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Is lithium from geothermal brines the sustainable solution for Li-ion batteries?

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ABSTRACT

The rising demand for Li, paramount for energy storage, necessitates expanded supply. As the supply is concentrated in a few countries, this poses supply chain risks for Li-ion battery makers. To diversify suppliers, alternative Li ore deposits such as geothermal brines are being explored. However, Li extraction from geothermal brines is challenging due to the unique chemistry and elevated temperatures. Since Li-extraction from geothermal brines is in its infancy, data availability and quality are still poor, hampering life cycle assessments. Hence, this study provides a parametrized life cycle inventory model of Li carbonate production from geothermal brines. The model accounts for site-specific environmental conditions and technological features. Life cycle impacts at the Salton Sea in the US (1686 cases) and the Upper Rhine Graben in Germany (1982 cases) are quantified. The high case numbers are chosen to mitigate the high uncertainties in input parameters. Specifically, the brine chemistry, adsorption yield, drilling required and energy inputs are varied. Climate change impacts of selected cases vary within 18–59 kg CO_2eq/kg Li carbonate at the Salton Sea and within 5.3–46 kg CO_2eq/kg Li carbonate at the Upper Rhine Graben, compared to 2.1–11 kg CO_2eq/kg Li carbonate in existing ecoinvent data sets. The wide range of potential impacts underscore the necessity of early-stage assessments of the technologies. In case of high drilling demand and use of fossil energy, climate change impacts of Li-ion batteries using Li carbonate from geothermal brines can increase by 30–41 % compared to literature values.

1. Introduction

The mitigation of climate change requires the implementation of Li-ion batteries as a core technology for energy storage [1]. However, the growing metal demand is in conflict with current production volumes [2,3]. One of the most relevant metals is Li, classified as a critical raw material in the United States and the European Union [4,5]. On a global level, Li is mined from pegmatites and brine deposits in Australia, Chile, China and Argentina, with the expectation for these sources continuing to lead the global supply in the future [6–9]. To reduce the risk of Li supply chain disruptions for the US and Europe, domestic Li deposits such as geothermal brines have gained increasing interest [10]. They do not only contain significant Li resources but also provide geothermal energy that can be used concurrently [11–14].

Geothermal brines with elevated Li concentrations (100–400 mg/L) exist in the Salton Sea in California [15] and the Upper Rhine Graben in Germany and France [13,16]. These brines have been used for geothermal plants in the past. Before re-injecting the brine into the reservoir, it can be sent to an additional processing sequence to extract Li and produce either Li carbonate (Li_2CO_3) or Li hydroxide monohydrate ($LiOH{\cdot}H_2O$) [11,17].

Due to the combination of complex brine chemistry, elevated temperature and pressure, Direct lithium extraction (DLE) is suggested for lithium extraction from geothermal brines. Assessments of DLE from geothermal brines in the US and Germany claimed economic feasibility [18–20]. The key to this technology is the selective recovery of Li from the brine. Different types of DLE (e.g. liquid–liquid extraction,

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Abbreviations: GHG, greenhouse gas; DLE, direct lithium extraction; LCA, life cycle assessment; LCI, life cycle inventory; LCIA, life cycle impact assessment; PM, particulate matter; $PM_{2.5}$, particulate matter with a size below 2.5 micrometer; Li_2CO_3, Lithium carbonate; $LiOH.H_2O$, Lithium hydroxide monohydrate; NMC811, Li-ion battery type (cathode: 80 % Nickel, 10 % Manganese, 10 % Cobalt); DALY, disability adjusted life years; mg/L, milligram per Liter; kg, kilogram; wt. %, weight percent; L/s, Liter per second; η, adsorption yield (in %); m, meter; a, years; m^3, cubic meter; $m^3_{\text{world-eq.}}$, cubic meters world equivalent; kg CO_2eq, kilogram carbon dioxide equivalent; kWh, kilowatt hour

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Li-ion selective extraction, electrochemical technologies) have been studied, mainly at laboratory scale [11,21–23]. A comparison of these technologies revealed that difficulties in up-scaling exist for all [23]. However, the authors highlight that in particular Li-ion selective adsorption is technically feasible according to the prevailing technology readiness level. The brine needs to be treated to remove unwanted ions (e.g., Fe, Si, Mn, Zn) before extracting Li from the brine. The geothermal brine is sent through adsorption columns storing resin to specifically adsorb Li. Water and/or acids are required to desorb the Li and produce a Li-containing solution. Residual impurities are removed from the Li-containing solution by precipitation reactions and/or ion exchangers [24]. The solution is significantly reduced by reverse osmosis and mechanical evaporation. If Li_2CO_3 is produced, soda ash is added to precipitate the Li_2CO_3 at technical grade (min. 99 wt.%) which needs further purification to obtain battery grade (min. 99.5 wt.%) [21,24,25]. If $LiOH{\cdot}H_2O$ is produced, electrolysis is used [24]. Since DLE is in its infancy, comprehensive assessments of DLE are currently limited – including the environmental perspective [17].

Another reported challenge of Li extraction from geothermal systems is the uncertain development of the Li concentration over time. While geothermal boreholes are commonly optimized to ensure stable heat extraction over a long time, the Li concentration in the brine could potentially decrease earlier. At the Upper Rhine Graben, a drop in Li concentration in the well would be observed after 5–10 years of extractive operation, while the heat would remain unaffected [26]. Therefore, drilling efforts to maintain elevated Li concentrations are expected to be higher; however, there is no practical evidence of this so far. This complicates a reliable evaluation of the economic and environmental aspects of the Li production life cycle.

Life cycle assessment (LCA) is a method to quantify environmental impacts of a product or service. LCAs of Li mining have mainly been performed considering currently producing sources (ores and salar/salt lake-related brines) [27–30]. With regard to Li mining from geothermal brines, LCA studies of the Salton Sea and the Upper Rhine Graben have been conducted [18,31]. However, both assessments lack transparency, completeness or standardized assumptions, which hamper the direct comparison between the two sites as well as with other studies. A holistic and transparent assessment including all relevant processes, to improve the understanding of related environmental impacts and to allow for a comparison among geothermal sites, remains absent. As Li extraction from geothermal brines is still not performed at industrial scale, potential uncertainties regarding up-scaling need to be taken into account when assessing the environmental impacts. Hence, the objective of this study is to transparently evaluate the life cycle environmental impacts of Li_2CO_3 production from geothermal brines by including the crucial technological features and environmental conditions that have not been considered thus far. To achieve this, LCI are modelled and the environmental impacts of Li_2CO_3 production from geothermal brines are assessed. This study assesses implications caused by varying brine chemistry and the impact of drilling, which will become necessary if Li concentrations decrease over time. To define realistic conditions, Li_2CO_3 production from the Salton Sea and the Upper Rhine Graben is investigated and extended with scenario results. Climate change impacts, fully regionalized human health impacts from fine particulate matter formation and fully regionalized water scarcity impacts are assessed in this study. The results are compared with existing literature data. Finally, the implications of Li_2CO_3 from geothermal brines for Li-ion battery LCAs are presented and discussed.

2. Methods

LCA is used to determine the life cycle impacts of a product, process or service from a bottom-up perspective [32]. By including resource consumption and emissions, this systematic method allows a detailed assessment of a broad range of environmental impacts throughout all stages of a product's life cycle. LCA can highlight emission hot spots along the necessary supply chains of a product. According to ISO 14040:2006 and ISO 14044:2006, the LCA framework consists of four key steps: (i) goal and scope definition, (ii) LCI analysis, (iii) life cycle impact assessment (LCIA), (iv) interpretation of results [33,34]. The first step defines the boundaries of the assessed system and the selection of life cycle impacts (e.g., climate change impacts, water scarcity, acidification, land use, etc.). Subsequently, LCI analysis involves the systematic compilation of input (e.g. energy, materials, water) and output data (e.g. direct emissions into the environment, generated waste) linked with the evaluated life cycle stages. In the third step, life cycle impacts are quantified based on the obtained LCI and emission-specific characterization factors per chosen life cycle impact. The interpretation of the results is the last step of the ISO framework.

2.1. Goal and scope

This study follows an attributional LCA, using the ecoinvent data with the cut-off allocation approach [35]. The goal is to assess the environmental impacts of Li_2CO_3 (battery grade) production from geothermal brines in the US and Germany. Hence, 1 kg of Li_2CO_3 at battery grade is chosen as a functional unit. Since the geothermal brine for Li_2CO_3 production is used after the geothermal plant and is considered as a waste stream pumped underground, no burden from extracting and injecting the brine used in the geothermal plant is allocated to the final product in this study (cut-off allocation approach). The system boundaries are set from the geothermal brine outlet of the power plant to the Li_2CO_3 processing sequence until Li_2CO_3 (battery grade) is produced (gate-to-gate approach). Drilling activity is included in the system boundaries when assessing the implications of additional drilling for Li_2CO_3 production. Additional drilling activity to maintain the Li concentration of the brine is entirely allocated to Li_2CO_3. Finally, a functional unit of 1 kWh capacity is used to assess the overall contribution of Li_2CO_3 from geothermal brines to the climate change impacts of Li-ion batteries.

2.2. LCI modelling

LCI are modelled by a process-based approach, to allow for a detailed assessment of the system and the influences of brine chemistry and drilling activities. The following processes of Li_2CO_3 production are included in this study and are based on Featherstone et al. [24] (the following italic process-names refer to Fig. 1):

1. Pre-treatment of geothermal brine: Limestone is added to remove Fe- and Si-hydroxides (*Fe & Si removal*) and quicklime to precipitate Mn- and Zn-hydroxides (*Mn & Zn removal*) without Li losses. Due to the added limestone and quicklime, the pH is elevated and hence is then reduced by adding hydrochloric acid (*acidification*).
2. *Li-ion selective adsorption*: Li is selectively adsorbed by a cationic resin (Al-hydroxides) from the geothermal brine. Li is desorbed by using freshwater, and the resulting Li-containing solution is then sent to the next treatment step.
3. Pulp purification: Residual Ca and Mg ions in the solution are removed by adding sodium hydroxide and soda ash (*Ca & Mg removal*). Residual divalent ions (Ca, Mg, B) are removed by the *ion exchanger*.
4. Volume reduction: *Reverse osmosis* as well as a subsequent *triple evaporator* are required to reduce the solution volume and increase the Li concentration.
5. Li_2CO_3 (technical grade) precipitation: Soda ash is added to precipitate Li_2CO_3 at technical grade (*Li_2CO_3 precipitation*). *Washing* and de-watering by *centrifugation* of Li_2CO_3 are the subsequent steps.

Table 1
Energy provision cases for this study.

	Electricity	Heat	Steam
Geothermal electricity & heat (*Geo*)	Geothermal electricity provision	15 % of the data set for geothermal electricity production	Steam from geothermal power plant as a waste product
Natural gas and electricity mix (*Conv*)	Country-specific electricity mix	Natural gas	Steam from natural gas

6. Li_2CO_3 (battery grade) precipitation: Li_2CO_3 needs to be dissolved in water at low temperature (*dissolution*) and then reheated to precipitate Li_2CO_3 at battery grade (*Li_2CO_3 precipitation*). *Washing*, *centrifugation* and drying (*rotary dryer*) of Li_2CO_3 are the final steps.

Environmental parameters (e.g. brine chemistry, elevation, average annual air temperature) and technical parameters (e.g. pumped brine volume, adsorption yield, annual production) are used to quantify process-specific mass flows for the Salton Sea and the Upper Rhine Graben. For example, the Li concentration and adsorption yield determine the incoming mass flow of geothermal brine to produce 1 kg Li_2CO_3. A specific focus of this study is the LCI modelling of processes such as the pre-treatment, Li-ion selective adsorption as well as the pulp purification (details can be found in Table A.2). Li_2CO_3 precipitation and the following treatment are based on the modelling approach by Schenker et al. [30]. Each process has a set of pre-defined inputs (e.g., chemicals, water, energy) and outputs (e.g., waste, recycled water). Chemicals and waste are modelled by stoichiometry using the brine chemistry or process-specific literature values as proxies. Heating demand is modelled based on the operating temperatures and temperature of the incoming mass flow. Electricity is based on literature values per kg mass in the modelled process. For both locations, site-specific base cases are defined. The base case of the Salton Sea is defined by the reported Li concentration of 0.018 wt.% Li and a pumping rate of 437 L/s [11]. The base case of the Upper Rhine Graben is defined by the reported Li concentration of 0.019 wt.% Li [36] and a pumping rate of 80 L/s [22]. For both cases, an adsorption yield of 50% is used. This is the more conservative value in the reported range by Goldberg et al. [23]. This set and other required input parameters of both locations are summarized in Table A.1.

To get more insights into how case-specific assumptions influence LCI and the resulting environmental impacts, the following parameters are systematically varied:

1. Li concentration (affecting the overall estimated Li_2CO_3 production and drilling metre per kg Li_2CO_3): 0.001–0.03 wt.% Li. The highest and lowest modelled Li concentrations are chosen based on existing geochemical analyses from the Salton Sea [15] and the Upper Rhine Graben [16].
2. adsorption yield (η) (affecting the mass of Li adsorbed in the Li-ion selective adsorption process and hence the mass flows into the Li-ion selective adsorption unit): 30–100%. Goldberg et al. [22] emphasized that the adsorption yield at lab scale ranges between 50% and 90% based on Isupov et al. [37] and Jiang et al. [38]. In the model, the range is widened to cover further potential scenarios.
3. brine chemistry (affecting the process input and output of the pre-treatment steps i.e. Fe and Si removal, Mn and Zn removal, acidification): Geochemical analyses from Neupane et al. [15] and Kovacs et al. [16] are used to assess the influence of brine composition variability on environmental impacts.
4. drilling activity: As the Li concentration might decrease faster than the brine temperature, additional drilling might be needed to maintain sufficiently high Li concentrations. At the Salton Sea, the depth of the production and re-injection wells is reported to be between 500 and 1500 m [39]. To represent a conservative case, 1500 m is chosen in the model. At the Upper Rhine Graben, 3500 m is used as a drilling depth based on Nitsch et al. [40].

As a background system, ecoinvent v3.9.1 cut-off version is chosen [41]. Background LCI changes based on ecoinvent v.3.9.1 data are specifically implemented on the energy provision level, as shown in Table 1. The first one is a geothermal energy provision case (*Geo*). Regionalized data sets are used for geothermal electricity provision. Since no data set on heat provision of geothermal power plants exists in ecoinvent v3.9.1, the geothermal electricity data set with its electrical efficiency of 15% is scaled to an efficiency of 100% to represent geothermal heat supply. The second scenario (*Conv*) assumes that electricity is sourced from the country-specific grid and heat from natural gas.

Life cycle impact assessment

Brightway 2.0 is used to perform the LCIA [42]. Climate change impacts (Global Warming Potential 100a) are assessed by the IPCC 2021 method [43]. Regionalized impacts for water scarcity and fine particulate matter-related human health impacts are evaluated by AWARE [44] and Oberschelp et al. [45], respectively. PM-related human health impacts are expressed in disability-adjusted life years (DALY) which is composed of the premature mortality and the loss of health due to diseases. Water scarcity impacts are expressed in $m^3_{world-eq.}$ which describes the water scarcity impact of water use based on the water availability and the demand of that specific location/region.

3. Results and discussion

3.1. Life cycle inventories

One of the core aspects of this study is the modelling of process-based LCI. For illustration, Fig. 1 presents the modelled mass flows of the earlier defined base case for 1 kg of Li_2CO_3 at the Salton Sea (results of the Upper Rhine Graben are found in the supplement A). The input of geothermal brine amounts to 2102 kg per kg of dry Li_2CO_3 at battery grade. In the pre-treatment (i.e. Fe & Si removal, Mn & Zn removal and acidification), chemicals and water are added to precipitate impurities as waste. The mass flows until the Li-ion selective adsorption process remains relatively constant. After the Li-ion selective adsorption, the depleted geothermal brine is re-injected into the reservoir. Required water (2348 kg) to desorb Li from the resin is partially substituted with water produced from reverse osmosis (1174 kg) and the triple evaporator (978 kg). In total, 2348 kg of LiCl-containing solution is sent to the Ca & Mg removal step and the subsequent ion exchanger, which do not change the mass flow significantly. Regeneration of the ion exchanger is highly water intensive (2583 kg). The reverse osmosis and the triple evaporator decrease the inter-process mass flows from 2348 kg to 196 kg, resulting in a highly concentrated LiCl solution, as described in Featherstone et al. [24]. Soda ash slurry is added to the solution to precipitate Li_2CO_3 at technical grade. The generated liquid waste is discarded or partially sent back to the Mn & Zn removal step. Finally, 2 kg of wet Li_2CO_3 is sent to the subsequent processes to yield 1 kg of dry Li_2CO_3 at battery grade.

Fig. 2 demonstrates the resulting change of mass flows needed for 1 kg of Li_2CO_3 due to a decreased Li concentration at the Salton Sea. A declining Li concentration from 0.03 wt.% Li to 0.001 wt.% Li would raise the required primary amount of geothermal brine from 1252 kg

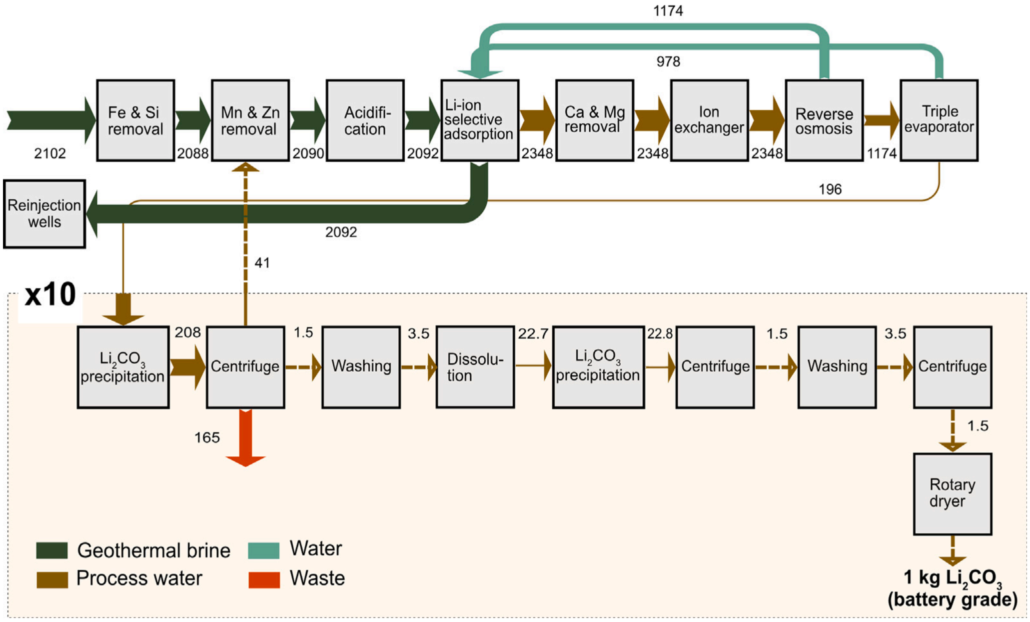


Fig. 1. Li_2CO_3 production at the Salton Sea. The base case is used to quantify the displayed mass flows in kg for 1 kg of Li_2CO_3 (data in Table B.3, additional details in Figure A.1).

to 37 570 kg per kg Li_2CO_3. The increase in mass flows also influences the inputs (i.e. required mass of limestone to precipitate Fe- and Si-hydroxides) and waste production of the pre-treatment phase. Fe & Si and Mn & Zn removal processes depend on the chemical composition of the geothermal brine. When lowering the Li concentration in the model, these concentrations (Fe, Si, Mn and Zn) remain fixed, and hence both the amount of required chemicals and the waste produced increase. On the other hand, the Li-ion selective adsorption process is not affected by the decreased Li concentration since the mass of the concentrated Li solution (=process output) is fixed to produce 1 kg Li_2CO_3 (i.e. the concentration of the solution is fixed as output of this step). Hence, a change of mass flows after the Li-ion selective adsorption process is not observed for 1 kg Li_2CO_3. When changing the adsorption yield of the Li-ion selective adsorption process, the mass flows upstream vary in a similar way as described for the varying Li concentration.

3.2. Life cycle impact assessment

The sections below present the LCIA results as a function of the uncertain input parameters to allow for a solid assessment with a spectrum of outcomes.

3.2.1. Li concentration and adsorption yield

Life cycle impacts of Li_2CO_3 production when varying the Li concentration and the adsorption yield are presented. First, the results are discussed for the lowest adsorption yield modelled (η = 30%) with different concentrations and geothermal energy provision (*Geo*).

As seen in Fig. 3, Li_2CO_3 from the Salton Sea generally has higher impacts in all impact categories than the Upper Rhine Graben. Regarding the dynamics of Li concentration, climate change impacts increase from 17.5 kg CO_2eq (0.03 wt.% Li) to 75.3 kg CO_2eq (0.005 wt.% Li) at the Salton Sea. Below 0.005 wt.% Li, climate change impacts escalate, and they result in a 4.7-fold increase (up to 353 kg CO_2eq/kg Li_2CO_3 for 0.001 wt.% Li). At the Upper Rhine Graben, the climate change impacts are considerably lower (e.g. 5.3 kg CO_2eq/kg Li_2CO_3 at 0.03 wt.%) but show the same overall trend with a maximum of 24.4 kg CO_2eq/kg Li_2CO_3. When raising the adsorption yield from 30% to 90% for 0.03 wt.% Li, climate change impacts at the Salton Sea are reduced by 44%, while the impacts are only decreased by 8% at the Upper Rhine Graben. Assuming the lower Li concentrations at both locations, the reduction potential of an increased adsorption yield becomes more apparent. The same dynamics are observed for $PM_{2.5}$-related human health and water scarcity impacts of both locations (Table B.7 and B.11).

The magnitude of the impact and the implications of declining Li concentrations substantially differ between the sites. The impurity concentration, which requires enhanced treatment at the Salton Sea, is higher than at the Upper Rhine Graben. Hence, pre-treatment processes and the related supply chains (i.e. waste disposal from Fe & Si removal and Mn & Zn removal; production of hydrochloric acid used in acidification) are more relevant in the case of the Salton Sea. In general, environmental impacts are relatively stable until a low Li concentration (0.01 wt.% Li) and at varying adsorption yield for both locations. This is a main result of the Li-ion selective adsorption step in the presented model. The amount of required water to desorb Li is not dependent on the initial brine chemistry but rather on the Li-adsorption capacity of the resin. Hence, the output of that process is a fixed mass of LiCl solution (Fig. 2). By decreasing the Li concentration and adsorption yield, only the pre-treatment processes are affected in terms of mass flows. Thus, the pre-treatment processes become more relevant as the required inputs and generated outputs increase, with more brine being treated to obtain 1 kg Li_2CO_3. Lower Li concentrations can also lead to lower adsorption yields as the likelihood of the Li finding a suitable adsorption site decreases and the average distance of Li and adsorption sites increases in the adsorption columns. Furthermore, lower Li concentrations at stable trace impurity concentrations can increase the competition for the same types of adsorption sites. As highlighted by Goldberg et al. [22], adsorption yields between 50 and 90% could be achieved by long storage times in the Li-ion selective adsorption process. If Li concentrations decrease, more geothermal brine would need to be treated, leading to shorter treatment duration and potentially lower adsorption yields.

3.2.2. Energy provision

Changing the energy provision from geothermal power and heat (*Geo*) to country-specific electricity mix and natural gas as a heat supply (*Conv*) for Li_2CO_3 production elevates climate change impacts by a factor of 4.2 at the Salton Sea and 8.2 at the Upper Rhine Graben for a given Li concentration. This leads to similar climate change impacts from both sites. For example, Li_2CO_3 from a geothermal brine with 0.03 wt.% Li and adsorption yield of 30% results in 49.5 kg

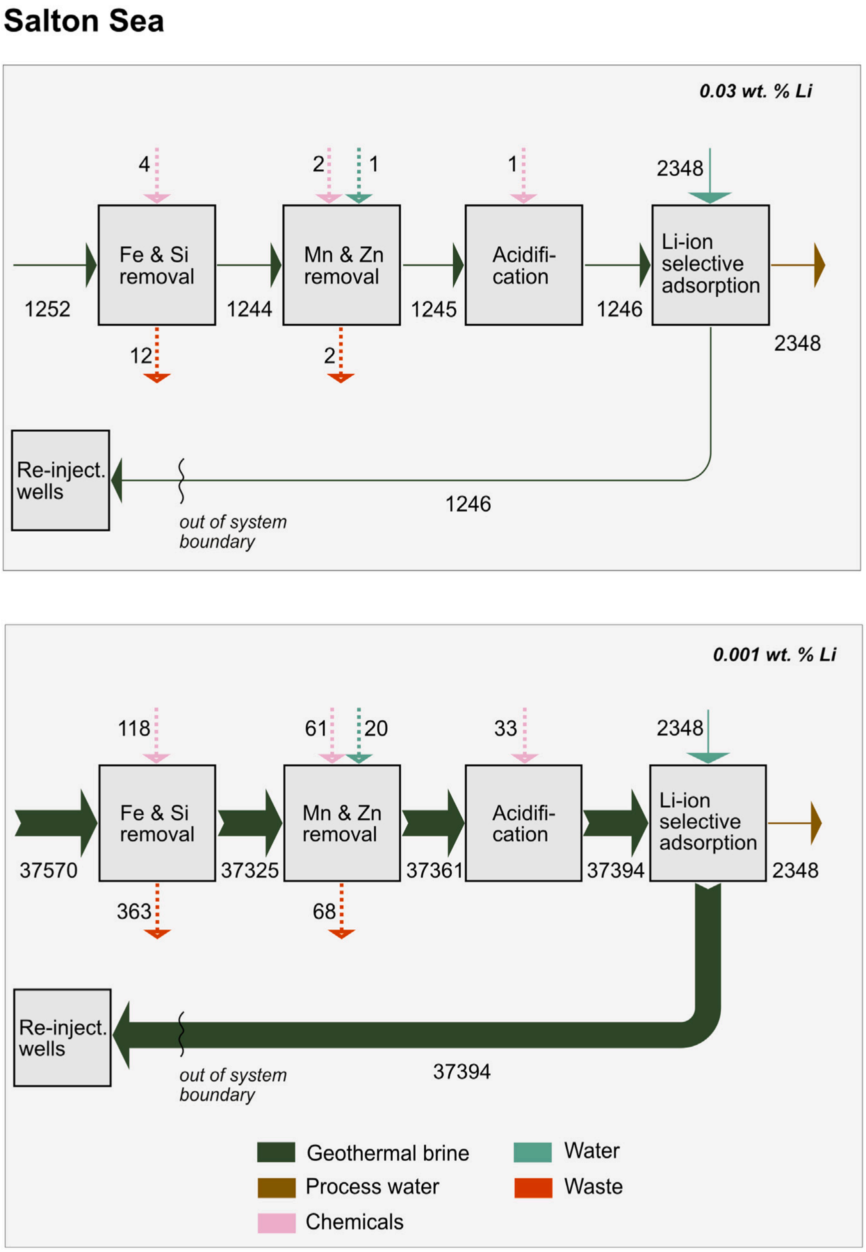


Fig. 2. Modelled mass flows in kg of maximum (0.03 wt.% Li, top) and minimum (0.001 wt.% Li, bottom) Li concentration of a geothermal brine at the Salton Sea (η = 50%). The mass flows are modelled for 1 kg Li_2CO_3. Re-inject. wells = Re-injection wells.

CO_2eq/kg Li_2CO_3 at the Salton Sea and 42.7 kg CO_2eq/kg Li_2CO_3 at the Upper Rhine Graben. The substantial increase of climate change impacts is explained by the following: The German grid accounts to 0.48 kg CO_2eq/kWh, while the western US grid accounts to 0.39 kg CO_2eq/kWh, which is a result of the different grid mixes [41]. The German grid is more dominated by electricity from lignite and hard coal, while the western US grid is based on electricity from natural gas. Regarding $PM_{2.5}$-related human health impacts, the energy shift also leads to an offset to higher impacts at both locations. The impacts are up to 1.5-fold higher at the Salton Sea. However, the relative offset almost diminishes with declining Li concentrations as seen in Fig. 3 (entire data sets in Table B.7 and Table B.9 for Salton Sea). To produce 1 kg Li_2CO_3 from a brine with a lower Li-concentration requires higher mass flows in the pre-treatment phase which does not require any energy inputs but chemicals (as discussed for Fig. 2). At the Upper Rhine Graben, the impacts even increase by a factor of 3.2–5.1 when changing the energy provision. The more pronounced offset at the Upper Rhine Graben is explained by the characterization factor there, which is significantly higher than at the Salton Sea due to the higher population density. A characterization factor for $PM_{2.5}$ of 1.94×10^{-5} DALY/kg $PM_{2.5}$ at the Upper Rhine Graben and one of 6.95×10^{-6} DALY/kg $PM_{2.5}$ at the Salton Sea are used from Oberschelp et al. [45]. Water scarcity impacts are less affected by the change of energy provision, showing only minor increases for both sites. Since the major share of the water scarcity impacts of both the Salton Sea and the Upper Rhine Graben come from chemicals production (e.g. soda ash), a shift in energy provision results in an increase but is relatively small compared to the impacts of produced chemicals used for processing. The results demonstrate the importance of assessing the energy provision level of Li_2CO_3 production. If the geothermal energy is sold to costumers, and natural gas and the grid provide the energy for Li_2CO_3 production, the impacts drastically change. Hence, the results emphasize the necessity to test these sensitivities from an environmental perspective.

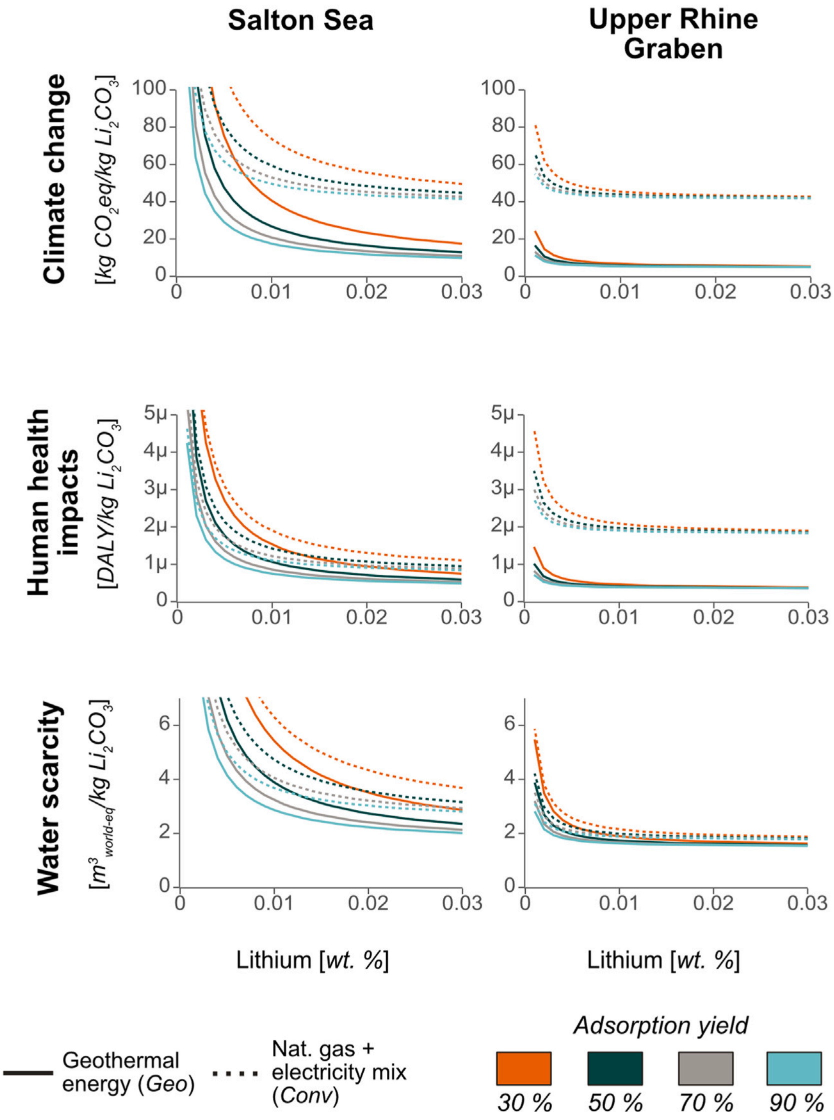


Fig. 3. Environmental impacts of Li_2CO_3 production at the Salton Sea and the Upper Rhine Graben. Li concentration is reduced in 0.001 wt.% Li steps, and the adsorption yield is varied between 30% and 100%. Nat. gas = natural gas, CO_2eq = carbon dioxide equivalents, DALY = disability adjusted life years, $m^3_{world-eq.}$ = cubic meters world equivalent, wt.% = weight percent.

3.2.3. Chemical composition of brines

The chemical composition of the geothermal brines at the Salton Sea and the Upper Rhine Graben may vary and hence yield variable environmental impacts. Geochemical analyses for the Salton Sea from Neupane et al. [15] and the Upper Rhine Graben from Kovacs et al. [16] were used (1) to model LCI and (2) to quantify life cycle impacts. Fig. 4 demonstrates the variability of climate change impacts for the observed ranges of chemical compositions with a fixed adsorption yield (η = 50%). Further results of $PM_{2.5}$-related human health and water scarcity impacts can be found in the supplementary A. Climate change impacts vary from 10 kg CO_2eq/kg Li_2CO_3 to 93 kg CO_2eq/kg Li_2CO_3 at the Salton Sea and from 5.2 kg CO_2eq/kg Li_2CO_3 to 24 kg CO_2eq/kg Li_2CO_3 at the Upper Rhine Graben. The variability of brine chemistry is higher at the Salton Sea, in consequence leading to higher variations of climate change impacts at the Salton Sea than at the Upper Rhine Graben. Changes in the overall brine chemistry may occur during operation because these reservoirs are geochemically heterogeneous (e.g. as shown by Sanjuan et al. [13]). This variability has a direct effect on the environmental impacts, as clearly indicated by the results.

3.2.4. Drilling activities

Due to the potential decrease of Li concentration of the geothermal brine over time, continued drilling of boreholes could maintain or even elevate Li concentrations. Hence, drilling activity using electrical power [46] at each site is included in the Li_2CO_3 production, for sensitivity assessment (Fig. 5). Different scenarios of drilling activity (0.5, 1, 2, 3 new boreholes per year (=the total annually pumped brine volume)) for both sites referring to the site-specific drilling depth are tested. As the installation of multiple new boreholes per year may not be feasible in practice, the case with no drilling is compared to the scenarios with continued drilling activities. Additionally, Li concentration is varied from 0.03 wt.% Li to 0.001 wt.% Li. Li_2CO_3 production (η = 50%) with geothermal energy provision (*Geo*) and the country-specific electricity mix used for drilling activities are employed. Environmental impacts at the Salton Sea hardly change by added drilling activity (3 boreholes), while it significantly affects the Upper Rhine Graben impacts. As seen in Fig. 5, climate change impacts at the Salton Sea increase by 4% and 7%, when using a brine containing 0.03 wt.% Li and 0.001 wt.% Li. At the Upper Rhine Graben, they increase by 247% and 1448%, respectively. With additional drilling activity, climate change impacts of Li_2CO_3 production at the Upper Rhine Graben reach the

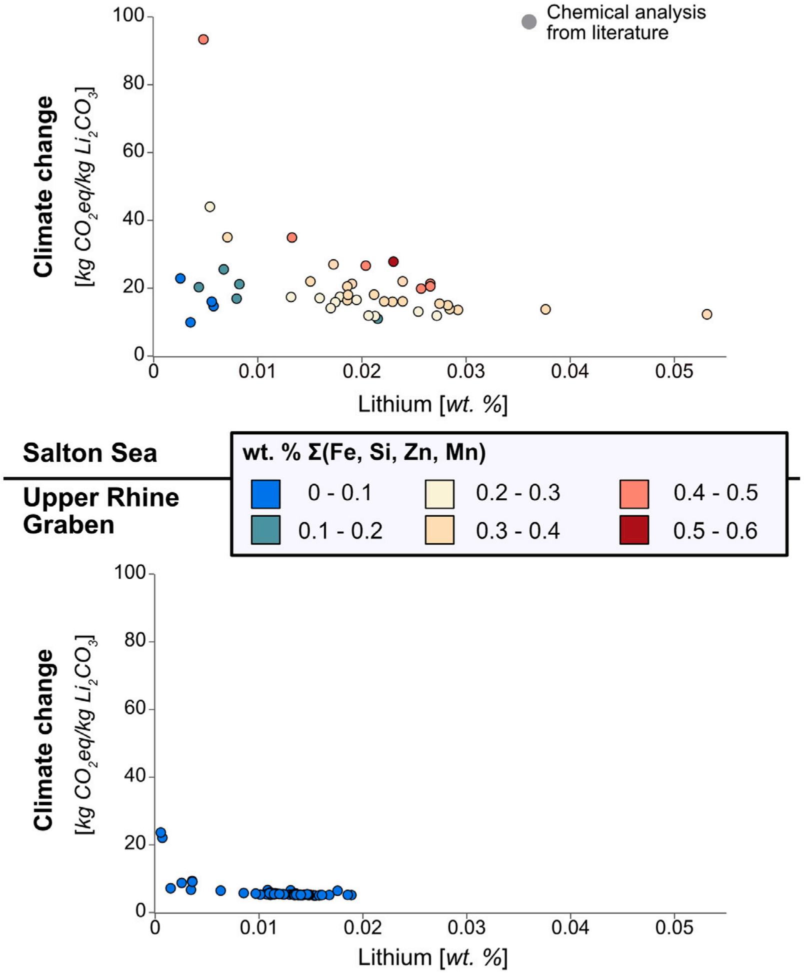


Fig. 4. Li_2CO_3 from geothermal brines with differing geochemistry [15,16]. The colour coding is based on the sum of the impurity (Fe, Si, Zn, Mn) concentrations. LCI are modelled with a fixed adsorption yield (η = 50%) and geothermal energy provision. CO_2eq = carbon dioxide equivalents, wt.% = weight percent.

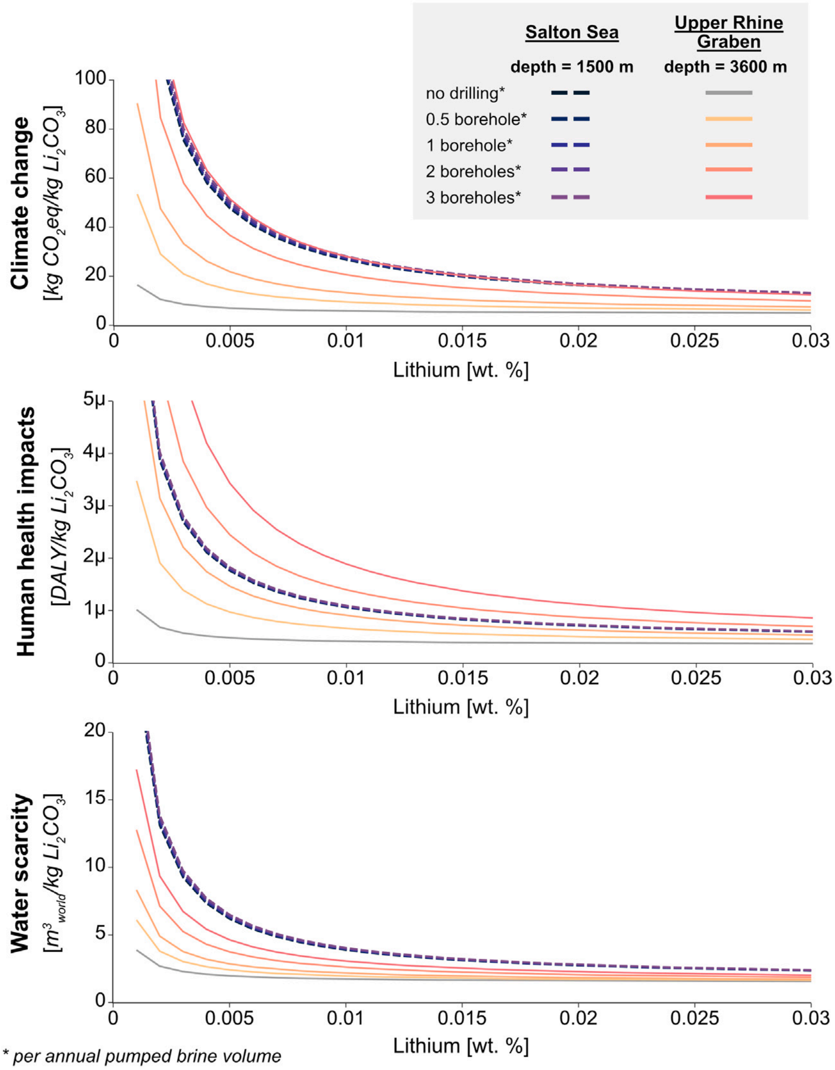
same range of impacts or even exceed the impacts of the Salton Sea. Drilling activity increases $PM_{2.5}$-related human health impacts by 2% (0.03 wt.% Li) up to 5% (0.001 wt.% Li) at the Salton Sea. At the Upper Rhine Graben, the impacts are amplified by a factor of 2.3 (0.03 wt.%) and 15.5 (0.001 wt.% Li). Drilling two boreholes at the Upper Rhine Graben exceeds $PM_{2.5}$-related human health impacts of the base case (*Geo*) without any drilling at the Salton Sea for all modelled brine concentrations. In terms of water scarcity impacts, a similar discrepancy for Li_2CO_3 production is observed at the Salton Sea and the Upper Rhine Graben. For the maximum drilling activity, water scarcity impacts only exhibit an increase of 2–6% compared to the geothermal base case at the Salton Sea. At the Upper Rhine Graben, water scarcity impacts are intensified by a factor of 1.3–4.4. However, in contrast to the other environmental impacts, water scarcity impacts at the Upper Rhine Graben remain lower than the impacts at the Salton Sea, also with three additional boreholes.

Based on these results, drilling activity at the Upper Rhine Graben appears to be more critical than at the Salton Sea. Two main reasons were identified regarding the large contribution of drilling at the Upper Rhine Graben in contrast to the Salton Sea: (i) The reported pumped brine volume (80 L/s) used in the model is smaller than that at the Salton Sea (437 L/s), and (ii) the drilling depth at the Upper Rhine Graben (=3600 m) is larger than at the Salton Sea (=1500 m). Both factors, the brine volume and required drilling depth, result in more drilling metres per kg Li_2CO_3 at the Upper Rhine Graben. In addition to these results, the German electricity mix used for drilling yields higher greenhouse gas (GHG) emissions, and thus elevated climate change impacts, at the Upper Rhine Graben. Regarding $PM_{2.5}$-related human health impacts, the characterization factors of Germany are generally higher than the ones from US-WECC due to higher population density.

3.3. Environmental impact drivers

In this section, life cycle impacts and their main drivers for selected cases at the Salton Sea and the Upper Rhine Graben are discussed (Fig. 6). The base cases (A) for the Salton Sea (0.018 wt.% Li) and the Upper Rhine Graben (0.019 wt.% Li), base cases with integrated drilling of one borehole per year (B) and Li_2CO_3 production from a brine containing 0.01 wt.% Li (C) are used to assess the drivers of the life cycle impacts. Furthermore, these scenarios were analysed with the two systems of energy provision for Li_2CO_3 production (*Geo* for geothermal heat and power, and *Conv* for heat from natural gas and the local electricity mix). The focus is on climate change and $PM_{2.5}$-related human health impacts. Water scarcity impacts and more detailed results can be found in Figures A.7 and A.8.

Climate change impacts sum up to 17.6 kg CO_2eq/kg Li_2CO_3 for the case *Geo-A* at the Salton Sea (Fig. 6). Climate change impacts of *Geo-B* only increase by <1% (17.7 kg CO_2eq/kg Li_2CO_3). Lowering the Li concentration to 0.01 wt.% Li (*Geo-C*) leads to 26.7 kg CO_2eq/kg Li_2CO_3. The pre-treatment of the geothermal brine largely contributes to the total climate change impacts (*Geo-A*: 66%, *Geo-B*: 65%, *Geo-C*: 77%). Quicklime used for MnOH and ZnOH precipitation and the disposal of the generated waste are responsible for the large contribution of these processes at the Salton Sea. Changing the energy provision to



* per annual pumped brine volume

Fig. 5. Environmental impacts of Li_2CO_3 production with annual drilling activity at the Salton Sea and the Upper Rhine Graben. Energy provision of Li_2CO_3 production is geothermal energy.

natural gas and country-specific electricity mix, the impacts increase by 282% (*Geo-A*), 281% (*Geo-B*), and 221% (*Geo-C*). In addition to the impacts caused by the pre-treatment, processes requiring either heat or electricity or both (i.e. ion exchanger, reverse osmosis, triple evaporator) are responsible for the higher climate change impacts. For example, volume reduction by reverse osmosis and the triple evaporator cause 37% of these impacts (*Geo-A*). In the context of Li_2CO_3 (technical grade) production from geothermal brines at the Salton Sea, environmental impacts based on LCIA methods TRACI 2.1 [47] are estimated to be substantially lower than those from this study [18]. Climate change impacts sum up to 1.2 kg CO_2eq/kg Li_2CO_3. The main reason for this significant difference is the incompleteness of included processes, inputs and outputs in the LCA study by Huang et al. [18]. For example, they did not include any processes to reduce the concentrations of unwanted ions (e.g. Fe, Si, Mn, Zn) by adding chemicals which cause the major share of environmental impacts, as shown in this study. Another reason is that the authors [18] assumed that the energy is provided by the geothermal power plant, which has lower environmental impacts than conventional energy sources (e.g. fossil fuels). Using the pre-existing geothermal power for that purpose, however, would create a supply gap for regional electricity demand that would need to be filled with conventional power, which was not covered in that study.

Regarding Li_2CO_3 production at the Upper Rhine Graben, climate change impacts amount to 5.3 kg CO_2eq/kg Li_2CO_3 (*Geo-A*), 6.6 kg CO_2eq/kg Li_2CO_3 (*Geo-B*), and 5.8 kg CO_2eq/kg Li_2CO_3 (*Geo-C*). The contributing supply chains are in great contrast to those observed at the Salton Sea. The pre-treatment only contributes 11% to the total impacts of *Geo-A*. Li-ion selective adsorption makes up 37% of the total impacts. The reason for this is the usage of a cationic resin to absorb Li. The usage of the ion exchanger to further purify the solution represents 31% of the impacts. The washing water required for the ion exchanger needs to be treated beforehand by reverse osmosis using electricity. Drilling integrated into the analysis (*Geo-B*) accounts for 21% of the climate change and hence increases the impacts from 5.3 kg CO_2eq/kg Li_2CO_3. Energy provided by natural gas and the German electricity mix (*Conv*) amplifies climate change impacts (*Conv-A*: 42.6 kg CO_2eq/kg Li_2CO_3, *Conv-B*: 46.5 kg CO_2eq/kg Li_2CO_3, *Conv-C*: 43.8 kg CO_2eq/kg Li_2CO_3). As for the case of the Salton Sea, energy-intensive processes, such as the ion exchanger, reverse osmosis and triple evaporator, account for the strong increase in climate change impacts. Regarding life cycle impacts of Li mining from the Upper Rhine Graben, a consulting company was commissioned to perform an LCA of 1 kg of $LiOH{\cdot}H_2O$ [31]. However, the lack of transparency of the LCA, such as methodological choices, hampers any evaluation or critical discussion of the presented results (e.g. negative GHG emissions per functional unit).

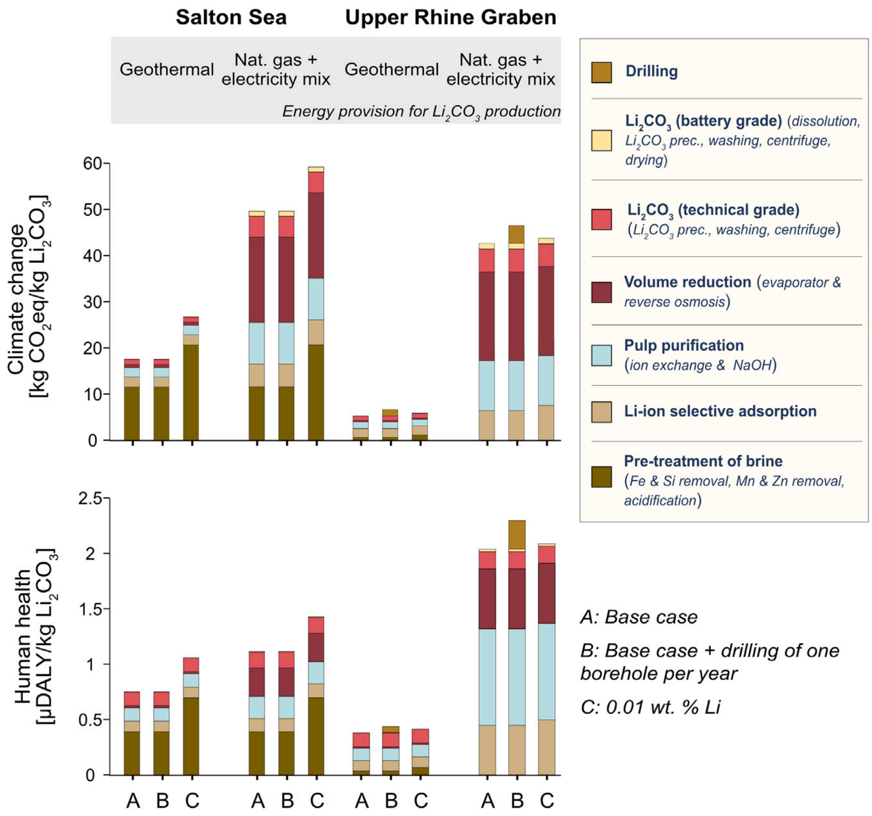


Fig. 6. Contributional analyses of climate change and $PM_{2.5}$-related human health impacts on a process basis for selected scenarios. Nat. gas = Natural gas, Li_2CO_3 prec. = lithium carbonate precipitation.

$PM_{2.5}$-related human health impacts at the Salton Sea amount to 0.75 µDALY/kg Li_2CO_3 (*Geo-A*), 0.75 µDALY/kg Li_2CO_3 (*Geo-B*), and 1.1 µDALY/kg Li_2CO_3 (*Geo-C*). As already observed for climate change impacts, the main contributing process sequence is the pre-treatment of the geothermal brine, with 51.9% contribution (*Geo-A*). The disposal of the generated waste accounts for 22.3% of the total impacts, followed by hydrochloric acid used in the acidification process with 20.3%. Li_2CO_3 (technical grade) precipitation comprises 16.2% of the impacts due to the usage of soda ash. Li-ion selective adsorption makes up 12.7%, attributed to the cationic resin used. Regarding case *Geo-B*, the contributions are the same as for case *Geo-A*, because the impacts of drilling are negligible. *Geo-C* results in an increasing share from 51.9% to 65.7% of the pre-treatment phase due to the larger mass of brine required to be treated. In contrast to the observations regarding climate change at the Salton Sea, these impacts do not show such a great proportional rise (*Conv-A*: 148%, *Conv-B*: 148%, *Conv-C*: 129%). Energy-intensive processes, such as the ion exchanger, reverse osmosis and triple evaporator, are the main drivers for the growth of impacts.

In contrast to the Salton Sea, $PM_{2.5}$-related human health impacts at the Upper Rhine Graben sum up to only 0.38 µDALY/kg Li_2CO_3 (*Geo-A*), 0.43 µDALY/kg Li_2CO_3 (*Geo-B*), 0.41 µDALY/kg Li_2CO_3 (*Geo-C*) when using geothermal energy for Li_2CO_3 production. For the case *Geo-A*, the most relevant contributor is the Li_2CO_3 (technical grade) precipitation (31.9%) sequence due to the use of soda ash. The extensive use of acids and bases to re-generate the ion exchanger makes up 29.3%, and cationic resin in the Li-ion selective adsorption 24.8% of the total $PM_{2.5}$-related human health impacts. The main cause for their contributions is the emission-intensive production based on fossil fuels. Adding drilling to this system (*Geo-B*) increases the impacts by 13% due to the electricity needed for drilling. Reducing the Li concentration in the geothermal brine (*Geo-C*) leads to an increase of impacts due to the increased mass going through the pre-treatment and Li-ion selective adsorption. Changing the energy provision for Li_2CO_3 production results in a substantial increase of impacts. More specifically, they increase by 292% in case *Conv-A*, 260% in case *Conv-B*, and 258% in case *Conv-C*. As already discussed for the Salton Sea, energy-intensive processes are the main drivers for the substantial increase. Especially in the German case, the characterization factor but also the electricity mix differ from the one in the US, as already discussed in Section 3.2.2.

3.4. Comparison with global Li_2CO_3 mining

In the context of global Li_2CO_3 production, the impacts of the selected cases from this study are compared with alternative sites and technologies as reported in the literature (Table 2). In general, climate change impacts of Li_2CO_3 production from the two sites are higher than the ones from Salar de Atacama, which is often used as a proxy for worldwide Li_2CO_3 production. Comparing these results with the other Li mining operations, *Geo-A* of the Upper Rhine Graben has lower impacts than Argentinian salars and pegmatites. The case *Geo-A* of the Salton Sea is more emission-intensive than the Argentinian salars and pegmatites and only less emission-intensive than Chaerhan salt lake. Shifting the energy provision from *Geo* to *Conv* in the Salton Sea and the Upper Rhine Graben results in climate change impacts that exceed all reported values in the literature. Impacts of *Conv-C* from the Salton Sea and *Conv-B* from the Upper Rhine Graben are 1.9 and 1.5 fold higher, respectively, than Chaerhan salt lake, the highest currently reported in the literature.

Regarding $PM_{2.5}$-related human health impacts, Li_2CO_3 from the Salton Sea and the Upper Rhine Graben amount to larger total impacts than the lowest one (Salar de Olaroz: 0.31 µDALY/kg Li_2CO_3) reported by Schenker et al. [30]. Selected cases (*Geo-A, Geo-B*) of the Upper Rhine Graben are in the same range of impacts reported for Salar de Atacama (0.43 µDALY/kg Li_2CO_3) but lower than the other Argentinian salars. This stands again in contrast to the Salton Sea. *Geo-A* of the Salton Sea is only lower than Li_2CO_3 from Chaerhan salt lake

Table 2
Comparison of Li_2CO_3 production from salars/salt lakes and pegmatites from the literature. Impacts are given in unit of impact category per kg Li_2CO_3 at battery grade.

Type	Location	Source	Climate change (kg CO2eq)	PM2.5-rel. impacts (μDALY)
Geothermal brines	Salton Sea	This study	17.6 (Geo-A) – 59.2 (Conv-C)	0.75 (Geo-A) – 1.4 (Conv-C)
	Upper Rhine Graben		5.3 (Geo-A) – 46.5 (Conv-B)	0.41 (Geo-A) – 2.2 (Conv-B)
Salars/salt lakes	Salar de Atacama	[41]	2.1	
		[28]	2.7–3.1	
		[30]	3.4	0.43
	Salar de Olaroz	[30]	7.4	0.31
	Salar de Cauchari-Olaroz		7.7	0.67
	Salar del Hombre Muerto		8	0.67
	Chaerhan salt lake		31.6	1.2
Pegmatites	Greenbushes	[41]	10.6	
	Greenbushes	[28]	20.4	

(1.2 μDALY/kg Li_2CO_3). Changing the energy provision, $PM_{2.5}$-related human health impacts of all selected cases of the Salton Sea and the Upper Rhine Graben exceed the literature values of Li mining from salars. Depending on the system setting, geothermal Li_2CO_3 has the potential to have higher impacts than other Li sources.

Li mines assessed in the literature are under continuous change, and the reported values are a snapshot of a specific point in time. The results of this study emphasize the relevance of environmental and technical parameters. Transparent data at an industry level are needed and will require sound assessment in the future.

3.5. Relevance for Li-ion batteries

To view the results within a larger context, the selected scenarios from above are used to test the implications regarding the environmental impacts of Li-ion battery production. LCI obtained in this study were integrated in the production of a Li-ion battery with a cathode of 80% Ni and 10% each of Mn and Co (NMC811) from ecoinvent [41] (results in Table B.21). The climate change impacts of a Li-ion battery with Li_2CO_3 from brines amount to 71.5 kg CO_2eq/kWh capacity [41]. Only 1% of these impacts is attributed to Li_2CO_3 from brines. In ecoinvent, this data mainly comes from the Salar de Atacama, which represents lower climate change impacts compared to other Li sources [30]. Integrating Li_2CO_3 from the Upper Rhine Graben (*Geo-A*) leads to an increase of 2% in the overall impacts. This stands in contrast to production in the Salton Sea (*Geo-A*), which increases the impacts of a Li-ion battery by 11%. The contribution of Li for the batteries becomes dominant in the cases that are more consistent with the assumption that geothermal electricity is sold to other customers (*Conv-A*: Salton Sea +34%, Upper Rhine Graben +29%). The Li impact share of a Li-ion battery becomes even more important when considering drilling activity or the drop of Li-concentration (*Conv-C*: Salton Sea +41%, *Conv-B*: Upper Rhine Graben +30%). These results emphasize the necessity of early-stage assessments in order to showcase the potential impacts but also to highlight which supply chains require more transparent data. Improved data, especially in the upstream industries of the Li-ion battery, are crucial for any decarbonization strategies for this growing sector [48].

4. Conclusion

Geothermal brines are a promising source of Li to diversify the Li market of economic hubs and to contribute to meeting the expected future demand. To prevent environmental burden shifting and overall increases of environmental impacts, it is crucial to assess the specific impacts of Li from geothermal brines. This study provides a model and specific LCI and assesses the environmental impacts of Li_2CO_3 production from geothermal brines in two locations having planned Li production. It further investigates future potential changes in terms of brine chemistry. The environmental impacts of Li_2CO_3 production from geothermal brines can be higher than those from other Li sources, depending on the actual brine chemistry, drilling metres required and energy sources used. This study emphasizes that Li from geothermal brines is not necessarily more environmentally sustainable than Li from other sources. The relevance of brine chemistry in terms of environmental impacts is clearly supported by the current findings and highlights the necessity of site-specific assessments. The development of Li concentration over time is indicated to be crucial from an environmental perspective and hence requires more geological research in the future. Additionally, the investigation on varying brine chemistry demonstrated the importance of the pre-treatment phase of Li_2CO_3 production. The environmental impacts of geothermal brines with high impurity concentrations in particular, such as the Salton Sea, are driven by the pre-treatment phase. This stands in great contrast to geothermal brines having lower impurity concentrations. In this case, the Li-ion selective adsorption and pulp purification are highly relevant in terms of environmental impacts. Due to the importance of these processes, more transparent data on operational settings and enhanced assessments at an industry level are required for the future. Another finding is that the energy provision may significantly contribute to the amplification of environmental impacts, and thus renewable energy should be used. The importance of drilling requires integration of drilling scenarios in future assessments, especially for brines with low impurity concentrations. Continued efforts are required to improve LCI, not only for Li (e.g. Li-chemicals such as $LiOH{\cdot}H_2O$ and other future Li sources) but also for other battery materials and to assess their contribution to Li-ion batteries.

CRediT authorship contribution statement

Vanessa Schenker: Conceptualization, Writing – original draft, Methodology. **Peter Bayer:** Conceptualization, Methodology, Supervision. **Christopher Oberschelp:** Data curation, Writing – original draft. **Stephan Pfister:** Conceptualization, Methodology, Supervision.

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.

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Appendix A. Supplementary data

The supporting information contains two files. Supporting information A documents life cycle inventory modelling and further results. Supporting information B (excel file) reports on characterization factors, modelled life cycle inventories of base cases, input data on the brine chemistry and corresponding LCA results (raw data).

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.rser.2024.114456.

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