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Towards the determination of metal criticality in home-based battery systems using a Life Cycle Assessment approach

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ABSTRACT

The operation and production of batteries is associated with environmental impacts that can be quantified with Life Cycle Assessment methodologies. Current life cycle impact assessment methodologies do not assess metal criticality: they are based on geological availability or resource depletion only and do not consider socio-economic factors. Such factors are included by the concept of metal criticality. This paper determines the metal criticality of six home-based battery systems (Li-Ion: LFP-C, NMC-C, NCA-C, NCA-LTO; VRLA battery and the VRFB) for a photovoltaics self-consumption application based on a Life Cycle Assessment approach. Cumulative life cycle inventory results on extraction of metal resources are coupled with characterization factors of 13 metals derived from three state-of-the-art criticality methodologies. The results are presented for two functional units: (1) the installed battery system per kWh of energy delivered (per cycle); (2) additionally including necessary replacements of battery packs during the system lifetime. Due to substantial differences in terms of battery lifetimes between battery technologies, the latter functional unit turns out to be more meaningful. In general, there is a correlation between lower metal criticality scores (i.e. better performance) and batteries with a higher specific energy, longer battery lifetime and lower mass of metal consumption. LFP-C battery shows both low metal criticality scores and comparatively robust results, while VRFB exhibits low metal criticality but associated with relatively high uncertainties. In contrast, the VRLA battery performs the worst due to low discharge efficiency and relatively short battery lifetime. We argue that metal criticality could be reduced by improving the specific energy of the battery, by selecting low metal-intensive and low-critical metal containing components, by increasing the use of secondary metals and by selecting batteries with longer battery lifetimes.

1. Introduction

In 2016, two thirds of new electricity generation capacity installed was from renewable sources, mainly wind and solar power (IEA, 2017). According to the International Energy Agency (2017), the global electricity capacity of solar and wind is expected to expand by another 759 GW from 2017 until 2022. Due to this expected increase in solar photovoltaics and wind power, electricity generation becomes increasingly intermittent and in order to match supply and demand, different balancing solutions are required (Auer and Haas, 2016). Storing electricity in stationary batteries is one of the promising solutions to overcome this issue (Schmidt et al., 2017). Stationary batteries are preferable for short- to mid-term energy storage due to their fast response, low standby losses and high round-trip efficiency (Chen et al., 2009).

However, the operation and production of batteries is inevitably associated with costs and environmental impacts (Baumann et al., 2017; Schmidt et al., 2018). Potentially scarce or 'critical' metals are used within batteries (Simon et al., 2014). Since the 1900s, the amount of global resource extraction increased with a factor of ten and in the business-as-usual scenario of a study published by UNEP (2011), it is expected that global extraction of resources could further increase three-fold by 2050 compared to the levels in 2000 (Krausmann et al., 2017). Furthermore, critical resources are increasingly used in renewable energy technologies to reach a low-carbon economy (Hertwich et al., 2015; Vidal et al., 2013). It is expected that this increasing demand will result in increasing unit

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List of abbreviations

BMS	Battery Management System
CFs	Characterization Factors
DoD	Depth-of-Discharge
EC	European Commission
EI	Economic Importance
EMS	Energy Management System
ESU	Energy Storage Unit
EU	European Union
FU	Functional Unit
HHI	Herfindahl-Hirschman-Index
kWh	kilowatt-hour
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LCSA	Life Cycle Sustainability Assessment
LFP-C	Lithium-Iron-Phosphate/graphite
LIB(s)	Lithium-Ion Battery (-ies)
NCA-C	Lithium-Nickel-Cobalt-Aluminium-Oxide/graphite
NCA-LTO	Lithium-Nickel-Cobalt-Aluminium/Lithium-Titanate-Oxide
NMC-C 111	Lithium-Nickel-Manganese-Cobalt-Oxide/graphite
NSTC	National Science and Technology Council
PCS	Power Conditioning System
PGU	Power Generation Unit
PVSC	Photovoltaics Self-Consumption
SR	Supply Risks
VRFB	Vanadium Redox Flow Battery
VRLA	Valve Regulated Lead Acid
WGI	Worldwide Governance Indicator

costs of production in terms of capital investments as well as associated environmental burdens (Frenzel et al., 2017). Consequently, resource utilization is raising concerns by different organizations, in terms of Supply Risks (SR) of materials and economic shortfalls (National Research Council, 2007). Different studies aim to evaluate the scarcity of (metal) resources and risks related to their supply and introduce the concepts of material and metal criticality (BGS, 2012; EC, 2014; Graedel et al., 2012; Morley and Eatherley, 2008; Moss et al., 2013; NSTC, 2016; Zepf et al., 2014).

Characterization Factors (CF) in existing Life Cycle Impact Assessment (LCIA) methodologies assess primarily geological availability or depletion of metals, omitting broader dimensions of metal criticality (Sonnemann et al., 2015). Metal criticality can be identified as a multidimensional sustainability concept, consisting of the economic, environmental and socio-political dimension. Although it is still debatable if Life Cycle Assessment (LCA) methodology should include socio-economic indicators, the inclusion of metal criticality indicators in LCA or in the Life Cycle Sustainability Assessment (LCSA) framework generates a more holistic approach of metal utilization (Mancini et al., 2018; Schneider et al., 2014).

Numerous studies have been conducted comparing life-cycle emissions and life-cycle costs of different batteries for the electric mobility application (Ellingsen et al., 2014; Kim et al., 2016; Majeau-Bettez et al., 2011; Notter et al., 2010; Peters and Weil, 2018). However, only a limited number of studies focus on the environmental performance of stationary battery systems (Baumann et al., 2017; Hiremath et al., 2015; Schmidt et al., 2018). Metal criticality as such is often analyzed from the perspective of the entire economy, while metal criticality assessments for particular products, and more specifically for batteries, are largely missing. Peters and Weil (2016) solely examine resource depletion of Lithium Ion Batteries (LIBs) using current LCIA methods applied. While Simon et al. (2014) focus on the development of a metal criticality assessment of the active material in stationary LIBs, they do not include the entire battery system. In addition, Helbig et al. (2017) identify the SR of metals in 6 different LIBs, however focus on SR of batteries in electric vehicles only. Gemechu et al. (2015) identify the geopolitical SR related to one type of electric vehicle to include material criticality applied in the LCSA framework. The work of Cimprich et al. (2017) extends the work of Gemechu et al. (2015), and compares the geopolitical SR related to an electric vehicle with those related to a conventional vehicle.

None of the previous studies compare metal criticality of stationary battery systems, including a wide set of criticality dimensions including SR, environmental considerations and economic importance. In addition, there is no consensus on how metal criticality should be evaluated in LCA (Sonnemann et al., 2015). Furthermore, batteries are usually assessed on a mass or specific energy basis and lack consideration of essential key parameters of battery technologies, such as the calendric and cycle lifetime of the battery (Peters et al., 2017). These issues represent research gaps which we aim to close: we use the most comprehensive and harmonized Life Cycle Inventory (LCI) data available for stationary battery systems as recently developed in Schmidt et al. (2018) and link the metal resource consumption from the cumulative LCI results of these battery systems with three different metal criticality methodologies, to determine metal criticality of battery systems considering their lifetime performances.

Our study includes 4 LIB technologies: Lithium-Iron-Phosphate/graphite (LFP-C), Lithium-Nickel-Cobalt-Aluminium-Oxide/graphite (NCA-C), NCA/Lithium-Titanate-Oxide (NCA-LTO) and Lithium-Nickel-Manganese-Cobalt-Oxide/graphite (NMC-C 111). LIBs offer high specific energy, lightweight and high performance (Xu et al., 2008) and are preferred and applied more frequently today. However, other batteries might offer benefits in terms of metal criticality and are therefore included here, namely the Valve Regulated Lead Acid (VRLA) battery and the Vanadium Redox Flow Battery (VRFB). Our evaluation of critical metals in battery systems could help battery designers to identify potential improvements related to costs and risks in metal supply chains for the future development of home-based battery systems (Graedel and Reck, 2016; Simon et al., 2014).

The paper is organized in the following way: in section 2, the methods and data will be discussed. First, the concept of criticality is introduced. After that, state-of-the-art criticality methodologies are selected to determine metal criticality CFs which are related to the scope of this research. Next, the individual metal criticality CFs are linked to the cumulative battery LCI. In section 3, the results are provided for two FUs to demonstrate the importance of the inclusion of battery lifetimes. In sections 4 and 5, the discussion and conclusions are presented.

2. Methods and data

2.1. Material criticality

Critical materials can be defined as those that have ‘the quality, state, or degree of being of the highest importance’ (Graedel and Nassar, 2013) or those ‘that have a supply chain to disruption, and that serve an essential function in the manufacture of a product, the absence of which would cause significant economic or secure consequence' (NSTC, 2016) or whereof 'the risks of supply shortage and their impacts on the economy are higher than for most of the other materials' (EC, 2014). Derived from the variety of definitions, it can be concluded that material criticality is still a vague term and that there is no agreement on the definition of criticality (Frenzel et al., 2017; Jin et al., 2016).

Consequently, there is a wide set of criticality methodologies available (Jin et al., 2016). They widely differ in scope, in terms of time-horizon, geographical scale, materials covered and organizational level represented as well as in terms of dimensions, indicators, weighting and aggregation of indicators (Gemechu et al., 2015; Graedel and Reck, 2016). We refer to the work of Achzet and Helbig (2013) for a detailed explanation on how criticality methodologies are usually developed.

2.2. Selection of criticality methodologies

Three material criticality literature reviews are considered to select suitable criticality methodologies for the battery application. These are Graedel and Reck (2016), Sonnemann et al. (2015) and Jin et al. (2016). The criticality of a material and its CF highly depends on the organizational level of the user, the geographical scope and the time horizon assessed (Graedel and Reck, 2016). We believe that raw materials in battery applications are expected to be used for energy storage in the medium-term time scale (5–10 years) because of the ongoing progress in the battery industry (Chen et al., 2009; Simon et al., 2014). Our work focuses on metals, as these are considered as the most critical and relevant ones in technology assessments (Graedel et al., 2015), and we perform our evaluation from a global economic perspective. In addition, the selected criticality methodology should cover and assess the most used metals in the battery application.

Two criticality methodologies included in previous reviews were selected as appropriate considering the scope of this study. These are the methodologies of Graedel et al. (2012) and EC (2014). In addition, a more recent methodology of the National Science and Technology Council (NSTC) (2016) is included, which was not covered in literature reviews but matches the scope of our research. Other studies are not selected because of a lack of or inappropriate data (e.g. Morley and Eatherley, 2008; Moss et al., 2013; Roelich et al., 2014), a different time horizon (e.g. BGS, 2012) or a different geographical focus (e.g. National Research Council, 2008). The characteristics of the selected methodologies are summarized in Table 1.

2.2.1. *Graedel et al. (2012)*

The methodology of Graedel et al. (2012) aims to quantify metal criticality of 62 metal elements of the periodic table. The methodology includes three dimensions: SR, vulnerability to supply restrictions and environmental implications of metal utilization. The SR differ on the temporal scale applied, and the vulnerability to supply restrictions differ on the organizational level used (corporate, national level or global level). For the SR, we use the medium-to long-term time scale and for vulnerability to supply restrictions the 'global' organization level, as this matches the scope of this research most appropriately. In this case, there is a focus on the global environment and medium-to long-term time scale. The indicators used (see Table 1) are equally distributed over the three dimensions, although some dimensions include more indicators and thus have different sets of weighting factors within each dimension. The calculation of the overall criticality score is visualized in a three-dimensional criticality plot. The final criticality of a material is obtained by calculating the distance of the origin from the criticality plot and yields the 'criticality vector magnitude' (Graedel et al., 2012).

2.2.2. *NSTC (2016)*

The subcommittee of the Critical and Strategic Mineral Supply Chain of the National Science and Technology Council (NSTC) in the US aims to evaluate 78 materials to identify a set of critical materials. Their set of critical materials can serve as an early-warning in relation to economic risks and security interests. The final criticality score is derived from three dimensions: SR, production growth and market dynamics. The SR dimension composes the Herfindahl Hirschman Index (HHI) and the Worldwide Governance Indicator (WGI) to determine possible supply disruptions. The production growth defines the annual growth rate of a material based on its global primary production. The market dynamics aims to identify the robustness of a material when it is subjected to price fluctuations. The market dynamics dimension is generated by fluctuations of the material's price over a specified time horizon. The NSTC (2016) study focuses on material trends in the medium-term on a global geographical scope.

2.2.3. *EC (2014) SR*

The second report of the European Commission (EC) on critical materials aims to determine critical materials for the European Union (EU). The EC (2014) methodology is focused on the medium-term and limits the geographical scope to continental Europe. 54 materials are assessed and indicated as either critical or non-critical

Table 1
Relevant characteristics of selected criticality methodologies.

	Graedel et al. (2012)	NSTC, (2016)	EC (2014) SR
Number of materials assessed	62 metals	78 materials	54 materials
Geographical focus	Global	Global	Global
Concept	Three-dimensional criticality space	Final dimension Criticality	Criticality matrix with x and y axis (here only y-axis considered)

Dimensions & Indicators	- SR:
	  - Depletion time (reserves)
	  - Companion metal fraction
	- Vulnerability to supply disruptions:
	  - Percentage of population utilizing a particular metal
	  - Substitute performance
	  - Substitute availability
	  - Environmental impact ratio
	- Environmental implications:
	  - ReCipe endpoint human health
	- ReCiPe endpoint ecosystems
		- SR:
		  - HHI
		  - WGI
		- Production growth:
		  - Annual growth rate of a material's global primary production
		- Market dynamics:
		  - The price of a material
			- SR:
			  - Substitutability
			  - Recycling rate
			  - HHI
			  - WGI

in a criticality plot with two dimensions: the economic importance (EI) of materials on the x-axis and the SR of materials on the y-axis. The EC (2014) methodology uses certain thresholds per dimension to determine critical materials. Materials are identified as 'critical' when both dimensions exceeding these thresholds.

The SR dimension is based on the poor governance indicator, and includes the WGI. The HHI is modified to include country production concentrations to be useful in the WGI index. Also, end-of-life recycling rates and substitutability are included as these factors can influence the SR score (EC, 2014). Consequently, the SR score demonstrates the substitutability, recycling rate, country concentration and governance of a material from a global perspective.

The EI dimension is defined to determine the amount of applications of a material, and is related to the economic values of industrial megasectors of the EU. We decide to solely use the SR dimension to represent metal criticality for the EC (2014) methodology due to the following reasons. First, the EI dimension is focused on the economic value of a material for the EU, hence this dimension is not in line with this research. In contrast, the SR dimension is focused on the global perspective. Except for the selection of the exponent, there are no subjective elements applied in the SR dimension (Mancini et al., 2018). In addition, there is no aggregation method given by the EC (2014) to aggregate the two dimensions into one single criticality score. Consequently, the aggregation of the two dimensions would lead to subjective decisions which we want to avoid. One should notice that metal criticality is a broad concept and is often focused on the SR dimension (e.g. Moss et al., 2013).

Now that we have selected three criticality methodologies, we have to incorporate these methods for application in the LCIA of our evaluation.

2.3. Life cycle assessment and characterization factors

The most common and wide-ranging method to evaluate environmental performance of products and systems is the LCA framework (Hellweg and Milà i Canals, 2014). The strength of an LCA is that it includes all environmental-relevant flows, including emissions, energy, materials, natural resources and waste, of a process or product. According to the International Organization for Standardization, an LCA entails of four steps: goal and scope definition, LCI analysis, LCIA and an interpretation of the results (ISO, 2006a, 2006b).

Currently, LCIA methodologies lack CFs to determine metal criticality based on elementary flows (Cimprich et al., 2017; Sonnemann et al., 2015). Consequently, the metal criticality scores obtained from the selected criticality methodologies in this study are used as CFs and are coupled to cumulated metal inventories to demonstrate the total metal criticality for each battery system. The CFs of these methodologies can be found in Table 2. Note that the color formatting visualizes the criticality of a metal in each methodology (with red as the highest criticality and green as the lowest criticality). Furthermore, iron is used as 'reference metal' in each methodology and therefore assigned with a CF value of 1. The original metal criticality scores can be found in Appendix A of the online Supplementary Information (Tables 1–3). Metals should be considered as relative critical to each other for comparison (Graedel and Reck, 2016). Therefore, the original CFs are normalized to the criticality of the reference metal iron within each methodology. This means that the metal criticality score of iron is set to one to increase comparability between methodologies and metals. The metals are normalized by dividing their scores by the reference score of iron. This generates a metal criticality score in iron equivalent, which is dimensionless. We selected 13 metals, as these metals are most relevant in batteries, they are available as elementary flows in the ecoinvent v3.3 database (Ecoinvent, 2016) and are assessed by the selected criticality methodologies.

It should be noticed that there are substantial differences between the identification of critical and non-critical metals between the criticality methodologies. Graedel et al. (2012) identify gold, silver and lead as most critical metals within our metal selection, while NSTC (2016) indicates cobalt, tin and chromium as most critical metals. The EC (2014) methodology identifies cobalt, chromium and tin as the metals with the highest SR. The largest differences are found on the CFs of gold and lead. Gold and lead are indicated as relatively critical metals by Graedel et al. (2012), but are identified as low critical metals by the EC (2014).

Moreover, the methodologies differ in variation of CFs, which can be explained by their methodological choices. For instance, the CFs of EC (2014) have more variability than the CFs of Graedel et al. (2012). In other words, the EC (2014) indicates more differences between metals of high and low SR than Graedel et al. (2012) identify on their metal criticality score. This should be taken into consideration when interpreting the results. We refer to the original methodologies and articles for a more comprehensive explanation of the individual metal criticality scores, as the focus of our work is not to explain why metals are identified as critical.

Now that we have transformed original criticality scores into CFs, we have to link these to the cumulated masses of consumed metals to produce the battery systems.

2.4. Life cycle inventory

Harmonized LCI data of stationary battery systems have previously been compiled by some of the authors in the work of Schmidt et al. (2018). These are used to quantify the cumulative LCI of metal resource extraction for the FUs. The systems of LIBs and VRLA battery include battery pack (i.e. stacks of battery cells), Battery Management System (BMS), Energy Management System (EMS) and Power Conditioning System (PCS). The VRFB system is structured differently due to its capability of separate sizing on energy- and power-based components, consisting of an Energy Storage Unit (ESU) and a Power Generation Unit (PGU). The ESU is mainly for electrolyte storage and pumping, while the stacks and other major components are mostly in the PGU.

Brightway2 is used to calculate the cumulative LCI results per Functional Unit (FU) (Mutel, 2017). The ecoinvent v3.3 database with the system model 'Allocation, cut-off by classification' (Wernet et al., 2016) is used as background database. From now on, we use the term 'battery' for the entire battery system (including all system components) and the term 'battery pack' referring to the stacks of battery cells with module and pack housing.

The distinction of primary and secondary metal (recovered by recycling) utilization can be identified as a driving factor for metal criticality, as high utilization of secondary metals can decrease metal criticality (Buchert et al., 2009). However, specific recycling of battery systems is not considered in this study as battery demand is expected to increase substantially and there are currently no large-scale recycling infrastructure and regulation mandates in place (Richa et al., 2014). Furthermore, there are no reliable sources available which assess the recycling of battery systems (Baumann et al., 2017). Because of the uncertain potential of the recycling of battery systems, we use market activities of the ecoinvent v3.3 database which represent current global market of materials supply that are not specific for battery applications. For instance, using the ecoinvent global market activity of lead results in a primary and secondary lead use of 45% and 55%, respectively. The actual recycling rate of lead within VRLA battery applications might be higher depending on the country of interest.

Table 2
CFs derived from three criticality methodologies.

	Graedel et al. (2012)	NSTC (2016)	EC (2014)
Aluminum	0.63	1.25	0.86
Chromium	1.34	1.38	2.02
Cobalt	1.06	2.13	3.26
Copper	0.99	0.75	0.44
Gold	1.64	0.56	0.30
Iron	1.00	1.00	1.00
Lead	1.48	1.31	0.80[1]
Lithium	0.58	0.81	1.30
Manganese	1.16	1.19	0.86
Nickel	0.76	1.19	0.48
Silver	1.62	1.06	1.46
Tin	1.22	1.44	1.64
Zinc	1.21	0.94	0.90

[1]Lead not assessed in EC (2014) report, therefore 'global SR' value of lead from EC (2017) adopted.

Table 3 presents the selected key parameters of battery performance and operation. Details on the quantification of the provided characteristics of battery technologies can be found in Schmidt et al. (2018)). Operation conditions depend on the application of the battery, and strongly influences the maintenance and lifetime of the battery (Baumann et al., 2017). Therefore, it is essential to identify performance parameters in the context of a specific battery application. We limit our scope to the Photovoltaics Self-Consumption (PVSC) application (Schmidt et al. (2018), as this is currently one of the most dominant applications in stationary battery projects (Malhotra et al., 2016). With the PVSC application, end-users generate value to store the electricity produced from their own PV panels to decrease electricity utility bill (Alskaif et al., 2017). The required energy size in terms of energy and power delivered are assumed to be 5 kWh per cycle and 2.5 kW respectively, representing the smallest scale of distributed battery application, for instance, for a single-family house.

However, the actual installed storage capacity is oversized based on the required energy delivered, considering the discharge efficiency, the guarantee of 80% of initial installed capacity at the end-of-lifetime, and the Depth of Discharge (DoD). See equation (1) below Schmidt et al. (2018). Note that Table 3 presents the values of the parameters in equation (1).

$$C_E^i = E_{del}^i * RE^{i-0.5} * EoL^{i-1} * DoD^{i-1}, \tag{1}$$

where C^i_E is the installed energy capacity of battery 'i' [kWh]; E^i_{del} is the energy delivered per cycle, required by the application of battery 'i' [kWh]; RE^i is the roundtrip efficiency of battery 'i' [%]; EoL^i is the energy capacity at the end-of-lifetime as a percentage of initial installed energy capacity of battery 'i' [%]; DoD^i is the depth-of-discharge of battery 'i' [%].

The round-trip efficiency determines the amount of energy lost per charge/discharge cycle. Only the discharge efficiency should be considered when oversizing the storage capacity; hence this explains the associated power (i.e. 0.5) for the roundtrip efficiency. For all batteries, an additional oversizing consideration is included to ensure the energy required by the application at the end of the system lifetime, since the usable energy storage capacity degrades over time. This percentage is set to 80% for all battery technologies. The DoD indicates the fraction of the installed battery capacity that is discharged/charged per cycle. The DoD highly influences the total lifetime of the battery pack, as a high DoD decreases battery pack lifetime for most battery technologies (Baumann et al., 2017).

In our work two FUs are used to demonstrate the influence of the lifetime of the battery pack. Both FUs are based on a cradle-to-gate analysis and include material flows generated by all upstream processes to produce the battery systems. The first FU focuses on the battery system installed, while the second FU additionally includes replacements of battery packs during the system lifetime.

First, metal criticalities are evaluated on the basis of battery system installed per kWh of energy delivered (per cycle) for the PVSC application (see Fig. 1). 'Energy delivered (per cycle)' is specified for the PVSC application. The first FU is based on the metal criticality score per kWh 'Energy delivered (per cycle)' from battery as described in Fig. 1.

Equation (2) is applied for LIBs and VRLA battery, and equation (3) for VRFB, as VRFB is a battery technology with a different system design compared to the other battery technologies examined in this study. The difference is particularly on the split of components that are power- and energy-based (e.g., VRFB stores energy in electrolyte, and is outside of the electrochemical cells). The numerators in both equations quantify the amounts of metals embedded in the installed battery systems from a life cycle perspective, and they are normalized by the energy delivered per cycle. Note that Table 3 presents the values of the parameters for the following equations.

$$\rho_{sc}^i(x) = \left[\left(M_{PACK}^i(x) + M_{EMS}^i(x) + M_{BMS}^i(x)\right) * C_E^i + M_{PCS}^i(x) * C_P^i\right] \Big/ E_{del}^i, \tag{2}$$

where ρ^i_{sc} is the amount of metal 'x' per kWh of energy delivered (per cycle) of battery 'i' [kg/kWh]; M^i_{PACK} is the amount of metal 'x'

Table 3
Battery performance data and quantification of results are obtained from Schmidt et al. (2018). N.A. stands for not applicable.

Battery	LFP-C	NMC-C	NCA-C	NCA-LTO	VRLA	VRFB
Specific energy at battery pack level [Wh/kg]	109.8	126.3	123.3	48.6	35.0	10.5
Installed energy capacity (C_E) [kWh]	7.21	7.09	7.09	6.53	13.1	7.70
Energy delivered per cycle PVSC (E_{del}) [kWh/cycle]	5.0	5.0	5.0	5.0	5.0	5.0
Installed power capacity (C_P) [kW]	2.5	2.5	2.5	2.5	2.5	2.5
Roundtrip efficiency (RE) [%]	86.6%	89.4%	89.4%	91.5%	75.3%	65.9%
Depth-of-discharge (DoD) [%]	93.2%	93.2%	93.2%	100%	55.0%	100%
Cycle lifetime [Cycles]	6529	4996	2498	15000	1500	13000
Equivalent years based on cycle lifetime [Years]	26	20	10	60	6	52
Calendrical lifetime [Years]	12	12	12	23	9	19
Lifetime of battery system (LT_{SYST}) [Years]	20	20	20	20	20	20
Battery packs needed during system lifetime (N_{PACK})	1.67	1.67	2.00	0.87	3.33	N.A.

per unit energy capacity of the battery pack installed of battery ‘i’ [kg/kWh]; M^{i}_{EMS} is the amount of metal ‘x’ per unit energy capacity of EMS installed of battery ‘i’ [kg/kWh]; M^{i}_{BMS} is the amount of metal ‘x’ per unit energy capacity of BMS installed of battery ‘i’ [kg/kWh]; M^{i}_{PCS} is the amount of metal ‘x’ per unit power capacity of PCS installed of battery ‘i’ [kg/kW]; C^{i}_{P} is the installed power capacity of battery ‘i’ [kW].

$$\rho_{sc}(x) = (M_{ESU}(x) * C_E + M_{PGU}(x)*C_P)/ E_{del}, \quad (3)$$

where ρ_{sc} is the amount of metal ‘x’ per kWh of energy delivered (per cycle) of VRFB [kg/kWh]; M_{ESU} is the amount of metal ‘x’ per unit energy capacity of ESU installed of VRFB [kg/kWh]; M_{PGU} is the amount of metal ‘x’ per unit power capacity of PGU installed of VRFB [kg/kW]; C_P is de power capacity of VRFB [kW]; E_{del} is the energy delivered per cycle, required by the application of VRFB [kWh].

Secondly, battery systems are evaluated on the basis of storing 1 kWh of electricity in the selected application, considering necessary battery pack replacements throughout the system lifetime. The distinction between lifetime of battery system and lifetime of battery pack is made in order to include possible replacements of battery pack during system lifetime, as the lifetime of a battery pack is often shorter than the lifetime of other battery system components. For LIBs and VRLA battery, the lifetime of the battery pack is either based on the calendric lifetime or cycle lifetime. The calendric lifetime is the number of years after which the battery pack does not fulfill its requirements in terms of energy delivered per cycle, and is usually influenced through stress factors such as temperature and state-of-charge of the battery (Rohr et al., 2017). The cycle lifetime is defined as the number of cycles the battery packs last. The cycle lifetime can be converted to year-equivalents when dividing by the number of cycles required in a specific battery application per year. The number of cycles per year is defined as 250 cycles (i.e. a charge and a discharge) based on Schmidt et al. (2018). For instance, the NCA-C battery pack has a total amount of 2498 operation cycles, which is equivalent to a cycle lifetime of 10 years, given 250 cycles per year. The calendric lifetime is longer than the cycle lifetime, hence the cycle lifetime is limiting the lifetime of the battery pack. Consequently, the lifetime of the NCA-C battery pack and NCA-C battery system is 10 and 20 years, respectively. In other words, there are two NCA-C battery packs needed during the battery system lifetime. For VRFB, the battery system consists of a power generation unit and an energy storage unit, for which we assume their lifetimes are equal to the battery system lifetime. Similar to the first FU, see equation (4) for LIBS and VRLA battery, and equation (5) for VRFB.

$$\rho^{i}_{LT}(x) = \left[M^{i}_{PACK}(x)*C^{i}_{E}*N^{i}_{pack} + \left(M^{i}_{EMS} + M^{i}_{BMS}\right)(x)*C^{i}_{E}\right] \Big/ \times \left(LT^{i}_{SYST}*N_{annual}*E^{i}_{del}\right), \quad (4)$$

where ρ^{i}_{LT} is the amount of metal ‘x’ of using battery ‘i’ for storing 1 kWh of electricity [kg/kWh]; N^{i}_{pack} is the number of battery packs needed during lifetime of battery ‘i’; LT^{i}_{SYST} is the lifetime of battery ‘i’ [years]; N_{annual} is the annual number of cycles required by the application.

$$\rho_{LT}(x) = (M_{ESU}(x)*C_E + M_{PGU}(x)*C_P)/(LT_{SYST} * N_{annual}*E_{del}), \quad (5)$$

where ρ_{LT} is the amount of metal ‘x’ of using VRFB for storing 1 kWh of electricity [kg/kWh]; LT_{SYST} is the lifetime of VRFB [years].

The cumulative LCIs used to generate the metal inventories per FU can be found in Appendix B of the online Supplementary Information (Tables 4–8). The equations below are used to determine the total metal criticality score for each battery technology. Equation (6) represents metal criticality of battery system installed per kWh of energy delivered (per cycle), and equation (7) is for metal criticality of battery system for storing 1 kWh of electricity in the selected application.

$$CS^{i}_{SC} = \sum_{i=1}^{m^{i}} \left(CF_{method}(x) * \rho^{i}_{SC}(x)\right), \quad (6)$$

where CS^{i}_{SC} is the metal criticality score of battery ‘i’ installed per kWh of energy delivered (per cycle) [CS/kWh]; ‘m’ is the number of selected metals in battery ‘i’; CF_{method} is the CF of metal ‘x’.

$$CS^{i}_{LT} = \sum_{i=1}^{m^{i}} \left(CF_{method}(x) * \rho^{i}_{LT}(x)\right), \quad (7)$$

where CS^{i}_{LT} is the metal criticality score of using battery ‘i’ for storing 1 kWh of electricity [CS/kWh]; ‘m’ is the number of selected metals in battery‘i’; CF_{method} is the CF of metal ‘x’.

Ultimately, applying three sets of CFs (one from each methodology in section 2.2) to the cumulative LCIs for two different FUs, leads to the determination of metal criticality in battery systems.

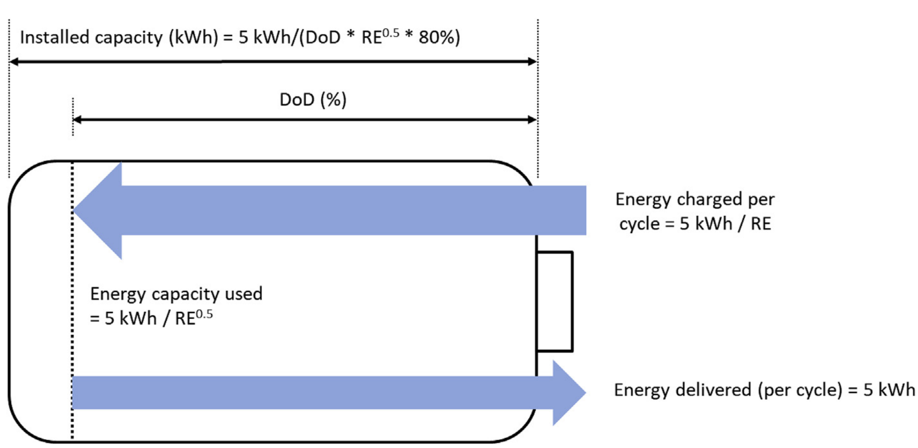

Fig. 1. Illustration of the different characteristics and parameters of the battery system.

3. Results

In this section, we present the total metal criticality scores for the two FUs, and discuss the main driving factors behind these scores as well as the differences that arise from the metal criticality methodologies applied. The numerical, cumulative life cycle inventory results for metal resource extraction and results for metal criticality by type of metal are provided in Appendix C and D of the online Supplementary Information (Tables 9–10, 11, 13). Results for metal criticality separating contributions from the production of materials used within batteries and other processes (e.g., transportation of materials, manufacturing energy, chemicals required in processing, etc.) are provided in Appendix E for selected battery technologies using the metal criticality method from the EC. For LIBs, the contributions from materials directly part of the batteries to criticality scores are above 80%, whereas for VRLA and VRFB, contributions from those materials are more than 90%. The contributions to metal criticality scores from manufacturing energy are minor (less than 3%). Interpretation of results regarding contributions by specific system components later in this section is closely related to the results shown in Appendix E.

3.1. Metal criticality of battery system installed per kWh of energy delivered (per cycle)

The results in Fig. 2 show a clear relationship between the specific energy of battery packs and metal criticality of battery systems. Note that the triangles show the specific energy at battery pack level (Wh/kg), whose values are depicted on the secondary y-axis. In most cases, higher specific energy of the battery pack corresponds to lower metal criticality score. VRFB is an exception, since it shows the lowest metal criticality scores while having the lowest specific energy of all batteries. This can be explained by the lower amount of metal consumption in the VRFB battery system. In fact, the largest mass share of a VRFB pack is electrolyte, which is less metal-intensive and mainly consists of water and sulfuric acid (93%). Metal consumption in VRFB is largely dominated by iron and copper in the power conditioning and connection system of PGU. In addition, VRFB has a maximum DoD value of 100%, which means oversizing is only required to compensate its loss in discharge and the guaranteed capacity required by the application at the end of lifetime. However, VRFB has the lowest roundtrip efficiency among the selected batteries and this partially counterbalances its beneficially high DoD. In general, the low metal criticality score of the VRFB can be explained by its low metal consumption in battery system.

All LIBs consume comparable amounts of iron and copper. Metal criticality in LIBs is largely dominated by metal consumption in PCS, cathode, module and pack housing. The lowest metal criticality score is obtained from the LFP-C battery, which can be explained by its cathode material that mainly consists of Lithium-Iron-Phosphate, and does not contain any cobalt as in other LIBs, which is indicated as a critical metal. In addition, NCA-C and NMC-C batteries perform well, because of their high DoD and roundtrip efficiency, and thus less oversizing is required. Despite its high DoD, NCA-LTO battery has the highest metal criticality of all LIBs and this can be explained by its low specific energy. In other words, higher weights of metals are required per unit capacity of energy. More specifically, this battery uses higher amounts of metals in the electrodes (e.g. nickel, lithium and cobalt) and copper in the electrode current collectors. In addition, this battery contains a mass-intensive LTO anode instead of a graphite anode, which is reflected by comparatively higher lithium consumption.

All methodologies assign VRLA battery with the highest metal criticality score, which can be explained by its relatively poor technology performance. The VRLA technology has lower DoD and roundtrip efficiency which requires a battery with larger overcapacity. Consequently, the VRLA battery consists of large amounts of lead for electrode production and iron for packaging. Contributions from zinc are caused by the co-mining of lead and zinc. Because such co-mining process is allocated based on the economic values of products, some extraction of zinc is allocated to the burden of lead production. Furthermore, the consumption of secondary lead in the VRLA battery pack does not sufficiently compensate for the high amount of metals demanded. However, this factor might be underestimated by using the global market dataset for lead from the ecoinvent database, which contains 55% of recycled lead, while the recycling rate of lead within the lead-acid battery industry might be higher.

3.2. Metal criticality throughout the system lifetime

Secondly, metal criticality of battery systems are evaluated for storing 1 kWh of electricity in the selected application, considering necessary battery replacements throughout the system lifetime. As shown in Fig. 3, it turns out that in addition to specific energy, there is also a strong correlation between longer lifetime of the battery pack (i.e. less amounts of battery replacements) and lower metal criticality score. Note that the diamonds show the battery lifetimes of the battery packs, whose values are depicted on the secondary y-axis.

We found that VRFB has the lowest metal criticality scores. This can be explained by the long battery lifetime of 20 years, although this is subjective to relatively high uncertainty of input data because less redox flow batteries have been installed worldwide than LIBs and lead-acid batteries. The ranking among LIBs is changed compared to what is shown by the first FU, specifically by the better performance of NCA-LTO battery when necessary replacements during system lifetime are considered. This can be explained by the longest lifetime of the NCA-LTO battery pack. Again, the LFP-C battery performs the best mainly because it has no consumption of cobalt in the battery. The NCA-C and NMC-C battery systems perform moderate, as their battery pack lifetimes are in the mid-range of the assessed technologies.

Similarly, the highest metal criticality score is obtained from the VRLA battery. The inclusion of the lifetime of battery pack increases the difference between VRLA battery and the other batteries. Based on the Graedel et al. (2012) methodology, the metal criticality of VRLA battery is 6 times higher in comparison with the second highest metal criticality score of NMC-C battery. In comparison, the difference on metal criticality score between VRLA battery and NCA-LTO on a storage capacity base (i.e. the first FU) was a factor of two. The high criticality score can be explained by the shorter battery pack lifetime and lower DoD of VRLA battery. Although the calendric lifetime is 9 years, the cycle lifetime is only 6 years which makes the lifetime of the VRLA battery pack limited by the number of cycles.

Based on the second FU, a ranking of preferred battery systems can be derived in terms of metal criticality. VRFB shows the best performance, followed by the LFP-C, NCA-C, NCA-LTO and NMC-C batteries, respectively. It turns out that VRLA battery has by far the worst performance in terms of metal criticality.

3.3. Variation between criticality methodologies

Figs. 2 and 3 show small variations between different methodologies. Nevertheless, there are some differences which should be acknowledged. For LIBs, the total metal criticality scores are comparable between the methodologies. However, there is a significant difference in the CFs of cobalt between the methodologies and this

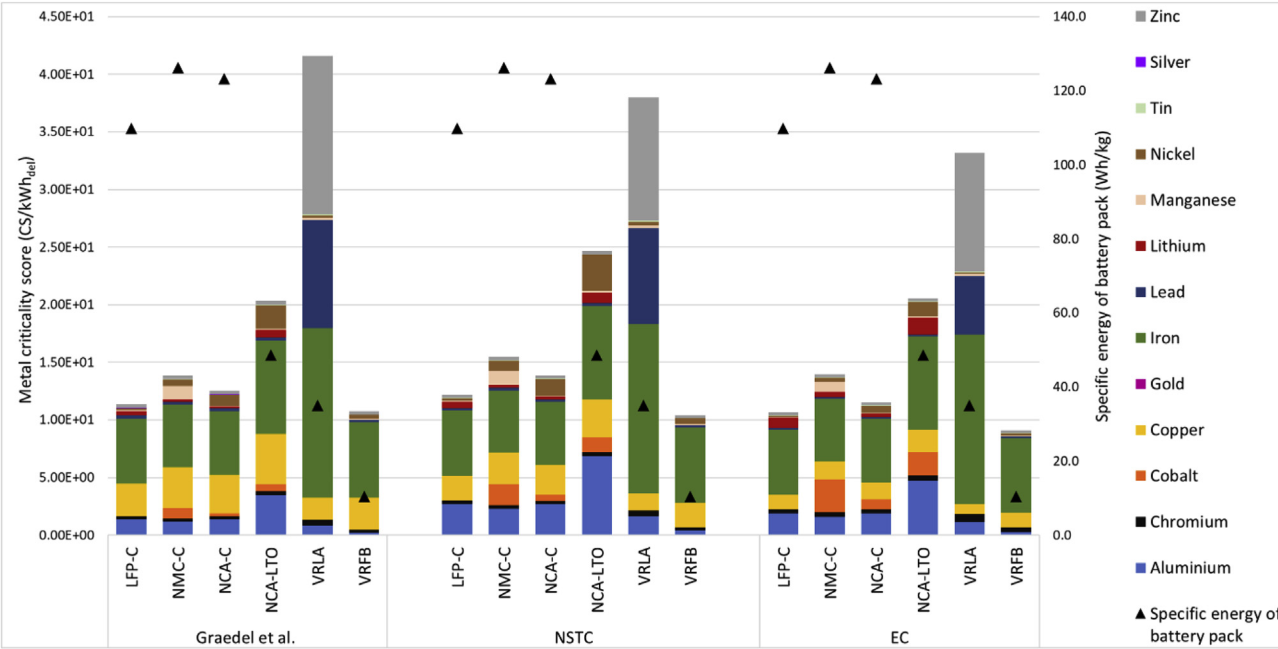

Fig. 2. Metal criticality scores of the batteries per kWh energy delivered (per cycle).

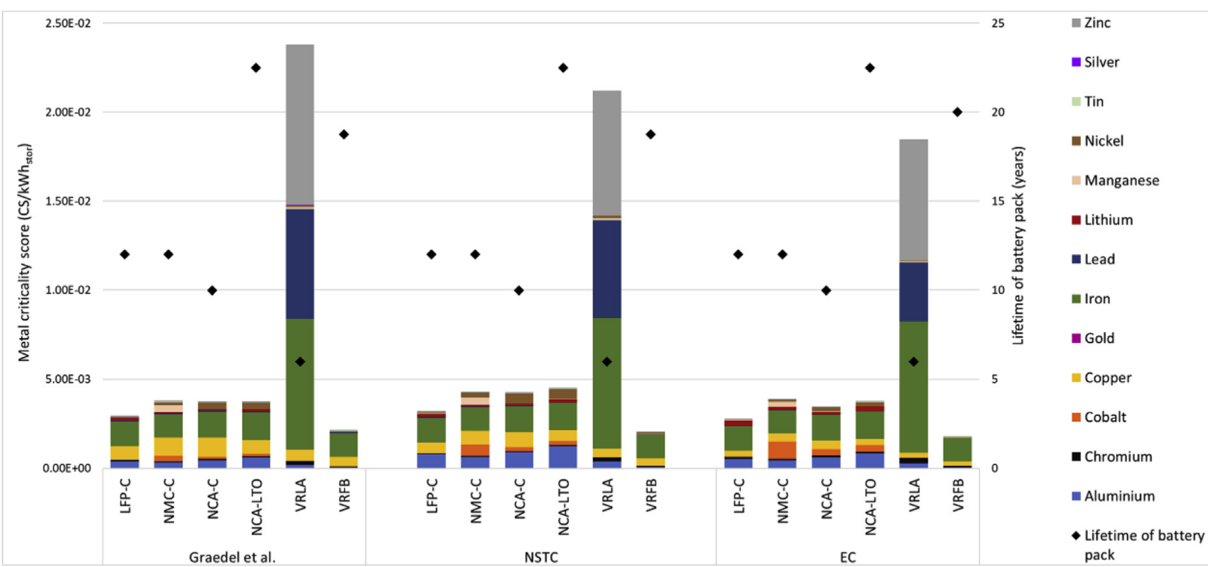

Fig. 3. Metal criticality scores of the batteries throughout the system lifetime, per kWh stored.

is reflected in the batteries (NMC-C, NCA-C and NCA-LTO) where cobalt is needed in the cathode materials. As shown in Table 2, cobalt is characterized as more critical by the NSTC (2016) and EC (2014) in comparison with the Graedel et al. (2012) methodology. However, copper is characterized as more critical in the Graedel et al. (2012) methodology in compared to the NSTC (2016) and EC (2014) methodologies. Ultimately, these different CFs for cobalt and copper compensate each other in this evaluation, which results in comparable total metal criticality scores between the methodologies. For the NCA-LTO battery, the total metal criticality scores assessed by using Graedel et al. (2012) and EC (2014) methodologies are lower than the NSTC (2016) methodology. This can be explained by the relative higher criticality of nickel and aluminum in the NSTC (2016) methodology.

The largest differences are found on the total criticality scores of VRLA battery. The metal criticality score is the highest when applying the Graedel et al. (2012) methodology, whereas the metal criticality scores using the EC (2014) and NSTC (2016) methodologies are substantially lower. T, due to lower CFs of lead and zinc in these methodologies.

In general, the total metal criticality scores are largely dominated by non-critical metals (eg. iron, copper, lead, zinc). In addition, the results demonstrate that the consumptions of non-critical metals have a larger effect than the consumptions of critical metals due to its higher amount of metal consumptions, which outweighs the higher criticality of individual critical metals.

4. Discussion

The results demonstrate that the masses of the metal resource consumption clearly dominate the total metal criticality scores of batteries which confirms the findings of earlier studies (Mancini et al., 2018; Schneider et al., 2014). This can mainly be explained by the low variability of CFs (i.e. variability of CF between highly critical metals and iron is low in compared to the difference between consumption of these metals) in criticality methodologies. A question which should be asked is whether the CFs of metal substances are in correct proportions for application in LCA. Manipulation of original CFs has been proposed to give higher importance to more critical materials (e.g. Mancini et al., 2018). However, such manipulations are subjective and more research is needed to deepen the understanding of the consequence of these manipulations. A second option is to improve the applicability of proposed methodologies, which aim to determine material criticality CFs to be applied in the LCA framework (e.g. Cimprich et al., 2017; Schneider et al., 2014).

In addition, the selected criticality methodologies differ in methodological choices and therefore the results differ to some extent. As already indicated, a widely accepted criticality methodology or framework is missing. Different authors advocate that new material criticality assessments and methodologies are needed which should rely on a wide set of indicators based on different dimensions (Frenzel et al., 2017; Glöser et al., 2015; Sonnemann et al., 2015). Our work also emphasizes that economists, social and environmental scientists should work together to integrate different dimensions into one reliable criticality framework to increase reliability and comparability between criticality assessments. Furthermore, it could be interesting to split the single metal criticality score into more dimensions when a certain criticality methodology is developed or chosen. In this way, the importance of a dimension to the criticality CF of a metal can be identified.

Furthermore, specific recycling measures for battery systems are not considered in this work. Instead, market activities for metal supply chains, as part of the ecoinvent database, are used which are not specific to the recycling in battery industry. How the recycling rate on a global market compared to the recycling rate specific to battery industry is metal-dependent, and requires further investigations when more data on the commercial-scale recycling of lithium-ion batteries is available. An increase of recycling measures could increase the utilization of secondary metals and could in turn reduce the metal criticality of batteries (Binnemans et al., 2013; Duclos et al., 2010). The work of Dewulf et al. (2010) showed that recycling of cobalt and nickel in LIBs could save 51% natural resources when switching from primary materials to secondary materials, which shows that recycling can be an influencer for metal criticality assessments.

The CFs used in this research are based on a current snapshot of the analysis year. However, criticality is a dynamic concept and is subjected to dynamic changes over time (Knoeri et al., 2013; Roelich et al., 2014). This work identifies the metal criticality in the batteries at the moment and additional assessments are required to determine potential future metal criticality. In addition, battery industry experiences fast development (Chen et al., 2009) and therefore, other promising battery and manufacturing technologies should be considered. Furthermore, to increase reliability for technology-specific criticality assessment, metal criticality CFs should be based on technology-specific applications as every technology has its own characteristics and influencers (Simon et al., 2014).

In addition, there are no CFs available from the criticality methodologies for some frequently used metals in battery systems (e.g., zirconium and molybdenum, which are used in stainless and alloy steel), hence these metals are not included in our work. Solely the NSTC (2016) methodology presents CFs for both zirconium and molybdenum. Therefore, the inclusion of these two metals on this methodology is tested (Appendix D, Tables 12 and 14): the influence of molybdenum is minimal in relation to the total metal criticality scores of batteries. It turns out that the metal criticality score of zirconium is comparably higher for the NCA-LTO battery. However, the inclusion of these metals result in non-significant differences on the total metal criticality scores.

Another question related to the quantification of life cycle inventories of metal extraction using the ecoinvent database is how to deal with the metals extracted as mixed ores. This raises concerns on how to allocate a certain material fraction contained in a mixed ore which could result in double-counting (Peters and Weil, 2016). In fact, what is more problematic is the issue of missing elementary resource flows in the background database or unmatched elementary flow names between background database and CFs provided by LCIA methods for some important metals, vanadium being the most important in the context of our analysis: vanadium is not included as an elementary resource extraction in the default list of biosphere flows in ecoinvent; in addition, vanadium is modeled to be recovered from steelmaking slags in our evaluation, and since we use the cut-off system model of ecoinvent, this slag is classified as waste and therefore available free of burden to the vanadium recovery process. Thus, even if vanadium were included as a resource extraction in the biosphere flows, it would not be accounted for in the metal criticality scores due to the cut-off system model. Vanadium is an essential metal for the VRFB, and is identified as a relatively critical metal by most of the criticality methodologies (CFs of 0.99, 2.19, 1.64 according to (Graedel et al., 2012), (NSTC, 2016), EC (2014), respectively, when the methodology in section 2.3 is applied), so we believe that this non-inclusion of vanadium leads to an underestimation of the metal criticality of VRFB.

5. Conclusions and future work

This research aims to determine and compare the metal criticality of 6 home-based battery systems using an LCA approach. Our study includes six battery technologies: LFP-C, NMC-C, NCA-C, NCA-LTO, VRLA and the VRFB. The assessment includes 13 metals used in battery systems. We started with the examination of criticality methodologies to define useful CFs which can be adopted in an LCA approach. Three state-of-the art criticality methodologies were selected. The metal criticality scores derived from the methodologies were normalized to the reference metal iron, to allow for comparisons between the methodologies. Next, harmonized LCIs for all batteries were adopted to quantify metal resource consumption per FU. We focused on the PVSC application to deliver 5 kWh electricity per cycle and 2.5 kW power. The results were quantified on two FUs: the installed battery system per kWh of energy delivered (per cycle) for the PVSC application as first FU, while the second FU additionally included necessary replacements of battery packs during the system lifetime.

The VRFB turns out to have the lowest metal criticality score on both FU. The metal criticality results of the LIBs were comparable, although the LFP-C battery shows better results on both FUs. The VRLA battery is least preferable in terms of metal criticality. The results substantially changed when the lifetimes of battery packs were considered with the second FU. This was reflected in a much better performance for the NCA-LTO battery and an even worse performance of the VRLA battery. These results showed the importance of the inclusion of the battery lifetime in battery assessments. In addition, we confirmed relationships between lower metal criticality scores and higher specific energies and longer lifetimes of the battery packs. One exemption for the specific energy relationship is the VRFB, and this can be explained by the low metal intensity in the electrolyte, which leads to a low energy density but at the same time low metal criticality score. This emphasizes that metal intensity of specific components can be another important factor for metal criticality. In addition, vanadium is an essential metal for VRFB in the electrolyte and is not included as resource flow in the ecoinvent database, which leads to a potential underestimation of the metal criticality of the VRFB. Therefore, the assessment of VRFB should be improved to give reliable recommendations related to this battery. From a metal criticality perspective, we argue to choose LIBs, and in more particular the LFP-C battery, as these technologies show robust results and perform excellent on a lifetime base.

Considering the three methodologies, we found small variations between total metal criticality scores of the methodologies. Cobalt was indicated as the most critical metal in our metal portfolio. Furthermore, the CFs derived from the metal criticality methodologies generated small differences on total metal criticality scores compared to masses of metals. Based on the CFs applied, masses (i.e. bulk metals) can be considered as more important than the variation of CFs of single metals.

We argue that battery designers could reduce metal criticality by improving specific energy, decreasing metal intensity of battery components, decreasing the use of critical metals (e.g., cobalt), utilizing more secondary metals and designing battery packs and systems with longer lifetimes. Future work should be directed towards knowledge improvement in metal consumption of promising and less assessed battery technologies (e.g., VRFB). Furthermore, there is still a research gap on recycling rates of most battery technologies. There is a need to develop efficient recycling technologies and to determine their influence on resource criticality. In addition, it is highly recommended to improve criticality methodologies and characterization models which easily can be adopted for future technology-specific criticality assessments to include a wider set of metals. One option is to integrate resource criticality into the current LCA framework. This integration could be a task for the LCA community to generate a more holistic approach for resource utilization.

Declarations of interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2019.02.250.

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