Regionalized life cycle assessment of present and future lithium production for Li-ion batteries

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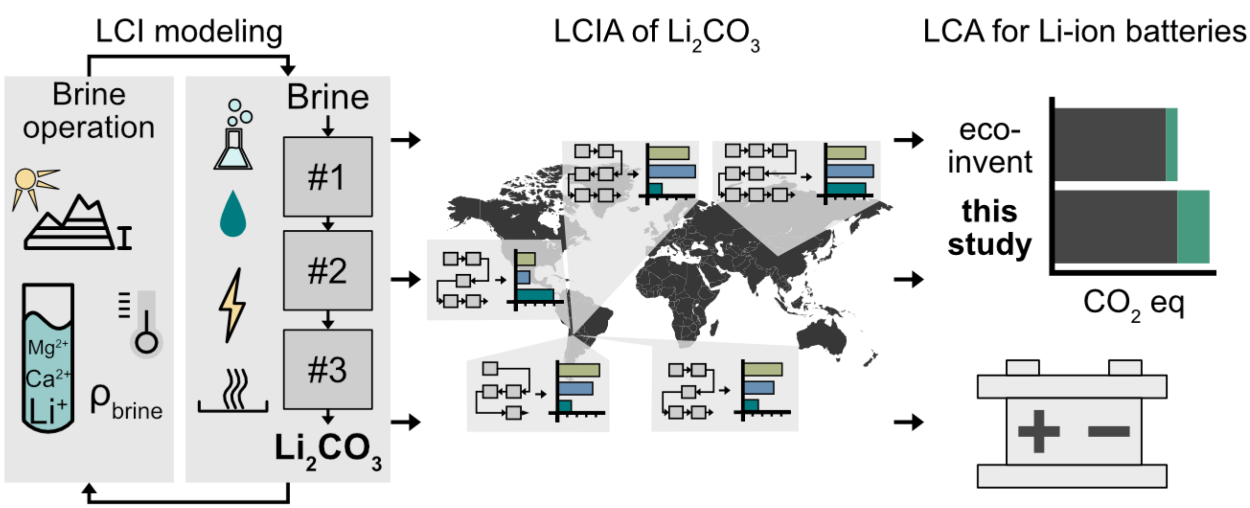
Abstract

Existing life cycle assessments (LCA) of lithium carbonate production from brines are mainly based on one single brine operation site, while many different lithium carbonate production routes have been developed in the past. Hence, current life cycle inventories do not capture the variability of brine sites and misestimate life cycle impacts. This study presents a systematic approach for LCA of existing and future lithium carbonate production from brines, which can furthermore be applied to geothermal brines or seawater. It has been used to model life cycle inventories of three existing and two upcoming brine operations in Argentina, Chile, and China and combined with regionalized life cycle impact assessment. Impacts on climate change, particulate matter human health impacts, and water scarcity from lithium carbonate production differ substantially among sites. Existing life cycle inventories for lithium-ion battery production underestimate climate change impacts by up to 19% compared to one from our study.

Keywords: Lithium, Environmental impacts, Life cycle assessment, Life cycle inventory, Brines

Graphical abstract



1 Introduction

The development of energy storage led to an increased demand for battery metals (Christmann et al., 2015; Wanger, 2011; World Economic Forum, 2019). By 2030, the battery demand is forecasted to grow by 1400 % and hence, the demand for Lithium (Li) used in Li-ion batteries is expected to increase by a factor of 6 with respect to 2018 (World Economic Forum, 2019).

More than two-thirds of the Li resources are located in Argentina, Bolivia, Chile, and China as brine deposits which hold a great supplying potential in the future (Bertau et al., 2017; Kesler et al., 2012; Munk et al., 2016). The primary producer of lithium carbonate (Li_2CO_3) from brines is Chile, followed by Argentina and China (S&P Global, 2021). Brine operations produce Li_2CO_3 with a technical grade (min. 99 wt. %) and battery grade (99.5 wt. %). The latter is used to manufacture Li-ion batteries (Dai et al., 2020).

Various production routes for Li_2CO_3 from brines have been developed in the past (Tran and Luong, 2015). Generally, the processing can be subdivided into three main processes: Brine's mass reduction in solar evaporation ponds, brine purification, and Li_2CO_3 precipitation (Garrett, 2004; Tran and Luong, 2015). The brine is pumped from an aquifer/salt lake into evaporation ponds to reduce the brine volume by solar evaporation. When a specific Li concentration of the brine is reached, the brine is sent to the processing plant. The purification part consists of a variety of processes in different arrangements to remove impurities (Calcium (Ca), magnesium (Mg), or boron (B)) from the Li-enriched brine, such as by adding quicklime to remove Mg, using organic solvent extraction to remove B, or using ion exchangers to remove Mg, Ca, or B. The selected processes and their order depend on the site-specific brine compositions. Once the pulp has been purified, Li_2CO_3 is precipitated by heating the pulp and adding soda ash. Crystallized Li_2CO_3 (technical grade) is dissolved in water at low temperature. The solution is re-heated at 80 °C, and Li_2CO_3 (battery grade) precipitates. The final product is dried in a rotary dryer (Garrett, 2004; Tran and Luong, 2015). In addition to this approach of extracting Li from brines, other processing techniques include selective Li recovery. The Li-ion selective adsorption technique uses adsorbents (manganese oxide, titanium oxide, or alumina) to selectively uptake Li from the brine sent through the ion exchangers (Garrett, 2004; Tran and Luong, 2015). Once the adsorbents are saturated with Li, the columns are washed and take up Li. The Li-containing solution is then sent to evaporation ponds to further concentrate Li (Garrett, 2004). Once a specific concentration is reached in the solution, Li_2CO_3 precipitation (technical grade) is forced by heating the pulp and adding soda ash, as explained for the previous production route. Crystallized Li_2CO_3 (technical grade) is dissolved in water at low temperature by adding pressurized gaseous CO_2. The solution is re-heated at 80 °C, and Li_2CO_3 (battery grade) precipitates. The product is dried in a rotary dryer (Garrett, 2004; Tran and Luong, 2015).

Regarding environmental impacts, energy provision for Li_2CO_3 production is mainly based on fossil fuels contributing to climate change (Stamp et al. 2012, Kelly et al. 2021). Stamp et al. (2012) published life cycle inventory (LCI) data for brine-related Li_2CO_3 production from the Salar de Atacama in Chile. This data was integrated into the ecoinvent LCI database in 2012 and has not been updated or expanded (ecoinvent, 2021). Kelly et al. (2021) used more updated recent technical data from the Salar de Atacama to quantify impacts on climate change and water scarcity but also did not improve on the coverage in terms of different brine sites. Ambrose and Kendall (2019) slightly extended the coverage by including lab-scale data from the Salar de Uyuni in Bolivia when assessing climate change impacts.

When assessing water scarcity impacts related to Li-ion battery storage, water scarcity impacts of Li from brines were classified as critical, according to Schomberg et al. (2021). They included brine consumption in the water scarcity footprint (WSF) when applying the LCA midpoint indicator AWARE. However, brines are not directly used by ecosystems or humans as a water source and should, thus, not be considered when applying this LCA method (Boulay et al., 2018). Brine pumping affects the hydrogeological systems with wetland and lake ecosystems at the Salar de Atacama but these direct and indirect effects of brine pumping are only measurable by assessing the hydrogeology of these salt flats (Liu et al., 2019; Liu and Agusdinata, 2021; Marazuela et al., 2019).

The collection of LCI data is extremely time-consuming (Kawajiri et al., 2022), and so the scientific focus of LCI development for Li_2CO_3 production has been on the Salar de Atacama site in Chile with comparably good data availability and a high share of global Li_2CO_3 production (57 % of the LCE production from brines in 2018 (S&P Global, 2021)). However, processing techniques from other brine operations differ from the one used at Salar de Atacama since they vary in their chemical composition (Flexer et al., 2018; Houston et al., 2011; Munk et al., 2016; Tran and Luong, 2015). Thus, production routes adapted to the brine chemistry and other environmental parameters were developed (Garrett, 2004; Swain, 2017; Tran and Luong, 2015). Hence, what is missing so far in literature is a detailed assessment of other Li_2CO_3 production pathways and their related environmental impacts. Furthermore, the existing LCA studies are difficult to compare since the goal and scope of these studies vary. Differences in system boundaries and degree of transparency hamper the direct comparison of these studies. Hence, the main objective of this paper is to develop a systematic approach to model site-specific LCIs of Li_2CO_3 production from brines when operational data from the companies are not publicly available. We apply our approach by assessing environmental impacts of Li_2CO_3 (battery grade) production from five brine operations in Chile, Argentina, and China. We cover climate change impacts, regionalized human health impacts from fine particulate matter (PM) formation and partly regionalized WSFs. Finally, by integrating Li_2CO_3 from different brine operations, the consequences on climate change impacts related to Li-ion battery production were assessed.

2 Methods

2.1 Framework to assess environmental impacts of Li_2CO_3 from brines

We present an approach to quantify environmental impacts of Li_2CO_3 production from brines (see Figure A-1 in supplemental information A). Specifically, we developed a modular approach to model site-specific LCIs, which allows for flexible adjustments to future process updates at each extraction site and can also be applied to other brines in future research. The approach follows the ISO 14040:2006 and ISO 14044:2006 standards to allow a standardized LCA (ISO, 2006a, 2006b). Hence, four steps need to be examined: Goal and scope definition, LCI analysis, life cycle impacts assessment, and the final interpretation.

Step 1: Goal and scope

The goal and scope should be defined in the first step according to ISO 14044 (ISO 2006). We suggest setting the functional unit to 1 kg Li_2CO_3 (battery grade). For example, this facilitates the integration of LCIs in Li-ion batteries when performing LCAs on future low-carbon mobility systems. System boundaries should be set accordingly to the project's scope (e.g., the system boundaries (cradle-to-gate approach) could be set from pumping the brines to the surface until the final product (Li_2CO_3, battery grade) leaves the processing plant).

Step 2: LCI analysis

Step 2 consists of modeling the LCI for Li_2CO_3 production from one or multiple brine sites if site data are not publicly available. We propose using the approach as developed in this study (Figure 1): (1) identification of relevant processes and site-specific environmental and technical parameters and (2) process-specific modeling of energy and material demand.

(1) Identification of relevant processes and site-specific parameters

Literature research (e.g., company reports, patents, scientific papers) is required to identify the relevant processes to produce Li_2CO_3. The process configuration determines the mass flows of the Li-containing pulp and thus, requires a detailed assessment for the latter LCI modeling. The types of input and waste production for all identified processes need to be defined. Environmental and technical parameters (e.g., chemical composition of the brine, evaporation rate, or annual average air temperature) need to be considered because they influence resource demand of Li_2CO_3 production (e.g., heating demand of Li_2CO_3 precipitation or the chemical demand to remove B from the pulp). Sources of thermal energy can be retrieved from company reports. Identifying processes and relevant parameters is an iterative process since the type of Li_2CO_3 production also guides the number and type of parameters required for LCI modeling.

(2) Process-specific modeling of material and energy demand

The reported Li concentration of the enriched brine serves as an approximation for the required mass of brine entering the processing plant. The input demand for processes, such as purification (Mg removal by adding quicklime) and Li_2CO_3 precipitation (adding soda ash), is dependent on the mass flows (pulp entering the process). The chemical composition of the brine affects required industrial chemical demand (e.g., the mass of Mg in the brine is proportional to the amount of quicklime if it is added to brine stored in the evaporation ponds (Flexer et al. 2018)). Mass of process-related chemicals (e.g., quicklime to remove Mg in the pulp) and produced waste (e.g., NaCl precipitation due to soda ash) are suggested to be stoichiometrically calculated. The required mass of chemicals should be adapted by adding a percentage to account for the incompleteness of chemical reactions according to available data. Those inputs and outputs need to be investigated to determine the mass of pulp going into the next process. Further affecting mass flows within the processing plant is residual pulp re-circulation to previous purification steps, which needs to be considered if reported. Energy demand can be quantified once process-specific mass flows are determined. Thermal energy demands are influenced by the mass, temperature difference, and heat capacity of the pulp (Figure 1). Literature values per kg pulp for process-specific electricity demands are available and can be used to determine the operational electricity requirement on-site.

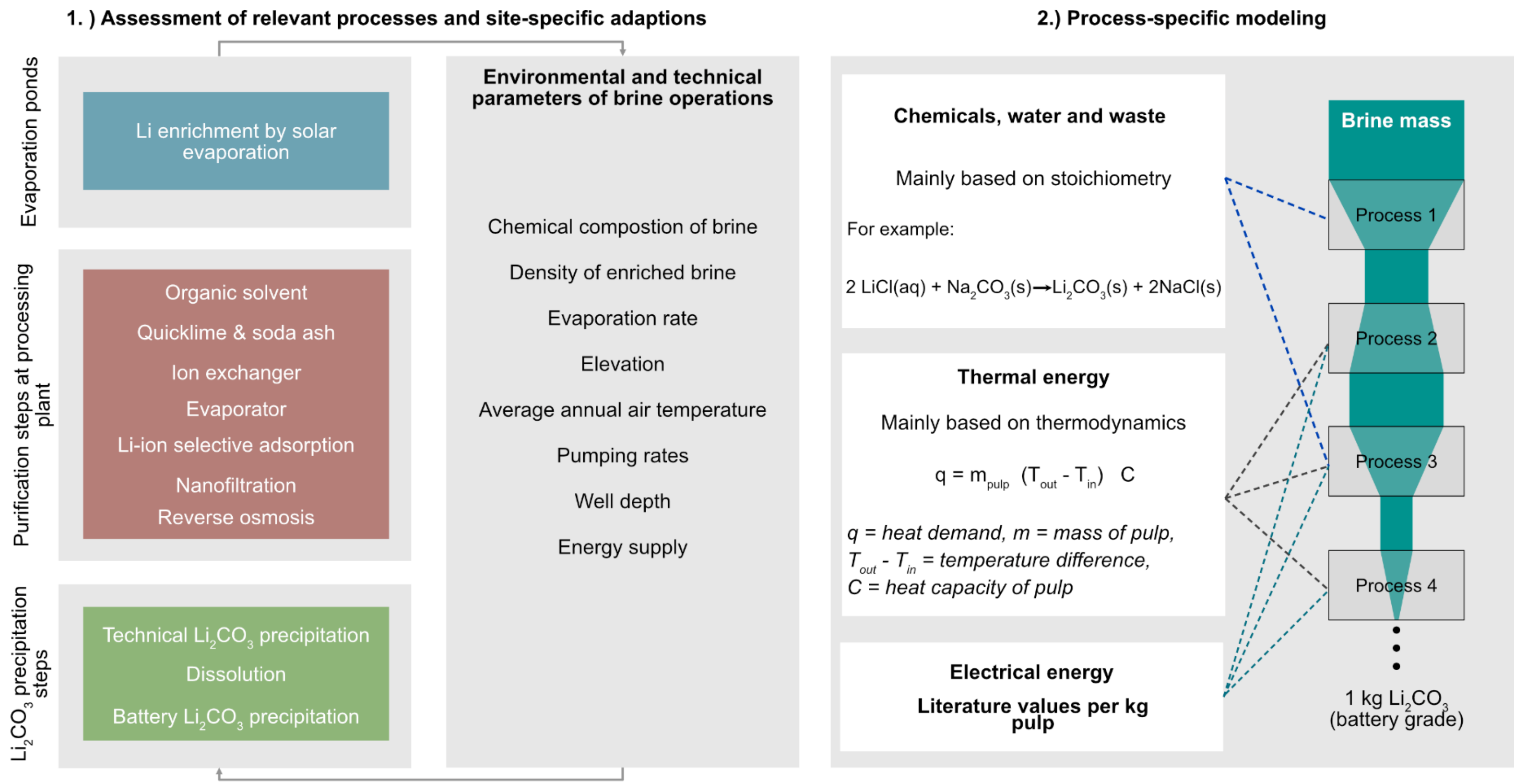


Figure 1: Proposed approach to model LCI for Li_2CO_3 production from brines.

Step 3a: Life cycle impact assessment

To assess environmental impacts of Li_2CO_3 (battery grade) production from each brine operation, we suggest to consider impacts on climate change (GWP 100a) (IPCC 2013), fully regionalized LCA impact assessment of fine PM formation (Oberschelp et al. 2020), and partly regionalized WSF based on AWARE (Boulay et al., 2018). The selection of impact categories (impacts on climate change and water scarcity) is based on existing literature (e.g., Stamp et al. 2012; Schomberg et al. 2021; Kelly et al. 2021). Since the energy requirement is mainly based on fossil fuels (Kelly et al., 2021), PM-related human health impacts should also be assessed, as it was shown in Oberschelp et al. (2019).

Step 3b: Sensitivity analyses

Monte Carlo simulations for brine operations allow to analyze the robustness of the results. Uncertainty distribution types (e.g., triangular or log-normal distributions) need to be carefully chosen. If many data are available, a random sampling of several datasets should be performed. Changing parameters based on physical relations could be a less preferred option to test the results. For the case of limited data, the data quality should be expressed by the Pedigree matrix described in Wernet et al. (2016).

3 Results and discussion

3.1 Application of the approach to present and future brine sites

To test our presented framework, the lithium extraction sites (Salar de Atacama, Salar de Olaroz, Salar de Cauchari-Olaroz, Salar del Hombre Muerto (North), Chaerhan salt lake) were environmentally assessed. Of the five selected sites, Salar de Atacama, Salar de Olaroz, and Chaerhan salt lake have been producing, whereas Salar de Cauchari-Olaroz is currently in the construction phase and plans to start Li_2CO_3 production in 2022 (S&P Global, 2021) while Salar del Hombre Muerto (North) is at an early exploration stage. For the latter, the start of mining activity and extraction technology is not yet clearly set (S&P Global, 2021), but a Li_2CO_3 production pathway has been suggested by Knight Piésold Ltd. and JDS Energy & Mining Inc. (2019).

This paper covers 70 % of the brine-related lithium carbonate production worldwide (sum of current production from the Salar de Atacama, Salar de Olaroz, and Chaerhan salt lake) (S&P Global, 2021). We also cover future production sites at Salar de Cauchari-Olaroz and Salar del Hombre Muerto North (reported and estimated production given in Table 1).

Step 1: Goal and scope

The goal is to quantify environmental impacts of Li_2CO_3 (battery grade) production from brine operations. First of all, this allows a comparison among the sites in terms of related environmental impacts. Additionally, the implications for LCA of Li-ion battery production can be assessed in high resolution. An attributional LCA with the cut-off allocation approach from ecoinvent was performed (Wernet et al., 2016). The functional unit was 1 kg Li_2CO_3 (battery grade). We used a cradle-to-gate approach. The system boundaries from pumping the brine to the surface until the final product (Li_2CO_3, battery grade) leaves the processing plant for the South American salt lakes. At Chaerhan salt lake, the brine is first sent to a K-fertilizer plant, and Li_2CO_3 production uses the effluent, which is considered as a waste stream and, therefore, without burden in the cut-off allocation approach. Since only Li_2CO_3 (technical grade) is produced (Gansu United testing services Co Ltd (2018), Lanke Lithium (2018)), we added the processes (dilution and re-heating the Li-bearing solution) required to manufacture Li_2CO_3 (battery grade). We further assessed the environmental impacts of Li-ion battery production. We incorporated our modeled LCIs of Chaerhan salt lake and Salar de Atacama in the Li-ion battery production based on ecoinvent v3.8 cut-off (ecoinvent, 2021). The functional unit was 1 kg of rechargeable Li-ion battery. Brightway 2 by Mutel (2017) and Activity Browser by Steubing et al. (2020) were used to conduct the assessment.

Step 2: LCI analysis

(1) Identification of relevant processes and parameters

We identified the relevant processes and their input demand for the chosen brine sites (Table 1, graphical illustration Figure 2). Required environmental and technical parameters are presented in the SI for each brine operation. Salar de Atacama, Salar de Cauchari-Olaroz, and Salar del Hombre Muerto (North) have similar general Li_2CO_3 production routes but with varying purification steps. While Salar de Olaroz uses ion exchangers to remove impurities from the pulp, Chaerhan salt lake uses Li-ion selective ion exchangers to adsorb Li.

Table 1 Differences in main processing techniques for the investigated brine operations. Production in metric tons of Salar de Atacama, Salar de Olaroz, and Chaerhan salt lake for the year 2018 are based on S&P Global (2021). Estimated production at Salar de Cauchari-Olaroz is given by Andeburg Consulting Services Inc and Montgomery & Associates (2019) and at Salar del Hombre Muerto (North) by Knight Piésold Ltd. and JDS Energy & Mining Inc., (2019).

Brine operation	**Processing techniques**
Salar de Atacama, Chile (88 100 t Li_2CO_3)	The brine is pumped into evaporation ponds to enrich Li from 0.15 wt. % to 6 wt. % and then transported to the processing plant (Garrett 2004). Subsequently, purification steps consist of organic solvent extraction to remove B and adding quicklime, respectively soda ash to remove Mg and Ca (Wilkomirsky, 1999). Then soda ash is added to the heated brine to let Li_2CO_3 (technical grade) precipitate (Kelly et al., 2021; Wilkomirsky, 1999). Li_2CO_3 (technical grade) is dissolved at low temperatures and re-heated to produce Li_2CO_3 (battery grade).
Salar de Olaroz, Argentina (12 000 t Li_2CO_3)	The pumped brine first reacts with quicklime and is then enriched from 0.06 wt. % Li to 1.2 wt. % Li in the evaporation ponds. In the processing

	plant, the brine reacts with soda ash to let impure Li_2CO_3 precipitate (Ehren and De Castro Alem, 2018; Orocobre, 2019). Then, Li_2CO_3 is dissolved in deionized water at low temperatures and the solution is sent through ion exchangers to remove residual Mg, Ca, and B. To precipitate Li_2CO_3 (battery grade), the pulp is re-heated as a last step (Ehren and De Castro Alem, 2018).
Salar de Cauchari-Olaroz, Argentina (40 000 t Li_2CO_3)	The pumped brine is enriched from 0.05 wt. % until 4 wt. % Li in evaporation ponds. Quicklime is added to remove Mg (Tran and Luong, 2015). B is removed via organic solvent extraction followed by removing Mg and Ca salts by adding quicklime and soda ash (Perez et al., 2014). The pulp is then heated to remove residual sulfates (Andeburg Consulting Services Inc and Montgomery & Associates, 2019). An evaporator is then used to decrease the volume of the Li-containing pulp, which is followed by an ion exchanger to remove any residual impurities (Andeburg Consulting Services Inc and Montgomery & Associates, 2019). In the next step, Li_2CO_3 (technical grade) is forced to precipitate by heating the pulp and adding soda ash. Subsequently, Li_2CO_3 is dissolved in water at low temperatures,and the solution is re-heated to produce Li_2CO_3 (battery grade) (Perez et al., 2014).
Salar del Hombre Muerto (North), Argentina (5 000 t Li_2CO_3)	A processing sequence similar to Salar de Atacama has been suggested (Knight Piésold Ltd. and JDS Energy & Mining Inc., 2019). As a first step, the brine would be enriched from 0.07 wt. % Li until 4 wt. % Li in evaporation ponds. Quicklime would be added to the evaporation ponds (Knight Piésold Ltd. and JDS Energy & Mining Inc., 2019). In the processing plant, purification steps would consist of B removal by organic

	solvent extraction, Mg and Ca removal by adding soda ash and quicklime. Then, Li_2CO_3 (technical grade) precipitates by heating the pulp and adding soda ash. Li_2CO_3 (technical grade) is then dissolved in water at low temperatures and re-heated to precipitate Li_2CO_3 (battery grade).
Chaerhan salt lake, China (8 000 t Li_2CO_3)	This Li_2CO_3 production plant uses the residual K-depleted pulp of a K fertilizers production plant. This pulp contains 0.022 wt. % Li (Lanke Lithium, 2018). The first step is a Li-adsorption technique, where alumina hydroxide is used as a resin to adsorb Li selectively. Deionized water is used to remove Li from the adsorbent again. The Li-containing solution is sent through ion exchangers to remove still existing impurities. Nanofiltration and reverse osmosis are then required to reduce the volume of the Li-containing solution (Wen et al., 2006). Solar evaporation continuously reduces the brine volume from which subsequently Li_2CO_3 (technical grade) is produced (Gansu United testing services Co Ltd, 2018; Lanke Lithium, 2018; Li et al., 2020). Due to the aforementioned required system expansion, we added the process sequence for Li_2CO_3 (battery grade).

(2) <u>Process-specific modeling of material and energy demands</u>

The resource demand of each process was calculated based on mass and energy balances as proposed by our approach. We calculated the mass required in the evaporation ponds to produce 1 kg Li_2CO_3 (battery grade) based on the reported Li concentration of the brine. For the evaporation ponds, waste production is not considered because the wastes from the different sites consist mainly of precipitated salts discarded in the near-by salt flats and are thus expected to have limited environmental impacts for the covered types of impact categories. The Li concentration of the enriched brine then served as an approximation of the mass going into the processing plant. Based on the calculated mass flow, chemicals were estimated based on stoichiometries. To account for the incompletion of the chemical reaction, 20 % mass is added to the modeled quicklime consumption based on Flexer et al. (2018) and 10 % to the modeled soda ash consumption (Li et al., 2020). 98.5 % of the organic solvent required in the B removal step is assumed to be recycled (CELIMIN, personal communication). Since deionized water is required in various processes, we estimated the required mass of water for each process. The brine operators reported water purification steps. However, they are not explicitly stated. Hence, for all brine operations we assumed that brackish water is treated by a reverse osmosis and an ion exchanger at all salt lakes (e.g., Lithium Americas Corp. 2019). Due to the lack of site-specific information regarding waste treatment at the processing plants, we did not include waste treatment in the LCI, in contrast to the existing dataset in ecoinvent v3.8 (ecoinvent 2021). Sources of thermal and electrical energy (i.e., heat from natural gas, heat and power co-generation from natural gas, and power from the location-specific grid) were based on company reports. Efficiencies of thermal processes are assumed to be 85 % due to the lack of information in used literature (U.S. Department of Energy Energy Efficiency and Renewable Energy, 2003).

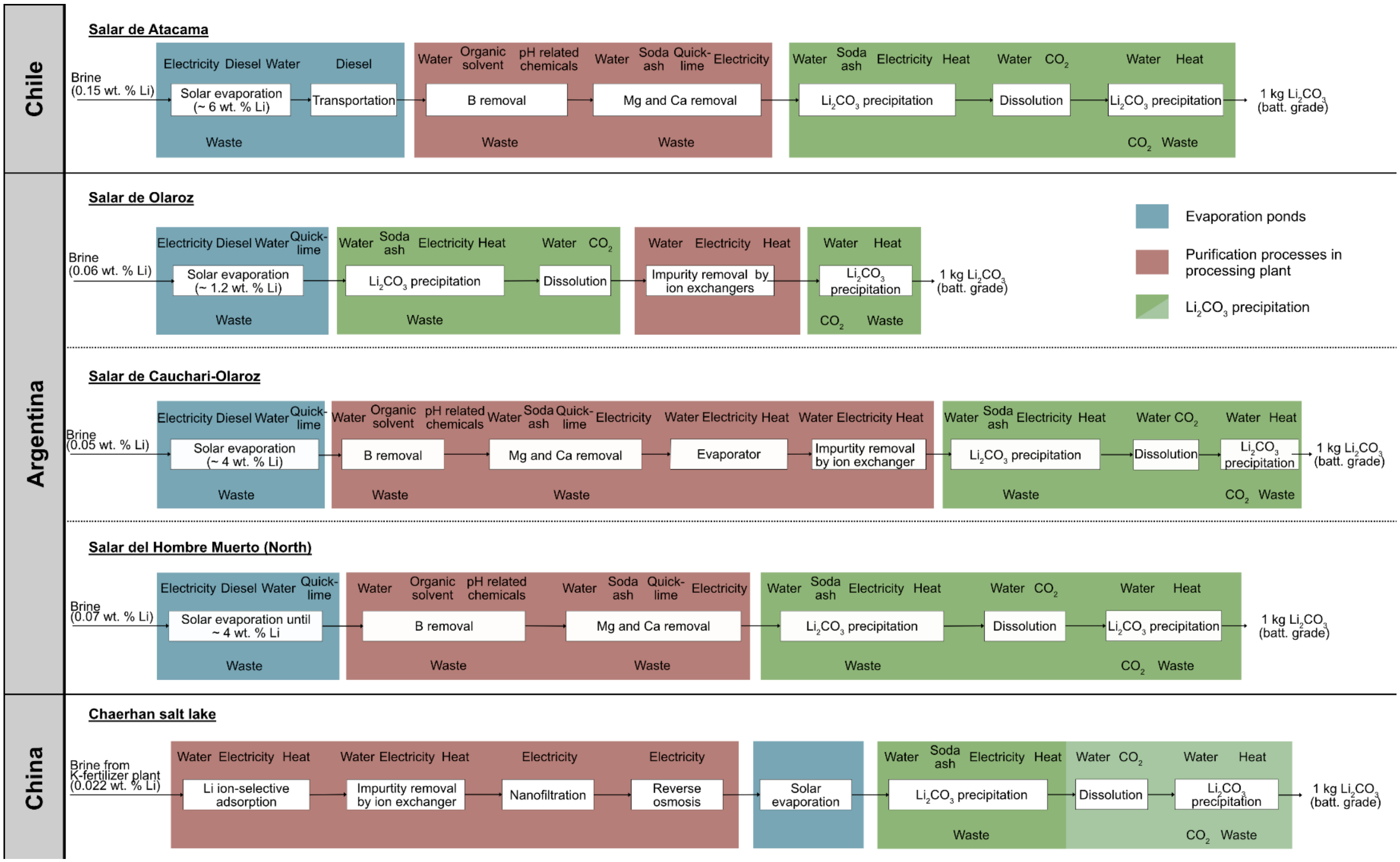


Figure 2 Process sequence of the five brine sites. The blue squares indicate that the processes are related to evaporation ponds, while red squares represent purification steps in the processing plant. Green squares represent Li_2CO_3 precipitation steps. Bright green squares within the processing sequence of Chaerhan salt lake indicate the required system expansion.

Modeled resource consumption

Table 2 presents the modeled inputs per kg Li_2CO_3 (battery grade) from the five sites. We find that Chaerhan salt lake has the highest consumption in electricity (27.8 kWh/kg Li_2CO_3), heat (298 MJ/kg Li_2CO_3), and water (474 kg/kg Li_2CO_3) due to the specific Li-ion adsorption, ion exchangers and the following nanofiltration, respectively reverse osmosis used for purification. Water (219 kg/kg Li_2CO_3) and electricity (1.5 kWh/kg Li_2CO_3) demand at Salar de Olaroz are higher than the other South American salt lakes. The water and electricity consumption originates from the intensive use of ion exchangers for removing impurities and re-generating ion exchangers. Since Salar de Olaroz relies on removing impurities by ion exchangers, Salar de Olaroz does not require any B removal-related chemicals in contrast to Salar de Atacama, Salar de Cauchari-Olaroz, and Salar del Hombre Muerto (North). Quicklime demand in the evaporation ponds to remove Mg is highest at Salar de Olaroz, while Salar de Atacama requires the lowest quicklime demand per kg Li_2CO_3. At Salar de Atacama, quicklime is only required to remove residual Mg from the pulp in the processing plant. Soda ash is used at all sites, mainly due to the Li_2CO_3 precipitation step.

Table 2 Modeled life cycle inputs for 1 kg Li_2CO_3 (battery grade) production at selected salt lakes.

Input demand/kg Li_2CO_3	*Salar de Atacama*	*Salar de Olaroz*	*Salar de Cauchari-Olaroz*	*Salar del Hombre Muerto*	*Chaerhan salt lake*
Electricity [kWh]	0.4	1.5	0.7	0.8	28
Heat [MJ]	19	19	28	14	298
Water [kg]	38	219	46	43	474
Quicklime [kg]	0.04	4.1	2.7	3.1	-
Sodium hydroxide [kg]	0.06	-	0.35	0.08	-
Organic solvent [kg]	0.04	-	0.1	0.7	-
Hydrochloric acid [kg]	0.10	-	0.5	0.9	-
Soda ash [kg]	1.9	1.6	2.1	1.6	1.6

Step 3a: Life cycle impact assessment

To assess environmental impacts of Li_2CO_3 (battery grade) production from each salt lake, GWP 100a (IPCC 2013), globally regionalized LCA impact assessment of PM formation (Oberschelp et al. 2020), and partly regionalized WSF based on AWARE (Boulay et al., 2018) were chosen.

Climate change impacts

Figure 4-A presents the climate change impacts to produce 1 kg Li_2CO_3 (battery grade) and their causes. In addition, we compare our results with two datasets (for Li_2CO_3 from brines and spodumene-bearing pegmatites) provided in ecoinvent v3.8 (ecoinvent, 2021).

We find that Li_2CO_3 production from the Salar de Atacama has the lowest climate change impacts (3.4 kg CO_2eq/kg Li_2CO_3). Heat (41 % of the climate change impacts) and on-site chemicals use (38 %) are the predominant contributors to the overall impacts. Soda ash for Li_2CO_3 precipitation is responsible for 31 % of the total climate change impacts, while other chemicals, like organic solvents, quicklime, and hydrochloric acid, only contribute minor shares. Hence, we find that Li_2CO_3 (technical grade) precipitation followed by Li_2CO_3 (battery grade) precipitation are the major processes contributing to climate change per kg Li_2CO_3 (battery grade) at Salar de Atacama (Figure 3). This is also in accordance with the findings by Stamp et al. (2012) and Kelly et al. (2021) when assessing environmental impacts related to the Li_2CO_3 production at the Salar de Atacama.

Climate change impacts related to the Argentinian brines are up to 235 % higher than for Li_2CO_3 extracted from the Salar de Atacama (Salar de Olaroz: 7.4 kg CO_2eq/kg Li_2CO_3; Salar de Cauchari-Olaroz: 7.7 kg CO_2eq/kg Li_2CO_3; Salar del Hombre Muerto (North): 8 kg CO_2eq/kg Li_2CO_3). Quicklime required in the evaporation ponds to remove impurities contributes to the total climate change impacts (Salar de Olaroz: 67 % of the total impact; Salar de Cauchari-Olaroz: 43 %; Salar del Hombre Muerto (North): 49 %), while heat and electricity on-site contribute to a minor extent. Quicklime production, in general, is associated with significant GHG emissions (Eula 2014). First, the production is heavily energy-intensive and mainly based on fossil fuels. Second, the chemical reaction to produce quicklime from limestone emits CO_2 as a co-product (European Lime Association, 2014). Those two factors are reflected in the overall impacts if quicklime is used in these evaporation ponds (Figures 3 and 4-A). Hence, evaporation ponds and Li_2CO_3 (technical grade) precipitation are the most critical contributors due to the usage of quicklime, respectively soda ash for the Argentinian salt lakes. At Salar del Hombre Muerto (North) and Salar de Cauchari-Olaroz specifically, the B removal step additionally makes up a significant share of climate change impacts.

We find that Li_2CO_3 from Chaerhan salt lake has by far the highest climate change impacts (31.6 kg CO_2eq/kg Li_2CO_3) resulting from the heat and electricity demand for the Li-ion selective adsorption technique. This technique mainly includes two phases (adsorption and desorption phase) which require the solution being heated up to a specific temperature using natural gas. Furthermore, electricity from the provincial electricity grid is required for the ion exchangers and membrane separation (nanofiltration and reverse osmosis) to remove impurities in the Li_2CO_3 bearing solution.

Climate change impacts of Li_2CO_3 from brines in ecoinvent v3.8 (2.1 kg CO_2eq/kg Li_2CO_3) and a recent study by Kelly et al. (2021) (2.7 – 3.1 kg CO_2eq/kg Li_2CO_3) are in the same range as the ones of our modeled LCI for Salar de Atacama. However, these numbers are lower than the other brines in Argentina and China, underestimating the climate change impacts of average Li_2CO_3. If Li_2CO_3 is extracted from spodumene-bearing pegmatites, as described with the dataset provided by ecoinvent v3.8 (ecoinvent, 2021), the climate change impacts add up to 10.7 kg CO_2eq/kg Li_2CO_3. Kelly et al. (2021) estimated 20.4 kg CO_2eq/kg Li_2CO_3 from the Australian pegmatitic mine. Both estimations are higher than our results for the Argentinian salt lakes but still lower than the one for Chaerhan salt lake.

Figure 3 Process-related contributional analyses regarding climate change impacts assessed with GWP (100 years) shown in percentage. The blue squares indicate that the processes are related to evaporation ponds, while red squares represent purification steps in the processing plant. Green squares indicate Li_2CO_3 precipitation steps. Bright green squares within the processing sequence of Chaerhan salt lake indicate that processes were added to hypothetically produce Li_2CO_3 (battery grade) to provide the same functional unit as the other systems.

PM-related human health impacts

The results indicate a large variability in PM-related health impacts (Figure 4-B), which underlines the necessity to perform regionalized LCAs for brine operations in this impact category. The highest PM health impacts occur due to the Li_2CO_3 production at Chaerhan salt lake (1.2 micro-disability adjusted life years (µDALY)/kg Li_2CO_3) followed by Salar del Hombre Muerto (0.67 µDALY/kg), Salar de Cauchari-Olaroz (0.67 µDALY/kg), Salar de Atacama (0.43 µDALY/kg), and Salar de Olaroz (0.31 µDALY/kg). For all sites, the background processes predominantely contribute to PM health impacts in contrast to foreground processes (i.e., heat or diesel consumption) from the remote location of all salt lakes and low local population densities. Electricity use in China, India, and Indonesia for various products and services in the background system makes up a significant share of the overall impacts of all brine sites (Salar de Atacama: 31 %; Salar de Olaroz: 44 %; Salar de Cauchari-Olaroz and Salar del Hombre Muerto (North): 56 %; Chaerhan salt lake: 48 %). The contribution of electricity required or generated on-site varies largely for PM health impacts. While the Argentinian electricity mix contributes to the overall PM health impacts with less than 1 %, the Chilean and Chinese electricity mix significantly contribute (27 % and 17 %) due to particulates, < 2.5 µm, sulfur dioxide, and nitrogen oxides from coal power generation. 43 % of the Chilean electricity mix comes from coal power. The Qinghai province-specific electricity mix, which was chosen for Chaerhan salt lake, consists of 22 % coal power, while the main source in this region is hydropower. In contrast, the Argentinian electricity mix includes little coal power (1 %) and does not significantly contribute to the PM health impacts.

Soda ash is a relevant contributor to the PM health impacts at all sites. The specific contributions range from 4 % at Chaerhan salt lake to 16 % at Salar de Atacama. During soda ash production, ammonia is released into the atmosphere contributing significantly to PM health impacts in highly populated areas, such as Europe. However, the location of soda ash production is highly uncertain since our results rather reflect the LCI in ecoinvent v3.8 than the actual resource supplier due to missing operational data.

Water scarcity footprint

The WSF for each salt lake using the AWARE method (Boulay et al., 2018) is presented in Figure 4-C. Chaerhan salt lake has the highest impact on water scarcity (35.25 $m^3_{world\ eq}$/kg Li_2CO_3), followed by Salar de Atacama (4.77 $m^3_{world\ eq}$/kg Li_2CO_3). The Argentinian salt lakes are in the same range (1.36 $m^3_{world\ eq}$/kg Li_2CO_3 at Salar del Hombre Muerto, 1.62 $m^3_{world\ eq}$/kg Li_2CO_3 at Salar de Cauchari-Olaroz, and 1.73 $m^3_{world\ eq}$ /kg Li_2CO_3 at Salar de Olaroz). The water scarcity impacts of Salar de Atacama predominantely originate from the direct use of freshwater at the processing plant (81 %). However, Salar de Atacama has the lowest water demand on-site compared to all other salt lakes. Nevertheless, due to its high aridity (e.g., Munk et al. (2016)) the location-specific characterization factor is the highest with 94.7 $m^3_{world\ eq}/m^3$ amongst these salt lakes, which is reflected in the overall water scarcity impacts.

The Argentinian brines have the lowest WSF due to their relatively low characterization factor (2.7 – 5 $m^3_{world\ eq}/m^3$). This is particularly important for Li_2CO_3 production at Salar de Olaroz, which has a high water requirement due to the intensive use of ion exchangers in the foreground system. The water demand originates from the regeneration of the resin used to remove residual impurities and then allow a Li_2CO_3 (battery grade) precipitation. Nevertheless, the overall impact is lower than the one from Salar de Atacama. In contrast to Salar de Olaroz, water scarcity impacts in the foreground are minor compared to the ones in the background at Salar de Cauchari-Olaroz (85 %) and Salar del Hombre Muerto (North) (91 %).

The WSF from Li_2CO_3 production at Chaerhan salt lake originates from the extensive water use (see chapter resource consumption) in the processing plant due to the Li-ion specific adsorption technique. Furthermore, the location-specific characterization factor (70.6 $m^3_{world\ eq}/m^3$) contributes to the relatively high WSF. The water demand in the background only accounts for 5 % of the total WSF. In general, it has to be noted that the background water consumption was not allocated to specific regions and was assessed with the global average AWARE characterization factor, which is rather high (Boulay et al., 2018). Therefore, background water stress might be overestimated in some cases.

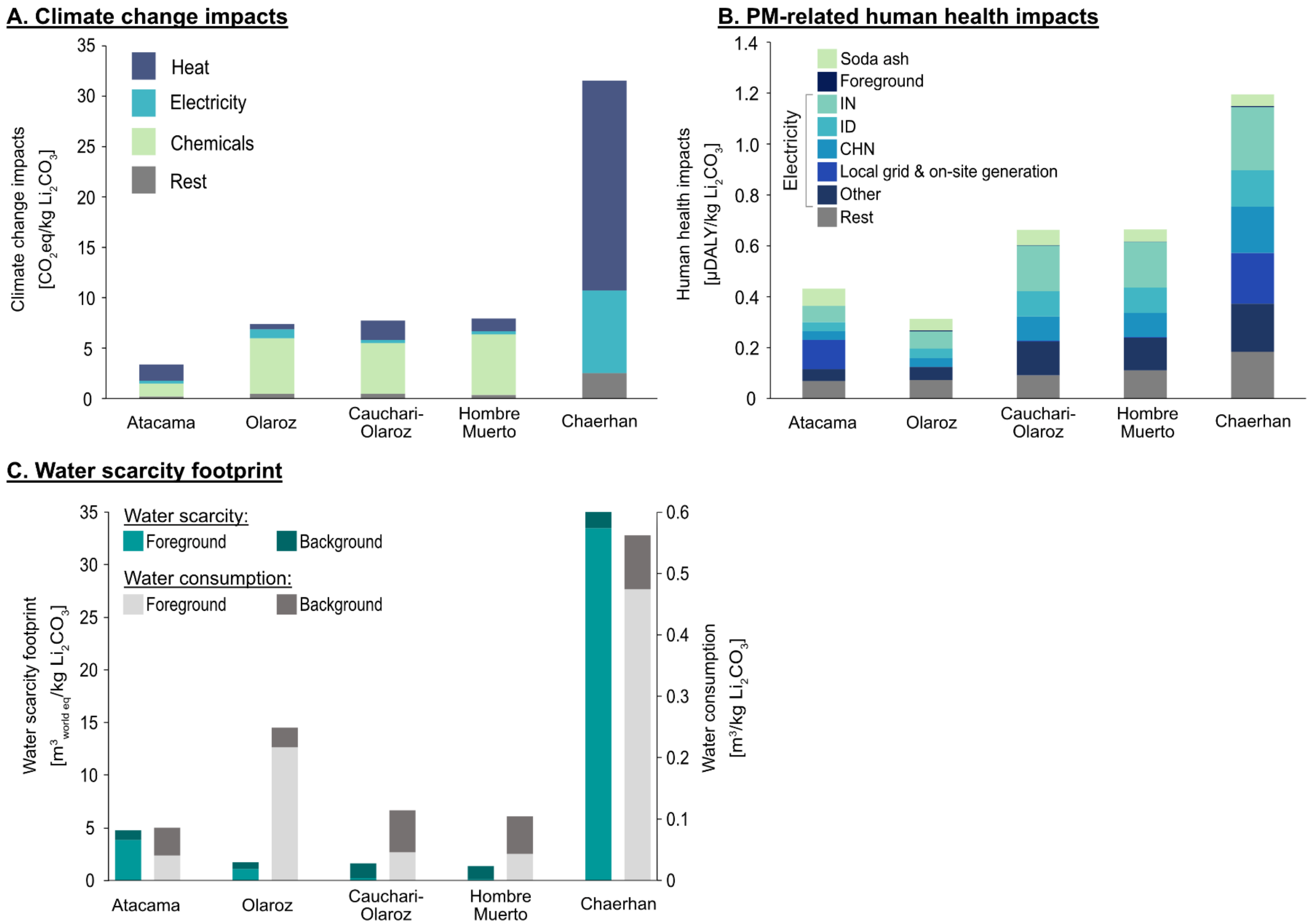


Figure 4 Environmental impacts of Li_2CO_3 production from brines. (A) Impacts on climate change (GWP 100 yrs), (B) PM-related human health impacts and (C) WSF (primary y-axis), and water consumption (secondary y-axis). (A) and (C) are shown on a midpoint level and (B) on an endpoint level.

Step 3b: Robustness and limitations of the approach

We ran Monte Carlo simulations with n = 5000 runs for the foreground system of each salt lake using GWP (IPCC 2013) and PM health impacts (Oberschelp et al. 2020) to assess the robustness of our results (more information in the SI). The relative standard deviation ranges from ± 33 % (Salar de Atacama) to ± 98 % (Chaerhan salt lake) for impacts on climate change, while the Argentinian salt lakes are in-between. For PM-related health impacts, the relative standard deviation lies between ± 46 % (Salar de Cauchari-Olaroz and Salar del Hombre Muerto (North)) and ± 73 % (Chaerhan salt lake). The relatively higher standard deviation of Chaerhan salt lake is explained by the lowest data quality of all assessed sites. In general, the origin of inputs is mostly unknown, thus contributing to the uncertainties. We, therefore, relied on ecoinvent data. In order to decrease these uncertainties, site-specific information regarding the input supply is crucial. If these data are not available, country-specific trading data could be obtained to decrease the uncertainties in the future.

There are also data gaps in our modeling approach which need to be discussed. Energy, water, and chemical demand modeled for this study were compared with annually reported company data or technical reports from exploration activity (more information in SI). We used company data as an indicator rather than as a fixed reference because specific boundaries and further documentation were generally not provided. Quantitative chemical demand at Salar de Cauchari-Olaroz has not been reported to the authors' knowledge and thus, could not be compared with our results. Furthermore, the LCI modeling of Li_2CO_3 (battery grade) from Chaerhan salt lake predominately relies on parameters reported in construction plans by Gansu United testing services Co Ltd (2018), impeding to test robustness of the model for that site. Annual changes of the brine chemistry are challenging to include in the LCI, but may affect resource consumption (especially chemical and heating demand) on-site. We could not estimate the salt crystallization sequence and hence, calculate waste production in evaporation ponds. However, these wastes are mainly deposited on-site, requiring little transport.

3.2 Implications for LCA of Li-ion battery production

To set this study into a broader context, modelled LCIs of two brine sites (Chaerhan salt lake and Salar de Atacama) were implemented in an ecoinvent v3.8 dataset, which represents the production of 1 kg rechargeable Li-ion batteries. This type of battery is used for a variety of electrical vehicles (Crenna et al., 2021). Furthermore, we also adjusted the ecoinvent dataset to only use Li from the brine dataset in ecoinvent v3.8. This leads to three battery datasets:

1. 100 % of the Li_2CO_3 production originates from the dataset for Li_2CO_3 from brines in ecoinvent v3.8 (based on Salar de Atacama).
2. 100 % of the entire Li_2CO_3 production is assumed to be from Salar de Atacama.
3. 100 % of the entire Li_2CO_3 production is replaced by Li_2CO_3 from Chaerhan salt lake.

As already Stamp et al. (2012) and Kelly et al. (2021) highlighted, the source of lithium affects the amount of GHG emissions related to Li-ion battery production. The maximum increase of climate change impacts is 19 % when implementing 100 % Li_2CO_3 production by Chaerhan salt lake (dataset 3) compared to the baseline (dataset 1). The leading cause is the high thermal and electrical energy demand based on fossil fuels of Chaerhan salt lake. Li_2CO_3 from Salar de Atacama (dataset 2) only increases climate change impact of a Li-ion battery by < 1 %. In the future, more lithium production might be sourced from high-impact mines since an increase in demand and price might make energy-intense production routes profitable. Multiple studies emphasize the range of reported or modeled GHG emissions related to Li-ion battery production (e.g., Raugei and Winfield 2019; Crenna et al. 2021). For our study, these findings indicate that the overall share of Li_2CO_3 from brines regarding impacts may change in the future due to more detailed and transparent Li-ion battery supply chains. Thus, LCA and carbon footprinting of future battery production should consider the potential for high-impact Li_2CO_3 supply and consider the existing LCIs as highly uncertain. Furthermore, it is vital to communicate these findings appropriately to a non-scientific audience, which could be done in future work by following the recently developed recommendations in Salemdeeb et al. (2021).

4 Conclusion and outlook

The Li supply is key for the transition towards a global decarbonized society. Li expects higher growth rates than other metals in the future. Hence, it is inevitable to assess environmental impacts of current and future Li extraction to avoid severe environmental burden shifting. Currently used LCIs of Li_2CO_3 from brines do not represent the global market nowadays and even less for the future. Thus, we developed a framework to update LCIs of Li_2CO_3 production from brines site-specifically. This methodology was applied to existing and future production sites. Our framework helps to treat data gaps and to derive process-specific parameters from patents. Furthermore, an approach to assess sites using waste streams as a Li source, like Chaerhan salt lake, was developed. Regionalization of foreground and background data as well as using regionalized impact assessment methods were examined and discussed in detail. Our results demonstrate the necessity of defining a framework to assess various sites and resulting variabilities in global production. The case studies show that available literature data underestimate environmental impacts of Li_2CO_3 production from brines. This mainly is a consequence of only assessing Li_2CO_3 production at Salar de Atacama and assuming that this data is representative of Li_2CO_3 production in general, which is not the case. Furthermore, the variability of our results is a consequence of the brine composition, the applied processing technique, and the brine location. For instance, water scarcity and PM impacts need to be site-specifically assessed since the location of impact (background or foreground) varies among these sites. Future improvements regarding the assessment of Li should focus on other Li sources, such as pegmatites, geothermal brines, and seawater.

The integration of obtained LCIs in Li-ion batteries demonstrates that the overall impacts on climate change increase to up to 19%. Hence, supply chains of Li-ion batteries need to be assessed in detail, especially for future scenarios. Improvements regarding the resolution of supply chains are crucial to transition towards low-carbon technologies sustainably. This includes site-specific assessment of other minerals for batteries like Aluminum and Cobalt, such as recently done for copper mine tailings (Adrianto et al., 2022). Our framework serves as a starting point for enhancing LCI and regionalized LCIA of other battery minerals.

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Declaration of interest

The authors declare no competing interests.

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