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Review

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

Mudit Chordia [*], Sanna Wickerts, Anders Nordelöf, Rickard Arvidsson

Division of Environmental Systems Analysis, Chalmers University of Technology, Sweden

ABSTRACT

Life cycle assessment studies of large-scale lithium-ion battery (LIB) production reveal a shift-of-burden to the upstream phase of cell production. Thus, it is important to understand how environmental impacts differ based on the source and grade of extracted metals. As lithium is highly relevant to several current and next-generation cell chemistries, we reviewed the effect of varying grades in different sources of lithium (brine and spodumene) worldwide. The review covered the Ecoinvent database, scientific literature, and technical reports of several upcoming production facilities. The results showed that lower-grade lithium brines have higher environmental impacts compared to higher-grade brines. However, spodumene-based production did not show such a trend, due to different technical process designs of the facilities reviewed. Water use impacts are higher in lower-grade sources and are expected to increase with decreasing lithium concentration. This could specifically be an issue in brine-based production, where brine is extracted from already water scarce regions and evaporated, thus increasing the risk of freshwater availability. However, these aspects of water use are not addressed in existing life cycle impact assessment methods. In the context of large-scale LIB cell production, the reviewed lithium hydroxide production routes account for 5–15% of the climate change impacts.

Abbreviations

AWARE	available water remaining
CF	characterization factor
CSI	crustal scarcity indicator
EST	energy storage technology
EV	electric vehicles
Li_2CO_3	lithium carbonate
LCA	life cycle assessment
LCIA	Life cycle impact assessment
$LiOH \cdot H_2O$	Lithium hydroxide monohydrate
LIB	Lithium-ion battery
SI	Supporting information
SOP	Surplus ore potential

1. Introduction

Lithium is at the core of the current energy transition and finds application in a wide array of energy storage technologies (Hussain et al., 2020). It is an active constituent in several commercially available (Blomgren, 2016) and next-generation battery chemistries (Edström et al., 2020). Thus, lithium is important for both present and possibly also future energy storage technologies (ESTs) (Ambrose and Kendall, 2020a). Meeting the rising lithium demand from an increase in ESTs has become a matter of concern (Castelvecchi, 2021), leading to several strategic joint ventures being formed between technology and mineral exploration companies to secure an uninterrupted supply of lithium (USGS, 2021). However, Kushnir and Sandén (2012) conclude that it is not the scarcity of lithium as such, but rather the ability to ramp up production that will determine its long term availability, as long battery lifetimes and multiple uses mean that recycling is unlikely to meet short-term supply (Olivetti et al., 2017).

Lithium is mainly extracted from brine aquifers and mineral ores, typically spodumene. Brine-based sources are being actively developed in the salars located in Chile, Argentina, and Bolivia in what is collo-quially known as the "lithium triangle" (López Steinmetz and Salvi, 2021). Spodumene-based sources are being developed in Australia and China, as well as at various stages of development in other parts of the world. The processing facilities are tailored for profitable extractive operations based on the lithium content at source. This affects the

* Corresponding author at: Department of Technology Management and Economics, Chalmers University of Technology, Gothenburg, Sweden.
E-mail address: mudit@chalmers.se (M. Chordia).

energy demand and chemical usage at the production sites (Calvo et al., 2016; Magdalena et al., 2021), and thus also the environmental impacts (Ambrose and Kendall, 2020a). Lithium mining operations in the salars have received criticism for their socio-environmental impacts (Giglio, 2021) and concerns regarding damage to the ecosystem in the region (Flexer et al., 2018). Hence, considering the growing adoption of ESTs, the environmental and resource impacts of current and future lithium supplies merit further consideration.

Life cycle assessment (LCA) can be applied for analyzing environmental impacts of products or services (Hellweg and Milà i Canals, 2014). Several LCA studies assess the lithium supply chain independent of subsequent applications. For example, Stamp et al. (2012) model the production of lithium carbonate (Li_2CO_3) from brines in Chile (Salar de Atacama) and Bolivia (Salar de Uyuni), as well as spodumene in Australia (Talison) and the Democratic Republic of Congo (Manono-Kitotolo). Li_2CO_3 is an important material in the context of battery production, as it is an intermediate product in the manufacturing of battery grade lithium hydroxide monohydrate ($LiOH \cdot H_2O$) used in some lithium-ion battery (LIB) chemistries. High-purity Li_2CO_3 can also be used for some next-generation battery chemistries. Jiang et al. (2020) collect primary data to assess the production of Li_2CO_3 from concentrated spodumene ore in China. Ambrose and Kendall (2020a) take a broader view and develop a resource model for predicting the future demand of lithium and link that to different environmental impact indicators. In a sequel paper, Ambrose and Kendall (2020b) use data collected by Stamp et al. (2012), An et al. (2012) and a technical report (Laferriére et al., 2012) to model life-cycle environmental impacts of lithium supply from brine and spodumene. More recently, Kelly et al. (2021) assessed the production of $LiOH \cdot H_2O$ from the brine and spodumene sources in Chile and Australia, respectively. The inventory data modeled in Stamp et al. (2012) and Jiang et al. (2020) are also incorporated in the Ecoinvent v3.8 database, which is currently the largest LCA database for inventory data (Ciroth and Burhan, 2021). The geographical representation of the data used in these LCA studies primarily covers lithium production from brines in Salar de Atacama (Chile), and spodumene in Talison (Australia). These two sources combined represent approximately 70% of current lithium production globally (USGS, 2021). However, additional brine and spodumene based sources with varying lithium concentrations at source are being explored and developed world-wide. Hence, there is a need to review data representing additional lithium sources, to better understand the implications of varying grades on environmental impacts of producing lithium.

Contributions to climate change impacts and energy use are frequently covered in LCA literature on lithium supply (Jiang et al., 2020; Kelly et al., 2021). However, Stamp et al. (2012) point out that the inclusion of water scarcity, specifically in relation to brine processes, is a neglected issue. Recently, Schomberg et al. (2021) used a spatially explicit water footprint method and compared the change of state of freshwater availability based on the safe operating space outlined by the Sustainable Development Goals. Kelly et al. (2021) differentiate between various classes of water based on their consumptive uses. They classify brine as being unfit for direct human consumption, which is why the water content of brine (brine water) was not included in their assessment. Since lithium is extracted from brines by evaporation and chemical treatment, only a small fraction of the brine water can be pumped back into the aquifers, resulting in a drop in aquifer height (Ejeian et al., 2021). This could lead to a pressure drawdown and freshwater from neighboring regions could permeate the brine aquifers (Houston, 2006; Houston et al., 2011), thus diluting them and indirectly reducing the local freshwater availability (Kesler et al., 2012). Another issue in brine extraction in the salars is that of slow lithium recharge in brines. Although geological studies suggest a continuous lithium recharge in brines (Langbein, 1961; Steinmetz, 2017), the recharge rates are several orders of magnitude lower than the variations caused by commercial scale exploitation of the salars, which can lower the lithium concentration in the brine due to the combined effect of dilution from neighboring freshwater aquifers and slow lithium recharge (Flexer et al., 2018). However, current life cycle impact assessment (LCIA) methods for assessing water use impacts do not address such issues arising from brine extraction in salars.

The main purpose of this study is to assess the environmental implications of varying lithium brine and ore grades on mining and processing of lithium from cradle to gate, up to the production of battery grade $LiOH \cdot H_2O$. As ore grades at mining and production sites decline due to continuous extraction operations, lower lithium grades at certain sites potentially represent the future declined state of higher-grade ores. For this, we conduct a detailed review of multiple lithium supply routes with varying lithium content at the source. The high-lithium content sources are incidentally also the starting point of today's dominant lithium supply routes globally, i.e., brines in Salar de Atacama (Chile) and spodumene in Australia. We refer to these as the "current" supply routes. The lower lithium content supply routes are represented by two brine-based (Salar de Maricunga in Chile and Salar de Cauchari in Argentina) and two spodumene-based (Whabouchi in Canada and Central Ostrobothnia in Finland) lithium sources, representing upcoming facilities at various stages of development. We refer to these as the "future" supply routes. A sub-goal of the study is also to contribute towards closing the gap in the current LCA literature for assessing brine extraction in relation to the hydrological cycles in the salars. We accomplish this by calculating the water content of brine at various sources and water volumes evaporated. Lastly, considering the growing relevance of lithium in ESTs, especially electric vehicles (EVs), we update a previously published study on large-scale LIB production (Chordia et al., 2021) with the current and future lithium supply data reviewed in this study, to estimate the influence of different supply routes on LIB production. The target audiences of this study are professionals involved in LIB production and the lithium supply chain industry, as well as LCA practitioners assessing life-cycle impacts of ESTs.

2. Method

Table 1 summarizes the lithium supply routes reviewed in this study and provides information about the data sources, geographic locations, lithium concentration at the source, and type of source (brine or spodumene). To produce $LiOH \cdot H_2O$ from raw brine, the brine is first extracted from the aquifers and pumped into concentration ponds. As the water in the brine evaporates due to solar radiation, some salts precipitate. Chemicals could also be added to precipitate the salts. This increases the lithium concentration in the brine. This process is referred to as "concentrated brine production". Next, the concentrated brine is subjected to heat treatment, chemical polishing, and solvent extraction

Table 1

Data sources and geography for the current and future lithium supply sources reviewed. The complete list of underlying references for these sources is provided in Table S1–1 of the SI.

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

to produce Li_2CO_3. This process is referred to as "lithium carbonate production". Li_2CO_3 is then converted to $LiOH \cdot H_2O$ by treating it with calcium hydroxide, which is referred to as "lithium hydroxide production". For $LiOH \cdot H_2O$ production via spodumene, the extracted ore is concentrated by mechanical separation and chemical treatment to remove excess gangue and increase the lithium concentration. This process is referred to as "concentrated spodumene production". The concentrated spodumene is then heat treated and chemically leached to produce Li_2CO_3 or $LiOH \cdot H_2O$. All reviewed product systems are modeled without any intermediate transport processes to be independent of specific processing site locations, as these might change in the future.

In Section 2.1 and 2.2, inventory data for current and future battery-grade $LiOH \cdot H_2O$ is described. In addition, technical process descriptions, detailed inventories, and overall results are presented in the SI. When calculating and comparing impacts from the reviewed lithium supply routes, a functional unit of 1 metric ton of battery-grade $LiOH \cdot H_2O$ is used along with an attributional, i.e., process-based approach (Yang, 2019). This is because $LiOH \cdot H_2O$ is a commonly traded battery material, and several datasets were available until the production of $LiOH \cdot H_2O$. Thus, for comparability, datasets with Li_2CO_3 as final product were supplemented with additional unit process data to represent the conversion of Li_2CO_3 to $LiOH \cdot H_2O$. The foreground system consists of the complete $LiOH \cdot H_2O$ production supply chain from the reviewed sources, starting with the extraction of lithium as brine or spodumene ore and ending with the production of battery-grade $LiOH \cdot H_2O$. The Ecoinvent v3.8 database (Steubing et al., 2016; Wernet et al., 2016) was used to model background processes linked to the foreground system.

2.1. Current supply routes

The current supply routes from brine and spodumene sources reviewed in this study are based on the unit-process data provided in the Ecoinvent v3.8 database and Kelly et al. (2021). Note that in Ecoinvent, multiple production processes and geographies can be combined into so-called "market processes" that contain average shares of each production process or geography. However, in this study, we do not consider aggregated market processes, but instead consider the specific supply chain for each lithium source.

The brine-based process for production of $LiOH \cdot H_2O$ described in Ecoinvent is a three-step process. In the first step, raw brine is treated to produce concentrated brine, followed by the production of Li_2CO_3 from the concentrated brine, and lastly the production of $LiOH \cdot H_2O$ from Li_2CO_3. The data sources used in Ecoinvent describing the unit processes until the production of Li_2CO_3 (Notter, 2021a,b), were originally published by Stamp et al. (2012), who in turn base the data on the lithium concentration provided in Yaksic and Tilton (2009), process design based on SQM (Sociedad Quimica y Minera S.A.), company reports (SQM, 2006, 2007, 2010) and personal communication. The spodumene-based route to produce $LiOH \cdot H_2O$ described in Ecoinvent is also a three-step process, beginning with the production of concentrated spodumene from mined spodumene ore, followed by the production of Li_2CO_3 from concentrated spodumene and lastly the production of $LiOH \cdot H_2O$ from Li_2CO_3. The inventory data used in Ecoinvent till the production of Li_2CO_3 is mainly based on data presented in Stamp et al. (2012) and Jiang et al. (2020). The unit process data for production of concentrated spodumene (Hischier, 2021) is based on the lithium concentration data in the ore provided in Stamp et al. (2012) and Ober (2017). The technical processes for iron milling, crushing, and limestone milling are used as proxies for the concentrated spodumene production (Hischier, 2007; Kellenberger et al., 2007). However, the Ecoinvent documentation report for this process states that output from the unit process is "milled spodumene". This implies that several activities that follow milling, such as floatation and concentration, which are necessary to produce concentrated spodumene, are not covered in the technical scope of the Ecoinvent process. The process for converting

concentrated spodumene to Li_2CO_3 is based on the LCA data provided in Jiang et al. (2020), who collected primary data from one of the largest lithium plants in China, accounting for nearly 14% of their national capacity.

The final process, i.e., production of $LiOH \cdot H_2O$ from Li_2CO_3 described in Ecoinvent (Sutter, 2021), is modeled based on inputs from several sources (Gendorf, 2000; Sutter, 2007; Wietelmann and Bauer, 2000; Wietelmann and Steinbild, 2014). It represents the production of $LiOH \cdot H_2O$ and calcium carbonate by reacting Li_2CO_3 with calcium hydroxide. The process is modeled based on stoichiometric calculations and data from a chemical plant (Gendorf, 2000). The applications listed in the Ecoinvent documentation include lubricating greases, production of dyes, adsorbents for carbon dioxide and additive to the alkali-silica reaction. However, batteries are not stated among the applications. $LiOH \cdot H_2O$ used in batteries requires very high purity and none of the applications stated in the Ecoinvent documentation require such high purity. Hence, this implies that the inventory data reported in Ecoinvent might not be fully representative of the technical process, materials, and energy requirements for producing battery-grade $LiOH \cdot H_2O$. Still, this data has been used in several LCA studies of LIBs (Kallitsis et al., 2020; Raugei and Winfield, 2019; Yin et al., 2019), including previous work by the authors (Chordia et al., 2021). It is therefore included as baseline in this review for benchmarking against other production routes reviewed in this study.

The brine-based process to produce $LiOH \cdot H_2O$ described in Kelly et al. (2021) is also a three-step process. The inventory data for these processes was collected via a joint effort between Argonne National Laboratory and SQM to conduct an LCA of Li_2CO_3 and $LiOH \cdot H_2O$ production. The spodumene-based process to produce $LiOH \cdot H_2O$ described in Kelly et al. (2021) is a two-step process beginning with the production of concentrated spodumene from the mined ore, and then directly converting the concentrate to battery-grade $LiOH \cdot H_2O$. The reference for the concentrated spodumene production process in Kelly et al. (2021) is Wietelmann and Steinbild (2014). The subsequent process for the conversion of concentrated spodumene to $LiOH \cdot H_2O$ is based on data from the environmental impact assessment report of a cathode plant (Tianqi Lithium Corporation) in China.

Note that although the supply routes for both brine and spodumene modeled in Ecoinvent and Kelly et al. (2021) have the same geographical origin (Salar de Atacama and Western Australia, respectively), the reported lithium concentrations at the source are different in all these datasets.

2.2. Future supply routes

The future supply routes reviewed in this study are based on data gathered from technical reports and environmental impact assessment reports of several lithium mining and processing companies (Lithium Americas Corp., Minera Salar Blanco, Nemaska Lithium Inc., and Keliber Oy) that are at various stages of developing their lithium assets. These are considered future supply routes as they are not yet active but are expected to be operational in 5–10 years. The lower lithium concentrations at these brine-based sources also reflect the possible future lithium grades of current supply routes from brines, whose lithium concentrations will likely decrease over the years due to extraction.

The brine-based future supply routes for production of $LiOH \cdot H_2O$ are similar to the current supply routes and represent operations at Salar de Cauchari, Argentina and Salar de Maricunga, Chile. These are two other locations within the "lithium triangle", where considerable lithium reserves exist. The average lithium concentrations at Cauchari and Maricunga are 0.05% and 0.09%, respectively. This influences the energy and chemical use, and to some extent the technical process designs at these facilities. The process information for the brine extraction operations in Salar de Cauchari was gathered from several technical reports prepared for Lithium Americas Corporation (Burga et al., 2020; Worley Parsons, 2011). Similarly, process information for operations in the

Salar de Maricunga was derived from technical reports prepared for Minera Salar Blanco (Atacama Water Consultants, 2022). When impacts from these future supply datasets for the brine route were calculated for comparative purposes, data from the specific reviewed sources were used until Li_2CO_3 production. After that step, Ecoinvent v3.8's unit process for converting Li_2CO_3 to $LiOH·H_2O$ was again used to model the final production step, and thereby fill the data gaps in the complete production process, for comparability with the other datasets modeled in the study.

The future supply routes for spodumene are described in technical reports of operations at the Whabouchi site, Canada and the Central Ostrobothnia region, Finland. While neither Canada nor Finland are among the countries with the largest lithium reserves (USGS, 2021), they represent examples of new lithium supplies that might become exploited in response to increased lithium demand. The average lithium concentration in spodumene at the Whabouchi site is about 0.7% and in Central Ostrobothnia region it is 0.6%. In these future supply routes, concentrated spodumene is directly converted to $LiOH·H_2O$ without producing Li_2CO_3 as an intermediate. The datasets representing mining operations in Canada and Finland are based on the technical reports prepared for Nemaska Lithium Inc. (Golder Associates, 2013; Laferriére et al., 2012; Nemaska Lithium Inc., 2013; SENES Consultants Ltd., 2013) and Keliber Oy (Keliber Oy, 2020a, b; Sweco, 2016), respectively. Specific data for chemical inputs was not available for the $LiOH·H_2O$ production unit process (from concentrated spodumene) in the Nemaska Lithium reports. Therefore, the chemical usage in the Keliber Oy reports were used as proxy data when comparing the different lithium supplies.

2.3. Impact assessment

The impact assessment focuses on impact categories based on their relevance to the technical system being assessed and the broader interest in the research community around LIB production. Foremost, climate change impacts, besides having high global relevance, are included since LIB storage technologies are mainly assessed for their potential to reduce greenhouse gas emissions (Kelly et al., 2021). Hence, this impact category is reported not only for $LiOH·H_2O$, but also a reference LIB (Chordia et al., 2021). Next, mineral resource scarcity is particularly relevant in the context of LIB production due to the use of rare metals (Nordelöf et al., 2014). To complement the shorter-term resource scarcity perspective with a long-term, we use surplus ore potential (SOP) method provided in the ReCiPe package (Huijbregts et al., 2016) and the crustal scarcity indicator (CSI) (Arvidsson et al., 2020), respectively. Water use impacts are particularly relevant for brine-based extraction processes, where large volumes of brine and freshwater are used in the processing, often in water scarce regions of the world. For this, we compare the water use indicator from the ReCiPe package with the Available Water Remaining (AWARE) impact assessment method (Boulay et al., 2018). The AWARE method quantifies the potential of water deprivation to humans and the ecosystem. Lastly, connected to freshwater extraction is also the aspect of water pollution. This is considered in terms of the USEtox method for assessing freshwater ecotoxicity (Rosenbaum et al., 2008), since USEtox is the most recent consensus method for assessing toxicity impacts in LCA. Complete ReCiPe package indicator results are reported for both $LiOH·H_2O$ and a reference LIB in the SI.

The mineral resource scarcity impact assessment for the supply routes starting with brine was conducted by considering the elemental composition of the brine at the respective salars. For the spodumene-based supply routes, the lithium concentration in the ore was considered. For the brine-based supply route modeled by Kelly et al. (2021), the average brine composition at Salar de Atacama (Worley Parsons, 2019) was used. For the spodumene supply route modeled by Kelly et al. (2021), a lithium concentration of 0.8% in spodumene ore as specified in their study was used. For both the brine and spodumene future supply datasets, the lithium concentrations in the brine and spodumene reported in the respective technical reports were used.

3. Results and discussion

First in this section, data for the current and future supply routes are reviewed at the unit-process level. This is followed by a discussion on impact assessment results for climate change, mineral resource scarcity, water use and freshwater ecotoxicity from the reviewed data sources. Finally, implications of varying lithium supplies for large-scale LIB production are discussed.

3.1. Unit-process comparison for LiOH·H₂O production from brine

The unit-process comparison for the current and future lithium supply routes with brine as the lithium source is presented in Table 2. Differences in inputs between Ecoinvent v3.8 and Kelly et al. (2021), such as the amount of brine extracted and electricity use, point to modifications in the technical processing over the years or that updated information regarding the process has become available.

3.1.1. Concentrated brine production

Part A of Table 2 presents unit processes representing the concentrated brine production in the different supply routes. The geographical starting point for all processes is the salars in Chile and Argentina. However, the lithium concentration at each source varies, being lowest at Cauchari (0.05%) and highest at Atacama (0.15%−0.17%). The process output is concentrated brine, which has higher concentration of lithium for the purpose of further processing, approximately 4–6% in three of the datasets. However, the Salar de Maricunga's reference report (Atacama Water Consultants and Worley 2022) states that the high concentration of dissolved solids, such as calcium, magnesium and lithium, implies that 3–4% lithium concentration is not attainable at ambient conditions in the Maricunga salar. Furthermore, it also states that maintaining higher than 1.5% lithium could lead to significant losses of lithium as entrapped salts. Thus, the process design was set to a lower target concentration of 0.9%.

The amount of brine extracted for processing depends on a combination of the initial concentration of lithium in the brine as well as the target concentration at the end of the process. The Cauchari brine undergoes the highest change in concentration due to its low initial lithium content and the Maricunga brine undergoes the lowest change amongst all supplies. This explains the order-of-magnitude difference between their respective brine inputs. In addition, the brine is pumped out from aquifers using pumps operating on electricity. Thus, brine volumes pumped out can be expected to correlate with the amount of electricity used in the process. This partly explains the difference in electricity usage between the Cauchari (high brine – high electricity) and the Maricunga (low brine – low electricity) brines. However, the brine input is highest in the process described by Kelly et al. (2021), despite a relatively modest increase in lithium concentration over the process. Even so, Kelly et al. (2021) does not report the highest electricity use. Hence, conclusive relationships between brine amounts extracted and electricity use are therefore difficult to derive based on the four reviewed datasets.

3.1.2. Lithium carbonate production

Part B of Table 2 provides data for the Li_2CO_3 production unit process. The highest input of concentrated brine in the lithium carbonate production is in the Maricunga dataset, which is explained by its lowest lithium concentration at the start of the process. This implies that a large volume of concentrated brine is required for producing the same mass of battery grade Li_2CO_3. This is also reflected in the high electricity and heat requirements in the Maricunga dataset. The Atacama datasets have similar initial lithium concentrations and seem to match well in terms of concentrated brine input versus electricity and heat requirements. In the Cauchari dataset, the concentrated brine input is slightly higher than in

Table 2
Unit process comparison for the current and future lithium supply routes from brine.

	Current supply route		Future supply route		
A: Concentrated brine production unit process (per ton of concentrated brine)					
Data source	Ecoinvent v3.8	Kelly et al. (2021)	Lithium Americas	MS Blanco	Unit
Geography	Chile (Atacama)	Chile (Atacama)	Argentina (Cauchari)	Chile (Maricunga)	
INPUT					
Lithium in raw brine (%) (1)	0.15%	0.17%	0.05%	0.09%	
Raw brine	4.45E+01	2.30E+02	1.27E+02	1.02E+01	t
Brine water	(2)	1.65E+02	9.43E+01	7.52E+00	t
Electricity	7.14E-03	2.11E-01	2.79E-01	2.81E-02	MWh
OUTPUT					
Evaporated water	(2)	(2)	8.76E+01	7.14E+00	t
Concentrated brine (Li%)	1 (6.7%)	1 (6%)	1 (4.3%)	1 (0.9%)	t
B: Lithium carbonate production unit process (per ton of lithium carbonate)					
Data source	Ecoinvent v3.8	Kelly et al. (2021)	Lithium Americas	MS Blanco	Unit
Geography	Chile (Atacama)	Chile (Atacama)	Argentina (Cauchari)	Chile (Maricunga)	
INPUT					
Concentrated brine	4.19E+00	4.00E+00	5.15E+00	3.69E+01	t
Energy					
Electricity	5.80E-01	4.17E-01	2.46E+00	5.51E+00	MWh
Heat	2.96E+00	3.20E+00	(2)	4.30E+01	GJ
Chemicals					
Soda ash	2.12E+00	2.00E+00	1.86E+00	2.32E+00	t
Lime/quicklime	6.08E+00	(2)	7.10E-03	9.28E-02	t
Sodium hydroxide	1.88E-04	(2)	2.00E-02	1.86E-01	t
Sulfuric acid	2.92E-03	(2)	2.30E-02	N/A	t
Hydrochloric acid	4.00E-02	(2)	4.50E-02	9.44E-01	t
Solvent and extractant	5.66E-04	(2)	(2)	4.77E-03	t
Other	1.44E-02	8.00E-02	(2)	1.02E-02	t
OUTPUT					
Lithium carbonate	1	1	1	1	t
C: Lithium hydroxide monohydrate production unit process (per ton of lithium hydroxide monohydrate)					
Data source	Ecoinvent v3.8	Kelly et al. (2021)	Lithium Americas	MS Blanco	Unit
Geography	Chile (Atacama)	Chile (Atacama)	Argentina (Cauchari)	Chile (Maricunga)	
INPUT					
Lithium carbonate	1.62E+00	1.05E+00	(3)	(3)	t
Energy					
Electricity	6.75E-01	1.39E+00	(3)	(3)	MWh
Heat	3.81E+00	2.40E+01	(3)	(3)	GJ
Chemicals					
Lime	1.62E+00	1.15E+00	(3)	(3)	t
Water	2.94E+01	5.00E-01	(3)	(3)	m^3
OUTPUT					
LiOH·H$_2$O	1	1	1	1	t

1 The lithium concentration in raw brine is based on the data reported in the reference.
2 Data not available.
3 Modeled the same as the unit process in Ecoinvent v3.8.

the Atacama datasets, which might explain the higher electricity and heat requirements in the process. Regarding input chemicals and reagents, missing data and differences in reporting makes comparisons challenging. However, it is noteworthy that the amount of soda ash (Na_2CO_3) input is similar between the datasets and close to the estimates by Dry (2018) who calculated chemical use for various brine sources. A reasonable explanation is that soda ash is mainly added to provide the carbonate in Li_2CO_3, which should be similar given that 1 ton of Li_2CO_3 is produced in all four cases.

3.1.3. Lithium hydroxide monohydrate production

Part C of Table 2 provides data for the LiOH·H_2O production unit process, which is the final process in the brine-based supply routes. For both the future brine supply datasets (Cauchari and Maricunga), the unit process information was available until production of Li_2CO_3 only, hence only the Ecoinvent and the Kelly et al. (2021) unit processes are compared. The unit processes described in Ecoinvent and Kelly et al. (2021) have reasonable agreement in terms of Li_2CO_3 and lime input, while the electricity, heat and water inputs in Ecoinvent are notably lower, which could be linked to the uncertainty in the proxy data used in Ecoinvent to represent LiOH·H_2O production from Li_2CO_3.

3.2. Unit-process comparison for LiOH·H_2O production from spodumene

The unit-process comparison for the current and future spodumene-based lithium supply routes is presented in Table 3. In the first step, concentrated spodumene is produced, and in the second step, LiOH·H_2O is produced from concentrated spodumene.

3.2.1. Concentrated spodumene production

Part A of Table 3 provides data for the concentrated spodumene production unit process. The starting point for all unit processes is the spodumene mining and extraction operations occurring at mining sites in Australia, Canada, and Finland. The lithium concentration in the ore varies between 0.6% and 1.9% at the different locations considered, with the highest in Talison, Western Australia. The concentrated spodumene contains 2.3–2.9% lithium. A notable difference is seen in the energy and chemical use in the four datasets, without any easily observable relationship between lithium concentration and energy use. The dataset with the highest lithium concentration (Ecoinvent v3.8) also reports relatively high energy requirements, and the dataset with the lowest lithium concentration (Canada) reports the lowest energy requirement. Furthermore, differences are seen in the balance between electricity and heat supplied on site for the ore processing operations. This can be due to the availability or lack of infrastructure, local

Table 3
Unit process comparison for the current and future lithium supply routes from spodumene.

	Current supply route		Future supply route		
A: Concentrated spodumene production unit process (per ton of concentrated spodumene)					
Data Source	Ecoinvent v3.8	Kelly et al. (2021)	Nemaska Lithium	Keliber Oy	Unit
Geography	Australia	Australia	Canada	Finland	
INPUT					
Lithium in spodumene (%) (1)	1.86%	0.8%	0.7%	0.6%	
Ore	1.98E+00	4.50E+00	4.79E+00	3.94E+00	t
Energy					
Electricity	3.39E-02	1.25E+00	4.60E-01	2.78E-01	MWh
Heat	1.18E+02	N/A	2.47E+02	1.74E+03	MJ
Chemicals					
Sodium hydroxide	(2)	(2)	2.07E-03	1.94E-03	t
Sulfuric acid	(2)	(2)	3.37E-04	6.06E-04	t
Collector	(2)	(2)	1.31E-03	6.30E-03	t
Flocculant	(2)	(2)	4.64E-05	1.52E-04	t
Dispersant	(2)	1.50E-02 (3)	4.74E-04	N/A	t
Other chemicals	(2)	(2)	1.56E-03	2.97E-04 (2)	t
Water	4.17E-01	3.00E+00	5.88E+00	9.88E+00	m^3
OUTPUT					
Concentrated spodumene (Li%)	1 (2)	1 (2.3%)	1 (2.9%)	1 (2.4%)	t
B: Lithium hydroxide monohydrate production unit process (per ton of lithium hydroxide monohydrate)					
Data source	Ecoinvent v3.8	Kelly et al. (2021)	Nemaska Lithium	Keliber Oy	
Geography	China	China	Canada	Finland	Unit
INPUT					
Concentrated spodumene	1.35E+01	6.42E+00	5.81E+00	1.04E+01	t
Energy					
Electricity	6.62E+00	3.50E+00	1.22E+01	1.49E+00	MWh
Heat	9.22E+01	7.13E+01	4.25E+01	2.07E+01	GJ
Chemicals					
Sodium hydroxide	8.54E-01	1.18E+00	1.12E-01 (4)	1.12E-01	t
Sodium carbonate	3.51E+00	2.50E-02	1.58E+00 (4)	1.58E+00	t
Sulfuric acid	4.69E+00	1.52E+00	(2)	(2)	t
Hydrochloric acid	2.80E-01	(2)	7.20E-02 (4)	7.20E-02	t
Other	2.99E+00	6.00E-01	1.82E+00 (4)	1.82E+00	t
Water	7.69E+01	1.12E+01	(2)	7.49E+01	m^3
OUTPUT					
LiOH·H$_2$O	1	1	1	1	t

1 Lithium concentration in spodumene is based on the data reported in the reference.
2 Data not available.
3 The dispersant input in unit process described by Kelly et al. (2021) includes sodium carbonate.
4 Modeled same as the Finland supply route dataset.

environmental legislation, availability of resources or economic reasons specific to the sites. For example, Kelly et al. (2021) describe a process where all electricity requirements on site are met by burning diesel in a generator. Contrary, the data presented for mining operations in Finland and Canada describes wood chips and diesel, respectively, as fuels for heat generation on site, whereas electricity is sourced from the grid. Another notable aspect is the use of chemicals, which are provided in detail only in the two future supply datasets but not in the current supply datasets. Thus, comparisons of chemical inputs are again hampered by data gaps and differences in reporting.

3.2.2. Lithium hydroxide monohydrate production

Part B of Table 3 provides the unit process data for the production of LiOH·H$_2$O. The data presented in the future supply datasets represents a process where LiOH·H$_2$O could be produced directly from the concentrated spodumene. In the current supply datasets, Kelly et al. (2021) present the data representing production of LiOH·H$_2$O (or, alternatively, Li$_2$CO$_3$) from concentrated spodumene. For comparability with the other unit processes described, the unit process for Li$_2$CO$_3$ production in Ecoinvent has been merged with the step for LiOH·H$_2$O production from Li$_2$CO$_3$ and normalized to the LiOH·H$_2$O reference flow. There are notable similarities between the datasets regarding spodumene concentrate input and energy requirement, at least at an order-of-magnitude level. Chemical inputs show less similarities between datasets, but this might again be due to data gaps and different reporting approaches.

3.3. Recommendations based on unit process comparison

Each of the datasets reviewed in this study have their strengths and weaknesses in terms of data completeness, representativeness, reliability, age, and temporal scope. The Ecoinvent v3.8 brine dataset aims at representing current production, but it is based on data from before 2010. Also, it is unclear whether the LiOH·H$_2$O grade is high enough for battery production. Kelly et al. (2021) also aims at representing current battery grade LiOH·H$_2$O production and is based on more recently acquired industry data. Although Kelly et al. (2021) do not provide detailed chemical use, this dataset could be recommended for modeling the current LiOH·H$_2$O production from brine, especially if complemented with additional information on chemical usage. Considering the large lithium reserves at Salar de Atacama, Kelly et al. (2021)'s dataset could be relevant also for prospective modeling of future LiOH·H$_2$O production. However, accounting for the likely increase in brine extraction from the salars, future market processes might also include shares of the lower lithium grade Cauchari and Maricunga datasets, preferably based on relevant projections and forecasts. These datasets are relatively comprehensive, although the Maricunga dataset is based on more recent data.

The spodumene datasets from Ecoinvent v3.8 and Kelly et al. (2021), rely on secondary data until the concentrated spodumene production. However, in the subsequent steps, Kelly et al. (2021) considered the production of LiOH·H$_2$O directly from spodumene concentrate based on industry data. The Ecoinvent v3.8 dataset covers Li$_2$CO$_3$ production from spodumene concentrate, and then production of LiOH·H$_2$O. It only has updated information until the production of Li$_2$CO$_3$ based on industry data from Jiang et al. (2020). Thus, when comparing the two, Kelly et al. (2021) is the preferrable option for modeling LiOH·H$_2$O production, with the caveat that data for spodumene concentrate production is updated with more representative data. Kelly et al. (2021) could also be considered for prospective studies due to the large reserves of spodumene in Western Australia, which is likely to remain a major source in the years to come. In terms of future supply, both the Finnish and Canadian datasets represent production until battery grade LiOH·H$_2$O. They are also comprehensive, recent and based on industry and environmental permit data. The Canadian dataset could be further improved with specific information on chemical use. The Finnish and Canadian datasets might also be applied in prospective modeling of future local lithium supplies in Europe and North America, respectively.

3.4. Climate change results

Fig. 1 shows the climate change impacts for the current and future lithium supply routes. Overall, the impacts are lower in the lithium extraction and concentration processes for both brine and spodumene, and higher when converting the concentrate (brine or spodumene) to Li$_2$CO$_3$ and LiOH·H$_2$O. For the brine-based supply routes, the emissions are highest for the Cauchari and Maricunga brines, which represent
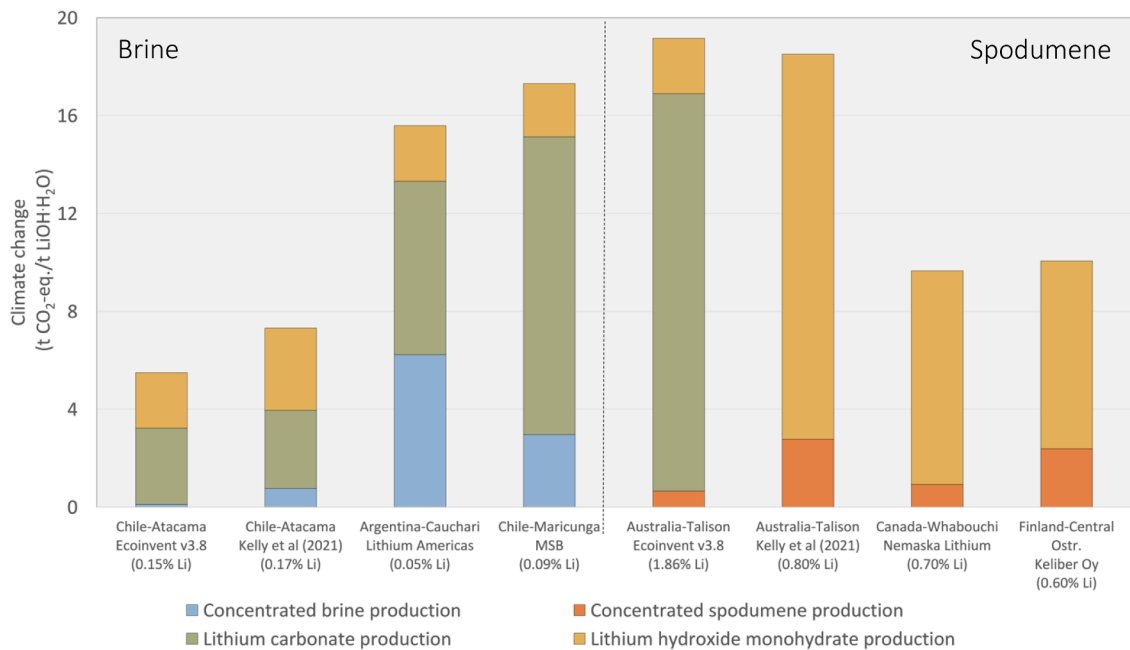
Fig. 1. Climate change impacts for brine- and spodumene-based lithium supply routes.

future supply routes. One explanation is the lower lithium concentration in the brine at these locations, which implies that greater volumes of brine need to be processed to produce equivalent grades of LiOH·H₂O, as compared to the brines in Atacama, where the initial lithium concentration is approximately twice that of Cauchari or Maricunga. Processing high brine volumes in the future supply routes implies that more energy and chemicals are required for processing which leads to higher overall emissions. A potential way to address this would be to invest in and adopt mechanical processes for brine processing instead of chemical precipitation (Flexer et al., 2018) as this might lower the overall energy demand and chemical use, thereby also lowering environmental impacts.

In the case of spodumene-based lithium supply, impacts are highest from spodumene extraction in Australia, although the lithium concentrations in those ores are higher (1.86% and 0.8%) compared to the Canadian (0.7%) and Finnish (0.6%) ores. This is due to carbon-intensive sources being used to generate energy for the extraction and processing operations in the Australian operations. Specifically, for the process considered by Kelly et al. (2021), diesel is the only source of fuel used for site operations to produce spodumene concentrate. In addition, the subsequent processing of the spodumene concentrate into LiOH·H₂O is carried out using the carbon-intensive Chinese energy mix. For the future supply production routes (Canada and Finland), the climate change impacts are distributed between the energy and chemical use as well as a small share to other processes, such as blasting and management of tailings from the spodumene concentrate production.

3.5. Mineral resource scarcity results

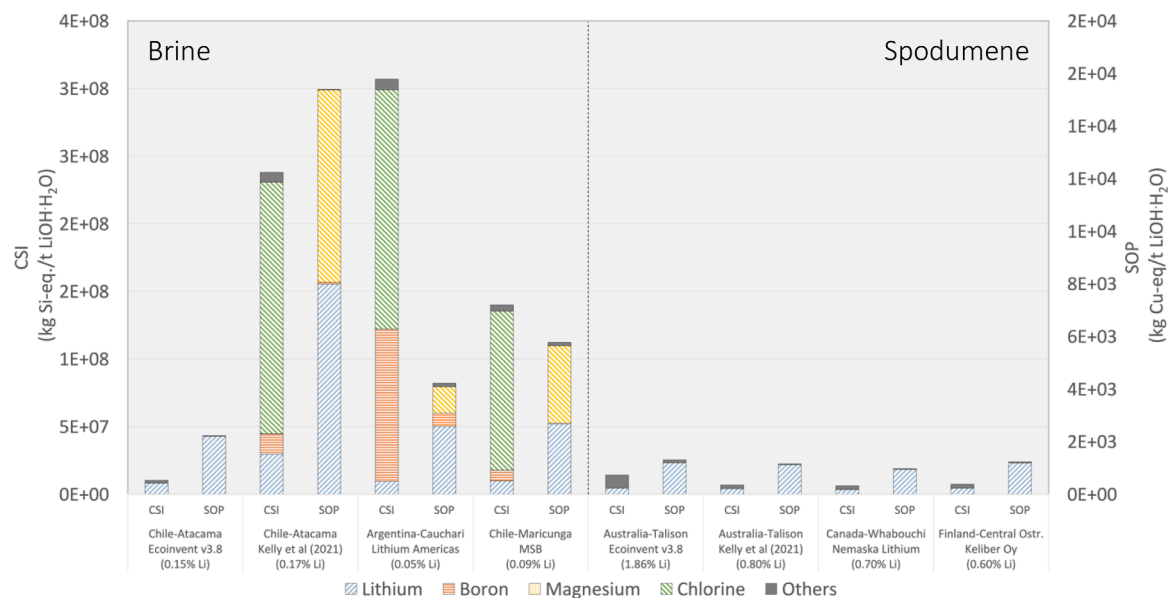Fig. 2 shows the results for the mineral resource scarcity impacts

Fig. 2. Mineral resource scarcity impacts for brine- and spodumene-based lithium supply routes. The figure compares the mineral resource impacts using the Crustal Scarcity Indicator (CSI, left-axis) and the Surplus Ore Potential (SOP, right-axis) indicators. The CSI and SOP are not numerically comparable.

from the current and future LiOH·H$_2$O supply chains assessed by the CSI and SOP methods. Overall, the resource impacts from brine-based routes show higher impacts as compared to the spodumene based routes. This is mainly due to the co-extraction of other dissolved substances in the brine, primarily chlorine, boron and magnesium, whose characterization factors (CFs) are higher than the CF for gangue (rock and soil) that are recovered along with spodumene during ore mining. Although other metals apart from lithium are also extracted when spodumene ore is mined, their relative amounts compared to the brines are much lower. Lastly, when comparing the brine- and spodumene-based processes in terms of the ratio of net lithium extracted to the amount used in the final product (i.e., LiOH·H$_2$O), the brine-based processes point to higher lithium extracted per unit of final product compared to the spodumene-based processes. For the brine-based routes, this ratio of lithium extracted to lithium in final product is approximately 3–10, whereas in the spodumene-based routes this ratio is approximately 1–3. This is due to significant losses of lithium during the evaporation steps of concentrated brine production, thus pointing to the difference in the efficiency of the two processes in terms of resource use. However, several salts produced as by-products during brine-based processes could potentially be used in other applications. For example, potassium chloride, commonly referred to as potash, is naturally precipitated during evaporation and can be used as a fertilizer. However, in this study, the use of potash and other potentially useful salts produced are not considered, and all burdens are allocated to LiOH·H$_2$O production.

When assessed by the CSI, the impacts of the brine-based routes are highest due to chlorine extraction, followed by lithium. Brines contain a high share of dissolved solids (25–30%) and amongst them the chlorine concentration is the highest. In the Cauchari and the Maricunga brines, impacts are also high due to the boron content in the brines of those salars. Although the boron content in the brine is comparatively low, the CF for boron is higher than for lithium and chlorine due to its relative geochemical scarcity in the earth's crust as per the CSI method. For the spodumene-based supply routes, the impacts are similar for spodumene extraction in Australia (0.8% lithium), Canada (0.7% lithium) and Finland (0.6% lithium). The impacts mainly depend on the lithium content in the spodumene and the consumption of sodium carbonate. A precursor material for sodium carbonate is sodium chloride, which contribute to the CSI of LiOH·H$_2$O mainly through extraction of chlorine from the crust. Chlorine has a relatively high CF according to the CSI method because of its low concentration in the crust, but this rarity is less problematic in practice due to the high abundance of chlorine from sea water.

When assessing the brine-based routes by the SOP, the impacts are almost equally shared between magnesium and lithium in the Atacama and the Maricunga brines. In the Cauchari brines, impacts are highest for lithium, followed by magnesium and boron. The magnesium/lithium ratios in the Atacama (5.6) and Maricunga brines (6.5) are almost double that in Cauchari (2.3), whereas the boron concentration in Cauchari brines is almost 8 times that of Atacama and Maricunga. In the spodumene based routes, the impacts are also mainly due to lithium when assessed by the SOP. It is worth noting that in the SOP method, there are no CF for chlorine, which means that the chlorine extracted through brines or as sodium chloride from the crust remains unassessed by the SOP.

3.6. Water use results

Water use impacts in the current and future lithium supply chains are shown in Fig. 3. The figure shows both the total freshwater use in the supply chain (as per ReCiPe) and the regionalized impact of water use (as per AWARE). Amongst the brine-based supply routes, the Cauchari and Maricunga salars have the highest water use impacts. A plausible explanation is the low initial concentration of lithium in those salars as compared to the Atacama salars, which implies that more brine needs to be extracted and then diluted with equivalent high shares of freshwater for lithium extraction. In terms of water scarcity from the AWARE method, the Maricunga (Chile) salars have the highest impacts. This is an important consideration in the context of the Atacama salars: also in Chile, as the lithium concentration in the brine aquifers decreases overtime, the water scarcity impacts will increase. Amongst the spodumene-based supply routes, high water use is seen in the Canadian and Finnish supply routes. However, due to large availability of natural water in these regions, the water scarcity impacts are lower.

The water content of the brine in the various salars was used to estimate the amount of brine water extracted per ton of LiOH·H$_2$O produced, except for the brine route represented in Ecoinvent, which does not provide water content data for the brine. Results show that the brine water content extracted is the highest in the case of the Cauchari salar (788 m^3/t LiOH·H$_2$O), followed by Atacama (693 m^3/t LiOH·H$_2$O) and then Maricunga (450 m^3/t LiOH·H$_2$O). When compared to the freshwater use determined using ReCiPe for these salars, i.e., 307, 71 and 326 m^3/t LiOH·H$_2$O, respectively, brine water volumes are notably higher than the freshwater use. Further, over 90% of the brine water is
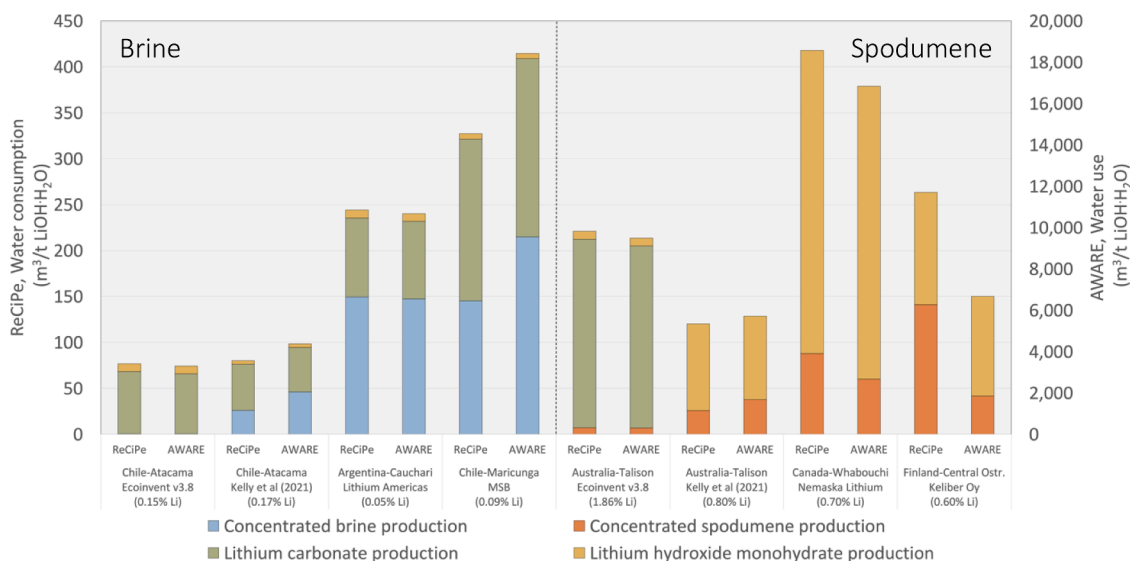

Fig. 3. Water use impacts for brine- and spodumene-based lithium supply routes. The figure compares the water use impacts for brine and spodumene based supply routes based on ReCiPe (left axis) and AWARE (right axis).

evaporated. This implies that depending on the correlation between brine water extraction and freshwater seepage into brine aquifers, the overall water use impacts could be significantly higher. As in the case of addressing climate impacts, this could be argument for introducing mechanical separation techniques, as they would lower chemical usage and possibly allow processed brine water to be pumped back into the reservoir, instead of evaporating it (Flexer et al., 2018). The idea then is that if the brine aquifers are replenished with processed brine water, it could reduce the risk of pressure drawdowns and thereby reduce the risk of water seeping in from freshwater aquifers in neighboring regions.

3.7. Freshwater ecotoxicity results

Fig. 4 shows the freshwater ecotoxicity results for the current and future lithium supply routes. The ecotoxicity impacts in all supply routes mainly occur in background processes due to tailings produced from mining and processing of sulfidic ores, followed by the treatment of hard coal ash linked to coal production in the supply chain. In the brine-based routes, impacts are seen due to sanitary landfills in the background supply chain. In the spodumene-based supply routes, blasting, which is done prior to ore extraction and processing, leads to toxic emissions. In the spodumene mining, extraction and processing operations also lead to the production of tailings during the production of the concentrate. However, as spodumene is a non-sulfidic ore which generally produces non-sulfidic tailings (Aylmore et al., 2018; Lemougna et al., 2019), the likelihood of tailings from spodumene mining being sulfidic is low. However, adjoining soil or chemicals used in the processing of the ore could lead to the production of toxic tailings, which then need to be carefully isolated from the rest of the environment to mitigate any harmful effects on the surrounding ecosystem (Nguyen et al., 2021).

3.8. Implications for large-scale LIB production

Fig. 5 shows the cradle-to-gate climate change impact of a large-scale LIB production facility (giga-factory) coupled with the data for current and future lithium supply routes reviewed in this study. The giga-factory is modeled based on the LIB production data published in Chordia et al. (2021). The results show that the supply route to produce LiOH·H₂O could account for 5–15% of the total climate-change impacts from LIB production at giga-factory scale when using the Swedish energy mix for cell production. Note that the Swedish energy mix consists of a large share of renewables (e.g., hydropower, bioenergy, and wind) and

nuclear power and is representative of a low-carbon intensity mix (approximately 40 g CO_2 eq./kWh). Amongst the spodumene-based routes, the highest impacts are seen when lithium is sourced from Australia. This is mainly due to carbon-intensive sources used for energy generation on site. Lithium sourced from spodumene mined in Canada or Finland has lower emissions, even though the average lithium content in the ore at these sites is lower (0.7% and 0.6%) than in Australia (0.8–1.86%). This is due to the low-carbon intensity of the energy generation at those sites. In the brine-based supply routes, Maricunga (Chile) and Cauchari (Argentina) have the highest emissions. This is largely due to the high-carbon intensity of the electricity supplies as well as the high use of chemicals for precipitating the various salts from the brine, including the production of Li_2CO_3. The lower lithium contents in the Cauchari and Maricunga brines mean that more energy and chemicals are used for the extraction and production of battery grade materials such as LiOH·H₂O. Low-carbon intensity energy sources could offset some of the emissions. However, emissions due to chemical production and use will likely remain high unless other technologies are developed and used for lithium extraction from brines.

4. Conclusion

In this study, we reviewed existing and new inventory data and analyzed how varying (declining) ore grades change the environmental impacts of the lithium supply chain. The results show that the amount and types of impacts depend on the type of lithium source (brine or spodumene). Also, starting from a lower lithium content at source influences concentrating processes in both brine- and spodumene-based routes. Low-lithium content brines showed higher climate change, water use, and freshwater ecotoxicity impacts compared to high-lithium content brines, which is linked to high chemical and energy use in processing of low-grade brines. Further, as raw brines are continually extracted, the lithium concentration in the brine decreases, possibly due to dilution effects from underground seepage of freshwater into brine aquifers (Houston et al., 2011). Hence, larger raw brine volumes need to be extracted to produce an equivalent amount of battery grade LiOH·H₂O. This means that unless there are significant technological improvements in existing operations, impacts may increase as brines with lower lithium concentration will be extracted. One possible option is to adopt mechanical processes for brine processing instead of chemical precipitation (Flexer et al., 2018).

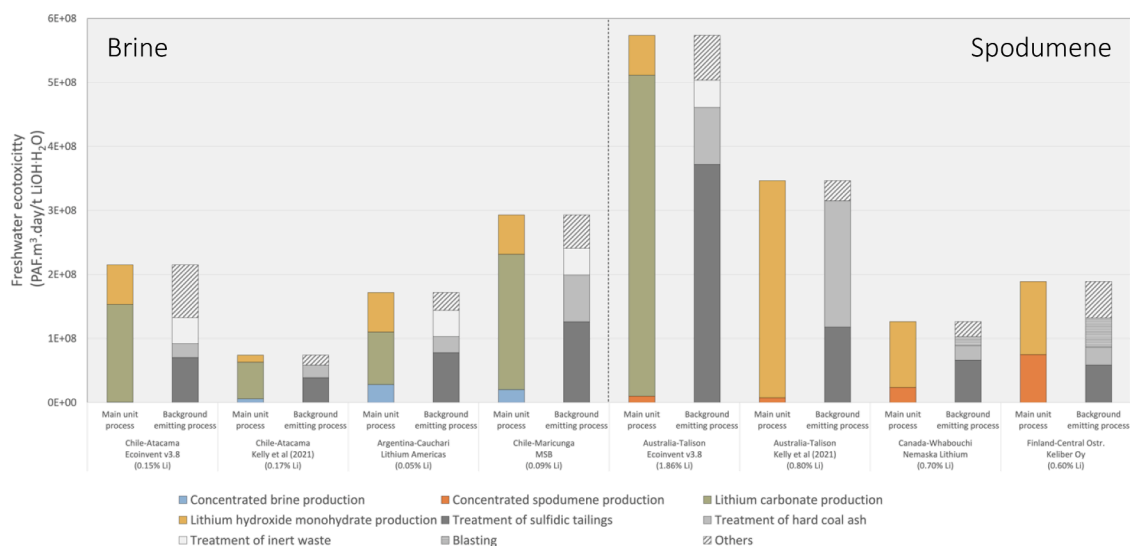Similar to the brine-based production of LiOH·H₂O, continual

Fig. 4. Freshwater ecotoxicity impacts for brine- and spodumene-based lithium supply routes. The figure shows results divided in two ways: (i) the main unit processes in the foreground system and (ii) the background processes where the toxic emissions occur.
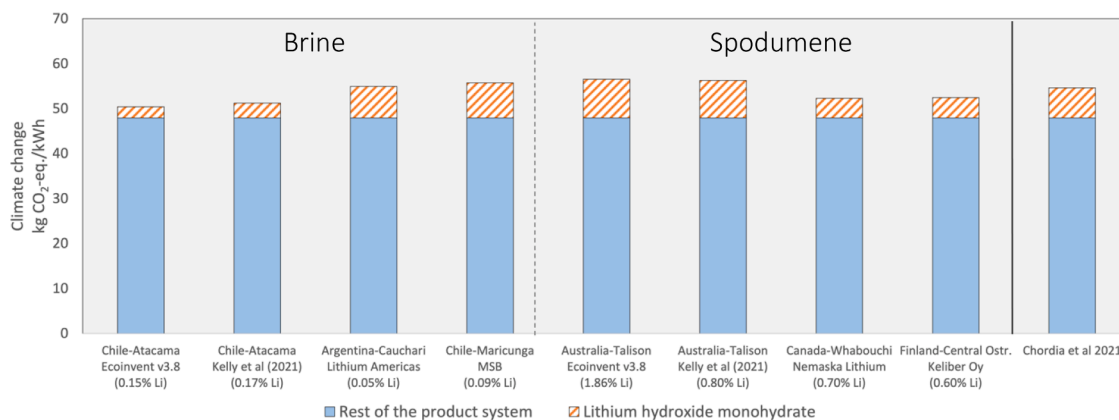
Fig. 5. Implications of lithium supply routes on large-scale LIB production in Sweden. Previous results from Chordia et al. (2021) (right-most bar) are provided for comparison.

spodumene-based production could be expected to lead to declining ore grades and subsequent higher energy demand and chemical usage in spodumene concentration operations. However, this effect is not clearly seen in this study. Instead, impacts are linked to the specific processes used for extraction and the processing operations at the different sites. For example, the site operations described in Australia point to diesel being used exclusively to meet the electricity requirements on site for the concentrated spodumene production unit process, whereas in the in the future lithium supply routes, heat from either diesel, natural gas or wooden pellets and electricity from the grid is used to produce concentrated spodumene. Such a difference in energy sources can influence the overall environmental impacts from the production processes. More conclusively, for both the brine- and spodumene-based LiOH·H$_2$O production routes, using fossil-free sources of energy for all operations is an obvious, albeit challenging, step with a considerable potential to reduce the impacts of lithium supply.

Water use impacts are expected to increase as the lithium concentration in brines decreases over time. This issue might worsen in regions that are inherently more arid or face water scarcity and continue to produce lithium from brine aquifers. When quantified, brine water volumes pumped up to the surface and evaporated are higher than direct freshwater use in LiOH·H$_2$O production. Thus, depending on the correlation between brine water extraction and underground freshwater seepage into brine aquifers, the overall water use impacts of brine extraction could be significantly higher than what is shown by existing LCA results. Therefore, to further assess the water use impacts specific to brine extraction in the salars, new LCIA methods need to be developed that address the complex hydrological cycle occurring in the region. This includes developing regional CFs that consider interactions between underground freshwater and brine aquifers.

Finally, a key message to stakeholders in the LIB production and the lithium supply chain industry is to find ways to reduce the demand for primary lithium when producing cells, for example by reducing scrap in existing production processes and ensuring availability of less burdensome secondary lithium by means of recycling of spent cells.

Declaration of Competing Interest

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

Data Availability

Data will be made available on request.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2022.106634.

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