity for these seaweed-based bioplastics to serve as a means of delivering nutrients to agricultural lands. In their EoL, the seaweed-based bioplastics can release the N and P, enabling the nutrients to be available for crop uptake and supporting the circularity of nutrient flows in agricultural systems. This consideration highlights the potential for seaweed-based bioplastics to not only address plastic waste concerns but also contribute to the efficient utilization of resources in agriculture.

Finally, as future research in a broader perspective, it would be relevant to further assess the potential market share that this novel seaweed-based bioplastic may cover within the entire plastic industry. Understanding the market coverage and acceptance of these seaweed-based bioplastics will provide valuable insights into their application and potential future market demand (Lau et al., 2020). However, it is crucial to acknowledge the limitations associated with these types of seaweed-based bioplastics like the ones previously discussed. These limitations may include factors such as restricted applicability or challenges in meeting specific performance requirements, which could influence their widespread adoption in various industries. Assessing these limitations and potentials more in detail would be essential in determining the feasibility and scalability of these seaweed-based bioplastics as viable alternatives to conventional plastics (Lau et al., 2020; PEW Charitable Trusts and SystemIQ, 2020).

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APPENDIX A

ARTICLE 1

Using quantitative story telling to identify constraints in resource supply: The case of brown seaweed

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Title: Using quantitative story telling to identify constraints in resource supply: The case of brown seaweed

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Keywords: Industrial ecology, Consequential Life Cycle Assessment, Marginal suppliers, Bioplastic, Blue bioeconomy, Macroalgae.

Abstract: Seaweed is increasingly considered a promising resource to produce high-value products such as bioplastics due to potential environmental benefits such as carbon uptake and no land-use change. However, the environmental assessment of emerging technologies for producing bioplastic from seaweed remains challenging due to the difficulties in modeling future seaweed supply and demand. Within the consequential approach to Life Cycle Assessment (LCA), an increase in demand for seaweed is met by the marginal suppliers in the market: those that are not constrained in their capacity to increase supply in response to an increase in demand. Current methods to identify marginal suppliers are however based on quantitative information and do not consider qualitative aspects and uncertainties inherent in the study of emerging technologies. This study, therefore, proposes and tests the use of quantitative story telling to identify marginal suppliers. The results show that there are two main groups of countries that are expected to be the marginal suppliers of brown seaweed for different reasons. Asian countries are currently the main brown seaweed suppliers and are expected to keep increasing and be marginal suppliers in the future. However, these countries have well-established brown seaweed aquaculture and their growth is expected to be steady. On the other hand, brown seaweed suppliers in Northern Europe and North America are still emerging but are expected to grow faster in the future due to their production capacity and technological development.

1. INTRODUCTION

The concept of blue bioeconomy, defined as an economy based on aquatic renewable bioresources, is gaining momentum and seaweed is among the most important feedstocks (Addamo et al., 2021). Seaweed aquaculture can potentially bring environmental benefits such as providing ecosystem services (Thomsen & Zhang, 2020), contributing to global carbon, oxygen and nutrient cycles (Brodie et al., 2014; Seghetta et al., 2016; Visch et al., 2020) or reducing eutrophication (Xiao et al., 2017). Additionally, seaweed does not compete with other land-use resources and, hence, there is no land-use change, use of fertilizers, pesticides, or freshwater involved in seaweed aquaculture, like in other bioresources (Duarte et al., 2017, 2021; Giercksky & Doumeizel, 2020; Hasselström et al., 2020).

The global seaweed production was 35,7 million tons of wet biomass in 2019, with human consumption being the main market (Cai et al., 2021), which poses a potential limit to how much of existing seaweed supply can be used for the production of bioplastic. Brown seaweed production grows annually by approximately 11%, from 13.000 tons in 1950 to 17,6 million tons in 2021 (Cai et al., 2021). Currently, 96,5% of the total global production of brown seaweed comes from aquaculture (FAO, 2023), while in Europe comes primarily from wild harvesting (Araújo et al., 2021; Stanley et al., 2019). There is a recent increasing interest in seaweed aquaculture (Peteiro et al., 2016), especially in Europe (Bak et al., 2018; van den Burg et al., 2016), to meet the future increase in demand (Stévant et al., 2017).

In recent years, the use of seaweed has been assessed as a potential source to produce bioplastic (Carina et al., 2021; Lim et al., 2021; Sudhakar et al., 2021; Zanchetta et al., 2021; Zhang et al., 2019). In the transition to a bio-based economy, bioplastics are a potential solution to reduce the dependence and extraction of fossil resources (Bishop et al., 2021; Spierling et al., 2018). First-generation and second-generation bioplastics, made from edible crops and plants and agro-industrial waste respectively, require intensive use of land and water, contributing to direct and indirect land-use change (Brizga et al., 2020; Ita-Nagy et al., 2020). The use of food crops for bioplastic production is questionable due to the risk of creating competition with the food market, which could lead to higher food prices and food insecurity. Third-generation bioplastics are still under development and relate to the use of living organisms to produce plastics not using land

resources (Brizga et al., 2020), including seaweed. Seaweed-based bioplastics potentially offer a more sustainable alternative compared to other type of bioplastics, as seaweed is a renewable resource that does not require arable land for its supply.

Novel brown seaweed-based bioplastic is considered an emerging technology from a Life Cycle Assessment (LCA) perspective. If this emerging technology is implemented, it is important to understand which suppliers will respond to a future increase in demand for seaweed, to anticipate the impact that will be induced by such demand (Bergerson et al., 2020; Blanco et al., 2020; Giesen et al., 2020). In a consequential LCA approach those suppliers are referred to as marginal suppliers (Consequential-LCA, 2020; Pizzol & Scotti, 2017; Weidema et al., 1999). In theory, this identification is intended to be an analysis of the possible constraints to increasing the supply of a specific product. In practice, however, the methods rely on a combination of assumptions and modeling, e.g. regression analysis and trade statistics (Buyle et al., 2018; Sacchi, 2018; Weidema et al., 1999). These quantitative methods have mainly been applied to established technologies with a relatively large amount of data available, fully addressing the challenges related to the assessment of emerging technologies, including data scarcity and uncertainties in forecasting future scenarios.

Current methods to identify marginal suppliers do not apply well to the case of brown seaweed because data on brown seaweed supply is scarce and unreliable. There are two main statistic sources for seaweed production: The Joint Research Centre (JRC, 2021) for brown seaweed statistics in Europe and the Food and Agriculture Organization (FAO, 2023) for global statistics. However, countries have no legal requirements to publish precise data, the available data on brown seaweed is, therefore, incomplete (Araújo et al., 2021). Studies that attempt to make future projections on brown seaweed supply are qualitative or based on simple extrapolations (Duarte et al., 2021). While qualitative information can serve as a starting point in identifying marginal suppliers, many constraints to upscale the production can only be identified qualitatively, e.g. the current state of emerging trend in optimal growth conditions, including locations and cultivation designs, regulatory regime shift and technological development.

Summing up, a better framework to identify marginal suppliers for emerging technologies is needed, that can handle the intrinsic uncertainty, and combine both quantitative and qualitative information

on existing and emerging development of the brown seaweed production and brown seaweed-based bioplastic market.

In this context, the objective of this study is to propose the use of quantitative story telling (QST), a mixed-methods approach proposed by Saltelli and Giampietro (2017) for supporting policy and decision-making in the sustainability domain, to identify marginal suppliers of brown seaweed. We investigate the global brown seaweed market and determine the marginal mix of suppliers to use in consequential LCA. The marginal mix represents the relative share of suppliers with a positive growth rate in the total supply. Combining qualitative and quantitative information, QST is used to investigate the current market trends and the consequences of increasing demand for brown seaweed to produce bioplastic. We also evaluate challenges in upscaling reflecting on the marginal supply, including production capacity, technology development, regulatory constraints and the market niche for brown seaweed bioplastic. This research contributes with new methodological insights on the identification of marginal suppliers of emerging technologies, where uncertainty and qualitative information play an important role.

2. METHODS

The quantitative story telling (QST) approach has been recently proposed by Saltelli and Giampietro (2017) as a complementary approach to traditional evidence-based policy. QST explores systematically the multiplicity of frames that are potentially legitimate in a scientific study. It assumes that in an interconnected society, multiple frameworks and worldviews are legitimately upheld by different entities and social actors (Saltelli & Giampietro, 2017). More recently, Cabello et al. (2021) propose a six-step procedure to further operationalize QST. In this study, we adapted the procedure proposed by Cabello and co-workers to our case (Figure 1) and followed the six steps to identify brown seaweed marginal suppliers: 1) Map actors, 2) Identify narratives, 3) Quantify nexus relations, 4) Link qualitative & quantitative information, 5) Participatory assessment of narratives and 6) New narratives.

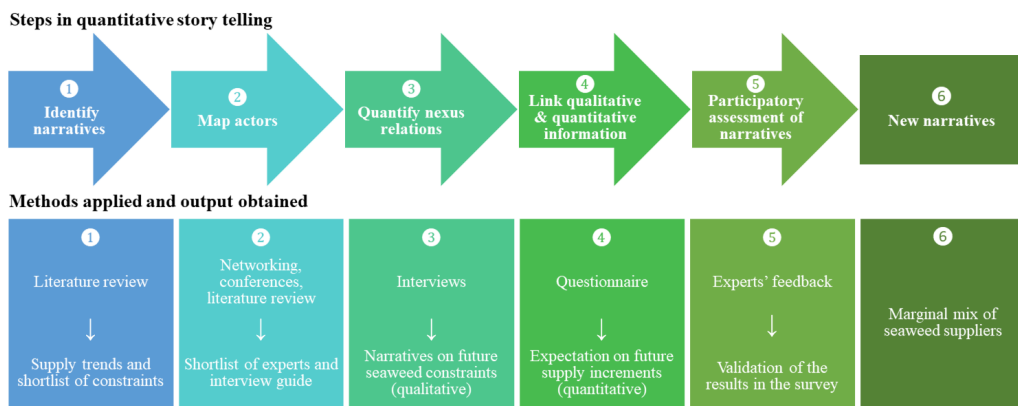


Figure 1: Steps in quantitative story telling as described in Cabello et al. (2021) and corresponding methods applied in this study.

Step 1: Identify narratives

A review of existing literature was first conducted to shortlist typical constraints to supply that are often considered in consequential LCA studies. Initially, we focused on the LCA literature and used various boolean combinations of the keywords: “Consequential”, “LCA”, “Marginal suppliers” and “Constraint” to identify scientific articles performing marginal suppliers’ identification in general, then described the type of data used in such studies, the processing applied to the data, the constraints considered and the research gaps. Two main sources were particularly relevant: Buyle et al. (2018) provided an overview of existing consequential studies and Weidema et al. (2009) introduced a series of possible constraints. Based on these, other relevant references were identified (Buyle et al., 2018; Ekvall & Weidema, 2004; Frischknecht & Stucki, 2010; Ghose et al., 2017; Lund et al., 2010; Pizzol & Scotti, 2017; Sacchi, 2018; J. Schmidt & de Rosa, 2020; J. H. Schmidt, 2008; J. H. Schmidt & Weidema, 2008; Thonemann & Pizzol, 2019; Weidema et al., 1999, 2009; Wernet et al., 2016). The identified scientific articles were organized in a table, including the main method used to identify marginal suppliers and the considered constraints. The literature review table is provided in Supporting Information S1. As a result of this literature review, we concluded that the constraints most typically considered in consequential LCA studies are *geographical market delimitation; market trend; most sensitive suppliers to change; production capacity; and technology development*.

We focused on the literature on brown seaweed and retrieved information on the global state of seaweed harvesting and farming, growing conditions, production methods, aquaculture, spatial planning and economic feasibility. Statistics on brown seaweed suppliers from FAO and JRC were also accessed. Seaweed cultivations appear to be geographically constrained by environmental conditions and country-specific regulations. As a result, two more constraints were included to identify brown seaweed marginal suppliers: *policies and regulatory constraints* and *natural constraints*. Even if the constraints were not explicitly considered in current quantitative consequential studies to calculate the marginal mix, we evaluated these as necessary to understand the brown seaweed market and its future development.

Summarizing the results of the review on marginal suppliers and brown seaweed, the identified constraints were grouped into four main constraints based on the information they provide. *Market trends* and *most sensitive suppliers to change* were grouped under *geographical market delimitation* considering that they provided the same information regarding the current status and future projections of the global brown seaweed supply market. *Natural constraints* and *production capacity* were grouped as they both give information about productivity. The last constraint is *technological development*, which included upscaling. These constraints can potentially condition the future brown seaweed supply.

- *Geographical market delimitation*: Market trends, market boundaries, predominant brown seaweed suppliers and market prospects.
- *Policies and regulatory constraints*: Regulations constraining the brown seaweed trade and market.
- *Natural constraints and production capacity*: Productive brown seaweed species and environmental parameters and techniques to increase productivity.
- *Technological development and upscaling*: Technological prospects to upscale brown seaweed production.

The four constraints were used as a starting point to design the interviews with experts in the seaweed domain (cf. *Step 3: Quantify nexus relations*).

Step2: Map actors

Experts in brown seaweed were identified using information from the literature, participation in conferences on seaweed science and contacts in the authors' research network. The list of experts included phycologists, experts in seaweed aquaculture, the Asian and European seaweed market, seaweed farmers and harvesters, and experts with knowledge of bioplastics. In the selection of the interview sample, experts from different countries and both academia and the private sector were targeted.

An interview guide was prepared (cf. Supporting Information S2), organized into four blocks of questions: one block for each constraint plus a fifth block of questions to focus on the use of seaweed for producing bioplastic. The questions about market delimitation and geographical market boundaries included questions about China's success in the past, present and future, other potentially predominant future suppliers and the prospects of the brown seaweed supply. The respondents were asked to reflect on whether brown seaweed is a local or global market and the limitations and possibilities of trading seaweed. The productivity, environmental and technical parameters conditioning the seaweed growth were also explored. The questions on technological development explored upscaling the production in the future, technological development and learnings from predominant suppliers. Finally, the questions on seaweed-based bioplastic were about the potential species for bioplastic production, the importance of the proximity of bioplastic production facilities to the seaweed farms and competition with the current main target markets.

Step 3: Quantify nexus relations

Using the interview guide and the shortlisted experts, interviews were conducted to obtain the most relevant qualitative information to identify the main brown seaweed suppliers. A total of 11 experts with different expertise and nationalities were interviewed: six academics, three seaweed farmers, a senior researcher and a policy maker. Interviewees were from Denmark, Norway, Sweden, the Faroe Islands, France, the Netherlands, the United Kingdom, Portugal, Korea and China. The interviews were held online and recorded with the consent of the interviewees. The interviews were then transcribed and analyzed using a matrix approach. The first column included all quotes from the interviews and the first row included the interview questions. In each quote/question intersection cell, a "condensed meaning" of the quote (e.g., the most relevant keywords) were then extracted and the keywords were written under their corresponding question. In the case of having

quotes corresponding to various questions, they were coded under different questions. With this method, we could filter the answers to each question. The answers were summarized systematically and key quotes with exemplary or recurring information were extracted (cf. Supporting Information S3). In the case of having similar quotes providing the same information, only the quotes with more complete information were used in the summary of the interview. Afterward, we summarized the answers based on the constraints (cf. Results section). The result was a narrative about each future constraint to brown seaweed production.

Step 4: Link qualitative and quantitative relations

LCA is a quantitative assessment tool and quantitative information is needed in the modeling process. In this step, the interviewees were contacted in a second round for a short expert elicitation questionnaire. Five of them were willing to contribute and thought could provide meaningful answers. The aim was to obtain quantitative projection estimates of production increments to define a marginal mix of seaweed suppliers needed in consequential LCA modeling.

All the countries mentioned during the interviews as possible future brown seaweed suppliers were listed in the questionnaire. First, the experts were asked to select from the list of countries the most competitive suppliers in the middle-long term: between 2025 and 2035. Afterward, they were asked to estimate the growth rate in the brown seaweed supply from each of the selected countries. The following questions were used to estimate the marginal mix of brown seaweed suppliers in 2025, 2030 and 2035. The last question referred to the expected market shares of brown seaweed for bioplastic in 2025, 2030 and 2035. The template of the expert elicitation questionnaire with the specific questions and answers options can be found in Supporting Information S4.

Step 5: Participatory assessment of narratives

To validate the results obtained in Step 4, a validation questionnaire was conducted with the experts who participated in the interview (Step 3) and the expert elicitation questionnaire (Step 4). In addition, a broader group of experts was contacted in person in a conference in seaweed science to obtain different and more representative perspectives. The aim of this validation questionnaire was to assess the level of agreement of the experts with the findings about marginal suppliers. In the validation questionnaire, the results on the marginal suppliers were

presented as a starting point for the questions. The experts were asked to express to what extent these results matched their expectation, on a five points Likert scale ranging from "Very poor match" to "Excellent match". The experts were also asked a follow up question to identify the variable that they found most uncertain. The three variables provided as options were: the listed countries, the percentage of shares and the scenario trends. Both multiple-choice questions were accompanied by an open text question box giving the opportunity to elaborating on the provided answer. The validation questionnaire template and the answers are presented in Supporting Information S5.

Step 6: New narratives

We analyzed quantitatively the results of the expert elicitation questionnaire. The countries that had been selected by at least 60% of the respondents were considered and calculated their average estimated growth rate across respondents. It was calculated as in equation (1):

$$\text{Average annual growth rate} = \frac{\text{Growth rate given by respondents}}{\text{No. of respondents}} \quad (1)$$

Based on the obtained average growth rates, we then derived a marginal mix by calculating the percentage of each supplier to the total supply. In the expert elicitation questionnaire, the experts were asked to directly provide their estimate of the marginal mix (cf. Supporting information S4). Equation (2) was then used to calculate the marginal mix:

$$\text{Marginal mix (\%)} = \frac{\sum \text{Respondent marginal mix (\%)}}{\text{No. of respondents}} \quad (2)$$

Respondent marginal mix is the marginal mix estimated by each respondent in the questionnaire. Following these steps, the marginal mix of marginal suppliers was obtained. This marginal mix will form the basis for consequential LCA of a transition into seaweed-based bioplastic.

Finally, the average market share for bioplastics was calculated. Like the marginal mix, the experts were asked to estimate the annual share of the total production of brown seaweed supply for bioplastic. We calculated the average market share for bioplastic according to equation (3) to calculate the average market share for bioplastics:

$$\text{Average market share for bioplastics (\%)} = \frac{\sum \text{Respondent market share (\%)}}{\text{No. of respondents}} \quad (3)$$

Respondent market share is the expected market share for bioplastic from the total brown seaweed production volumes. All data and calculations are provided in Supporting Information S6.

3. RESULTS

3.1. Geographical market delimitation

China is the main global brown seaweed supplier. China's success can be explained due to its long tradition cultivating and consuming seaweed, knowledge of seaweed aquaculture and breeding species to obtain high yields, large demand and an established market. Most experts agreed that Chinese seaweed producers will be front-runners at least in the next decade and even if European production is expected to grow, it will not overtake Asian production volumes.

The interviewees made a high-growth projection of brown seaweed production. The main driver of the expected increase in brown seaweed demand is an increasing global interest in using seaweed for different applications and the need for new resources in the coming years, bioplastic being one potential new market. Another driver to cultivating brown seaweed mentioned by experts was the environmental benefits of seaweed cultivation, including ecosystem services and carbon capture. Technological development in brown seaweed aquaculture was mentioned as another driver, including selective breeding and mechanizing the production process to optimize productivity and area utilization.

When interviewees were asked about brown seaweed suppliers in the future, all of them agreed with the potential of Norway to become a predominant brown seaweed supplier due to its favorable environmental conditions and technological development. Another factor is Norway's aquaculture and offshore technology industries which can support the upscaling and industrialization of seaweed cultivation. Atlantic European countries and islands were the most repeated potential future suppliers by experts. These included the Faroe Islands, Iceland, Greenland, Ireland, the UK, the Netherlands, Belgium, France, Sweden, Denmark, Portugal and Spain. Other countries outside Europe were considered: The United States, Canada, Chile, Australia, Korea and Japan. A few experts named New Zealand, Namibia and Kenya.

3.2. Policies and regulatory constraints

Experts agreed that seaweed is a global free market and there are no regulatory limitations for the global seaweed trade and dry seaweed can easily be transported. Some experts mentioned that even if seaweed can be traded globally, regulations are needed to ensure the quality of the seaweed.

Buying local or imported seaweed is market dependent. Asian countries already have a large internal brown seaweed demand for food applications and the volumes of seaweed that they export are small. In Europe, consumers are more inclined to buy locally, mainly for food safety and sustainability reasons. The international trade of non-food applications is less constrained than the food and feed market, and it is an increasing market. According to the interviewees, seaweed origin is less significant for non-food uses.

In some countries, obtaining permits to cultivate seaweed is another regulatory constraint. Moreover, in most countries, it is required to cultivate native species. The regulations to cultivate seaweed in Asia are more flexible.

3.3. *Natural constraints and production capacity*

To upscale brown seaweed production, various aspects regarding natural constraints and production capacity need to be considered. First, the selection of the most productive species. Different species are cultivated globally, but *Saccharina latissima*, *Saccharina japonica* and *Macrocystis pyrifera* are the fastest growing species according to brown seaweed experts. The farm site selection with favorable environmental conditions is important to cultivate seaweed. The main environmental parameters that condition brown seaweed growth are a combination of cold water, nutrient and light availability, proper salinity, a big sea area, wave activity and tidal zone differences. There are different techniques to increase productivity and upscale brown seaweed production. Some experts highlighted the importance of seaweed cultivation instead of wild harvesting to increase production volumes and preserve the local marine environment. Selective breeding was repetitively mentioned as a well-known and popular technique to naturally obtain the most productive seaweeds.

3.4. *Technological development and upscaling*

Some experts mentioned other emerging technologies to upscale seaweed production, such as offshore seaweed farms, together with already established activities, such as wind farms, seeding

lines between windmills, or submersible farms to resist adverse environmental conditions. Technological development would ease seaweed production and the shared infrastructure will make it more economically feasible. Experts mentioned that technological development could entail Western countries achieving similar production volumes to Asia. Limiting factors to upscale include high-production costs, the limiting carrying capacity of marine ecosystems, social licensing, sea space limitation, the vulnerability of monoculture, environmental laws constraining seaweed cultivation and the lack of an established market for the entire value chain in Western countries. Asian suppliers have a long tradition and there are, therefore, different practices for smaller suppliers to learn from them. Breeding techniques, for instance, are highly developed in Asia. In general terms, experts answered that current Asian intensive farming practices cannot be directly applied in Europe. The reasons are mainly environmental protection, social licensing, supply chain transparency and the differences in harvesting techniques. Asian suppliers use traditional and labor-intensive techniques, which is unfeasible in Europe due to high labor costs making automation necessary.

3.5. *A new market for seaweed-based bioplastics*

All brown seaweeds can potentially be used to create alginate-based bioplastic due to their alginate content. When it comes to selecting the most suitable species for this purpose, the key factors are the alginate content and the growth rate of the species. Therefore, *Saccharina latissima* and *Saccharina japonica* were mentioned by experts as the most suitable species to cultivate and harvest as feedstock for bioplastics.

To know the importance of having the facilities to produce bioplastic close to the seaweed farms, it is important to know the quality of brown seaweed required to create the bioplastic. Even if dry seaweed can be easily transported globally, most experts mentioned various advantages of having seaweed facilities close to seaweed farms. The main advantages are reducing transportation costs and preserving the biomass to maximize the recovery of high-value components from the biomass. These two reasons could make a difference for bioplastics because large volumes and at a competitive price are used to create this biomaterial.

Experts agreed that food is the main target market for brown seaweed, followed by feed, pharmaceuticals, cosmetics, fertilizers and the alginate market. Other materials, including

bioplastics, would be one of the lowest-ranked target markets. The potential of using brown seaweed to produce bioplastics is a matter of how much the market is willing to pay for biomass. Therefore, most of the experts did not see a potential for the current production to shift towards the bioplastic market because it is not as economically profitable as higher-value applications.

There are various potential scenarios for using brown seaweed biomass to produce bioplastic. When the brown seaweed production upscales and the volumes increase, the production prices would gradually decrease. In some decades, depending on investment in research and development to increase the TRL of seaweed plastic production, it could be economically feasible to produce bioplastics from brown seaweed. Another potential solution would be using seaweed of lower quality compared to that used for human consumption. This lower-grade seaweed is collected during the second harvest to achieve higher yields and is associated with an increased risk of biofouling, that may not meet the standards for human consumption but can still be utilized for bioplastic production.

Combined production methods or sequential extraction were also mentioned by some experts as another opportunity for bioplastics. Other high-value compounds would be used for other purposes and the alginate for bioplastic (X. Zhang & Thomsen, 2021). In this case, the seaweed would be cultivated to capture Carbon and Nitrogen and the seaweed is not harvested as frequently as when targeted for use in human consumption. Hence, this seaweed could be used to produce bioplastics.

Some experts foresee the importance of working with bioplastics as a consequence of moving away from fossil fuels. There is a possible scenario for prioritizing bioplastics if the prices for conventional plastics increase or with potential government incentives for companies adopting biopackaging. The extended information on all the interview answers, including relevant quotes, can be found in Supporting Information S3.

3.6. Identified marginal mix for global brown seaweed suppliers

When the five experts were asked to select the main brown seaweed suppliers in the middle- long-term, between 2025 and 2035, all of them selected China, Norway and South Korea; four Japan; and three Chile, Iceland and the United States. It is worth mentioning that Australia, Canada, the Faroe Islands, North Korea and the United Kingdom were selected by at least two experts as main suppliers

in the middle-long term. The complete results with the answers can be found in the Supporting Information S6.

Figure 2 represents the expected annual growth rate of the marginal suppliers. The graph shows that the largest expected annual growth is from Northern European and North American countries, followed by Chile and then the Asian seaweed producers.

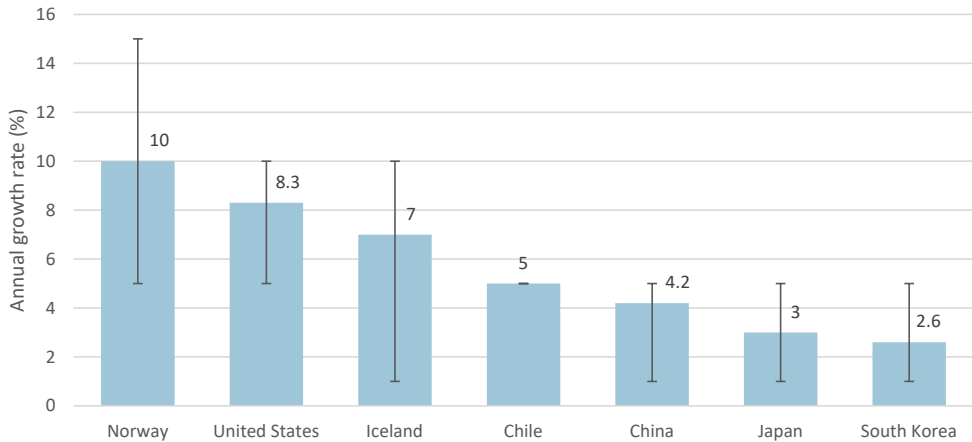


Figure 2: Expected annual growth rate (%) in a medium-long term. The values are averaged across respondents. In the error bar: the minimum and maximum value provided by experts.

Table 1 displays the expected marginal mix in 2025, 2030 and 2035. The mix shows that an increase in demand for brown seaweed is primarily met by China as the dominant marginal supplier and then by the second group of important suppliers (South Korea, Norway) and in minor share by other emerging suppliers. The result also shows that over time the marginal supplier mix is expected to change with a larger contribution from the emerging suppliers and a decrease in the contribution from the incumbent suppliers.

Countries	Marginal mix in 2025	Marginal mix in 2030	Marginal mix in 2035
China	73% (40-89)	69% (30-84)	65% (25-80)
South Korea	10% (2-20)	11% (2,5-20)	10% (2-19)
Norway	3% (0,5-5)	6% (2-15)	10% (2-25)
Japan	8% (1-30)	6% (2-20)	5.5% (2-15)

Chile	4.5% (5-10)	5% (5-10)	5% (5-8)
United States	1% (1-3)	2% (1,5-5)	3% (2-8)
Iceland	0,5% (0,5-1)	1% (1-2)	1,5% (1-4)
Total	100	100	100

Table 1: Marginal mix of global brown seaweed suppliers in 2025, 2030 and 2035 (ranked from larger to smaller in 2035). Values are averaged across respondents. In parenthesis: the minimum and maximum value provided by experts.

The results on the market share for bioplastics are listed in Table 2 shows the total production of brown seaweed expected to be allocated to bioplastics. There is no substantial difference in the share of bioplastics between the suppliers. The production is expected to grow significantly in the long term and, according to these numbers, with an upscaled production in 2035, the market share for bioplastics will be larger than in the middle term.

Countries	2025	2030	2035
Chile	1% (1-1)	5% (5-5)	10% (5-20)
Iceland	1% (1-1)	3.7% (1-5)	8.7% (1-20)
United States	1% (1-1)	3.7% (1-5)	8.7% (1-20)
Norway	2.6% (1-5)	5.2% (1-10)	8.2% (1-20)
South Korea	1% (1-1)	4.2% (1-5)	8.2% (1-20)
China	1,8% (1-5)	3.4% (1-5)	5.2% (1-10)
Japan	1% (1-1)	3% (1-5)	4.3% (1-10)

Table 2: Expected market share for bioplastics (ranked from larger to smaller in 2035). Values are averaged across respondents. In parenthesis: the minimum and maximum value provided by experts.

3.7. Validation of results

The responses to the validation questionnaire indicated that most of the experts considered the identified marginal suppliers as a fair match to their expectations. Specifically, most participants indicated a “fair match”, one participant indicated a “good match”, with only one participant rated the match as “excellent”. No participants rated the match as “poor” or “very poor”.

The most uncertain variable identified by the experts was the percentage of shares, with some experts pointing to uncertainties in identifying marginal suppliers as the main reason. Some experts noted that Norway had a bigger share than expected, while others expected other European countries to appear in the list of marginal suppliers. Additionally, there was a general opinion that Asian suppliers would have a slightly bigger share in the future than the one anticipated in the results. In general, the predictions about growth rates were viewed as uncertain. The insights from the validation step indicate that while there is a general agreement among experts that the identified marginal mixes are valid to a fair to good extent, there is not a full agreement among experts regarding the specific shares of each supplier and the assessment is characterized by uncertainties. This result was to some extent expectable as the shares are obtained averaging the estimates of different experts and predictions about the future are intrinsically uncertain and, therefore, all results are here provided with ranges.

4. DISCUSSION

In general, we observe a good alignment between interviews, both questionnaires and literature information, and this suggests that the proposed method provides results that are sufficiently robust for the case of brown seaweed. The expert elicitation questionnaire results (step 4) indicate that Northern European and North American countries are expected to grow in the next years. That aligns with the experts' insights: these countries are still in the early stage of brown seaweed cultivation but have the necessary tools to upscale the production. The literature and the interviewed experts agree that Norway has the technology, infrastructure, knowledge and the optimal environmental conditions to increase the production (Broch et al., 2019; Handå et al., 2013; Stévant et al., 2017; van den Burg et al., 2021). Chile is also expected to keep growing in the future. Currently, it is one of the main brown seaweed producers and its production is mainly based on wild harvest. In the interviews and the literature, we see that a transition toward aquaculture is necessary to upscale Chilean production (Buschmann et al., 2008). The Asian countries are expected to have a smaller growth rate than the European and American countries. Based on the interviews and the literature (Hurtado et al., 2019; Kim et al., 2017; J. Zhang, 2018), this can be explained because the Asian countries have an established market, intensive production, many exploited areas and a high internal demand. internal brown seaweed demand (Hu et al., 2021; Hwang et al.,

2019). China is foreseen to be the main supplier, but the rest of the countries are expected to increase their future share in the marginal mix.

The marginal mix results can be used in consequential LCA studies for brown seaweed-based products, such as bioplastic. The marginal mix is intended to develop country-specific inventories of seaweed farming technologies, for the listed countries in the mix and combine them based on the proportions provided in this study. For instance, for a reference flow of 1 kg of seaweed demanded globally, 0.73 kg of seaweed is produced in China (cf. Table 1).

Although some limitations to the methodology can be discussed, the results of this study were validated in several means. It can be argued if 11 interviewees are enough to obtain valid results and how it affects the quality of the results. The selected experts for the interviews and both questionnaires were highly qualified and diverse in their backgrounds, providing a solid foundation for valid information. The focus was on selecting the right experts over receiving many respondents. After a few interviews, a satisfactory level of data saturation was reached. Moreover, the results from the interviews were triangulated with the answers from both questionnaires, validating the questionnaires' answers likewise. Therefore, by selecting different seaweed experts, the results would be very similar and reproducible to a good extent. Regarding the analysis of interview data, the answers to the questions were summarized systematically (cf. Supporting Information S3) and it is likely that practitioners analyzing the interviews to derive the same results. Another limitation comes with the questionnaire on step 4, as experts found it challenging to make accurate predictions given the uncertainty about the future. Experts were instructed to state their expectations based on their current level of knowledge on the topic and we acknowledge the limitations of an expert-based assessment. The validation questionnaire (step 5) helped to ensure the reliability and accuracy of the findings. The idea behind this research is that market trends are also based on guesses and, in the case of brown seaweed, there are other factors to consider that are not reflected in the market analysis.

This method is intended to be used in the assessment of emerging technologies and conditions of data scarcity and high uncertainty. The proposed mixed qualitative/quantitative methods approach might not apply to established technologies and where larger amounts of quantitative data are available. In those cases, quantitative methods might be more appropriate (Buyle et al., 2018; Ghose

et al., 2017; Sacchi, 2018; J. H. Schmidt & Weidema, 2008). However, this expert prediction-based method is appropriate for identifying and understanding marginal brown seaweed suppliers.

5. CONCLUSION

In this research, we investigated how quantitative story telling can be applied to identify marginal suppliers in the case of brown seaweed. The main theoretical highlights from the study are that current methods to identify marginal suppliers do not properly address the complexity of emerging technologies where uncertainty is substantial, data are scarce and there is a need to rely on and combine systematically both qualitative and quantitative information. For these reasons, existing methods do not apply well to the case of brown seaweed production intended as an emerging technology. We conclude in this study, that the steps in the method of quantitative story telling, proposed by Cabello et al. (2021), can be applied successfully to identify marginal suppliers in cases of scarce quantitative data or when qualitative information is essential to consider.

In terms of novelty for seaweed and LCA research, this research goes beyond previous studies. On the one hand, it gathers data on brown seaweed supply, such as the global production, market, natural constraints, production, technology to upscale the production and provides novel information on using brown seaweed to produce bioplastics. Finally, novel information on brown seaweed marginal mix is provided which can be used to identify marginal suppliers in consequential LCA studies.

The results show that China is the main marginal supplier and is expected to be the marginal supplier in the middle- long term. We also see that seaweed aquaculture is key to upscale brown seaweed production because it enables obtaining larger biomass volumes. The brown seaweed suppliers in Northern Atlantic, still in an early development stage, are forecasted to increase significantly in the middle- long term. According to our results, a large market share for brown seaweed bioplastic is not feasible in a short term given that the price of the seaweed for this purpose is lower than for other applications. There are different options to increase the market share of bioplastics in the current market. Findings suggest that the use of brown seaweed to produce bioplastics could be a co-product of a pricier target markets. We identify different alternative scenarios to materialize this outcome: using the parts of the seaweed not destined for food, using the lower quality seaweed that is not suitable for consumption, or a multi-functional biorefinery model where the alginate is

used to produce this bioplastic and the remaining components for other purposes. With the expected global growth in brown seaweed production, the market share for bioplastics is expected to be higher in the long term. We conclude by recommending these scenarios and production factors to be considered in future LCA studies of seaweed-based products.

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SUPPORTING INFORMATION

Supporting Information

Supporting information is linked to this article on the *JIE* website:

Supporting Information S1: This supporting information provides the literature review on marginal suppliers (Step 1).

Supporting Information S2: This supporting information contains the interview questions (Step 2-3).

Supporting Information S3: This supporting information includes the summary of interview answers (Step 3).

Supporting Information S4: This supporting information provides the expert elicitation questionnaire template (Step 4).

Supporting Information S5: This supporting information contains the validation questionnaire (Step 5).

Supporting Information S6: This supporting information contains the analysis of the expert elicitation questionnaire (Step 6).

APPENDIX

APPENDIX B

ARTICLE 2

Life Cycle Assessment of pilot scale production of seaweed-based bioplastic

Maddalen Ayala, Marianne Thomsen, Massimo Pizzol

The article is published in the *Algal research* journal.

Note: An error was found in the Article. A corrigendum is therefore attached to the article.

Corrigendum to “Life Cycle Assessment of pilot scale production of seaweed-based bioplastic”

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The authors regret to inform that a mistake was found in the Life Cycle Inventory (LCI) of this study. Consequently, the identified error has implications on the results reported in the original publication. The amount of crude extract from the biorefinery needed in the film fabrication step was underestimated. More specifically, in the original article 1.223L of crude were reported to produce 3.75kg of bioplastic, and the corrected amount is 9.058L for a kilogram of bioplastic. For that reason, the contribution in the Global Warming impact of the biorefinery, offshore farm and hatchery stages respectively is higher than in the original article, and the recirculation of mannitol shows lower impact compared than the base scenario. Besides these differences all other main conclusions of the study remain unchanged: the highest contributor to the global warming impact remains the film fabrication stage due to the use of glycerol; recirculation of cellulose still does not allow for impact reduction, and consistently across all scenarios the composting end-of-life option has still the lowest impact.

The updated results are provided in the following:

3.1 Carbon balance

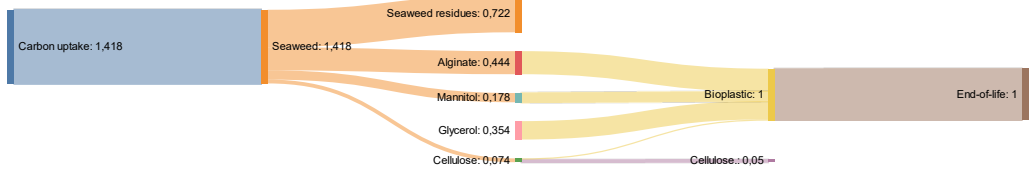
(1)



(2)



(3)



(4)

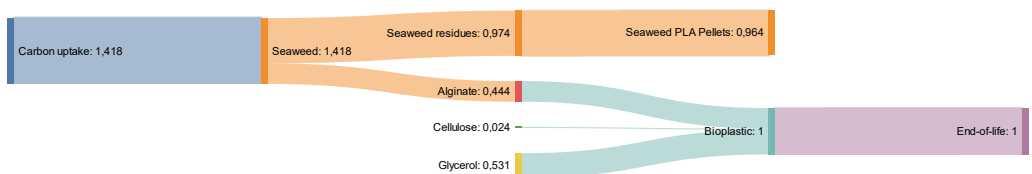


Fig. 2: Sankey diagrams of the carbon balance. Base scenario, BASE (1); cellulose recirculation scenario, CELL (2); and cellulose and mannitol recirculation scenario, MANN (3); PLA substitution 5% and 30%, PLA5 and PLA30 (4). Units: kg carbon.

3.2 Scenarios and contribution analysis

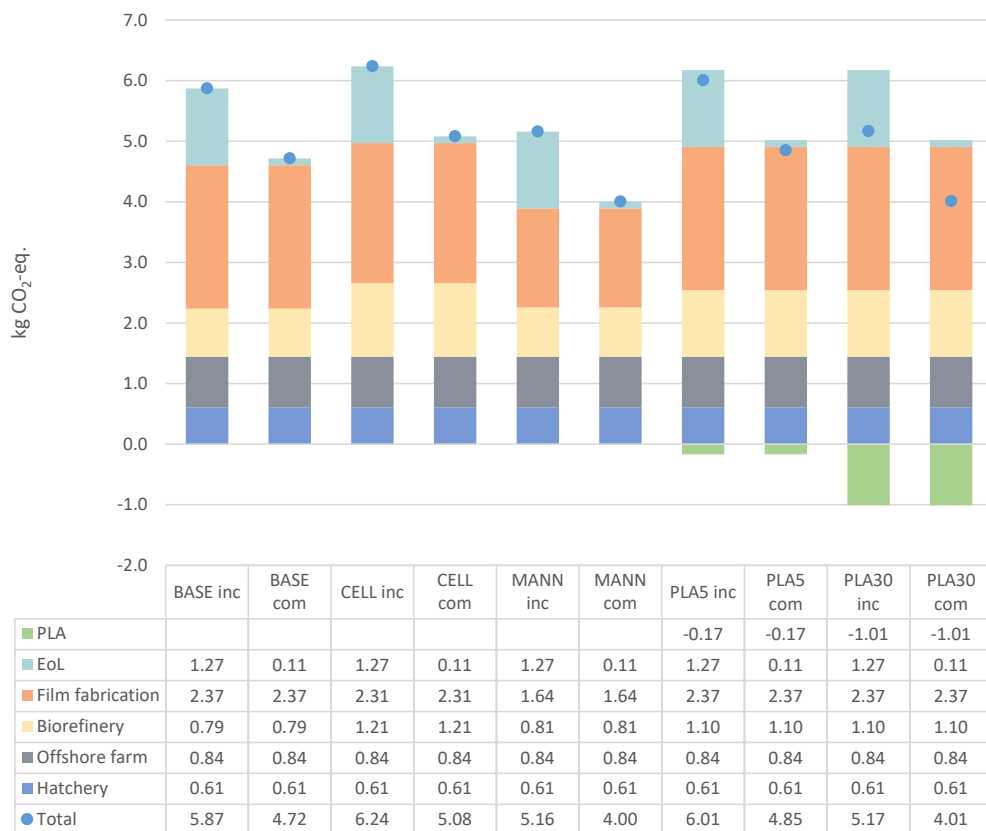


Fig. 3: Contribution analysis in the impact category Global Warming (GW) impact of all the scenarios and sub-scenarios: BASE, CELL, MANN, PLA5, PLA30, incineration (Inc.) and composting (Com.).

Supplementary information

Consequently, the following supplementary materials were also updated:

- A. LCI
- C. Carbon balance
- D. LCA results
- E. Uncertainty



Life Cycle Assessment of pilot scale production of seaweed-based bioplastic

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ABSTRACT

The use of seaweed as a bioresource for plastic production is gaining momentum. However, the environmental impacts of the production of this novel bioplastic are still unknown. In this research, we assess the environmental impacts of the production of a bioplastic film at an experimental pilot scale using Life Cycle Assessment (LCA). The system boundaries chosen for this analysis include seaweed cultivation accounting for its carbon uptake, alginate extraction, production of bioplastic film at the pilot scale and different end-of-life pathways. The recirculation of different seaweed co-products from the alginate extraction step into the bioplastic film production is also assessed using scenario modelling and the analysis is completed with a carbon balance and an uncertainty analysis. The results show the main hotspot at the pilot scale is the last step in the production, *film fabrication*, mainly due to the glycerol in this process. The results also vary significantly depending on the end-of-life of the bioplastic, composting reduces the impacts by 30 % compared to incineration.

1. Introduction

Plastic is a widely used material due to its versatility, durability, resistance and low price [1]. However, conventional plastics are made from non-renewable sources and they threaten marine environments [2–5]. Bioplastics could potentially represent a solution to these problems but first- and second-generation bioplastics produced from biomass have been shown to exert a substantial impact in terms of direct and indirect land-use change [6–9]. Instead, seaweed is considered a third-generation feedstock for bioplastics because there is no land use involved in its cultivation [10,11]. Furthermore, seaweed cultivation delivers regulating and supporting ecosystem services and habitat provisions, such as carbon and nutrient cycling. Hence potentially contributing to the reduction of eutrophication, ocean acidification and climate change, among other benefits [12–16].

While the term bioplastic is often ambiguous as it is used to refer to either bio-based and/or biodegradable plastic [17], in this article the term bioplastic is used to describe plastic that is both bio-based and biodegradable. The production of such bioplastic from seaweed has gained interest in recent years, even though there are still doubts regarding the potential of this technology to substitute, partly, the production of plastic from fossil sources [6,18]. As an emerging technology, it is important to identify at the early stages of technological development (i.e., at a pilot scale) the environmental hotspots of

seaweed-based bioplastic production and opportunities for optimising the design of this process before it is implemented at an industrial scale [19].

Recent studies investigate and discuss the potential use of seaweed as a bioresource for bioplastics. Rimundo [20] is among the first to investigate the use of alginate as possible material for food packaging and, more recently, Carina et al. [21] expand the state of the art on seaweed polysaccharides as food packaging material using not only alginate but also other polysaccharides extracted from brown, red and green seaweed. Pacheco et al., Zhang et al. and Zanchetta et al. [17,22,23] describe the general status of seaweed polymers and their properties to produce different plastic types. Shrivaya et al. and Lim et al. [24,25] review the processes to extract seaweed polymers for use in bioplastic production. Some studies perform an experimental study on the production of seaweed-based plastics. Albertos et al. [26], for instance, complete an experimental production of edible films using brown and red seaweed, and Aragao et al. and Lim et al. [27,28] bioplastic using alginate in brown seaweed. While these studies review different seaweed-based plastic types, they mainly focus on the physical properties and the potential methods to produce seaweed-based plastics, some of them using experimental data, but do not assess quantitatively the environmental impacts of the production and end-of-life (EoL) of those seaweed-based plastics.

Instead, Helmes et al. [29] perform an LCA study of a seaweed-based

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plastic where they study the production of lactic acid (PLA) from the green seaweed *Ulva* spp. and show that the main impacts of producing 1 kg of purified lactic acid are the result of using electricity during land-based seaweed cultivation. There are then other studies that perform an LCA of the cultivation of the brown seaweed *Saccharina latissima* [30] and drying [31], as well as seaweed cultivation and processing of this seaweed specie to produce biofuels [32–34] and highlight that the main hotspots are seaweed processing, and fuel and drying respectively. Nonetheless, according to van den Burg et al. [35], the use of *Saccharina latissima* to produce bioplastics has not been documented yet.

Some LCA studies of algae-based bioplastic do not include the entire life cycle of the bioproduct but only consider cradle-to-gate [34,36]. It is important to account for the emission from the EoL to fully assess the impacts of the product and avoid misleading conclusions. For example, as the carbon accumulated in seaweed-based bioplastic is released in the EoL, excluding the EoL from the LCA study can erroneously display the bioproduct as a net carbon sink if only the carbon uptake is accounted for. It is moreover important to explore different EoL scenarios, as how the bioplastic is processed after use can vary substantially depending on local conditions.

In summary, while the potentials of alginate-based plastic and seaweed cultivation have been respectively addressed in the literature, the environmental impacts of seaweed-based bioplastic over its entire life cycle remain uncertain. In this context, an LCA study is conducted using primarily experimental data from a lab- and pilot-scale alginate-based plastic production using the brown seaweed *Saccharina latissima*. A carbon balance is also performed to keep track of the carbon flows and ensure a representative carbon accounting over the entire life cycle. The LCA is complemented with an uncertainty analysis.

This assessment is conducted early in the technology development and should help to identify the hot spots already present at the pilot scale. The study aims to use the consequential approach to determine impacts that arise when demanding 1 kg of bioplastic produced from brown seaweed *Saccharina latissima*.

2. Methodology

2.1. Goal and scope of the LCA, system boundaries, and co-products

The consequential LCA was performed using SimaPro v9.2.0.2 and Brightway2 [37] with the ecoinvent v. 3.8 background database [38]. ReCiPe 2016 Midpoint (E) was used as the Life Cycle Impact Assessment method. SimaPro was used initially to calculate the impacts and Brightway2 to perform an uncertainty analysis. The functional unit (FU) is the production of 1 kg of bioplastic film. This bioplastic is a transparent, flexible and thin plastic film. The system under analysis (cf. Fig. 1) was modelled using a cradle-to-grave approach. The system boundaries included all the necessary processes to produce the bioplastic film, from the cultivation of the seaweed in off-shore farms, including the hatchery and CO₂ uptake of the seaweed during its growth in the sea, the seaweed biorefinery and film fabrication with plasticizers and the EoL treatment of the bioplastic. The recirculation of different co-products in the seaweed biorefinery step was assessed in five scenarios.

2.2. Life cycle inventory

The life cycle inventory (LCI) was completed using experimental data from a pilot-scale production from the PlastiSea project [39], where the project partners covered the entire supply chain: seaweed cultivation and harvesting, seaweed processing, acid wash and alkaline extraction, film fabrication at the lab- and pilot-scale.

The modelled location was Norway, where the seaweed was cultivated. It was further assumed that the bioplastic film was produced close to the seaweed farm, as there are numerous advantages to producing seaweed-based plastic in proximity to seaweed farms. The main advantage being reducing transportation costs: since the process to

produce this bioplastic uses wet seaweed, and given the high-water content of seaweed, the transportation costs of seaweed for this purpose are especially high [40]. The table with the LCI can be found in Supplementary material A.

2.2.1. Hatchery and offshore farm

Saccharina latissima is the seaweed used and its cultivation usually requires a hatchery or nursery step for seedling production. Seaweed spores are settled into ropes and incubated in tanks, which contain filtered seawater and a growing solution containing nutrients and pesticides to support their growth [41,42]. The nutrients consist of a solution called *West and McBride's Modified ES Medium*. The gametophytes grow into juvenile sporophytes for 4–6 weeks [43]. This process helps to ensure their later growth in the ocean. The result of the hatchery is small seaweeds attached to a rope that is later deployed into the ocean. With direct seeding, seeding the lines without the hatchery step, the yields are usually lower as the holdfast is often underdeveloped and they are more likely to get detached from the seeding ropes [37].

Once the seedlings are 1–2 cm long, the seeding lines are rolled around sturdy ropes and deployed into the sea in autumn [42]. The seaweed farm is organised in horizontal lines attached between buoys. The seaweeds grow for 6–7 months and are regularly monitored and checked. The length of the seaweed varies between 1 and 2 m depending on the harvesting time. At the beginning of the harvesting season, in April, seaweeds are around 1 m long and at the end of the harvesting season, in June, the seaweed can be up to 2 m long.

The seaweed farm is based on a mooring frame with its structural horizontal lines suspended at 3 m depth. It is a square shape of 440 m in length, divided into sectors of 110 m each, in water depths of approximately 10–30 m. The farm is located approximately 2 km east of Sistranda and Frøya Island, and 3 km from its land base. It is protected by a nearby Island in the south and partially protected on the east and northeast sites, allowing swell and wind waves to enter with estimated maximum heights of 2 m. The long lines connected to the mooring frame are stretched at a depth of 1.5–2 m, with a spacing of 15 m. The seeded substrate is connected horizontally between long lines with an average spacing of 1.5–2 m. There is annual planned maintenance of the farm structure between the harvest season and the deployments (July–August), and minor corrections are done throughout the year. Typically, aquaculture catamaran-type workboats are used for accessing the structural elements, as cranes are needed.

The LCI data from the hatchery and sea cultivation was taken from the reports of the GENIALG project [42,44] with minor modifications provided by the seaweed farmers involved in this project and updated processes from ecoinvent 3.8. This data included the materials to build and maintain the farm structure.

2.2.2. Seaweed biorefinery

This seaweed biorefinery aims to extract alginate, cellulose, mannitol, laminarin and fucoidan from *Saccharina latissima* and fresh seaweed is used for this purpose. In this research, alginate was considered the main product, and mannitol and cellulose were considered co-products. Laminarin, fucoidan and other seaweed compounds are potentially also extracted in this biorefinery process but are not used in this bioplastic film. The polysaccharide extraction is based on the method proposed by Wahlström [45], with some modifications. The insoluble fraction is sequentially treated with hydrogen peroxide (H₂O₂) and ethanol to extract high-quality cellulose. Ethanol is used to remove pigments and fatty acids [45].

The biorefinery starts with seaweed milling, followed by an acid step where hydrochloric acid (HCl, 0.2 M) is added. Laminarin, fucoidan and a mannitol-rich fraction are extracted in the acid and washing steps, which can have a high economic value and are important for economic viability [46]. The acid helps to break the cell wall structures and remove minerals such as calcium crosslinking the alginate within the biomass [47] and the washing, using tap water, helps to extract the

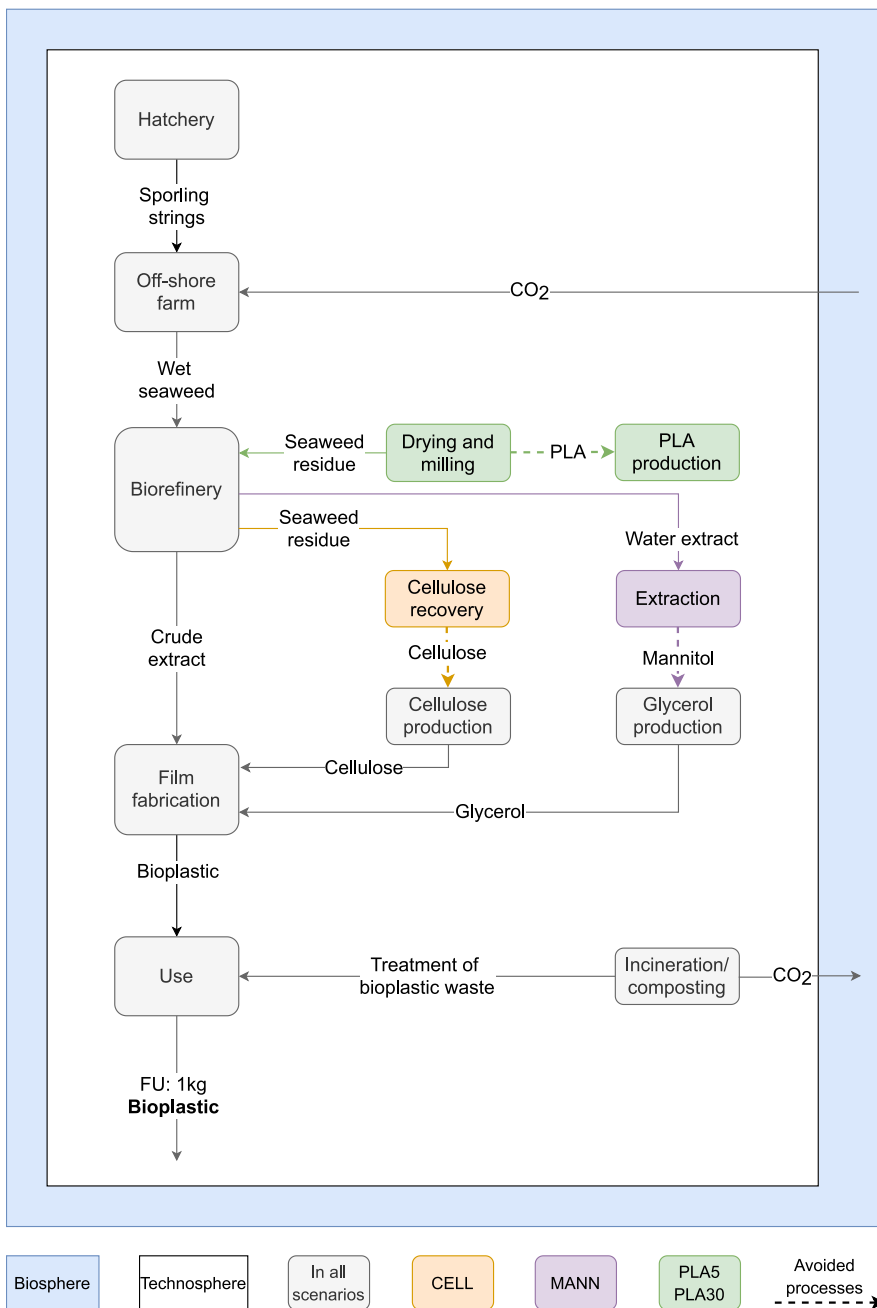


Fig. 1. Foreground product system. System boundaries of all scenarios. BASE: baseline scenarios; CELL: cellulose recirculation scenarios; MANN: cellulose and mannitol recirculation scenarios, PLA5: 5 % PLA substitution; PLA30: 30 % PLA substitution. Each scenario has two EoL sub-scenarios: incineration and biodegradation.

remaining laminarin, fucoidan and mannitol fractions. Depending on the time of harvest, mannitol can constitute up to 25 % of the seaweed's dry weight [48]. In this research, around 12 % of the dry weight was considered mannitol based on previous studies [49] and approximately 80 % of the mannitol was considered to be extracted in this biorefinery process. The process continues with an alkaline step to solubilise and extract alginate. The acid step, washing and alkaline treatment are carried out in a continuous stirred-tank reactor. The alkaline extraction starts by adding 0.2 M NaHCO₃. Centrifugation is the following step, which aims to separate the supernatant mixture and the seaweed residue [50,51], containing alginate and cellulose respectively. An alginate-rich crude extract is the main product and the seaweed residue is the co-product. The seaweed residue has a high cellulose content and two co-products could be obtained with the seaweed residue: cellulose and processed seaweed pellets. The cellulose is extracted using ethanol and H₂O₂ and the seaweed pellets are obtained by drying and milling the seaweed residue. A flow diagram illustrating the biorefinery process is presented in Supplementary material B.

2.2.3. Film fabrication

Alginate is water sensitive and would have limited applications as the sole component in a bioplastic on its own. A film compounding process is, therefore, recommended to lower the final film moisture sensitivity [52]. Three steps can be distinguished in the film fabrication process where different plasticizers and polymers are used to produce this transparent and flexible bioplastic: (1) compounding and homogenization, (2) casting and (3) crosslinking.

In the compounding and homogenization step, the alginate extracted in the biorefinery step is mixed with cellulose and glycerol, which are used as functional additives within the formulation. Glycerol is a widely used plasticizer with hydrophilic biopolymers [23], improving the mechanical properties of alginate-based plastic, such as water permeability and thermal resistance [53]. Cellulose is used as a reinforcing filler. Considering the mechanical properties of cellulose, strength properties with high stiffness and tensile strength, the cellulose nanofibers (CNF) can also be used to produce bioplastic film [54] without affecting its compostability [55]. The homogenization step is accomplished by using an Ultra-Turrax for mixing and a vacuum pump for degassing.

The casting step consists of pouring the homogeneous viscous mixture onto a flat surface. Due to the properties of the mix, the method used to cast the bioplastic at the pilot scale is an adapted cast film extrusion to allow the use of aqueous mixes instead of thermoplastics. The mix is dispensed in a liquid pump, and afterwards, the material is homogeneously distributed on the roll. However, at the pilot scale, an adapted cast extrusion machine is used: the homogeneous mixture is dosed using a liquid pump and the volume to be cast is controlled by a dye.

The crosslinking step included spraying a solution of calcium chloride (CaCl₂) onto the casted solution in order to improve the mechanical properties, moisture sensitivity and visual appearance of the bioplastic, including homogeneity and thickness [52,56]. Calcium ions in combination with glycerol are claimed to be an optimum combination to enhance the mechanical properties of the alginate-based bioplastic films to increase their water resistance and flexibility [53]. Finally, the film fabrication process is concluded by a drying step, in which the water content of the film is reduced to around 20–30 %.

2.2.4. End-of-life

Since the true end-of-life of the plastic is currently unknown and might differ depending on the location where the plastic is used and collected, two different EoL pathways were modelled: incineration and composting. Incineration was chosen because it is a realistic possible pathway e.g. in Nordic countries like Denmark and Norway. Composting was chosen because this novel bioplastic is biodegradable as confirmed by both qualitative (observational — soil-burial) and quantitative (screening — closed chamber bioreactor) laboratory tests carried out

within the consortium of the research project funding this study (data not yet published). Since this bioplastic is not recyclable with conventional plastics, a mechanical recycling EoL scenario was discarded.

Both EoL scenarios were modelled using the background data from ecoinvent and modifying the background data. The background process for the composting EoL was *Bio waste {CH} | treatment of bio waste, industrial composting | Conseq, U*. The electricity mix of the background process was modified to include the electricity mix in Norway instead of Switzerland.

For incineration we used the following process: *Waste polyethylene {CH} | treatment of, municipal incineration | Conseq, U*. It was considered that polyethylene had similar properties to this seaweed-based plastic when it comes to the incineration process. This process produces electricity and heat waste. In Norway energy from the incineration process is recovered, but the dataset in ecoinvent only provides the emissions and not the recovery heat and electricity. According to the dataset documentation, 5 MJ/kg and 10.02 MJ/kg are respectively the net waste of electric and thermal energy that can be recovered for burning polyethylene in a municipal solid waste incinerator [57]. Therefore, electricity and heat were modelled as avoided products in the incineration EoL.

2.3. Carbon balance

The carbon balance accounted for the carbon uptake in the seaweed cultivation process, the carbon content from the bioplastic film components and the EoL of the bio-based plastic. The carbon content of the compounds used in all the processes to produce the bioplastic plastic was accounted for, i.e. biorefinery and film fabrication. This carbon (C) content was calculated based on the measured mass and molecular weight of polymers (alginate, cellulose and glycerol) in the produced bio-based plastic at the lab and pilot scale biorefinery. The point of departure to calculate the carbon balance was 1 kg of carbon in the bioplastic film. The carbon content of alginate, glycerol and cellulose was calculated considering their molecular weight, the number of carbon molecules and the molecular weight in each molecule, and their proportion in the bioplastic. The carbon balance was calculated using a substance flow analysis approach [58]. The carbon flows in the EoL were calculated assuming the oxidation/combustion of the polymers in 1 kg of bioplastic. The biogenic carbon was maintained separately, i.e. the carbon from the alginate, mannitol and glycerol from the seaweed, and the fossil carbon, i.e. carbon of glycerol and cellulose added in the crosslinking step not derived from the seaweed.

The carbon balance was calculated for the first three scenarios later explained (cf. *Scenario analysis* section). A detailed explanation of how the carbon balance was accounted for can be found in Supplementary material C.

2.4. Scenario analysis

A scenario analysis was developed to assess the circulatory of cellulose and mannitol in the seaweed biorefinery processes to be later used in the film fabrication step. Cellulose and mannitol are seaweed co-products which were originally modelled as waste diluted in the water and acid waste. Scenarios were developed with the recirculation of each co-product, mannitol and cellulose. The following figure (Fig. 1) represents the system boundaries of all the scenarios.

All the modelled scenarios included the seaweed hatchery, offshore cultivation farm, biorefinery, film fabrication and two end-of-life scenarios. The baseline scenario (BASE) was modelled simply considering the production of the main product, alginate-rich crude extract in the biorefinery (Fig. 1), excluding the recirculation of the co-products in the biorefinery. In the cellulose recirculation scenario (CELL), the cellulose from the seaweed was recovered in the alkaline extraction step of the biorefinery process. The recovered seaweed cellulose was recirculated to replace the commercial cellulose nanofibers (CNF) used in the film

fabrication step. Mannitol and glycerol have similar mechanical properties: both are sugar alcohols and have similar molecular structures. Mannitol can theoretically be used to substitute glycerol [59]. In the third scenario (MANN), the recirculation of mannitol from the acid wash step in the seaweed biorefinery was assumed.

Experiments conducted within the PlastiSea project [39] showed that the seaweed residues from the alkaline extraction step in the biorefinery, once dried, could be used as a filler for polylactic acid (PLA). The experiments were conducted by substituting 5 % of PLA (PLA5) and 30 % (PLA30). With a 5 % of substitution, the mechanical properties of the PLA plastic were better preserved than with a 30 % of substitution. Table 1 presents a summary of the key features for each of the modeled scenarios.

2.5. Uncertainty analysis

Since the technology under analysis was at the pilot scale, it is important to understand the uncertainties in the impact scores produced by the model. An uncertainty analysis of the following indicators was performed: *reliability, temporal correlation, geographical correlation, completeness and further technological correlation*. In this study, a stochastic approach was adopted and propagated the uncertainty in the inputs to the outputs using a Monte Carlo simulation, i.e. randomly sampling input values in their range and iterating the calculations 1000 times.

In a first-of-its-kind approach, the total uncertainty was decomposed between the uncertainty in the foreground system, i.e. the primary data about a pilot scale technology, and uncertainty in the background system, i.e. secondary database data about established technologies. While uncertainty information such as location, scale, and distribution type for each exchange in the database are available fromecoinvent, the uncertainty in the foreground system is unknown. Due to the lack of repeated measurement data, and consistency with the background database, the pedigree matrix approach was used to provide an expert-based estimate of the uncertainty in each exchange in the foreground system and assumed a lognormal distribution to avoid negative values. This is a simplified approach but pragmatic given the resources available. The Monte Carlo simulation was then performed considering the uncertainty in the foreground system only, in the background system, and the two together respectively. This allows us to appreciate the uncertainty due to this model alone and combined with the inevitable uncertainty of the database data used in the LCA.

3. Results

3.1. Carbon balance

The results in the carbon balance illustrate the carbon flow within the different scenarios (cf. Fig. 2). The first scenario (BASE) (Fig. 2, 1) assumes that alginate is the only seaweed compound used in the bioplastic and this accounts for 0.44 kg C in 1 kg of bioplastic. I.e., 44 % of the carbon is biogenic in this scenario, while cellulose and glycerol were added externally. In this scenario, 69 % of the carbon in the seaweed goes to seaweed residues. The second scenario (CELL) (Fig. 2, 2) shows the recirculation of alginate and cellulose from the seaweed to the bioplastic film. In this scenario, 12.5 % of the carbon in the cellulose nanofibers is avoided when recirculating the cellulose in the seaweed.

Table 1
Summary of the scenarios modelled in the study.

Scenario acronym and name	BASE Base scenario	CELL Cellulose recirculation	MANN Mannitol and cellulose recirculation	PLA5 PLA substitution 5 %	PLA30 PLA substitution 30 %
Co-products	None	Cellulose	Cellulose and mannitol	PLA filler pellets	PLA filler pellets
End-of-life sub-scenarios	Incineration composting	Incineration composting	Incineration composting	Incineration composting	Incineration composting

The third scenario (MANN) (Fig. 2, 3) displays that the mannitol in the seaweed is lower than the glycerol required for the bioplastic. The mannitol from the seaweed avoids 5 % of the carbon from the glycerol. The carbon balance in scenarios PLA5 and PLA30 (Fig. 2, 4) shows how seaweed residues in the base scenario are later used to substitute 5 % or 30 % of PLA. When using seaweed residues as PLA filler, more of the original seaweed biomass is integrated into products and less is discarded (cf. Fig. 2, 4).

Overall, the results in the carbon balance show that by recirculating cellulose and mannitol by substituting PLA in scenarios CELL, MANN, PLA5 and PLA30, more of the carbon originally absorbed by the seaweed is converted into products, and less external carbon inputs are required. In other words, the BASE scenario requires the most external carbon inputs.

3.2. Scenarios and contribution analysis

The results (Fig. 3) reflect the contribution analysis of the Global Warming (GW) impact of all the processes across all life cycle stages of 1 kg of bioplastic film. The scenarios with the highest impact are the BASE, CELL and MANN with the incineration EoL, each with an impact of 3.72 kg CO₂-eq., 3.79 kg CO₂-eq. and 3.77 kg CO₂-eq. respectively. The scenarios with the lowest impacts are the PLA substitution scenarios with the composting EoL, the lowest being PLA30 substitution with 2.3 kg CO₂-eq. and PLA5 with 2.53 kg CO₂-eq. closely followed by BASE composting, 2.56 kg CO₂-eq.

The highest impact in all cases derives from film fabrication, mainly due to the high contribution from the production of glycerol with an impact of 2.11 kg CO₂-eq. The recirculation of cellulose and mannitol in the biorefinery, both used in the film fabrication in scenarios CELL and MANN, requires the use of oxygen peroxide and ethanol in the biorefinery step. This means that the GW in biorefinery, in both CELL and MANN, is 0.11 kg CO₂-eq., which is significantly higher when compared to 0.03 kg CO₂-eq. in the BASE scenario. Moreover, a substantial amount of glycerol is needed in the film fabrication step when considering the relatively low proportion of mannitol in the seaweed with 80 % recovery in the biorefinery process. The PLA substitution scenarios (PLA5 and PLA30) have a lower impact than the BASE scenario. In the incineration sub-scenario, the impact is 3.68 kg CO₂-eq. (PLA5) and 3.46 kg CO₂-eq. (PLA30). In the composting sub-scenario, it is 2.53 kg CO₂-eq. (PLA5) and 2.3 kg CO₂-eq. (PLA30).

In all the scenarios, the GW impacts of the offshore farm are significantly low compared to other processes, due to, for instance, the carbon uptake in seaweed cultivation. Regarding the EoL, there is a considerable difference between incineration and composting in all scenarios, with a GW impact of 1.27 kg CO₂-eq. in incineration and 0.11 kg CO₂-eq. in composting. The results from all the impact categories can be found in Supplementary material D.

3.3. Uncertainty analysis

The results of the uncertainty analysis are visualised in Fig. 4. To reproduce the results, the code used in the study is available in a GitHub repository [60]. Summary statistics for all distributions and a matrix of the differences between paired samples of all distributions are provided in Supplementary materials E and F.

These results confirm the conclusion from the static analysis that

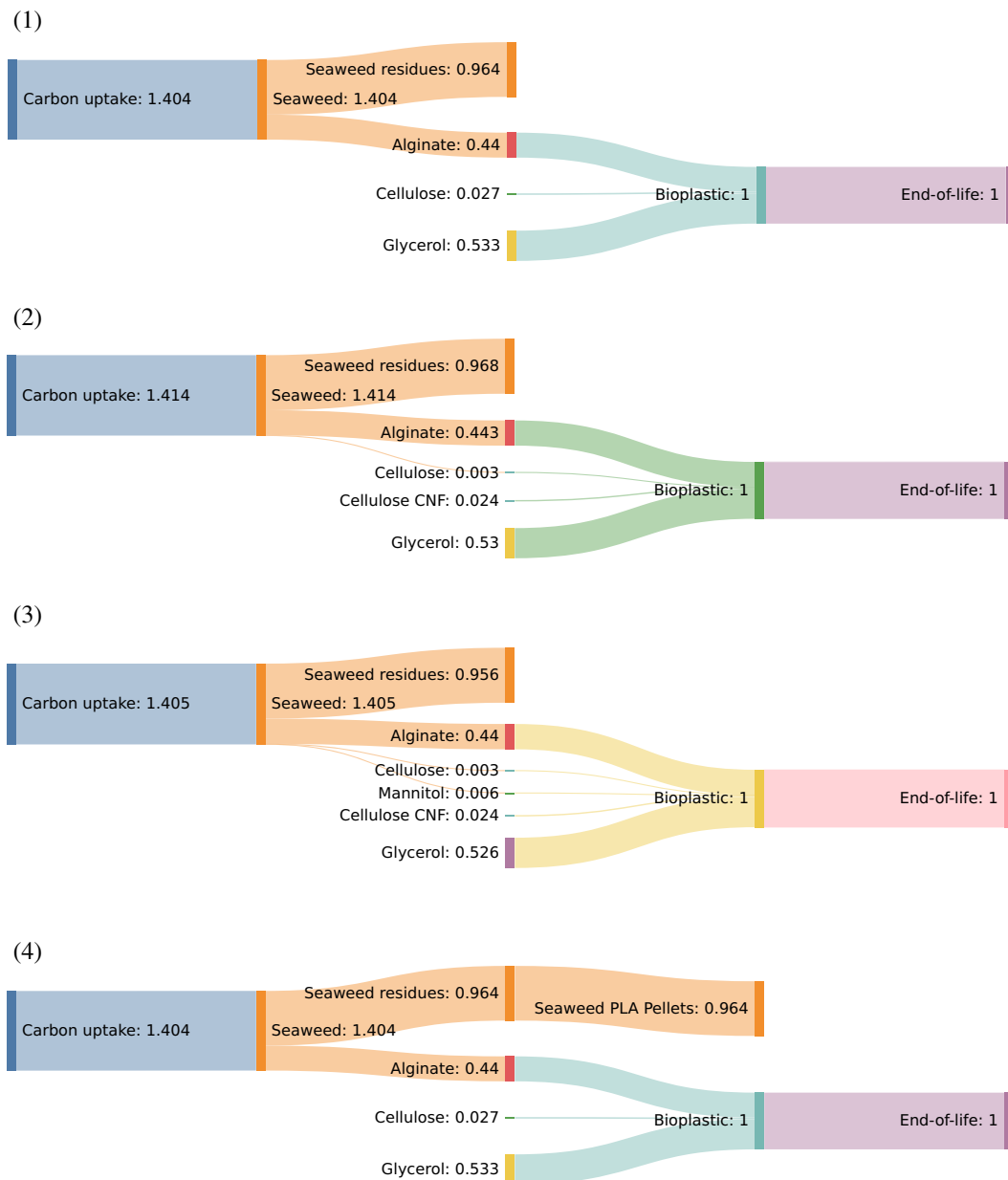


Fig. 2. Sankey diagrams of the carbon balance. Base scenario, BASE (1); cellulose recirculation scenario, CELL (2); and cellulose and mannitol recirculation scenario, MANN (3); PLA substitution 5 % and 30 %, PLA5 and PLA30 (4). Units: kg carbon.

indeed the composting alternatives (“composting”) perform better than the incineration counterparts, as more than 96 % of the time, the results in the composting scenarios are lower than those in the incineration scenarios. Visually speaking, Fig. 4 also confirms that the distributions for the composting alternatives are always shifted downwards compared to the distribution of the incineration alternatives.

Given the uncertainty, the ranking across the various scenario can,

however, not be confirmed with confidence. Results for the MANN, CELL and PLA5 scenarios are lower than the BASE scenario in less than 50 % of the samples, and less than 60 % for the PLA30 substitution scenario. Given the uncertainties, these scenarios are not clearly distinguishable from the BASE scenario. This result reflects two aspects: first, the technology is still at the pilot scale and uncertainties are expected to be high; second, the pedigree approach used for estimating these uncertainties

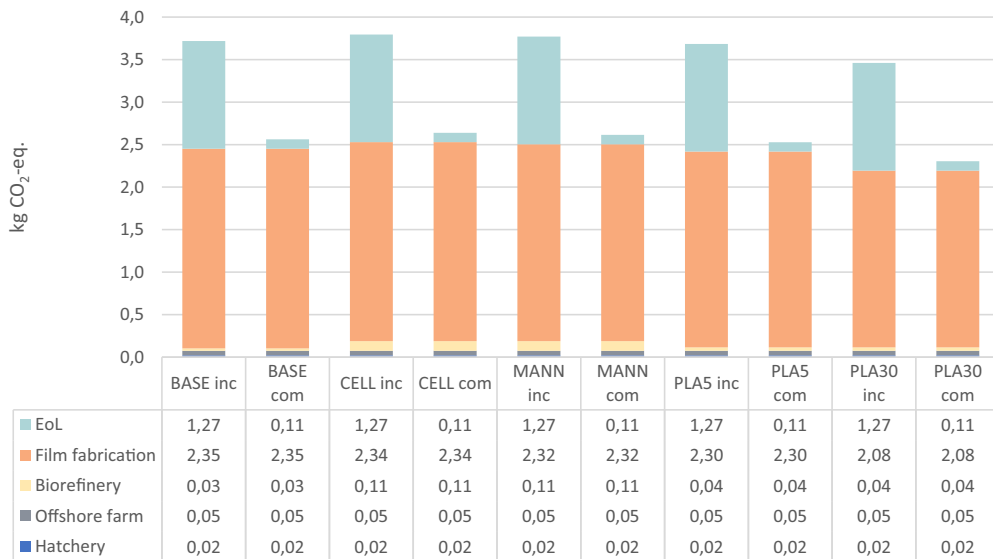


Fig. 3. Contribution analysis in the impact category Global Warming (GW) impact of all the scenarios and sub-scenarios: BASE, CELL, MANN, PLA5, PLA30, incineration (Inc.) and composting (Com.).

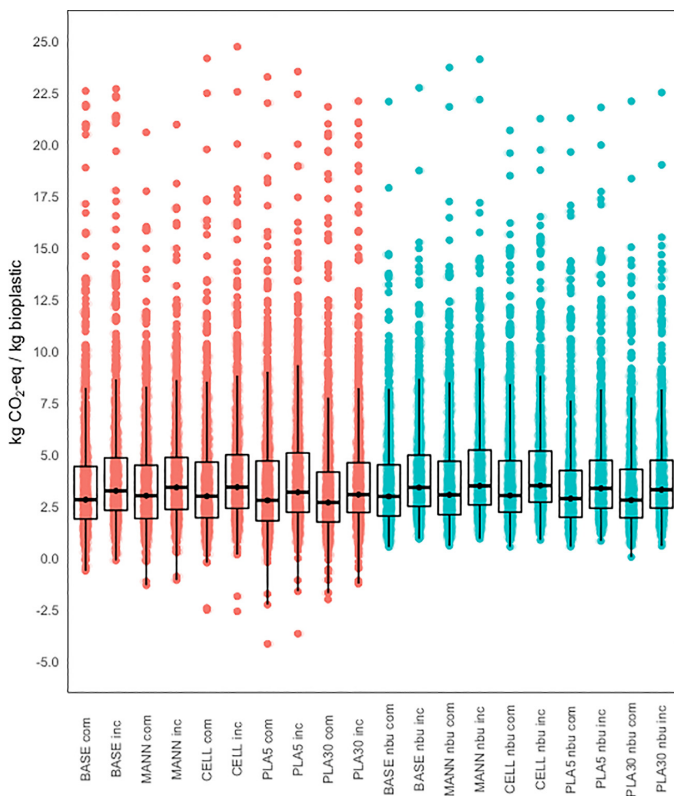


Fig. 4. Uncertainty analysis in all scenarios. The figure shows the result of the Monte Carlo simulation (1000 paired samples) across scenarios both by including foreground and background uncertainty (left side, red colour) and by excluding the background uncertainty (right side, light blue colour, label “nbu” — no background uncertainty) respectively. Boxplot: box bottom = 25 % quantile, box top = 75 % quantile, bold line = 50 % quantile. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

has low precision and is likely to overestimate the uncertainty.

The comparison between the uncertainty generated when considering both the foreground and background uncertainty, and the foreground uncertainty only also provides interesting insights. Fig. 4, shows no substantial difference in the spread of the distributions with or without background uncertainty. Interestingly, without background uncertainty, the distributions tend to remain more positive while removing negative outliers — a sign that the uncertainty in specific background activities in the database might skew the results. When comparing the summary statistics, it can be seen more clearly that the uncertainty in the distributions obtained without background uncertainty is reduced as the standard deviations of all distributions decrease. A more accurate comparison between the summary statistics of the distributions obtained with and without background uncertainty is not possible, as the sampling is not paired across the two versions of the background database (with and without uncertainty) used in the simulation.

4. Discussion and conclusion

This study explores the environmental impacts of a novel seaweed-based plastic using *Saccharina latissima* and the potential of recirculating some co-products in the biorefinery step, to reuse in the later film fabrication step. This potential is assessed via carbon balance and LCA.

Previous studies stress the importance of using the entire seaweed biomass to increase its value [46]. That is not only relevant from an environmental perspective but also from an economic perspective. Seaweed biomass has a high demand in the food and feed sector, where a high economic value is given to biomass. The price of the seaweed biomass for plastic production is significantly lower, thus the utilization of all the seaweed biomass and co-products from the biocrude extraction increases its value [35]. The results of this study, however, show that the scenarios where mannitol and cellulose are recirculated have a slightly higher impact at the pilot scale than the base scenario, although due to the substantial uncertainties involved this difference can only be confirmed with low confidence. It needs to be considered that we assumed that 12 % of the dry matter of the seaweed is mannitol and 5 % cellulose and the mass of mannitol and cellulose obtained from the biomass is insufficient to produce this bioplastic film. Nevertheless, other studies show the influence of using whole seaweed biomass versus extracting a single component when the biorefinery processes are optimised [46,61].

In the different scenarios, we see that the end-of-life scenario reduces the carbon footprint by approximately 30 % via composting compared to incineration. These results show the importance of having an EoL aligned with the properties of the bioplastic. In this case, the bioplastic is 100 % compostable and is, therefore, important to compost this bioplastic film to reduce its impacts and increase its circularity.

It needs to be acknowledged that the carbon release is postponed in time with composting. In the composting scenario, 42 % of the carbon is stored but this carbon will eventually be released in the long term. A time-dependent analysis would be needed to appreciate this delay in emissions [62] but it was beyond the scope of this study. The contribution to the GW impact from the electricity consumption is not substantial due to the high percentage of hydropower and wind power in the Norwegian consequential electricity mix. Patel et al. [63] confirm the benefits of using renewable energy to develop algal products.

The base scenario has a carbon footprint of 3.7 kg CO₂-eq. in the incineration sub-scenario and 2.6 kg CO₂-eq. in the composting scenario. For comparison, the carbon footprint of 1 kg of polypropylene and polypropylene is 1.71 and 2.4 kg CO₂-eq. respectively using ReCiPe 2016 Midpoint (E) and default datasets from the ecoinvent database [38]. Looking at other bioplastics, we can see that the carbon footprint of bio-based PLA is higher than fossil-based plastics, 4.21 kg CO₂-eq. [38]. For contextualisation, it needs to be considered that conventional fossil-based plastics are well-stabilised industries and the impacts are

analysed at an industrial scale while the bioplastic production technology modelled in this study is at a pilot scale.

It is also worth mentioning the importance of including the entire life cycle of bio-based products [17]. If the carbon uptake is accounted for in seaweed cultivation, the stage where the carbon is released, the EoL should also be considered. Otherwise, an excessive benefit is given to the system. In this regard, performing a carbon balance is recommended to ensure how the carbon is distributed within and released from the system.

In this study, the carbon emissions for 1 kg of wet-weight (WW) seaweed is 0.082 kg CO₂-eq. This value is within the range of estimates from other studies assessing the carbon footprint of seaweed cultivation or harvest. Thomas et al. [30] report 55.2 kg CO₂-eq./tonne WW (0.0522 kg CO₂-eq./kg WW) in cultivation and Zhang et al. [61] 5187.6 kg CO₂-eq./tonne dry weight (approximately 0.5 CO₂-eq/kg WW) in harvesting.

Helmes et al. [29] is the closest previous assessment study of a seaweed-based plastic. They assess the environmental impacts of producing lactic acid from *Ulva* spp., a type of green seaweed with different properties than brown seaweed. The polylactic acid under assessment is a precursor of polylactic acid (PLA), but this is excluded from their system boundaries because the system only includes seaweed cultivation and processing. *Ulva* spp. has a different cultivation technique compared to brown seaweed, as it is cultivated in both land-based and off-shore systems. Due to the onshore cultivation needed for cultivating *Ulva* spp., electricity consumption is a hotspot in Helmes et al. [29] with a GW impact of 1.47 kg CO₂-eq./kg of purified lactic acid. The offshore cultivation of *Ulva* spp. is currently in progress and that would decrease the impacts of land-based [64]. Beckstrom et al. [36] assess the production of bioplastic feedstock from microalgae, with a worst-performing scenario showing a carbon footprint of 0.66 kg CO₂-eq./kg of bioplastic. Nilsson et al. [34] analyse a seaweed-based biorefinery where the GW impact is 2.73 kg CO₂-eq./kg of sodium alginate. In their case, drying the seaweed is a hotspot, but this processing step is not needed in the current research. Those studies do not include the entire lifecycle of the product as the EoL is excluded from their system boundaries. Even if the EoL only contributes to 5 % of emission composting and 30 % of incinerating, partially explains why their reported carbon footprints are lower than those reported in the current study. Another remarkable aspect is the fact that the current study shows pilot scale impacts and the processes are not optimised. That is noticeable in the case of the film fabrication stage. On that account, an LCA should be repeated in the future on an upscaled version of the product system.

There are some uncertainties given that the data for the LCI is done at a pilot scale. Following the uncertainty factors for the pedigree matrix [65], we consider a low uncertainty in *reliability*, *temporal correlation* and *geographical correlation* as the data is real experimental pilot-scale data, in a specific time and location. The *completeness* indicator has a higher uncertainty because the data is only at the pilot scale. Regarding the *further technological correlation* indicator, some processes were modelled with lower uncertainty and some with higher. Performing an uncertainty analysis also quantifies the confidence of the model. The processes at the lab- and pilot scale are not optimised. Therefore, and according to previous studies on the upscaling of emerging technologies [66–68], it could be reasonable to think that a reduction in the impacts would be possible when the system is upscaled to an industrial scale as the processes will be optimised, including the recirculation of co-products, and, consequently, fewer resources, energy and waste will be needed per FU.

The validity of the results of the uncertainty analysis needs to be discussed critically. Although 50 % of the data are contained approximately within a factor two range, several samples differ by a factor three from the median and outliers up to a factor ten are obtained in the simulation. The stochastic procedure is somehow artificial as it generates a large number (1000) of virtual instances of the system under analysis, and it is expectable to obtain a few very high and very low values in the sampling, leading to a large range overall, that needs to be

contextualised for better interpretation. On the one hand, the uncertainty range in the foreground system obtained with the pedigree matrix, and thus with a substantial degree of subjectivity in the assessment, might lead to overestimation. On the other hand, being some of the processes such as film fabrication are still at the pilot scale, it makes sense to obtain a large uncertainty on the results. The use of pedigree matrices and Monte Carlo simulation has been criticised in the LCA literature [69–71] and it is, hence, recommended to consider the results of the uncertainty analysis only in a comparative context to explore the degree of confidence with which we can say one alternative is better than the other given the uncertainty. Furthermore, we stress that the approach here used is a rough estimation of the uncertainty related to the assessment of technology at the pilot scale, and that more fine-tuned approaches to modelling upscaling should be applied to reduce model uncertainty, such as performing process simulations to upscale the biorefinery and processes and should be the topic of further research.

Another point of discussion is the limitation of using LCA for the overall environmental analysis of seaweed bioplastics compared to fossil plastics. For instance, current impact categories do not reflect the impacts of microplastics coming from fossil plastics, and thus, do not benefit bioplastics compared to fossil plastics.

While other studies assess the potential of using seaweed to produce bioplastics, this is one of the first studies to quantify its impacts. It can be concluded that seaweed-based plastic is a promising alternative to conventional land-based. The identified hotspots at the pilot scale are glycerol and cellulose in the film compounding step in the film fabrication.

Regarding the overall properties of this bioplastic film and its applicability, alginate is partially diluted in water. Thus, this bioplastic could be used as food packaging for greasy or dry food, until water barrier properties are improved by novel formulations and manufacturing methods. This bioplastic has also been proven to be suitable for fresh fruits and vegetables. It could likewise potentially be used as packaging for the fashion industry, cosmetics or other dry goods. Jabeen et al. [72] describe that storage tests, including oxygenation and water solubility, are necessary before upscaling and commercializing new bioplastics.

Although seaweed-based bioplastics are not the main climate solution, they help to reduce dependency on fossils and, hence, slightly contribute to reducing carbon emissions. Overall, it can be argued that one of the main environmental advantages of these seaweed-based plastics compared to first- and second-generation bioplastics are the low impact on land use in seaweed production. However, seaweed-based plastics are still in the early R&D stage and further research is needed to upscale and commercialise this novel bioplastic. Future work on upscaled industrial-scale impacts and upscaling scenarios using different techniques will be carried out.

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CRedit authorship contribution statement

Maddalen Ayala: Conceptualization, Data curation, Formal analysis, Software, Visualization, Investigation, Methodology, Validation, Writing – original draft, Writing – review & editing. **Marianne Thomsen:** Conceptualization, Supervision, Validation, Investigation, Methodology, Writing – review & editing. **Massimo Pizzol:** Funding acquisition, Conceptualization, Supervision, Formal analysis, Software, Visualization, Investigation, Methodology, Validation, Project administration, Writing – review & editing.

Declaration of competing interest

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

Data availability

The data is shared in the supplementary material

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APPENDIX

APPENDIX C

ARTICLE 3

Prospective LCA of brown-seaweed-based bioplastic: Upscaling from pilot to industrial scale

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Prospective LCA of brown seaweed-based bioplastic: Upscaling from pilot to industrial scale

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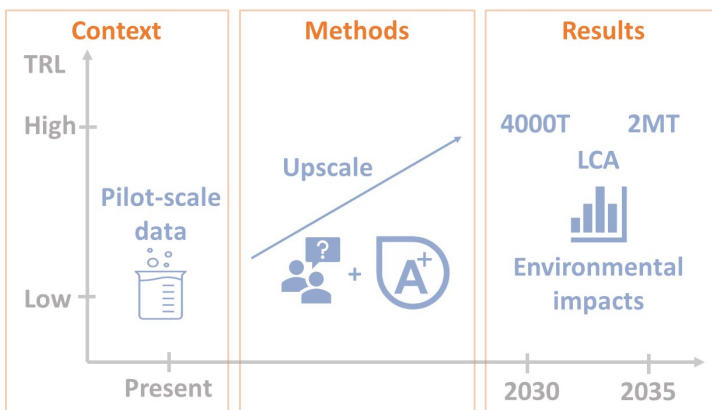
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Keywords: Process simulation, Aspen Plus, Emerging technology, Ex-ante LCA, *Saccharina latissima*, biobased.

Graphical abstract



Highlights

- Industrial-scale production of brown seaweed-based bioplastic was simulated.
- All production processes were upscaled to 4000 t/yr. in 2030 and 2Mt/yr. in 2035.
- Expert opinion and process simulations were used, complementing each other.
- Upscaling reduces impact down to 1.58 kg CO₂-eq. in the 2Mt/yr. scenarios.

- Uncertainties in assessing emerging technologies remain high.

ABSTRACT

Seaweed-based bioplastics are considered a possible alternative to conventional fossil-based plastics due to their potential environmental advantages. The cultivation of seaweed is a fast-growing practice that requires no arable land, freshwater, or fertilizers, perceiving it as an advantageous option for bioresource production. However, research on the environmental impacts of seaweed-based bioplastics is still limited, highlighting the need for a life cycle assessment (LCA) to evaluate their potential. In this article, a prospective LCA is conducted to assess the environmental impacts of brown seaweed-based bioplastic production, from pilot to industrial scale. Upscaling techniques are combined for each life-cycle stage, using interviews to upscale seaweed production and process simulation for the biorefinery and film fabrication steps, and the end-of-life scenario is modelled as composting. All the processes were upscaled to 4000 tonnes per year in 2030 and 2 million tonnes per year in 2035. The results show that the production of one kilogram of brown seaweed-based bioplastic resulted in approximately 1.37 kg CO₂-eq. in the best-performing scenario, producing 2 million tonnes per year in 2035 accounting for the carbon uptake, which is lower than LDPE with an impact of 3.6 kg CO₂-eq. This study provides for the first-time estimates of prospective industrial-scale impacts of the emerging seaweed-based bioplastic and shows how different upscaling techniques can be successfully combined, i.e., interviews and process simulation, to conduct a prospective LCA of seaweed-based bioplastics. The results demonstrate the potential of seaweed-based bioplastics as a sustainable alternative to conventional plastics.

1. Introduction

Plastic waste is a major environmental concern that has been growing over the years. Bioplastics have emerged as a promising alternative to fossil-based plastics, offering a potential solution to the plastic pollution problem (Bishop et al., 2021; Ita-Nagy et al., 2020; Rosenboom et al., 2022). Seaweed-based bioplastic, in particular, is an emerging product that is gaining significant attention in recent years. Seaweed cultivation has environmental advantages compared to first- and second-generation feedstocks currently used for bioplastic production that come from arable crops. Seaweed cultivation requires no arable land or freshwater (Bishop et al., 2022), sequesters carbon (Duarte et al., 2020), and reduces eutrophication (Duarte and Krause-Jensen, 2018; Prasad et al., 2022), contributes to emission capture and reuse through by nitrogen, phosphorus and carbon cycling (Seghetta, Marchi, et al., 2016; Seghetta, Tørring, et al., 2016; Thomsen & Zhang, 2020; Zhang et al., 2022). Seaweed cultivation can also provide habitat for marine organisms, enhancing biodiversity and ecosystem services (Thomsen and Zhang, 2020) and it is a fast-growing bioresource (Duarte et al., 2021). The literature also increasing opportunities to grow seaweed cultivation in Norway (Stévant and Rebours, 2017), in Europe (Araújo et al., 2021; van den Burg et al., 2021) and globally (Ferdouse et al., 2018). China dominates as the leading producer of fertilised brown seaweed globally, with an annual production volume of approximately 13 million tonnes of wet weight, food being the primary target market (Cai et al., 2021; FAO, 2021). This significant presence in the market poses both competitive pressures and constraints for the seaweed-based bioplastic industry, particularly considering China's focus on the food market (Ayala et al., 2023).

Several recent studies have investigated the potential use of seaweed as a bioresource for bioplastics. Studies by (Carina et al., 2021; Lim et al., 2021; Lomartire et al., 2022; Pacheco et al., 2022; Rinaudo, 2014; Shravya et al., 2021; Zanchetta et al., 2021; Zhang et al., 2019) have reviewed the use of seaweed polysaccharides for different types of plastic production and extraction processes. (Albertos et al., 2019; Aragão, 2022; Lim et al., 2018) have performed experimental studies on the production of seaweed-based plastics. While those studies have conducted experiments on the production of seaweed-based plastics, most have focused on the physical properties and methods for producing these plastics, without assessing their environmental impacts. Accordingly, a life cycle assessment is needed to fully understand the environmental

benefits and drawbacks of seaweed-based bioplastics compared to other types of bioplastics and traditional plastics.

Prospective life cycle assessment (LCA) is particularly important in assessing the environmental potential of seaweed-based bioplastics, as it can determine potential environmental impacts associated with the entire life cycle of the product in an early-stage technological development (Cucurachi et al., 2018). In this low Technology Readiness Level (TRL 4-6) only laboratory and pilot-scale data are available. In prospective LCA the systems are usually modelled at a future time, performed before the industrial-scale data is available and it is defined as an LCA in early development stages, i.e., low TRL, of emerging technologies when there are chances to make improvements (Arvidsson et al., 2018). By using prospective LCA, potential environmental issues early in the development process can be identified and addressed. The latter by minimizing negative environmental impacts and maximizing the potential benefits.

Ayala et al., (2023a) assess the environmental impacts of this bioplastic at the pilot scale. While pilot-scale LCAs are valuable for assessing potential environmental impacts and identifying areas for improvement, they return a rather uncertain estimate of the impacts since the use of materials and processes is usually not optimized at low TRL pilot scales (Bergerson et al., 2020). Industrial scale LCA of emerging technology provides the opportunity for a more realistic comparison with full-scale industrial products already on the market (Thonemann and Schulte, 2019).

A range of methods can be utilized in prospective LCA that can be used alone or in combination, including prospective scenario analysis (Langkau et al., 2023), utilization of prospective LCI background databases (Sacchi et al., 2022), advanced uncertainty propagation techniques (Jouannais et al., 2022) or the application of learning curves (van der Hulst et al., 2020), among others. Upscaling the processes is another commonly employed method, which can also be combined with the previously mentioned methods. Various methods can be applied to upscale the processes from lab or pilot scale to industrial scale, which can be summarized in manual calculations, molecular structure models (used for chemical technologies to calculate key LCI parameters using the molecular structure of a chemical), use of proxies and process simulations (Tsoy et al., 2020). Process simulations are considered one of the most accurate methods to upscale in prospective LCA (Parvatker and Eckelman, 2019; van der Giesen et al., 2020). Various research use process simulations to upscale lab or pilot scale procedures to industrial scale for the foreground system to

perform an LCA of emerging technologies (Budzinski and Nitzsche, 2016; González-García et al., 2018; Lozano et al., 2022; Lu et al., 2020; Mazzoni et al., 2019; Nitzsche et al., 2016). Process simulations has previously been used as a method to upscale performing a consequential prospective LCA (Thonemann and Schulte, 2019).

Current methods for upscaling processes primarily rely on quantitative approaches, i.e., the ones mentioned above (Tsoy et al., 2020), which may be limited as certain factors that need to be considered during upscaling the seaweed cultivation are not included as standard simulation units processes in existing process simulation tools. These factors include techniques to increase productivity, the availability of raw materials, technical feasibility and challenges and opportunities to upscale,. Therefore, it is recommended to complement quantitative methods with qualitative assessments, such as expert interviews or expert judgment, to ensure a comprehensive and accurate evaluation of the upscaling process (Buyle et al., 2019). In this context, it would be beneficial to get deeper insights into the emerging technology development within the seaweed cultivation field.

This article provides a prospective LCA of the upscaled process of lab and pilot scale bioplastic using alginate, a polymer found in brown seaweeds, also known as alginic acid, to industrial scale (TRL 9). The bioplastic under assessment is a transparent film, with a similar aspect to low-density polyethylene (LDPE) but different mechanical properties, i.e. it is biodegradable, with higher moisture content, water absorption and breathability. Based on an experimental pilot-scale production, we aim to upscale the processes of producing seaweed-based bioplastic at an industrial scale using a combination of qualitative and quantitative information on different life stages of the production processes, resulting in an LCA. This study employs a novel approach by integrating upscaling methods across different life-cycle stages, combining 1) qualitative information from expert opinion and interviews to upscale seaweed cultivation and 2) quantitative data obtained from process simulation using Aspen Plus to upscale the biorefinery and film fabrication. This study represents the first instance in which these upscaling approaches have been merged to enable a prospective LCA of industrial-scale seaweed-based bioplastic production.

2. Methodology

2.1. LCA

2.1.1. Goal, scope and system boundaries

The goal of the present study is to conduct an LCA of an industrial-scale seaweed-based bioplastic product system to evaluate the associated environmental impacts. The scope of the study encompasses the entire life cycle of this novel bioplastic, from seaweed cultivation, through processing to end-of-life (EoL) disposal options. The functional unit (FU) is defined as 1 kg of seaweed-based bioplastic film. To determine the potential environmental performance of using seaweed-based bioplastic as an alternative to traditional petroleum-based plastic packaging films, the system boundaries of the study have been established to include seaweed hatchery and cultivation with CO₂ and nutrient (phosphorus and nitrogen) uptake, biorefinery for alginate extraction, bioplastic film fabrication, and EoL modelled as composting. The seaweed cultivation includes a hatchery step, the biorefinery consists of an acid wash, wash and alkaline treatment, and film fabrication has crosslinking, compounding, casting and drying. Figure 1 represents the system boundary and all the steps involved in the system and each step is detailed in section 2.2.

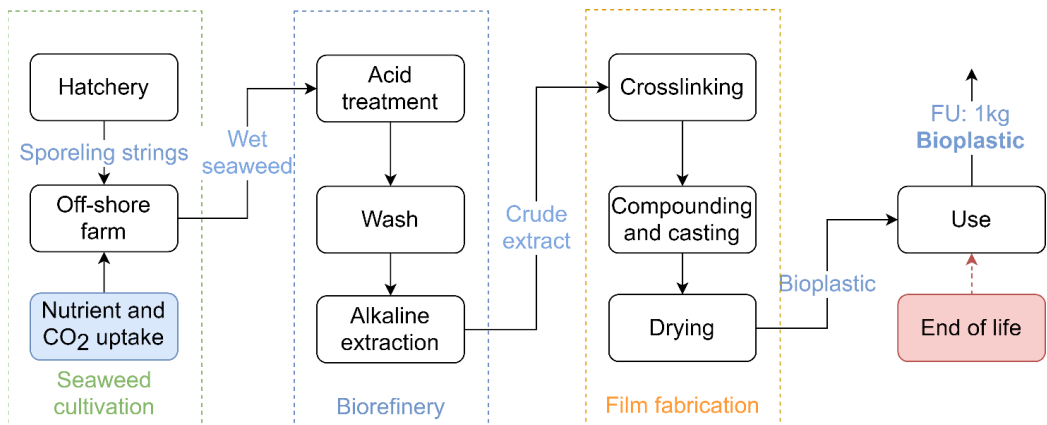


Figure 1: System boundary with each step in the seaweed cultivation (green), biorefinery (blue), film fabrication (orange), use and end of life.

Consequential LCA was chosen for this study because it considers the potential consequences of changes in the system under study and the implications of demanding 1kg of bioplastic (Ekvall and Weidema, 2004). The electricity marginal market mix was modelled according to the marginal suppliers based on Ayala et al. (2023b). For this analysis, ReCiPe 2016 Midpoint (E) was the life cycle impact assessment (LCIA) method used. In this study, two different approaches are employed to calculate the global warming impact score: including and excluding biogenic carbon respectively

Originally, the ReCiPe method excludes biogenic carbon and, hence, the carbon uptake during the seaweed growth from deployment to harvest. In order to show results accounting for biogenic carbon as well, thus focusing exclusively on the physical effect of uptaking and releasing carbon dioxide without normative assumptions about whether this should be accounted or not, the ReCiPe method was modified by assigning a characterization factor (CF) of 1 to the CO₂ classified as biogenic, for example, the CO₂ uptaken during seaweed cultivation and released during composting. The use of two approaches allows to appreciate the methodological differences considering that indications and guidance on how to include carbon in the assessment of biobased systems are not univocal across sources. The LCA was modelled using Simapro v9.2.0.2 with the background database ecoinvent v. 3.8 (Wernet et al., 2016).

The experimental data from the PlastiSea project (SINTEF, 2023) served as the primary data input for upscaling the production process in this study. The data obtained from the primary data from the PlastiSea project at low technology readiness levels (TRL) was utilised as the baseline for the upscaling approach.

2.2. System description and Life Cycle Inventory

The procedure used to produce the bioplastic is described in detail in Ayala et al., (2023). The upscaling process involved the use of interviews and process simulation techniques to gather information and predict the performance of the bioplastic production process at an industrial scale.

2.2.1. Seaweed cultivation

The data used for hatchery and seaweed cultivation was obtained from Seaweed Solutions AS, Trondheim based on the operation of a *Saccharina latissima* farm located off the coast of Frøya, Norway (63°42'15"N, 8°52'40"E), which produced 200T wet weight in the year 2022. The cultivation starts with a hatchery or nursery step to produce seedlings. The hatchery process involves settling seaweed spores into ropes and incubating them in tanks with a growing solution containing nutrients and pesticides to support their growth. Once the seedlings are 1-2 cm long, they are rolled around sturdy ropes and deployed into the sea in autumn. The seaweed farm is organized in horizontal lines attached between buoys, with a mooring frame at a depth of 3m. The seaweeds grow for 6-7 months and are regularly monitored and checked (Dias et al., 2021, 2020). The farm structure is subject to annual planned maintenance and minor corrections throughout the year.

2.2.2. Biorefinery

The biorefining process described here was based on (Birgersson et al., 2023; Nøkling-Eide et al., 2023) with some modifications based on internal experiments. The biorefining process aims to extract the components in the seaweed, alginate being the components of interest for this research and laminarin, fucoidan, mannitol and cellulose being the co-products. All the seaweed components are assumed to have a recovery rate of 80% based on experimental data.

The biorefinery processing starts with an acid step, where the *S. latissima* biomass is immersed in hydrochloric acid (HCl, 0.2 M). The acid treatment liquor is later processed with ultrafiltration steps, extracting 64% of mannitol, laminarin and fucoidan components in the biomass. The extraction of those components is followed by a washing step with fresh water, where an additional 16% of mannitol, laminarin and fucoidan are extracted. An alkaline step is used to solubilise and extract alginate with a sodium bicarbonate solution (0.2M NaHCO₃). The acid step, washing and alkaline treatment were carried out in a continuous stirred-tank reactor (CSTR). The next step is centrifugation, aiming to separate the supernatant solution and the seaweed residue, which contains alginate and cellulose-rich spent biomass respectively. The alginate-rich supernatant labelled 'crude extract' is the main product and the seaweed residue is the co-product.

Two co-products can be extracted from the seaweed residues: cellulose and seaweed pellets, which are mainly ashes and the 20% of the seaweed components not recovered. The cellulose is extracted, adding ethanol and H₂O₂ in a CSTR, followed by centrifugation to separate the cellulose (liquid) and seaweed pellets (solids). The seaweed pellets contain seaweed ashes, proteins, lipids and 20% of the remaining seaweed components not extracted in the previous steps.

2.2.3. Film fabrication

The film fabrication step can be summarized in 4 steps: (1) compounding and homogenization, (2) casting (3) crosslinking and (4) drying. In the first step, compounding and homogenization, the alginate extracted in the biorefinery is blended with glycerol and cellulose and then dispensed with a liquid pump. The process uses a technique known as wet casting, whereby the material is extruded from a head to form a sheet shape. This is followed by the crosslinking step where a solution of calcium chloride (CaCl₂) is sprayed onto the casted solution to improve its mechanical properties. The material is then placed on a conveyor belt with several drying stations, typically consisting of

fans or infrared lamps. At the pilot scale, there are 13 infrared lamps and 4 fans used for drying the bioplastic gel.

2.2.4. End-of-life

Two biodegradation tests were conducted within the PlastiSea project to assess the degradation of the bioplastic: A soil-burial test comparing the films to an LDPE control sample, and a disintegration test measuring the weight loss percentages. The preliminary results show that after 2 months, the bioplastics lost 50% of their weight. According to the biodegradability criteria, a minimum of 90% disintegration after 6 months is required. These preliminary results indicate the high biodegradability of seaweed-based films. Therefore, the end-of-life was modelled as composting using the *Biowaste {CH}* | *treatment of biowaste, industrial composting* | *Conseq, U* process in ecoinvent.

2.3. Upscaling procedure

2.3.1. Seaweed cultivation and interviews with seaweed experts

The qualitative information in this study is derived from two different sources, providing insights into the upscaling process of seaweed cultivation. Firstly, the upscaling of seaweed cultivation was based on expert opinions from seaweed farmers who provided estimations on resource utilization and infrastructure usage at the pilot scale. Their input helped determine an upscaling factor that considers the expected increase in resource utilization efficiency and subsequent yield improvements on larger scales.

Secondly, 11 seaweed experts were interviewed to further explore the potential to upscale seaweed cultivation and the techniques to increase productivity. The interviews aimed to gather insights into the potential for increasing brown seaweed production, as well as the limiting factors and techniques that could be used to upscale existing production. The respondents were selected based on their extensive expertise in seaweed cultivation, their industry experience, and their academic background. The respondents were further also selected based on their extensive expertise in various domains, including phycology, seaweed aquaculture, the Asian and European seaweed market, seaweed farming and harvesting, and bioplastics. The aim was to include experts from different countries and sectors, both academia and private sector. The interviews were conducted with the consent of the interviewees and all the interviews followed the same interview guide.

During the interviews, the experts were asked about their views on the prospects of increasing production, the techniques to improve productivity, the potential to upscale existing production and the limiting factors and challenges that need to be addressed to successfully upscale seaweed cultivation.

The interviews were analysed using a matrix approach to extract condensed meanings of quotes based on relevant keywords for each question. Key quotes were then summarised systematically, filtered, and organized based on constraints to produce a narrative about the constraints and opportunities to upscale brown seaweed production. More details on methods used during the interviews can be found in Ayala et al., (2023b).

2.3.2. Process simulations

Process simulation was deemed appropriate for the design and technical assessment of the processes, incorporating mass and energy balances. The simulation software Aspen Plus utilized lab-scale or pilot-scale operation parameters as inputs, which resulted in the calculation of energy flows, material flows, and elementary flows (Parvatker and Eckelman, 2019). In this study, the process simulations were conducted using Aspen Plus v9 (Aspen Technology Inc., 2023). The process model design results were used for simulating material and energy balances of chemical production, processing and chemical optimization, specifically to simulate the biorefinery and film fabrication life cycle stages. The biorefinery and film fabrication processes were simulated at scales of 4000 tonnes of bioplastic/operational year (t/yr.) in 2030 and 2 million tonnes/operational year (Mt/yr.) in 2035 based on growth projections from the bioplastic manufacturers within the project. The results from the simulation on Aspen were used to compile an industrial-scale inventory.

The process was simulated assuming ambient conditions ($P = 1\text{atm}$; $T = 25\text{C}$). The non-random two-liquid property method (NRTL) was used as thermodynamic model due to its ability to model the non-ideal nature of the interaction between compounds encountered in the feedstock. In light of the fact that the seaweed polysaccharides (alginate, laminarin and fucoïdan) are not included as standard compounds in the Aspen database, cellulose was used as a proxy to model them. For the model, the seaweed ashes were assumed to be composed of calcium chloride, given its inert nature as a salt. The selection of these proxies was made after consultation with experts in chemistry and seaweed processing.

The process simulation starts with a *mixer* simulating the grinder to mill the seaweed. At the pilot scale, continuous stirred tank reactors (CSTR) are utilized, and for this process simulation, they were represented as *stoichiometric reactors*. In these reactors, HCl and NaHCO₃ function as catalysts rather than reactants based on the established conversion rates of reactants to products. In reality, the acid and alkali additions serve to control the solubility of polysaccharides. At pH below the pKa of alginate (pH 3.6) fucoidans, laminarin and mannitol are soluble while alginate remains in the solid biomass, so the former components are removed from the biomass with the acid treatment and subsequent wash, in order to reduce the impurities in the alkaline stage. At high pH and in a solution rich in Na⁺ ions, sodium alginate becomes readily soluble, and becomes the primary component in the aqueous phase. The seaweed polysaccharides (alginate, laminarin, fucoidan, and cellulose) and sugars (mannitol) were modelled as solids. When the seaweed components are mixed with HCl, water, or NaHCO₃ in their respective stirred tank reactors, the seaweed components become solubilized and, in the model, transformed from a solid to a liquid state. The model includes two additions of these components, one represented as a solid and the other as a liquid. Each step in the CSTR was followed by a *centrifuge* to separate the solids from the liquids. *Mixers* were used in the crosslinking, compounding, and casting steps of the film fabrication to add inputs. *Calculator* unit operations were linked to all input streams, allowing them to scale up proportionally with the main flow. The last process step in the simulation is drying and this was modelled with a *compressor*, *dryer*, *flash tank* and *design specification*. The *compressor* was utilized as a simulation tool to introduce air, simulate a fan, and facilitate the drying process of the slurry mix. This process simulated the use of a fan, which enabled the movement of air necessary for the drying process. To calculate the amount of hot air required to reduce the moisture content of the product, the *design specification* tool was employed. Finally, the *multiplier* tool was used to increase all the flows to achieve the desired production rate. In this case, to obtain a specific annual production rate, i.e., 4000T and 2MT, by scaling up the input parameters accordingly. The flowsheet (Fig. 2) represents the processes modelled on Aspen Plus.

Supplementary Material A provides further details on process simulation performed on Aspen Plus, including the list of the modelled components, the assumptions of the proportions of the seaweed components, the efficiency of the equipment, the reactions in the CSTR, multiplying factors, details about the calculations carried out in the calculators and design specifications.

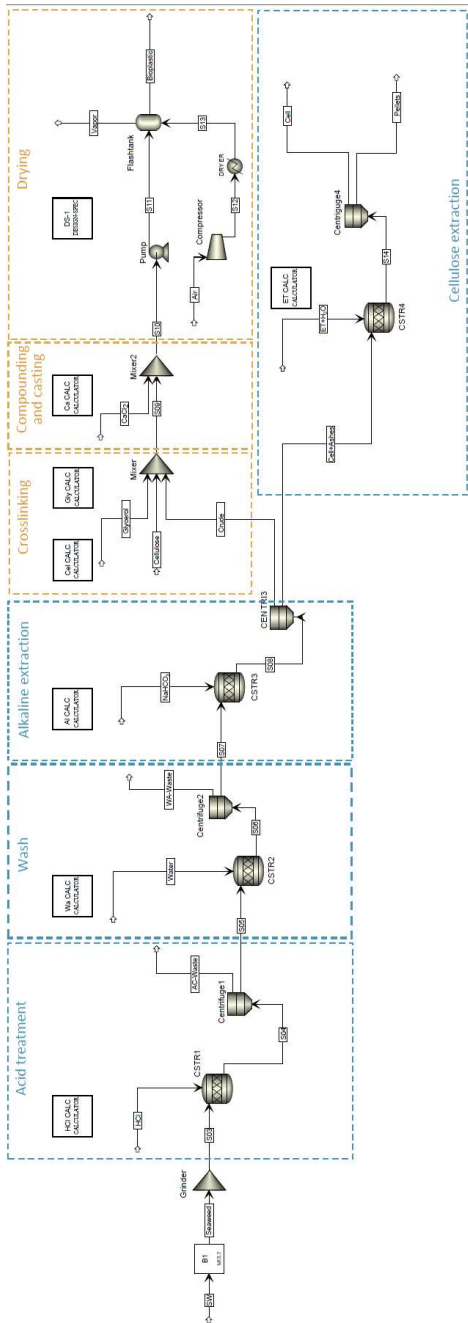


Fig. 2: Flowsheet of the process simulation on Aspen Plus with the steps in the biorefinery (blue) and film fabrication (orange).

3. Results

3.1. Seaweed cultivation and interview results

Based on the seaweed farmers' opinion, the hatchery was assumed to be scaled linearly (with a factor of 1) and the seaweed cultivation scaled with a factor of 0.8. The assumption behind these upscaling factors reflect the expectation that the hatchery is already efficient and will not be substantially different at larger scale, while the farming structure could be used for longer and larger vessels with higher capacity might be used in a large-scale cultivation. Increasing resource efficiency would therefore result in improved productivity and higher yields on larger scales, where cultivation processes and materials would be 20% more efficient. Supplementary Material C includes information regarding the life cycle inventory (LCI) and the results obtained from upscaling the seaweed cultivation based on the seaweed farmers' judgment, where the input processes are multiplied with a factor of 0.8.

To complement this upscaling, seaweed experts were interviewed to understand the complexity of upscaling seaweed cultivation. When experts were asked about the potential and the techniques to increase productivity, both inbreeding and outbreeding were mentioned by most experts as essential techniques. According to our respondents, a thorough understanding of the cultivation technology and the local environmental conditions was identified as key elements to increasing seaweed cultivation. That also includes using the existing knowledge on seaweed farming in other countries with well-established activities, namely China, Japan or Korea, or alternatively, in other sectors, such as fish farming.

To scale up seaweed production, the respondents stated that it is recommended to explore new areas for cultivation, including offshore and more exposed regions. Offshore cultivation involves submersible farms that can withstand adverse weather conditions, as well as the integration of seaweed farms with existing activities such as wind farms and oyster farms as part of integrated multitrophic aquaculture (IMTA). These techniques were mentioned in the interviews as viable options for offshore cultivation.

The mechanization of seaweed cultivation and harvesting was also emphasized repeatedly by the respondents as a critical factor in scaling up the production process. Experts mentioned that this would not only increase efficiency but also reduce labour costs. Long-term investments were also

highlighted as required for upscaling production. Additionally, the potential impact of large-scale seaweed cultivation on the carrying capacity of ecosystems and the availability of sea space was also identified by the respondents as potential limiting factors that need to be carefully considered, which is also discussed in the literature (Duarte et al., 2021). This limitation is related to the carrying capacity of ecosystems that could be affected by large-scale seaweed cultivations. Therefore, it is crucial to assess and monitor the environmental impacts of seaweed production to ensure that it remains within the limits of the carrying capacity of the ecosystems. Even if monoculture is a productive technique to have large-scale production, it can affect the local environment and the seaweed are likely to be more vulnerable to diseases. The use of fertilizers is known to enhance productivity, although it is not widely accepted or endorsed in most countries, especially in Europe. In conclusion, while the majority of experts acknowledge the potential for scaling up seaweed production, they also agree that accomplishing this goal will require a significant investment of time and resources. Additional insights regarding market prospects of seaweed cultivation, obtained from the interviews with seaweed experts, are available in Ayala et al. (2023b).

3.2. Process simulation results

The outcomes of the process modelling are presented concerning the overall mass and energy flows across the various process streams, including their elemental composition and potential utilization within the process. These findings indicate that scaling up to the 2Mt scenario can lead to improved resource efficiency compared to the 4000t scenario, as it achieves the same level of seaweed-based bioplastic production with fewer resources. This highlights the importance of considering larger-scale production for enhanced resource utilization and reduced environmental impacts.

Figures 3 and 4 present the results of the process simulation, providing insights into material flows and resource requirements between the two scenarios. In the 4000t scenario (Figure 3), the seaweed-based bioplastic flow accounts for 2.7% of the total material flow, while in the 2Mt scenario (Figure 4), it constitutes 4.8% of the material flow, indicating a wider flow and reduced material usage per unit of output. Notably, the seaweed input flow decreases in the larger scenario. Based on the process simulation results, the 4000t scenario requires 17.12 kg of seaweed per kilogram of seaweed-based bioplastic, whereas the 2Mt scenario only requires 9.57 kg of seaweed

per kilogram of seaweed-based bioplastic. These results demonstrate the potential for improved resource efficiency with larger-scale production.

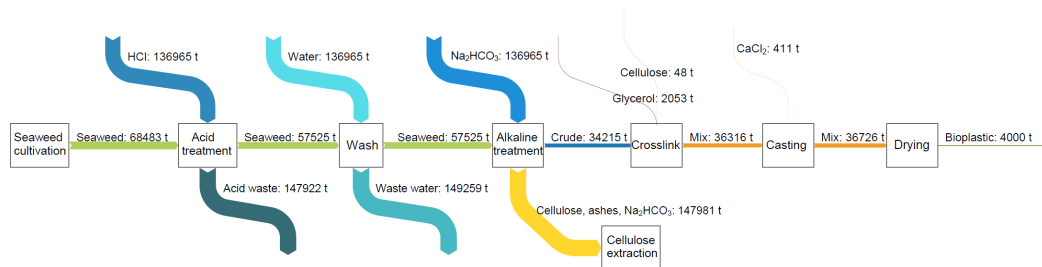


Fig. 3: Mass flow in the 4000t scenario resulting from the process simulation illustrating the inputs and outputs within the system.

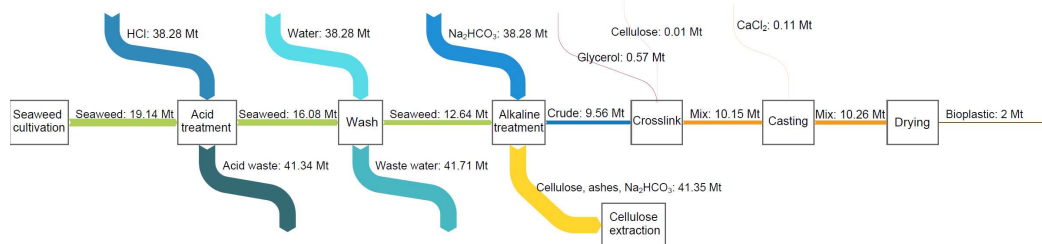


Fig. 4: Mass flow in the 2Mt scenario resulting from the process simulation illustrating the inputs and outputs within the system.

The extended results from the simulation including all the flows and unit operations can be consulted in Supplementary Material B. The results from the process simulation, together with the upscaling of the seaweed farm, are used as the LCI to perform the LCA at larger scales. The LCI is presented in Supplementary Material C.

3.3. LCA results

Figure 5 presents the contribution analysis of the LCA results for the 4000 t/yr. and 2 Mt/yr. scenarios in three impact categories: global warming potential, marine eutrophication, and freshwater eutrophication respectively. Each scenario is evaluated by accounting and not accounting for biogenic carbon. The values shown in the figure represent the contribution of each scenario to the respective impact category per functional unit (FU) of 1 kg of bioplastic.

As expected, we see a difference between accounting and not accounting for biogenic carbon as a contributor to climate change or climate change mitigation. Figure 5 illustrates the decrease in global warming impacts with increasing production scale due to increased efficiency. In the upscaled scenario of 2 Mt/yr., the bioplastic exhibits the lowest impact, with 1.37 kg CO₂-eq. per kg of bioplastic when accounting for the biogenic carbon. The impacts are higher when not accounting for the biogenic carbon, i.e., 3.85 kg CO₂-eq in 2030 and 2.49 kg CO₂-eq in 2035. The impacts increase in the smaller production scale scenario, 4000t/yr., 1.75 kg CO₂-eq. accounting for biogenic carbon. The highest impact contribution in all the scenarios is from the crosslinking step in the film fabrication stage, primarily due to the use of glycerol.

Regarding marine eutrophication and freshwater eutrophication, an important aspect to highlight is the nutrient uptake of phosphorus and nitrogen during seaweed cultivation. The impact in marine eutrophication is -0.017 kg N-eq. in the 4000 t/yr. scenario and -0.009 kg N-eq. in 2 Mt/yr. scenario, and in freshwater eutrophication -0.0037 in 4000 t/yr. and 0.002 kg P-eq. in 2 Mt/yr. This uptake process results in negative values, indicating a positive impact in reducing nutrient levels in marine and freshwater environments. The 4000 t/yr. scenario demonstrates the most significant positive impact since more seaweed is needed in this scenario. The extended LCA results with the contribution analysis in all the impact categories are available in Supplementary Material D.

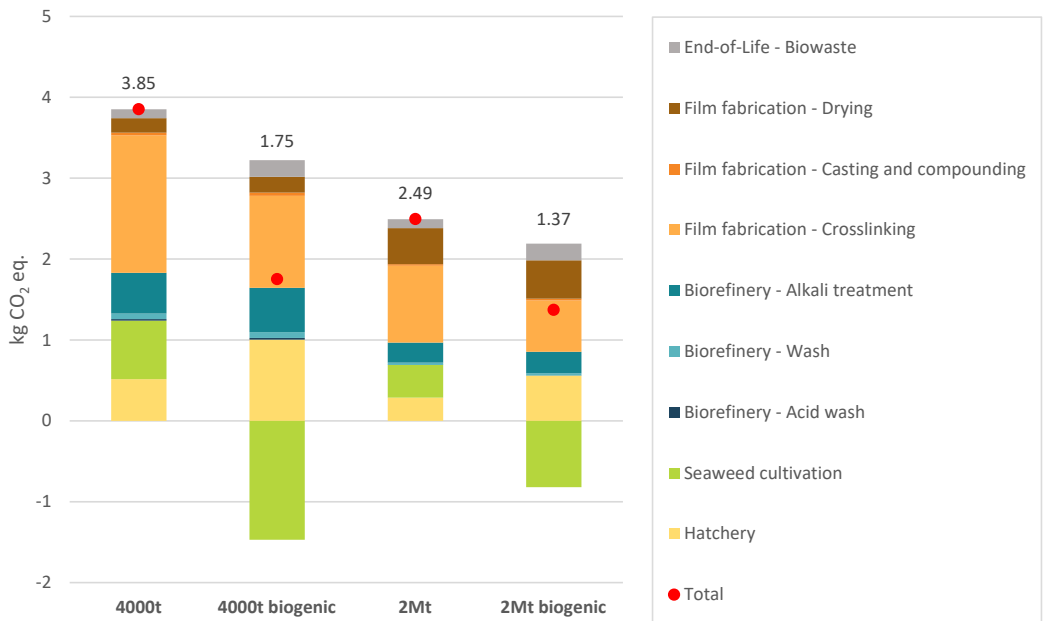


Fig. 5. Environmental performance of 4000 t/yr. and 2 Mt/yr. scenarios in the global warming potential impact category not accounting for biogenic carbon (i.e., carbon assimilation has been assigned a CF of 0) (4000 t/yr. and 2 Mt/yr.) and accounting for biogenic carbon and carbon uptake (4000 t/yr. biogenic and 2 Mt/yr. biogenic). The values presented in the figure represent the impact per FU of 1 kg of bioplastic.

4. Discussion

The results presented in this study show a trend towards lower environmental impacts with the increasing scale of the seaweed-based bioplastic production process. Specifically, the findings disclose that as the production volume of bioplastic increases, the environmental impact decreases. For instance, in the 4000 t/yr. scenario, the impact was measured at 1.75 kg CO₂-eq., while in the 2 Mt/yr. scenario, it reduced to 1.37 kg CO₂-eq. in global warming accounting for biogenic carbon. Data presented in figure 5 show that reduction in environmental impact scales together with the growth of seaweed-based bioplastic production. As it was the case for the pilot scale production (#cite your paper Ayala et al. 2022) The glycerol component has the highest impact contribution, especially in terms of global warming potential. Reducing the use of glycerol, such as optimizing

formulation ratios or substituting it with a compound with better environmental performance, can significantly decrease overall environmental impacts, particularly in the carbon footprint.

The adaptation of greener and renewable energy sources is recommended for the bioplastic production process. Although the biorefining and the product drying stages, which mainly require energy, do not significantly contribute to global warming, utilizing greener energy sources in these processes can further increase the positive environmental impact. In our study, we modelled the energy mix using the same assumptions as in Ayala et al. (2023), which indicates a predominant use of non-renewable energy in China, with a projected mix of 69% in 2030 and 65% in 2035. Despite the relatively small contribution of these activities to the overall environmental impacts, transitioning to a greener energy mix would help further reduce the carbon footprint of bioplastic production.

The findings in the marine eutrophication and freshwater eutrophication impact category are consistent with previous studies conducted by (Seghetta et al., 2016b; Thomsen and Zhang, 2020), supporting the argument that seaweed cultivation plays a significant role in reducing both marine and freshwater eutrophication. These studies have demonstrated that the uptake of nutrients by seaweed helps mitigate excess nutrient levels in aquatic ecosystems, thereby eutrophication.

When comparing the global warming impacts across the various scenarios, it is evident that all the scenarios exhibit lower scores compared to the pilot scale system. The impacts of the pilot scale system amount to 3.55 kg CO₂-eq. per kg of bioplastic accounting for the carbon uptake during seaweed cultivation and 4.72 kg CO₂-eq. without accounting the carbon uptake. Furthermore, when considering LDPE as a benchmark, the GW impacts of the seaweed-based system, including the EoL impacts, are lower at 3.6 kg CO₂-eq (Wernet et al., 2016). In addition to the lower global warming impacts observed in the seaweed-based system compared to the pilot scale system and LDPE, it is important to highlight that global warming is just one impact category among several. The bioplastic derived from seaweed cultivation offers additional environmental benefits in other impact categories, particularly in freshwater and marine eutrophication. The reduction of eutrophication impacts in both freshwater and marine ecosystems is a significant advantage of the seaweed-based system.

The present study recognizes some limitations and uncertainties that should be taken into consideration when interpreting the findings. As an emerging technology, the system's performance, and scalability are still under research and development and, thus, the estimated impacts on an industrial scale are unknown. An uncertainty analysis was outside the scope of the present study but would strengthen and nuance the interpretation of results.

Another aspect to consider is the potential biases and limitations in the methodology employed. Using Aspen software for process simulation comes with some limitations. The interpretation of simulation results may be challenging and a comprehensive understanding of the principles of the simulation is important for effectively interpreting the results. Additionally, the validity of assumptions made in the simulation, such as the modelled seaweed components or selected equipment to perform the simulation (e.g. steering tank reactor, decanter) can influence the accuracy of the results. Aspen software may not be well-suited to model certain types of processes, especially those systems involving biomaterials due to the lack of bio-based chemical data, such as alginate or laminarin. However, in this research, cellulose, which is available in the database, was used as a proxy for alginate, fucoidan and laminarin. That was considered a reasonable approximation to overcome the data limitation and enabled effective modelling of the biorefinery process, although it comes at the cost of less accurate data for the actual compounds employed during processing.

In the context of LCA, it is important to note that the current methodologies may not fully acknowledge the utilization of renewable seaweed biomass, as highlighted by (Zhang et al., 2021). This limitation means that the environmental benefits associated with the use of renewable seaweed biomass in the bioplastic production process may not be fully captured. Further advancements in the LCA frameworks are needed to encompass the full sustainability potential of seaweed-based bioplastics. The exclusion of biogenic carbon in the ReCiPe method, for instance, highlights a limitation of current LCA methods. Another example is that this novel bioplastic offers other advantages related to the cultivation of seaweed or the lack of microplastics produced in the degradation of this alginate-based material, which is not directly captured within the LCA methodology. It should be stressed that since this is a cradle-to-cradle LCA study the biogenic carbon is accounted for both in the cultivation stage (carbon uptake) and the film degradation stage (carbon release). This can be clearly observed in Fig. 5, where the scenarios accounting for biogenic carbon

show a negative value for the cultivation stage. However, there is no net carbon sink effect modelled in this study nor any misleading claim of the plastic being “carbon negative”: all the carbon is released again in a short cycle and mass balances are maintained (cf. also Ayala et al. (2023)). As a further limitation of the LCA, it should be noted that the measurement of microplastics generated from the degradation of alginate-based materials is not currently accounted for in LCA methodologies.

The choice of the 4000T and 2Mt scenarios in this study was based on different considerations. While the 2Mt scenario may be deemed impractical for implementation in a single plant due to its extremely large scale, it was included to test the upper limit and assess the efficiency of such a high-volume production, which is nevertheless still far from the scale of the current production of fossil-based plastic. By modelling both scenarios, we aimed to better understand the relationship between scale and efficiency and identify if a possible break-even point existed. Interestingly, the results demonstrated that the efficiency did not significantly differ between the two scenarios, indicating that even at a higher production volume, the overall efficiency remained relatively consistent. This suggests that the 4000T scenario, which is more realistic and achievable in terms of production volume at a single plant, still provides favourable outcomes in terms of resource utilization and environmental performance.

In order to further advance the understanding and applicability of the findings presented in this study, some areas for future research should be considered. Firstly, it would be beneficial to expand the scope of interviews beyond seaweed experts and involve a broader range of stakeholders and experts in the bioplastic industry. Engaging with professionals from various disciplines can provide valuable insights and perspectives on the challenges and opportunities associated with upscaling and a more comprehensive understanding of the upscaling process and its implications can be achieved. One suggestion is to further explore the combination of interviews and process simulations in prospective LCA, as this approach has demonstrated its potential in providing valuable insights.

One specific area for future research involves enhancing the representation of seaweed components, or other components of bioresources, within process simulation software. This would enable more accurate and reliable results by ensuring a better match between the modelled system and the actual process. Incorporating additional components into the Aspen database or refining

existing data can help improve the fidelity of the simulation and enhance the overall reliability of the environmental assessment.

Additionally, conducting a techno-economic assessment alongside the environmental evaluation would provide a more comprehensive understanding of the bioplastic production process. This assessment would consider not only the environmental impacts but also the economic feasibility and viability of implementing the technology, thus addressing the economic sustainability pillar. Such an analysis would consider factors such as production costs, market potential, and competitiveness, providing valuable insights for decision-makers and industry stakeholders. The social sustainability aspect would also require further attention and consideration. Integrating social factors such as labour conditions, community engagement, and societal impacts would contribute to a more holistic evaluation of seaweed-based bioplastics and ensure that all three pillars of sustainability are addressed effectively.

By addressing these areas for future research, it is possible to further refine and expand upon the knowledge gained from this study, leading to a more comprehensive understanding of the environmental and economic implications of bioplastic production from seaweed.

5. Conclusions

In conclusion, the study has provided valuable insights into the environmental performance of seaweed-based bioplastic production. The main findings reveal that the estimated environmental impacts, particularly in terms of global warming potential, are 1.75 kg CO₂-eq. in the 4000T scenario in 2030 and 1.37 kg CO₂-eq. in the 2MT scenario in 2035 accounting for biogenic carbon. The results do not only demonstrate that upscaling seaweed-based bioplastic production can effectively lower the environmental impacts per kilogram of bioplastic produced but also that the overall impacts from an LCA perspective are lower than the fossil-based LDPE.

Another important highlight from the study is how the combination of mixed methods, including interviews and process simulation, has proven to be appropriate in evaluating emerging technologies. By incorporating qualitative data through interviews alongside quantitative information, a more comprehensive assessment of the complexities and challenges involved in seaweed-based bioplastic production is achieved.

It is further important to acknowledge the uncertainties inherent in the assessment of emerging technologies. The immaturity and novelty of the systems under analysis translate into several unknowns about their future state, both regarding the technology itself and regarding its surrounding environment and socio-economic system, highlighting the need for ongoing research and development efforts to improve the accuracy and reliability of environmental assessments in this field.

CRedit authorship contribution statement

Maddalen Ayala: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization.

Neill Goosen: Conceptualization, Resources, Writing - review & editing, Supervision.

Leszek Michalak: Investigation, Data curation, Validation Writing - review & editing.

Marianne Thomsen: Conceptualization, Validation, Investigation, Methodology, Writing - review & editing, Supervision.

Massimo Pizzol: Conceptualization, Formal analysis, Investigation, Resources, Methodology, Validation, Writing - review & editing, Visualization, Supervision, Project administration, Funding acquisition.

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Supplementary Material

- A. Details of the processes modelled on Aspen Plus
- B. Aspen simulation results
- C. Life cycle inventory (LCI)
- D. LCA results - Contribution analysis

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APPENDIX D

ARTICLE 4

A supply-chain perspective on producing and upscaling bioplastic from brown seaweed

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A supply-chain perspective on producing and upscaling bioplastic from brown seaweed

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Abstract

Plastic pollution is an environmental emergency and finding sustainable alternatives to traditional plastics has become a pressing need. Seaweed-based bioplastic has emerged as a promising solution, as it is biodegradable, renewable, and seaweed cultivation provides various environmental benefits. However, the feasibility of implementing a brown seaweed-based bioplastic supply chain in a realistic setting remains unclear, as previous research focused either on single processing steps or on virtual supply chains aggregating data from different studies. This study describes the steps made to build a real-world supply chain of seaweed-based bioplastic within the PlastiSea research project: from seaweed cultivation and preservation to bioplastic development at the lab and pilot scale, thus providing insights into its feasibility. Adopting a multidisciplinary approach, the study employs multiple methods to characterize each stage in the supply chain and provides an overall life cycle assessment (LCA) as well as lessons learned throughout the process. The analysis shows substantial potential for producing multiple co-products from the same seaweed source. The use of residual biomass from processing seaweed for food as a source of alginates for producing bioplastics offers a low-cost and sustainable biomass supply currently not competing with other markets. Additionally, alginate crude extracts prepared by low-cost and low-energy intensive processing were found suitable for producing bioplastic materials similar to commercially refined alginate. The LCA results indicate the potential for reducing the environmental impact of seaweed-based bioplastic production through upscaling and increasing process efficiency.

1 Introduction

Seaweed cultivation is growing globally, especially in Northern Europe (Hasselström et al., 2020; Stévant and Rebours, 2017; van den Burg et al., 2021). Seaweed cultivation is increasingly being recognized for its positive impact on marine environments through carbon sequestration and reduction of eutrophication, as well as its potential for generating fast-growing and sustainable raw materials for biorefining (Prasad et al., 2022). In comparison to feedstocks derived from agriculture, seaweed has an advantage as it does not require arable land, freshwater, fertilizers, or pesticides (Atiwesh et al., 2021), and contributes to the nitrogen, phosphorus, and nutrient cycles (Seghetta et al., 2016). It can also provide a habitat for marine organisms, enhancing biodiversity and ecosystem services (Thomsen and Zhang, 2020). Moreover, it is a very versatile resource that has been utilized in various applications, including food, animal feed, medicine, cosmetics, and biofuel (Araújo et al., 2021).

One of the emerging applications of seaweed is bioplastics (Lim et al., 2021; Lomartire et al., 2022; Shravya et al., 2021; Zanchetta et al., 2021). The potential of seaweed-based bioplastic as alternative to fossil-based plastic lies in its biodegradability and renewability (Lim et al., 2021) (Thushari and Senevirathna, 2020) and its virtually absent use of arable land that minimizes its competition with food production (Zanchetta et al., 2021).

Existing studies on seaweed-based bioplastics focus on a particular aspect of the bioplastic life cycle, for example, the processing of seaweed feedstock or film fabrication (Albertos et al., 2019; Aragão, 2022; Helmes et al., 2018). Others have aggregated secondary data from different studies to envision a potential supply chain for the industrial production of bioplastic (Lomartire et al., 2022; Pacheco et al., 2022; Rinaudo, 2014; Shravya et al., 2021; Zanchetta et al., 2021). It remains however unclear whether processing and production steps that have been developed and optimized in isolation could be coupled together into a real-world supply chain. The environmental impact of a complete value chain of seaweed-based plastics is also underexplored. Thus, the feasibility and potential of producing seaweed-based bioplastics in real-world settings remains unclear. To address this knowledge gap, a more systemic perspective is needed, which is based on primary data covering an existing entire supply chain from seaweed cultivation and harvesting to biorefinery, bioplastic production, and disposal of the novel seaweed-based bioplastic.

The goal of the research presented here is to comprehensively analyze the supply chain for seaweed-based bioplastic, including all steps from seaweed cultivation and preservation, seaweed biorefining, to bioplastic development at the lab and pilot scale, as well as the co-products generated during the biorefinery and film fabrication processes (Figure 1). By providing a full overview of the entire supply chain based on a real-world implementation, coupled with simulation data also derived from real-world conditions, we developed a holistic perspective of the feasibility and sustainability of the emerging seaweed-based bioplastic. Through this approach, we were able to identify potential challenges, pitfalls, and key knowledge gaps that need to be addressed for seaweed-based bioplastics to become viable from a technological, commercial, and sustainable viewpoint.

This research is developed along the PlastiSea project (SINTEF, 2023), incorporating the latest level of knowledge on seaweed-based bioplastics. PlastiSea is an interdisciplinary research project aiming to develop seaweed-based bioplastics in the European context. Therefore, this research benefits from the latest research and development on seaweed-based bioplastics.

A comprehensive analysis of an existing bioplastic supply chain is presented, specifying the detailed processes and steps up to the production of the bioplastic (Figure 1), and based on a structured summary of existing findings from the project. Additionally, the study contains novel elements such as an evaluation of the potential of utilizing the stipe and holdfast fraction of cultivated seaweed, a by-product from the processing of biomass for food applications, to produce bioplastics. The performance of alginate from this fraction extracted in an established biorefinery process is compared with commercial alginate in the film fabrication step. The research is supported by the Life Cycle Assessment (LCA) of two extraction methods in the biorefinery and an LCA comparing the pilot scale production with prospective industrial-scale scenarios. Valuable insights into the feasibility and sustainability of seaweed-based bioplastics as a practical solution to the plastic pollution problem are gained through this approach.

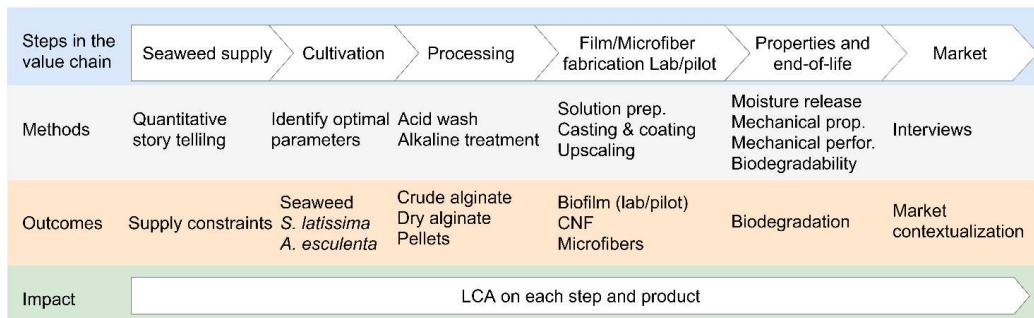


Figure 1: Overview of the supply chain considered in this study

2 Methods

2.1 Analysis of seaweed supply

As large-scale, global implementation of brown seaweed-based bioplastic will substantially increase the demand for brown seaweed, it becomes crucial to identify the suppliers that would respond to this future surge in demand.

Currently, China is producing approximately 13 million tons (wet weight) of brown algal biomass by cultivation per year, with food as the main target market (Cai et al., 2021). Even if China is the current main supplier, other emerging suppliers need to be considered to meet increasing demand and have a more local supply for biorefineries outside of Asia. The analysis thus focused on identifying suppliers that have the highest potential room to grow in the long term. In the industrial ecology domain, common methods to identify such “unconstrained” suppliers rely on numerical extrapolation from time-series data (Buyle et al., 2018). However, in the case of brown seaweed, statistical data are scarce and other factors, such as optimal growth conditions, regulatory constraints, and technological development in cultivation designs, also need to be considered. Thus, a mixed-methods approach was chosen as documented in Ayala et al. (2023b), and the “quantitative story telling” method, based on interviews and surveys with experts in seaweed cultivation and production was used to derive a set of suppliers likely to provide seaweed in the future.

2.2 Seaweed cultivation

In a context of growing demand for seaweed, investigating optimal seaweed cultivation conditions is necessary for scaling up production, which again is necessary to reduce costs per ton biomass and meet volume demands for bioplastics applications. Furthermore, an efficient use of the entire biomass needs to be achieved. The research involved the testing of cultivation conditions and working with both the entire seaweed and the holdfast fraction, which is currently discarded during processing for food applications, as it often carries contaminating marine organisms.

Two species of brown algae, *Saccharina latissima* (S.L.) and *Alaria esculenta* (A.E.) were cultivated and used as feedstock. The cultivation starts with a nursery phase, where mature zoospores are released from wild *parent algae*, which were collected at the cultivation site outside the island of Frøya in central Norway (63°42'15"N, 8°52'40"E). The zoospores develop into young sporophytes, which were seeded onto polyester ropes and grown until a size of 1-2 cm in the hatchery in Trondheim, Norway. After the seedlings were grown, they were transported to the cultivation site in Frøya (NO) and deployed on the sea farm. The S.L. and A.E. seedlings were deployed at different times: October 2020 and January 2021, and left to grow until spring 2021. The biomass was then sampled every 14 days to assess seasonal variation in algininate yield and structure. The biomass was frozen within 24 hours after harvest, either as whole algae, or the holdfast, which is the anchoring structure of the seaweed (Zhang and Thomsen, 2019), were removed and frozen separately before analyses and processing. Alginates have also been isolated from acid-preserved biomass as a more cost-friendly alternative to biomass freezing.

2.3 Seaweed processing

Once harvested, the seaweed feedstock needs to be further processed for subsequent use in bioplastic production. The processing includes extraction of alginate, where the seaweed first is washed with acid, followed by an alkali treatment to solubilize alginate. This alkaline "crude" alginate extract can be used directly as a low-cost substrate for bioplastic films. Alternatively, alginates can be precipitated from the extract and washed to reduce contaminants (minerals, proteins, other carbohydrates) and color. In the present study, both crude alginate extracts and refined alginates were prepared from the same batch of A.E. holdfasts, for comparison of structure,

composition, and performance in bioplastic materials. The residual solids after alginate extraction can be dried and used as a fiber-rich filler in composite materials or as a substrate for cellulose extraction, which can be used in bioplastics and other materials. The alginate extraction starts by treating the seaweed with 0.2 M HCl ("acid wash"), to remove impurities and keep alginate insoluble while fucoidan and other side stream soluble. The acid is then removed, and the seaweed is rinsed with ion-free water. Then, the seaweed is treated with 0.2 M NaHCO₃ (alkaline treatment) to convert insoluble alginic acid to water-soluble Na-alginate, which is extracted from the seaweed. The alginate extract is separated from the solids by centrifugation and precipitated with 50 % ethanol or by lowering the pH below the pKa of alginate (3.5). Further washing of the precipitated material can be done with acid and/or alcohol depending on the required purity for the alginate. Previously work by the authors (Nøkling-Eide et al., 2023a) has investigated conditions for maximizing alginate yield under scalable, low-energy processing conditions that maintain the quality of the alginates and allow extraction of multiple components in an integrated biorefinery. This has been further elaborated in the present study, where different ratios of seaweed to liquid ratios (1:2, 1:4, 1:6, and 1:8) were evaluated to allow volume- and energy-efficient scaling. Additionally, as the molecular weight (MW) and thus viscosity of the alginate is an important parameter for subsequent processing and bioplastic formulation, variation of the temperature and time during the acid wash was investigated to tune the MW of extracted alginates. The experiments were conducted in 50 mL tubes with 5 g of frozen milled biomass (whole plant or holdfast fractions), in triplicates.

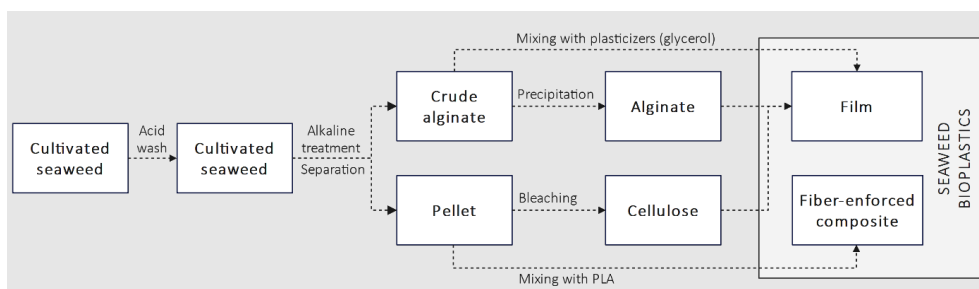
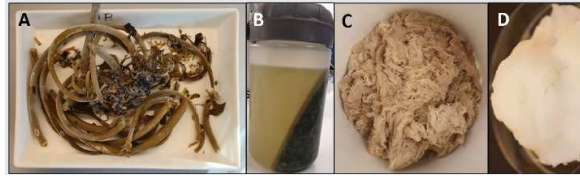


Figure 2: Overall process from seaweed biomass, via alginate and cellulose extraction, to final bioplastics (films or fiber-reinforced composites).



*Figure 3: Seaweed biomass and its four products applied in seaweed bioplastics. A) Holdfast fraction from *S. latissima*, B) Crude alginate and pellet separated by centrifugation, C) Alginate, and D) Cellulose.*

2.4 Bioplastic development at the lab scale

Bioplastic films

The alginates extracted in the processing step were mixed with solvents, additives, and plasticizers (water, cellulose nanofibers, and glycerol, respectively) to form films. The research focused on defining suitable fabrication protocols and optimized process parameters to obtain homogeneous films in a reproducible manner using both refined alginates and crude extracts mainly to save processing time and costs. The lab scale development work on film fabrication included three steps: film forming solution preparation, processing with casting and coating methods, and characterization (assessment of moisture release, mechanical properties, and biodegradability).

Specifically, two types of alginates were tested to produce the bioplastics in this study: refined Sodium Alginate in the dry state, and crude extracts solutions in water (both commercial ones and extracted in the biorefinery from the previous step). Two main processes were employed to obtain non-soluble films from the film-forming water-based solutions: a casting method and a coating method. The casting method consisted in casting 100 mL of film-forming solution in a 25cm x 25cm petri dish and letting it dry overnight. Once the film had dried, 100 mL of crosslinking solution was poured onto the dish and left for 10 seconds. Thereafter, the crosslinked films were rinsed with 100 mL of water for another 10 seconds and then left to dry at room temperature. When using the coating method, the solution was poured onto a Plexiglas board and spread by hand with a blade coater to form a homogeneous layer. The crosslinking solution was sprayed over the wet film surface and the film was dried at room temperature. See Figure 4 for a schematic depiction of the two processing techniques described.

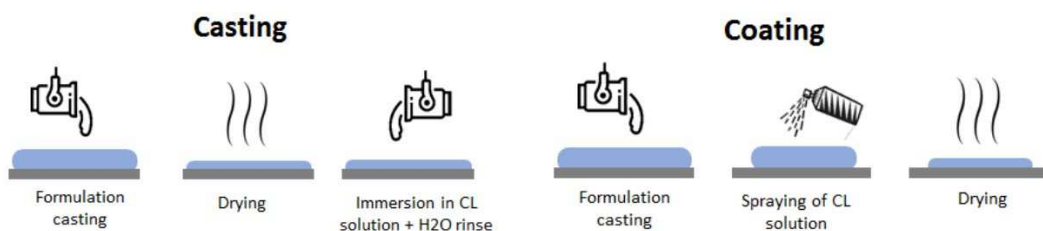


Figure 4. Schematic representation of casting method and coating method respectively.

In order to understand how different formulations and processing parameters can affect the final properties of the seaweed-based bioplastics, the films fabricated were characterized, focusing specifically on mechanical properties and their dependence on the crosslinking degree and cellulose content, and moisture absorption and release. Finally, a preliminary study on the film biodegradability was performed by benchmarking seaweed-based bioplastic with petroleum-based alternatives such as low-density polyethylene (LDPE). Further details on specific protocols and methods used can be found in Supplementary Material.

Bioplastic microfibers

Cellulose constitutes a substantial fraction of the whole seaweed biomass. It is, therefore, important to understand what use could be made of such fraction in the context of bioplastic production. Seaweed cellulose nanofibrils (CNFs) that are three orders of magnitude smaller than the extracted seaweed cellulose fibers can be produced on a large scale, the same as those from wood pulp fibers. We developed a technology to utilize the high mechanical performance of CNFs, obtained from seaweed processing residues, combined with alginate from biorefinery of cultivated seaweed *A.E.*, for the fabrication of all seaweed-based bioplastic microfibers of high performance. The work included three steps: preparation of cationic CNFs, preparation of CNF/alginate composite microfibers, and characterization. The experimental details are summarized in SM. The extraction of seaweed cellulose is described in the above section (2.3 *Seaweed processing*) and in the literature (Nøklung-Eide et al., 2023b). The cationic CNFs were prepared from extracted cellulose following a previously reported method (Pei et al., 2013). CNF/alginate composite microfibers were produced by the interfacial polyelectrolyte complexation (IPC) drawing method (Grande et al., 2020), in which neither coagulation solvent nor specially designed spinning device is required. To optimize the

drawing process of CNF/alginate composite microfibers, the effect of the cationic charge density of CNFs on the microstructure and mechanical performance of the composite microfibers using commercial alginate was investigated, providing important information for possible further scale-up production with alginate from the *A.E.* biorefinery. The mechanical performance of the CNF/alginate composite microfibers was compared with other microfibers obtained from various bio-based or biodegradable polymers to elucidate the significance and application potential of the seaweed bioplastic microfibers.

2.5 Bioplastic development at pilot scale

Formulations and seaweed sources characterized and validated at laboratory scale were taken to the pilot scale to manufacture the bioplastic in a production line that is closer to real-world implementation. The transition from the laboratory to the pilot scale presents several challenges, process continuity being the most demanding and critical aspect to address. Standard plastic manufacturing technologies used in the packaging sector such as extrusion, film blowing, thermoforming, and injection molding, are highly time-cost efficient, since these processes are nowadays highly optimized. Alongside the manufacturing technologies, fossil-based polymers such as LDPE or PP have also gain high standards in terms of performance and efficiency. Nevertheless, industrial biobased materials such as PLA, or any of the newly developed polymers are already able to perform at such level and compete with fossil-based ones in the market (Porta et al., 2022). To address these challenges, the possibility of achieving a continuous process that can efficiently handle the targeted seaweed-based material was investigated. The processes already implemented at the lab scale were analyzed, and the bottlenecks for their scalability were identified. Among these, the evaporation of water emerged as the most critical point affecting the efficiency and time-cost effectiveness production. It is key to remark that polymers only spend a few minutes being processed by industrial machinery, making time for evaporation the main issue in the processing of seaweed-based films. To produce the seaweed-based films, the pilot scale developed machinery set the thickness of the deposited solution at around 600 μm and width of 25 cm, and used nine of the 10 meters of length of the machine to the evaporation process, which was tested through the combination of different state-of-the-art drying processes, i.e. hot air, infra-red and drying air.

In a second experiment seaweed pellets, which are the co-products from the alginate extracted in the biorefinery, were incorporated as reinforcement into polylactic acid matrix (PLA 3251 D) through conventional extrusion processes (Coperion ZSK 26 twin screw extruder) with a temperature profile between 180°C and 190°C, validating the use of natural fibers coming from seaweed as reinforcements for the production of parts through injection molding (BOY 25 E), where the temperature profile was going from 155 °C to 185 °C and 3D printing (Ender 3 with an V4 pellet extruder).

Mechanical characterization for the composites generated was done on specimens manufactured through injection molding. The tensile characterization was performed following the ISO 527, the flexural characterization following the ISO 178 and the impact characterization following the ISO 179 guideline.

2.6 Analysis of market potentials

Seaweed experts were interviewed to understand the current seaweed market and assess the potential of the novel bioplastic in the market. The interviews were conducted with 11 experts of diverse backgrounds and nationalities, primarily from Europe and Asia, to obtain qualitative information about the constraints in the global brown seaweed suppliers and the biomass utilization and market niche for brown seaweed bioplastic. The selection of experts was based on their extensive knowledge of seaweed cultivation, industry experience, and academic background. More details are provided in Ayala et al. (2023b).

2.7 Life cycle assessment of bioplastic production

The environmental performance of the value chain can be assessed through an LCA following the four step-procedure indicated in the guidelines set by ISO 14044:2006. ReCiPe 2016 Midpoint (E) was used in the analysis, modeled on Simapro v9.2.0.2 with the ecoinvent v. 3.8 background database, modifying the characterization factor to include biogenic carbon. The LCA was performed using the consequential approach, which aims to assess the potential consequences of changes in the system being analyzed and the implications of demanding of the product (Ekvall and Weidema, 2004). Two LCAs were performed: A comparative LCA of the production of crude alginate and dry

alginate, and a comparative LCA of pilot production and upscaled production. All the LCA models were built based on primary experimental data from this research.

The goal of the first LCA is to compare the production methods to extract alginate to assess the best-performing method from an environmental perspective. The functional unit was 1 kg of alginate. The volumes of alginate in the crude extract and dry alginate were based on (Nøkling-Eide et al., 2023a). The system boundaries include hatchery, seaweed cultivation, and harvest and biorefinery.

In the pilot and upscaled production LCA, the goal was to evaluate the environmental impacts in the entire value chain of the bioplastic, from seaweed cultivation to end-of-life disposal options. The functional unit was defined as 1 kg of seaweed-based bioplastic film, and the system boundaries include all the stages in the value chain, namely seaweed hatchery and cultivation, biorefinery, film fabrication, and end-of-life (EoL). The process simulation software Aspen Plus was used to upscale the processes to later use the upscaled values as input for the LCA on an industrial scale. Two prospective upscaling scenarios were modeled. 4000 tons in 2030 and 2 million tons in 2035. Finally, the EoL was modeled as composting. Further details are available in Ayala et al. (2023a).

3 Results and discussion

3.1 Constraints to supply of seaweed on a global scale

During the last two decades, Western countries including Spain, Portugal, Faroe Island, Iceland, Norway, Scotland, Ireland, Denmark, Sweden, United States, Canada, and more have started to cultivate seaweed (Bak et al., 2018; Forbord et al., 2020). The volumes are still low, but with an estimated potential of increasing the production of cultivated seaweed in Europe to 8 mill tons by 2030 (Vincent, 2020). According to the findings from (Ayala et al., 2023a), China is currently the primary marginal supplier of brown seaweed and is anticipated to maintain this position in the mid to long term. Meanwhile, Northern Atlantic brown seaweed suppliers, although still in their early stages of development, are projected to experience significant growth in the future. These results suggest that there are two distinct groups of countries that are expected to become future brown seaweed suppliers for different reasons. While Asian countries already have established brown seaweed aquaculture and are expected to continue growing, Northern Europe and North American

seaweed producers are emerging as strong competitors due to their production capacity and technological advancements.

The supply of seaweed on a global scale is constrained by various factors. Firstly, while there are no significant regulatory constraints for global trade, local markets often prefer locally sourced seaweed. Secondly, the carrying capacity of ecosystems and understanding optimal cultivation conditions are important to obtaining high yields and increasing the seaweed production. Lastly, the technology for cultivating and harvesting seaweed is emerging and it needs to be optimized to upscale. Addressing these constraints will require collaborative efforts to establish sustainable and efficient seaweed supply chains.

3.2 Use of cultivated seaweed as a bioplastic substrate

In 2022 a total of 221 tons of cultivated *S.L.* and *A.E.* were harvested in Norway (Directorate of Fisheries, 2023) and the estimated production in 2023 is around 500 tons. Seaweed Solutions has since 2015 been harvesting seaweed (*S.L.* and *A.E.*) in the 50–20-ton scale. In 2020 the harvested volume was 60 tons wet weight, 150 tons in 2021, and 200 tons in 2022 and 2023. Given the growing demand for cultivated seaweed, Seaweed Solutions, the seaweed provided in the project, is planning for a larger up-scaling in the next 2-5 years period, aiming at reaching 1000 tons within this period of time.

The analysis of the holdfast fraction of *A.E.* and *S.L.* showed that despite a lower fraction of guluronic acid (F_G) and shorter G-blocks ($N_{G>1}$) in the alginates from holdfast fraction compared to the whole seaweed (Table 1), these biomasses are giving biofilms with promising properties (see Figure 5). Deployment and harvesting date of the cultivated seaweed also seem to influence the yield and content of guluronic acid (F_G) in the extractable alginate (Figure 5) and should thus be taken into consideration when developing bioplastics. The alginate from *S.L.* and *A.E.* that are harvested at the end of the seaweed season (late May-June) represents higher yields and higher F_G , respectively. Late harvest of the seaweed could therefore be a good alternative when developing bioplastics, especially considering that fouling and less delicate mouthfeel is a challenge when using late-harvested seaweed for food applications.

Table 1: Average fraction of guluronic acid (F_G), mannuronic acid (F_M), all possible diads, the triad of guluronic acid (F_{GGG}), G-blocks (F_{GGM} , F_{GMM}), single Gs (F_{MGM}) and the average length of G-blocks ($N_{G>1}$) in alginates from whole *S.L.* and *A.E.* and their associated holdfast alginates. The fractions are based on the characteristic signals for M and G residues obtained from the anomeric region in their recorded NMR spectra.

Alginate	F_G	F_M	F_{GG}	$F_{GM, MG}$	F_{MM}	F_{GGG}	$F_{GGM, MGG}$	F_{MGM}	$N_{G>1}$
<i>S. latissima</i> whole	0.47	0.53	0.27	0.20	0.33	0.24	0.03	0.16	9
<i>S. latissima</i> holdfast	0.42	0.58	0.24	0.19	0.39	0.18	0.05	0.14	6
<i>A. esculenta</i> whole	0.55	0.45	0.41	0.14	0.31	0.38	0.03	0.11	14
<i>A. esculenta</i> holdfast	0.44	0.56	0.29	0.16	0.40	0.24	0.04	0.12	8

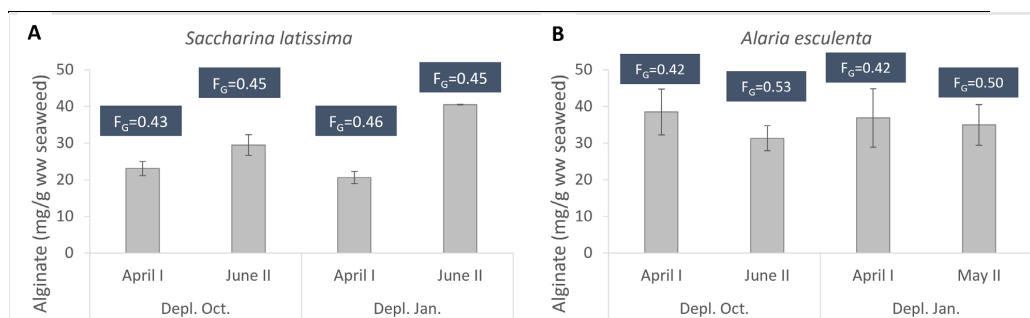


Figure 5: Yield (plotted as mg dry alginate/g wet weight seaweed) and fraction of guluronic acid (F_G) in alginate extracted from *S.L.* (A) and *A.E.* (B) deployed in October or January 2021, and harvested in early April (April I), late May (May II), or late June (June II). Alginate is extracted during two rounds of alkaline treatment, following a 20-hour acid wash.

3.3 Seaweed processing

Alginate yield and quality are key considerations for developing biorefinery processes for the utilization of cultivated seaweed in bioplastic applications. These are dependent on seaweed species and to an extent seasonal variations, but primarily on the processing conditions. In a previous study

by the authors (Nøkling-Eide et al., 2023a, 2023b) , experiments varying pH, extraction time and temperature on the yield and structure of alginates from *S.L.* and *A.E.*, showed that high-Mw alginates are efficiently extracted under relatively mild conditions compared to other species used in commercial alginate production, due to structural differences in the biomasses.

These high-Mw alginates confer significant viscosity to the alginate extracts, complicating separation from the seaweed residues. In the present study we found that extracting alginates using a seaweed-alkali ratio of 1:2 or less liquid resulted in significantly reduced yield (Figure 6). As a way to circumvent this issue and still retain a high alginate concentration in the crude fraction, increasing the temperature during the acid wash was found to reduce the alginate Mw in a controlled manner by partial hydrolysis of glycosidic linkages (Figure 7). Another insight from previous work is that the molecular weight of alginates is reduced during storage of acid-preserved biomass, which will be a necessary approach to ensure a year-round biomass supply and prevent processing bottlenecks at a lower cost than freezing or drying (Nøkling-Eide et al., 2023a). Increasing process efficiency and utilization of side streams will be important for economic feasibility and scaling, and retrieval of water-soluble polysaccharides in an integrated biorefinery has been demonstrated in previous work (Birgersson et al., 2023). While not directly relevant for bioplastic applications, side stream products such as fucoidan and laminarin can have other high-cost applications increasing the valorization of the biomass.

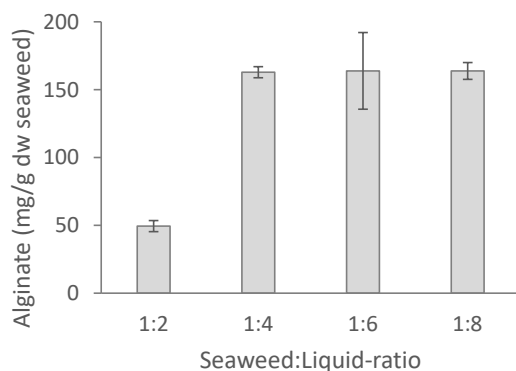


Figure 6: Alginate yield from different ratios between seaweed (wet weight) and alkali (NaHCO₃).

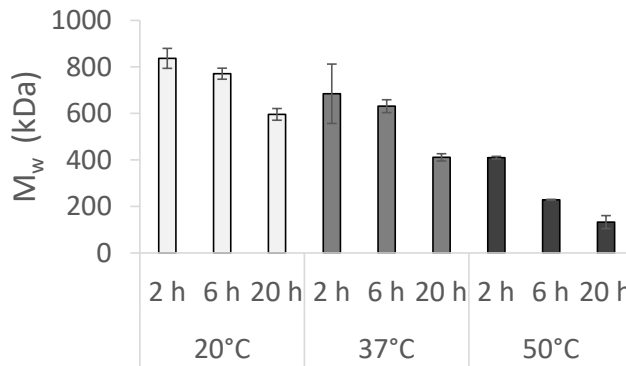


Figure 7: Weight average molecular weight (M_w) of alginates extracted from *A.E.* with different temperatures (20, 37 and 50 °C) and different extraction times (2, 6 and 20 hours) in the acid wash.

3.4 Bioplastic development at lab scale, main findings

Bioplastic films

Insights on optimizing film fabrication

The concentrations of Sodium Alginate (SA) and Micro Fibrillated Cellulose (MFC) were carefully chosen in order to obtain solutions with a viscosity of between 0.1 to 0.3 psi. These concentrations were optimized since a fundamental parameter that can dramatically affect the homogeneity of the film and processing throughput is the concentration of the precursor solution, which directly affects its viscosity. If the solution has an excessively low viscosity, this will cause the casted solution to be extremely sensitive to possible slopes or tilts of the casting surface, leading to relevant variations in the final thickness of the film or even to solution dewetting from the underlying substrate. If the solution is too viscous, this might lead to partial gelification preventing homogeneous spreading. The viscosity of alginate solution typically depends on Mw, G/M ratio, and concentration. For the G/M ratio and the Mw tested in this work (between 150 and 500 kDa), the optimum alginate concentration in the precursor solution identified to obtain homogeneous films ranged between 1.5 and 2.5 wt%.

The mixing time of the polysaccharides in water was limited to 10 minutes since a long time led to overheating of the solution and consequent loss in viscosity, attributed to the breakage of molecular chains by heat and stress-induced degradation (Oates and Ledward, 1990). Similar behavior was observed for Sodium Alginate solution stored at room temperature for more than 48h, hence in this study, the film-forming solutions were used immediately after their preparation. Finally, the weight ratios among Sodium Alginate, MFC and glycerol were selected in order to obtain flexible and easy-to-handle films after the crosslinking process (Giz et al., 2020; Paula et al., 2015; Rhim, 2004; Russo et al., 2007; Sirviö et al., 2014). Figure 8 depicts films fabricated via the coating technique using *S.L.* dry alginate extracted in the biorefinery step and using purified commercial alginate. Both biomasses were suitable to fabricate smooth and homogeneous films, with sufficient mechanical properties for handling, nevertheless, several differences between the two films were identified.

Refined commercial alginate provided smooth, transparent to translucent films, depending on the number of cellulose fillers used in the formulation, while using dry alginate from the biorefinery, the films obtained were semi-transparent and colored, with more or less intense colors ranging from yellowish to brownish to greenish. Film coloration depended on the seaweed species and part of the seaweed used, and small inclusions are visible in the film depending on the degree of purity of the alginate extract. Finally, films fabricated using the alginate extracted in the biorefinery also presented a more intense “sea” smell. While these aspects give the final bioplastic film a “natural” appearance, they also might represent a limitation that should be addressed (e.g., using bleaching strategies) for their use in certain commercial applications.



Figure 8: Seaweed based films fabricated with alginate from the biorefinery (SINTEF, left) and highly refined commercial alginates (Algaia, right).

Findings about mechanical properties of the biofilms

Alginate water solutions exposed to cationic divalent ions form hydrogels. During this process, the G and M blocks belonging to the alginate backbone arrange themselves around the positively charged ions following the so-called egg box structure (Ardiles and Rodríguez, 2021). This ionic crosslinking of the polymeric matrix typically results in stiffer and tougher films once the hydrogel dries out.

The crosslinking degree and therefore the final mechanical and chemical properties of the obtained films depend on several factors, such as the molecular structure of sodium alginate (Mw, G/M ratio), the chain's mobility (i.e. type of solvent and concentration of the alginate in solution), concentration and type of divalent ions, among others (Costa et al., 2018). This is well documented in literature (Benavides et al., 2012) and a full understanding of the effect of these parameters on the final properties of the films goes beyond the scope of this article. However, for reproducible processing and rigorous characterization of alginate-based composite films, it is fundamental to control the ionic crosslinking degree during film fabrication. The stiffening effect induced by the formation of the egg box structures can dramatically affect the mechanical properties of the produced films, hindering the effect of other phenomena or misleading the correct interpretation of data.

In order to provide a useful guideline for the correct characterization of this class of bioplastic materials and of their composite, in this study we highlight the effect of two main processing parameters that affect the crosslinking degree of the film, and hence the mechanical properties. These would be (1) the number of cationic ions the alginate matrix is exposed to and (2) the film thickness.

To systematically study the effect of the amount of crosslinking solution on the final crosslinking degree of the film, different film-forming solutions were sprayed with an increasing amount of crosslinking solution. The final concentration of Ca²⁺ ions in the films was considered proportional to the amount of crosslinking solution sprayed assuming homogeneous dispersion of the Ca²⁺ ions, as schematized in Figure 9.

It can be observed from Figure 9 (a) that increasing the amount of crosslinking solution to which the liquid precursor is exposed, will bring a steady increase of the film stiffness (Young modulus), while

tensile strength will reach a plateau. It is worth underlining that particularly for mild crosslinked films (between 0 and 10 sprays) just by doubling the amount of CL solution a 10-fold increase in Young modulus was observed. It can be concluded that the mechanical properties of the films are extremely sensitive to small changes in the film crosslinking degree. If on the one hand, this can be a powerful tool to easily tune the final film properties, on the other hand, when using the coating method careful control of the amount of crosslinking solution sprayed on the film is, therefore, a fundamental point for experiments aiming to analyze the effect of other parameters (i.e., changes in formulation) on films mechanical properties.

Film casting is another widely used method to fabricate films at a laboratory scale, however, thickness control is scarcely mentioned in other works in literature. Figure 9 (b) shows the mechanical properties obtained on casted films with equal crosslinking protocol, and different initial thicknesses. It can be observed that films present on average a lower stiffness compared to films fabricated using the coating method and that the stiffness also strongly depends on initial film thickness. This can be easily explained considering the Ca^{2+} ions will only partially diffuse from the exposed surface to the rest of the film during immersion in the crosslinking solution. This will result in a lower crosslinking degree of casted films compared to coated ones and also in the fact that thinner films will have a higher crosslinking degree compared to thicker ones, as schematized in Figure 9. During the casting method two competing phenomena occur: on one hand the films crosslink, becoming insoluble, but on the other hand the soluble components in the liquid precursor, among which glycerol and non-crosslinked Sodium Alginate, will dissolve into the CL solution. These phenomena will further contribute to the stiffening of the film, since glycerol acts as the plasticizer of alginate films (Avella et al., 2007; Paixão et al., 2019).

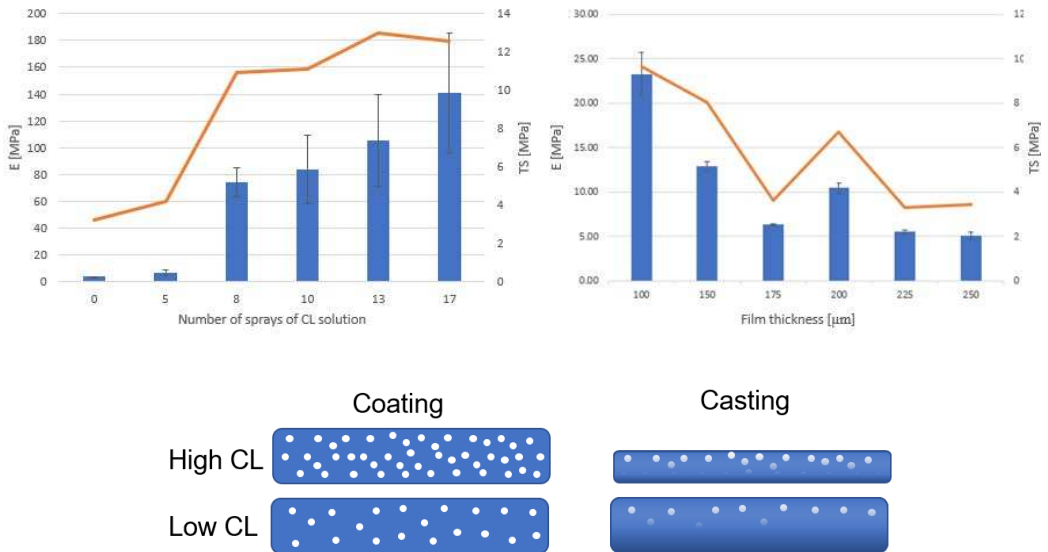


Figure 9: Mechanical properties of coated (left, (a)) and casted (right (b)) films with different crosslinking process parameters. Schematic of crosslinking degree in coated and casted films. Ca²⁺ ions are represented by white dots.

Film thickness control can be affected by both specific components in the liquid precursors and by processing parameters (concentration, solid content, type of additives, type of alginate, total volume cast). From the results on mechanical properties of casted films with different thicknesses (Figure 6), it can be concluded that if film thickness is not carefully controlled it is impossible to obtain reproducible and reliable results for further comparisons. Figure 10 shows the effect of adding an increasing content of MFC on the mechanical properties of alginate films. The well-known tendency of MFC to enhance the stiffness of alginate films (Sirviö et al., 2014) can be clearly seen in the top section of Figure 10(Sirviö et al., 2014), where films with the same thickness were prepared and characterized. When film thickness was not controlled (Figure 10, bottom section) an opposite trend was observed. This can be explained by attributing a stronger effect to the crosslinking degree compared to the reinforcement effect obtained by increasing the content of MFC.

Sample	MFC0	MFC1	MFC2	MFC3	MFC4	MFC5	MFC6	MFC7
Cellulose content (wt%)	0	1	2	3	4	5	6	7



Figure 10: Effect of MFC cellulose on film properties with (top) and without (bottom) thickness control.

Findings on moisture content and biodegradability

Results of the moisture content analysis showed that alginate films release up to half of their water content during the one hour and up to 1/3 during the first 30 minutes, following a quasi-linear decay. It is therefore of paramount importance that the mechanical characterization is performed during the first 5 to 10 minutes after the condition if the RH in the measuring environment differs from the one used for conditioning.

The disintegration test showed that after 2 months only 50% of weight loss was reached. To be considered biodegradable, the disintegration should be at least 90% after 6 months, so this preliminary result can be considered as a promising indication of the high biodegradability of seaweed-based films (cf. SI for images of degradation tests).

Bioplastic microfibers

As revealed by atomic force microscopy analysis (Figure 11), nanoscale discrete cationic CNFs were successfully prepared from the micrometer scale cellulose extracted from seaweed processing residues by chemical modification followed by mechanical disintegration. The width of the cationic CNFs was 4–9 nm and the length was in the range of 500 nm to 3 mm. The water suspension of the cationic CNF was transparent and colloidally stable (Figure 11). Composite microfibers from cationic CNF and alginate (Figure 12a) were successfully prepared by the IPC drawing method from the suspension of CNFs and solution of alginate. The results from the commercial alginate suggest the preparation of high-performance microfibers favors cationic CNFs with high charge density, which simultaneously improved the ultimate tensile strength and Young's modulus of composite microfibers owing to the enhanced interaction between cationic CNFs and alginate (Supplementary Material). By using alginate from the biorefinery of *A.E* and cationic CNFs of high charge density, both ultimate tensile strength and Young's modulus of microfiber were improved, compared to using commercial alginate, owing to the lower molecular weight of *A.E*. alginate and higher orientation of CNFs in the composite microfiber. The mechanical performance of CNF/alginate composite microfibers was of high significance when compared to microfibers prepared from various bio-based or biodegradable polymers (Figure 12 and Table S5, Supplementary Material) including alginate and chitosan from marine biomass, regenerated silk, bovine serum albumin and collagen from animal tissue extracts, polylactic acid and polyhydroxybutyrate from the fermented products of carbohydrate; polycaprolactone, a biodegradable polymer which can be produced from biomass, such as soybean oil. The tensile strength of CNF/alginate composite microfibers (299 MPa) was among the highest, comparable to BSA (279 MPa) and collagen (383 MPa). It also showed Young's modulus of 13.0 GPa, the highest among all listed microfibers. This bioplastic microfiber is produced entirely with the products from the seaweed biorefinery process and can be readily functionalized during the microfiber drawing procedure by incorporation of functional components

in the polyelectrolytes (Nechporchuk et al., 2017). These results highlighted the great potential of CNF/alginate composite microfibers as a green and strong alternative to the existing bio-based microfiber products, which find applications not only in textile, packaging, and food industry but also in various biomedical scenarios such as wound dressing, tissue engineering, and drug delivery (Wan et al., 2016).

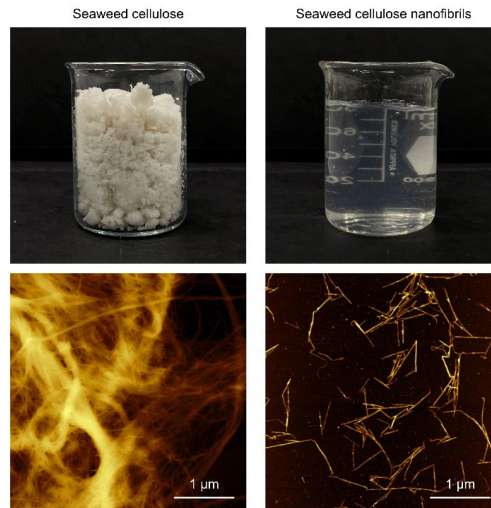


Figure 11. Photographs and atomic force microscopy images of the extracted cellulose from seaweed processing residues and the prepared cationic cellulose nanofibrils (CNFs).

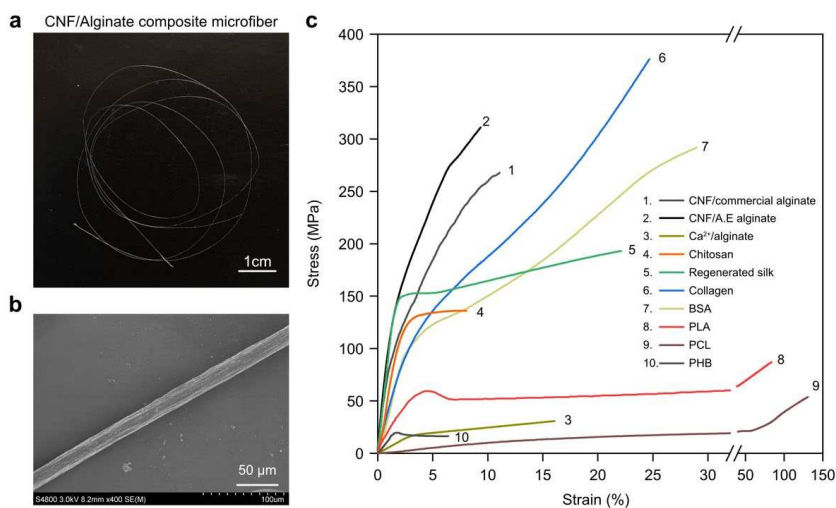


Figure 12. CNF/Alginate composite microfibers. a) Photograph of a single thread of the CNF/Alginate composite microfiber. b) Scanning electron microscopy image of the CNF/Alginate composite microfiber. c) Tensile stress-strain curves of CNF/Alginate composite microfibers as compared to other spun microfibers from various bio-based or biodegradable polymers (Mirabedini et al., 2015).

3.5 Insights from bioplastic fabrication at pilot scale

Two types of bioplastic materials were produced at the pilot scale. The first type is alginate-based films, obtained with an ad-hoc design machinery that allows to manufacture continuously the films with the coating methodology (Figure 4).

The conventional plastic industry usually employs dried materials instead of solutions. In fact, the industry currently relies on melting capacity of thermoplastic polymers, whereas in the case of this novel bioplastic film, it is dissolved in water. That implies that when upscaling the production of alginate films at a market level, significant modifications to the current machinery or acquisition of new equipment were necessary.

In particular the production at pilot scale was achieved using machinery for feeders and thickness control, for a curing process, and for a drying process respectively.

By synchronizing these three types of equipment, it become possible to achieve automatic and continuous manufacturing of alginate-based bioplastic films as shown in Fig 8. While this research has focused on understanding the importance of each point and identifying bottlenecks, no experimentation was performed regarding automatization or the interrelation of parameters. The challenges that emerged during the proof of concept for the pilot-scale manufacturing included achieving a homogeneous thickness, preventing voids in thin films, and ensuring a thorough material drying. Nevertheless, the developed machinery demonstrates the capacity to manufacture biobased thin films with pure seaweed extracts.

the tensile, flexural and impact properties of the seaweed reinforced PLA obtained, as well as images. In the case of impact resistance, the presence of the fiber in the PLA matrix drops the impact resistance of the material.

3.6 Results of the life cycle assessment

Based on the results in Figure 13, there was no substantial difference observed between the impacts on global warming (kg CO₂ equivalent) of the crude extract and dry alginate. The total impacts for the crude extract and dry alginate were 6.2 and 5.8 kg CO₂-eq., respectively to produce a kg of alginate. The hatchery phase contributed the least to the total impacts with 1.7 kg CO₂-eq., while the seaweed cultivation and biorefinery phases contributed more with 2.3 and 2.2 kg CO₂-eq. for the crude extract and 2.3 and 1.9 kg CO₂-eq. for the dry alginate. These results suggest that the dry alginate production process may have a slightly lower impact on global warming compared to the crude extract, but the difference is not large enough to derive strong conclusions when considering the uncertainties.

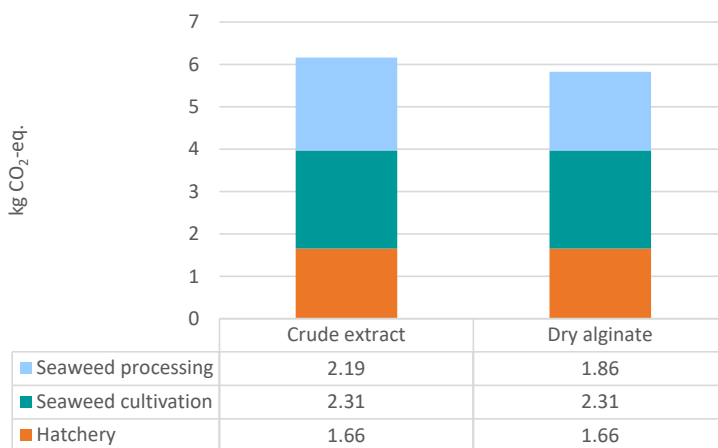


Figure 13: Global warming impacts of the crude extract and dry alginate.

The results show in Figure 14 that the impacts of global warming decrease significantly with the increase in production scale. The 2 million upscaled T/year scenarios have the lowest impact, with a total of 1.37 kg CO₂-eq. per kg of bioplastic (Ayala et al., n.d.). The pilot scale production has the highest impact, with a total of 3.55 kg CO₂-eq. per kg of bioplastic. The film fabrication step contributes the most to the impacts of global warming in all scenarios. This is due to the use of glycerol in the process, the production of which is associated with a large global warming impact. The hatchery and seaweed cultivation steps have a relatively low impact in all scenarios (Ayala et al., 2023b). Overall, these results suggest that the upscaling of seaweed-based bioplastic production could lead to a significant reduction in greenhouse gas emissions compared to pilot-scale production.

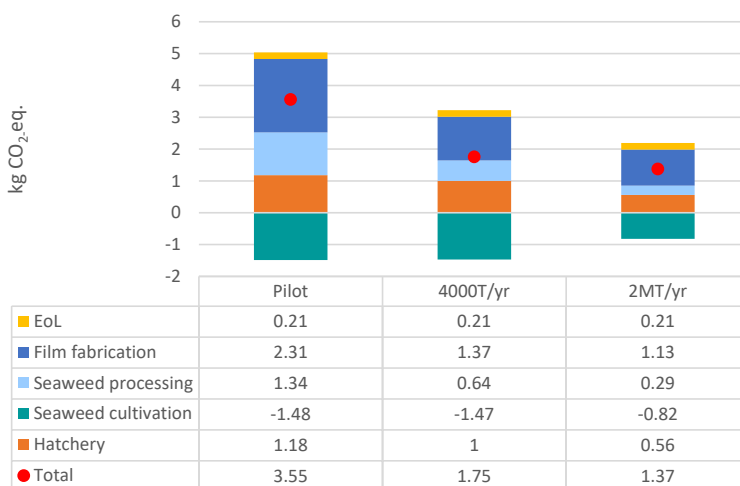


Figure 14: Comparison of global warming impacts per kg of bioplastic in different production scenarios.

The study carried out by (Ayala et al., 2023b) shows a significant difference in the end-of-life (EoL) choices, since composting has a carbon footprint of 0.22 kg CO₂-eq./kg bioplastic and the incineration EoL 1.27 kg CO₂-eq./kg bioplastic.

3.7 A market for bioplastics and biomass utilization

One of the main constraints of seaweed-based bioplastic is the competition with other biomass uses. The primary market for brown seaweed is currently food, followed by feed, pharmaceuticals, cosmetics, fertilizers, and the alginate market. Those high-value application uses to make the current price of biomass relatively high. In contrast, bioplastics are considered one of the lowest-ranked target markets. Therefore, the potential for the current production to shift towards the bioplastic market is limited because using biomass for bioplastics is not as economically profitable in the short term as higher-value applications. According to the conducted interviews conducted by Ayala et al. (2023b), the price of seaweed needs to be low to be able to compete with fossil-based plastics and other bioplastics.

There are various potential scenarios for making brown seaweed biomass economically profitable to produce bioplastics. As the production of brown seaweed scales up and volumes increase, the

production prices would gradually decrease. Another potential solution would be using lower-quality seaweed to produce bioplastic. Lower-quality seaweed is collected in the second harvest or late summer with an increased risk of biofouling. The economic feasibility would improve when targeting seaweed feedstock that is not suitable for human consumption, which is the case of the seaweed holdfast, discarded for the food industry. The concept of sequential extraction or cascade model was also mentioned by some experts as another opportunity for bioplastics. High-value compounds would be used for other purposes and the alginate for bioplastic. Some experts also foresee the importance of working with bioplastics as a consequence of moving away from fossil fuels. There is a possible scenario for prioritizing bioplastics if the prices for conventional plastics increase or with potential government incentives for companies adopting bio-based packaging (Ayala et al., 2023a).

4 Conclusions

This study provides a thorough evaluation of the seaweed-based bioplastic supply chain, outlining the specific procedures involved in each stage of the bioplastic production process. An important aspect of this study is the potential for producing other co-products, such as cellulose for PLA substitution or to produce CCNFs, which revalorizes the biomass as multiple valuable substances and products can be extracted from the seaweed. The production of various co-products from the same seaweed source can significantly increase the economic viability of the biorefinery.

The results of this study also demonstrate the remarkable potential of using seaweed holdfast as a source of high alginate content to produce bioplastics. The use of this waste biomass from the food industry not only avoids competition with other uses of biomass but also contributes to the valorization of underutilized seaweed resources.

One of the key findings of this study is that both commercial alginate and alginate extracted in the biorefinery were found to be suitable for producing smooth and homogeneous films with adequate mechanical properties. This is a significant advantage because the alginate extracted from the cultivated seaweed in the project performs just as well as the commercial alginate, and the quality of the alginate is high and comparable with commercial alginate.

The LCA results showed that both crude alginate and dry alginate have similar impacts on global warming, with a total impact of 4.7 kg CO₂-eq. However, the impact in prospective scenarios decreased, which shows the potential for reducing the environmental impact of bioplastic production by upscaling the technology and increasing the efficiency of the processes. For comparison, the carbon footprint of LDPE in its entire life cycle is 3.6 kg CO₂-eq./ kg of plastic (Wernet et al., 2016), which is similar to the impacts of the alginate-based bioplastic in the pilot scale, but lower than the upscaled scenarios. It needs to be considered that global warming is only one impact category, and this seaweed-based bioplastic offers significant advantages compared to fossil-based plastics, such as its biodegradability, not using fossil-based resources, and all the benefits associated with seaweed cultivation, to mention some.

Future research in this field should focus on various aspects. The seasonality of seaweed is a constraint that needs to be further addressed in the future. For future large-scale production of packaging materials from seaweed, less energy-demanding stabilization methods will be more favorable than freezing, acid preservation is a promising candidate, which is currently being developed and tested in other ongoing research projects.

Secondly, upscaling the technology to an industrial level is necessary to realize the full potential of seaweed-based bioplastic production. This would require mechanization of the seaweed cultivation and harvesting, further optimization of the biorefinery process, and large production facilities to produce the bioplastic. Although promising results have been obtained in this study, advancements are necessary to increase the TRL and bring this technology to an industrial scale. Therefore, more research is needed to optimize the cultivation and harvesting process and to increase the yield of the biorefinery process.

Finally, a techno-economic assessment should be conducted to evaluate the economic feasibility of the technology. This assessment would provide insight into the economic viability of the production process and help identify potential bottlenecks that need to be addressed to make the technology more competitive. Factors such as the cost of seaweed cultivation and harvesting, the cost of the biorefinery process, and the price of the final product need to be considered in this analysis. Moreover, the potential environmental and social benefits of the technology should also be evaluated.

Supplementary Material

Supplementary Material: Details on the methods and extended results.

Acknowledgement

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SUMMARY

The PhD thesis focuses on assessing the environmental impacts of bioplastic production from brown seaweed within the context of the emerging field of Blue Bioeconomy as a consequence of an increasing interest in biomaterials. The research is conducted in parallel with the PlastiSea project, which aims to develop bioplastics using alginate extracted from brown seaweed. The main objective of the PhD is to evaluate the environmental consequences of producing bioplastics from brown seaweed through a consequential Life Cycle Assessment (LCA) perspective.

The thesis is a compilation of scientific articles and the research is structured into four parts, each addressing a specific aspect of the study and corresponding to an article. The first part involves identifying marginal suppliers of brown seaweed, analysing the supply chain and determining key suppliers in the market. The second part focuses on evaluating the environmental impacts of the pilot-scale seaweed-based bioplastic production system and the potential of re-circulating seaweed co-products and different end-of-life pathways, while the third part examines the impacts of an industrial-scale system. The fourth part takes a broader perspective by considering seaweed-based bioplastic production from a supply chain standpoint.

The thesis presents relevant results in different areas. In terms of marginal suppliers, China emerged as the primary supplier and is projected to maintain its significance. Additionally, suppliers from northern Europe and America are anticipated to witness a moderate increase in their market share, but in smaller proportions.

The LCA at the pilot scale identifies the potential for recovering mannitol from seaweed, which could be utilized in the subsequent seaweed-based bioplastic production step. This approach exhibits a slight advantage compared to the base scenario; however, the observed difference is not deemed significant. The assessment of the end-of-life scenarios shows a significant advantage of composting end-of-life compared to incineration.

Moreover, the prospective LCA, which involves upscaling the processes to larger scales, reveals a noteworthy trend of decreasing environmental impacts. As the processes increase in volume, the impacts associated with their operation lower, emphasizing the potential benefits of scaling up production in terms of reducing environmental burdens. The inclusion or exclusion of biogenic carbon shows a substantial impact on the results, showcasing the importance of accounting for this factor in the assessment. The disparities in the outcomes highlighted the significance of considering the biogenic carbon component in LCA analyses.

The findings of the PhD reveal promising potential for brown seaweed-based bioplastics, particularly in terms of greenhouse gas emissions, which show lower impacts compared to conventional fossil-based plastics at larger scales. Moreover, seaweed-based bioplastic exhibits significant advantages in other impact categories, such as marine and freshwater eutrophication. The results collectively also provide valuable insights into the dynamics of marginal suppliers, the potential recovery of seaweed co-products and the assessment of different end-of-life pathways, the influence of biogenic carbon accounting, and the environmental implications of upscaling processes. However, some constraints need to be considered, including competition with other uses of the seaweed biomass and the specific properties and limited applications of the seaweed-based bioplastic, such as its high moisture absorption.

In conclusion, the thesis highlights the challenges and opportunities associated with the production of bioplastics from brown seaweed. It emphasizes the need for further research to address the identified constraints and explore potential solutions. The discussion and conclusion provide a comprehensive overview of the research outcomes, implications, and future directions for the development and utilization of brown seaweed-based bioplastics from an environmental perspective.