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A comparative assessment of value chain criticality of lithium-ion battery cells

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ABSTRACT

As the global transport sector ramps up the transition towards electromobility, the value chain of raw materials for lithium-ion battery (LIB) development is becoming crucial. Assessing the criticality of material value chains identifies potential supply risks within these value chains and can better inform battery technology development. This study uses the ESSENZ method to systematically assess eleven (11) criticality aspects of ten (10) LIB cells. The criticality scores of the LIB cells are evaluated by aggregating the criticality scores of eleven (11) constituent value chains. These criticality scores are further complemented by twelve (12) environmental midpoint impacts performed using life cycle assessments. For the value chains, cobalt dominates the criticality scores in political stability, mining capacity, trade barriers, the feasibility of exploration projects, and the occurrence of coproduction. Lithium dominates the criticality scores in demand growth, concentration of reserves, concentration of production, and primary material used. Nickel dominates the criticality scores in price volatility, while natural graphite dominates the criticality scores in company concentration. We further explain in our results the reasons driving the criticalities in the value chains. For the comparative LIB cell assessment, we developed a quadrant matrix chart depicting the relative performance of the LIB cells based on their aggregated elemental criticality and environmental impact scores. This analysis identifies LIB cells with low value chain criticality and environmental scores and those that need improvements on either the criticality or the environmental impact scores. We propose a series of measures, such as the transition towards cobalt-free batteries, material efficiency improvements, and end-of-life recycling to alleviate the criticality and environmental impacts associated with these LIB cells. This research emphasizes the need to include all sustainability dimensions for comprehensive and holistic insights to positively shape the course of action towards sustainable LIB production systems.

1. Introduction

The transport sector currently accounts for approximately a quarter of direct emissions from fuel combustion, with land transport accounting for about 75% of total transport emissions [1]. Curbing anthropogenic emissions from the land transport sector through electrification of road transport using electric vehicles (EVs) has gained a significant momentum in the last decade. The EV demand is projected to grow steadily in the next decades [2], causing a great need for battery raw materials [3,4]. As these raw materials form the pivot of decarbonization efforts, understanding the criticality of their value chains as well as their environmental impacts become imperative for battery

stakeholders [5].

The success of EV deployment and the transition to electromobility will depend, in part, on the ability of battery value chains to keep up with the growing demand while mitigating supply risk. Scarcity of battery raw materials can lead to supply disruptions, which would likely hamper the transition to electromobility and stationary storage. Additionally, a growth in raw material prices, consequently increasing the cost of battery production could slow the adoption rate of EVs. Further, supply disruptions could lead to intense competition for metals, which could aggravate geopolitical conflicts [6]. The battery industry is constantly investigating ways to mitigate these supply risks by developing innovative chemistries with high densities, substituting materials

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whose value chains are critical, and reducing the reliance on concentrated production of battery chemicals and cells. Regarding innovation of new chemistries, there are efforts towards developing new generation cells that present high performance [7]. This is observed in the development of high nickel-content battery cells, with increased energy density [8]. Furthermore, as concerns the transition away from critical elements, there are optimistic ambitions in the battery industry to move towards cobalt-free batteries, identified as one of the most critical battery metals [9]. In addition, circular economy strategies like the recycling of end-of-life batteries have become a focal research point to recover some of the critical metals [10]. The demand for batteries is expected to grow exponential for the next 10-20 years before reaching its deflection point. As batteries are expected to be in use for 5–10 years and targeted to last for double that time through second life applications, the material recovered from recycling will only become substantial in the next 20–30 years. Consequently, the need for virgin battery materials will remain significant in the next decade of battery manufacturing. Furthermore, to reduce concentrated production of battery chemicals and cells and the potential risks with foreign imports particularly from Asia, there is a surge of battery factories in various parts of the world, especially in Europe. This is geared towards reducing the dependence on imports on the one hand and the other hand, taking advantage of the enormous business opportunities that battery production systems offer. In this vein, it is projected that Europe's battery manufacturing capacity will reach 960 GWh by 2030, increasing 20 folds compared to 2020 and accounting for 33% of globally announced production capacity (2900 GWh global capacity) by 2030 [11]. However, Europe will still depend on primary materials from Asia, particularly China, because many resource stocks are located there.

In view of the foregoing, there are several challenges that condition the deployment of EVs. Some of these are constraints towards the availability of the raw materials, while others are the constraints posed by the impacts of the manufacturing processes on both humans and the environment.

Constraints towards the availability of raw materials have been addressed in the academic literature through criticality assessments. These criticality assessments can be conducted at various levels including country or region-specific level, company-specific level, and at product-levels [12]. Country or region-specific methods focus on criticality along value chains related to a country or region (e.g. [13–15]). Company-specific methods are used to obtain specific criticality information for a given company's supply chain, for example, the CS-ESSENZ [16] and the approach by Kolotzek et al. [17]. Product-level assessments are conducted using methods such as GeoPolRisk [18–20], Strategic Metal Index (SMI) [21], thermodynamic rarity [22,23], and the integrated method to assess resource efficiency (ESSENZ) [24,25]. The ESSENZ and GeoPolRisk methods have been identified as the best approaches for evaluating criticality at the product level [26,27].

In addition to resource constraints, the environmental and societal impacts are also garnering a growing attention across many stakeholders. To this end, life cycle assessment (LCA) has emerged as an effective method to assess the potential environmental impacts of LIB production systems. With LCA, it is possible to choose more environmentally friendly LIB products, properly measure the effects of mitigation strategies, and appropriately allocate the contributions of processes or technologies on the overall footprints.

A significant number of LCAs of LIBs [28–35] and value chains [36] have been conducted to shed light on the accompanying impacts of LIB production systems. However, only a few studies address criticality aspects of LIB cells (e.g Helbig et al. [37], Olivetti et al. [38], and Wentker et al. [39]). Furthermore, as shown in Dolganova et al. [40], criticality aspects are barley addressed in case studies of electric vehicles. For LIB criticality, Helbig et al. [37] addressed the supply risk associated with LIBs by aggregating the scores of the supply risk of raw materials in the LIBs. In addition, Olivetti et al. [38] analyzed the potential risks associated with supply of battery raw materials and Wentker et al. [39]

examined the supply risk and environmental impact metrics for a number of LIB cathode technologies. In the studies of Helbig et al. [37] and Olivetti et al. [38], few chemistries are discussed and there is a lack of distinction between variants of the lithium nickel manganese cobalt oxide (LiNi_xMn_vCo_zO₂; denoted NMC_{xvz}, where x, y, z represents the molar fractions of the elements Ni, Mn, and Co, x + y + z = 1) chemistries. They therefore do not provide a clear understanding of how much the supply risk will change during the transition for example from NMC111 to NMC811 chemistries. Furthermore, these studies do not complement the criticality with environmental metrics. Wentker et al. [39] covers a wider range of LIB cathode technologies and post lithium ion battery (PLIB) cathodes. However, their study is limited to the cathode components and thus excludes the criticality and environmental impacts arising from the anode components (graphite, LTO and Si), current collectors (copper and aluminium), and electrolyte components (lithium and phosphorus in lithium hexafluorophosphate) in the LIB cell.

It is therefore vital to re-assess the criticality and environmental aspects of LIB cells by including all vital cell components and for a wide range of LIB cell chemistries. This study aims to fulfill this objective by examining the criticality aspects and environmental impacts of LIB cells by including the elements in the cathode, anode, electrolyte, and current collectors. Furthermore, this study addresses new LIB cell chemistries that have not been previously studied. We put forth two main research questions to achieve our objectives:

- a) how do various battery cells affect the criticality scores and the environmental impacts of value chains?
- b) and what implications do these have on current and future cell chemistries?

This paper provides the answer to these questions by applying the ESSENZ method for value chain criticality assessment and an LCA for the environmental impacts of the value chains. ESSENZ was chosen for this assessment because some of the authors of this paper have developed the method and are therefore familiar with its challenges and overall implementation. The remaining parts of the manuscript are divided as follows. Section 2 provides a background of the ESSENZ method, including its different indicators. In Section 3, we describe the methods and provide a step by step procedure of how the results are obtained. We analyse and discuss the results in Section 4 together with the limitations of the study and provide conclusions in Section 5.

2. Background - ESSENZ method

ESSENZ is a method for determining the resource efficiency of abiotic materials by assessing products' criticality from a sustainability point of view and originally considers a portfolio of 36 materials. The method focuses on economic, social, and environmental aspects, however, for the purpose of this study, only the economic sustainability dimension is considered. The economic dimension consists of the following eleven (11) socio-economic availability categories assessing the potential criticality of the abiotic raw materials within a product:

- Concentration of mine production, reserves and companies: only few countries and/or companies mine and trade resources, which can lead to potential supply disruptions; measured with the Herfindahl-Hirschmann-Index (HHI) [41]
- Price volatility: unexpected price fluctuations can lead to higher prices of a resource that might no longer be affordable; measured with the volatility indicator [42]
- Occurrence of co-production: resources might be restricted when mined as companion metals; measured with data by Angerer et al., (2009) [43]
- Political stability: governance stability in mining countries; measured with Worldwide Governance Indicators (WGIs) [44]

- Demand growth: production cannot keep up with increasing demand; measured with data by [45,46].
- Feasibility of exploration projects: Political and societal factors affecting the opening of new mines; measured with policy potential index (PPI) [47]
- Primary material use: limited use of secondary material; measured by the recycled content [48]
- Mining capacity: remaining time until a certain resource is fully extracted; measured by reserve-to-annual-production ratio.
- Trade barriers: existing barriers to raw material trade; measured with Enabling Trade Index (ETI) [49]

The material characterization factors (CF) for each category are based on the ecological scarcity approach [50,51], where the individual indicator results are set in relation to the category targets. The targets are unique for each of the 11 categories and are based on expert recommendations and stakeholder survey. By dividing the indicator results with the target, the distance-to-target (DtT) values are determined. DtT above 1 indicates a potential criticality, while values under 1 show absence of risk and therefore are set to 0. Next, the DtT values are then normalized using a global normalization factor. The values of the factor are determined based on USGS [46] and BGS [45] and represent the global production amount of the given raw material. The production value is used in order to consider the effect of material quantity produced within a year. The results obtained are in very low numbers, which makes their weight rather insignificant.

The final results are determined by multiplying the CFs with the raw material flows. Each of the categories is calculated separately, thus the results are not aggregated due to the lack of adequate weighting. That means that the categories and their trade-offs need to be considered individually similarly to LCA studies.

3. Method

3.1. Goal and scope of the study

A LIB cell consists of a positive and negative electrode (cathode and anode) attached to current collectors, which are separated by a separator soaked with a liquid electrolyte. The electrode consist of chemically active materials that participate in electrochemical reactions, binder materials that provide adhesion within the electrode and conductive carbon particles that increase conductivity of the porous layer. Cell components are enclosed in a cell casing made of either aluminum or nickel-plated steel material or in a pouch. The electrolyte used in most cells is usually lithium hexafluorophosphate (LiPF₆), which is mixed with ethylene and dimethyl carbonates or similar hydrocarbons.

This study considers a functional unit of 1 kWh of LIB cell. Battery accessory components like battery management system (BMS) and other pack components are excluded from the study. The default anode active material for these LIB cells is natural graphite. However, for the LIB cells that do not use natural graphite as anode active material, we specify the type of anode used. The nomenclatures of these cells are:

- 1. Lithium iron phosphate (LiFePO₄: LFP),
- 2. Lithium manganese oxide (LiMn₂O₄: LMO),
- 3. Lithium nickel cobalt aluminum oxide (LiNi_{0.8}Co_{0.15}Al_{0.05}O₂: NCA),

four Lithium-Nickel-Manganese-Cobalt-Oxide (NMC) cell variants,

- 4. LiNi_{1/3}Mn_{1/3}Co_{1/3}O_{2:} NMC111,
- 5. LiNi_{0.5}Mn_{0.3}Co_{0.2}O₂:NMC532,
- 6. LiNi_{0.6}Mn_{0.2}Co_{0.2}O₂:NMC622,
- 7. LiNi_{0.8}Mn_{0.1}Co_{0.1}O₂:NMC811,
- 8. composite cathode made of 50% NMC532 and 50% LMO (NMC532/LMO),

- 9. an NMC111 with a silicon anode (NMC111-Si), and
- 10. LMO cell with a lithium titanate ($Li_4Ti_5O_{12}$:LTO) anode denoted (LMO-LTO).

Eleven elements (Li, Ni, Mn, Co, Al, Cu, Fe, P, natural graphite, Si, and Ti) are considered in this study for all the LIBs because their primary sources are usually from mining activities for which criticality assessments are well suited. However, elements such oxygen, polymer binder materials, conductive carbon particles, hydrogen and other organic compounds used in the LIB cell are excluded from this study. Furthermore, synthetic graphite that is often used as anode active material in the place of natural graphite is also excluded from this analysis. The exclusion of these elements (and materials) is due to the fact that they are usually synthesized chemically or through other industrial process routes rather than being mined.

3.2. Material inventory of LIB cells

The material composition of the different LIB cells is based on data from the battery performance and cost (BatPac) model [52], which is reprocessed by [53]. To calculate the elemental composition for each LIB cell, we obtained the material flow data for the LIB components (cathode, anodes, electrolytes, current collectors, and casing) from [53]. In addition to the material flow data, we also evaluated the percentage composition of each element for the LIB cells using the molar fractions of the elements and the chemical formulae of the LIB cells. The resulting elemental inventory for each cell is the product of the material flow and the elemental percentage composition. Visualized in Fig. 1 is the total elemental mass and contribution for each LIB cell. Details of the elemental mass compositions for the different cell components and the total material flow are displayed in Tables S1- S3 of Supplementary Material.

3.3. Establishing criticality scores and environmental impacts for LIB cells

The criticality score for each ESSENZ category is obtained by multiplying the ESSENZ CFs with the total elemental material flows. For each category, the elemental criticality score is aggregated to obtain the final criticality score for the LIB cell. The absolute values of the criticality scores by category for all the LIB cells investigated are shown in Table S4 of Supplementary Material. Further, environmental impacts for the different elements or value chains are obtained from Ecoinvent 3.7 [54] using ReCiPe 2016 midpoint characterization method [55], complemented by climate change (CC) impacts from [36]. The impacts of the LIB cells are only the embodied impacts of the constituent value chains which excludes the impacts of the battery grade processing and cell manufacturing phases. The following twelve (12) environmental impact categories are considered: CC, fossil depletion potential (FDP), freshwater ecotoxicity (FETP), freshwater eutrophication (FEP), human toxicity (HTP), marine ecotoxicity (METP), marine eutrophication (MEP), metal depletion (MDP), ozone depletion (ODP), particulate matter formation (PMFP), photochemical ozone formation (POFP), and terrestrial acidification (TAP). The impacts displayed are not the total impacts of the LIB cell but only the aggregate impacts of the elements embodied in the cell. Absolute values of the environmental footprints are displayed in Table S5 of Supplementary Material.

3.4. Rescaling criticality and environmental metrics for comparative assessments

As the criticality and environmental metrics are calculated for each category, it becomes challenging to compare the performance of these LIB cells due to many categories under consideration in this study. We adopt a contextualized approach that enables a relative comparison of the LIB cells for each criticality and environmental category, applying equal weighting. For this relative comparison, we first rescale the ab-

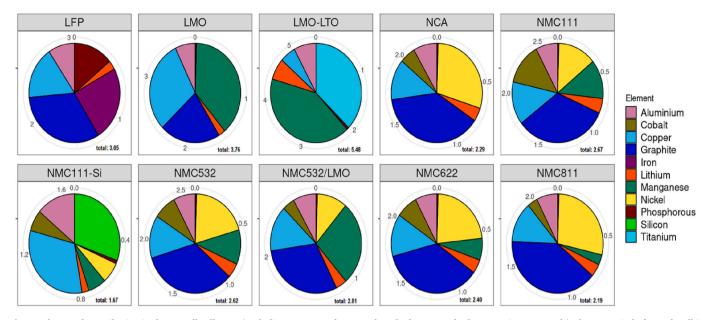


Fig. 1. Elemental contribution in the overall cell mass (excluding oxygen, polymer carbon, hydrogen, and other organic compounds): the outer circle for each cell is expressed in units of kg/kWh shows the mass spread per element; the total mass of the elements per unit capacity in kg/kWh is also displayed at bottom of each frame.

solute values for each criticality and environmental metric to a reduced interval ranging from zero (0) to one (1) (with zero being the best score and one the worst score). Rescaled values (c_r) for each criticality category are obtained by dividing the absolute criticality values (c) with the maximum criticality value of all the LIB cells as shown in eq. 1.

$$c_{r_{ij}} \in [0,1] = \frac{c_{ij}}{max(c_j)} \tag{1}$$

i represents LIB cell ($1 \le i \le 10$).

j represents the criticality category (1 $\leq j \leq 11$)

 $c_{ij}\xspace$ is the absolute value of the criticality category $j\xspace$ for cell i

 $c_{r_{ii}}$ the rescaled criticality value of the criticality category j for cell i

c_j is the maximum of the absolute criticality value for the criticality category j for all the LIB cells

The concept in eq. 1 is also used to calculate the reduced environmental values e_r . Values of c_r and e_r are shown in Tables S6 and S7 of the Supplementary Material.

4. Results and discussions

In this results section, we first present the criticality results followed by the environmental impact results. We adopt a comparative perspective in the results across the LIB cells to enable a profound understanding of which LIBs perform best and worst for a given criticality and

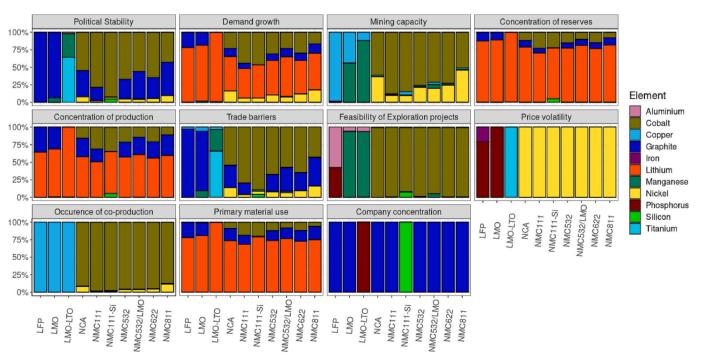


Fig. 2. Elemental contributions in the criticality scores.

environmental impact category.

4.1. Criticality of battery value chains and aggregate cell chemistry scores

The contribution of the aggregated score for each LIB cell and criticality category is displayed in Fig. 2. For the category of political stability, the graphite value chain drives the criticality score for the LFP and LMO cells, while cobalt is the main contributor of the criticality scores for the NMC cell variants, with added contribution from the graphite value chain. Graphite's high political (in)stability is caused by high production concentration in China, a country characterized by a high world governance indicator (WGI) score, which is an indication for unstable government [46,56]. Brazil, Mozambique, Madagascar, and India are also graphite producing countries with high WGI scores. In addition, the high political (in)stability of the cobalt value chains is due to the concentration of production in the Democratic Republic of Congo (DRC), which is significantly affected by political conflicts and instability, leading to high WGI [57,58]. For the LMO-LTO chemistry, titanium and manganese are the main contributors to the criticality score for political stability, due to the large material quantity used in the battery cells. In addition, a significant part of the manganese's global supply originates in Gabon, China, and South Africa, characterized by high WGI value. Titanium's production is also partly concentrated in China (35% of the global production).

Lithium is driving high demand growth scores across all chemistries, followed by cobalt for cobalt-containing chemistries. Between 2016 and 2017, the demand for lithium increased by >50% due to fast development of electric vehicles (EVs) requiring specific lithium materials [59]. The demand growth scores for cobalt reduces during the transition from NMC111 to NMC811, while nickel demand grows progressively.

Cobalt has the largest impact in the NMC cell variants, followed by nickel within the category mining capacity. The high criticality score of cobalt within the mining capacity category is largely due to the fact that 56% of global cobalt is mined in the DRC, which poses supply risks within this category. The copper value chain drives the mining capacity score for the LFP cell chemistry due to the large share of copper in the current collector tab used in the cell [37]. Additionally, for the LMO and LMO-LTO chemistries, manganese is responsible for the highest impacts within the mining capacity category due to the large mass share of manganese used in the cellcrode.

For the categories of concentration of reserves and production, lithium has the highest score across all chemistries. It is identified as a hotspot in the category concentration of reserves because almost half of the global reserves are in Chile, while the rest of the reserves are spread across a few countries like Australia, Argentina, China, and USA [46]. Moreover, lithium production is mostly concentrated in Australia (near 80% of the global production) [46] increasing its criticality score for concentration of production.

Graphite and cobalt value chains dominate the criticality scores within the category of trade barriers for all chemistries except for the LMO-LTO cell, which is impacted by titanium and manganese. Restrictions on supply due to trade barriers for graphite and cobalt reflects the Enabling Trade Index (ETI) for graphite and cobalt producing countries (China and DRC, respectively) that are significantly low. The free flow of goods is limited due to policies, infrastructure, and services in these countries with low ETI, which poses supply risks to these raw materials [60].

For all cobalt-containing cells, cobalt dominates the criticality scores within the category feasibility of exploration projects. In contrast, manganese is the value chain driving the scores within the feasibility of exploration project category for LMO cells. For the LFP cell, aluminum and phosphorus are the value chains responsible for high impacts within this category. The category feasibility of exploration projects is defined by the Policy Potential Index (PPI) of the countries containing mineral reserves and considers the regulations and policies regarding mining activities [47]. Thus, the materials identified as hotspots are due to the origin of the reserves and the policies promoting the mining of these reserves. In the DRC, which holds cobalt reserves, the PPI is extremely low, which translates into a high criticality score. Similarly, aluminum and phosphorus have reserves origins in Guinea, Indonesia, Morocco, Syria, Brazil with low PPI.

The risk of occurrence of co-production is principally triggered by the cobalt value chain for cobalt-containing cells and copper for the LFP, LMO and LMO-LTO cells. The high impacts of cobalt in the occurrence of co-production are driven by the fact that cobalt is produced as a companion metal, and there are so far very few independent cobalt mines in operation [61].

Regarding nickel-containing cells, the nickel value chain poses the highest risk regarding price volatility, while the titanium value chain drives this risk in LMO-LTO cells. The LFP and the LMO cells experience price volatility risks that come from the phosphorus value chain. The price fluctuation might be influenced by the increasing popularity of the materials, and respectively, by the increasing demand. For the case of phosphorus, along with its use as a cathode element in battery cells, the material is mostly used in agriculture as a fertilizer, and thus its demand is largely influenced by population growth and developing farming technologies [62].

The risk for primary material use is dominated by lithium through all the cells due to low recycling potential of lithium at the end-of-life [37]. In addition, graphite and cobalt contribute to criticality scores in the primary material use indicator due to low recycling potentials and recoveries. Graphite has the highest impact contribution for all the chemistries within the category company concentration except for NMC-Si and LMO-LTO, which are respectively driven by silicon and copper. As concluded by [6], a major amount of graphite is produced by a small number of companies located in India, China, and Brazil, making the material a hot spot in the category.

After analyzing the results for all the cells, we can identify several main takeaways. It is important to note that graphite is a hotspot in numerous categories among all the cells excluding the NMC111-Si, which contains silicon instead of graphite in its anode. In addition, lithium is a very large contributor to the overall criticality of all the cells within the primary material use, demand growth, concentration of reserves, and production categories. Another value chain identified as critical is cobalt, which displays high criticality in most of the categories (excluding company concentration and price volatility) for all cobaltcontaining cells. Cobalt demand is increasingly growing, accompanied by a high production share from DRC, a country that scores poorly in most of the indicators. It is important to note that the quantities of cobalt and lithium contained in the batteries are low compared to other metals. Thus, the higher criticality results are delivered from the higher CFs indicating restricted availability due to different socio-economic factors. The last material contributing to criticality of the chemistries is nickel, being the only dominating material within the price volatility category and partly influencing the mining capacity category. The material seemingly with the lesser impact to the overall criticality is iron, followed by aluminum.

Additional results of the criticality scores grouped by cell chemistry are shown in Fig. S1 of Supplementary Material.

4.2. Environmental performance of battery cells based on value chains

Like the criticality, the environmental footprint of each cell is the aggregated value along the value chains that make up the cell. The elemental contribution in the total impacts is displayed in Fig. 3. The observable trend in the impact contributions is that most of the impacts from the nickel value chain increase from NMC111 to NMC811 due to increase metal content, while impacts from cobalt value chain decrease due to a reduction in the metal content. The impact of copper dominates in the categories freshwater ecotoxicity (FETP), freshwater eutrophication (FEP), human toxicity (HTP), and marine toxicity (METP) for all chemistries due to the high toxicity of free hydrated copper ions (Cu^{2+})

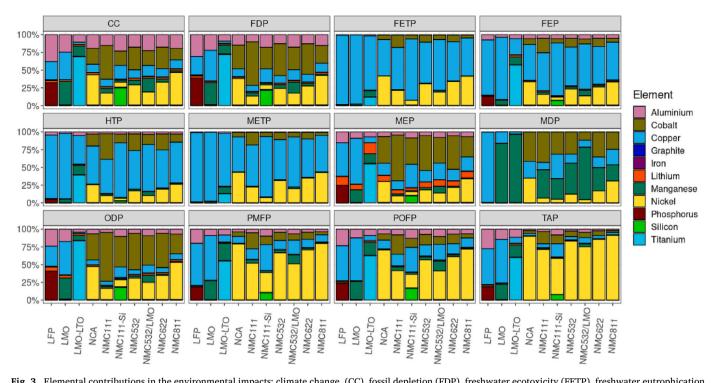


Fig. 3. Elemental contributions in the environmental impacts: climate change. (CC), fossil depletion (FDP), freshwater ecotoxicity (FETP), freshwater eutrophication (FEP), human toxicity (HTP), marine ecotoxicity (METP), marine eutrophication (MEP), metal depletion (MDP), ozone depletion (ODP), particulate matter formation (PMFP), photochemical ozone formation (POFP), and terrestrial acidification (TAP).

to especially aquatic organisms [63]. Nickel has the percentage highest contributions in particulate matter formation (PMFP), photochemical ozone formation (POFP), and terrestrial acidification (TAP) impact categories for nickel-containing cells. For TAP category, the smelting of sulphidic ores results in the emission of substantial amounts of sulphur dioxide into the atmosphere, which results in large contributions in the TAP [64]. High POFP is driven by large volumes of volatile organic compounds (VOCs) that are emitted during nickel mining and smelting. Similarly, PMFP is attributed to massive quantities of particulate matter emitted during the process of sulphidic and oxidic ore mining and smelting. Climate change (CC) is a relevant impact category due to its vitality in shaping knowledge of decarbonization strategies and will be discussed in detail. As observed in Fig. 3, for NMC variants, nickel, cobalt, aluminum, and copper are the four main elements responsible for high embodied metal CC impacts in the cells with a cumulative percentage score >65% in all chemistries [36]. This suggests that mitigation strategies to reduce the CC impacts of these four metals will significantly reduce the embodied metal CC of the cells. For LFP chemistry, copper, aluminum, and phosphorus are the main drivers of the embodied CC impacts. The contribution of manganese, which usually has low CC impacts compared to other cathode metals is significant in LMO due to large mass share of manganese in the LMO cell, which is estimated to be about 47%. For LMO-LTO cell, titanium is the major driver in the total impacts contributing >70% of the CC impacts

Contrary to the criticality indicators, the impacts from lithium and natural graphite are less significant and sometimes non-existent within certain impact categories in the environmental dimension. This is because the reported impacts from natural graphite and lithium are low compared to other elements used in the electrodes. Furthermore, copper shows a significant contribution in most of the environmental impacts but little to no to criticality in all categories except for occurrence of coproduction and mining capacity. Nickel has considerable environmental impacts but less criticality except for the price volatility category. Aluminum shows impact in the CC and FDP impact categories but no significant contribution in any criticality category. Cobalt simultaneously has high criticality scores and environmental impacts, while iron displays no major criticality nor environmental impacts. In Fig. S2 of the Supplementary Material, we add the environmental impacts grouped by LIB cell.

4.3. Contextualizing indicators and impacts for comparative analysis

Depicted in Fig. 4 top is the scores for the criticality categories. The points in the figure represent the scaled values of the categories. Three key aspects are observed: the position of a category in a chemistry relative to the same category in other chemistries, the position of the box plots with respect to vertical axis, and the position of the median line on the boxplots. It is observed from the position of the categories how well a chemistry performs for a given category compared to other chemistries. For example, for the category company concentration, NMC111-Si has a rescaled score of 0.02 while the other chemistries have rescaled scores ranging from 0.8 to 1.0. This implies that, within the category company concentration, other chemistries are 40 to 50 times worse off than NMC111-Si. Similarly, for the category political stability, LMO-LTO has a rescaled score of 0.0625 while NMC111 has a score of 1.0 suggesting the latter is approximately 16 times more liable to supply risks due to political stability than the former. Such comparative analysis is deduced for all the indicators using Fig. 4.

For the second key observation that deals with the position of the box plots and complements the position of the category, it can be observed that the boxplots of LFP, LMO, and NMC111-Si are lower on the scale than the boxplots of NMC111, NCA, NMC622, etc. Thus, LFP, LMO, and NMC111-Si perform better than the other chemistries on most of the categories. The transition from NMC111 to NMC811 yields lower criticality scores, and therefore reduces the criticality that could arise from the 11 indicators as illustrated by the boxplots and indicator points in Fig. 4 (top). For cells under development at laboratory or pilot scales, we observed that, changing the anode of an NMC111 from graphite to entirely silicon yields a significant reduction in the overall criticality for NMC111-Si. However, changing the anode of LMO cell from graphite to entirely LTO instead increases the criticality and therefore makes this switch more susceptible to supply risk disruption. Making the active

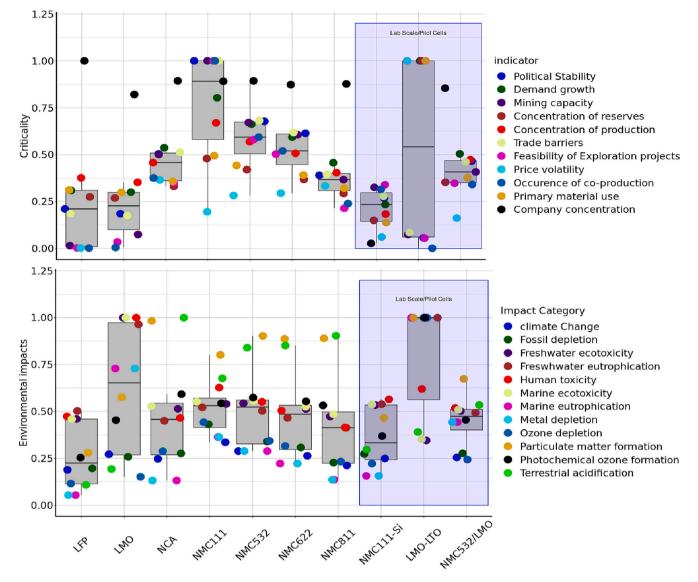


Fig. 4. Criticality scores (top) and environmental impacts (bottom) rescaled [0–1] on material composition basis. One (1) represents maximum relative criticality/impact and zero (0) represents minimum relative criticality/impact.

cathode material 50% NMC532 and 50% LMO reduces criticality, thereby making the switch more favorable from a criticality point of view.

Based on the two observations, it can be shown that some cells perform well in certain criticality category but equally worse in others. As earlier seen, a cell chemistry with a low political stability criticality score may simultaneously have a high company concentration. Considering political stability indicator, for example, NMC111 emerges as the cell with the highest criticality score as it has the largest amount cobalt used in the cathode. The next chemistries in order of decreasing political stability criticality scores are NMC532, NMC622, NCA, NMC532/LMO, NMC811, NMC111-Si, LFP, LMO and LMO-LTO. Contrary to NMC111 (with graphite anode), NMC111-Si (with silicon anode) shows a political stability criticality score four times lower than NMC111 due to the absence of graphite and a reduction in the mass of cobalt owing to a higher density of the NMC111-Si cell. LMO-LTO ranks best in category political stability due to no cobalt in the cathode and no graphite in the anode, which are elements with high WGI and high political stability criticality scores. With regards to the concentration of production, LMO-LTO has the highest criticality score due to the increased amount of lithium used to make both the cathode and anode. Further, for the category feasibility of exploration projects, the

chemistries in order of decreasing criticality scores are NMC111, NMC532, NMC622, NCA, NMC111-Si, NMC532/LMO, and NMC811. The criticality scores for the feasibility of exploration projects are directly proportional to the amount of cobalt in the cell chemistries due to low PPI for the cobalt sourced from the DRC. The chemistries with low criticality scores within this category are non-cobalt containing cell chemistries like LFP, LMO, and LMO-LTO. The category primary material use is principally driven by lithium due to low end of life (EOL) recycling potential. Therefore, chemistries with high lithium content per kWh of cell tend to have high criticality scores within this category. In this vein, LMO-LTO has the highest primary material use criticality score while NMC111-Si has the lowest criticality score due to the lowest lithium content. From the third observation that indicates the position of the median line on the box plot, we deduce a qualitatively ranking of each LIB cell.

Similarly, in Fig. 4 (bottom), we display the impact scores for the environmental impact categories. Using the analogy employed for the reduced criticality scores, achieving trade-offs in impact categories is more difficult in the LMO and LMO-LTO cells due to the large spread in the impactvalues than for the LFP and NMC532/LMO chemistries that are more clustered. The transition from NMC111 to NMC811 leads to a reduction in the total embodied elemental impacts for all categories

except for the TAP and PMFP. The TAP and PMFP increase mainly due to large contributions from the nickel value chain. The embodied CC impacts are least in the LFP chemistry due to low emissions from the cathode active material metals and highest in the LMO-LTO due to high emissions from titanium used in the anode to make LTO. Analogous to the criticality categories, we draw qualitative interpretation of the relative performance of the cell chemistries from the environmental dimensions. Based on the position of the median line, LFP, NMC111-Si and NMC811 are the top three chemistries. We complement Fig. 4 with a traffic light color gradient to depict the qualitative ranking of each LIB cell per criticality and environmental category shown in Figs. S3 and S4 of Supplementary Material.

4.4. Implication of results on current and future cell chemistries

The above results highlight the importance of considering the synergies that should exist between the criticality and environmental categories to arrive at an informed decision. Failure to do so may result in a skewed outcome towards one criticality or environmental category, which may offer a high resolution in that regard. However, analyzing a single criticality or environmental category fails to provide a holistic picture of the LIB cell's performance. In this regard, we present in Fig. 5 a quadrant matrix chart, which depicts the relative performance of the chemistries based on their median criticality and environmental impact scores.

From a technological level comparison, LFP has the best overall performance on the criticality and embodied environmental impact categories, which increases its potential as the chemistry to dominate the future market. So far, the large-scale deployment of LFP in electromobility has been inhibited by its low energy density, which reduces the driving range when deployed in EVs as compared to chemistries like the NMC with comparatively higher energy density [65]. However, there has been recent and prospective gravitation towards using LFP in electromobility. The automobile manufacturer Tesla has announced the shift to LFP cell chemistries globally for standard range vehicles [66]. It is also hinted that, other automobile manufacturers like Daimler, Ford, and Volkswagen will also adopt the LFP technology in low range electric vehicles [67]. LFP therefore could be a technology that mitigates both the criticality and the environmental impacts through large scale deployment in EVs. LMO, which has overall low criticality scores, presents a relatively large spread in the values for the different environmental impact categories. Therefore, choosing LMO is advantageous from a reduction in the criticality but may lead to higher scores in certain environmental impact categories. For NMC variants, the transition from NMC111 to NMC811 (traversing NMC532 and NMC622) yields a substantial decline in the overall criticality scores, though the NMC variants use identical value chains. This decline is accounted for in two ways. Firstly, nickel-rich NMC cathodes have a higher energy density, reducing the total elemental mass per kWh LIB capacity. As the criticality scores are directly proportional to the elemental mass, nickelrich NMC cathodes lead to lower criticality scores. Further, a transition to nickel-rich NMC cathodes reduces the cobalt content in the LIB cells. Since cobalt is an element with significant criticality factors across most of the indicators, reducing its content significantly reduces the criticality arising from the cobalt value chain. Consequently, aiming for low content cobalt in NMC cells or completely cobalt-free chemistries becomes beneficial from the criticality perspective. Simultaneously, the NMC variants experience a slight decline in the overall embodied elemental impacts as well during the transition from NMC111 to NMC811, except for TAP and PMFP.

Observing the chemistries existing at laboratory scale like the NMC111-Si, we see that the criticality is reduced in the NMC111 when graphite is replaced with silicon anodes. However, the technical challenges, principally volume expansion and contraction (+300%) of silicon during cycling inhibits its use as an alternative to graphite in the anode [68]. Silicon is therefore only used in small quantities to make a graphite-silicon composite anode [69]. By increasing the proportion of silicon in the graphite-silicon composite anode and reducing the proportion of graphite, it is possible to further reduce the criticality that would have otherwise occurred with a purely graphite anode.

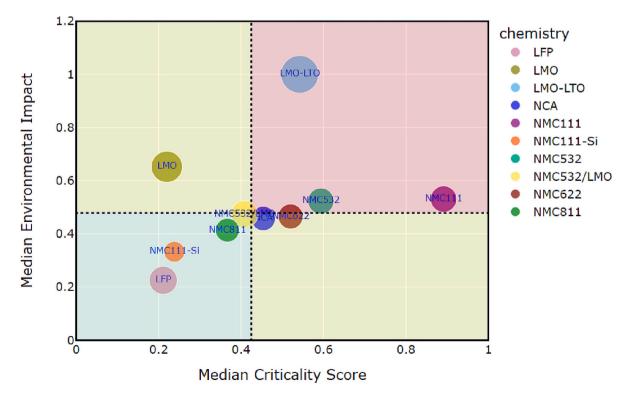


Fig. 5. Criticality and environmental impact quadrant matrix (size of the bubbles are proportional to the total elemental mass per unit LIB cell capacity, dotted lines represent the median criticality and environmental values across all the LIB cells).

Graphite-silicon composite anodes therefore could be adopted to mitigate criticality associated with the anodes in present cell chemistries. Additionally, the aggregated elemental impacts in the switch from graphite to silicon anodes also decrease in the NMC111 cell due to the increase in the cell energy density of NMC111-Si, which reduces the material used. However, it should be noted that these environmental impacts could potentially change if nano engineered silicon, such as silicon nanowires (SiNW) and silicon nanotubes (SiNT) are considered instead of metallurgical grade silicon (MGS) that has been used in this study. Studies have shown that SiNT and SiNW anodes have higher impacts than graphite anode especially in the categories of CC, TAP, PMFP, and MDP [70]. Therefore, the results of the environmental impacts only represent the basic elemental composition of the cell without considering the downstream processing steps of these base elements.

Introducing the LTO anode for the LMO cell increases the criticality compared to the LMO with a graphite anode, suggesting that such a switch yields little or no benefits from the criticality perspective. Equally, for the environmental dimensions, the aggregated elemental impact scores do not show any remarkable improvement in switching from LMO to LMO-LTO. Therefore, introducing LTO anodes as a substitute for graphite for the LMO chemistries and other chemistries may not bring any benefits both from the overall criticality and the environmental performance perspective. Approximately 3% of total commercialized anodes are LTO [71]. However, large scale deployment of LTO anodes as substitutes for natural graphite is less likely to solve the criticality problems and environmental impacts facing natural graphite. This is firstly due to a drop in the theoretical capacity for LTO which is estimated at 175 mAh/g as opposed to natural graphite at 372 mAh/g [72]. This leads to more material demand and consequently higher criticality scores per unit capacity in LIB cells with LTO anodes than those with graphite anodes. Further, LTO anodes require additional amounts of lithium and titanium which also have significant criticality scores and embodied environmental impacts.

Composite cathode active material comprising 50% NMC532 and 50% LMO (NMC532/LMO) produces criticality indicator results midway between LMO and NMC532, and elemental impacts that are comparatively lower than both LMO and NMC532. Therefore, we deduce that this composite active material could yield positive results and thus holds potential for deployment to overcome future supply risks. However, the technical specifications relating to the performance of such cathode composites are beyond the scope of this work.

It is imperative that improving current cells and developing future LIB cells must focus on attaining the bottom left quadrant of Fig. 5 where the criticality and environmental impacts are relatively lower. In addition, as the necessity to produce more sustainable batteries increases, LIB cells at the top right quadrant of Fig. 5 (where the criticality and embodied environmental impacts are relatively higher) risk being phased out and thus require more innovative and improvement actions. In this regard, a cohesive collaboration is needed between battery research and development, policy makers and sustainability experts. Though the assessment in this work principally involves primary value chains, it however implicitly prompts a need for more EOL recycling capacity globally. Increased used of secondary materials will curb the criticality associated with primary material value chains. Data from Graedel and colleagues estimates the global average EOL recycling input rate at <1% for lithium, approximately 34% for aluminum and nickel, 37% for manganese, 32% for cobalt and between 22 and 37% for copper [73]. For the European Union, these EOL recycling input rates are estimated at 22% for cobalt, 0.1% for lithium, 9% for manganese, 3% for natural graphite and 16% for nickel [74]. Evidently, existing recycling potentials are quite low and thus, the criticality associated with primary raw materials will potentially decrease if metal recycling rates were to increase. However, it is pertinent to emphasize that the challenges of recovering high purity battery grade materials suitable for LIB applications are technical concerns still facing some of the existing recycling routes [75]

4.5. Limitations

There are limitations to the study. First, some of the impact categories for the elements considered, especially those relating to ecotoxicity potential, have significant uncertainties that may affect the overall ranking within the environmental dimension. As argued by Pizzol et al. [76], ecotoxic impacts of metals have presented large disagreements in LCA results, which often prompt the exclusion of ecotoxicity impacts by LCA practitioners. Further, the footprints of the elements investigated could vary greatly due to several value chain parameters as illustrated in [36]. In addition, as the environmental impacts of the value chains depend on the chosen ecoinvent process, variability potentially arises in the results when the ecoinvent processes are changed. Choosing ecoinvent processes which are representative of the value chains for battery production presents a potential limitation. As demonstrated by Schmidt and colleagues [77], several process routes exist for nickel and cobalt but only a limited few are applicable for battery production. To overcome the limitations presented by varying background processes, we have analyzed our results from a relative perspective, and thus the extent to which the overall comparative LIB assessments will be altered with different background processes is relatively small. Nonetheless, the choice of the background processes are central to evaluating the environmental impact contributions of value chains in the LIB cell.

Moreover, the criticality indicator scores are evaluated for the average global supply risks pertaining to the cells investigated. However, it is worth noting that region specific criticality scores might differ significantly from the global average [6]. It is vital to investigate how these criticality scores could potentially change for different regions of the world. Further, the ESSENZ CFs are currently only valid for primary materials as the data required to calculate CFs for secondary materials is not available. As within this study the materials are considered to be primary ones, no distortions of the results occur. However, as battery recycling plants will be established more and more in Europe, there is a need for including secondary materials as well.

The criticality assessment is not carried out over the entire value chain, because the ESSENZ CFs are only reflecting criticality in mining countries. All indicators and underlying data used to determine the ESSENZ CFs face the challenge of data quality. Poor data quality leads to higher uncertainties. However, several well-established indicators are applied, which tend to have lower uncertainties due to constant improvement.

While this study has articulated the aggregated criticality scores and the environmental impacts of the value chains at the cell chemistry level, the social dimensions were not taken into account.

As this study primarily focuses on how LIB cells affect criticality and environmental impacts of value chains, the environmental footprints assessed do not include the footprints of the cell manufacturing phase. Further research that includes the whole LIB cell impacts is also recommended. Moreover, the material inventory for the LIB cells is subject to uncertainties, which could consequently affect the results, particularly for cells that are not yet commercialized such as NMC111-Si, NMC532/LMO, and LMO-LTO. The accuracy of the results could therefore be increased by applying updated material inventory for each LIB cell.

5. Conclusion

The insight from this research addresses the current and short and medium term criticality with the value chains of LIB cells. Criticalities from cobalt, lithium, nickel, and graphite are present in minimum of 4 of the 11 categories (>36%) across all LIB cells and jointly account for >85% of the overall criticality score in the LIB cells containing them. The comparative assessment show that LFP, NMC111-Si, NMC811, and NMC532/LMO have relative criticality scores and environmental impacts that are lower than the median values of all chemistries. In contrast, the relative criticality values and environmental impacts of

LMO-LTO, NMC111, and NMC532 are higher than the median of all chemistries. Furthermore, LMO presents a lower criticality score and higher environmental impacts while NCA and NMC622 have relatively lower impacts and higher criticality scores. The future of battery development depends on how well the battery industry reduces the criticality and environmental impacts associated with battery production. A series of measures could be adopted to mitigate potential criticality concerns and environmental impacts. The first of these measures is the transition towards low cobalt content or cobalt free batteries. In addition, material efficiency improvements that consist of reducing the total elemental mass per unit cell capacity seems beneficial. Additionally, ramping up end of life recycling would significantly reduce the criticalities and environmental impacts associated with primary material extraction. Furthermore, switching from natural graphite anodes to graphite-silicon composite anodes or nano silicon anodes would reduce the criticality associated with graphite but also increase the energy density and consequently reduce the overall material demand in the LIB cell. As the demand for LIB cells continues to grow due to large EV deployment, it is essential to include all the sustainability dimensions for a holistic and systematic assessment of current and future LIB cells. Such assessments are to be periodically updated to account for changes in innovations and geopolitics, enabling policy, researchers, and industries to positively shape the course of action towards sustainable LIB production systems.

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CRediT authorship contribution statement

Nelson Bunyui Manjong: Conceptualization, Methodology, Formal analysis, Visualization, Writing – original draft. Vanessa Bach: Methodology, Formal analysis, Writing – review & editing. Lorenzo Usai: Formal analysis, Writing - review & editing. Sylvia Marinova: Methodology, Formal analysis, Writing – review & editing. Odne Stokke Burheim: Supervision, Funding acquisition, Writing - review & editing. Matthias Finkbeiner: Methodology, Writing - review & editing. Anders Hammer Strømman: Conceptualization, Supervision, Funding acquisition, Formal analysis, Writing - review & editing.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.susmat.2023.e00614.

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