Silje Christin Pilskog Olsen

Parametric LCA Modelling of Battery Materials

The case of Graphene

Master's thesis in Energy and Environmental Engineering
Supervisor: Anders Hammer Strømman
Co-supervisor: Nelson Manjong

June 2021

Master's thesis

NTNU
Norwegian University of Science and Technology
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for
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Parametric Life Cycle Modelling of Graphene

Background and objective

Electrification is a promising option for deep decarbonization of key land transport segments through the deployment of battery-electric vehicles. How large climate change mitigation benefits electrification might yield is dependent on the footprint from the manufacturing of the vehicle, battery, and the electricity fuelling the car. Further, this transition to green mobility through electrification of land transport systems has pushed for the development of batteries with high capacity to increase their technical attributes(such as energy density and product lifetimes). Lithium sulphur (Li-S) batteries have a theoretical capacity estimated to be five times the capacities of conventional batteries currently being used. However, Li-S batteries suffer from instability leading to low cycle life. Experiments on graphene show it has a critical role in improving the stability of Li-S batteries and increasing the battery cycle life. However, the environmental footprints of graphene reported in the literature have witnessed significant variabilities and therefore needs further investigation.

Aim and Scope

This thesis aims to develop a parametric model that provides flexibility for testing various combinations of process parameters for graphene to harness the spread in the footprints. Parameters of interest include yield, graphene oxide conversion technologies, the intensity of the electricity mix, etc. In addition, parametric modelling provides a novel approach to understanding variability in LCA and gives a new technique in presenting several LCA simulations for a given functional unit. This study will contribute to the broader analysis of understanding the footprints of value chains and performing high-resolution life cycle assessments of new battery chemistries. This thesis gives the student a solid bridge between environmental modelling and process calculations.

The following tasks are to be considered:

1. Literature Review and Parameterisation

This section reviews graphene value chain and identifies key process levers that could potentially affect the material's environmental footprint. Using engineering and regression models from different literature resources, the thesis creates equations linking process levers to life cycle inventory.

2. Compilation of Life Cycle Inventories (LCIs)

Using the parametric model described in section 1, the student compiles and creates a parametric inventory model with the flexibility to test value chain levers.

3. Application of Life Cycle Impact Assessment (LCA) methods.

In this section, the parametric model created in section 2 should assess the environmental impacts using the in-house modelling software ARDA. Levers tested should be within defined engineering ranges.

4. Analysis of the results.

The analysis of the results should compare the footprints (specifically greenhouse emissions) as a function of the lever combinations with details for each process in the value chain. The results of the thesis should capture how changes in the parameters produce changes in the overall footprints.

5. Documentation

The findings of this research are expected to be documented according to the MSc thesis standards of EPT.

The work shall be edited as a scientific report, including a table of contents, a summary in Norwegian, conclusion, an index of literature etc. When writing the report, the candidate must emphasise a clearly arranged and well-written text. To facilitate the reading of the report, it is important that references for corresponding text, tables and figures are clearly stated both places. By the evaluation of the work the following will be greatly emphasised: The results should be thoroughly treated, presented in clearly arranged tables and/or graphics and discussed in detail. The candidate is responsible for keeping contact with the subject teacher and teaching supervisors.
Risk assessment of the candidate's work shall be carried out according to the department's procedures. The risk assessment must be documented and included as part of the final report. Events related to the candidate's work adversely affecting the health, safety, or security, must be documented and included as part of the final report. If the documentation on risk assessment represents a large number of pages, the full version is to be submitted electronically to the supervisor and an excerpt is included in the report.

According to "Utfyllende regler til studieforskriften for teknologistudiet/sivilingeniørstudiet ved NTNU" § 20, the Department of Energy and Process Engineering reserves all rights to use the results and data for lectures, research, and future publications.

Submission deadline: **11th June 2021**

☐ Work to be done in lab (Waterpower lab, Fluids engineering lab, Thermal engineering lab)
☐ Field work

Department for Energy and Process Engineering,

____
Supervisor: Prof. Anders Hammer Strømman
Co-Supervisor: Nelson Manjong (PhD Candidate)

Abstract

Substantial investments are being made in new battery technologies with focus on renewable energy in the industry. Currently, Lithium-Sulfur (Li-S) batteries are considered the most promising successor for Lithium-ion (Li-ion) as it represent several advantages. However, Li-S batteries are facing challenges where graphene hold promises in eliminating the barriers for commercialization. Graphene is a highly conductive, flexible and mechanically robust material. This thesis considers the production route; synthetic and natural graphite production, Hummers' process for graphite oxidation and chemical reduction for the last stage in production.

This study investigates the environmental impacts from producing 1 kg graphene by utilizing a parametric life cycle assessment (LCA) model. This model enables an assessment of the interplay between the key parameters in the graphene value chain and detect the variation of impacts by altering them dynamically. This detailed assessment highlights relevant processes and increases the precision of the assessment. A total of four variables (graphite production route, modifications of the Hummers' process, electricity intensity and graphene yield) were parameterized, resulting in 108 different scenarios for graphene production.

The results provided by this study show that the two processes: graphite oxidation and chemical reduction have the largest contribution to GWP. This indicates that the parameters electricity intensity, as both processes are energy-intensive, and graphene yield in the chemical reduction process are significant. In addition, evaluating the chemical composition of the graphite oxidation process is also crucial in order to minimize the environmental impact from graphene production.

Sammendrag

I dag blir det gjort betydelige investeringer i nye batteriteknologier med fokus på fornybar energi. Foreløpig er Li-S batterier ansett som den mest lovende etterfølgeren for Li-ion, da disse representerer flere fordeler. Imidlertid står Li-S overfor utfordringer hvor grafen holder løfter om å eliminere hindringene på veien for kommersialisering. Grafen er et svært ledende, fleksibelt og mekanisk robust materiale. Denne oppgaven tar for seg produksjonsveien: syntetisk og naturlig grafittproduksjon, Hummers' prosess for grafittoksidasjon og kjemisk reduksjon for siste trinn i produksjonen.

Denne studien undersøker miljøpåvirkningen ved å produsere 1 kg grafen ved å bruke en parametrisk livssyklusvurdering (LCA)-modell. Dette muliggjør en analyse av samspillet mellom nøkkelparametrene i grafenverdikjeden og også avdekke variasjonen av miljøpåvirkninger ved å endre dem dynamisk. Denne modellen gir en detaljforståelse av relevante prosesser og øker presisjonen av analysen. Totalt er fire variabler (grafittproduksjon, modifikasjoner av Hummers-prosessen, strømintensitet og grafenutbytte) parametrisert, noe som resulterte i 108 forskjellige scenarioer for grafenproduksjon.

Resultatene gitt i denne studien viser at de to prosessene; grafittoksidasjon og kjemisk reduksjon bidrar mest til GWP. Dette impliserer at parametrene som gjelder for disse to prosessene er betydelige, dvs. strømintensitet, ettersom begge prosessene er energiintensive, og grafenutbytte i den kjemiske reduksjonsprosessen. I tillegg er evaluering av den kjemiske sammensetningen av oksidasjonsprosessen for grafitt også avgjørende for å minimere miljøpåvirkningen fra grafenproduksjon.

Acknowledgements

This thesis was written in the course of spring 2021 at the Norwegian University of Science and Technology (NTNU) in Trondheim, Norway. This thesis serves to fulfil my master's degree in Energy and Environmental engineering at the Department of Energy and Process Engineering (EPT).

First of all I would like to thank my supervisor, Professor Anders Hammer Strømman and my co-supervisor, Nelson Manjong for invaluable help and guidance throughout my master thesis. Their help have been much appreciated.

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Silje Christin Pilskog Olsen

June 2021, Trondheim Norway

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Abbrevations

EPD Environmental product declarations

FU Functional unit

GO Graphite oxide

GWP Global warming potential

IPCC Intergovernmental Panel on Climate Change

LCA Life cycle Assessment

LCI Life Cycle Inventory

M1 Modification 1 of the Hummers' process

M2 Modification 2 of the Hummers' process

M3 Modification 3 of the Hummers' process

rGO Reduced graphite oxide

1 Introduction

1.1 Background and motivation

The purpose of the Paris agreement is to prevent an overall global temperature rise of 2 degrees Celsius above pre-industrial levels. The agreement also aims to toughen the global response and execute measures to limit the temperature rise even further to 1.5 degrees Celsius (United Nations Climate Change, 2020). In this context, decarbonization of the transport sector is crucial, as it is responsible for approximately 23% of total energy-related CO_2 emissions (6.7 $GtCO_2$) (IPCC AR5 WG III, 2014). Here, electrification of transport segments hold great promise of approaching the goal set in the Paris agreement. However, the level of climate change mitigation benefits from implementing batteries into the transport section, depends on the footprint from each life cycle stage, i.e. production, use-phase and end-of-life treatment. This includes the electricity fuelling the vehicle.

Today, substantial investments are being made in new battery technologies with focus on renewable energy in the industry. There is an ongoing global race for knowledge within the field that covers the whole battery life cycle and its value chain (Sintef, 2021; European commission, 2020). China has for a significant period of time been dominating this industry, but now the European Union is taking action to secure infrastructure and production of vital components in Europe. This enables a more sustainable industry and creates a greater market for battery production.

The market evolution of electric vehicles (EVs) depends primarily on the capacity of the battery. Today, EVs traditionally operate with Lithium-ion (Li-ion) batteries. However, the theoretical limit of capacity is approaching (Benveniste, Rallo, Canals Casals, Merino, & Amante, 2018). In order to keep enhancing the capacity, active research on Li-ion is undertaken, in addition to more attention to new battery systems with higher density. Currently, Lithium-Sulfur (Li-S) batteries are considered the most promising successors for Li-ion (Azimi, Xue, Zhang, & Zhang, 2015).

Li-S represents several advantages from an economic and environmental point of view, in addition to abundance of sulfur and prominent theoretical capacity. Still, Li-S is held back by its short lifespan and quick capacity decay due to the insulating property of sulfur and the significant solubility of lithium polysulfides. Here, Graphene holds promises in eliminating the barriers on the way of commercialization of Li–S batteries. Graphene is a highly conductive, flexible and mechanically robust material. This thesis considers the production route; natural and synthetic graphite production, Hummers' process for graphite oxidation (GO) and chemical reduction for the last stage in production. The constructed porous structures of reduced GO, i.e. graphene, along with high conductivity lay the foundation for excellent electron and ion transferability, empowering higher usage of active material (Zhang, Gao, Song, He, & Li, 2018; Yu, Li, Wu, & Shi, 2015). In addition, this material with its range of applications have great prospective for future development.

In order to enhance the understanding of the environmental impacts from implementing batteries and improve the environmental performance, key materials in the battery value chain, such as graphene, must be assessed. This can contribute to a more sustainable production. Life cycle assessment (LCA) is a powerful tool to assess all stages in the graphene life cycle and is a well suited method to uncover environmental trade-offs. This thesis applies a parametric LCA model in order to achieve a more detailed assessment, utilizing a dynamic inventory.

1.2 State of the art

A number of publications on environmental assessments of lithium-ion batteries have been made, but there are limited studies assessing the environmental impacts of key materials for the lithium sulfur battery value chain, including graphene. This study considers cradle-to-gate LCAs from various literature sources. Each assessment evaluate graphene production with the same system boundaries and functional unit, as described in section 1.3. There are multiple production routes for graphene, this study reviews both natural and synthetic graphite production, the Hummers' process for graphite oxidation and chemical reduction for the final stage in production.

Cossutta, McKechnie, and Pickering conducted an LCA of three graphene production routes: chemical vapour deposition (CVD), electrochemical exfoliation of graphite rods and graphite chemical oxidation and subsequent chemical or thermal reduction. This study presents five different modifications for the graphite oxidation process, i.e. Hummers' process, where it is shown that the chemistry composition of this process has major impact on the final contribution to GWP. This study also presents that the least impacting production route remains Hummers' process followed by thermal reduction (90 kg CO_2 eq./kg graphene). However, the impact from combining the Hummers' process with chemical reduction, as in this thesis, was estimated at 150 kg CO_2 eq./kg graphene (Cossutta et al., 2017).

Serrano-Luján et al. investigated the environmental implications from graphene production. This study compared two modifications of the graphite oxidation process along with chemical reduction. The final global warming potential from producing 1 kg graphene, was measured to be 586 kg CO_2 eq./kg graphene (Serrano-Luján et al., 2019). In addition, a study by Khanam, Popelka, Alejji, and AlMa'adeed presented the GWP from producing graphene, with the same production routes, at approximately 90 kg CO_2 eq./kg graphene.

A study by Arvidsson, Kushnir, Sandén, and Molander performed a prospective LCA of graphene production, by investigating the production routes: chemical reduction and ultrasonication. Although this study did not estimate the GWP, they focused on potential differences in energy use, human toxicity, ecotoxicity, and blue water footprint. In addition, the study provided an inventory that were utilized to compare and evaluate with the remaining studies mentioned above. Here, the data utilized where based on patents and scientific papers. The results show that the chemical reduction route has almost twice the energy requirement compared to the other alternative. This is mainly due to the chemical reduction process (roughly 75%). However, this production route has a lower contribution to both human and ecotoxicity (Arvidsson et al., 2014).

A common feature of these studies is that each assessment applied conventional LCA to estimate the environmental impacts from graphene. This thesis provides a more detailed assessment as it includes a dynamic inventory that to a greater extent shows the variations of impacts (scenarios) that can occur by altering key parameters. This is beneficial as several preconditions are important in determining the final result, such as process efficiency, electricity mix, etc.

1.3 Objective of this study

The main objective of this thesis is to assess the environmental impacts from producing graphene, as it is a key material in the value chain of Li-S batteries. As mentioned, there are multiple production routes. This study investigates both natural and synthetic graphite, the Hummers' process for graphite oxidation and chemical reduction for the last stage of graphene production, namely a cradle-to-gate perspective as described in Figure 1. Due to unique features, these methods have been extensively utilized in Li–S batteries (Zhang et al., 2018).

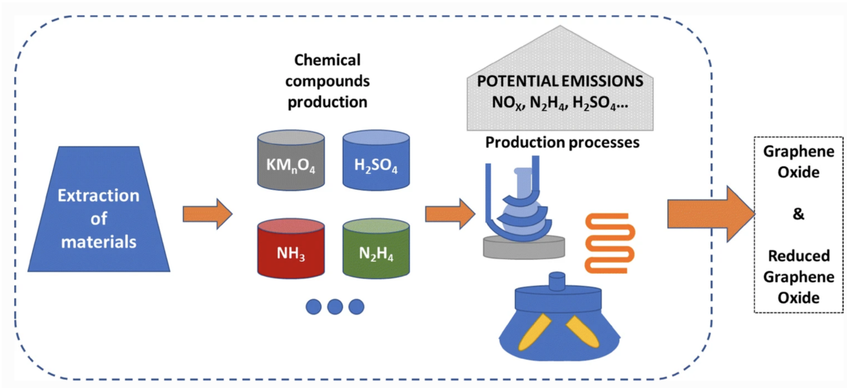

Figure 1: Cradle-to-gate system boundary of the graphene production route: Hummers' process and chemical reduction (Serrano-Luján et al., 2019).

In order to achieve a more detailed and precise understanding of the environmental impacts from graphene, a parametric LCA model is deployed. On the basis of the substantial influence on the environmental performance, graphite production route (synthetic and natural), three different modifications of the Hummers' process, graphene yield (%) and electricity intensity (kg CO_2 eq./kWh) were chosen as parameters in the model. By expanding from conventional LCA to parametric modelling, it enables an analysis of the interplay between the key parameters in the graphene value chain and a detection of the variation of impacts by altering them dynamically.

The main focus in this study is to assess the GWP from each process of graphene production. For evaluating the influence of the parameters mentioned above, they are assessed in reference to a base case scenario that are discussed in section 3.4. Further, an presentation of the overall results that includes specifying preconditions, i.e. parameter settings, for the best and worst case scenario. In addition, a presentation of the contribution analysis from each foreground process.

The system boundary in this study is illustrated by the flow diagram presented in Figure 4, along with an illustration of which processes that are affected by the various parameters. The flow diagram is divided by a stippled line to clarify background and foreground processes. Background processes are generic database processes, while foreground processes are compiled specifically for this study. This figure, along with the various processes and parameters will be further discussed in section 3. The functional unit is one kilogram (kg) graphene produced and is selected to enable a comparison of the results with previous and future studies within the topic.

1.4 Structure of work

The thesis is divided in six sections in addition to the appendices that includes all data code utilized in both MATLAB and Python as well as inventories and background data for the parametric LCA model. Section 2 covers the methodology that has been applied in this thesis. Here, an introduction to conventional LCA together with the parametric LCA model. This section also includes the tools used and implementation of method. Section 3 presents the case description, where each process and parameter are discussed. It also includes a description of the base case scenario and inventory analysis. This will be followed by section 4 where all results generated by the model are assessed. The results are presented per functional unit. Finally, section 5 and 6 presents the discussion and conclusions of the thesis.

2 Methodology

In this section a description of the methods utilized in this study will be presented. This involves a brief explanation of the life cycle assessment (LCA) framework as a method for environmental impact assessment, along with mathematical aspects. Further, a description of the parametric LCA model. Lastly, an overview of the tools used and implementation of method is given. The methodological choices and inventory analysis are described in the following section.

2.1 Life Cycle Assessment

Life cycle assessment (LCA) is a well-established method to assess the environmental impacts of a product, process or service. The objective of the assessment is to conduct consistent comparisons of various technological systems in reference to environmental impacts. In order to achieve this, the whole life cycle must be considered, i.e everything from raw material extraction to end of life treatment. LCA is also an essential tool of revealing matters of "problem shifting". This means to uncover when one problem is shifted to another phase of the production value chain, outside of the the system boundary.

In order to carry out an LCA analysis and estimate potential environmental impacts, one must first gather sufficient inventory of material and energy requirements for the whole value chain. Then it is possible to detect the overall footprint of the product, process or service evaluated and uncover which phase is most critical. By doing so, it enables an overview of possible improvements that can lower the environmental impacts (Strømman, 2010).

LCA is the foundation of environmental product declarations (EPDs) and included in multiple international (ISO) standards. ISO 14040:2006 document the LCA framework, principles and also include various limitations to this method. Today, companies, institutions and organisations implement LCA to document and support their environmental performance (ISO 14040:2006, 2016).

The LCA framework is generally divided into four methodical phases. Figure 2 illustrates the phases included in the ISO standard 14040. Namely, goal and scope definition, inventory analysis, impact assessment and interpretation. Note that the last stage, interpretation, is closely linked to each phase. In the following subsections, each phase will be further explored.

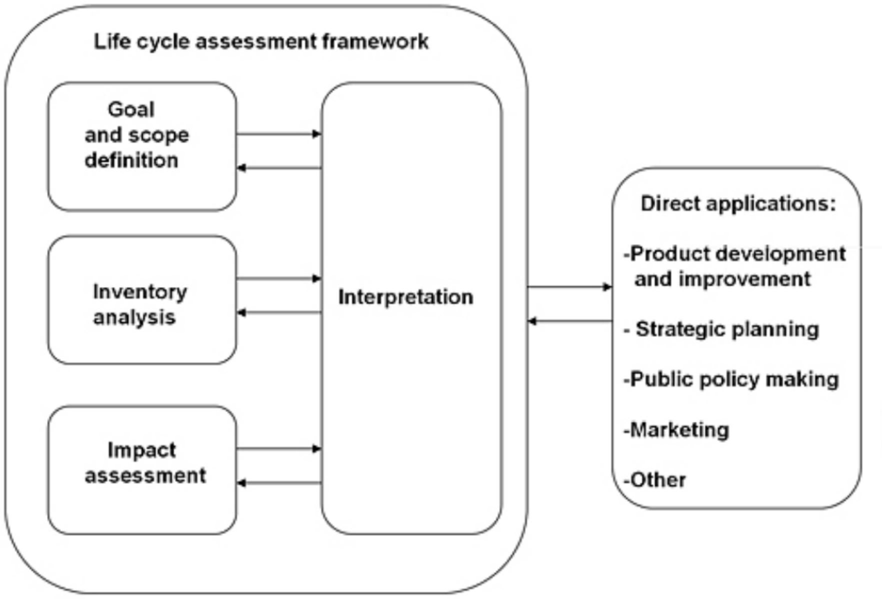

Figure 2: The LCA framework (ISO 14040:2006, 2016).

2.1.1 Goal and scope definition

An LCA study starts with defining an explicit statement of the goal and scope of the objective to be investigated. The goal is key for the analysis, as it presents the context and motivation for conducting the analysis, as well as the intended audience and intended application for the study.

The scope should present which impact categories that will be considered and addressed, in addition to the functional unit (FU) and system boundaries. The functional unit should express the "function" of the object in question and is the reference for comparison of and analysis between different alternatives. The system boundaries can be in relation to time, technical systems and geographical borders. The definition of system boundaries will substantiate whether the study follows a cradle-to-gate or a cradle-to-grave approach. As the assumptions made in this phase will affect the final results of the study, it is vital that they are reviewed in the interpretation phase (ISO 14040:2006, 2016).

2.1.2 Inventory analysis

In order to organize the various flows within the system boundary, flowcharts or flow diagrams are often implemented. Here, all production steps or processes are included, in addition to the flows between them. Typically, processes or products are represented by boxes and the various flows are represented by arrows. In LCA modelling, the illustrated system or flowcharts are usually divided in background and foreground systems. The background system incorporate processes that are established on generic databases and becomes inputs to the foreground system. While the foreground system incorporate processes that are modeled with data specific to the study (Strømman, 2010). The flow diagram in Figure 4 presents the system evaluated in this thesis, and illustrates which part of the system is background and foreground.

Further, matrix algebra or LCA software are used to systematize the quantitative data. Generally, a sufficient LCA often requires large amounts of data, which can be found from several sources. Typical sources are usually measurements, engineering process data from manufacturers, LCA databases, other LCA studies, or other literature and expert judgments. A further explanation of the mathematical framework of LCA methodology is found in section 2.1.4.

2.1.3 Impact assessment and Interpretation

The third and fourth phase of the LCA framework is the impact assessment and interpretation. In the impact assessment, all emissions or "stressors" in general LCA terminology, are transformed to impact unit is that are understandable and comparable. As mentioned, this thesis will focus on the impact category global warming potential (GWP) that presents climate change impacts in terms of CO_2 equivalents. In this study the ReCiPe method is utilized in order to quantify the environmental impacts from various stressors. This method will be further discussed in section 2.3. This phase can be divided into four steps, namely classification, characterization, normalization, and weighting. However, normalization and weighting are evaluated as non-mandatory (ISO 14040:2006, 2016).

The last step includes an interpretation of all previously reviewed phases; goal and scope definition, inventory analysis and impact assessment. In order to achieve an adequate interpretation, it is important to understand the analysis and identify critical assumptions, parameters, as well as important contributors from stressors (pollutant or resource) and various activities. Different uncertainties and assumptions must also be recognized and evaluated. The aim of this phase is to draw a conclusion based on the findings and identify doable improvements to decrease environmental impacts.

In accordance with ISO 14040:2006 (2016), the interpretation should also include a contribution and sensitivity analysis in order to substantiate the LCA conclusions. The contribution analysis evaluates the contribution of each process step of the system, while the sensitivity analysis aims to assess the robustness of the final results.

2.1.4 Mathematical framework of LCA

This section presents the mathematical framework that lays the foundation for life cycle assessments. The technicalities that differentiate the parametric LCA model from conventional LCA is presented in section 2.2.1.

The framework of LCA is based on vector and matrix calculations that enables input data to transform into environmental impacts. Each of the following technicalities (matrices, equations, etc.) are assembled from Strømman (2010).

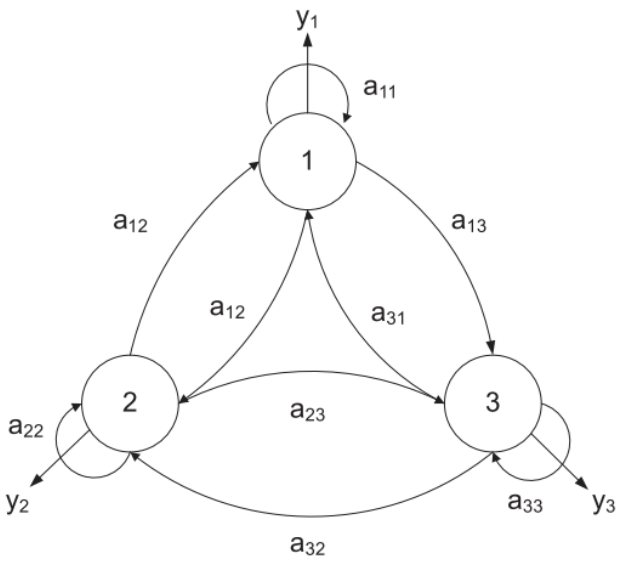

Figure 3: Production nodes and inter-connectivity (Strømman, 2010).

Figure 3 provides a schematic presentation of a three production nodes system, as in this thesis. Here, the coefficients a_{ij} represents requirements denoted by process i per unit output of process j:

$$aij = \frac{\text{amount of i required}}{\text{output of j}}, \tag{1}$$

The coefficient y_j represents external demand of products. In this thesis all requirements are normalized so they are represented per kilo graphene produced. Once the requirements for all production nodes are identified, the A-matrix, i.e. the requirements matrix, in equation 2 can be established.

$$A = \begin{bmatrix} A_{ff} & 0 \\ A_{bf} & A_{bb} \end{bmatrix} \tag{2}$$

The A_{ff} represents the requirements between the foreground processes, that are data compiled specifically for this study. The A_{bf} represents upstream inputs of background processes to the foreground system. Lastly, the A_{bb} represents the background processes in the requirement matrix.

This matrix is utilized to identify the activity developed in all production nodes as the result of the demand for the functional unit. The matrix also organize the production balance shown in equation 3:

$$x = Ax + y \tag{3}$$

The x vector represents the production output in each node and is a function of intermediate demand, Ax, and external demand, y. This vector is usually referred to as the output vector. Note that x can also be expressed as a function of the Leontief inverse, L and external demand, y:

$$x = Ax + y \Leftrightarrow (I - A) = y \Leftrightarrow x = (I - A)^{-1}y \tag{4}$$

Where

$$L = (I - A)^{-1} \Rightarrow x = Ly \tag{5}$$

The coefficients, l_{ij}, in the L-matrix represent the output of process i that is required per unit of final delivery of process j. For example, l_{12} represents the output of process 1 needed per unit external demand of product from process 2.

In order to calculate the environmental loads and total emissions generated from the external demand in question, the stressor intensity matrix S needs to established. This matrix contains, for a particular process, the vector of stressors (*stressors* are used as a more general terminology compared to *emissions*) per unit of output. The stressor matrix is further utilized to calculate the total amount of stressors for a given external demand, e in equation 6. Note that stressor data ought to be assembled analogously to the coefficients in the A-matrix.

$$e = Sx \tag{6}$$

Further the characterization matrix, C, must be established in order to calculate the total impacts, namely the vector d (shown in equation 7). The C matrix includes characterization factors that enable a conversion of emissions with the same environmental impact into equivalents, e.g. N_2O into CO_2-equivalents.

$$d = Ce = CSx \tag{7}$$

It is vital to understand how the different processes in the system contribute to the various impact categories. To enable this understanding, the D_{pro} matrix must be established. The calculation behind this matrix is shown in equation 8 to 10.

$$D_{pro} = CS\hat{x} \tag{8}$$

$$D_{str} = C\hat{e} = C\hat{S}x \tag{9}$$

$$d = \sum_{pro} D_{pro} = \sum_{str} D_{str} \tag{10}$$

it is also of interest to calculate which stressors contribute to the different impact categories, namely the D_{str} matrix in equation 9. Equation 10 is the sum of the columns in D_{str} and the rows in D_{pro}, thereby represents the vector of total impacts.

There are multiple frameworks that can be utilized in order to evaluate various environmental impacts. This study applied the ReCiPe framework.

2.2 Parametric LCA model description

Limitations associated with LCA modelling is acknowledged by both international standards of LCA and scientific literature. The key standards, ISO 14040:2006 and ISO 14044:2006, are quite transparent with gaps and challenges related to the method. Thus, each study must document their limitations (ISO 14040:2006, 2016; Finkbeiner et al., 2014).

Some of the LCA challenges is associated with modelling value chains. Conventional LCA struggle to capture variations in key elements, as it applies static values for the impact calculations. Thereby, increases the uncertainty of the results. In addition, there are limitations associated with several aspects in the LCI phase; due to differences or uncertainties in aggregation and allocation procedures, setting the system boundaries that do not include all inputs and outputs of every unit process, etc. A summary of limitations and uncertainties can be found in Finkbeiner et al. (2014).

The main objective of the parametric LCA model that is utilized in this study, is to address some of the limitations of conventional LCA. In particular, this study aims to provide an assessment that considers a certain variation of the key parameters, thereby providing a dynamic inventory. This enables an analysis of the interplay between the key parameters in the value chain and detects the variation of impacts by altering them dynamically. This model gives a detail understanding of the relevant processes and increases the precision of the analysis.

This study investigates the environmental impacts from producing 1 kg graphene, which is part of the Li-S battery value chain. Four key parameters in graphene production were considered which resulted in 108 different "scenarios" for graphene production.

Figure 4 illustrates which parameter affects the various process steps in graphene production. As shown in the figure, altering the intensity of electricity will affect the Hummers' process and chemical reduction, which are both energy intensive processes. Therefore, it is considered to be a crucial parameter for this production. For graphite production, an global average electricity mix is applied as the environmental impact was proven to be less significant. The figure also indicates that the modifications of the Hummers' process affect the graphite oxidation and altering the graphene yield affects the chemical reduction. Note that due to data availability, only graphite production includes direct emissions. A thorough assessment of adjusting each parameter and different scenarios for graphene production is further discussed in section 4.

2.2.1 Mathematical framework of the parametric LCA model

The primary technical differences between traditional LCA and the parametric LCA model occur in the A-matrix. Here, the goal is to achieve several variants of the A-matrix. To enable this, one must identify key elements in the value chain and then consider the area in which these may vary. This will then constitute the parameters in the model. For this thesis the parameters are as mentioned; graphite production route (synthetic and natural), three different modifications of the Hummers' process, graphene yield (%) and electricity intensity (kg CO_2 eq./kWh).

All parameters are upstream inputs from processes of the background system to the foreground system, they are therefor included in the A_{bf} section of the requirement matrix, i.e. A-matrix. The graphene yield will also change the value of the foreground process chemical reduction in A_{ff} matrix and graphite production route will determine the foreground values that apply to the processes; natural graphite production (mining and processing) and synthetic production. For example, if the model is simulating a scenario with natural graphite, the value for synthetic graphite production would automatically be zero and vice versa.

By utilizing the tools, discussed in section 2.3, these parameters dynamically change for each simulation, creating multiple inventories, which are referred to as *scenarios* in this thesis.

As this thesis applied four parameters, where;

- parameter 1 contains a range of 2 values
- parameter 2 contains a range of 3 values
- parameter 3 contains a range of 6 values
- parameter 4 contains a range of 3 values

It creates $2 \times 3 \times 6 \times 3 = 108$ scenarios for graphene production.

2.3 Tools used and implementation of method

The software Arda developed by the Industrial Ecology Department at the Norwegian University of Science and Technology (NTNU), lays the foundation for life cycle assessments, including the parametric LCA model. This software is used to perform the LCA calculations by utilizing the ReCiPe impact assessment methodology and Ecoinvent v3.2 database. The Ecoinvent database is based on industrial data that have been compiled by internationally recognized LCA consultants and research institutes. It presents transparent and consistent life cycle inventory data. it is recognized as the most finalized database for LCA purposes and for superior quality (Ecoinvent, 2013).

The ReCiPe method includes eighteen "midpoint" level categories and three "endpoint" level categories. At midpoint level, the life cycle inventory results are transformed into category indicators. At endpoint level, the impacts of the LCI results are divided or quantified in three endpoint indicators. This framework is also associated with three different cultural perspectives; individualist, hierarchist and egalitarian. What set them apart is the expectations of future technology development and time frame. (Goedkoop et al., 2008). This study will focus on evaluating midpoint indicators, more precise the global warming potential (GWP), as it is associated with lower uncertainty and apply the default hierarchist perspective.

In order to achieve a parametric LCA model that vary parameters dynamically, some alterations have been made to the Arda template coupled with MATLAB vR2019a, a product by MathWorks. The model development was performed by the co-supervisor Nelson Manjong, however some alterations have been made to adapt the model for graphene production (Manjong, Strømman, Burheim, & Usai, n.d.).

Firstly, a input file containing all possible scenarios were programmed and assembled in Python, see Appendix A.2.2. By importing this file to MATLAB as described in in Appendix A.3, it enabled a dynamical alteration of the the inventory, while running/simulating each time a parameter changed and thereby creating different scenarios. For all model runs, each "scenario" were saved in an excel format. As for graphene, this resulted in 108 excel sheets for further processing and layout. Note that the modifications M1, M2 and M3 are labeled GO2, GO1 and GO3 respectively in the input file. This change was made due to practical considerations.

It was challenging to process and visualize such comprehensive data where all parameters changed dynamically. For this reason, it was considered best to utilize the high-level programming language Python 3.8. This software contains packages for data processing, analysis and visualisations. In addition, a base case scenario was constructed to assess the variation of one parameter, while others are kept constant. Note that inventory, MATLAB and Python code are available in Appendix A.1 to A.3.

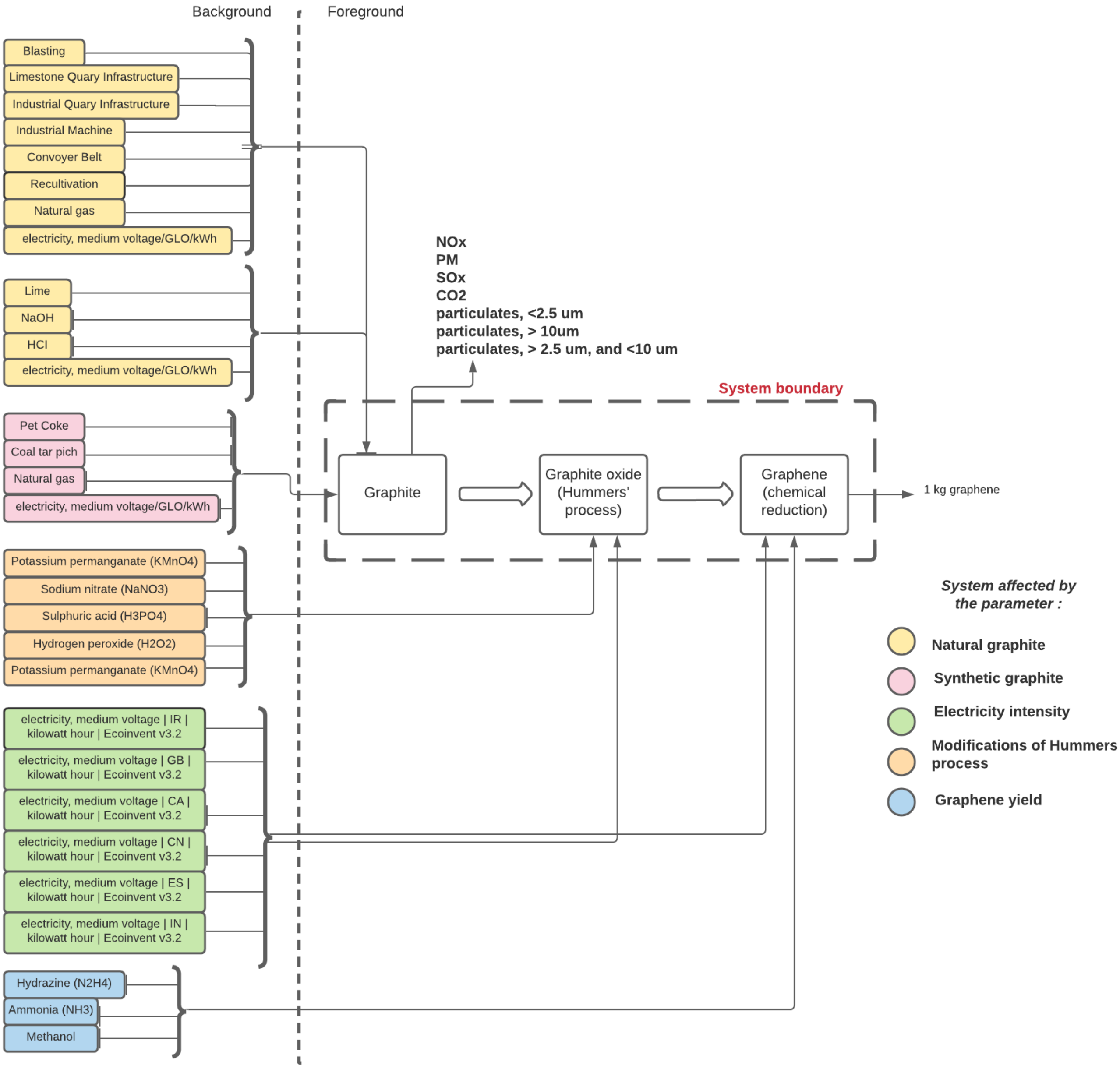

Figure 4: Flow diagram of the studied system that illustrates which processes that are affected by the various parameters.

3 System description

Graphene is a two-dimensional, single layer of bonded carbon atoms arranged in a hexagonal pattern shown in Figure 5 (Chen, Yan, & Bangal, 2010). This material has attracted much attention due to its properties and potential derivatives. At present, there are five methodical choices for graphene production; mechanical exfoliation of graphite (Paton et al., 2014), chemical vapor deposition (CVD) (Y. Zhang, Zhang, & Zhou, 2013), solvothermal synthesis (Parvez, 2019), epitaxial growth on electrically insulating surface (Lu, Yu, Huang, & Ruoff, 1999), and reduction of graphite oxide (GO) (Serrano-Luján et al., 2019; Cossutta et al., 2017; Arvidsson et al., 2014).

This material is a highly conductive, flexible and shows exceptional mechanical properties. For Li-S batteries, graphene and its derivatives hold promises in eliminating the barriers on the way of commercialization. It is capable of enhancing the electrochemical performance of the battery. Here, graphene has been widely used for different purposes. In addition to cathodes, this material has been exployed for other purposes such as interlayers or separators to hinder the movement of dissolved polysulfides, by reducing the shuttle effect and better the usage of the active material. Recently, it was discovered that utilizing graphene in Lithium anodes prevents spontaneous degradation and Lithium dendrites growth. Also, a graphene constructed current collector empower outstanding rate ability and lifespan due to the ability to entrap polysulfides and better the battery conductivity (Zhang et al., 2018).

Further development of the Li-S battery is currently at a crossroads due to low energy density, which is affected by low sulfur utilization, low sulfur loading and cost issues from Li anode. These are factors that hinder commercialization and scale-up production. Graphene and its derivatives are expected to play a key role in game-changing Li–S batteries.

This section will firstly present an overview of each process in graphene production along with discussion of the parameter selection for the parametric LCA model. Lastly, a description of the base case scenario in this thesis and a review of the inventory analysis. This study will examine both natural and synthetic graphite production, the Hummers process for graphite oxidation and chemical reduction for the final production step towards graphene. This choice of method was made on the basis of suitable properties for Li-S battery production (Zhang et al., 2018; Karki & Ingole, 2020).

It should be noted that in literature there are often confusion associated with the term ”graphene”. This term is frequently utilized when speaking of graphene derivatives that possess different mechanical and chemical properties as shown in figure 5 (Kumar & Pattammattel, 2017). To clarify, this thesis speak of graphene produced from chemical reduction, i.e. reduced graphene oxide when using the term ”graphene”.

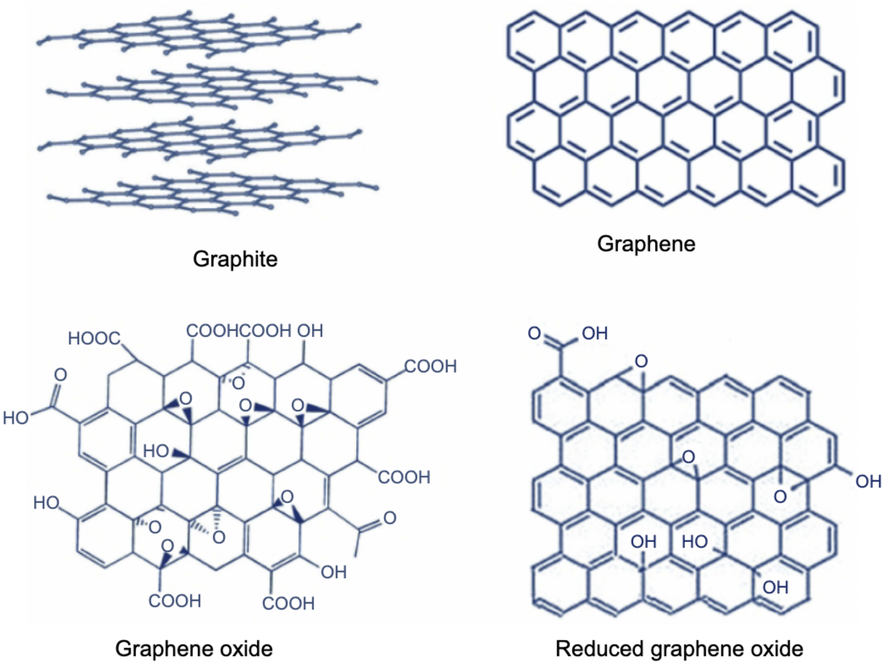

Figure 5: Structure of graphite, graphite oxide, reduced graphene oxide and graphene (Geetha Bai et al., 2019).

3.1 Graphite production

The baseline for graphene production is graphite. Graphite is a material with notable characteristics and properties. It is also significant for several current and future industry applications (Yu et al., 2015). This raw material can either be synthetically produced or mined. Between these two options there are some fundamental differences that also contribute to different environmental impacts occurring during production.

The consumption of energy is one of the main differences that separates these two production routes, This requirement is generally higher for synthetic graphite. Synthetic graphite is primarily manufactured by graphitization of selected carbon precursors, such as petroleum and coal tar-based cokes under an oxygen-free environment and with temperatures higher than 2,500°C (Ambrosi et al., 2012). In order to improve the final quality and material properties of graphite, the baked mixture is often impregnated with pitch and rebaked prior to the graphitization furnace.

The petroleum is usually made from delayed coking of residues from the thermal processing of crude oil. While, coal tar-based cokes, is often manufactured from the coke oven for steel production as a byproduct (Dunn et al., 2015).

As a natural mineral, graphite is found mostly in metamorphic rocks. The further production can be divided in four steps; namely mining, beneficiation, purification and processing (Gao, Gong, Liu, & Zhang, 2018a). In total, the world production of natural graphite is approximately 600 000 tonnes each year were some of the most important production regions are China, India, Mexico and Brazil (Raade, 2020). Thus, these regions have been taken into account for the analysis. Especially with regard to the intensity of electricity. As there are two alternatives for graphite production, both methods are included as parameters in the model utilized for this thesis.

3.2 The Hummers' process

In order to construct graphene by utilizing chemical reduction, the graphite must first undergo an oxidation process creating graphite oxide (GO). The most common oxidative treatment is the Hummers' process, it is therefore applied for this assessment (Ambrosi et al., 2012; Arvidsson et al., 2014). The oxidation occurs when an oxidative agent is added to an acid solution containing graphite powder. This solution is further cooled to avoid vigorous reactions from the oxidative agent and to control the temperature.

This thesis considers a chemical composition of sulfuric acid, sodium nitrate and potassium permanganate as the oxidative agent. Furthermore, hydrogen peroxide is required to lower excess potassium permanganate and some deionized water for washing and dissolution (Arvidsson et al., 2014). However, several studies describe various modifications of the Hummers' process, leading to improved the efficiency from the process (Arvidsson et al., 2014; Ambrosi et al., 2012; Cossutta et al., 2017; Marcano et al., 2010). To clarify the influence by the graphite oxidation (the Hummers' process) on the total environmental impacts from graphene, three modifications obtained from Cossutta et al. (2017) are included as parameters in the model.

In this study the modifications are referred to as M1, M2 and M3. M1 has a relatively short process time (approx. 30 min at 35 °C) compared to M2 and M3 that need 3 hours at roughly 100 °C. Other factors that differentiate the modifications are the input values of sulphuric acid (H_2SO_4), hydrogen peroxide (H_2O_2), deionized water and energy consumption. Table 1 provides a detailed description of the differences between the chemical composition of M1, M2 and M3.

Table 1: Chemical composition of modifications M1, M2 and M3.

Inventory	**Unit**	**M1**	**M2**	**M3**
Potassium permanganate (KMnO4)	kg/kg graphene	2,675	2,675	3,85
Sodium nitrate (NaNO3)	kg/kg graphene	0,45	0,45	0,4875
Sulphuric acid (H2SO4)	kg/kg graphene	37,75	41	310
Hydrogen peroxide (H2O2)	kg/kg graphene	1,55	3,25	8,3625
Input graphite	kg/kg graphene	0,8875	0,8875	0,9625
Electricity	MJ/kg graphene	12,5	37,5	137,5
deionized water	MJ/kg graphene	278,75	290	528,75

3.3 Chemical reduction process

The last step in graphene production is the chemical reduction of graphite oxide. In this assessment, hydrazine (N_2H_4) is utilized as the reducing agent, which is currently the most used agent for its purpose. This process consists of incorporating hydrazine with ammoina (NH_3), deionised water, methanol with graphite oxide. In addition, chemical reduction requires a significant amount of energy. During the reduction the oxygen functionalities are eliminated so a porous structure of single/few-layered graphene sheets, i.e. reduced graphene oxide (rGO) remains (Arvidsson et al., 2014; Cossutta et al., 2017; Pei & Cheng, 2012). Figure 6 illustrates the production route from graphite materials to graphene sheets.

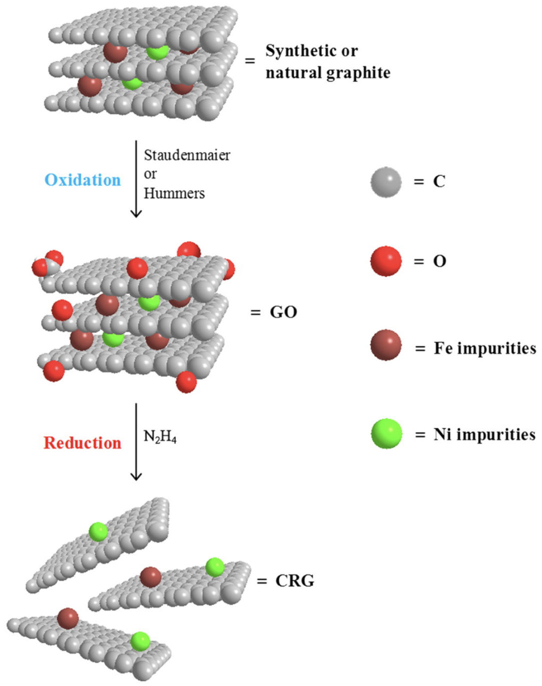

Figure 6: Schematic preparation of chemically reduced graphene (Ambrosi et al., 2012).

The properties of reduced graphite oxide, are highly tunable via chemical reductions or annealing. The material can be further improved by multiple feasible methods. Due to these unique features, graphite oxide and reduced graphite oxide, have been greatly utilized in Li–S batteries.

However, both Ambrosi et al. (2012) reveal that graphene production by both natural and synthetic graphite impart metallic impurities that still remains after chemical reduction of graphite oxide. Such metallic impurities can effect the recognized properties of graphene. This study propose a thermal treatment of reduced graphene oxide for purification in halogen atmosphere.

By reviewing several studies, it appears that the graphene yield can vary depending on several aspects. Such as chemistry composition, various requirements, scalability, energy consumption etc. The graphene yield has an substantial influence on the total environmental impacts (Serrano-Luján et al., 2019). Thus, this becomes an important parameter and is thereby taken into account in this thesis. The model includes a yield from 60 to 80 percent.

As previously mentioned, the intensity of the electricity is also included as a parameter in the model. The selection of electricity intensity for this study, was based on the location of the most prominent manufacturers of graphene today. These locations represent a fairly large spread of values when it comes to quantifying kg CO_2 eq./kWh. Consequently, the following values are included in the parametric LCA model; 0.2, 0.4, 0.6, 0.8, 1.1, 1.4 kg CO_2 eq./kWh. The parameterization of electricity intensity applies to the Hummers' process and chemical reduction. For graphite production a global average is utilized. In addition, it was assumed *medium* voltage for all processes as the consumption is directed at industry production.

3.4 Base case scenario

In order to assess the various parameters thoroughly (in section 4.1), they are all assessed in reference to a base case scenario. By doing so, all the parameters that are not under evaluation, are kept constant and only the impacts by the parameter(s) in question are displayed in different plots and figures.

The base case scenario is chosen on the basis of representative values and locations for graphene production. In addition, some assumptions have been made. Table 2 provides an overview of the parameter settings in the base case scenario:

Table 2: Parameter settings in base case scenario.

Parameter	**Setting**
Graphite production	Natural
Modification of the Hummers' process	M2
Electricity intensity [kg CO_2 eq./kWh]	0.2
Graphene yield [%]	80

Due to cost considerations, natural graphite was selected for the base case scenario. In addition, natural graphite provides a smaller contribution to the environmental impacts compared to the synthetic alternative. However, both natural and synthetic graphite can be utilized in batteries and the majority of consumers will employ a blend of the two options, depending on the application (Leading edge materials, 2020).

Evaluating the Hummers' process modifications, it is clear that M1 and M2 have quite similar requirements. However, energy consumption is one of the factors that creates a distinction between them. The M1 modification has the lowest energy requirement in addition to overall lower quantity of input materials. To represent the range of modifications included, M2 is selected to represent the Hummers' process in the base case scenario.

The selection of electricity intensity for the base case scenario, was concluded based on one of the most prominent location for graphene manufacturers today, namely Canada (Rashotte, 2020).

As mentioned in section 3.3, graphene yield varies depending on several aspects including precursor requirements, production route, chemistry, etc. The studies Cossutta et al. (2017) and Serrano-Luján et al. (2019), which lay the foundation of inventory data for GO and chemical reduction, report a wide rage of values for the different methodological options they investigate, including a graphene yield at 80%.

3.5 Inventory analysis

In order to gather sufficient data for this assessment, several studies have been reviewed. However, there are some limitations with regard to sources that provide sufficient background data for the production methods chosen in this study. There are also noticeable variations between the results and the reported impacts from the different studies. This also applies to studies that investigate graphene impacts with the same production route, i.e. Hummers process and chemical reduction. Figure 7 illustrates the variation in GWP results from three different studies that all utilize the Hummers' process and chemical reduction.

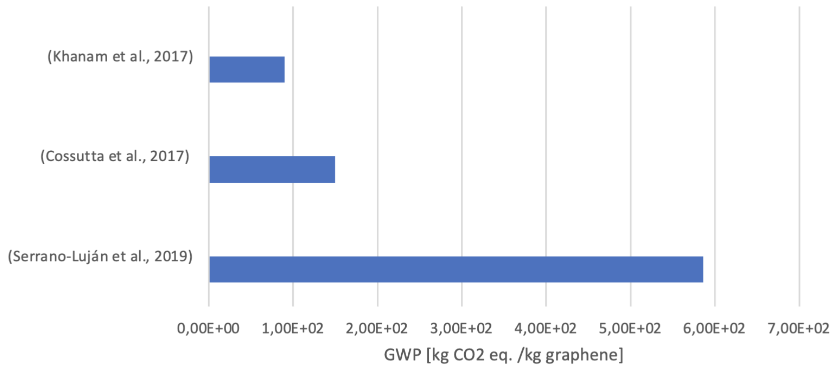

Figure 7: Comparison of the GWP value from different studies that all utilize the same production route, Hummers' process and chemical reduction.

After a closer evaluation of Arvidsson et al. (2014), Cossutta et al. (2017) and Serrano-Luján et al. (2019), it was decided that Cossutta et al. (2017) and Serrano-Luján et al. (2019) would lay the foundation for the Hummers' process and chemical reduction inventory. This was decided due to the uncertainty from Arvidsson et al. (2014), as it utilizes values that were significantly higher in comparison to the other studies and consequently leads to higher environmental impacts. Further, these two studies were compared and evaluated, both in terms of inventories and environmental impacts. Due to the data limitations, only direct emissions which derives from graphite production, were included in the model.

One of the reasons why there is limited access to data is that graphene productions are currently under development. For that reason, it is mainly laboratory data that is available and not commercial. this leads to a certain degree of uncertainty in the LCA analysis. Each study mentioned above is based on lab-scaled data, and consequently this thesis is also scaled as such. However, for the majority of the input values, the gap between laboratory and commercial scale are not substantial, with the exception of the energy requirement. The lab scaled energy requirement will be slightly higher compared to commercial scale (Cossutta et al., 2017).

Originally a total of six parameters were considered, whereas three derived from graphite production. After assessing the distribution of the environmental impacts, it was clear that the contribution from graphite was considerably smaller compared to the remaining processes. In addition, six parameters would result in approximately 1000 scenarios to assess. Therefore, it was decided to only include parameters that were considered significant to the final result. That is, graphite production route, Hummers' process modifications, electricity intensity and graphene yield, which generated 108 scenarios. As the simulation of all scenarios is a rather time-consuming process, individual runs were performed to ensure that the results were in an expected range prior to simulating all 108 scenarios.

Data collection for the graphite production, both synthetic and natural, were performed by the co-supervisor of this thesis, Nelson Manjong (Manjong et al., n.d.). For natural graphite the data were obtained from Gao, Gong, Liu, and Zhang (2018b) and Q. Zhang, Gong, and Meng (2018), while for the synthetic the main data source alternative was Dunn et al. (2015).

4 Results and analysis

This section presents the variation of environmental impacts from graphene production, including all 108 scenarios. The results are presented per functional unit. In order to create a sufficient basis for comparison for both the various parameters and the scenarios, the main focus is on global warming potential (GWP).

Firstly, a review of the significant parameters and their influence on the overall result for graphene production. Here, the parameters are assessed in reference to the same base case scenario defined in section 3.4 (Natural graphite, Hummers' process 2 (M2), 0.2 kg CO2 eq./kWh and 80% graphene yield). This is to disclose which parameters that are significant to the final environmental impact.

Further, a presentation of the overall spread of impacts for all 108 scenarios and highlighting the characteristics around best and worst case scenario. Lastly, an investigation of the contribution analysis for each production step. Note that due to data availability, only GWP from graphite production is calculated with both indirect and direct emissions. For the remaining processes, GWP is calculated based on indirect emissions.

4.1 Parameter assessment in a base case perspective

In reality, environmental impacts from various productions, activities, etc., change depending on several aspects. As mentioned previously, conventional LCA modelling struggle to capture these variation and therefore increases the level of uncertainties. Through dynamically changing the parameters, rapid changes in impacts are captured, enabling a more detailed and precise assessment.

Figure 8 provides an insight to the spread of environmental impacts by changing vital parameters from the base case scenario described above. Here, it is the electricity intensity (CO_2 eq./kWh) and graphene yield (%) that alter within the given range. The figure illustrates how large the spread of impacts can be by simply changing two parameters. This result shows a clear advantage when using a parametric LCA model for assessing such a value chains.

To clarify the influence from the parameters, this section will present both theoretical trends in addition to actual values from the model, in parallel. Note that only parameters that provide a great influence on the final GWP for graphene are presented in this section, i.e. electricity intensity, graphene yield and the Hummers' process modifications. Assessment of the graphite production routes will be discussed later in section 4.3.

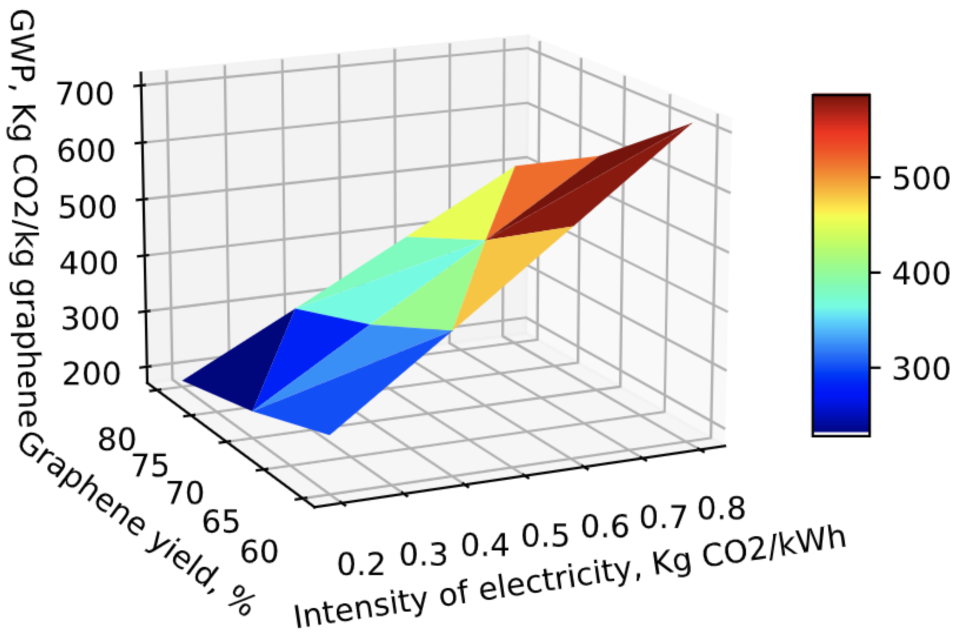

Figure 8: GWP variations in the base case scenario by altering two variables, graphene yield (%) and electricity intensity (kg CO_2 eq./kWh).

4.1.1 Electricity intensity and graphene yield

By evaluating the results provided by the parametric LCA model, it emerges that the intensity of electricity and graphene yield have a great impact on the final value of GWP for graphene production.

Figure 9 shows the theoretical development of the total GWP value by altering both electricity intensity and graphene yield in the given range. The lines with their respective colors illustrate different combinations by changing the electricity intensity on the x-axis and the graphene yield on the y-axis. As shown in the figure, an efficient process in addition to low-carbon energy consumption, is crucial to bring the environmental impacts from graphene production to a minimum.

Input values for electricity intensity utilized in the model range from 0.2 to 1.4 CO_2/kWh. This wide range affects the environmental impacts, including the GWP value, significantly. Figure 10 presents actual values generated from the parametric LCA model. The figure shows a major increase in GWP from approximately 180 to 800 kg CO_2 eq./kg graphene when the graphene yield is at 80%. The main reason for this significance is the energy consumption in the Hummers' process and the chemical reduction. This consumption is namely 380 MJ/kg graphene for the chemical reduction and varies between 12.5, 37.5 and 137.5 MJ/kg graphene for the Hummers' process, depending on choice of modification.

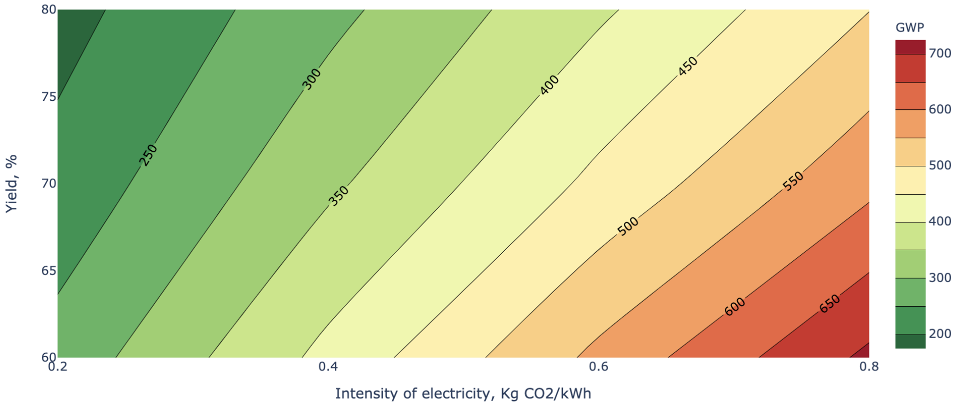

Figure 9: Contour plot of theoretical GWP values when varying graphene yield (%) and electricity intensity (kg CO_2 eq./kWh).

The graphene yield in the last stage of production, that is chemical reduction, also affects the environmental impacts substantially. This model tested 3 different values that were considered probable after a review of several relevant studies, namely 60, 70 and 80 percent.

Both Figure 9 and 10 presents a wide range of impacts when going from 60% to 80% graphene yield. Figure 10 also shows that higher energy intensity, causes a wider range in GWP values when altering yield from 60% to 80%. In the base case scenario (electricity intensity of 0.2 kg CO_2 eq./kWh), the GWP varys from 182 to 269 CO_2 eq./kg graphene due to alteration in yield. However, in a scenario where the electricity intensity is at 1.4 kg CO_2 eq./kWh, the GWP range from 808 to 1141 CO_2 eq./kg graphene.

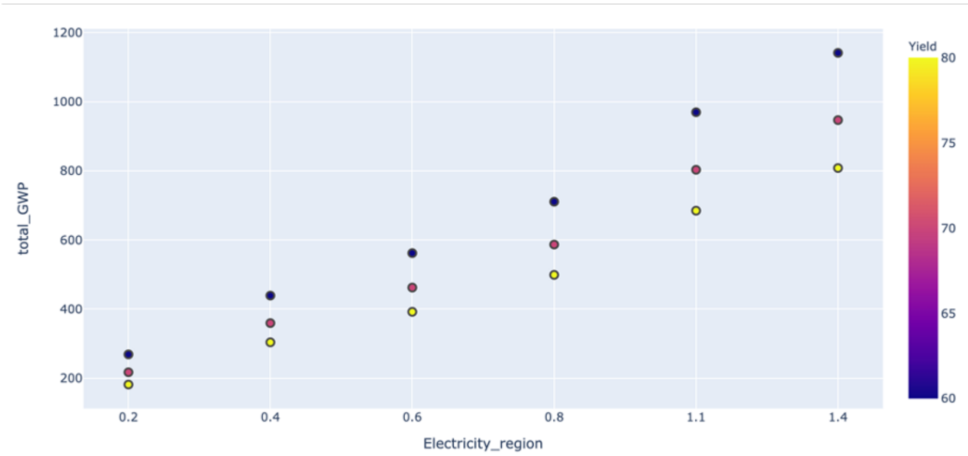

Figure 10: Actual GWP values in base case scenario when electricity intensity and graphene yield alter.

4.1.2 Modifications of the Hummers' process

Three different modifications of the Hummers' process were included as a parameter in the parametric LCA model. The different modifications are subsequently referred to as M1, M2 and M3. What separates the various modifications is the input value for sulfuric acid (H_2SO_4), hydrogen peroxide (H_2O_2) and the energy consumption. A detailed inventory can be found in Appendix A.1.4.

Figure 11 and 12 illustrate show the development of the environmental impact of the three modifications when electricity intensity increases on the x-axis. Theoretical trends is presented by Figure 11, whereas 12 presents the actual values developed by the parametric LCA model, for all six electricity intensities.

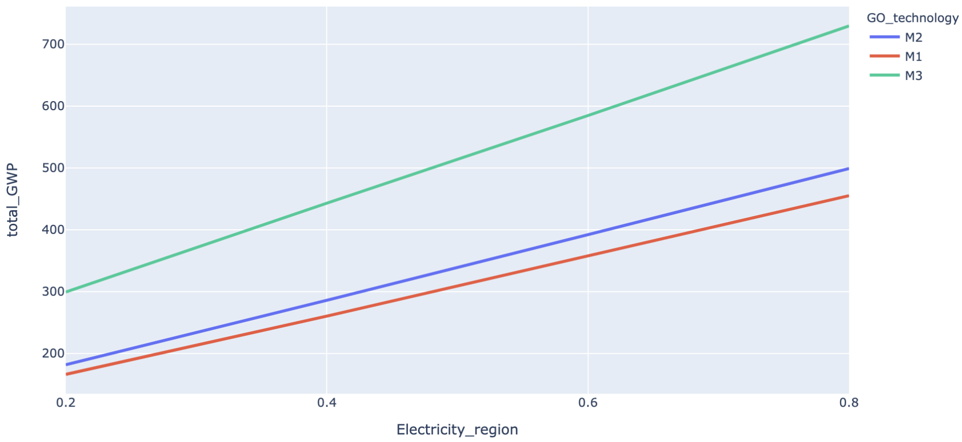

Figure 11: Theoretical trends of GWP values when varying electricity intensity (kg CO_2 eq./kWh) and modifications of the Hummers' process.

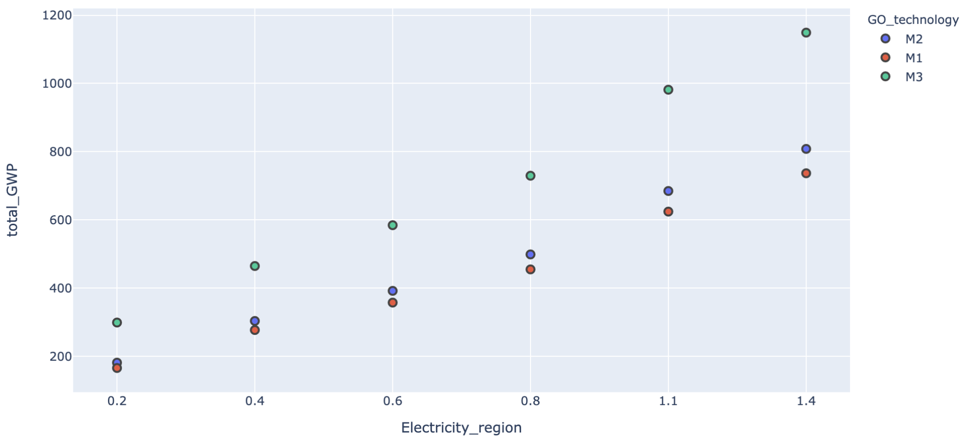

Figure 12: Actual values produced by the model of GWP values when varying electricity intensity (kg CO_2 eq./kWh) and modifications of the Hummers' process.

Both figures uncover that M1 and M2 affect the final GWP impact quite similarly, however the impact from utilizing M3 is considerably higher. For the M3 modification, the consumption of sulphuric acid and energy is almost 10 times higher compared to M1 and M2. In addition, the input value for hydrogen peroxide is almost tripled.

Investigating the base case scenario (electricity intensity of 0.2 kg CO_2 eq./kWh and graphene yield of 80%), M1 and M2 contribute to a final environmental impact at 165.1 and 181.6 kg CO_2 eq./kg graphene. Whereas the M3 scenario almost double the impact of M1 at 299.1 kg CO_2 eq./kg graphene. Note that M3 represent a worst-case scenario where no acid recovery takes place.

Overall, the scenario with the lowest impact on GWP utilize M1 for the graphite oxidation process. This is mainly due to moderate use of sulphuric acid, potassium permanganate and relatively short process duration and thereby the energy consumption is kept to a minimum. For M1 it is graphite and acid consumption that are the main contributors to GWP.

4.2 Best and worst case scenarios

As stated, the parametric LCA model generated 108 different scenarios that all represent possible environmental profiles for graphene production. Between these scenarios, there are substantial variations in terms of environmental impacts. Figure 13 illustrates the spread of impacts for the total GWP value. The figure also illustrates the variation in contributions from each stage of production. Here, the green line represents the median and the box extend one standard deviation from the median. The "whiskers" illustrate the spread of GWP values outside the box. Lastly, the circled dots represent outliers for the processes.

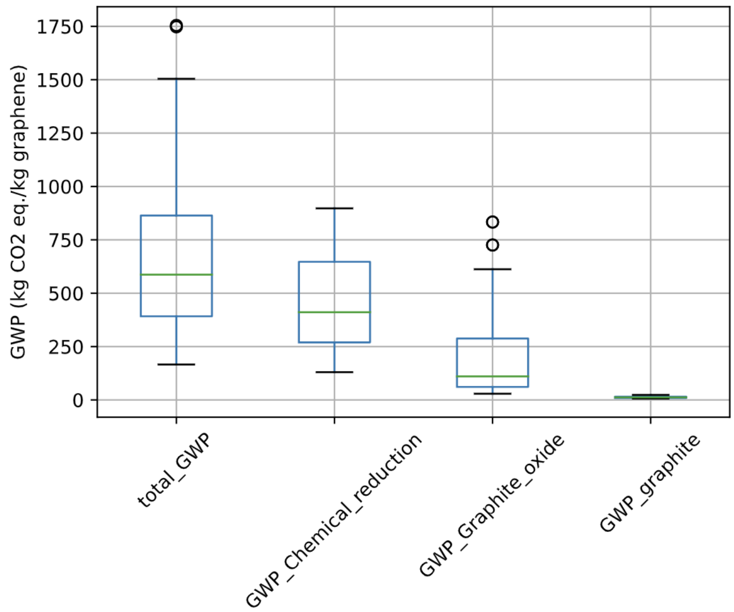

Figure 13: Illustrates the spread of impacts were all 108 scenarios are included.

By investigating the total GWP, it appears that the final impact can vary from 166 to 1750 kg CO_2 eq./kg graphene. Thus, these values constitute the best and worst case scenario for graphene production. The preconditions underlying these two scenarios are what causes the remarkable inequality. Table 3 presents an overview of the parameter settings in the best and worst case scenario for graphene production.

Table 3: Overview of the parameter settings for the best and worst case scenario.

Parameter	Best case	Worst case
Graphite production	Natural	Synthetic
Modification of the Hummers' process	M1	M3
Electricity intensity [kg CO2 eq./kWh]	0.2	1.4
Graphene yield [%]	80	60

4.3 Contribution analysis

To disclose how the total GWP result from graphene production is distributed among the various processes, contribution analysis is applied. This section will investigate the spread of results from all 108 scenarios, for each process in graphene production.

This section also investigates which parameters are significant for each of the foreground processes. Firstly, evaluate the contribution to GWP from chemical reduction, further the Hummer's process and lastly the graphite production.

An overview of the GWP contribution from each process in graphene production was presented in Figure 13 (section 4.2). Figure 14 provides a more detailed understanding of how the impacts or contributions change as there is an increase in energy intensity. As mentioned in section 4.1.1, the majority of the contribution to the final GWP stem from chemical reduction and the Hummers' process. Note that this figure does not distribute a clear understanding of the contribution from graphite production, as these impacts are considerably lower compared the remaining processes. In addition, all impacts are presented in the same scale for GWP. Nonetheless, the contribution from graphite production will be discussed later in section 4.3.3.

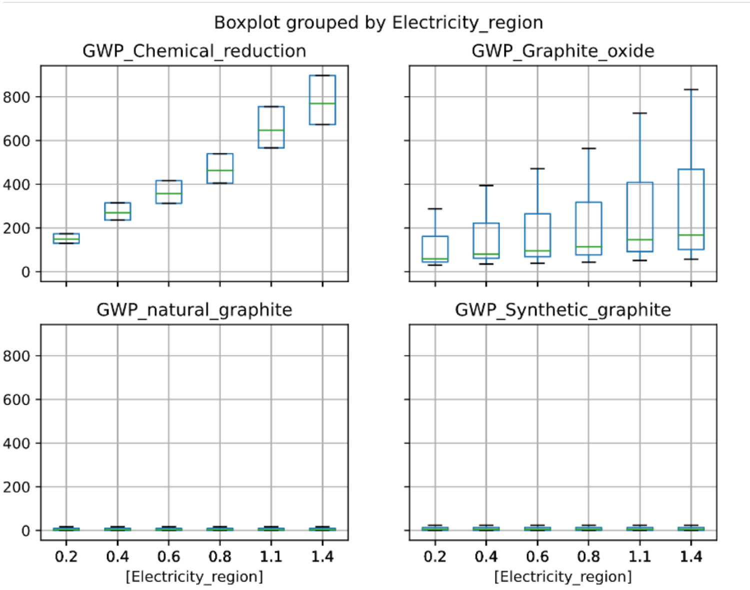

Figure 14: Contribution to GWP from each process in graphene production as the electricity intensity increases.

4.3.1 The chemical reduction

It has already been established that chemical reduction has a major impact on the overall GWP for graphene. Figure 15 and 16 investigate by what means the two parameters, graphene yield and electricity intensity affects the GWP contribution from the chemical reduction process. The figures show that the quantity of CO_2 equivalents in the energy mix utilized for the process has a remarkable effect on the final impact level from chemical reduction. The graphene yield plays an important role as well in minimizing the environmental impacts from the process. The chemical reduction process alone has a GWP contribution between 130 and 897 kg CO_2 eq./kg graphene depending on the preconditions.

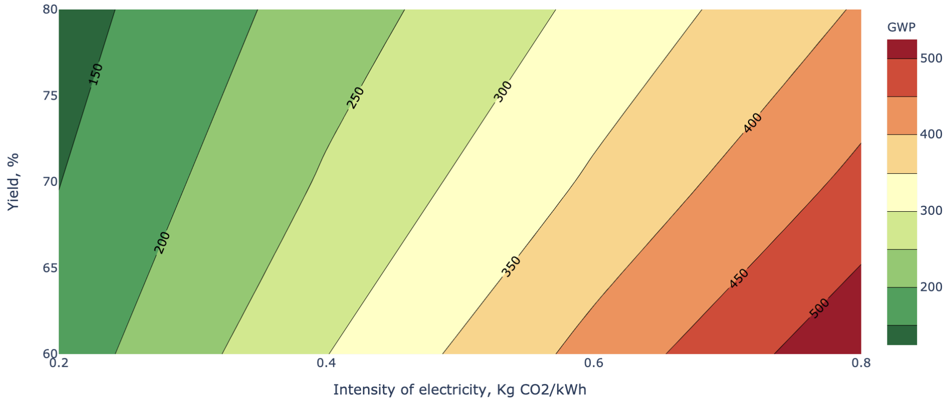

Figure 15: Contour plot of theoretical GWP contribution of the chemical reduction when varying graphene yield (%) and electricity intensity (kg CO_2 eq./kWh).

The main contributors to GWP in the chemical reduction process is the energy consumption (approximately 75% of the total consumption in graphene production) in addition to hydrazine and ethanol. Here, the energy consumption represents approximately half of the contribution. This process requires as much as 380 MJ/kg graphene produced. As Figure 16 illustrate, the graphene yield can also have a major impact on the contribution to GWP. By evaluating the best case scenario for this production stage (electricity intensity of 0.2 kg CO_2 eq./kWh) the GWP can vary from 130 to 174 kg CO_2 eq./kg graphene. For the worst case scenario (electricity intensity of 1.4 kg CO_2 eq./kWh) the GWP can vary from 673 from 897 eq./kg graphene.

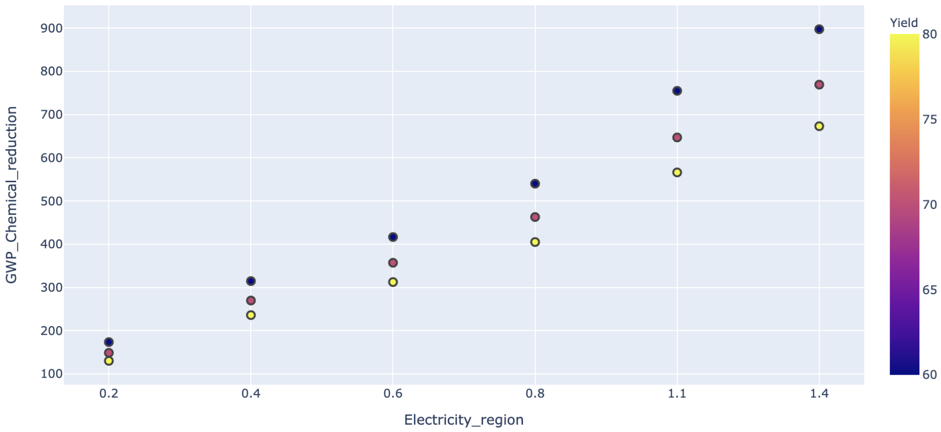

Figure 16: The GWP contribution by the chemical reduction process (kg CO_2 eq./kg graphene).

4.3.2 The Hummers' process

Similar to the chemical reduction process, the electricity intensity has a major impact on the contribution to GWP from the Hummers process. Figure 17 demonstrate the theoretical trends, here the contribution to GWP, by altering between the three different modifications. Whereas figure 18 represent actual values generated from the model. The figures illustrates that there are significant variations in impacts for all modifications, but especially M3. In a scenario with M3, the GWP contribution from this process alone can vary between 162 and 317 CO_2 eq./kg graphene by altering the electricity intensity from 0.2 to 1.4 kg CO_2 eq./kWh. This is due to the significant energy requirement, namely 12.5, 37.5 and 137 MJ/kg graphene for M1, M2 and M3 respectively.

The choice of modification for the Hummers process will also be substantial to the GWP contribution. it is clear that a careful assessment of the chemical composition for the Hummers' process is crucial. The more efficient the process and an increased focus on recovering and reusing inputs wherever feasible, will minimize the impact. Note that M1 and M2 have the same yield (approx. 88% yield), but M3 is simulated with a somewhat lower yield for graphite oxide (approx. 77%). In addition, virtually no acid recovery is included for the modification M3.

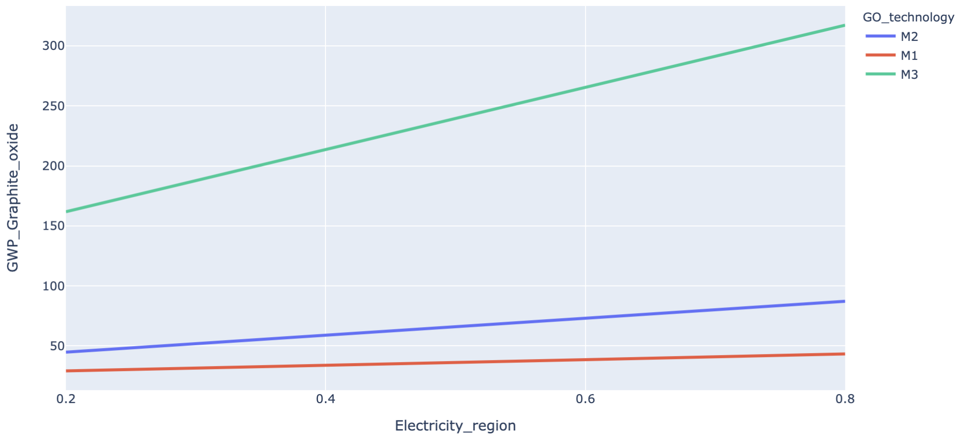

Figure 17: Theoretical trends of the GWP contribution by the Hummers' process in (kg CO_2 eq./kg graphene).

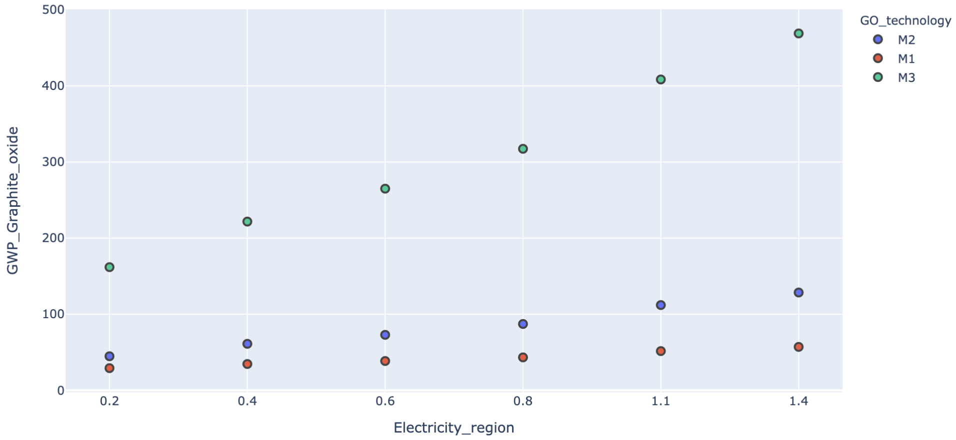

Figure 18: Actual values produced by the model of the GWP contribution by the Hummers' process in (kg CO_2 eq./kg graphene).

4.3.3 Graphite production

This study includes both synthetic and natural graphite production as parameters in the parametric LCA model. After analyzing all scenarios, it is clear that the impact level from synthetic graphite production is slightly higher compared to natural production. However, in light of the study's functional unit, the difference is not substantial.

Figure 19 shows the the overall impact from both synthetic and natural graphite production. In addition, it illustrates the variations from all 108 scenarios developed by the parametric LCA model. When graphite production is consider isolated from the remaining processes, the difference in impacts are noticeable. Investigating the base case scenario from section 4.1, the graphite production contribute with approximately 4-5 percent depending on the graphite production route. Thus, the graphite production route becomes the least significant parameter for the overall GWP result in this study.

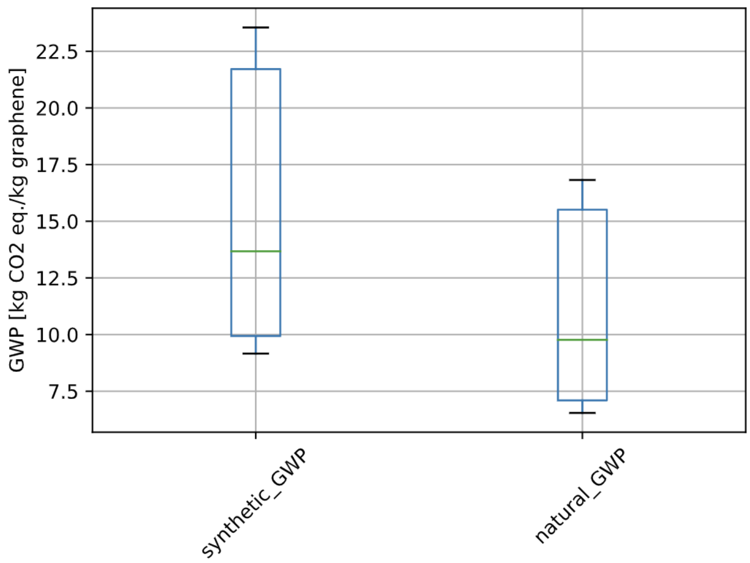

Figure 19: The variation in contribution to GWP from synthetic and natural graphite production. All 108 scenarios are included.

Despite a relatively small contribution from graphite production, the level of environmental impact varies between natural and synthetic graphite production. As shown in Figure 19, the contribution to GWP from synthetic graphite varies from 9.2 to 23.6 kg CO_2 eq./kg graphene, while the contribution from natural graphite varies from 6.5 to 16.8 CO_2 eq./kg graphene. The variation of impacts from this production stage is due to different process efficiencies in the remaining production steps. As mentioned in section 3.5, it was first intended to include more parameters from the graphite production, but as the influence on the outcome were insignificant, they were not included. This means that some of the values from graphite production are set statically. Namely, beneficiation recovery efficiency at 95% and the ore grade at 15%.

The GWP contribution from natural graphite consists of contributions from both graphite mining and processing. Between these to sub-processes, the distribution of GWP was fairly even. One of the main contributions to environmental impacts for natural graphite production is the energy consumption followed by the requirements of raw coal, coke oven gas, diesel, NaOH and others. Here, the electricity consumption and raw coal contribute with approximately 21.98% and 24% to the total carbon emission respectively. The direct emissions derived from the production process and fuel combustion.

The energy requirement is also an important contributor to environmental impact in synthetic graphite production. The graphitization and baking are the two most energy-intensive processes. It should be mentioned that the data source for synthetic graphite did not account for emission control technologies.

5 Discussion

The objective of this study was to investigate the environmental impacts from producing 1 kg graphene by utilizing a parametric life cycle assessment (LCA) model. Thus, enable an analysis of the interplay between the key parameters in the graphene value chain and detect the variation of impacts by altering them dynamically. By utilizing this model, rather than conventional LCA, the aim was to achieve a more detailed understanding of the relevant processes and increase the precision of the analysis. A total of four variables (graphite production route, modifications of the Hummers process, electricity intensity and graphene yield) were parameterized, which resulted in 108 different scenarios for graphene production.

This section will revisit the mission statement and assess to what extent the goal of this study was achieved, in addition to presenting some of the main findings of the assessment. Further, a discussion of some key assumptions and limitations of the study, as well as the environmental implications. Lastly, recommendations for further research are provided. Complete inventory are available in Appendix A.1.

5.1 Mission statement revisited

A parametric LCA for graphene production is preformed were graphite production routes, modifications of the Hummers' process, electricity intensity and graphene yield are chosen as key parameters. This resulted in 108 different scenarios. In order to investigate the influence of the significant parameters, they where assessed in reference to a base case scenario described in section 3.4. To create a sufficient basis for comparison for both the various parameters and the scenarios, the main focus was on global warming potential (GWP). Since the results showed large variations within the 108 scenarios, the best and worst case scenario where presented with the respective parameter settings and preconditions. Lastly, a contribution analysis is also performed to detect which process the majority of life cycle emissions occur and disclose the causing effect.

By analyzing the results generated from the model, it is clear that the parameters; graphene yield, modifications of the Hummers' process and electricity intensity have a great influence on the level of environmental impact from graphene production. In this context, the choice of graphite production method became less significant as the impact from this process only represent approximately 4-5 percent (in base case scenario) of the total GWP. Due to the immense energy consumption of the chemical reduction and the Hummers' process, the energy intensity (kg CO_2 eq./kWh) is crucial to minimize the environmental impact from the production.

In the previously described base case scenario, it was shown that altering the electricity intensity from 0.2 to 1.4 CO_2/kWh, would cause an increase in GWP from 180 to 800 kg C_2 eq./kg graphene. These results substantiates the importance of closely evaluating the energy mix utilized both in production and when preforming a life cycle assessment.

In section 4.2, the spread of environmental impacts from all scenarios are presented. These results show that the parametric LCA model is able to detect possible variations by dynamically changing the parameters, where each result represents a possible scenario for graphene production. This is a property that conventional LCA does not possess. Thereby, a more detailed and precise assessment is obtained.

5.2 Key assumptions and limitations

In order to perform an LCA analysis, it entails collecting large amounts of data from various sources and it often involves certain assumptions. This can lead to a level of uncertainty to the assessment. Therefore, it is necessary to consider this aspect, both internal and external with the object of assessing the overall quality of the study.

5.2.1 Internal evaluation

One of the biggest challenges in order to preform parametric LCA on graphene production, was to gather sufficient and up-to-date data for the entire graphene value chain. Although more and more research are being published, there are multiple production routes and derivatives for graphene. This study has, as previously mentioned, considered a production that are commonly utilized in the context of battery production. Therefore, there were some limitations as the inventory did not include direct emissions from the Hummers' process or the chemical reduction, only graphite production. For the Hummers' process and the chemical reduction, the inventory was based on two sources that were considered credible. However, before the final simulation of all 108 scenarios, several single runs, with different parameter settings, were tested and then compared with several applicable studies. Thereby obtained confirmation that the GWP value(s), were within an acceptable range.

Due to limited data availability, there are uncertainties in connection with the chemical reduction and Hummers' process' inventory as they are at laboratory scale, and not commercial scale. Such data is based on key assumptions associated material recovery , graphene yield and energy efficiency. However, for the majority of the input values, the gap between laboratory and commercial scale are not substantial, with the exception of the energy requirement. The lab scaled energy requirement is slightly higher compared to commercial scale (Cossutta et al., 2017).

Finally, it is also important to mention that some parameter compositions are more likely than others. For example, the modification of the Hummers' process, M3, is not commonly utilized, but it is included to signify the influence of the chemical composition/scaling of the input values have on the total GWP for graphene.

Despite these uncertainties, the results provided in this thesis are important to uncover the main drivers for environmental impacts derived from graphene. It is also possible to connect the impacts from key parameters in production.

5.2.2 External evaluation

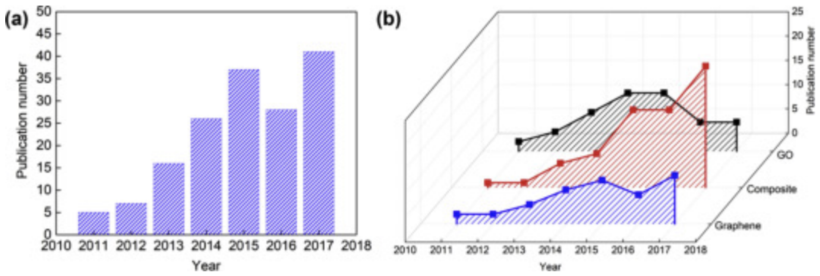

Figure 20: Statistics on graphene/graphite oxide publications for Li–S batteries. (a) Publications peaked in 2015 and slowed down in 2016 but bounced back in 2017. (b) Publications on graphene/graphite oxide based cathodes peaked in 2014. More studies followed up quickly on composites after 2015, indicating that graphene/graphite oxide are promising for high-performance Li–S batteries (Zhang et al., 2018).

Figure 20 presents the amount of publications on graphene and graphite oxide given the time period. This illustrates the increased interest and research of the material and its applications. Note that in year 2017, approximately half of the publications are on composites, indicating that graphene-based composites are promising for Li-S batteries with high performance (Zhang et al., 2018). However, despite the fact that there is ever-increasing research on the material, there are limited publications concerning the environmental impacts.

After evaluating several relevant studies, it is clear that the estimated environmental impacts between different graphene production routes vary greatly. This also applies to studies that consider the same production route. Such as the Hummers' process for graphite oxidation followed by chemical reduction. Consequently, increases the uncertainty associated with the results in this thesis.

A study by Serrano-Luján et al. (2019), conducted an cradle-to-gate LCA of graphene from the same production route as described in this thesis. This study utilize experimental data that are laboratory scaled. As the various data have been measured at a Spanish laboratory, Spanish-2015 electricity mix is utilized. The final global warming potential from producing 1 kg graphene, is measured to be 586 kg CO_2 eq./kg graphene.

A cradle-to-gate LCA performed by Cossutta et al. (2017) investigated several production routes and concluded that the least impacting production route remains Hummers' process followed by thermal reduction (90 kg CO_2 eq./kg graphene). However, the impact from combining the Hummers' process with chemical reduction, as in this thesis, was estimated at 150 kg CO_2eq./kg graphene. They also find that environmental impacts vary greatly between the production routes. In addition, a study by Khanam et al. (2017) presents the GWP from producing graphene, with the same production routes, at approximately 90 kg CO_2 eq./kg graphene. And preformed a cradle-to-gate LCA with laboratory scaled data.

These studies provide a specific range that are used as a reference for comparison of the results developed by the parametric LCA model. The base case scenario described in section 3.4 present a total GWP at 182 kg CO_2 eq./kg graphene, which is within the range provided by the previous studies. The overall results generated from all 108 scenarios, describe a GWP range to be approximately 166 to 1750 kg CO_2 eq./kg graphene. The highest values for GWP are outside the range described above, but they represent the worst case scenario where all parameters can be altered to decarbonize the graphene production.

5.3 Environmental implications

Conducting a parametric life cycle assessment on graphene production enables a more detailed and precise understanding of the environmental implications of the various processes in graphene production. Recent research on various graphene applications has been high on the agenda. However, research on environmental impacts from graphene production is inadequate as of today. The results provided in this study show that the two processes; graphite oxidation and chemical reduction are the largest contributors to GWP. This implies that the parameters that apply to these two processes are significant. Both processes are energy-intensive. This means that an increase from 0.2 to 1.4 kg CO_2 eq./kWh cause a remarkable impact on the total GWP for graphene production. In the base case scenario this lead to an increase from approximately 180 to 800 kg CO_2 eq./kg graphene. These results substantiates the importance of closely evaluating the energy mix utilized both in production and when preforming an life cycle assessment. A Nordic battery value chain would hold a major advantage due to the relatively low-carbon energy mix. They also verify the importance of assessing such variations of parameters, which are not achievable with conventional LCA.

An alteration of graphene yield (applies to the chemical reduction process) from 60 to 80 percent resulted in an decrease in total GWP from 269 to 182 kg CO_2 eq./kg graphene, also in the base case scenario. In addition, evaluating the chemical composition of the graphite oxidation process, here the Hummers' process, is also crucial in order to minimize the environmental impact from graphene production. As described in section 3.2, this study assessed three different modifications (M1, M2 and M3) to this process. The results show that M1 has the smallest contribution to GWP, and is therefore part of the best case scenario presented in section 4.2. From a base case perspective, M1, M2 and M3 contribute 165.1, 181.6 and 299.1 kg CO_2 eq./kg graphene respectively. These results also indicate that it is essential to focus on material recovery and reuse in order to decarbonize the production.

As publications associated with environmental implications of graphene production are limited, the parametric LCA model can contribute to reveal the influence of the various stages of production. As a total overview is attained This provides the opportunity to consider adequate measures to decarbonize the graphene production, which will be crucial in a larger context, since the material is part of several value chains, e.g. the Li-S battery. According to IPCC AR5 WG III (2014), without implementing ambitious mitigation policies, emissions derived from the transport sector in particular will increase faster compared to any other energy end-use sector.

5.4 Further research

Reduction of emissions, including from the transport sector, is gaining more and more focus. This has lead to substantial investments in new battery technologies with focus on renewable energy in the industry (Wagner, 2020). As battery production increases significantly it is crucial to assess relevant materials that are included in the battery value chain. Here it is substantial to detect the environmental impacts from various processes in order to implement measures that are adequate and efficient to reduce both direct and indirect emissions.

As this study processed extensive amounts of data from 108 scenarios, and aimed to provide the best possible assessment and visualization of the results, it was considered best to only examine the global warming potential from graphene production. In order to attain a more detailed understanding, other environmental impact categories should be included in the assessment. Such as Human health, particulate matter, etc. As mentioned, this study assessed graphene from a cradle-to-gate perspective. For future research, it is crucial that the entire life cycle is considered to ensure that environmental impacts from end-of-life treatments are included.

As mentioned, this study is primarily based on laboratory scaled data provided by Serrano-Luján et al. (2019) and Cossutta et al. (2017). By making commercial data available, an opportunity for further research is possible with a greater focus on the industry and more precise in terms of estimating environmental impacts (Kwade et al., 2018).

Despite challenges related to large-scale production of graphene, there has been a remarkable progression in the short time since the technological material was discovered. The prototypes of commercialized graphene-based products, e.g. sensors, graphene inks, various applications in batteries etc., give reasons to be optimistic about future production and applications of graphene (Phiri, Gane, & Maloney, 2017).

6 Conclusion

Graphene has attracted much attention due to its properties and potential applications. This material is a highly conductive, flexible and mechanically robust. Graphene hold promises in eliminating the barriers for commercialization of Li–S batteries. There are multiple production routes for graphene, this study reviews natural and synthetic graphite production, the Hummers' process for graphite oxidation and chemical reduction for the final stage in production.

This study investigates the global warming potential (GWP) from producing 1 kg graphene by utilizing a parametric life cycle assessment (LCA) model. Thus, enables an assessment of the interplay between the key parameters in the graphene value chain and detect the variation of impacts by altering them dynamically. This model provide a detail understanding of the relevant processes and increases the precision of the analysis. A total of four variables (graphite production route, modifications of the Hummers' process, electricity intensity and graphene yield) were parameterized, which resulted in 108 different scenarios for graphene production.

The results provided in this study show that the two processes: graphite oxidation and chemical reduction are the largest contributors to GWP. This implies that the parameters that concern these two processes are significant, i.e. electricity intensity and graphene yield in the chemical reduction process. In addition, evaluating the chemical composition of the graphite oxidation process is crucial in order to minimize the environmental impact from graphene production. In this context, the choice of graphite production method is less significant as the impact from this process only represent approximately 4-5 percent of the total GWP value from graphene.

There has been an increased interest in and research of the material and its applications. However, there are limited publications concerning the environmental impacts from graphene. Thus, there were limited data availability for the entire value chain of graphene. After evaluating several relevant studies, it is a clear that the estimated environmental impacts for different graphene production routes vary greatly. This also applies to studies that consider the same production route.

The inventory for graphite oxidation and chemical reduction in this thesis, is based on the sources Serrano-Luján et al. (2019) and Cossutta et al. (2017). In addition, These studies provide a specific range that are used as a reference for comparison of the results developed by the parametric LCA model. Similar studies were utilized to compare the inventories and results.

The results substantiate that the parametric LCA model is able to detect possible variations by dynamically changing the parameters, where each result represents a possible scenario for graphene production. This is a property that conventional LCA does not possess. When an overview is obtained of all production steps and the entire value chain at a such detailed level, an opportunity to consider adequate measures to decarbonize the graphene production is provided. This will be crucial in a larger context as the material is part of several value chains, e.g. the Li-S battery.

Since this study evaluates graphene production based on laboratory-scaled data, in addition to only assessing the global warming potential of graphene production, there are some limitations in the study. Further recommendations would be to extend the assessment by including more impact categories to achieve more details on the environmental impacts, in addition to performing assessments based on commercial data if available. It is also recommended that future research evaluate the entire life cycle of graphene, including impacts from end-of-life treatments.

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A Appendices

A.1 Inventories and background data

A.1.1 Foreground system data

	Label (PRO_f):			A_ff:	1	2	3	4	5	6	7	8
	FULL NAME	PROCESS I			Graphene	Chemical reduction	Graphite oxide	Graphite	synthetic graphite	Natural graphite	Processing of natural grap	Mining of natural g
1	Graphene	50 000 000	1,25									
	Chemical reduction	50 000 001	0		1							
2	Graphite oxide production	50 000 002	0			1,56E+00						
3	Graphite	50 000 003	0				1,109375					
4	synthetic graphite	50 000 004	0					0				
5	Natural graphite	50 000 005	0					1,0526				
6	Processing of natural graphite	50 000 006	0							1		
7	Mining of natural graphite	50 000 007	0								1,11	

Figure 21: The foreground system for a scenario with 80% graphene yield and natural graphite production.

A.1.2 The A_{bf} for the base case scenario

Background Process Name	***Foreground Process Name***	***(Matrix Rov***	***(Matrix column position)***	
Comment	***Comment***	***BACKGROU***	***FOREGROUND PROCESS ID #***	***VALUE***
Blasting	Mining of natural graphite	7300	50000007	2,0E-05
Limestone Quary Infrastructure	Mining of natural graphite	13171	50000007	5,3E-11
Industrial Machine	Mining of natural graphite	8966	50000007	2,3E-04
conveyor belt	Mining of natural graphite	8824	50000007	2,8E-08
recultivation	Mining of natural graphite	13129	50000007	6,5E-06
natural gas	Mining of natural graphite	10823	50000007	1,6E+00
Diesel	Mining of natural graphite	13604	50000007	6,7E+00
electricity, medium voltage/market group for electricity, medium voltage/GLO/kWh	Mining of natural graphite	10815	50000007	6,9E-01
Lime	Processing of natural graphite	4981	50000006	5,6E-03
NaOH	Processing of natural graphite	6612	50000006	5,0E-01
HCl	Processing of natural graphite	6470	50000006	4,5E-01
electricity, medium voltage/market group for electricity, medium voltage/GLO/kWh	Processing of natural graphite	10815	50000006	2,0E-01
Pet Coke	synthetic graphite	5862	50000004	9,9E-01
Coal tar pitch	synthetic graphite	5863	50000004	2,5E-01
Natural gas	synthetic graphite	10823	50000004	6,0E+00
electricity, medium voltage/market group for electricity, medium voltage/GLO/kWh	synthetic graphite	10815	50000004	4,1E+00
Potassium permanganate (KMnO4)	Graphite oxide production	552	50000002	2,7E+00
Sodium nitrate (NaNO3)	Graphite oxide production	6823	50000002	4,5E-01
Sulphuric acid (H2SO4) [ml]	Graphite oxide production	6846	50000002	4,1E+01
Phosphoric acid (H3PO4) [ml]	Graphite oxide production	536	50000002	0,0E+00
Hydrogen peroxide (H2O2) [ml]	Graphite oxide production	6255	50000002	3,3E+00
Canada mix	Graphite oxide production	10821	50000002	3,8E+01
Spain mix	Graphite oxide production	1145	50000002	0,0E+00
Great Britain mix	Graphite oxide production	1156	50000002	0,0E+00
Ireland mix	Graphite oxide production	10729	50000002	0,0E+00
China mix	Graphite oxide production	10812	50000002	0,0E+00
India mix	Graphite oxide production	10761	50000002	0,0E+00
deionized water	Graphite oxide production	11805	50000002	2,9E+02
hydrazine (N2H4)	Chemical reduction	6245	50000001	1,4E+00
ammonia (NH3)	Chemical reduction	6946	50000001	3,4E-01
Methanol	Chemical reduction	745	50000001	1,0E+01
Canada mix	Chemical reduction	10821	50000001	3,8E+02
Spain mix	Chemical reduction	1145	50000001	0,0E+00
Great Britain mix	Chemical reduction	1156	50000001	0,0E+00
Ireland mix	Chemical reduction	10729	50000001	0,0E+00
China mix	Chemical reduction	10812	50000001	0,0E+00
India mix	Chemical reduction	10761	50000001	0,0E+00

Figure 22: The A_{bf} matrix for the base case scenario described in section 3.4. Note that this inventory would change for each scenario.

A.1.3 Background system data

STRESSOR NAME	(Matrix row)	(Matrix column)	(Value)
Comment	**STRESSOR ROW #**	**FOREGROUND PROCESS ID #**	**AMOUNT**
NOX	27363	50000004	9,3E-03
PM	403	50000004	4,1E-03
SOX	26599	50000004	6,4E-02
CO2	26883	50000004	4,4E-01
Particulates, <2.5 um	394	50000007	8,9E-06
Particulates, >10 um	399	50000007	1,2E-04
Particulates, >2.5 um, and <10um	403	50000007	4,8E-05

Figure 23: Stressor inputs for graphite production.

A.1.4 Background data for the parametric model

		Graphite	Natural graphite			graphite oxide		
7								
8		Graphite route	Ore Grades	Beneficiation recovery efficiency Natural graphite (%)	Electricity region (Natural graphite)	Yield [g graphite oxide/graphene]	Electricity region	Graphite oxide prod. Route/technology
9	*Graphite*	Natural	15 %	95 %	CA	80,00 %	CA	M2
10			*These parameters will be static.*					

Figure 24: The parameters page with base case settings. Values in the A_{bf} were linked to these parameter settings.

Base Inventory [A]

		Unit	Battery Grade Graphite	Graphite Flakes (Mining+Beneficiation
	Processing of Natural graphite	kg	0	0
A_ff	Mining of Natural graphite	kg	1,05	0
A_bf	Blasting	p	0	0,0000773
	Limestone Quary Infrastructure	kg	0	5,3E-11
	Industrial Machine	kg	0	2,3E-04
	conveyor belt	m	0	2,8E-08
	recultivation	m2	0	6,5E-06
	natural gas	MJ	0	f(ore grade)
	Diesel	MJ	0	f(ore grade)
	Electricity	kWh	0	f(ore grade)
	Lime	kg	0,01	0,00
	NaOH	kg	0,50	0,00
	Electricity	kWh	0,20	0,00
	HCl	MJ	0,45	0,00

2. Synthetic Graphite

Carbonization	Units	Value	Ecoinvent ID
Pet Coke	kg	0,985625	5862
Coal tar pitch	kg	0,249	5863
Natural gas	MJ	6	10823
Graphitization Electricity	kWh	4,103078	10815

Emissions			
NOX	kg	0,0093	27363
PM	kg	0,0041	403
SOX	kg	0,064	26599
CO2	kg	0,44	26883

Figure 25: Base inventory for both natural and synthetic production route.

			M2	M1	M3
	chemical reduction of graphite oxide to graphene				
A_ff	Oxidation of graphite to graphite oxide	1,25			
A_bf	Graphite oxide	kg/kg graphene	1,25		
	hydrazine (N2H4)	kg/kg graphene	1,38		
	ammonia (NH3)	kg/kg graphene	0,34		
	Deionised water	kg/kg graphene	163		
	Methanol	kg/kg graphene	10		
	Electricity	MJ/kg graphene	380		
	Specific feature		M2	M1	M3
	Potassium permanganate (KMnO4)	kg/kg graphene	2,675	2,675	3,85
	Sodium nitrate (NaNO3)	kg/kg graphene	0,45	0,45	0,4875
	Sulphuric acid (H2SO4)	kg/kg graphene	41	37,75	310
	Phosphoric acid (H3PO4) [ml]	ml			
	Hydrogen peroxide (H2O2)	kg/kg graphene	3,25	1,55	8,3625
	Input graphite	kg/kg graphene	0,8875	0,8875	0,9625
	Output graphite oxide	kg/kg graphene	**1,25**	**1,25**	**1,25**
	Yield [g graphite oxide/g graphite]		1,408450704	1,408450704	1,298701299
	Electricity	MJ/kg graphene	37,5	12,5	137,5
	deionized water	kg/kg graphene	290	278,75	528,75

Figure 26: Base inventory for chemical reduction and the three modifications of the Hummers' process.

A.2 Python codes

A.2.1 Creating the inputfile

```
import pandas as pd
import numpy as np

graphite_tech=["Synthetic","Natural"]
mix=["CA", "ES","GB","IR", "CN","IN"]
GO=["GO1", "GO2","GO3"]
yield_GO=[60,70,80]
sc_name=[]
tech=[]
elect=[]
go=[]
yieldGO =[]
count=1


for i in graphite_tech:
    for j in mix:
        for l in GO:
            for n in yield_GO:
                    sc_name.append("sc_"+str(i)+"_Elec_"+str(j)+"_GO technology_"+str(l)+"_yield
                    _"+str(n))
                    count=count+1
                    tech.append(i)
                    elect.append(j)
                    go.append(l)
                    yieldGO.append(n)

df=pd.DataFrame()
df["Scenario"]=sc_name
df["Graphite production"]=tech
df["Electricity mix"]=elect
df["GO technology"]=go
df["GO yield"]=yieldGO
```

A.2.2 Input file

	Graphite prod.	El. mix	GO tech.	GO yield
sc_Synthetic_Elec_CA_GO technology_GO1_yield_60	Synthetic	CA	GO1	60
sc_Synthetic_Elec_CA_GO technology_GO1_yield_70	Synthetic	CA	GO1	70
sc_Synthetic_Elec_CA_GO technology_GO1_yield_80	Synthetic	CA	GO1	80
sc_Synthetic_Elec_CA_GO technology_GO2_yield_60	Synthetic	CA	GO2	60
sc_Synthetic_Elec_CA_GO technology_GO2_yield_70	Synthetic	CA	GO2	70
sc_Synthetic_Elec_CA_GO technology_GO2_yield_80	Synthetic	CA	GO2	80
sc_Synthetic_Elec_CA_GO technology_GO3_yield_60	Synthetic	CA	GO3	60
sc_Synthetic_Elec_CA_GO technology_GO3_yield_70	Synthetic	CA	GO3	70
sc_Synthetic_Elec_CA_GO technology_GO3_yield_80	Synthetic	CA	GO3	80
sc_Synthetic_Elec_ES_GO technology_GO1_yield_60	Synthetic	ES	GO1	60
sc_Synthetic_Elec_ES_GO technology_GO1_yield_70	Synthetic	ES	GO1	70
sc_Synthetic_Elec_ES_GO technology_GO1_yield_80	Synthetic	ES	GO1	80
sc_Synthetic_Elec_ES_GO technology_GO2_yield_60	Synthetic	ES	GO2	60
sc_Synthetic_Elec_ES_GO technology_GO2_yield_70	Synthetic	ES	GO2	70
sc_Synthetic_Elec_ES_GO technology_GO2_yield_80	Synthetic	ES	GO2	80
sc_Synthetic_Elec_ES_GO technology_GO3_yield_60	Synthetic	ES	GO3	60
sc_Synthetic_Elec_ES_GO technology_GO3_yield_70	Synthetic	ES	GO3	70
sc_Synthetic_Elec_ES_GO technology_GO3_yield_80	Synthetic	ES	GO3	80
sc_Synthetic_Elec_GB_GO technology_GO1_yield_60	Synthetic	GB	GO1	60
sc_Synthetic_Elec_GB_GO technology_GO1_yield_70	Synthetic	GB	GO1	70
sc_Synthetic_Elec_GB_GO technology_GO1_yield_80	Synthetic	GB	GO1	80
sc_Synthetic_Elec_GB_GO technology_GO2_yield_60	Synthetic	GB	GO2	60
sc_Synthetic_Elec_GB_GO technology_GO2_yield_70	Synthetic	GB	GO2	70
sc_Synthetic_Elec_GB_GO technology_GO2_yield_80	Synthetic	GB	GO2	80
sc_Synthetic_Elec_GB_GO technology_GO3_yield_60	Synthetic	GB	GO3	60
sc_Synthetic_Elec_GB_GO technology_GO3_yield_70	Synthetic	GB	GO3	70
sc_Synthetic_Elec_GB_GO technology_GO3_yield_80	Synthetic	GB	GO3	80
sc_Synthetic_Elec_IR_GO technology_GO1_yield_60	Synthetic	IR	GO1	60
sc_Synthetic_Elec_IR_GO technology_GO1_yield_70	Synthetic	IR	GO1	70
sc_Synthetic_Elec_IR_GO technology_GO1_yield_80	Synthetic	IR	GO1	80
sc_Synthetic_Elec_IR_GO technology_GO2_yield_60	Synthetic	IR	GO2	60
sc_Synthetic_Elec_IR_GO technology_GO2_yield_70	Synthetic	IR	GO2	70
sc_Synthetic_Elec_IR_GO technology_GO2_yield_80	Synthetic	IR	GO2	80
sc_Synthetic_Elec_IR_GO technology_GO3_yield_60	Synthetic	IR	GO3	60
sc_Synthetic_Elec_IR_GO technology_GO3_yield_70	Synthetic	IR	GO3	70
sc_Synthetic_Elec_IR_GO technology_GO3_yield_80	Synthetic	IR	GO3	80
sc_Synthetic_Elec_CN_GO technology_GO1_yield_60	Synthetic	CN	GO1	60
sc_Synthetic_Elec_CN_GO technology_GO1_yield_70	Synthetic	CN	GO1	70
sc_Synthetic_Elec_CN_GO technology_GO1_yield_80	Synthetic	CN	GO1	80
sc_Synthetic_Elec_CN_GO technology_GO2_yield_60	Synthetic	CN	GO2	60
sc_Synthetic_Elec_CN_GO technology_GO2_yield_70	Synthetic	CN	GO2	70
sc_Synthetic_Elec_CN_GO technology_GO2_yield_80	Synthetic	CN	GO2	80
sc_Synthetic_Elec_CN_GO technology_GO3_yield_60	Synthetic	CN	GO3	60
sc_Synthetic_Elec_CN_GO technology_GO3_yield_70	Synthetic	CN	GO3	70
sc_Synthetic_Elec_CN_GO technology_GO3_yield_80	Synthetic	CN	GO3	80

	Graphite prod.	El. mix	GO tech.	GO yield
sc_Synthetic_Elec_IN_GO technology_GO1_yield_60	Synthetic	IN	GO1	60
sc_Synthetic_Elec_IN_GO technology_GO1_yield_70	Synthetic	IN	GO1	70
sc_Synthetic_Elec_IN_GO technology_GO1_yield_80	Synthetic	IN	GO1	80
sc_Synthetic_Elec_IN_GO technology_GO2_yield_60	Synthetic	IN	GO2	60
sc_Synthetic_Elec_IN_GO technology_GO2_yield_70	Synthetic	IN	GO2	70
sc_Synthetic_Elec_IN_GO technology_GO2_yield_80	Synthetic	IN	GO2	80
sc_Synthetic_Elec_IN_GO technology_GO3_yield_60	Synthetic	IN	GO3	60
sc_Synthetic_Elec_IN_GO technology_GO3_yield_70	Synthetic	IN	GO3	70
sc_Synthetic_Elec_IN_GO technology_GO3_yield_80	Synthetic	IN	GO3	80
sc_Natural_Elec_CA_GO technology_GO1_yield_60	Natural	CA	GO1	60
sc_Natural_Elec_CA_GO technology_GO1_yield_70	Natural	CA	GO1	70
sc_Natural_Elec_CA_GO technology_GO1_yield_80	Natural	CA	GO1	80
sc_Natural_Elec_CA_GO technology_GO2_yield_60	Natural	CA	GO2	60
sc_Natural_Elec_CA_GO technology_GO2_yield_70	Natural	CA	GO2	70
sc_Natural_Elec_CA_GO technology_GO2_yield_80	Natural	CA	GO2	80
sc_Natural_Elec_CA_GO technology_GO3_yield_60	Natural	CA	GO3	60
sc_Natural_Elec_CA_GO technology_GO3_yield_70	Natural	CA	GO3	70
sc_Natural_Elec_CA_GO technology_GO3_yield_80	Natural	CA	GO3	80
sc_Natural_Elec_ES_GO technology_GO1_yield_60	Natural	ES	GO1	60
sc_Natural_Elec_ES_GO technology_GO1_yield_70	Natural	ES	GO1	70
sc_Natural_Elec_ES_GO technology_GO1_yield_80	Natural	ES	GO1	80
sc_Natural_Elec_ES_GO technology_GO2_yield_60	Natural	ES	GO2	60
sc_Natural_Elec_ES_GO technology_GO2_yield_70	Natural	ES	GO2	70
sc_Natural_Elec_ES_GO technology_GO2_yield_80	Natural	ES	GO2	80
sc_Natural_Elec_ES_GO technology_GO3_yield_60	Natural	ES	GO3	60
sc_Natural_Elec_ES_GO technology_GO3_yield_70	Natural	ES	GO3	70
sc_Natural_Elec_ES_GO technology_GO3_yield_80	Natural	ES	GO3	80
sc_Natural_Elec_GB_GO technology_GO1_yield_60	Natural	GB	GO1	60
sc_Natural_Elec_GB_GO technology_GO1_yield_70	Natural	GB	GO1	70
sc_Natural_Elec_GB_GO technology_GO1_yield_80	Natural	GB	GO1	80
sc_Natural_Elec_GB_GO technology_GO2_yield_60	Natural	GB	GO2	60
sc_Natural_Elec_GB_GO technology_GO2_yield_70	Natural	GB	GO2	70
sc_Natural_Elec_GB_GO technology_GO2_yield_80	Natural	GB	GO2	80
sc_Natural_Elec_GB_GO technology_GO3_yield_60	Natural	GB	GO3	60
sc_Natural_Elec_GB_GO technology_GO3_yield_70	Natural	GB	GO3	70
sc_Natural_Elec_GB_GO technology_GO3_yield_80	Natural	GB	GO3	80
sc_Natural_Elec_IR_GO technology_GO1_yield_60	Natural	IR	GO1	60
sc_Natural_Elec_IR_GO technology_GO1_yield_70	Natural	IR	GO1	70
sc_Natural_Elec_IR_GO technology_GO1_yield_80	Natural	IR	GO1	80
sc_Natural_Elec_IR_GO technology_GO2_yield_60	Natural	IR	GO2	60
sc_Natural_Elec_IR_GO technology_GO2_yield_70	Natural	IR	GO2	70
sc_Natural_Elec_IR_GO technology_GO2_yield_80	Natural	IR	GO2	80
sc_Natural_Elec_IR_GO technology_GO3_yield_60	Natural	IR	GO3	60
sc_Natural_Elec_IR_GO technology_GO3_yield_70	Natural	IR	GO3	70
sc_Natural_Elec_IR_GO technology_GO3_yield_80	Natural	IR	GO3	80

	Graphite prod.	El. mix	GO tech.	GO yield
sc_Natural_Elec_CN_GO technology_GO1_yield_60	Natural	CN	GO1	60
sc_Natural_Elec_CN_GO technology_GO1_yield_70	Natural	CN	GO1	70
sc_Natural_Elec_CN_GO technology_GO1_yield_80	Natural	CN	GO1	80
sc_Natural_Elec_CN_GO technology_GO2_yield_60	Natural	CN	GO2	60
sc_Natural_Elec_CN_GO technology_GO2_yield_70	Natural	CN	GO2	70
sc_Natural_Elec_CN_GO technology_GO2_yield_80	Natural	CN	GO2	80
sc_Natural_Elec_CN_GO technology_GO3_yield_60	Natural	CN	GO3	60
sc_Natural_Elec_CN_GO technology_GO3_yield_70	Natural	CN	GO3	70
sc_Natural_Elec_CN_GO technology_GO3_yield_80	Natural	CN	GO3	80
sc_Natural_Elec_IN_GO technology_GO1_yield_60	Natural	IN	GO1	60
sc_Natural_Elec_IN_GO technology_GO1_yield_70	Natural	IN	GO1	70
sc_Natural_Elec_IN_GO technology_GO1_yield_80	Natural	IN	GO1	80
sc_Natural_Elec_IN_GO technology_GO2_yield_60	Natural	IN	GO2	60
sc_Natural_Elec_IN_GO technology_GO2_yield_70	Natural	IN	GO2	70
sc_Natural_Elec_IN_GO technology_GO2_yield_80	Natural	IN	GO2	80
sc_Natural_Elec_IN_GO technology_GO3_yield_60	Natural	IN	GO3	60
sc_Natural_Elec_IN_GO technology_GO3_yield_70	Natural	IN	GO3	70
sc_Natural_Elec_IN_GO technology_GO3_yield_80	Natural	IN	GO3	80

A.2.3 Visualization of the results

Python plot for figure 8:

```
fig = plt.figure()
ax = fig.gca(projection='3d')
surf=ax.plot_trisurf(df_el['Electricity_region'], df_el['Yield'], df_el['total_GWP'],
cmap=plt.cm.jet, linewidth=0.2)
#ax.view_init(10,100)
ax.set_zlabel("GWP, Kg CO2/kg graphene")
ax.set_xlabel("Intensity of electricity, Kg CO2/kWh")
ax.set_ylabel("Graphene yield, %")

fig.colorbar(surf, shrink=0.5, aspect=6)
ax.view_init(elev=15, azim=245)
plt.show()
```

Python plot for figure 9:

```
fig = go.Figure(data =
   go.Contour( x=el_unique33,  y=Y_unique33, z=T_elY,

       #category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]}
       colorscale = 'rdylgn',
       contours = dict(
       #coloring = 'heatmap',
       showlabels = True,
       labelfont = dict(
           size = 12,
           color = 'black',
       )
   )))
fig['layout']['xaxis'].update(title='Intensity of electricity, Kg CO2/kWh')
fig['layout']['yaxis'].update(title='Yield, % ')
fig.update_traces(colorbar_title_text = "GWP", selector = dict(type = "contour"))
fig.update_traces(reversescale = True, selector=dict(type='contour'))
fig.update_layout(title_text = 'GWP for 1 kg graphene (kg CO2 eq/kg GO)', title_x = 0.5)
#img_bytes = pio.to_image(fig, format = "svg")
fig.show()
```

Python plot for figure 11:

```
fig = px.line(df_M, x='Electricity_region', y='total_GWP', color='GO_technology',
  category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]})
fig.update_layout(title_text = 'Total GWP affected by different modification of the
Hummers process',
title_x = 0.5)
fig.update_traces(line=dict( width=3))

fig.show()
```

Python plot for figure 12:

```
fig = px.scatter(df_el, x = 'Electricity_region', y = 'total_GWP', color = 'GO_technology',
          #facet_col = 'Graphite_production_route',
          category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]})
fig.update_traces(mode='markers', marker_line_width=2, marker_size=8)
```

Python plot for figure 13:

```
boxplot = df0.boxplot(column=['total_GWP','GWP_Chemical_reduction', 'GWP_Graphite_oxide',
                             'GWP_graphite'], rot =45)


boxplot.set_ylabel('GWP (kg CO2 eq./kg graphene)')
boxplot
```

Python plot for figure 14:

```
boxplot = df0.boxplot(column=['total_GWP','GWP_Chemical_reduction', 'GWP_Graphite_oxide',
'GWP_graphite'], by=['Electricity_region'],  figsize=(8,6))

boxplot.set_ylabel('GWP (kg CO2 eq./kg graphene)')

boxplot
```

Python plot for figure 15:

```
fig = go.Figure(data =
   go.Contour( x=el_unique33,  y=Y_unique33, z=T_CR2,

       #category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]}
       colorscale = 'rdylgn',
       contours = dict(
       #coloring = 'heatmap',
       showlabels = True,
       labelfont = dict(
           size = 12,
           color = 'black',
       )
   )))
fig['layout']['xaxis'].update(title='Intensity of electricity, Kg CO2/kWh')
fig['layout']['yaxis'].update(title='Yield, % ')
fig.update_traces(colorbar_title_text = "GWP", selector = dict(type = "contour"))
fig.update_traces(reversescale = True, selector=dict(type='contour'))
fig.update_layout(title_text = 'GWP for the chemical reduction process (kg CO2 eq/kg GO)',
title_x = 0.5)
#img_bytes = pio.to_image(fig, format = "svg")
fig.show()
```

Python plot for figure 16:

```
fig = px.scatter(df_el_NOT_MOD, x = 'Electricity_region', y = 'GWP_Chemical_reduction',
 color = 'Yield',
          #facet_col = 'Graphite_production_route',
          category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]},
        # size = 'total_GWP'
         )
fig.update_traces(mode='markers', marker_line_width=2, marker_size=8)
```

Python plot for figure 17:

```
fig = px.line(df_M, x='Electricity_region', y='GWP_Graphite_oxide', color='GO_technology',
  category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]})

fig.update_traces(line=dict( width=3))

fig.show()
```

Python plot for figure 18:

```
fig = px.scatter(df_HUM, x = 'Electricity_region', y = 'GWP_Graphite_oxide',
           color = 'GO_technology',
           #facet_col = 'Graphite_production_route',
           category_orders={"Electricity_region": ["0.2", "0.4", "0.6", "0.8", "1.1", "1.4"]})
fig.update_traces(mode='markers', marker_line_width=2, marker_size=8)
```

Python plot for figure 19:

```
boxplot = df0.boxplot(column=['synthetic_GWP', 'natural_GWP'], rot =45)

boxplot.set_ylabel('GWP [kg CO2 eq./kg graphene]')

boxplot
```

A.3 MATLAB codes

```
%%Simulation, master 2021

 %Importing the input file and arranging them:
 file_input = readtable('input_graphene.xlsx')

 B = file_input(1:height(file_input),2:6)

 %Writing values in Arda template and running scenarios using ARDA Client:

 n = height(B); %n = number of rows in B
 i = 0; %starts here

 while i < 3
    i = i+1;

    xlswrite('Graphite_graphene_inventory.xlsx',B.GraphiteProduction(i),'Parameters Page','B9');
    xlswrite('Graphite_graphene_inventory.xlsx',B.ElectricityMix(i),'Parameters Page','G9');
    xlswrite('Graphite_graphene_inventory.xlsx',B.GOTechnology(i),'Parameters Page','H9');
    xlswrite('Graphite_graphene_inventory.xlsx',B.GOYield(i)/100,'Parameters Page','I9');
    xlswrite('parameters_ArdaCLI.xls',B.Scenario(i), 'Parameters','K3');
    try
        run('arda_subrun.m')
    catch
        continue
    end
 end
```

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Norwegian University of
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