

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Taking stock of large-scale lithium-ion battery production using life cycle assessment

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Abstract

Battery electric vehicles are being increasingly favored as an alternative to internal combustion engine vehicles (ICEVs). This is mainly due to their lower environmental impact when compared to ICEVs over the vehicle's lifetime. Life cycle assessment (LCA) studies focusing specifically on battery electric vehicles (BEVs) have identified battery cell production as an environmental hotspot in the BEV's life cycle. However, lack of primary or industrial data, different technical scopes, and varying data quality, limit a thorough understanding of the environmental impacts of cell production. Further, with scaling-up of battery production (to meet the rising demand for BEVs), the source and level of impacts are expected to change. In response, the main aim of this thesis is to explore and understand the implications of upscaling in battery production. An example of such a change is provided at the mining sites where raw materials for lithium used in batteries are extracted and produced. As mining continues, over time, the ore grades at these sites decline. Thus, this thesis also aims to investigate the effect of declining ore grades on the overall impacts from cell production. A sub-goal is to understand the relevance of background data in LCA studies and its effect on overall results.

The technical scope of this thesis is the production of a graphite-NMC:811 21700 type cylindrical cell. To assess the environmental impacts of upscaling, production in a small-scale facility is compared to production in a large-scale facility. Next, the impact of declining ore grades on overall cell production is estimated by analyzing the data from multiple mining sites for lithium, with varying ore grades and different types of sources – spodumene and brine. To assess the effect of background database on overall results, the LCA model for cell production was coupled with different versions of the Ecoinvent background database. Lastly, a physics-based model platform, developed in cross-disciplinary collaboration, is proposed with the objective of filling data gaps in LCA of lithium-ion batteries (LIBs). The model platform will help link the cell design aspects such as power or energy optimization to changes in the individual cell production processes. Further, the model platform will help expand the technical scope to broadened set of cell geometries and chemistries, and increase the precision in use phase modeling as well.

The results show that the upscaling leads to a reduction in environmental impacts from cell production. This is due to higher energy and material efficiency of cell production at large scale. Further, when low-carbon intensive sources are used, then the impacts from cell production shift almost entirely to the raw material extraction and production phase. In the context of declining ore grades, the type of source and grade of lithium account for 5-15% of the global warming impacts from cell production. This implies that future environmental impacts from LIB production could increase, due to increased chemical and energy inputs, in response to declining ore grades at mining sites. The changes in the background data have a significant bearing on the overall results. These are due to evolving technical systems and an improved representation of these systems in terms of data quality and geo-spatial representativeness. Lastly, preliminary results from the physics-based model platform show that accounting for variations in cell design can further add variability in results.

Prologue

As a researcher working in the field of electromobility, I am often asked if I would consider buying an electric car. I suspect such conversations relate not only to my tastes but also to my moral values, especially when the follow-up questions are about the socially exploitative mining conditions in Congo or the damage to the ecosystem in the South American salars due to brine extraction.

“Are electric vehicles *better* from an environmental point of view, as compared to fossil-based ones?” “Are they *sustainable*?”. “What happens if *everyone* drove an electric vehicle?”.

[Better...? Sustainable ...? Everyone ...?]

Seeking answers to such questions is the prime reason I was drawn to research the field of electromobility back in 2015. Obviously, as I now know, the answers to such questions are complex and have numerous caveats. In my personal view, they are always unsatisfactory. The questions are, however, very engaging. I hope that my deliberation will come to some fruition by the time I am done with my doctoral studies.

Today, at the mid-way point of my doctoral studies, the one thing I can say for certain is that the goalposts feel further away than they did when I started. *The more I learn, I realize, the less I know*. For now, I am only just scratching the surface of a complex problem that is not just an environmental challenge, but also has underpinnings in the geopolitics of resource extraction and energy (in)security. I am still *taking stock* of the field.

In this licentiate thesis, I attempt to articulate what my stock-taking has led to so far. This is primarily from an environmental perspective. My hope is that through this thesis and the rest of my doctoral studies, my work may contribute to efforts to reduce the spectrum of unknowns and uncertainties related to electromobility and its impacts.

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Paper 2: CHORDIA, M., WICKERTS, S., NORDELÖF, A. & ARVIDSSON, R. 2022. Life cycle environmental impacts of current and future battery-grade lithium supply from brine and spodumene. *Resources, Conservation and Recycling*, 187, 106634.

Paper 3: CHORDIA, M., WIKNER, E., & NORDELÖF, A. 2022. A model platform for solving lithium-ion battery cell data gaps in life cycle assessment. *Electric Vehicle Symposium 35*, Oslo, Norway.

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1. Introduction

The road transport sector is in the midst of a transition. The conventional fossil-fuel based internal combustion engine vehicles (ICEVs), that have been at the core of the private transport sector for almost a century, are now being outperformed by alternative powertrain vehicles on several metrics such as efficiency, environmental pollution, economics of ownership, and broader energy security issues (Poornesh et al., 2020, Liu et al., 2021, Azzuni and Breyer, 2018, Buekers et al., 2014). Alternative powertrain vehicles commonly in use today are flexible-fuel, gas, battery electric, hybrid, and fuel cell electric vehicles (Ghadikolaei et al., 2021). Amongst these, battery electric vehicles (BEVs) in particular have seen a steep surge in demand (IEA, 2021a). This is due to technical factors such as high efficiency and low maintenance, environmental factors such as improving urban air quality, reducing environmental and noise pollution, and economic factors such as lower cost of ownership and resilience from fluctuations in oil prices. Specifically for oil importing countries, BEVs also reduce reliance on imported oil, providing higher energy security to these countries (Greene et al., 2013).

The idea of using energy stored in batteries as a source to power vehicles is a century-old idea. Several prototypes and technologies have been tested over the years, although none were successful enough for commercialization to compete with ICEVs (Matulka, 2014). It was, however, the advent of the portable consumer electronics industry in the 1990s and the concurrent improvements in energy storage technology (EST) that provided the right impetus for the implementation of EST at a large scale in BEVs as well (Terada et al., 2001, Pilkington and Dyerson, 2006). Arguably, a landmark moment in the automotive industry was the launch of Tesla's first fully electric "Roadster" in 2008 which helped challenge the prevailing perception around BEVs as being economically infeasible and technically inferior. This helped present BEVs as a viable alternative to ICEVs (Bohnsack et al., 2014). Despite the initial apprehension about the practicality and competitiveness of the BEVs as compared to the ICEVs (Dijk and Yarime, 2010), the automotive industry has moved along quickly in the past decade or so and the stocks of BEVs are growing rapidly in many regions all over the world (Figure 1).

Today, the sale of BEVs globally is still dwarfed by that of ICEVs, i.e., 6 million vs 80 million in 2021, respectively (IEA, 2022, Placek, 2022). However several leading car makers have announced plans to gradually phase out ICEVs and go all-electric in the coming years (Campbell, 2021) and governments worldwide are making similar policy announcements (Wappelhorst, 2021). The evidence indicates that economies across the world are taking definite strides towards a highly (if not fully) electrified road transport sector.

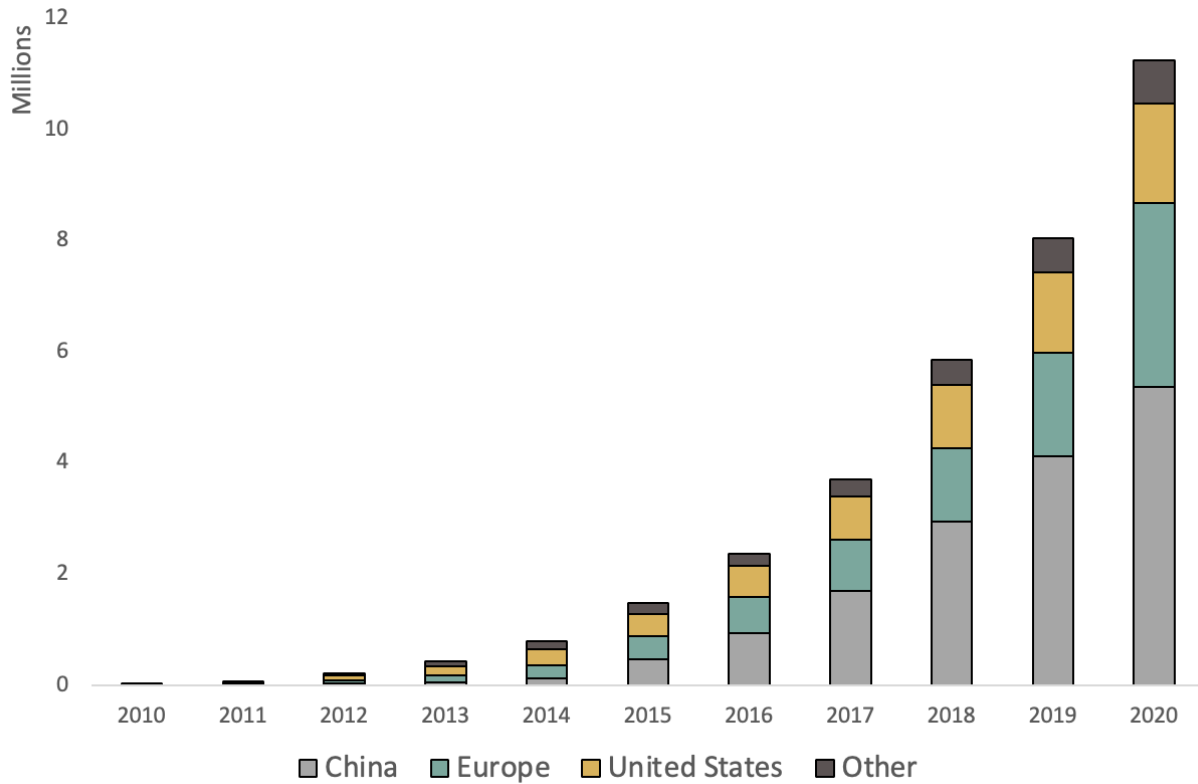


Figure 1: Global battery electric vehicle stock by region between 2010 and 2022 (IEA, 2021b)

Large-scale BEV adoption is being enabled by the upscaling of battery production facilities globally, especially for lithium-ion batteries (LIBs), which are the dominant technology for energy storage applications in BEVs (Blomgren, 2016, Ding et al., 2019). An indication of LIB production upscaling is seen in the increase in the average production capacity of LIB manufacturing plants, which has increased from 0.5 GWh to 7 GWh between 2015 and 2020 (Benchmark Mineral Intelligence, 2020). This is due to a combination of factors, such as, capacity expansion of existing production plants, installation of new large-scale production facilities, and maturing of the supply chains for key battery materials (Strauch, 2020). However, upscaling LIB production from lab or pilot scale to industrial scale faces challenges in product design, process scaling, facility optimization, raw material availability, and economics of demand and supply (Kwade et al., 2018, Keppeler et al., 2021, Wrålsen et al., 2021). Despite the challenges in upscaling, a key driver for BEVs is to reduce the environmental impacts of the transport sector. Therefore, understanding the environmental implications of large-scale BEV adoption becomes highly relevant.

For assessing the environmental impacts of producing and using BEVs, life cycle assessment (LCA) methodology is commonly used. LCA studies on BEVs point to LIB battery production as the main source of environmental impacts in the BEV life span (Nordelöf et al., 2014). However, LCA on battery production has pointed to a large variation in results (38-356 kg CO₂-eq./kWh) (Ellingsen et al., 2017). This can be explained by different technical scopes modeled in the various studies, and the lack of representative data for modeling battery and cell parameters such as lifetime, energy density, and energy demand in cell

production (Peters et al 2017; Peters and Weil 2018). Aichberger and Jungmeier (2020) point to small and underutilized facilities as reason for an overestimation of impacts from battery cell production. Similarly, high energy demand of throughput independent equipment (such as dry rooms) can result in high energy intensity in low throughput facilities (Dunn et al., 2015a). Further, production techniques and location of the facility could also play an important role in energy demand (Ellingsen et al., 2015). A number of studies rely on inventory data from the Greenhouse gases, Regulated Emissions, and Energy use in Transportation (GREET) database, partly due to its ease of availability (Dai and Kelly, 2019, Yuan et al., 2017, Wang et al., 2019, Deng et al., 2018, Kelly et al., 2019, Raugei and Winfield, 2019). Additionally, periodic updates to the GREET database make it suitable for investigating novel battery components (Wang et al., 2018, Dai et al., 2017, Dai et al., 2018, Dai and Winjobi, 2019). However, this also means that studies using GREET represent the same type of production facilities, technical products, and scopes, and rely on the same methodological assumptions. Peters et al. (2017) conducted a review of the LCA literature on LIBs and found that of the 79 studies reviewed, less than half provided sufficient data for extracting information about environmental impacts, and only seven relied exclusively on their own life cycle inventory (LCI) data for the processes investigated. This highlights the interdependency of studies in terms of LCI data. Peters et al. (2017)'s insight about the high interdependence amongst the studies and extensive use of the GREET database in modeling LIB production indicates a lack of representation of different production methods and technical scopes in LCA studies. Data from GREET specific to LIB production is now also implemented in the latest versions of the Ecoinvent database (Crenna et al., 2021), which is a commonly used database for modeling the background system in LCAs (Wernet et al., 2016, Steubing et al., 2016). This could lead to an increased dependency of LCA studies on limited data sources unless complemented with additional data sources.

Further, the interaction of battery chemistry and the cell performance is largely unexplored in LCA (Ambrose and Kendall, 2016, Aichberger and Jungmeier, 2020). Often LCA studies comparing different chemistries assume the same lifetime, whereas battery cycle life tests indicate that there could be large variations (Burke, 2014), which can affect the lessons drawn from the studies (Ambrose and Kendall, 2016). Similar conclusions are also drawn by Eddahech et al. (2015), who found a variation of more than 50% in battery degradation rates in various LIB chemistries based on the thermal and charge conditions during storage.

A number of LIB chemistries have been commercialized by the automakers for application in BEVs. The choice of chemistry depends upon several factors such the intended application, state of the current manufacturing technology, economics of production, and an assured supply of raw materials (Zhao et al., 2022, Jones et al., 2020). Cell chemistry generally refers to the materials used in the active material of the cell (both cathode and anode). Amongst the cathode materials commonly used in BEV applications, LiNiMnCoO_2 (lithium nickel manganese cobalt oxide) also referred to as NMC, is the most common today (Tsiropoulos et al., 2018). NMC chemistries are denoted by the share of nickel-manganese-cobalt present in the chemistry. For example, NMC-1:1:1 refers to an equal share of the respective metals whereas NMC-8:1:1 refers to high-nickel content chemistry. Other NMC chemistries in use in the industry include 5:3:2 and 6:2:2. High-nickel content chemistries (such as 6:2:2 and 8:1:1) are likely candidates for higher adoption in the transportation sector due to their advantages over other cell chemistries in terms of energy capacity,

energy density, power capability and cost (Manthiram, 2017). Higher nickel content in NMC implies a lower share of cobalt per unit mass of the active cathode material. Lower cobalt share in LIB is also favored, as nearly 70% of all the cobalt produced globally comes from the Democratic Republic of Congo (USGS, 2020). Such a high concentration of supply in a single region exposes cobalt to price fluctuations (Wentker et al., 2019). Additionally, mining in Congo is often highlighted for its perverse practices (Sharma and Manthiram, 2020), that adversely affect human health (Arvidsson et al., 2022). Thus, there are technical, economic, and social reasons for adopting high nickel content LIB chemistries.

Apart from nickel, manganese and cobalt, lithium is also an important constituent in the LiNiMnCoO_2 chemistry, which offers thermal stability and high capacity (Zhao et al., 2017). Lithium is also present in the electrolyte in the form of lithium hexafluorophosphate and is an active constituent in several upcoming battery chemistries as well (Edström et al., 2020). Some researchers have projected lithium demand to increase 20-fold by 2050 (Xu et al., 2020). The aspect of raw material supply, especially for lithium, is crucial for the current BEV market where carmakers have increasingly sought strategic tie-ups with mining companies and in some cases bought them out to ensure long term supply of raw materials for use in batteries (USGS, 2021). Thus, lithium is likely to play a critical role in the global battery supply chain in the upcoming years (Greim et al., 2020).

Lithium is commonly extracted from brine aquifers and mineral ores, typically spodumene (Wietelmann and Steinbild, 2014). Brine-based sources are primarily extracted in the salars located in South America, in a region colloquially known as the “lithium triangle” (López Steinmetz and Salvi, 2021). Spodumene-based sources are primarily extracted in Australia and China, although spodumene deposits are available in several other parts of the world (USGS, 2021). Lithium mining operations in the salars have been criticized for their socio-environmental impacts (Giglio, 2021) and damage to the ecosystem (Flexer et al., 2018) often connected to water use and pollution (Garcés and Álvarez, 2020). Several LCA studies have assessed the water use impacts in relation to lithium production from brine (Kelly et al., 2021, Schomberg et al., 2021, Stamp et al., 2012). However, brine is a part of a complex hydrological cycle occurring in the salars (Houston, 2006, Houston et al., 2011) and the current LCA methods do not address the complexity completely. This issue is further exacerbated by the fact that, as brine extraction operations continue over the years, the average brine grade at the mining sites will decline. Thus, requiring and increasing input of energy and chemicals for processing of lithium compounds (Calvo et al., 2016). Hence, decreasing ore grade correlates with increasing energy, chemical and water inputs and the associated environmental costs (Magdalena et al., 2021). This is also applicable to spodumene-based lithium production.

2. Research questions

Environmental impact of LIB production is an increasingly researched topic. However, a number of broader questions remain unanswered. This thesis answers specific research questions in an attempt to further understandings of large-scale LIB production. My hope is that this specific contribution may help build the evidence base to help tackle these broader questions. The following are the main research questions of this licentiate research:

1. What are the environmental implications of upscaling lithium-ion battery production?
2. How do the type of source and grade of lithium affect the environmental impacts of producing battery grade lithium hydroxide monohydrate?
3. How can differences in lithium-ion battery cell design be captured in life cycle assessment?

2.1. Scope

The LIB type studied in this research is a graphite-NMC:811 21700 cell. For specific details on the cell composition and design, the readers are referred to section 2.3 of Paper 1. The research focusses on the implications of large-scale cell production. Hence, all the relevant flows from cradle-to-gate, i.e., raw material extraction, and processing, and cell production in a factory are analyzed. Further, lithium input in the NMC chemistry is in the form of battery-grade lithium hydroxide monohydrate ($\text{LiOH}\cdot\text{H}_2\text{O}$). For this reason, $\text{LiOH}\cdot\text{H}_2\text{O}$ production from two types of sources are assessed, i.e., brine and spodumene, with different starting ore grades. For details on the technical process design of the various supply routes to produce $\text{LiOH}\cdot\text{H}_2\text{O}$, refer to section S2 and section S3 of Paper 2.

2.2. Structure of the thesis

This thesis comprises of six chapters and three appended papers at the end. Chapter 1 of this thesis provided a background to the selected research questions. This chapter, i.e., Chapter 2, states the research questions the thesis aims to answer. The following Chapter 3 describes the methods used in the thesis. Next, Chapter 4 provides the results including a synthesis of the main findings from the appended papers. Chapter 5 presents the main conclusions and limitations of the thesis along with an overview of possible directions for future research. Chapter 6 provides the list of references used in this thesis.

3. Methods

Environmental systems analysis (ESA) tools include environmental impact assessment, risk assessment, material and substance flow analysis, input-output analysis and LCA. These ESA tools are used to assess the impacts of the technosphere on the environment (Baumann and Tillman, 2004, Finnveden and Moberg, 2005). However, in this licentiate research, LCA methodology has been used in all the appended articles. The LCA methodology is describe briefly in the following section along with a summary of the key aspects of the methodology used in the appended papers. For the specific details regarding the scope (technical and temporal), functional unit, scenarios, and impact assessment methods, readers are referred directly to the methods section of the appended papers provided at the end of the licentiate thesis. Further, the licentiate research and the appended articles required the use of additional methods to address the research questions. These additional methods are described in this section as well.

3.1. Life cycle assessment

LCA methodology is applied to assess the impacts from the life cycle of a product or a service by quantifying the environmentally relevant flows between the technosphere and the environment (Baumann and Tillman, 2004, Hellweg and Milà i Canals, 2014). As per the LCA framework defined in ISO 14040 and ISO 14044, the key phases of an LCA study include the *goal and scope definition*, *inventory analysis*, *impact assessment*, and *interpretation*. The goal and scope definition phase typically defines the application and type of LCA, life cycle stages to include, and other technical system boundary, geographical and temporal scope, and the functional unit(s). The inventory analysis quantifies the inflows and outflows from the product system. The impact assessment phase translates these inventory flows into environmental impacts. Finally, in interpretation phase, conclusions are drawn, and recommendations provided.

3.1.1. Type of LCA

LCA studies are usually classified as either attributional or of the consequential type, and the choice of LCA type depends on the stated goals of the study (Yang, 2019). Attributional LCAs estimate the share of global environmental burden that can be linked to the life cycle of a product or a service. Consequential LCAs on the other hand, estimate how the environmental burdens change due the production or use of a certain product or service (Ekvall, 2019). Essentially, consequential LCA are used when responding to a change in the demand for the function of the product or service being assessed. In this thesis, attributional LCA was used for several reasons. Firstly, Paper 1 compares the production of LIBs at small and large scale to understand the implications of upscaling. For this LIB production and the associated environmental impacts were compared in two separate production systems for which attributional LCAs are better suited. However, if the goal of Paper 1 was to assess the impacts of repurposing a small-scale factory to a large-scale factory, a consequential LCA would have been better suited. Using attributional LCA also has the advantage of being more readily comparable with a number of studies in the field which tend to favor attributional type of assessments due to fewer uncertainties in data type and higher data availability compared to consequential type assessments. Further, the aim of Paper 2 is to assess the impacts of lithium

supply based on the source and the grade of lithium. For this, multiple lithium supply routes are compared to assess the environmental impacts associated with their supply chain for the production of $\text{LiOH}\cdot\text{H}_2\text{O}$. As the modeled supply routes represent multiple production systems, an attributional type of assessment is used. However, if the purpose of the study was to assess how the impacts from a particular supply route changes due to declining ore grades, a consequential assessment could have been used. Lastly, in Paper 3, a model platform is being developed to fill data gaps in LCA studies to cover multiple cell geometries and chemistries and to assess their environmental performance in the life cycle. Once again, attributional assessment is better suited for this, as the study aims to compare a wide range of cells and not the effect of using one type of cell over another.

3.1.2. System boundaries and the functional unit

An important aspect of LCA studies is a clear specification of the *technical system boundary*. This system boundary defines technical system considered relevant based on the goals of the study. So far, the licentiate research focusses on the aspect of cell production. Hence, the system boundary covers all the technical activities, from raw material extraction and production, up to cell production. Such a system boundary is also referred to as cradle-to-gate, where the gate refers to the factory gate before the product (LIB cell) is shipped to the customer.

The system under investigation in an LCA can also be divided into a *foreground and a background system*. The foreground system within the system boundary represents the part of the technical system for which (new) data is collected by the LCA practitioner. The data for the foreground system can be collected from multiple sources such as product, material or process specific databases, literature survey, technical reports, environmental permit applications, patents, expert judgement, stakeholder consultations, process modeling and simulation, engineering calculations etc. In Paper 1, foreground data was collected for modeling the large-scale LIB production from the technical reports and environmental permits published by Northvolt AB. Readers are referred to section S1.1 of Paper 1 for a detailed description of the data collection process. In Paper 2, the foreground data is collected from multiple technical reports and environmental permits of lithium mining and processing facilities. Readers are referred to section S1 of Paper 2 for detailed information on the data sources. The foreground data for Paper 3 is based on engineering calculations, cell teardown, cell performance modeling and simulations as well as expert judgement.

The data for the background system is typically based on databases such as Ecoinvent database (Steubing et al., 2016, Wernet et al., 2016). The Ecoinvent database is a global database and supports regionalized impact assessment. It was first released in 2003 and since then has been continuously updated. The background system in Paper 1 was modeled using Ecoinvent v2.2 and v3.7.1. The older version (v2.2) was used to remodel the small-scale factory as it was applied in the original study by Ellingsen et al. (2014). The background system in Paper 2 and Paper 3 is modeled using Ecoinvent v3.8.

The *geographical scope* of an LCA study specifies the geographical boundary of the technical system represented in the study. For detailed information on the geographical scopes assessed, the readers are referred to the methods section of the appended papers.

The *temporal scope* specifies the time horizon of the assessment. In Paper 1, the small-scale factory representing production in 2010 is compared to a large-scale factory using presently latest available production technology and methods, i.e., valid for 2022. Thus, the study, has both, a retrospective, and a current temporal scope. In Paper 2, data from upcoming lithium mining facilities is used to model the effect of ore grades on the overall environmental impacts. As these facilities are expected to become fully operational in the future, i.e., in the next five years, the temporal scope of the study is the near future (prospective). As the ore grades at mining sites will decline over time, the studies would have to be reassessed in the future to see if they are still representative of the mining processes at the various sites.

Every product or a service in an LCA is defined based on the purpose, or the function it serves. Thus, a *functional unit* is the quantified description of the function that the system is expected to provide, and the reference based on which all the impacts are calculated. In Paper 1, the functional unit was selected as 1 kWh of cell storage capacity for comparing LIB production in small-scale and the large-scale production facility. The main function of a cell is to store energy and expend it when needed (discharge cycle in the use phase). As the scope of the study was limited to cell production, the functional unit of 1 kWh of theoretical storage capacity was deemed suitable. However, if the scope of the study included the use phase, then a more suitable functional unit will be 1 person-kilometer driven. In Paper 2, the functional unit of 1 ton LiOH·H₂O is selected as the study aims to compare LiOH·H₂O production based on different supply routes. Further, when the LiOH·H₂O production is assessed in the context of LIB production in that study, a functional unit of 1 kWh cell storage capacity is chosen. The key aspects of LCA methodology applied in the appended papers are summarized in Table 1.

Table 1: Summary of the key aspects of LCA methodology applied in the appended papers

Paper	Object of study	Functional unit	Scope	Temporal scope*	Scale
1	NMC-111 pouch cell	1-kWh storage capacity	Cradle to factory out-gate	Retrospective and current	Small
	NMC-811 21700 cell				Large
2	LiOH·H ₂ O from brine and spodumene	1-ton battery grade LiOH·H ₂ O	Cradle to factory in-gate	Current and prospective	Large
	NMC-811 21700 cell	1 kWh storage capacity	Cradle to factory out-gate		
3	NMC- 811 21700 cell (energy and power optimized)	1 kWh storage capacity	Cradle to factory out-gate	Current	Large

*The temporal scope in this thesis is defined with 2022 as the reference year.

3.2. Impact assessment

The licentiate research focuses foremost on the climate change (global warming) impacts due to their global relevance and since BEVs (and LIBs) are primarily being promoted to reduce greenhouse gas emissions (Kelly et al., 2021). Further mineral resource scarcity is considered highly relevant in the context of LIB production due to the use of a number of rare elements (Nordelöf et al., 2014). Additionally, water use, and water pollution issues are identified as problematic in the context of brine extraction in the salars (Giglio, 2021, Flexer et al., 2018). For this reason, attention is paid to water use impacts as well.

Impact assessment in this licentiate research is mainly carried out using the ReCiPe midpoint method with the hierarchist perspective (Huijbregts et al., 2016). The ReCiPe package was used as it provides characterization factors covering global scale for a wide range of environmental impact categories such as particulate matter formation, photochemical ozone formation, ionizing radiation, stratospheric ozone depletion, human toxicity (carcinogenic and non-carcinogenic), climate change, water use, ecotoxicity, freshwater eutrophication, terrestrial acidification, land use, mineral resource scarcity and fossil resource scarcity (Huijbregts et al., 2016). The ReCiPe is also amongst the most commonly used LCIA methods used in LCA studies (iPoint Systems, 2018). The study by Ellingsen et al. (2014) on small-scale LIB production applied the ReCiPe package for impact assessment. Hence, for comparability reasons the ReCiPe package was selected for this licentiate research. Some additional LCIA methods were also applied to complement ReCiPe and gain further perspectives from the assessment of the technical systems. These involved the assessment of resource use impacts, water use impacts, as well as freshwater ecotoxicity impacts. These are further discussed further in following sub-sections.

3.2.1. Resource use impacts

In the ReCiPe package, resource scarcity impacts are assessed using the surplus ore potential (SOP) method. The SOP method relies on ore grades and commodity prices to characterize the minerals and metals. This implies that the results for the same resource type can vary over time provided the characterization factors (CFs) are updated regularly. Thus, the SOP method provides a more near term (or a short term) perspective and is relevant only as long as the CFs are updated to reflect the existing market. To supplement this near-term perspective with a long-term resource scarcity perspective, the crustal scarcity indicator (CSI) (Arvidsson et al., 2020) was applied. The CSI method characterizes the resources based on their availability in the earth's crust and not from other environmental compartments like the oceans or the atmosphere. Thus, the CSI method is agnostic of any supply and demand dynamics pertaining to extraction rates and commodity prices. However, neither the CSI nor the SOP method are regional resource scarcity indicators but consider the global scale. The CSI method was applied in Paper 1 and Paper 2 to complement the SOP method.

3.2.2. Water use impacts

In the ReCiPe package, despite being referred to as the midpoint indicator, water use impacts are reported at the inventory level (Arvidsson, 2021). The salar region already faces water scarcity and as brine operations require high volumes of freshwater input (Garcés and Álvarez, 2020, Bustos-Gallardo et al., 2021), water use impacts were further assessed using the Available Water Remaining (AWARE) method. The AWARE method is a regional indicator that quantifies the available water remaining per area once the demand of the humans and ecosystem have been met (Boulay et al., 2018). This method is also recommended by the United Nations Environment Programme and the Society for Environmental Toxicology and Chemistry (Frischknecht et al., 2016).

3.2.3. Freshwater ecotoxicity impacts

The freshwater ecotoxicity impacts are connected to the research objectives of Paper 2 as freshwater availability is connected to water pollution. The USEtox method provides characterization factors for freshwater ecotoxicity (as well as human toxicity), is continuously updated and is the result of a consensus process led by the United Nations Environmental Programme and the Society for Environmental Toxicology and Chemistry (Rosenbaum et al., 2008, Hauschild et al., 2008). For this reason, the USEtox method (model v2) was applied in Paper 2.

3.3. Remodeling of existing studies

The database used to model the background system in LCA studies are regularly updated to fill data gaps and replace old or outdated data. The new data added to the background databases are more representative of the technical system and to an extent can also highlight changes in the technical system over time, such as market averages and electricity supply mixes. Hence, when comparing LCA studies it is essential to understand what the data in the background system represents. For this licentiate research, three LCA studies were identified as relevant based on the selected technical systems and remodeled. This helped quantify changes in the background databases in terms of environmental impacts. Although the specific reason for remodeling the studies differed in each case, the broad aim of remodeling studies was to understand the influence that background databases can have on LCA results.

3.3.1. Ellingsen et al. (2014)

The study by Ellingsen et al. (2014) study was replicated and remodeled as a part of Paper 1. Ellingsen et al. (2014) collected data for a LIB production facility operating at small-scale, i.e., approximately 70 MWh of annual battery capacity production, and modeled it using Ecoinvent v2.2. To develop a better understanding of how impacts from battery production have changed over time and to establish a benchmark for comparing the small-scale factory to the giga-factory, the original Ellingsen et al. (2014) study was replicated using Ecoinvent v2.2 and then remodeled using the latest version of Ecoinvent available at the time of the study, i.e. v3.7.1. The replicated model showed less than 0.2% difference when

compared to the results published in Ellingsen et al. (2014). This small difference was deemed to be low and attributed to truncation and hence validated the replicated model. The same study was then remodeled to benchmark against the giga-factory model, also modeled using Ecoinvent v3.7.1.

3.3.2. Kelly et al. (2021)

Kelly et al. (2021) modeled their inventory using the GREET database. However, the background system in Paper 2 was modeled using Ecoinvent v3.8. Thus, for maintaining consistency amongst the datasets reviewed, Kelly et al. (2021) was remodeled using Ecoinvent v3.8.

For the brine-based route, Kelly et al. (2021) modeled the production of LiOH·H₂O using four allocation methods (facility level-mass, facility level-economic, product level and process level) and presented the results based on each of the allocation methods used. However, for the purposes of this research, only facility-level economic allocation was used for modeling the brine-based production route. The results of remodeling using Ecoinvent v3.8 matched for the brine-based route, whereas for the spodumene based route, the results of modeling with Ecoinvent v3.8 resulted in 18% higher values than the values published by Kelly et al. (2021) for global warming impacts, linked to the modeling of chemical inputs and differences between the different database versions.

3.3.3. Engels et al. (2022)

Graphite is the anode material of the studied NMC-8:1:1 cell. The study by Engels et al. (2022) provides industry data on natural graphite production, which complements the synthetic graphite production data available in Ecoinvent v3.8, originally based on the report published by Dunn et al. (2015b). The background system in the Engels et al. (2022) study was modeled using the Gabi database (v10.0 SP40). However for this licentiate research, Engels et al. (2022) was remodeled with the Ecoinvent v3.8 database linked to the inventory data published in that study. This remodeling was done for two reasons. Firstly, to have a common background database throughout the study, and secondly to complement the natural graphite data with synthetic graphite data for the production of the anode. To increase the life-time of the battery cells, a combination of synthetic and natural graphite in a predetermined ratio (85:15) could be used in cells (personal communication, Northvolt, 2022). The results from the remodeling of the Engels et al. (2022) study are presented in section 4.4 of this licentiate thesis, but not in the results reported in the appended papers. The results from the original Engels et al. (2022) study and the remodeling by Ecoinvent v3.8 differed by 34% for global warming impacts, which is largely attributed to differences between the different database versions.

3.4. Cell parameterization and performance modeling

This section is applicable to the research objectives of Paper 3 and is a part of an ongoing study aimed at developing a model platform to fill data gaps in LCA of LIBs. Refer to the "Methods" section of the appended Paper 3 for a detailed description of the methods used. Figure 2 shows the flowchart of the model platform

highlighting the cell design and its linkage to the LCA model. In the figure, the black boxes indicate the calculation steps from the software, and the blue boxes indicate the intermediate inputs and outputs between the calculation steps. The methodology includes the development of a model platform to estimate a cell design based on a manufacturer's tabulated data or cell teardown data. The design is made flexible such that based on a few user-specified inputs, a representative cell model for an application, can be established. The output from this cell design platform is fed to the cell performance model such that information about the energy losses from cell resistance during charging and discharging, as well as the cell energy content can be determined. The output from the cell design platform is also fed to the LCA inventory model for cell production described in the next paragraph. The output from the cell performance model is then sent to the battery pack and the vehicle powertrain model, which calculates the energy use in the operational phase of a generic EV operating on a specific drive cycle. The battery pack and the powertrain model also account for both the cell's share of the total vehicle mass and losses caused by this mass in the calculation of the total energy losses. This information is fed to the LCA model to account for the impacts from the use phase of the cell.

The input to the LCA inventory model from the cell design model is used to develop parameterized LCA inventory models covering a wide range of cell formats, chemistries, and performance applications. This parameterization is done based on a linear extrapolation of inventory flows of a cell whose design and manufacturing details are well established. This extrapolation is based on the pre-determined relationship between inventory flows and specific cell properties. Further, the parameterization model is validated against newly acquired cell design and manufacturing data.

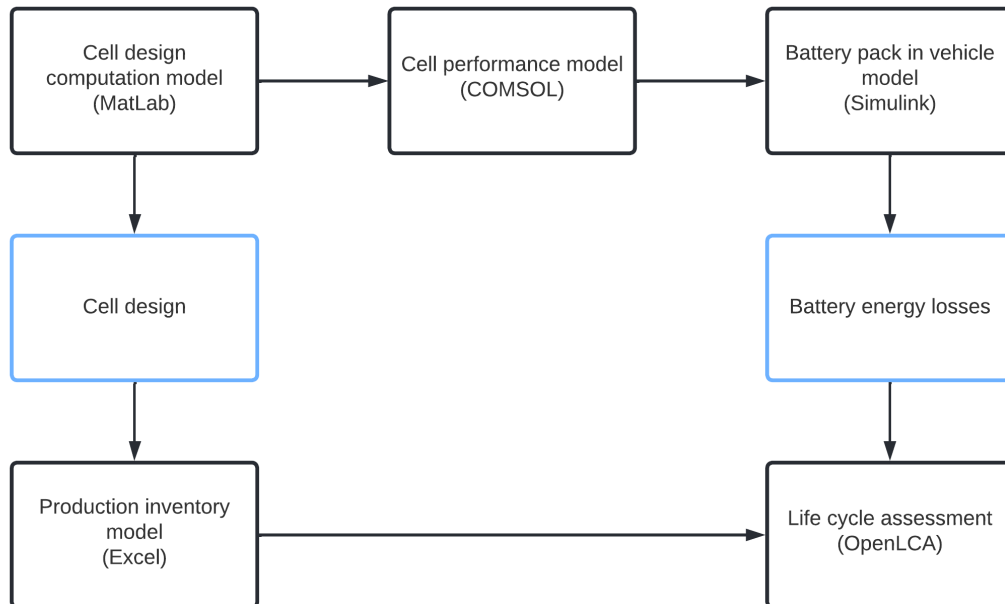


Figure 2: Overview of the model platform from cell design parameters to LCA, showing the main steps and software.

4. Results

This section presents a summary and the main findings of papers appended in the licentiate thesis. The section also includes a synthesis of the main findings in the appended papers to present an updated estimate of large-scale LIB production.

4.1. Paper 1: Summary of results and learning outcomes

The main research objective of Paper 1 was to assess the environmental implications of upscaling LIB production, i.e., to quantify the scale and type of environmental impacts when production volumes increase from small to large scale. LIB production in a small-scale factory was modeled based on the inventory data provided in Ellingsen et al. (2014), which represented a factory output of about 70 MWh cell production capacity annually. Ellingsen et al. (2014) used Ecoinvent v2.2 for modeling the background system in their study. LIB production in the large-scale factory was modeled based on the environmental and technical permit applications published by Northvolt AB, which is an upcoming battery manufacturer based in Sweden. The large-scale production represented about 16 GWh of cell production capacity annually. Specific details on the data collection process and the permit applications published by Northvolt are provided in section S1.1 of the supporting information document of Paper 1. The large-scale factory was modeled using the latest available version of Ecoinvent, i.e., v3.7.1, at the time of modeling. Further, the small-scale factory was remodeled using Ecoinvent v3.7.1 for establishing a baseline for comparing the results with the large-scale factory.

4.1.1. Background database in LCA

Global warming impacts for the small-scale factory modeled with Ecoinvent v2.2 and v3.7.1 are shown in Figure 3. The impacts are higher by nearly 30% when the small-scale factory is modeled using Ecoinvent v3.7.1 (Small-3.7), as compared modeling with Ecoinvent v2.2 (Small-2.2). This increase is due to a combination of improvements in data quality between the two Ecoinvent background database versions and changes in the technical systems over time. Specific to data quality, the data representing cobalt sulfate (CoSO_4) production improved significantly between the two Ecoinvent database versions. In the Ecoinvent v2.2, cobalt production was approximated with data from nickel mining and beneficiation. In particular for this version, the reduction process which is used to increase the metal concentration during nickel processing was modeled using stoichiometric calculations with an assumed yield of 95%. Additionally approximations were used to model energy consumption based on data from other chemical plants (Hischier, 2007). However, in Ecoinvent v3.7.1, cobalt hydroxide production is modeled using industry data representing 30% of the global production of refined cobalt (CDI, 2016). To model cobalt sulfate, data from a Canadian refinery was used that produced battery-grade cobalt sulfate with cobalt hydroxide as input (Ausenco, 2020).

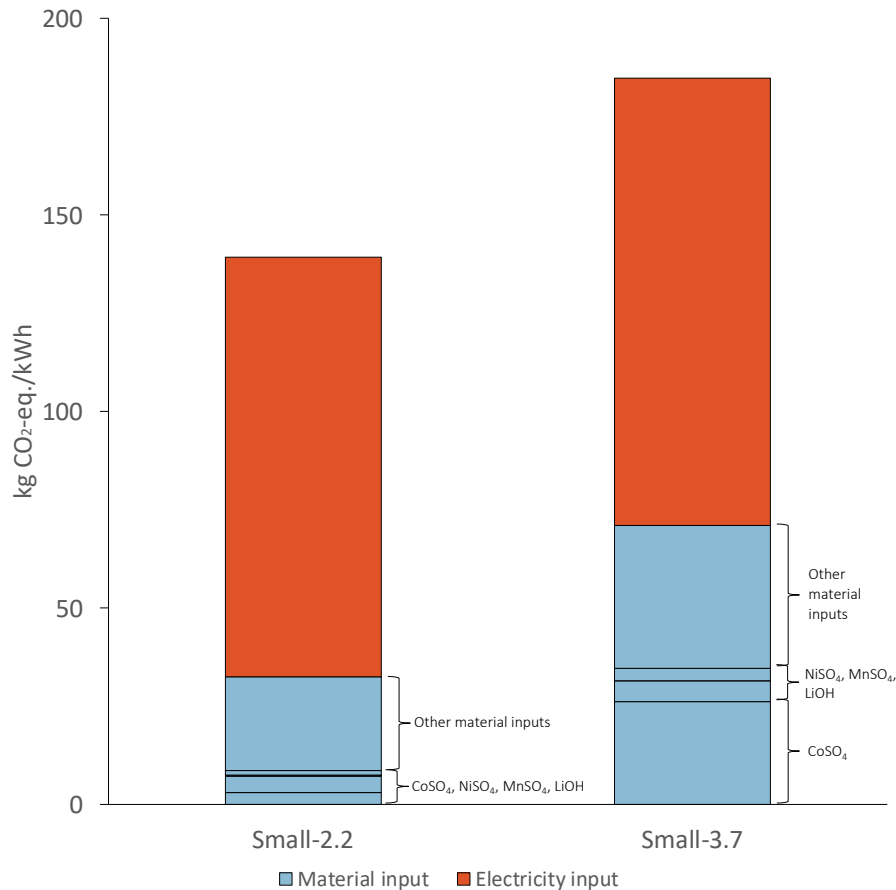


Figure 3: Comparison of global warming impacts for the small-scale factory modeled using different Ecoinvent versions (v2.2 for Small-2.2 and v3.7.1 for Small-3.7)

In terms of changes in the technical system, both the share of recycled copper – which is an input for making foils for current collectors – and the share of the various energy sources in the South Korean electricity mix changed between the two database versions. Thus, the increase in the global warming impacts for the small-scale factory model from changing the background database version, is due to both improvements in data quality and changes in the technical system. This highlights the importance of underlying data quality and the background technical systems when comparing past studies to more recent ones. Such an insight is valuable for LCA practitioners and points to the need to revisit the modeling data and context of past studies when drawing comparisons to more recent ones.

4.1.2. Energy demand in upscaling

Table 2 provides a summary of LCA studies on large-scale LIB production with details on the annual throughput (GWh/annum), cathode chemistries and the primary source of data. Only studies reporting NMC-based cathode chemistries are included in the table. The Ellingsen et al. (2014) study presented data for LIB production in a small-scale factory and is mentioned in the table for context when comparing to large-scale cell production facilities.

Ellingsen et al. (2014) collected monthly electricity use data in the factory over a span of 18 months. Due to the large variation in the data collected, Ellingsen et al. (2014) used three data points in their assessment for energy demand in cell production – a lower-bound value, an asymptotic value and an average value. However, the lower-bound value was deemed to represent the most energy-efficient months which in principle would also reflect an energy optimized large-scale production facility. Thus, the reported lower-bound value of 586 MJ/kWh (163 kWh/kWh) is used for comparison with the remaining large scale production facilities. Note that since electricity is the only source of energy used in the factory (Ellingsen et al., 2014), all heat and cooling requirements are met by equipment operating on electricity alone.

Large-scale LIB production facilities (2 GWh/annum) such as the one reported by Dai et al. (2019) state total electricity requirement of 8.3 kWh/kWh and 140 MJ/kWh of steam. Dai et al. (2019) collected data from a leading LIB manufacturer in China in 2017 and state that electricity is used to power eleven dehumidifiers, and four industrial chillers, while steam is used for dehumidification and drying. The electricity requirement for the remaining process equipment is considered to be negligible and are not reported. The facility reported in Paper 1 (Chordia et al., 2021) has an annual LIB cell production capacity of 16 GWh and reports demand for electricity (68 kWh/kWh), heat (27 MJ/kWh) and cooling (108 MJ/kWh). It is worth noting that the facility reported in Paper 1 was still under construction when the study was conducted. The energy and material input, as well as emissions data reported, were based on technical reports and permit applications. Data in such permit applications typically represents an upper limit for a facility and in the real-world, once the facility is operating and the energy efficiency measures are in place the energy input per produced cell will likely reduce. Whereas, the data reported in Dai et al. (2019) is from a production facility that is already operational. Hence, it is unlikely that the energy requirement in the facility will change much in the future. The technical system boundary of the facility described in Paper 1 and by Dai et al. (2019) also differ slightly. The input to the LIB production facility described in Paper 1 are the sulfate solutions of nickel (Ni), manganese (Mn), and cobalt (Co) which are then mixed, processed, and heat treated to produce NiMnCo oxide powder (also called the NMC precursor). Pre-treated LiOH·H₂O is mixed with the NMC precursor to produce LiNiMnCoO₂ (active cathode material) in a calcination furnace. In the facility described by Dai et al. (2019), the NMC precursor and treated LiOH·H₂O are the main inputs and the processing starts directly at the mixing step followed by calcination to produce LiNiMnCoO₂.

Sun et al. (2020) present data for a 30 GWh/annum LIB production facility stating a requirement of 20 kWh/kWh for electricity and 34 MJ/kWh for steam. Sun et al. (2020) collected onsite data from multiple Chinese manufacturers. In their scope, Sun et al. (2020) include the production of the NMC precursor. Per Sun et al. (2020), operating the dry room and the electrode vacuum drying process accounts for over 75% of the electricity requirement and 90% of the heat requirement from steam.

Jinasena et al. (2021) developed a theoretical process-based model to represent industrial-scale LIB production of 2 GWh/annum. Jinasena et al. (2021) state a net energy requirement of 45 kWh/kWh with drying process and the dry rooms accounting for over 95% of the energy requirement. Jinasena et al. (2021) take the mixing step as the starting point of their calculations, where the active cathode material is mixed with solvents and binders to form a slurry. Thus, the system boundary for cell production has a starting

point further downstream than the one presented in Dai et al. (2019). Degen and Schütte (2022) report theoretical data of a LIB production facility with an output of 7 GWh/annum. The engineering data is acquired from an upcoming advanced research facility for battery production in Germany, called the Fraunhofer Research Institution for Battery Cell Production (FFB). The facility considered is not operational yet and the data presented is based on engineering calculations and expert judgement. The system boundary for the LIB production facility is the same as that of Jinasena et al. (2021). See Figure 4 for a further elaboration of the differences in the foreground system boundary (factory) represented in the studies presented in this section.

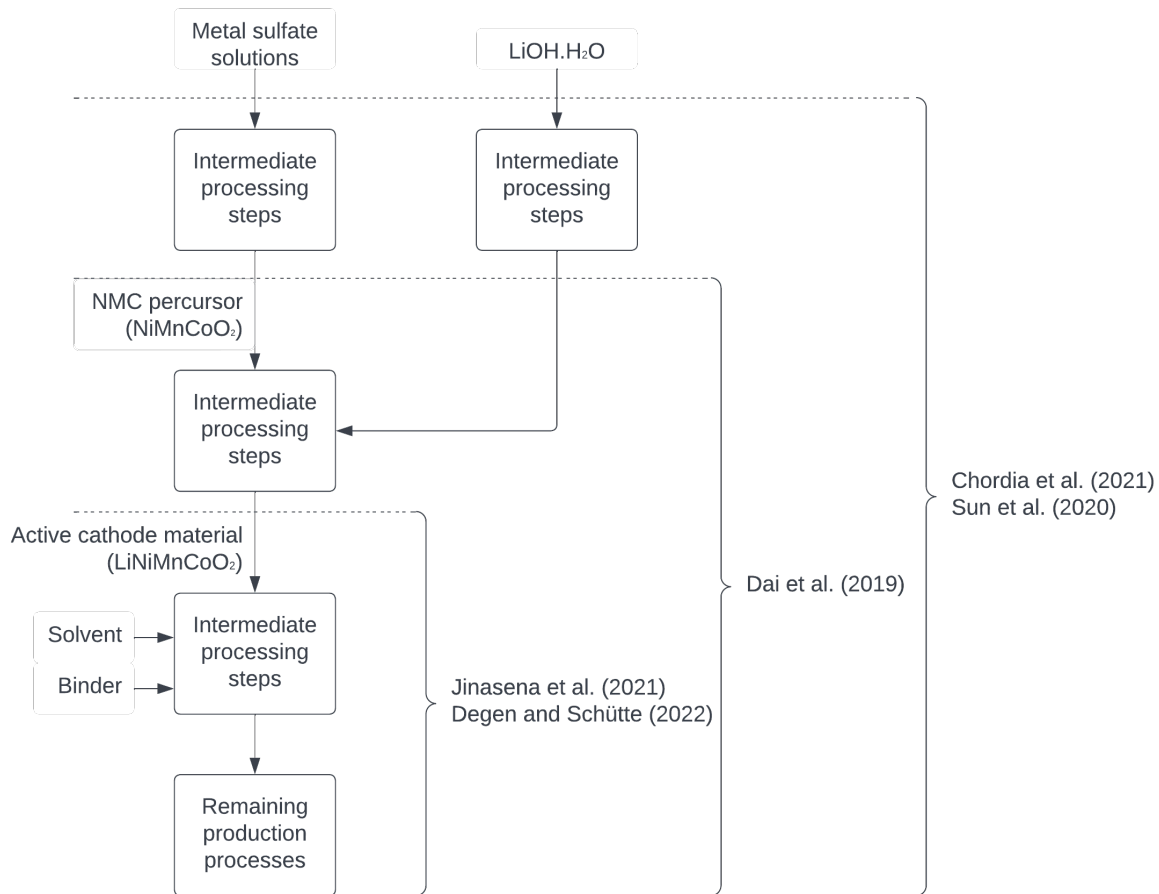


Figure 4: Example of varying upper technical foreground system boundary in different LCA studies

Process design of a factory is also influenced by the geographical location of the factory. Regional climatic factors such as average ambient temperature, humidity, and proximity to a water source affect facility design and hence the net energy consumption of cell production. This becomes relevant for process equipment, such as the dry rooms that require strict control of the operating environment (Ahmed et al., 2016). The influence of geography is likely more relevant in the design of high throughput facilities as compared to small-scale facilities. This is because the ambient environment could then be more effectively

used for optimizing heat and cooling in the facility – for example using lower temperature river water for cooling some process streams – instead of relying exclusively on industrial cooling equipment.

Table 2: Summary of LCA studies on large-scale LIB production. Paper 1 is highlighted.

Study	Cathode chemistry	Energy density (Wh/kg)	Production scale (GWh/a)	Data source
Ellingsen et al. (2014)	NMC-111	174	0.07	Miljøbil Grenland
Dai et al. (2019)	NMC-111	197	2	Chinese manufacturer
Sun et al. (2020)	NMC-622	180	30	Multiple Chinese manufacturers
Jinasena et al. (2021)	NMC-111	215	2	Theoretical calculations
Degen and Schütte (2022)	NMC-622	n.a.	7	FFB
Chordia et al. (2021)	NMC-811	225	16	Northvolt AB

4.1.3. Approaches to modeling energy consumption

A common approach when modeling energy consumption of technical processes is to use engineering principles, throughput volume, and the machine (or equipment) operational time to model the energy consumption of the facility (Piccinno et al., 2016). This is referred to as the "bottom-up" approach (Dunn et al., 2015a). A drawback of this approach is that when upscaling production volumes, different process equipment or technologies might be required, for which the engineering principles used to model the small-scale throughput may no longer be applicable (Kwade et al., 2018). For example, planetary mixers are used for slurry mixing for small-scale process operations, while at large scale continuous mixers/extruders can be used for producing slurry at large scale (Dreger et al., 2015). Often when there is a lack of sufficient information about specific technical processes, energy consumption can be modeled by means of suitable proxies that are principally similar to the processes being investigated. Thus, factors such as technical changes in upscaling and the suitability of proxies need to be accounted for when estimating the total energy demand of large-scale production facilities. This could severely affect the final estimates for total energy demand and provide an unrepresentative picture of high throughput facilities which are energy (and cost) optimized.

Contrast to the bottom-up approach is the "top-down" approach, where overall factory wide data is used in conjunction with the factory throughput for a given time span to calculate an average energy consumption. This was also the approach taken by Ellingsen et al. (2014), who modeled the energy demand of their small-scale factory based on the monthly electricity use of the battery manufacturer over an 18 month period. Care must be taken when using this approach to ensure the time intervals chosen for energy demand measurement are representative of the type of investigation being carried out. This includes the relation of the actual throughput during the time span investigated versus the designed throughput

capacity of the facility. A drawback of such an approach is that the information about the specific technical processes is limited by the high level (low resolution) of the data collected. Despite this, the overall estimates are likely to be more accurate than in the bottom-up approach.

In the absence of plant or equipment data, process simulation or engineering tools can be used to estimate energy consumption data. An alternative is to use proxies. Ultimately, it is important to ensure all processes in the factory are accounted for and correlate with the throughput level of the specific process or equipment and also the final throughput of the factory. As seen in the section 4.1.2, differences in technical scope of the various LIB production facilities assessed affect the overall energy consumption estimates, apart from variations caused due to the type of data used.

4.1.4. Shift-of-burden

Figure 5 shows the effects of upscaling LIB production in high- and low-carbon intensity scenarios represented by the South Korean (690 g CO₂-eq./kWh supplied electricity) and Swedish electricity mix (40 g CO₂-eq./kWh supplied electricity), respectively. The analysis of the small- and large-scale LIB production showed that global warming impacts reduced by almost 45% even in the high carbon intensity scenario. This points to higher energy and material efficiency of high-throughput facilities as compared to low-throughput facilities, irrespective of the carbon intensity of the energy source used during production. If, however, the carbon intensity of the energy source is as low such as the Swedish energy mix, then the global warming impacts are reduced by an additional 55%. As a result of upscaling and using energy from low-carbon intensity sources, the impacts shift almost entirely to the upstream phase of LIB production, i.e., to raw material extraction and production of battery materials (right most bar in Figure 5).

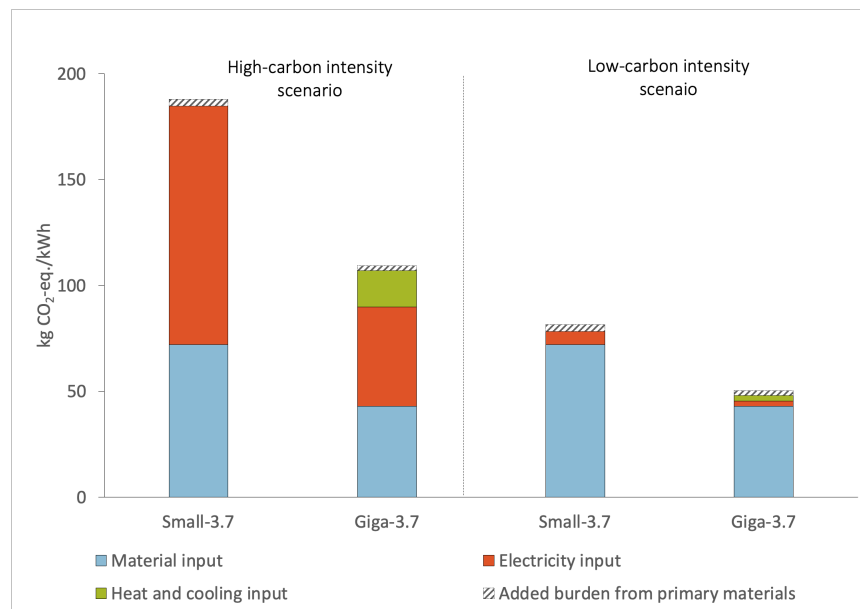


Figure 5: Global warming impacts for the small-scale (Small-3.7) and large-scale (Giga-3.7) factory models with different carbon intensity scenarios and data from Ecoinvent 3.7.1 for the background system

Several other environmental impacts, such as photochemical ozone formation, particulate matter formation, stratospheric ozone depletion, and ionizing radiation co-vary with the global warming impacts – i.e., impacts per unit of production reduce due to high energy and material efficiency at large-scale production, with a further reduction if low-carbon intensity energy sources are used. The main reason for this co-variance is that these environmental impacts also originate from emissions due to fossil fuel combustion, just like the global warming impacts. Thus, lower energy demand per unit production reduces emissions of nitrogen oxides, ammonia, and volatile organic compounds that cause hazardous ground level ozone and particulate matter, as well as chlorofluorocarbons that deplete ozone at high altitudes. However, not all impacts co-vary with global warming impacts. For example, the impacts due to ionizing radiation which are mainly caused by nuclear energy usage increased (Figure 6). Nuclear energy is common source of energy in low-carbon intensity energy mixes (like that of the Sweden). Similar tradeoff between low-carbon intensity energy sources and increasing impacts in environmental indicators is seen in the use of solar (photovoltaics) and wind power. For example, Lieberei and Gheewala (2017) conducted a LCA of various renewable energy production technologies based on photovoltaics, wind, and biomass and found that mineral resource depletion increased for photovoltaic and wind. Similarly, land use change and related environmental impacts due to biomass-based energy is another aspect that merit consideration when switching to low-carbon intensity sources for energy production (Hosseinzadeh-Bandbafha et al., 2021).

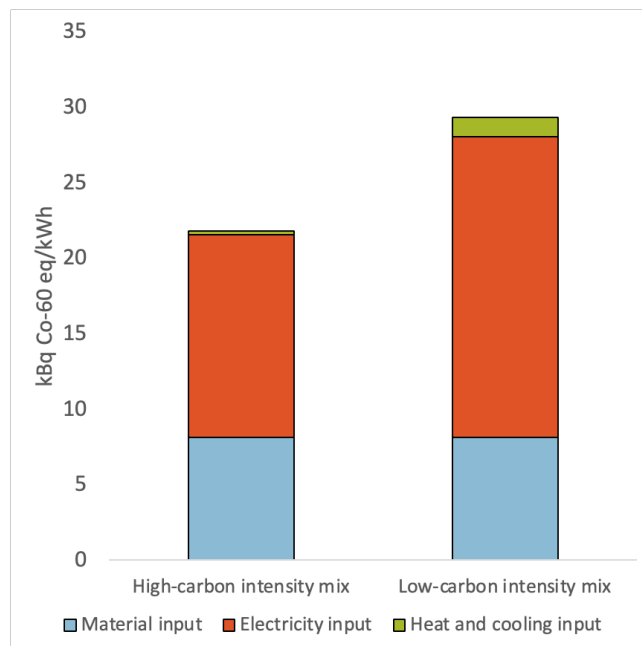


Figure 6: Comparison of ionizing radiation impacts between varying carbon intensity energy scenarios for large-scale LIB cell production

Regional environmental impacts such as terrestrial acidification and human toxicity (see Figure 4 and Figure 5 of Paper 1) also decrease due to upscaling of LIB production. However, these environmental impacts were already higher in the upstream part of the life cycle to begin with. Specific to acidification, the impacts are linked to the nickel sulfate and copper production, both of which are currently produced from sulfidic ores.

Interestingly though, nickel is primarily found in laterite ores (oxides), which constitutes for up to 80% of the nickel reserves (Mudd, 2010, Bhattacharya and Basak, 2016). Still, prohibitive costs associated with the processing of laterite ore implies that sulfidic ores are likely to remain the main source of nickel in the years to come. Insights such as these about ore types and future supply eventually led to motivations underpinning Paper 2. These are discussed in more detail in the next section 4.2.

Toxicity related impacts were notably affected by the choice between primary and secondary material inputs. This is relevant specifically in the context of the cell type analyzed in this large-scale factory study, which is a 21700 cylindrical cell using a steel container housing for the cell. Typically, low-alloyed steels are derived from steelmaking by using either electric arc furnace (EAF) technology or a route via blast furnace (BF) ironmaking and blast oxygen furnace (BOF) steelmaking (Björkman and Samuelsson, 2014). In the EAF, scrapped iron often accounts for all of the iron input, whereas the BF-BOF processes are ore-based, but often include a minor share of iron scrap added for cooling purposes (Ryman, 2007). Steel production where scrap is the main input requires treatment processes to handle the slag produced in the EAF. Especially, if the EAF produces a variety of alloys, including stainless steel grades, or uses stainless steel scrap as input, these treatment processes cause chromium emissions, which are highly toxic. The primary-only steel-making processes modeled in Paper 1 excludes all EAF processing, as well as all iron scrap inputs to the ore-based route (BF-BOF). In such a setup, toxic emissions of chromium are largely avoided in primary low-alloyed steel production, resulting in overall lower emissions as compared to producing recycled steels. However, the toxicity impacts due to chromium emissions seen here are due to modeling choice of data representing mixed steel scrap including stainless steel grades. The likelihood of a real-world situation where exclusively primary steel is used in manufacturing is likely limited to specific product applications.

Overall, there are several learnings from the assessment of regional environmental impacts such as acidification and toxicity. Firstly, as the impacts are already higher in the upstream phase of LIB production, the industry needs to start working with its suppliers to identify opportunities to mitigate or eliminate the impacts. Further, reducing fossil-based emissions is the obvious first step. However, not all impacts are caused by these emissions. Hence, the metal mining and processing industry needs to develop means to tackle regional impacts by investing in technology that and can help mitigate such non-fossil-based impacts as well. Secondly, the LCA practitioners assessing LIB life cycles need to investigate environmental impacts other than those from fossil combustion and highlight the processes where the emissions the highest. Thereby, preventing a shift-of-burden between different environmental impact categories, as these will be the ones remaining if the fossil fuels are phased out.

4.1.5. Long-term resource use implications

Resource use impacts were assessed using short- and long-term resource scarcity (Figure 7). Short-term resource use impacts, assessed using the SOP method available in the ReCiPe impact assessment package (Huijbregts et al., 2016), showed that nickel sulfate, followed by cobalt sulfate, lithium hydroxide and copper have the highest impacts. Whereas, long-term resource use impacts, assessed using the CSI method (Arvidsson et al., 2020), indicate copper as the most scarce resource, followed by nickel sulfate, cobalt

sulfate and lithium hydroxide. The SOP method characterizes minerals (metals) based on their ore grades and commodity prices and hence the results should expectedly vary over time if characterization factors are updated to reflect the prevalent economic scenario and market dynamics.

The CSI method is market, geography, and time agnostic and instead conveys information based on the crustal concentration of elements in the earth's crust. Arguably this is the strength as well as a limitation of the method. The CSI method, while decoupled from the technosphere, can provide useful information that can be used in long-term decision-making pertaining to technology adoption. On the other hand, policy and technology decisions are often reflective of the immediate circumstances or a near-term predictable future, influenced by market prices, security of supply, and contractual or trade agreements. Thus, for LCA practitioners, it is recommended to assess resource use impacts with both short- and long-term perspectives in the assessment of maturing and/or emerging technologies. Both CSI and SOP methods are non-regional indicators. Hence, they do not consider the geographical distribution of the resources into account. This is a limitation of both the methods and highlights the need for complementing resource use assessments with regional factors in LCA.

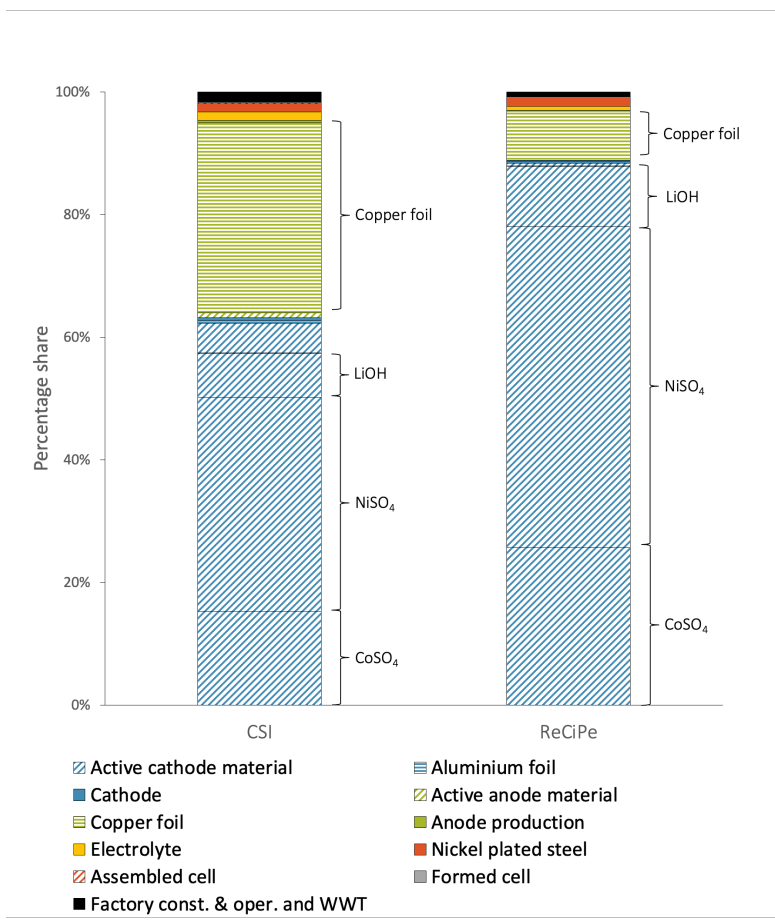


Figure 7: Resource use impacts of large-scale LIB production assessed using two different mineral resource scarcity assessment methods

4.2. Paper 2: Summary of results and learning outcomes

The underlying motivation for Paper 2 stemmed from insights gained from Paper 1 that showed a shift-of-burden to the upstream phase of LIB production resulting from upscaling and LIB production using low-carbon intensity energy sources. Further, an assessment of the battery supply chain materials led to the identification of data gaps and unrepresentative data especially pertaining to battery-grade LiOH·H₂O production in the Ecoinvent background database, which is a key input to LIBs. The data representing current nickel and cobalt supply chains, which are two other relevant materials used in the LIBs, have recently been updated by data from the Nickel Institute (Gediga et al., 2015) and the Cobalt Development Institute (CDI, 2016), respectively, and hence were not included in the scope for assessing the supply chains of relevant materials used in LIBs. However, no such update of data has been performed for the lithium supply.

The main research objective of Paper 2 was thus to review the data representing the current lithium supply chain while supplementing it with data representing potential future supplies. For this purpose, environmental and technical reports of a number of upcoming LiOH·H₂O production facilities were collected and their process data analyzed. A list of the current and future lithium supply routes considered along with the starting lithium grade, data source, and geographical location of the processing facility is provided in Table 3 for context. The assessment was conducted in the context of different lithium source types and varying lithium grades at source. A secondary objective of the study was to examine water use in the context of lithium extraction from brines. The motivation for this stemmed from the lack of attention paid to water use during brine extraction (Stamp et al., 2012) and the discussions in media regarding damage to the local ecosystem due to water shortage in the arid climate of the salars (Flexer et al., 2018, Gutiérrez et al., 2022).

Table 3: Data sources and geography for the current and future lithium supply sources reviewed

Temporal scope	Data source	Lithium source (%Li)	Geographical scope
Current supply	Ecoinvent v3.8	Brine (0.15%)	Salar de Atacama, Chile
	Kelly et al., 2021	Brine (0.17%)	Salar de Atacama, Chile
	Ecoinvent v3.8	Spodumene (1.86%)	Western Australia and China
	Kelly et al., 2021	Spodumene (0.8%)	Western Australia and China
Future supply	Lithium Americas Corp.	Brine (0.05%)	Salar de Cauchari, Argentina
	Minera Salar Blanco	Brine (0.09%)	Salar de Maricunga, Chile
	Nemaska Lithium	Spodumene (0.7%)	Whabouchi, Canada
	Keliber Oy	Spodumene (0.6%)	Central Ostrobothnia, Finland

4.2.1. Temporal shift-of-burden

The process design criteria for mining and processing facilities are based on technical, environmental, legal, and economic factors. The main consideration for the process design is the average ore grade at the start of the mining operations. As mining proceeds for years (or decades), the average ore grade available at the

site declines. This declining ore grade in reality is reflected in an increasing material, energy, and infrastructure input for processing the ore. To address this, a cut-off grade is set by mining operators beyond which mining operations at the site stop. Thus, decreasing ore grade correlates with increasing energy, chemical and water inputs and hence, the associated environmental costs (Magdalena et al., 2021). It is worth noting though, that the metal reserves are reported to be constantly increasing even though their demand has increased substantially (Jowitt et al., 2020). This is possible due to advancements in technology that enables exploitation of low-grade ores at costs comparable to exploitation from higher grade ores only a few years prior (Vidal et al., 2022). Still, care has to be taken to avoid falling in the trap of perpetual growth as technological improvements are reaching their potential thermodynamic limits (Vidal et al., 2022).

Figure 8 shows the global warming impacts from the current and future lithium supply routes. The figure also highlights the impacts based on the type of input (chemical, energy, etc.) and the unit process of origin. The brine-based processes that typically occur in the salars require less energy input for processing as compared to spodumene-based processes. This is because the brine-based extraction utilizes the heat from solar radiation for precipitation of the salts from the brine. The energy inputs on the brine extraction sites are typically for operating pumps, such as those pumping the brine up from the underground aquifers and transferring brine and freshwater between the evaporation ponds for sequential precipitation of salts. Some energy inputs are also required at the chemical plant for processing the concentrated brine to produce battery-grade $\text{LiOH}\cdot\text{H}_2\text{O}$. However, despite lower energy requirements on the site, the impacts from brine-based processes are calculated to be as high as the spodumene-based processes due to large amounts of chemicals that target the precipitation of specific salts from the brine. Furthermore, the impacts due to water use are also high in the brine-based processes as additional freshwater is needed for processing brine as well as transporting brine between the evaporation ponds once the water contained in the brine is evaporated (see also section 4.2.2). Hence, it is likely that as the lithium concentration in the underground aquifers decreases (due to dilution of the brine aquifers from neighboring freshwater aquifers or slow geological recharge of lithium in brines), the requirements for chemicals, freshwater, and energy will increase to compensate for the lower grade of lithium at source.

Spodumene-based $\text{LiOH}\cdot\text{H}_2\text{O}$ production processes are energy intensive. Processes such as the calcination, where α -spodumene is converted to β -spodumene occur at over 1000 °C, are particularly energy intensive and require large amounts of heat typically obtained from burning fossil fuels. Thus, for the spodumene-based routes, the source of energy used in production is an important consideration for reducing environmental impacts. This is especially important once the grade of lithium decreases at the mining sites over time, as then, larger volumes of α -spodumene will have to be calcinated to produce battery-grade $\text{LiOH}\cdot\text{H}_2\text{O}$. Freshwater ecotoxicity impacts are generally higher in the spodumene-based routes (see Figure 4 in Paper 2). The impacts are mainly linked to the treatment and handling of sulfidic tailings in the supply chain of the raw material inputs to the foreground system, which typically originate from the extraction and processing of sulfidic ores, not from the spodumene mining but from the mining of other metals.

Another example of potentially increasing environmental impacts over time is that of blasting during pit mining operations. Once again, as the lithium grade decreases, more rocks and boulders will need to be blasted to produce comparable volumes of battery-grade $\text{LiOH}\cdot\text{H}_2\text{O}$. Thus, a key message for LCA practitioners conducting prospective studies is to understand and communicate the temporal aspects of technologies or processes in their assessments. Secondly, an advice to the industry is to identify such trends and develop technologies that can help mitigate the impacts in the future, today.

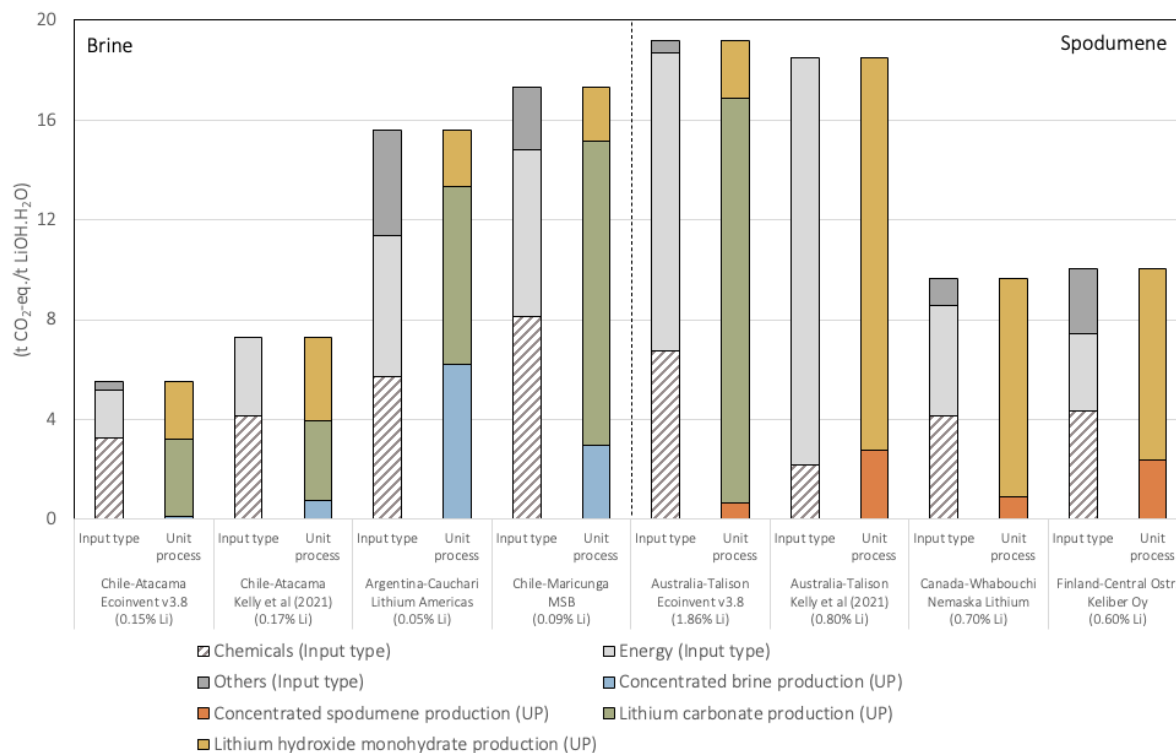


Figure 8: Global warming impacts from current and future lithium supply highlighting the type of input and unit process (UP)

In a recent study of future cobalt supply van der Meide et al. (2022) modeled cobalt production by variables such as the ore grade, share in the primary and secondary market, and energy transition scenarios. Here too, the environmental impacts of future cobalt production are expected to increase and highly dependent on the recycling market and the overall energy transition, with the likelihood of tradeoff between the environmental impacts based on the transition scenario chosen (sustainable development, business as usual etc.). The main recommendation for reducing the environmental impacts is to reduce the decline of high-grade ores (van der Meide et al., 2022). Per Usai et al. (2022), the demand for battery materials, such as aluminum, nickel, manganese and copper, does not constitute a concern. However, graphite, lithium and cobalt might be in short supply by 2035 due to the complexity of the supply chain and the lack of diversity in supply sources for some of these metals. Especially the production of cobalt, since it is mainly a co-product of copper and nickel mining, its production is linked to the market dynamics of these commodities (Stampatori et al., 2020).

When the source and grade of lithium is seen in the context of large-scale LIB production, overall global warming impacts can vary by 5-15% (see Figure 5 in Paper 2). Thus, the source and grade of lithium shows a small but non-negligible influence on the results. In a similar assessment, Kelly et al. (2021) estimated the share of $\text{LiOH}\cdot\text{H}_2\text{O}$ to be 20%. This is a relevant insight for the LIB manufacturers and the automotive industry, implying that they have to monitor their supply chain for environmental hot-spots and work with the suppliers to find opportunities to reduce the environmental impact of the metal extraction, processing, and production activities.

4.2.2. Characterizing the hydrological cycles in brine extraction

Water is an extricable part of ore extraction and processing steps during mining, especially during brine extraction for producing lithium. Brine by composition is essentially 70-75% water, and the remaining are dissolved solids. There are three pertinent issues with regards to characterizing water use in the context of lithium extraction from brines. First, it is the classification of brine itself. There persists an academic debate on whether to classify brine as mineral or water (Ejeian et al., 2021). The arguments in favor of treating brine as mineral are that brine is unfit for direct human consumption, or even direct agricultural use. Hence brine can potentially be treated just as another rock-based ore. However, the counter arguments point to the very composition of brine and the source of its chemical formation (Ejeian et al., 2021).

The second issue pertains to the interaction between brine and freshwater aquifers. Hydrological studies on the salars suggest that as brine is continually extracted from the brine aquifers, pressure drawdown effects could lead to seepage of freshwater on the periphery of the brine aquifers into the brine aquifers (Houston, 2006, Houston et al., 2011). This results in dilution of the brine aquifers and also lack of freshwater availability in the region. Characterizing such inter-aquifer mixing is not done in impact assessment methods applied in LCA.

The third issue is of water loss due to evaporation during brine concentration in the evaporation ponds. While the three issues described here are important and warrant further hydrological studies to better understand the issues, especially inter-aquifer mixing, another challenge is that of characterizing these aspects in impact assessment methods in LCA studies. Thus, a key message from this is directed at researchers working with water use in LCA to develop characterization methods to represent the hydrological cycles in relation to brine extraction operations in the salars.

In Paper 2, several issues pertaining to characterizing hydrological cycles in the salars were addressed. This is done by reporting the water content of brines at different brine extraction sites and calculating average water losses due to evaporation. Water use impacts assessed using the ReCiPe and the AWARE method point to high impacts in low lithium concentration brines (see Figure 3 in Paper 2). Low concentration of lithium in brine implies that large volumes of brine need to be extracted from aquifers for producing equivalent amount of $\text{LiOH}\cdot\text{H}_2\text{O}$. This, in turn, increases the inputs for energy, processing chemicals and freshwater. Results from Paper 2 show that the brine-water extracted is the highest in the case of low lithium content brines such as the ones in Cauchari ($788 \text{ m}^3/\text{t LiOH}\cdot\text{H}_2\text{O}$) followed by Atacama ($693 \text{ m}^3/\text{t}$

LiOH·H₂O) and then Maricunga (450 m³/t LiOH·H₂O). Compared to the freshwater demand, 307, 71 and 326 m³/t LiOH·H₂O, respectively, the brine-water volumes are notably higher. Additionally, since up to 95% of the water is evaporated from the evaporation ponds (Kaunda, 2020), the overall water use impacts could be significantly higher if the extracted and evaporated brine water affects the local groundwater. The impacts from water use, particularly in the salars, become relevant in the context of declining brine grades as demand for lithium or lithium-based products is expected to increase due to upscaling of LIB production. Thus, it is likely that the water use impacts could increase significantly in the future.

4.3. Paper 3: Update on model development

The motivations for Paper 3 stemmed from the insights gained from the review of the LCA literature on LIBs that highlighted the lack of primary data sources on large scale LIB production. With battery production ramping up all over the world, lack of industrial data on energy, resource use and emissions from battery production constitutes a hindrance in developing an understanding of how the impacts from cell production are affected by different cell formats, chemistries, and applications. This implies that little specific guidance on technology development can be provided to the industry or the policy makers on the current and future environmental impacts from LIB production. The development of a model platform to fill data gaps in LCA of LIBs is a step towards addressing this issue.

The preliminary results of the model platform highlight how an application-specific design of a given cell, i.e., power optimized vs energy optimized, or, changes in cell geometry, i.e., cylindrical vs prismatic, affects the environmental impacts from cell production. As a next step, the model platform will be further developed to address changes in cell chemistry, other prismatic cell formats, and pouch cells. Additionally, use phase calculations will be included to understand how the cell design affects the cell performance in the use phase of the BEV.

4.4. Synthesis of results

This section presents an update on results of large-scale LIB production presented in Paper 1, including the learnings from Paper 2 and Paper 3. The main objective of doing this is to present an updated estimate of the global warming impacts of large-scale LIB production in the context updated background and foreground data, effect of supply chains, and design for application of LIB cells. Figure 9 shows an updated estimate of the global warming impact for 1 kWh large-scale battery production. The updated results cover several aspects such changes in the background database, new industry data on battery grade graphite production, source and grade representing different LiOH·H₂O supply routes, and application-based changes in cell design. It is assumed that the large-scale factory is based in Sweden and uses only primary materials as inputs to cell production. The far-left bar in Figure 9 shows the results for the “Giga-3.7 model” (see Figure 3 of Paper 1) that represents large-scale LIB production in a low-carbon scenario modeled using the Ecoinvent v3.7.1 background database. The bars from left to right then highlight how the present estimate for large-scale LIB production can be updated based on considerations to data (both background

and foreground), supply route and application. These updates in estimates are discussed below in further detail.

4.4.1. Background database

The Giga-3.7 model used in Paper 1 was remodeled with Ecoinvent v3.8 (Bar #2 in Figure 9). As a result, the overall global warming impacts increased by 8%. The increase was mainly due to the updated dataset for $\text{LiOH}\cdot\text{H}_2\text{O}$ production available in Ecoinvent v3.8. Note that when a similar remodeling was done for the small-scale factory model in the Ellingsen et al. (2014) study, the impacts increased by 30% (see section 4.1.1).

The main update in terms of $\text{LiOH}\cdot\text{H}_2\text{O}$ production data between the two Ecoinvent database versions is in the dataset representing lithium carbonate production from concentrated spodumene. In the v3.7.1 this was modeled based on a material input estimate from Wietelmann and Bauer (2000), energy inputs based on Kim and Overcash (2003) and expert judgements. In v3.8 however, this dataset was replaced by primary industrial data collected at the largest rock-based lithium production company in China (Jiang et al., 2020). Almost the entire increase in impacts when the large-scale factory is modeled using v3.8 (instead of v3.7.1) was due to this single process dataset. This points to the effect of data quality on overall results and the need to contextualize LCA studies based on the background databases used for modeling.

4.4.2. Battery-grade graphite production

The battery-grade graphite production in Ecoinvent v3.8 is modeled based on Hischer et al. (2009), who in turn based their estimate on a combination of proxy data from the literature and oral communication. To improve the data quality for this process dataset, the battery-grade graphite production data was updated (Bar #3 in Figure 9) based on a recent study by Engels et al. (2022), who collected the data from an industrialized battery anode supplier based in China for the year 2019-2020. Overall, the impacts increased by a further 8% when the factory model is updated with improved battery grade graphite production data. Thus, this again emphasizes the importance of data quality and relevance in LCA studies. See section 3.3.3 for more details on the modeling of battery-grade graphite production process.

4.4.3. $\text{LiOH}\cdot\text{H}_2\text{O}$ supply routes

Different $\text{LiOH}\cdot\text{H}_2\text{O}$ supply routes are modeled to highlight the effect of source and grade of lithium on the overall impacts of large-scale LIB production (see Bar #4 Figure 9). See Table 3 for details on the type of source, location, and the starting grade of lithium used to model $\text{LiOH}\cdot\text{H}_2\text{O}$ input to large-scale LIB production. As seen in Figure 9 the supply of $\text{LiOH}\cdot\text{H}_2\text{O}$ could account for 5-15% of the overall impacts from cell production. This is a highly relevant insight for the LIB manufacturing industry as it highlights the significance of the supply chain for reducing environmental impacts. Further, varying grades assessed highlight how declining ore grades could affect environmental impacts in the future.

4.4.4. Application based cell design

As described in section 4.3, the design of cells used in BEVs could be optimized based on the desired application, i.e., power or energy optimized. Bar #5 in Figure 9 shows this in the context of large-scale LIB production. Based on the application, the global warming impacts could vary by about 4%. Although a relatively small amount, these differences can propagate when impacts are considered at pack level. Thus, in order increase the accuracy of their assessment, LCA practitioners might therefore want to account for specific changes cell design based on the application.

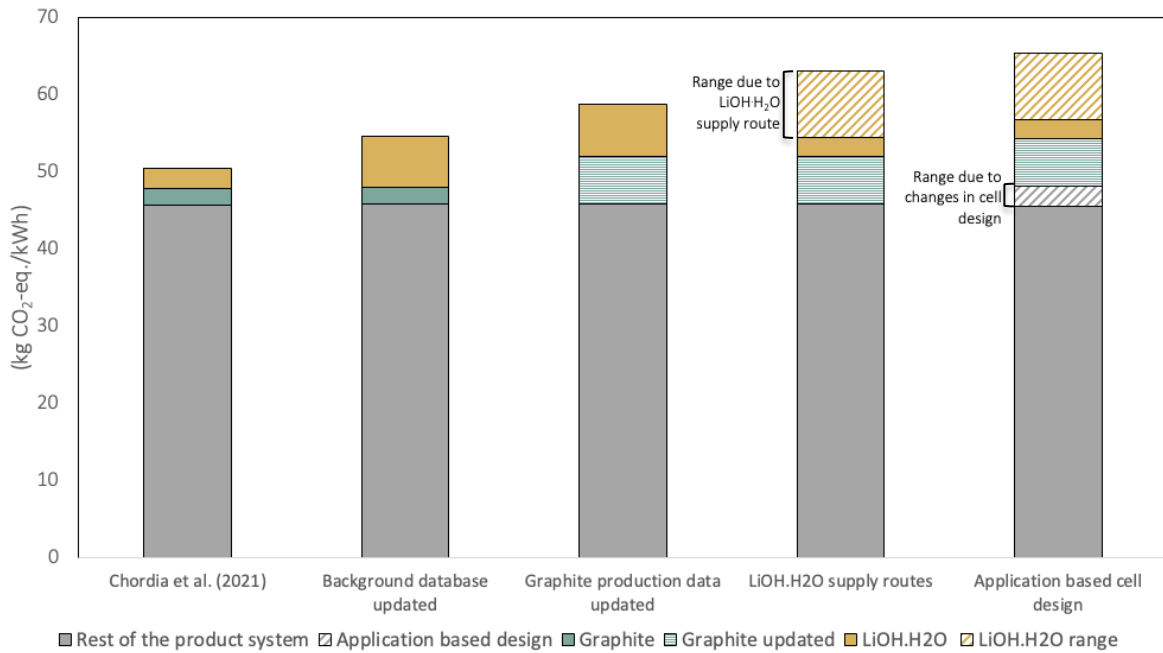


Figure 9: Updated estimates of global warming impacts of large-scale LIB production using low-carbon intensity mix as source

5. Conclusions and limitations

This licentiate thesis foremost aimed to examine the implications of upscaling LIB production. In response to the first research question, the thesis concludes that the environmental impacts reduce when LIB production is upscaled. This is due to the energy and material efficiency of production at large scale. Further, upscaling production leads to a shift in burdens to the raw material extraction and production phase for most environmental impact indicators. If, however, low-carbon intensity energy mix is used for cell production, the environmental impacts are almost entirely in the raw material extraction and production phase. Still, using low-carbon intensity energy sources can have other unintended consequences. For example, using nuclear energy could increase impacts from ionization radiation, wind and solar power could lead to higher mineral resource depletion, and energy from biomass could impact land use more.

To address the second research question, the thesis examined the effect of declining ore grades on overall environmental impacts from $\text{LiOH}\cdot\text{H}_2\text{O}$ production. Ore grades decline over time, due to mining activities. This corresponds to an increase in the energy and chemical inputs and thus an increase in the linked environmental impacts for producing an equivalent quantity and grade of the final product. The brine-based routes showed a rise in demand for energy and chemicals when a lower grade of brine was used to produce lithium. However, this trend was not as apparent for spodumene-based routes. This is because the type of energy sources, and the share of heat and electricity used at the various sites, differed. If the energy and material input data for each mining site considered in this research could be collected regularly and followed up, this would help develop a clearer understanding of the long-term implications of large-scale LIB production.

Combining the first and second research questions (as seen in section 4.4) shows that, overall, environmental impacts are still reduced due to upscaling LIB production. However, a key observation here is that while some impacts decrease, others increase. Thus, there is a shift in the type of environmental impacts occurring, due to upscaling LIB production.

To address the third research question, a physics-based model platform is under development with the aim of filling data gaps in LCA of LIBs. The model platform offers a novel methodology for combining cell design and engineering to fill data gaps in LCA such as, linking cell design to specific applications or performance.

Several methodological and data gaps in LCA of LIBs were identified during the course of this licentiate research. Specifically, in terms of methodology, remodeling older LCA studies (section 3.3) with different versions of background databases, exemplified the impact of background systems on LCA results. Further, changes in the background system represent how the technical systems evolve over time. An LCA study is only as good as the underlying data used to model it. And the responsibility is on the practitioner to investigate what the data represents. From the multiple examples of remodeling of LCA studies (section 3.3) using different versions of background databases and updating foreground data based on new industry data (see for example section 4.4), it is clear that the conclusions drawn from any study can vary significantly

based on the type of data used. Thus, LCA practitioners are recommended to look deeper into the source of data and to assess their results in the context of varying data quality.

Specifically, the primary data collection in Paper 1 and Paper 2, highlights the relevance of using technical reports and environmental permit data in LCA studies. Such data sources present an upper limit in terms of energy and material inputs and emissions permissible. Thus, they can be used for establishing a baseline for LCA studies assessing similar technical systems. As an example, the primary data on large-scale LIB production collected and analyzed in Paper 1 complemented the existing LCA data on large-scale LIB production already used in a number of published studies in the field. This provides context and highlights the differences in technology or processes being deployed in various regions all over the world. Similarly, Paper 2 presents LiOH·H₂O production data from a number of sites all over the world differing in terms of the source and starting grade of lithium. Using legal and environmental permit documentation to highlight the effect of changing ore grades over time is a novel approach taken in this thesis and complements the existing studies on LiOH·H₂O production.

Lastly, LCA studies need to be able to characterize the flows to and from the system when assessing specific technical systems. This might, at times, require the development of new impact assessment methods. For example, the case of brine extraction operations in the salars, that were assessed for their impacts on freshwater availability in the region (Paper 2). In this particular case there is a need to develop assessment methods that are able to address the complex hydrological cycles operating in the region. These include inter-aquifer mixing (i.e., brine and freshwater), evaporation, rainfall, and lithium discharge in brines. Additionally, specific issues such as that of brine extraction, could also benefit from the application of other ESA tools that assess water use in regions where brine is extracted.

5.1. Limitations and future work

There are two main limitations identified in the thesis. Firstly, in Paper 1, the impacts of upscaling production are presented by means of independent snapshots of small- and large-scale production. There is no investigation of how the individual processes, or the technology might change when LIB production is upscaled. This limits an understanding of how processes and technology change on a detailed level when production occurs at large scale. However, in the on-going development of the physics-based model presented in Paper 3, some aspects of process scaling such as, linking cell design to cell manufacturing, will be investigated which will address this limitation in Paper 1.

The second limitation of this thesis is, the type of data used in LCA. The thesis relies extensively on the environmental permit data which represents an upper bound in terms of the energy and material inputs and emissions. This presents a conservative estimate in terms of the results, which can be argued to contradict general LCA practices where the best available representation of the technical system is sought.

In the licentiate thesis, the technical scope was limited to the NMC-811 21700 cell. For future research the aim is to expand the technical scope to the full battery pack, followed by the vehicle and possibly the

charging infrastructure. The ongoing work described in Paper 3 is aimed at developing a model platform that could potentially fill data gaps in LCA studies of LIB. The ambition with the project is to expand the analysis to multiple cell geometries, chemistries, and application-based cell design. Additionally, expanding the technical scope would also entail broadening of the life cycle phases considered to the use and end-of-life phases. Furthermore, the licentiate depends entirely on the LCA methodology to assess the environmental implications of upscaling LIB production. For a broader understanding of upscaling production, additional environmental systems analysis tools could be applied to provide economy wide perspectives on sustainability of LIBs, including equity of resources, environmental justice, social impacts, and a safe operating space in terms of planetary boundaries.

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Paper 1

Environmental implications of upscaling lithium-ion battery production

The International Journal of Life cycle assessment

Paper 2

Life cycle environmental impacts of current and future battery-grade lithium supply from brine and spodumene

Resources, Conservation and Recycling

Paper 3

A model platform for solving lithium-ion battery cell data gaps in life cycle assessment

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