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Review article

Review of life cycle assessment on lithium-ion batteries (LIBs) recycling

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ABSTRACT

The recycling of Lithium-ion batteries (LIBs) waste is recognized as a viable solution for alleviating the pressure on natural resources caused by the increasing demand for materials used in LIBs production and the disposal of these hazardous wastes in landfills. Life Cycle Assessment (LCA) has been widely employed to evaluate the environmental impacts associated with LIBs recycling. However, a comprehensive synthesis of the lessons learned from these assessments, including methodological choices, findings, and implications, is lacking in the literature. Therefore, this study aims to summarize the available knowledge on the application of LCA for LIBs recycling. This study uses a systematic literature review method in combination with structured content analysis to identify and analyze 64 peer-reviewed LCA studies on LIBs recycling. The key findings reveal significant variations in potential impact results and divergent results regarding the environmental preference among the available recycling processes (hydrometallurgical, pyrometallurgical, direct recycling, and bioleaching). These discrepancies arise from different assumptions and methodological choices in LCA, including variations in system boundaries, inputs, the inclusion or exclusion of specific stages, unit process and flows, assumptions regarding the use of avoided products, functional units, impact assessment methods, and the use of secondary data due to the lack of primary data, especially on an industrial scale. While the Climate Change category receives considerable attention, other impact categories are often neglected, making it challenging to establish the environmental preference of a particular recycling technology. For direct recycling and bioleaching technologies lack assessments for all impact categories. Electricity consumption and chemical inputs are identified as hotspots for all recycling options. To enhance the sustainability of LIBs recycling, additional studies that focus on collecting primary data, particularly for the collection, pretreatment, and final disposal stages are recommended. To improve the transparency and reproducibility of future studies, this article provides recommendations and a research agenda for conducting LCA studies in the field of LIBs recycling.

1. Introduction

The global demand for Lithium-ion batteries (LIBs) is projected to grow rapidly in the coming years, with an annual growth rate of 30% [59]. By 2030, LIBs demand is expected to increase 14 times, driven by renewable energy storage and vehicle electrification [49]. However, this growth raises concerns about environmental and social burdens arising from the natural resources consumption for LIBs production and end-of-life management these hazardous waste [98,101,114,171]. The production of LIBs involves the supply of various metallic and chemical materials, leading to environmental impacts such as water pollution, air emissions, and hazardous waste generation [102]. The production of raw materials, chemical materials, and LIBs cells is energy and material intensive [131,45].

In social domain, the negative social impacts associated with the LIBs production also presents major concern [61,107]. The demand for metals in the production of LIBs places pressure on both local communities and ecosystems [145]. Mining activities associated with LIBs life cycle cause environmental degradation and social externalities, such as conflicts, displacement, poverty, and human rights violations [166,87,116]. Particularly, Lithium mining has been receiving increasing attention due to the social harm caused to fragile ecosystems and local communities, especially in operations conducted in South America, in the Lithium Triangle (Argentina, Chile, Bolivia) [35,117]. The excessive water consumption and production of chemical waste for lithium production cause severe socio-environmental impacts, such as water scarcity and pollution, decrease in access to clean water for local communities, biodiversity loss, local violation of indigenous

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communities rights, violations related to land access and rights, loss of livelihoods, destruction of cultural heritage, corruption, governance, and inequalities in wealth distribution [1,162,116,117]. Beyond lithium, serious social issues (e.g., forced labor, worst forms of child labor, unsafe and hazardous working conditions, political instability, lack of governance and transparency, and adverse effects on local communities health and livelihoods) are also reported for other raw materials of LIBs, such as cobalt [5,64], graphite [39], nickel, iron, and copper [87].

Furthermore, the increasing use of LIBs leads to the generation of millions of tons of LIBs waste by year [82,102,101]. By 2030, 2 Mt of electric vehicle batteries waste will be generated per year, escalating in the following decade [160]. This brings challenges related to the proper management of LIBs waste [90,101]. LIBs are classified as hazardous waste due to the presence of toxic and flammable substances in the composition of the materials, such as reactive salts containing fluorine and heavy metals [29,100,138]. Thus, improper disposal of LIBs can cause adverse effects on human health and the environment [106].

Due to these environmental and social issues associated with the production and waste management of LIBs, the proper management and recycling of raw materials from LIBs waste is an adequate solution to limit the need to extract natural resources and mitigate the pollution of inappropriate disposal [40,42]. However, for the recycling of LIBs to deliver these benefits, the development, implementation, and evaluation of adequate and environmentally friendly processes are essential [108, 102]. In the LIBs context, this assessment is even more important, since technologies for LIBs recycling are still in the initial stages of implementation on an industrial scale [129], it is still unclear which LIBs recycling methods are environmentally preferred [128], and data on the end-of-life of LIBs are scarce [47,118].

To guide environmental sustainability efforts and assess potential environmental impacts, Life Cycle Assessment (LCA) is recognized as an appropriate technique [146,135]. LCA is considered an ideal technique for investigating the environmental impacts of recycling processes, as it provides a standardized and systematic framework that raises questions about the environmental profile of recycling, helping to avoid secondary pollution [28]. However, few LCA studies evaluate LIBs recycling processes in detail [8,118]; the main interest has been in the evaluation of production stages (Pinegar & Smith, 2019). Thus, the available literature reviews on the environmental impacts of the LIBs life cycle have summarized LCA studies conducted in the LIBs production stages, in a Cradle-to-Gate approach, without including the end-of-life stages in the scope of the review (see [119]; [120]; [31]; [19]. Other reviews focused on the LIBs production include end-of-life in scope but present a less in-depth look at the potential impacts of recycling using different technologies [2,8]. Some recent reviews on LIBs recycling included investigation of LCA results (e.g., [89]; [163]. However, these studies also have limitations regarding study coverage and impacts, with the main focus on just two categories of environmental impact, namely, Global Warming Potential (GWP) and Cumulative Energy Demand (CED). Aichberger et al [2] review 16 LCA studies that evaluate the recycling of LIBs to compile results across 10 impact category groups. However, the study presents results of mapped impacts on recycling in the form of an aggregate statistical distribution without differentiating by type of recycling technology or functional units. Wenz et al. [163] analyze LCA results of LIBs recycling from only 3 studies for CED and GWP. Li et al. [89] review 24 studies and perform a meta-analysis of 111 impact observations for LIBs recycling by cathode type, energy matrix and recycling method, but they are also focused only on the impact results for GWP and CED and do not present detailed methodological choices and differences between studies. Although studies on the LCA of LIBs recycling have received increasing attention, a comprehensive review of the application of LCA considering methodological choices and all impact categories is still lacking in the literature. None of the previous reviews analyses in detail the LCAs conducted in LIBs recycling with a specific focus on all methodological choices of LCA and mapping of results for all environmental impacts categories. Questions about how and where LCA has been applied to LIB recycling systems and which lessons and best practices have been learned to improve the studies remain unanswered.

To answer these questions, this article aims to critically review LCA studies on LIBs recycling, seeking to summarize the current knowledge and provide recommendations to improve LIBs recycling and future LCA studies.

2. LIBs waste and recycling

This section presents a theoretical background of the composition of LIBs, applicable recycling processes, potential release of pollutants in LIBs recycling processes, and advances in legislation on LIBs life cycle in recent EU regulation batteries.

2.1. LIBs composition

A LIB cell is usually composed of six main components: anode, cathode, electrolyte, separator, current collectors, and housing [182,99]. The anode is composed of a copper current collector coated with a mixture of graphite, polyvinylidene fluoride binder, and additives [181]. The cathode is composed of an aluminum current collector and a mixture of ligands, additives, and active cathode material, which is composed of lithium intercalation oxides [181,186]. The active cathode material differentiates the different types of LIBs, as it can be composed of different materials, such as lithium cobalt oxide (LCO=$LiCoO_2$), lithium-nickel-cobalt-manganese oxide (NMC=$LiNixMnyCozO_2$), lithium iron phosphate (LFP=$LiFePO_4$), lithium-nickel-cobalt-aluminum oxide (NCA= $LiNiCoAlO_2$) and lithium-manganese oxide (LMO = $LiMn_2O_4$) [165,52,82]. Therefore, the term LIB covers several types of chemical composition for the cathode [61]. The electrolyte is usually made of a mixture of Li salts (e.g., lithium hexafluorophosphate), organic solvents (such as dimethyl carbonate and ethylene carbonate), and additives [51]. The polymeric separator is made of Polypropylene or Microporous Polyethylene [105]. The housing protects the LIB from the external environment and can be made of iron/steel or aluminum alloys and plastic components [165,31]. Table 1 summarizes the average composition and concentration of two types of LIB cells for notebooks and electric vehicles.

2.2. Processes for LIBs recycling

The LIBs waste recycling chain involves complex processes of reverse logistics, pretreatment, and metallurgical processing [164]. LIBs waste is recycled through thermal, chemical, or physical processing [142,102]. Technologies for recycling LIBs are classified into three main categories: pyrometallurgical, hydrometallurgical, and direct recycling, also called functional recycling [61,138]. Pyrometallurgical and hydrometallurgical methods are the most commonly applied [165]. Currently, most available commercial operations use a combination of pyrometallurgical and hydrometallurgical processes for the recovery of secondary raw materials from LIB waste [122]. As an advance of hydrometallurgy, new developments can make biohydrometallurgy a viable option for recycling LIBs [114].

The first stage of LIB recycling is the collection and sorting of LIBs waste [164]. Then, the pretreatment step separates the active cathode materials from other cell components such as housings, current collectors, electrolytes, binders, and separators [90,114,176]. The pretreatment process may involve thermal or mechanical methods [178]. Mechanical methods, such as comminution, classification, and magnetic separation, are more commonly used [66,125]. Thermal pretreatment is utilized to decompose electrolytes, remove binders and impurities [178]. Pretreatment is important to efficiently separate the metal-rich active substances, reducing impurities and facilitating the subsequent metallurgical treatment process or direct recycling [158,115]. Fig. 1 shows a simplified flowchart of pretreatment for LIBs.

Table 1
Material composition and average concentration of a LIB cell (LCO and NMC) used in Notebooks and Electric Vehicles. Source: Adapted from Wang et al [158] and Richa et al [131].

Product type	Portable electronics (Notebook)				Electric Vehicles			
Material	LCO		NMC		NMC		LFP	
	Mass (g)	Mass (%)	Mass (g)	Mass %	Mass (g)	Mass (%)	Mass (g)	Mass (%)
Al	2.40	5.21%	2.22	5.26%	45.08	8.46%	62.78	8.49%
Co	7.97	17.31%	4.08	9.67%	10.72	2.01%	0	0.00%
Cu	3.36	7.30%	3.29	7.80%	86.62	16.26%	117.40	15.88%
Li	0.94	2.04%	0.48	1.14%	12.56	2.36%	9.43	1.28%
Mn	0	0.00%	3.81	9.03%	39.97	7.51%	0	0.00%
Ni	0.56	1.22%	4.07	9.64%	42.70	8.02%	0	0.00%
Stell/Iron	7.60	16.51%	7.31	17.32%	0	0.00%	75.85	10.26%
Graphite	10.64	23.12%	7.26	17.20%	109.90	20.64%	119.57	16.17%
Carbon (non-graphite)	2.78	6.04%	2.55	6.04%	10.86	2.04%	14.44	1.95%
Electrolyte + Solvent	2.14	4.65%	2.56	6.07%	78.33	14.71%	155.74	21.06%
Binders	1.11	2.41%	1.02	2.42%	14.83	2.78%	18.33	2.48%
Plastics	2.20	4.78%	1.33	3.15%	25.88	4.86%	36.92	4.99%
Other	4.33	9.41%	2.22	5.26%	55.11	10.35%	128.94	17.44%
Total	46.03	-	42.2	-	532.56	-	739.4	-

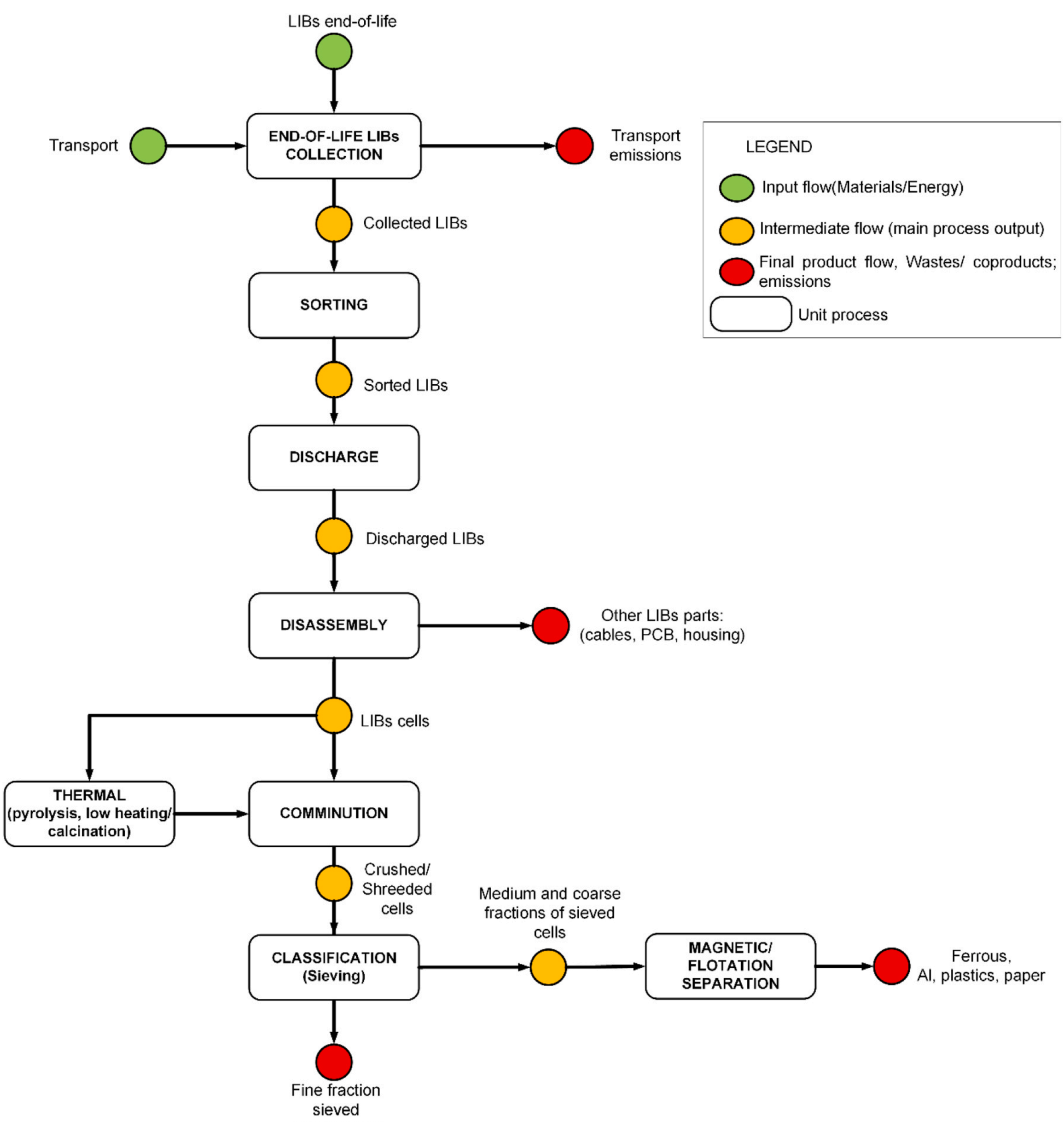

Fig. 1. LIBs pretreatment process.

Pyrometallurgical recycling is high-temperature thermal processing [61,114]. Typical processes are smelting, pyrolysis, incineration, or torrefaction/calcination [99]. Heating LIBs waste in high-temperature furnaces reduces metal oxides into metal alloys containing Cu, Co, Fe, and Ni [66,79]. Plastics, organic solvents, and graphite are degraded and provide heat during combustion [186]. Thus, the products of the pyrometallurgical process are a fraction of metallic alloys, slag, and gases [61]. The metal alloy is subsequently treated and purified in a hydrometallurgical process for refining and recovering metals [186]. The advantages of pyrometallurgical recycling lie in the fact that it is a simple process, in which a large amount of spent batteries can be processed, without the need for pretreatment processes [99,79,74]. Its main disadvantages are the low recovery rates of materials, the loss of metals such as lithium and manganese in the slag, the expensive treatment of gases (and, in many cases, the absence of this), the high energy consumption and the need for post-processing to recover pure metals [66,61,74]. If the objective is to produce battery-grade cathode materials, in a closed loop, additional hydrometallurgical treatment is required [171].

Hydrometallurgical treatment is chemical processing that uses aqueous solutions containing strong chemical reagents to leach metals from the active material (black mass) [61,125]. After leaching, metals are recovered through purification, precipitation, and solvent extraction at low temperatures [79,74]. Hydrometallurgy recovers metals with high efficiency and purity [27], but requires large amounts of chemical solvents and water [82]. The possibility of cross-contamination and the generation of contaminated effluents are also disadvantages of this process [61]. As an alternative to inorganic chemical reagents, some studies have suggested substitution by organic acids, reducing the potential for secondary pollution and treatment of wastewater and gases [114,122,74]. Biohydrometallurgy has been developed for LIBs recycling [61,138]. In biohydrometallurgy, inorganic chemicals are replaced by microorganisms in bioleaching that carry out metal solubilization. However, this technology is at an early stage of investigation; several parameters still need to be defined for industrial scale implementation [138,125].

Direct, or functional, recycling performs the reconditioning and recovery of negative and positive electrode materials for reuse in LIBs, avoiding the separation and dissolution of multiple metallic ions in their elemental form or the need for smelting or leaching [61,79]. It is considered a more economical and environmentally promising technology for closed-loop recycling [171]. The main advantage of direct recycling is that almost of all battery is recovered for reuse later, shortening the recycling cycle and the requirement for materials and energy [82]. However, there is still a lack of further developments in the direct recycling of the negative electrode. The current literature has focused on the cathode, due to the high value of its metals [171]. There are still many challenges to effective direct recycling, such as eliminating aluminum contamination during material separation from the current collector, which causes poor electrochemical performance of the recovered cathode material [61]. Direct recycling processes are still unable to handle mixed batteries with different cathode compositions [126].

Although the recycling of end-of-life LIBs contributes to achieving a circular economy and sustainable development, guidelines for proper collection, storage, transport and recycling are still under development [130]. There is no simple way to recycle LIBs, as they are complex and constantly evolving with product design, and all the available recycling methods have advantages and disadvantages [55]. The currently available recycling processes have the potential to consume a large amount of auxiliary resources, such as chemical inputs, energy, and water, and generate emissions and hazardous solid waste, which can contaminate the air, water, and soil [82,129,138]. Thorough investigation, in a life cycle approach, of the environmental loads of each unit process employed in the different recycling methods is important to guide improvements of LIBs recycling processes [55].

2.3. Risk of pollutants in LIBs recycling

Due to the complex composition of LIBs, all activities involved in end-of-life management of LIBs are particularly concerning regarding the potential for severe environmental contamination [141]. Disposal of LIBs in landfills can lead to soil and water contamination due to leaching of heavy metals [77] or other compounds added in the housings, electrolytes, or binders [106]. Although LIBs recycling provides an appropriate pathway for material recovery and avoiding of the disposal of LIBs in landfills, pollution can also occur during recycling through the release of toxic gases, generation of contaminated wastewater, and residual solid waste [8]. Wastewater from LIBs recycling may contain heavy metals [184] and other potentially toxic substances such as fluorine, hydrogen fluoride, and phosphates [141,106]. Toxic gases may be released during mechanical and thermal pretreatment [65] pyrometallurgical or leaching [133]. Mechanical and thermal processing applied in LIBs recycling can lead to the release of toxic gases due to electrolyte volatilization and degradation of organic components [65,154]. Thermal runaway in LIBs can occur due to physical, electrical, or thermal abuse, such as heating, puncture, short circuit, or crushing, posing risks of explosions, fires, and release of toxic smoke and hazardous gases [96,109]. Table 2 presents a summary of potential pollutants that may be released during different stages and processes applied in LIBs recycling due to chemical complex interactions of materials in LIBs.

Due to the high energy density and potential reactivity and flammability of LIBs the risk of fires caused by LIBs waste in transport, recycling, sorting, storage and landfilling operations is an important safety aspect in the management of LIBs life cycle [26,29,46]. In recent years several incidents of explosions and fires during the use, transport and treatment of LIBs waste have been reported [80,67]. A study carried out by the Environmental Protection Agency in the United States revealed that the number of material recovery facilities affected by fires caused by LIBs have increased exponentially in recent years, growing from only two fires being reported at a single facility in 2013 to 65 fires reported across 16 different facilities in 2020 [46]. In the United Kingdom it is estimated that 48% of waste fires are caused by LIBs, a total of 201 fires each year [21]. Fires caused by LIBs are of particular concern as they are difficult to control and can cause serious damage, such as deaths, environmental pollution and economic losses of facilities and valuable recyclable materials [26,29]. For a detailed and comprehensive view of the different risks across the LIBs life cycle, including a detailed analysis of end-of-life fire incidents see the study by Christensen et al. [29]. An important issue in facing the challenges related to fires caused by LIBs refers to the lack of knowledge about the potential environmental impacts of these incidents due to the release of toxic gases into the air and the contamination of the water used to extinguish the fires [29]. These issues should still be the subject of further studies, including LCA studies to verify the potential impacts on human health and ecosystems.

2.4. EU battery legislation and role of LCA

How LIBs are relevant to different key areas for achieving low carbon economies, such as the decarbonization of transport systems, resource efficiency, and waste management [11,58] the development and improvement of policies and legislation that addresses all stages of LIBs life cycle has been made in some countries and regions, as a most recent example the European Union (EU) [101,110]. In 2023, the EU approved a new comprehensive regulation EU-2023/1542 [50] on batteries and batteries waste, including LIBs. The new regulation aims to improve safety and sustainability standards, from the extraction of raw materials at the end-of-life management of batteries, as well as the industrial strengthening of the EU through the creation of value chains in the domestic market to guarantee the supply of critical materials and LIBs [58]. Although Europe does not dominate the LIB supply chain, as it is the second largest consumer, changes to battery legislation in the EU

Table 2
Risk of pollutants in LIBs recycling.

Source (material/component)	Pollutants	Chemical formula	Potential release in LIBs recycling	Emissions types	Reference
Combustion of graphite, organic materials and electrolyte oxidation	Carbon monoxide	CO	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[4,36,96,97,109, 106,113,180]
Combustion of graphite, organic materials and electrolyte oxidation	Carbon dioxide	CO_2	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[65,96,97,106, 113,180]
Leaching acids			Acid leaching step	Toxic gases	[133]
	Hydrogen	H_2		Toxic gases	[113,180]
White vapour			Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[106]
Discharging			Discharging with saline solution	Toxic gases	[23]
Separator	Methane	CH_4	Thermal treatment	Toxic gases	[4,65,113,180]
	Ethane	C_2H_6	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[113]; [65]; [106]; [4]
Leaching acids			Leaching step	Toxic gases	[4]
Decomposition from PVDF binder and LiPF6	Ethene	C_2H_4	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[4,113,180]
Decomposition from PVDF binder and LiPF6	Carbon sulfide	CS	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[109]
Decomposition from PVDF binder and LiPF6	Lithium fluoride	LiF	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[109]
Decomposition from PVDF binder and LiPF6	Fluorine	F	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[96]; [65]
Electrolyte	Fluorine		Leaching step	Toxic wastewater	[163]
Decomposition from PVDF binder and LiPF7	Hydrogen fluoride	HF	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[36,84,83,96,97, 109,180]
Electrolyte	Hydrogen fluoride	HF	Leaching step	Toxic wastewater	[106]
Disposal	Hydrogen fluoride	HF	Solid waste	Leaching of solid waste	[106]
Decomposition from organic and electrolyte	Phosphoryl fluoride	POF_3	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[65,83,180]
Decomposition from organic and electrolyte	Phosphoric acid	H_3PO_4	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[109]
Decomposition from organic and electrolyte	Phosphoric acid	H_3PO_5		Wastewater	[169]
Decomposition from organic and electrolyte	Boric acid	H_3BO_3	Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[109]
Leaching acids	Chlorine	Cl_2	Leaching step	Toxic gases	[133]; [38]
Leaching acids	Sulfur dioxide	SO_2	Leaching step	Toxic gases	[133]; [104]
White vapour			Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[106]
Leaching acids	Phosphorus	P	Leaching step	Toxic gases	[133]
Leaching acids	Nitrogen oxides	NO_x	Leaching step	Toxic gases	[38]
White vapour			Thermal pretreatment / Thermal runway / Heating /Fires/ Mechanical treatment	Toxic gases	[106]
Decomposition of numerous additives in the electrolytes, in which F, Cl, Br, N, etc. could be the essential elements	Hydrogen bromide	HBr	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[65]
	Carbonyl fluoride	COF_2	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[36,180]
	Acrolein	C_3H_4O	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[36,65,109,180]
Decomposition of numerous additives in the electrolytes, in which F, Cl, Br, N, could be the essential elements	Hydrogen chloride	HCl	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[36]; [65]
	Formaldehyde	CH_2O	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gases	[36]; [65]

(continued on next page)

Table 2 (*continued*)

Source (material/component)	Pollutants	Chemical formula	Potential release in LIBs recycling	Emissions types	Reference
Electrolyte organic solvents	Dimethyl carbonate	$C_3H_6O_3$	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gas / Wastewater	[36]; [65]
	Ethyl methyl carbonate	$C_4H_8O_3$	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gas / Wastewater	[36]; [109]; [65]
	Diethylene carbonate	$C_5H_{10}O_3$	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gas / Wastewater	[36]; [109]; [65]
	Ethylene carbonate	$C_3H_4O_3$	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gas / Wastewater	[36]; [109]; [65]
	Propylene carbonate	$C_4H_6O_3$	Thermal treatment (pyrolysis or combustion) / Mechanical treatment	Toxic gas	[36]
Separator	Styrene	C_8H_8	Thermal treatment	Toxic gas	[109]; [65]
Separator	Benzene	C_6H_6	Thermal treatment	Toxic gas	[109]; [65]
	Tuolene	$C_6H_5CH_3$		Toxic gas	[109]
	Biphenyl	$C_{12}H_{10}$		Toxic gas	[109]
All LIB	Particulate matter	(e.g., Dust, C, Al, Mn e F)	Dismantling / mechanical treatment / explosion / landfill	Toxic gas	[106]
	Carbon Tetrafluoride	CF_4	Recycling in general	Toxic gas	[180]
	Cyclohexylbenzene	$C_{12}H_{16}$	Recycling in general	Wastewater	[9]
Cathode	Lithium	Li	Recycling in general	Wastewater/ Leaching solid waste	[184]; [57]; [111]
Cathode	Aluminium	Al	Recycling in general	Wastewater/ Leaching solid waste	[184]
Cathode	Magnesium	Mg	Recycling in general	Wastewater	[184]
Cathode	Nickel	Ni	Recycling in general	Wastewater/ Leaching solid waste	[169]
Cathode	Iron	Fe	Recycling in general	Wastewater/ Leaching solid waste	[169,184]
Leaching acids	Sulfuric acid	H_2SO_4	Leaching	Wastewater	[169]
Disposal	Sulfuric acid	H_2SO_4	Solid waste from recycling processes	Leaching of solid waste	[106]
	Calcium	Ca	Discharge, leaching, extraction	Wastewater	[184]; [111]
Discharge, leaching, extraction	Sodium	Na	Discharge, leaching, extraction	Wastewater	[184]; [139]; [57]; [111]
Housing	Silicon dioxide	SiO_2	Discharge, leaching, extraction	Wastewater	[111]
	Dimethyl fluorophosphate	$C_2H_6FO_3P$	Solid waste	Leaching of solid waste	[106]
	Diethyl fluorophosphate	C_4H10FO_3P		Leaching of solid waste	[106]
Polyamide from some cell pouch housing	Ammonia	NH_3		Toxic gas	[4]
Recycling wastewater	Ethylene glycol	CH_2OHCH_2OH		Wastewater	[9]
Leaching acids		SO_4		Wastewater	[57]
Cathode		Co	Leaching / disposal	Leaching of solid waste	[77]

will mean changes to global value chains are made to meet new regulations, as well as contributing to the development of recycling markets [101].

The main changes highlighted in the new regulation (EU) 2023/1542 [50] to direct the implementation of more sustainable supply chains for LIBs include: i) the mandatory Carbon Footprint declaration per stage of the LIBs life cycle (from raw material extraction to end-of-life); ii) mandatory declaration of recycled content and minimum recycled content requirements; iii) labelling rules; iv) new targets for LIBs waste collection, efficiency of recycling and recovery of materials; v) mandatory systems due diligence and the implementation of the digital battery passport to increase the transparency on LIBs life cycle and potential social and environmental impacts [11,58,101].

The cradle-to-grave life cycle perspective is recognized as an important element in LIBs management, having been explicitly and directly incorporated into the new Regulation (EU) 2023/1542 of the EU through the mandatory disclosure Carbon Footprint of LIBs placed on the EU market [50]. The new regulation brings major changes and advances in the coverage of the sustainability impacts of the LIBs life cycle [101], highlighting the systematic and comprehensive use of assessments based on LCA approaches to provide information on the Carbon Footprint and other environmental and social impacts of LIBs, as well as guiding the definition of Carbon footprint performance classes and maximum carbon thresholds, important mechanism to reduce CO2 emissions throughout of LIBs life cycle [50].

With the mandatory disclosure of Carbon footprint along the LIBs life cycle, conducting LCA studies will be essential for producers, importers, distributors and recyclers to meet the objectives of the new regulation, providing information on environmental impacts of the production of raw materials, manufacturing, distribution, waste collection and different end-of-life management strategies for the LIBs, both to meet Carbon Footprint limits and for material recovery. In this sense, LCA studies of the impacts and benefits of potential recycling processes for the recovery of materials to meet established material recovery targets also have great relevance for meeting the maximum Carbon Footprint threshold requirements of LIBs.

3. Methods

The research method is a systematic literature review with content analysis. The systematic review framework uses previous recommendations, such as the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) [103] and STARR-LCA checklist items [188]. For the content analysis, this study used as a general reference the data classification categories developed for a previous systematic review of LCAs in waste management systems [86], according to the four phases of the LCA methodology in standards ISO 14040 [70], ISO 14044 [71], and ILCD guidelines [48].

3.1. Identification and collection of studies

The identification and collection of studies were carried out in January 2023 in Scopus and Web of Science (WoS) databases, applying the following search terms and Boolean operators in titles, abstracts, and keywords: ("Life Cycle Assessment" or "Life Cycle Analysis") AND ("Hydrometallurgy" or "Hydrometallurgical" or "Pyrometallurgical" or "Pyrometallurgy" or "Biohydrometallurgy" or "Biohydrometallurgical" or “Bioleaching" or "Recovery" or "Recycling") AND ("Battery" or "Batteries") AND ("lithium-ion" or "lithium" or "LIBs" or "lithium-polymer"). Search terms are derived from initial exploration of the scientific

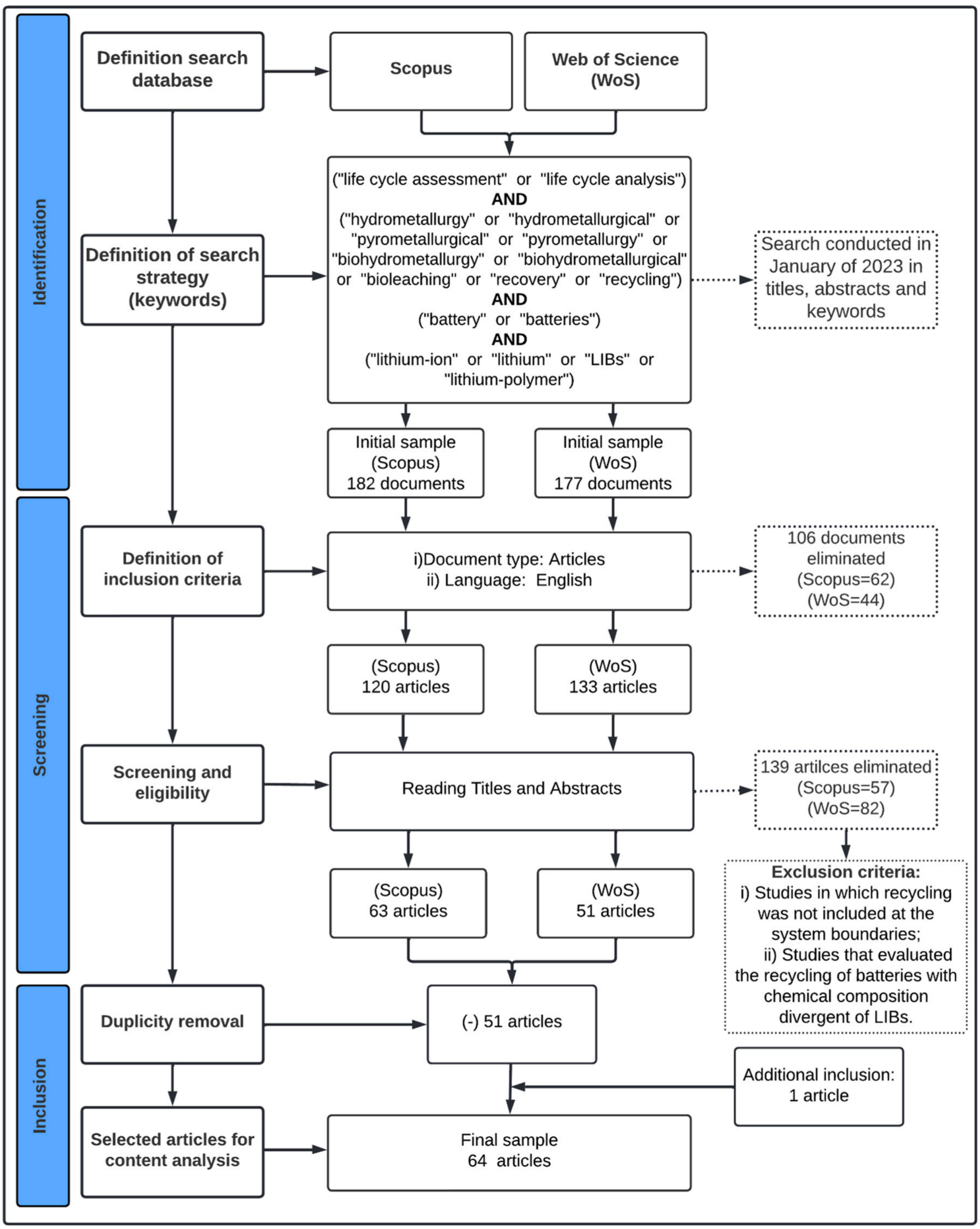

Fig. 2. PRISMA protocol of the research.

literature on LIBs waste and applicable recycling processes (e.g., [61]; [105]. The initial exploration allowed for identifying specific terms about the treatment technologies applied in LIBs recycling (e.g., “Hydrometallurgical”, “Pyrometallurgical”, and “Direct recycling”). Broader terms such as “recycling” and “recovery” also were included to allow identifying studies where terms related to the specific treatment process (e.g., Hydrometallurgical) are not described in the title, abstract or keywords.

The initial search resulted in 359 documents (182 in Scopus and 177 in WoS), including books, book chapters, research, and conference papers. Then, two inclusion filters were applied: i) only peer-reviewed article document type; ii) written in English. As a result of this step, 106 documents were eliminated (Scopus=62 and WoS=44) and 253 articles were selected for the next screening step. The screening was conducted by reading the titles and abstracts, with the objective of selecting only those in which LCA was applied in LIBs recycling, eliminating articles with the following criteria: i) studies in which recycling was not included in the system boundary (e.g., [95]; [170]; [91]; ii) studies evaluating the recycling of batteries with different chemical compositions from LIBs, such as NiMH (e.g., [140] or Zinc Alkaline (e.g., [152]. As a result of this step, 139 articles were eliminated (Scopus=57 and WoS=82) and 114 articles were considered adequate (Scopus=63 and WoS=51). Finally, duplicates were eliminated (51 articles) and 63 articles were selected for content analysis. In addition, a study published in a journal of another database was included because of direct recommendation [143], resulting in a final sample of 64 articles for content analysis. Fig. 2 shows the PRISMA research protocol and the review flow.

3.2. Content analysis

After collecting and screening the studies, a data classification framework was structured for content analysis. Data collection was organized in an Excel spreadsheet to extract data on methodological choices from each phase of the LCA. Categories of data collection are adapted from the data collection protocol carried out by Laurent et al. [86]. Some data categories were adapted to the context of the LIBs, for example, the LIBs type (chemical composition) and product of origin of LIBs waste (e.g., Electric vehicles or consumer electronics). Frame 1 in Supplementary Material S1 provides the data collection protocol.

4. Results and discussion

The results are presented and discussed according to the data collection protocol and following the sequence of the methodological phases of the LCA. First, an overview of the reviewed studies is presented, identifying the types of recycling evaluated, stages and processes included in the system boundaries, products avoided, and the distribution of publications by year and country, to provide a general picture of the state-of-the-art and temporal trend. Next, the methodological adequacy characteristics of the studies in each LCA phase are summarized. We highlighted in the main text the key findings from the content analysis. Some additional results may be found in Supplementary Material S3.

4.1. Overview of studies on the LCA of LIBs recycling

Fig. 3 provides an overview of the system boundaries, life cycle stages, recycling technologies assessed, avoided processes, and status of available data for each stage of LIBs recycling. The identification and

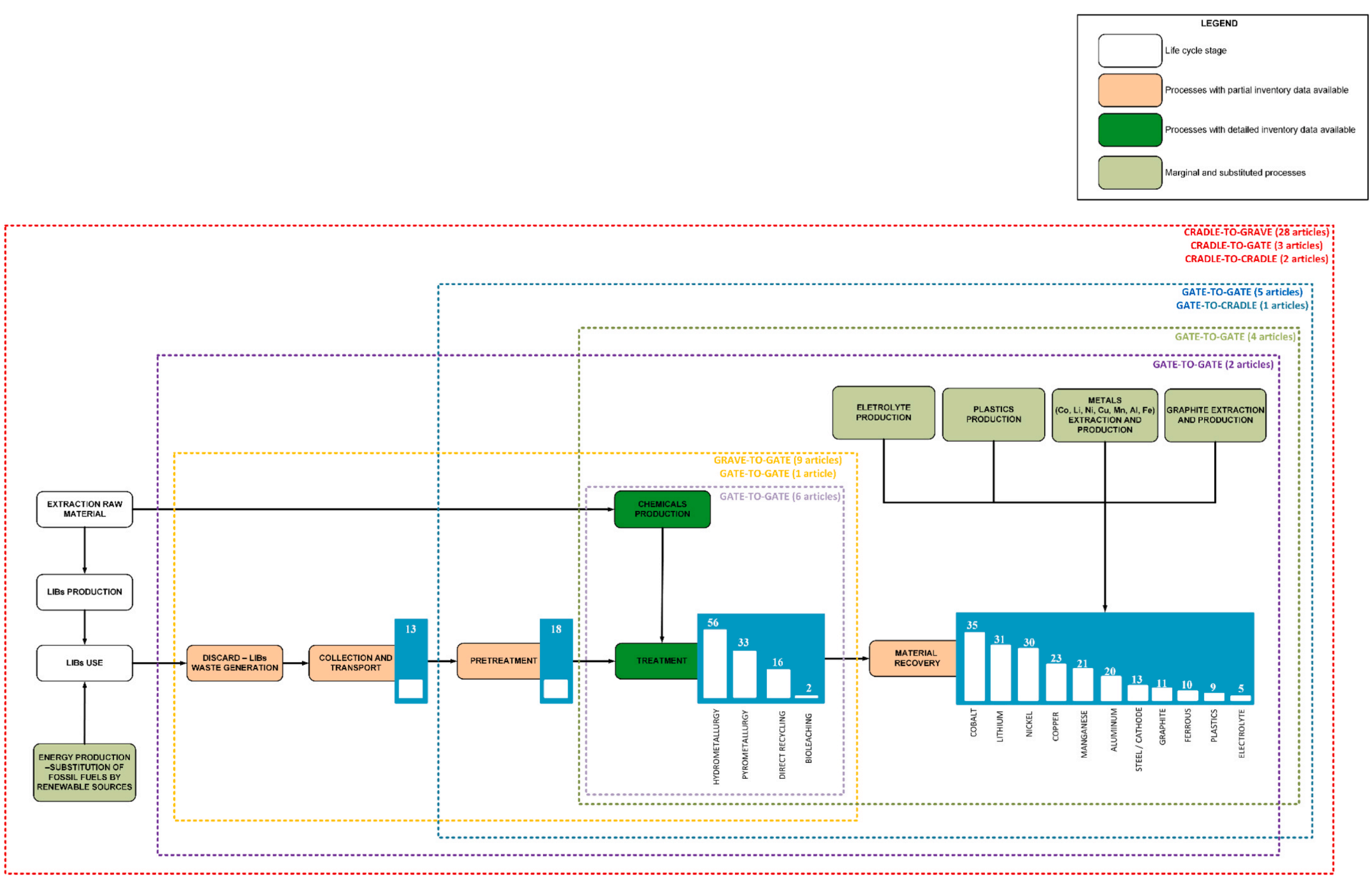

Fig. 3. Overview of LCA of recycling LIBs.

scope detailed, including LIB type, unit processes, inputs, functional unit, system boundaries, impact categories e impact methods, of 64 reviewed studies is provided in Supplementary material S2. The studies are identified by a number of Identification (ID) to allow the traceability of the results.

Fig. 3 shows that studies were identified in the four recycling processes: i) hydrometallurgical, ii) pyrometallurgical, iii) direct recycling, and iv) bioleaching. Hydrometallurgical and pyrometallurgical recycling are the most evaluated while Direct Recycling and Bioleaching have fewer evaluations. Highlights that the main target materials of LIBs recycling are metals Co, Li, Ni, Mn Cu, with few studies include electrolyte recycling.

Fig. 3 also presents the system boundaries found in the reviewed studies. Six different system boundaries have been identified: i) Cradle-to-Grave (e.g., [32]; [104]; ii) Cradle-to-Gate (e.g., [183]; iii) Gate-to-Gate (e.g., [3,22,93,167]; iv) Grave-to-Gate (e.g., [17,75], v) Gate-to-Cradle (e.g., [43,14]; and vi) Cradle-to-Cradle (e.g., [149]; [24]. It is important to highlight that our results indicate an inconsistency in the definition of the system boundary among the studies reviewed. As illustrated in Figs. 3, 18 studies assessed recycling LIBs using a boundary Gate-to-Gate but included different unit processes within the system boundary. For example, 6 studies declared the boundary as Gate-to-Gate, focusing only on the leaching and extraction of the materials processes, while others included within the same system boundary (Gate-to-Gate) the collection, pretreatment, recycling processes, and avoided products. Therefore, caution must be exercised when comparing studies with the same system boundary, carefully analyzing the specific unit processes included.

4.1.1. Distribution of publications by year and country

The sample of selected articles covers the period from 2011 to 2023 (Fig. 4). Most of the studies (69.76%; 43 articles) were conducted in the last three years, denoting that the assessment of impacts related to the recycling of LIBs is an emerging and growing issue. This growing interest in the impacts of recycling is explained by the fact that LIBs are at the centre of decarbonization strategies for mobility and energy storage from renewable sources, but at the same time consume much finite mineral resources for production [148,128].

4.1.2. Distribution of publications by country

The analysis of publications by country and by recycling treatment (Fig. 5) reveals that China and the USA dominate publications on LCA of LIBs recycling for the three main recycling technologies. Studies conducted in European countries account for a total of 31.25% of the studies. However, within the European context, studies mainly focus on pyrometallurgical and hydrometallurgical recycling. This domain of studies in China, USA, and European countries can be explained by the fact that together with Europe, China and USA are the three largest LIBs markets and concerns about LIBs end-of-life management have made recycling advanced in these countries [110,179]. Furthermore, studies have focused on pyrometallurgical and hydrometallurgical technologies, while direct recycling and bioleaching technologies are still investigated. Direct recycling is assessed only considering three countries (USA, China, Germany) and Bioleaching only two (USA and Spain).

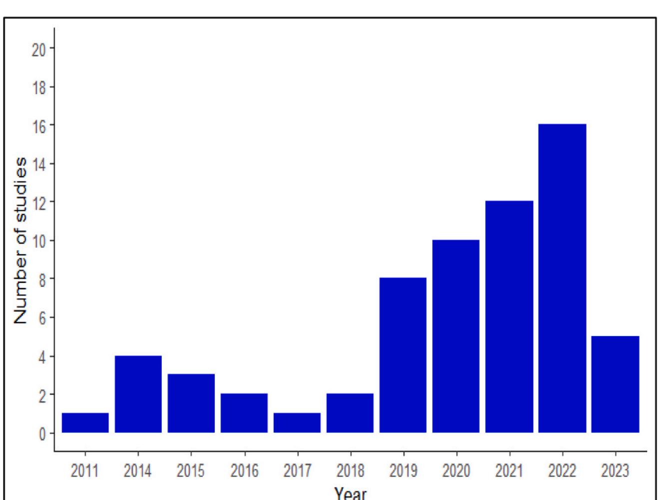

Fig. 4. Temporal distribution of reviewed studies.

The analysis of the geographical context reveals that research on recycling LIBs is focused on two countries, with many countries and regions of the world are still underrepresented in the assessment of LIBs recycling, mainly developing countries. Brazil and India, for instance, have a significant demand for LIBs in electric vehicles and consumer electronics, resulting in substantial flows of LIBs waste. The lack of research on the environmental impacts of recycling in different countries is a critical issue, mainly because the pollution potential of LIBs waste streams is high. Furthermore, most studies are conducted in the context of developed countries and cannot be used as models for the development of public policies in developing countries, as operational conditions, infrastructure, issues related to the energy matrix, and mitigation of secondary pollution differ greatly.

4.2. LCA methodological review

The results of the content analysis are presented according to the sequential LCA phases, indicating what was found in the studies and identifying best practices to overcome eventual methodological challenges.

4.2.1. Goal and scope

The six questions are not answered or partially answered in the goal definition (Table 3). 78.13% of studies do not provide answers for any of the six questions. The definition of limitations due to method and impact coverage was not present in any study. Questions 3, 4, 5, and 6 are rarely stated. The intended application is answered in 15.6% of studies. The lack of delimitation of the intended application of the results, the identification of the target audience, and limitations compromise the reliability of the studies [135].

Only one study explicitly mentions the context to justify the choice of LCA attributional modeling [127], and another study does so to justify the choice of consequential modeling, but without mentioning the reference guidelines [156]. Defining the decision context (iii) is an important step in goal and scope, as it determines the type of approach to inventory modeling, attributional or consequential, that should be applied, as well as the solutions to resolve multifunctionality [48].

These results indicate that the LCAs of LIBs recycling are being conducted in a more limited spectrum, with no in-depth analysis of the issues for defining the goal, context situation, and where the results can be applied, which limits further analysis. The choice of inventory modeling approach should be based on the context analysis, not arbitrarily. Future studies on LIBs recycling should clearly answer the six questions and the decision context for goal definition. For these definitions, practitioners are recommended to use the guidelines provided in the ILCD [48].

4.2.2. Function, functional unit, and reference flow

Two functions evaluated in LIBs recycling systems were identified: i) the treatment of LIBs waste; ii) the generation of secondary materials as substitutes of primary material. Although both functions are implicitly present in the studies, the definition of the system's function is not discussed in depth. The proper definition of which function is evaluated guides the definition of the Functional Unit (FU) and Reference Flow (RF) in LCA [16,54]. It is suggested that future studies clearly define which system functions are being evaluated, for example, delimiting

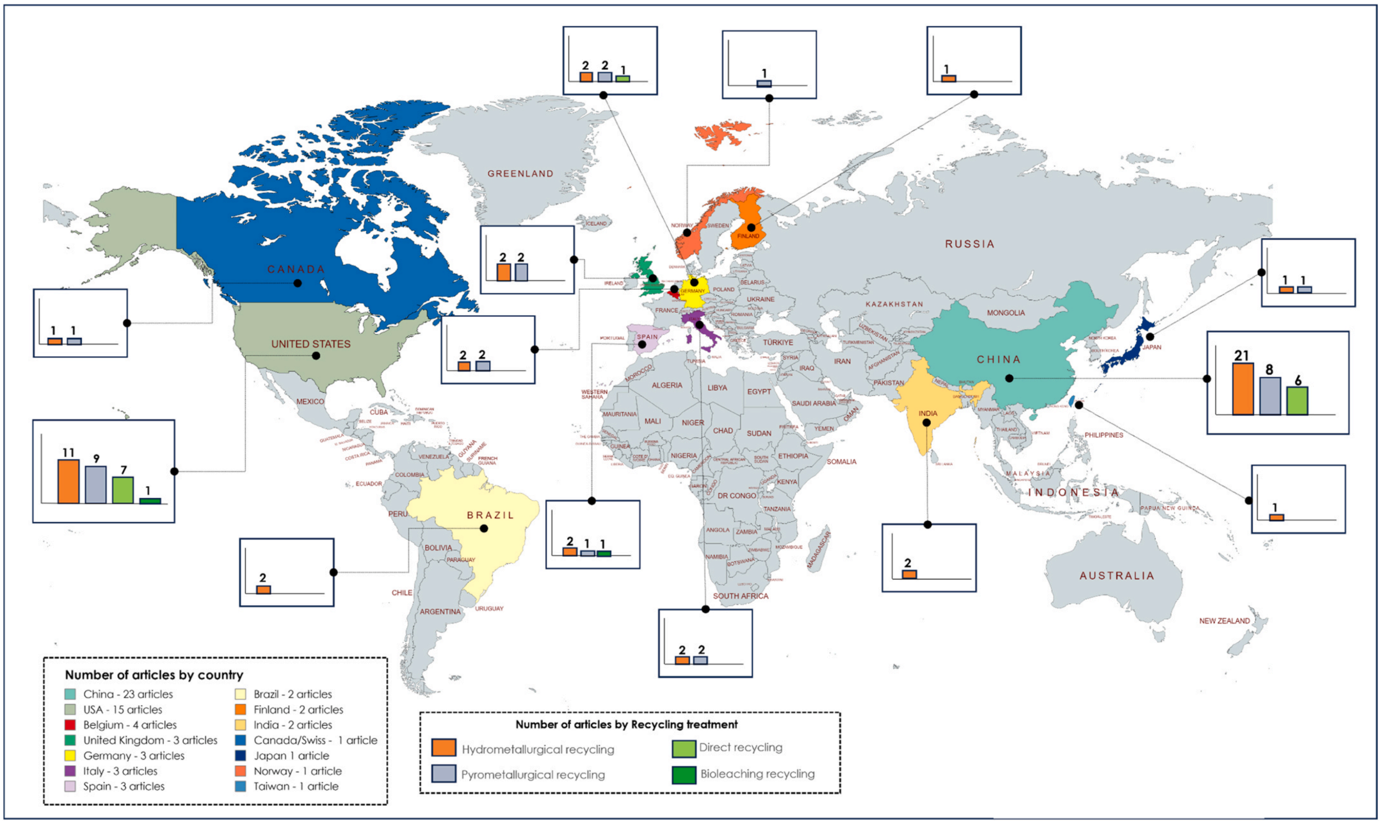

Fig. 5. Distribution of LCA studies by country and treatment type.

Table 3
Answers to the six questions for Goal definition according to ISO 14040 [70], ISO 14044 [71], and ILCD [47].

Six questions for Goal definition	% of answering studies	study ID
i)-Intended application	15.63%	19–22–25–28–29–30–31–32–33–47
ii)-Limitations due to the method, assumptions, and impact coverage	0	-
iii)-Reasons for carrying out the study and decision-context	3.13%	11–32
iv)-Target audience	3.13%	11–19
v)-Comparative studies to be disclosed to the public	1.56%	22
vi)-Commissioner of the study and other influential actors	3.13%	11–28
None	78.13%	-

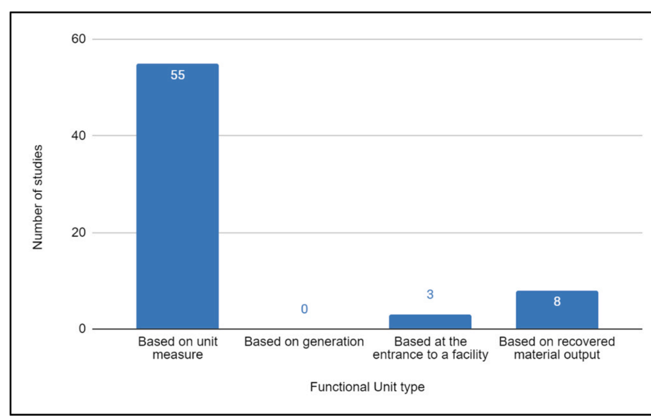

Fig. 6. Functional Unit types identified in the reviewed studies.

whether the function of waste treatment, recovery of recycled materials or both, are being evaluated.

Fig. 6 presents the distribution of FU types found in the revised LIBs recycling studies. Table S3.1 in Supplementary Material S3 provides examples of FU types by studies. The most chosen FU is based on unit measurement, with 55 studies (85.93%). The second most used type of FU was based on the output of the recovered material (8 studies). Three studies are based on the processing capacity of a recycling plant. Two studies use two FU for the pretreatment stage they use a unit measure “1 kg LCO obtained from spent LIBs” [168]; “Extraction of 1 kg of cathode” [143] and for the separation and metal extraction stages a FU based on the output “100% Co separated from 1 kg LCO” and “100% Recycled Co” [168]; “Production (recovery) of 1 kg of metallic cobalt” [143]. No study has been conducted based on the FU of total LIBs waste generation in a region in a period.

Another characteristic that emerges from the studies, due to the choice of the type of unit FU, is the fact that the reference flows are identical to the FU since the quantity described as a functional unit is already the physical flow of the system [16]. This may indicate that practitioners intend to provide a rounded reference stream or that they do not differentiate the meaning of FU and RF [86]. Therefore, there is a lack of more complete descriptions of the FU, which is a direct effect of the little detail of the function delivered. For example, describing whether the evaluated function is the treatment of waste or the recovery of materials or both, helps to better define the expected performance of the system (FU) and the amount of physical flows needed to deliver that performance (RF). It is recommended that studies include in the FU definition detailed descriptions of what, how much, for how long and where it is being evaluated [16]. For example, when defining a functional unit based on the recovery of materials, it is necessary to define the quantity of LIB cells that must enter the system to deliver this evaluated quantity as an output, as well of the LIBs chemical type.

Due to the different chemical compositions of LIBs currently available on the market, with different applications (e.g., electronics, electric vehicles, stationary storage) and that can be recycled with different methods, it is recommended that the FU of LIBs recycling describes the characteristics of the system, such as the composition of the treated LIB, source product, type of treatment used for recycling and geographic location (country where recycling process are executed). For example, "Treatment of 1 kg/ton of LIBs waste with NMC cathode from electric vehicles using hydrometallurgical treatment in country x" or "Treatment of 1 kg/ton of LIBs waste with NMC cathode from electric vehicles using hydrometallurgical recycling in country x to recovery x kg of Co, y kg of Ni, z kg of Li."

A characteristic of studies in which the battery production is included in the system limit (Cradle-to-Grave) is the use of a FU that reflects the amount of energy that the LIBs delivers during its life cycle (kWh/kg) or the total lifespan, expressed as the maximum distance driven (km) that a LIBs travels in electric vehicles [88,136,144,94]. However, for the analysis of the end-of-life stage, the FU can be converted to a unitary basis. It is recommended that studies using the FU of energy capacity describe the list of materials and density energy and capacity of LIB studied, specifically linking the recycling inventory data to the same functional unit. This is important because the list of materials in a LIB and the energy density and capacity can vary considerably with the cell type, package configuration and battery size, since that directly comparing results per kWh of LIBs with notably different characteristics is not appropriate [33].

4.2.3. Defining system boundaries

Collection and transport processes are not included in the system boundaries of 83% of the studies. Only 11 studies mention the inclusion of transport for the collection of end-of-life LIBs (Table 4). However, two of these studies only present an estimated average mileage for collection and transport, 100 km [157] and 650 km [94], but do not provide transport related impact results. The lack of discussion of the results of transport impacts are justified due to the small contribution to the total impact in all categories, but the values of these impacts are not available [94] or are aggregated to the total results of the whole transport involved in the life cycle of LIBs, when the Cradle-to-Grave system limit is evaluated, with no distinction made between the impacts of end-of-life transport only [157]. Assuming an average distance of 2.500 miles for the collection and FU of 1 kg of battery LIBs from electric vehicles, it was estimated that 0.33 kg of CO_2 equivalent per kg of battery are emitted by collection, which would represent approximately 3.5% of total CO_2 equivalent emissions when using a pyrometallurgical process, and 4% when using a hydrometallurgical process or direct recycling in USA [30]. A comprehensive analysis of a reverse logistics network for collecting electric vehicle LIBs for California (USA) evaluated the impacts of collection and transportation for LIBs recycling [62]. The study concluded that the collection and transportation of batteries significantly contribute to environmental burdens, highlighting the impacts of

Table 4
Collection and transport for LIBs recycling.

Transport data for Collection LIBs					
Transport type	Distance	FU	Country	Assumptions	ID
-	900 miles (1.448 km)	1 kg of recovered cobalt	USA	Transport of black mass	3
Truck	100 km	1 kg of recovered cobalt		Transport distance to recycling facilities and truck type (32 t) are assumptions based in literature (Zhou et al., 2021)	5
Train Truck	688 km 174 km	1 kWh of nominal capacity	China	LIB is first transported by freight train and then by freight truck to the sorting facility - based on average transport distances in China	8
Truck - Collection and Disassembly	100 miles (169 km)	1 kg of battery	USA	LIBs are recycled within the US and transported via truck.	10
Truck - To recycling facility	1.000 miles (1.609 km)		USA		
Truck - Collection and Disassembly	100 miles (169 km)	1 kg of battery	USA	LIBs are recycled within the USA and transported via train and truck.	10
Train - To recycling facility	1.000 miles (1.609 km)		USA		
Truck - Collection and Disassembly	100 miles (169 km)	1 kg of battery	USA	LIBs are recycled in China and transported via truck and ocean tanker.	10
Truck - port operations	260 miles (USA) 740 miles (China)		USA / China		
Ship	19.270 nautical miles (35.688 km)		China		
-	0.1 tkm (100 g *1 km)	1 kg treated	Belgium	-	14
Road	888.52 km	Production of 1 kg of NiCoMn ternary cathode materials	China	LIBs are recycled in China and transported by road using an average distance of cities in China.	17
Train Truck	688 km 174 km	1 kg of LIBs cell	China	LIBs are recycled in China and transported via freight train and freight truck.	19
-	-	1 kWh NCM811 Battery for an electric vehicle	USA	Transport included in the GREET model.	20
Truck	120 km	1 kg of Cobalt	Brazil	LIBs are recycled in Brazil and transported via small trucks with a load capacity of 3.5–7.5 tons.	21
-	650 km	Weight of a LIB of electric vehicle	China	LIBs are recycled in China and the average distance to transport is 650 km.	33
Truck	2.500 miles (4.023 km)	1 kg of battery	USA	LIBs are recycled in USA and transported via truck.	50
Truck Rail	243 miles (391 km)	Plant capacity: 7000 t/year	USA	LIBs are recycled in USA and transported via rail and truck. Battery collection points: car dealerships.	58
-	-	1 battery	China	LIBs are recycled in China and long transports were assumed in end-of-life treatment.	61

pollutants emitted into the air, such as particulate matter, volatile organic compounds, SO_2 and NOx.

Some studies justify the absence of collection and transport within the system boundaries due to a lack of data or high uncertainty associated with available data [104,127,172]. Another justification for exclusion is based on the lack of recycling systems operating on a large scale, which makes it difficult to quantify transport distances [177]. However, the vast majority (82.8% of studies) only mention that this stage is not included in the system limit, but without presenting a justification for the exclusion, contrary to the principles of the LCA standards. Another type of transport inclusion rarely mentioned in the studies is the transport of consumables. The quantification of the implications of transport distances incorporated in database processes, such as Ecoinvent, for chemical products, auxiliary materials and avoided primary materials can be explained and evaluated in terms of the percentage of contribution to the total impacts assessed [132]. Table 4 summarizes the distances, types of vehicles and assumptions made in studies that included collection and transport at the system boundary.

Although pretreatment techniques for LIBs recycling present importance in the efficiency of recovering valuable materials from LIBs [115] and reducing of energy consumption in subsequent processes [123], the environmental aspects of pretreatment processes are little explored in the LCA available in the literature. Some authors justify the absence of pretreatment due to the lack of reliable data, while others because the object of analysis is only the recovery process of the target material [128]. Another study justifies the exclusion of pretreatment due to a lack of data on the location and methods of discharge and dismantling [127]. Other studies exclude it from the system limit, stating that the impacts of these steps were considered small, due to the low temperatures used in the pretreatment [132]. However, the studies do not present details of the processes and the values of the impacts considered irrelevant.

Another missing aspect in the literature refers to the environmental implications of the final subproducts of recycling processes (e.g., characterization of final wastewater or solid wastes sent to landfills). This can be explained because data related to recycling and waste treatment are processes where direct measurements are possible and more reliable than landfill data which have to be partially modeled and where estimates are needed [112]. Landfill leachate composition and concentration can vary significantly depending on waste composition and moisture content, age, landfill type and meteorological condition [53, 77].

The generation of hazardous wastewater is a problem for the hydrometallurgical recycling of LIBs [61]. Pretreatment produces wastewater during brine discharge and the leaching process can generate wastewater with metals, plastic, electrolyte, graphite and solvents, which cause significant environmental damage [163]. Large volumes of wastewater contaminated with heavy and toxic metals are generated [168]. Recycling just one electric vehicle cell of 569 g can generate 6.6 kg of wastewater [23]. Extraction of one ton of metal from LIB leachate can generate 20.000 liters of oily effluents with a high potential for toxicity and carcinogenicity [168]. Wastewater from recycling LIBs may contain heavy metals, sodium, sulfates, and phosphate [23,78]. Direct emissions of phosphate and heavy metals to water contribute to impacts in freshwater eutrophication, freshwater ecotoxicity and marine ecotoxicity categories [23]. The recovery of acids, bases and precipitants, and the use of organic and biodegradable acids can improve the environmental profile of LIB recycling, reducing water consumption, wastewater generation and secondary pollution [23,78]. However, the relationships between the amount of waste treated and consumption of water and reagents must be carefully observed, as recycling processes with organic acids may require more water and reagents to treat the same amount of cathode than processes with inorganic acids [72].

4.2.4. *Definition of the LCI modeling framework*

The LCI modeling framework, that is, attributional or consequential is not stated in 55 articles (86%). This is a recurring problem in LCA studies, considered a serious failure since the definition of the type of LCI modeling is essential to select the range of collection and types of inventory data and the guidelines to solve multifunctionality problems [86,15]. Eight articles declared explicitly LCI modeling as Attributional [144,81,132,127,41,159,23,75]. Only one identified article modeled a Consequential LCI [156]. The rest of the studies apply an attributional approach, even if not explicitly stated.

In the only Consequential-LCA study, the displaced marginal processes were the energy production processes displaced by the storage of surplus renewable energy by the stationary LIBs and the marginal virgin material extraction processes avoided by recycling the materials at the end-of-life [156]. There is great uncertainty about the type of recycling for LIBs that will be dominant in the future, so the recycling processes were modeled as the current ones. They considered the recovery rates and displacement of production from virgin materials without an in-depth analysis of the changes in marginal processes due to recycling, not differing from the substitution approach commonly used in attributional studies.

Future studies need to deepen the discussion on the choice of the LCI modeling framework, considering the situation of the decision context, goal and scope of the study. A consequential approach in LCA of recycling LIBs is urgently required to clarify the consequences of changes due to material recovery in marginal process and environmental impacts profile.

4.2.5. *Types and chemical composition of LIBs in LCA studies*

The predominance of studies 68.75% (44 articles) evaluate the recycling of LIBs from electric vehicles. In the second position, 12.5% (8 studies) evaluate the recycling of LIBs from consumer electronics, such as cell phones and notebooks. Table S2 in Supplementary Material S3 provides the mapping of studies by type of LIBs waste stream and chemical composition. However, despite the importance of investigating the impacts of recycling LIBs from electric vehicles, other products also generate large flows of LIBs waste, such as portable electronics (e.g., smartphones, notebooks), widely consumed worldwide [20,100,7,110]. Further investigation is needed regarding the potential volume of waste LIBs generated by other products, as well as the impact of this volume on the environmental sphere since the type of collection, dismantling of products and amounts of recoverable materials can be very different for each waste stream. LIBs from Electric vehicles are larger and heavier, which may require more transport capacity and disassembly time than consumer electronics. In addition, there is a growing trend towards replacing LIBs chemistries for Electric vehicles with high cobalt and nickel content with lighter options such as LFP [68], which can difficult recycling of LIBs from portable electronic and Electric Vehicles together due to contamination issues.

LIBs cells can contain different chemical compositions, combining several metals to increase their capacity. Fig. 7 shows the LIBs chemical compositions recycled in the reviewed LCA studies. Seven types of chemical composition for the positive electrode were evaluated: LCO ($LiCoO_2$), NMC ($LiNiMnCoO_2$), NCA ($LiNiCoAlO_2$), LMO ($LiMn_2O_4$), LFP ($LiFePO_4$), LCP ($LiCoPO_4$) and LMP (Lithium-metal-polymer). The results indicate that NMC chemistry is the most investigated LIB chemistry in the studies, with 42 studies analyzing the potential impacts of recycling this type of cathode. This result can be explained because NMC is, currently, the most promising type of cathode for applications in Electric vehicles and consumer electronics [137]. The composition of LIBs is an important aspect because, depending on the composition of the input material, the economic and environmental viability of recycling is lost [90]. For example, the recycling of LFP cells is considered commercially unfeasible, because the chemical composition does not contain valuable metals, such as Co and Ni [90].

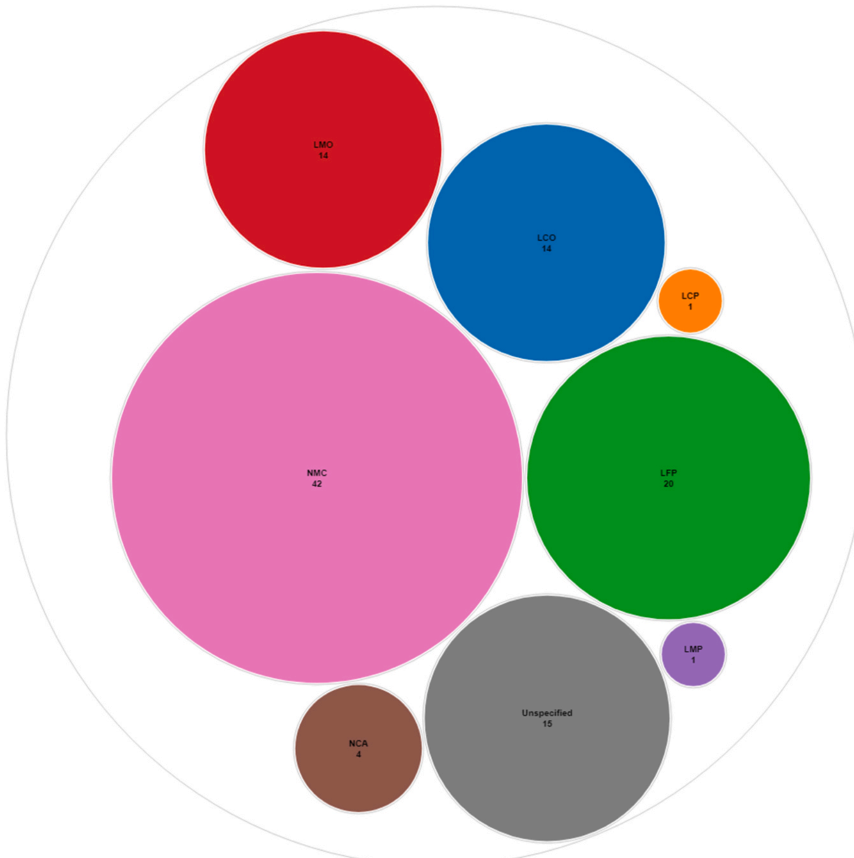

Fig. 7. Chemical composition of LIBs in reviewed studies.

4.3. Inventory analysis

Fig. 8 shows a mapping of data types and sources to inventory in the reviewed studies. Of the 64 reviewed studies, 57.81% (37 articles) use secondary data to conduct the studies and 42.19% (27 articles) use primary data, of these, 26.56% (17 articles) use data from laboratory scale, 4.69% (3 articles) use data from pilot scale, and 10.94% (7 articles) from industrial scale. For details of the type of data and sources used in each study see Supplementary Material S4.

Data from literature and LCA databases (e.g., Ecoinvent, GREET, BatPac (EverBatt)) have mostly been used to model foreground and background systems. Secondary data are used to model electricity supply, chemicals and other auxiliary materials production, waste treatment, as well as for parameter proxy of unit processes.

The primary data in a laboratory scale provides the energy demand, the amount of auxiliary materials, operational times needed and material recovery rate for unit operations, mainly leaching and material recovery experiments (e.g., [23,76,93,132]).

The primary data from pilot scale [34,183,81] provides data for processing 100 kg of material at the company ACCUREC in Germany are detailed in the study by [34], and the energy demand and the amount of auxiliary and operational materials required in a pilot plant of the industrial partner of the project DeMoBat Erlos GmbH from Germany [81].

The primary data from industrial scale for hydrometallurgy and

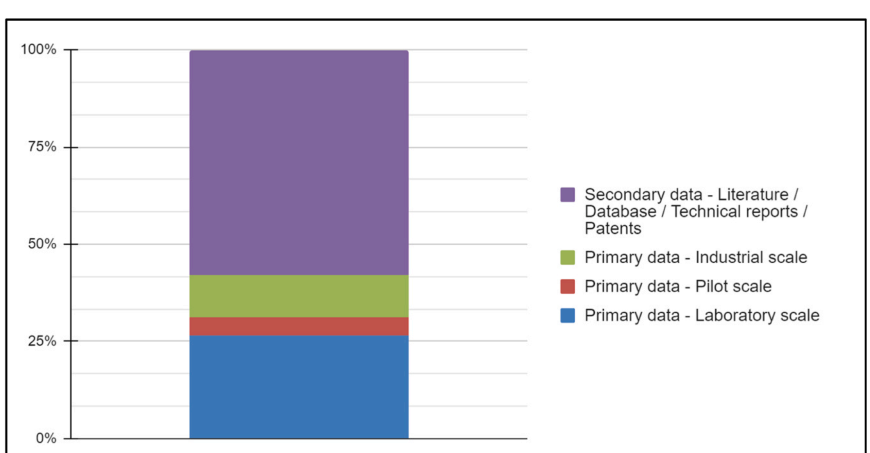

Fig. 8. Type and sources of inventory data.

pyrometallurgy were found in [60,147,104,127,73]. Primary hydrometallurgical recycling inventories were obtained from two recycling companies in China, one in Hunan province with an NMC recycling capacity of 100 kt/year and the other in Jiangxi province with an LFP recycling capacity of 25 kt/year [73]. Another study obtains primary data for an advanced hydrometallurgical process from Duesenfeld GmbH, Germany [104]. Industrial inventories for a DC plasma smelting for a pyrometallurgical process developed by Tetronics Company [127]. In addition, two studies [60,147] point out that they use primary data from recyclers in China, but they are not sufficiently transparent, as the data are provided in black-box, and it is not possible to track the inputs and outputs in each unit process.

Our results confirm that LCA data for LIB recycling are focused on a laboratory scale, with few studies presenting pilot and industrial scale data. Pilot and industrial plant data are mainly from plants in China and Germany. This scenario raises concerns, as the low availability of data on the scale of commercial production and the diversity of modeling options make it difficult and uncertain to assess the environmental impacts of LIBs [31]. Despite the number of studies in this field of LIBs recycling, a major problem remains the limited availability of transparent data based on different industries [12]. Lack of data leads to error propagation and reduces the reliability and robustness of several of the existing studies. There is often a lack of transparency in many LCA studies of LIBs, which makes it difficult to trace the results [8].

The analysis of the production scales evaluated in the studies shows that many studies are built based on laboratory data and then extrapolated to pilot or industrial scales. However, this should be interpreted with caution as yields, equipment, solvent types and resources may change depending on the scale analyzed. It is recognized that operations and processes on laboratory and pilot scales are inefficient, presenting low mass conversion yield [44,150]. For example, in many industrial plants the heat and power sources can be changed, as well as the solvents type and yield [121]. An industrial-scale plant has large-scale equipment that allows it to optimize many process material and energy flows, presenting large differences in process efficiency and operating conditions [121,134,153]. For example, leaching and metal extraction reactors can be quite different from the laboratory scale (reactor works in batches) while on the pilot or industrial scale the reactors can be continuous and present differences in the yield of inputs and outputs of the processes [44]. In the laboratory, the various steps are typically not directly linked and the type of equipment used are not comparable with reactors and machines in a commercial plant [121]. At larger production scales, some unit processes can be replaced or eliminated, such as separation or drying processes, reducing overall energy consumption [134]. But on the other hand, some parameters can be changed, such as the generation of more liquid effluents [134]. The tendency is that with increasing scale, energy consumption per kg produced/recycled decreases [121], and this may be influencing greater of the impact generation. As production scale increases, synergistic effects and automation can enhance efficiency, reduce material waste, affecting the entire inventory and generating environmental impacts [47]. To illustrate how different scales can have different resource consumption, Fig. 9 presents a brief comparison of the energy demand values for hydrometallurgical processes at laboratory and industrial scales, showing that the study at an industrial scale has lower energy consumption per kg of treated LIBs.

Therefore, for the case of LIBs recycling, recycling at laboratory and industrial scales do not seem to be directly comparable due to the high variability that parameters can present. In the example used at laboratory scale, a linear projection of energy consumption and impacts would result in much higher potential impacts, not reflecting an industrial operation of LIBs recycling. Ideally, future studies should aim to simulate plants and processes as close to feasible industrial operations as possible for the studied process.

Efforts are critical by the scientific community and industry to move towards transparent and comprehensive studies based on primary data of LIBs recycling [12]. Ignoring some data from the upstream and downstream processes can cause errors that seriously harm some practices, such as long-term decisions for policy formulation [69]. A priority area of study is collecting and disseminating industrial data, in a readily reusable format, to support future analyses.

4.4. Data representativeness

In the current LCA literature on LIBs recycling, data representativeness is little discussed: only 18.1% (8) studies present explanations about representativeness for the three scopes (Technological, Temporal, Geographic) [18,56,60,92,124,136,175,174]. However, some of these studies present unclear explanations about some of the scopes, for example, the geographic scope [56] states that the data are representative of China, however, the data are not detailed in the inventory [18,60,174]; these, one only mention that the evaluation brings data from the "current process".

Fig. 10 synthesizes the data representativeness of the articles. Technological and geographic scopes are most commonly defined, whilst the temporal scope is rarely presented, with almost 80% of the

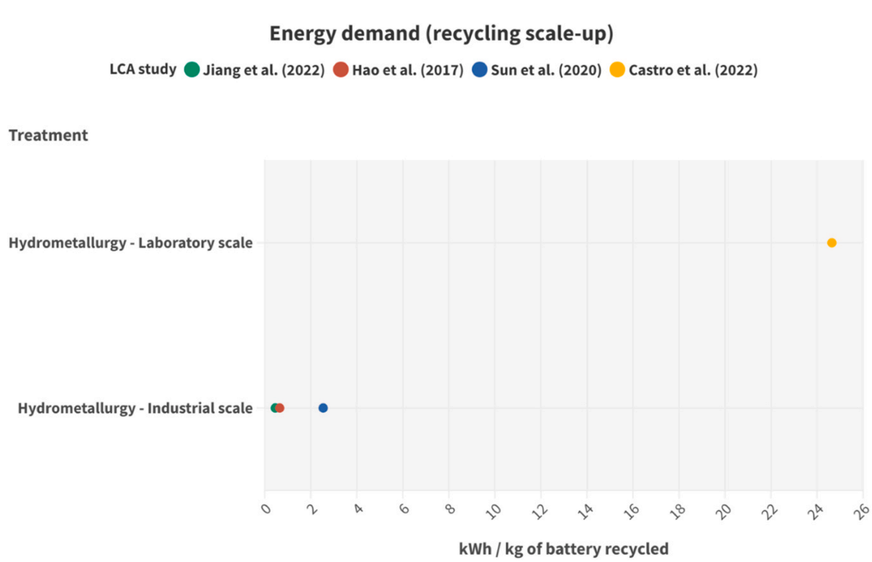

Fig. 9. Energy demand comparison between laboratory and industrial scales.

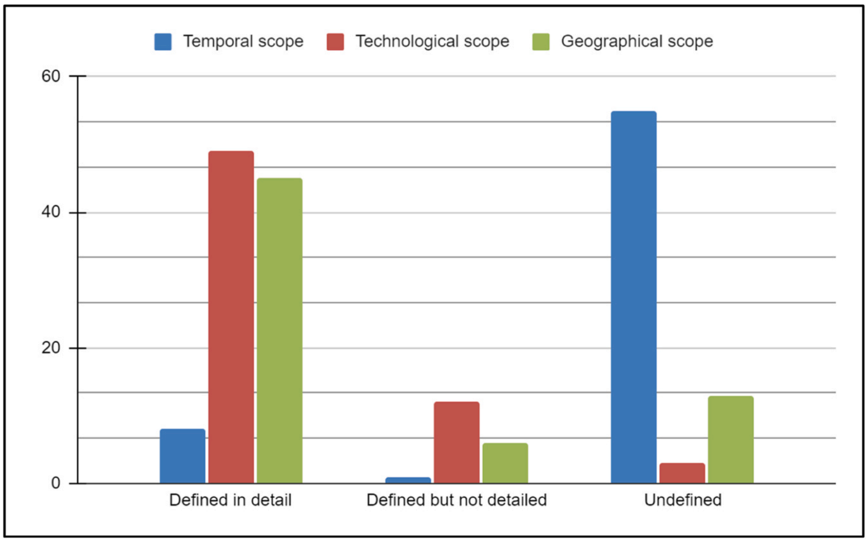

Fig. 10. Temporal, technological and geographic representativeness of the studies.

studies not discussing this scope. This result is in agreement with previous findings, in which considerations about temporal representativeness are still uncommon in LCA studies, raising concerns about the representativeness and relevance of currently available results [13]. By not clearly identifying in the studies the temporal representativeness of the data and the implications of the age of the data for the results, the level of uncertainty of results increases, since there is no way to attest that the data used in the evaluation represent the evaluated context [48, 63]. When there are considerations about temporality in studies, it occurs in vague statements such as "time of publication of the article" [132]. Of the studies that present the temporal scope, three do so only for the energy matrix data [60,136,174]. Other studies mention that the data refer to a date range (years) in which the data were collected [56, 92].

Geographic representativeness is a universal problem in LCA, particularly for studies from developing countries, as the lack of representative inventory data leads to studies using global average data or data from developed countries. However, for some datasets, this barrier is overcome, such as for electricity production, as energy matrix datasets from several countries are available in current databases. Another issue is the production of chemical inputs; usually, the production technology is homogeneous, which can help in choosing a product produced in another country, adjusting only the energy matrix.

The lack of discussion and transparency about data representativeness is a critical issue to be addressed in future studies on LIBs recycling. It is worth noting that most studies do not use updated and primary data, which may compromise representativeness in the three spheres [85]. It is recommended that studies utilize regionalized data to reflect the available recycling technologies in different locations, considering up-to-date information and the time horizon in which the data remains

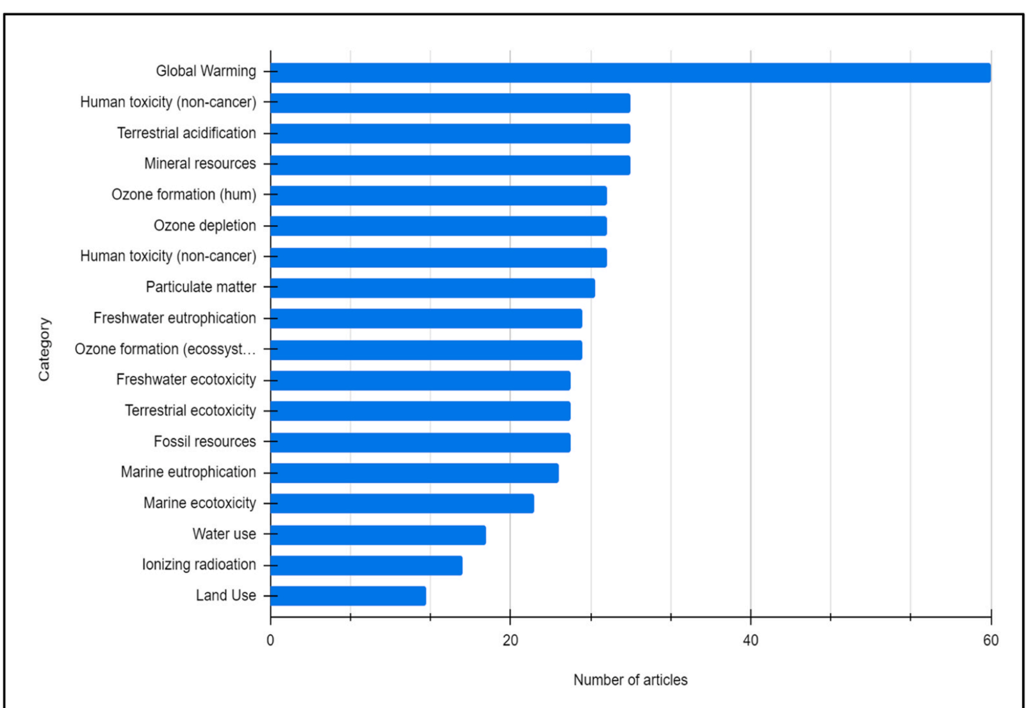

Fig. 11. Distribution of impact categories in reviewed studies.

valid. For example, Yu et al. [177] evaluated 30 different regions in China to assess how the carbon and water footprints can vary due to changes in LIB recycling locations within the same country.

4.5. Impact assessment

The assessed studies cover 18 different impact categories (Fig. 11). However, the coverage of impacts is very diverse across studies; some studies assess 18 categories (e.g. [37,128,156,185] and 17 categories [94], while others assess only one (e.g. [6]; [81] or two (e.g. [104]; [76], which makes direct comparisons between studies difficult. The most evaluated impact category related to Global Warming Potential (GWP), is present in 93% of the studies (60 articles). In second place, are the categories of Human toxicity (cancer), Terrestrial acidification and Depletion of Mineral Resources, present in 30 studies each. Less evaluated impact categories were Land use (13 articles), Ionizing radiation (16 articles), and Water use (18 articles).

4.5.1. Impacts of LIBs recycling by LIB type and different functional units

Figs. 12 to 17 present the comparison of impact results in kg of CO_2 eq for LIBs recycling considering the two main functional units found in the studies (1 kg of battery and 1 kWh of energy capacity) for pyrometallurgical, hydrometallurgical, and direct recycling technologies, and considering different cathode chemistries. The detailed data used to construct Figs. 12 to 17 can be found in Supplementary material S5.

The results demonstrate that the potential for Climate Change impact can vary widely depending on the cathode chemistries and the type of recycling applied. Additionally, the different methodological choices of the studies, such as different system boundaries, exclusion of unit processes, and flows of assessments, influence the generation of final impacts. For example, to pyrometallurgical recycling, the recycling of LFP and NMC chemistries showed higher impacts due to the addition of Ammonium hydroxide for LFP chemistry treatment [172] and due to the recycling taking place in China for NMC chemistry [43]; [177]. For the recycling of 1 kg of LFP battery, direct recycling based on defect-targeted healing that regenerate defects with acid citric without altering any other properties of LFP particles presents lower impacts compared to recycling with hydrometallurgy and pyrometallurgy [172].

Recycling in China seems to have higher impacts due to the Chinese energy matrix being more carbon-intensive than that of the USA [43]. The results indicate that recycling in China can have 10–20% more impacts than in the USA, regardless of cathode chemistry and recycling type. The Climate Change impact category of hydrometallurgical recycling in China can vary from 1.087 kg CO_2eq / kg of battery [167] to 2.7 kg of CO_2eq / kg of battery [73]. In the USA, hydrometallurgical recycling has a potential emission of 2.3 kg of CO_2 eq / kg of battery. Surprisingly, we found a study that uses a European energy matrix that shows higher environmental impacts, around 3.8 kg of CO_2eq to 4 kg of CO_2eq per kg of recycled battery with hydrometallurgical technology [132]. The result of higher impacts may be due to the fact that the process modeled in Rinne et al. [132] consumes more NaOH and includes an additional step of Rare Earths Elements recovery due to the joint treatment of LIBs and NiMH batteries.

Figs. 15, 16, and 17 depict the mapping of impact results in kg of CO_2 eq for recycling based on the functional unit of 1 kWh of energy capacity across pyrometallurgical, hydrometallurgical, and direct recycling technologies, considering different cathode chemistries. Overall, the results for recycling 1 kWh exhibit higher values compared to recycling per kg of battery, as the mass associated with 1 kWh of energy capacity is typically greater than 1 kg of battery due to the mass and energy density relationship of different cathode types. For instance, the mass of 1 kWh from an LFP battery can be approximately 8.64 kg, and 1 kWh from an NMC battery 4 kg [25].

4.5.2. Environmental impacts categories in different functional unit

Due to different methodological choices, such as the type of LIBs material composition, different chemical inputs, non-detailed inventories, choice of system boundaries, assumptions about local conditions, and different methods used, it becomes complex to compare the results of all impact categories provided by the studies. However, seeking to clarify the impacts of recycling we performed a comprehensive mapping and meta-analysis to demonstrate the environmental impacts considering different recycling technologies in different functional units. Figs. 18, 19 and 20 presents the absolute impact results mapped to each impact category. The results are organized with the average values of environmental impacts and the variations of minimum and maximum values observed in studies that use the same functional unit and technology. The minimum and maximum variations are highlighted by the error line. Detailed raw data for each impact category presented in Figs. 18, 19 and 20 are provided in Supplementary material S6 by recycling type and functional unit.

It is noteworthy that, although some studies present the results of the

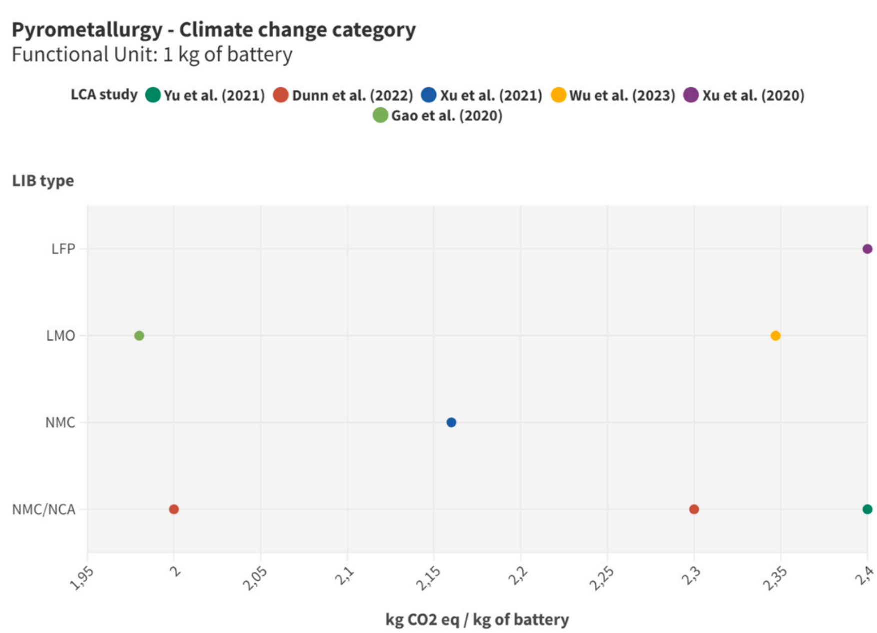

Fig. 12. kg CO_2 eq in pyrometallurgical recycling considering 1 kg of battery in different chemical cathode composition.

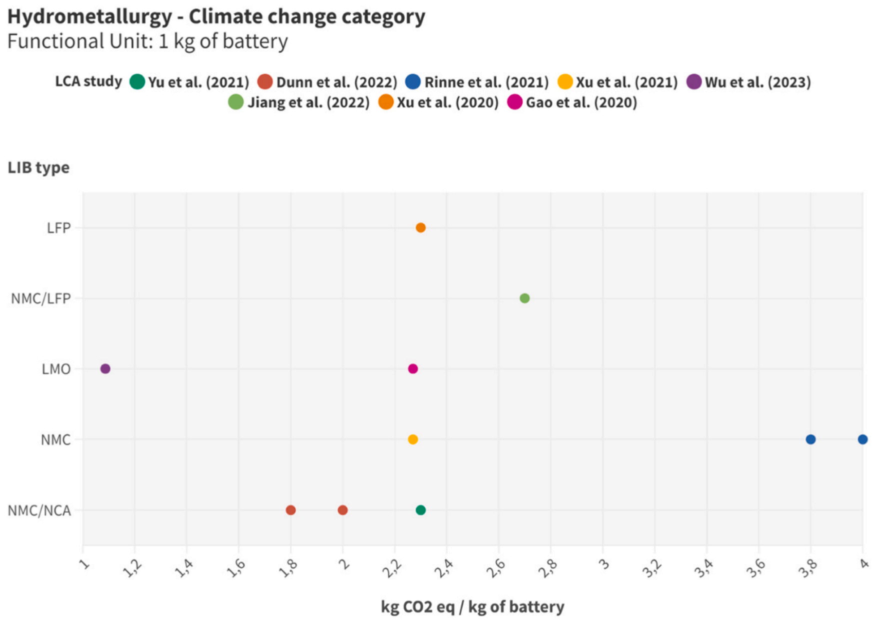

Fig. 13. kg CO_2 eq in hydrometallurgical recycling considering 1 kg of battery in different chemical cathode composition.

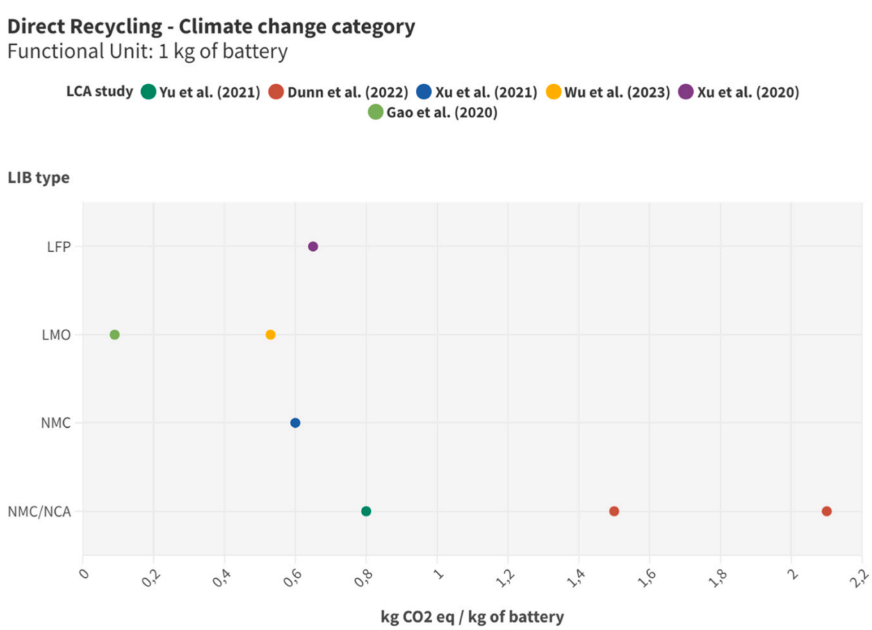

Fig. 14. kg CO_2 eq in direct recycling considering 1 kg of battery in different chemical cathode composition.

assessment of impacts in certain categories, there are no detailed data on the elementary flows and processes that cause the main contributions in all impact categories. The discussion is focused on the relative contribution of the processes, but without detailing the impact results. To increase transparency in reporting results, it is suggested that future studies deepen the discussion of results to the most detailed level of contribution, focusing on impact categories and not just on the overall performance of processes. In addition, some studies (e.g., [93,124,147] present only the net values of the impacts, obscuring the real value of the impacts of the processes and the credits provided by the recycling.

The results of the meta-analysis show that the impact results present wide variability between the different types of functional unit, and, even considering the same functional unit, and technology, there is a great variation in the results of environmental impacts in all impact categories of different studies. For example, in the Climate change category, for the functional unit of 1 kg of treated cathode using hydrometallurgical technology, results can vary widely from 4.21 kg CO_2eq to 299 kg CO_2eq per kg of treated cathode [76], due to differences in chemical inputs and types of unit processes used in leaching and recovery of materials. In addition, beyond the difference in inputs, another factor that determines this wide variation is the different limits of the system and the energy matrix of different countries. For example, Dunn et al. [43] point out that recycling LIBs in the USA causes lower impacts than recycling in China due to a shorter transport distance and a less fossil fuel intensive electrical grid. In addition, the exclusion of pretreatment, transport or final waste disposal stages may result in smaller impacts for the same functional unit.

Direct recycling and bioleaching technologies do not have impact assessments available for all impact categories and for all mapped functional units. For direct recycling we found impact values only for

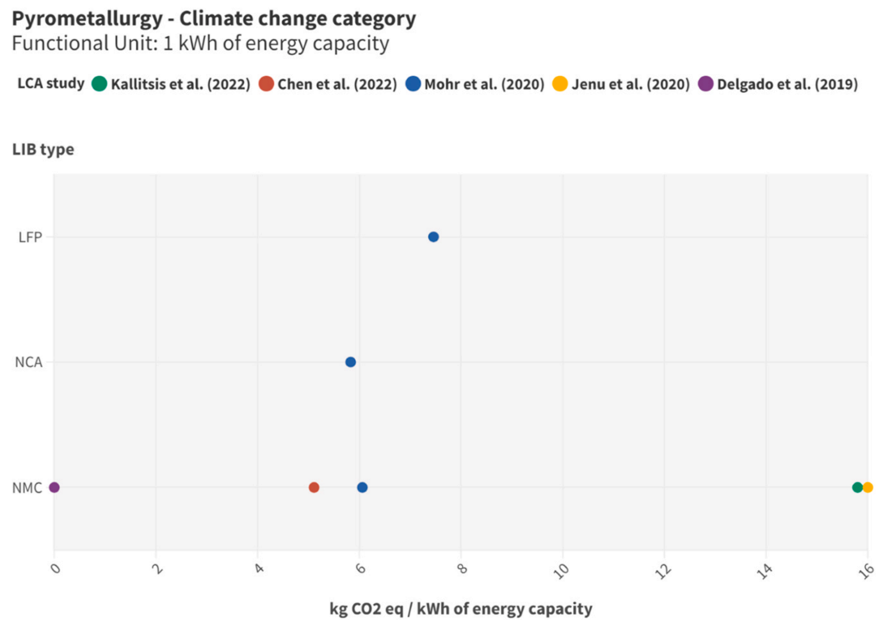

Fig. 15. kg CO_2 eq in pyrometallurgical recycling considering 1 kWh of energy capacity of different chemical cathode composition.

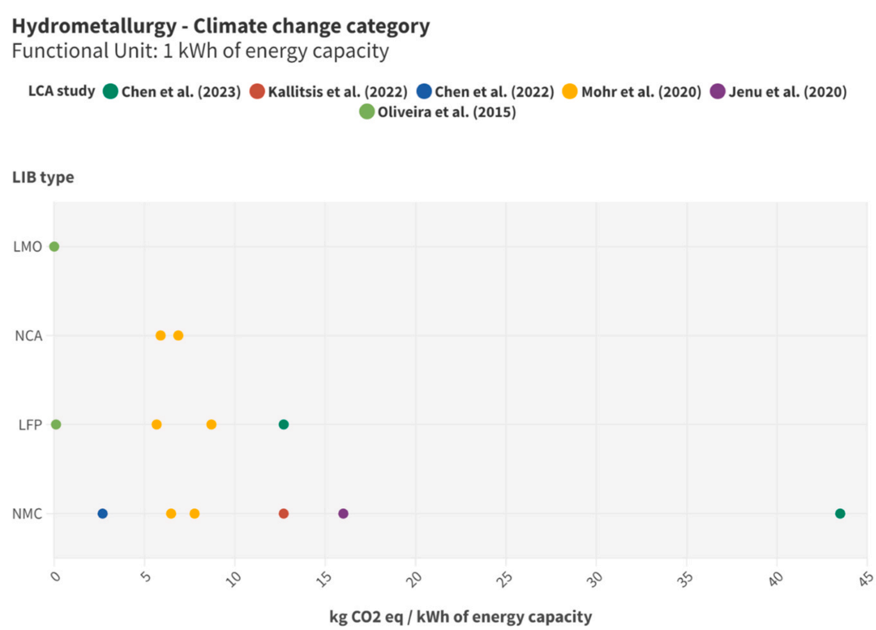

Fig. 16. kg CO_2 eq in hydrometallurgical recycling considering 1 kWh of energy capacity of different chemical cathode composition.

two categories, Climate change (Fig. 20-m) and Water use (Fig. 19-g) and for only two functional units, 1 kg of battery and 1kWh of nominal capacity. For the bioleaching technology there was only two studies available, which included 15 environmental impact categories evaluating the functional unit of 1 kg of recovered cobalt [3] and 18 impact categories evaluating 1 kg of treated cathode [72]. These results indicate that assessments of LIBs recycling technologies still need further development, especially considering the comparison of different technologies and with a comprehensive set of categories of environmental impacts.

With the results of impacts available in the literature, a comparison of these two technologies (direct recycling and bioleaching), considered more environmentally friendly than hydrometallurgy and pyrometallurgy, is limited. With this, there is a risk of displacing impacts to other environmental categories, since there are no complete assessments available to guide decision-makers as to the most environmentally clean option. For example, when compared to recycling 1 kg of cobalt recovered by bioleaching and hydrometallurgy, although bioleaching has lower impacts than hydrometallurgy in 7 categories (Climate change, Particulate matter, Water use, Human toxicity cancer and non-cancer, Ozone depletion), in the other 8 assessed categories (Freshwater ecotoxicity, Marine ecotoxicity, Terrestrial ecotoxicity, Freshwater eutrophication, Marine eutrophication, Fossil fuel depletion, Ozone formation ecosystem and human) it has potentially greater impacts. Thus, there is a knowledge gap regarding the impacts of direct recycling and bioleaching in all impact categories and considering different functional units.

For the Climate change category (Fig. 20-m), considering 1 kg of battery as a functional unit, direct recycling has the lowest impact when compared to hydrometallurgical or pyrometallurgical options, with a

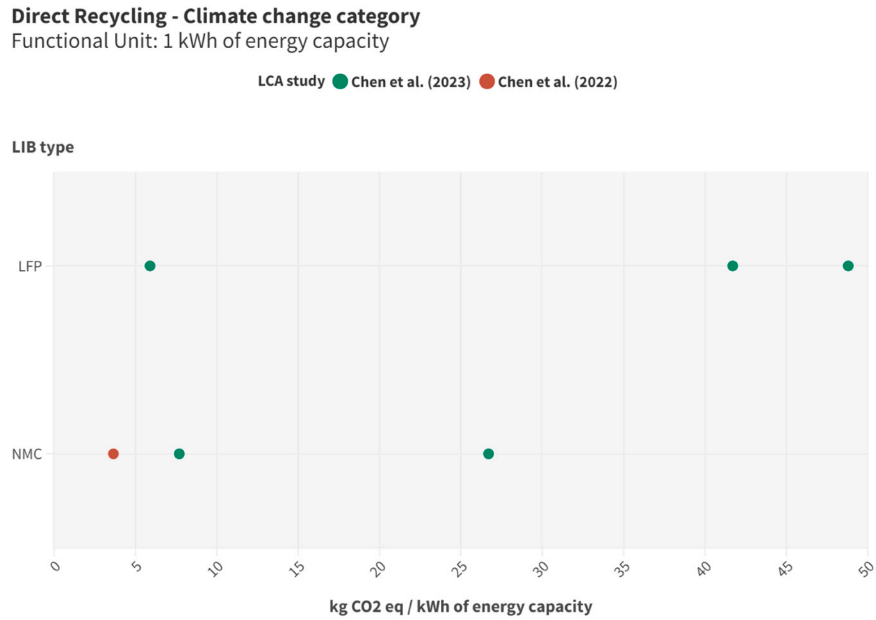

Fig. 17. kg CO_2 eq in direct recycling considering 1 kWh of energy capacity of different chemical cathode composition.

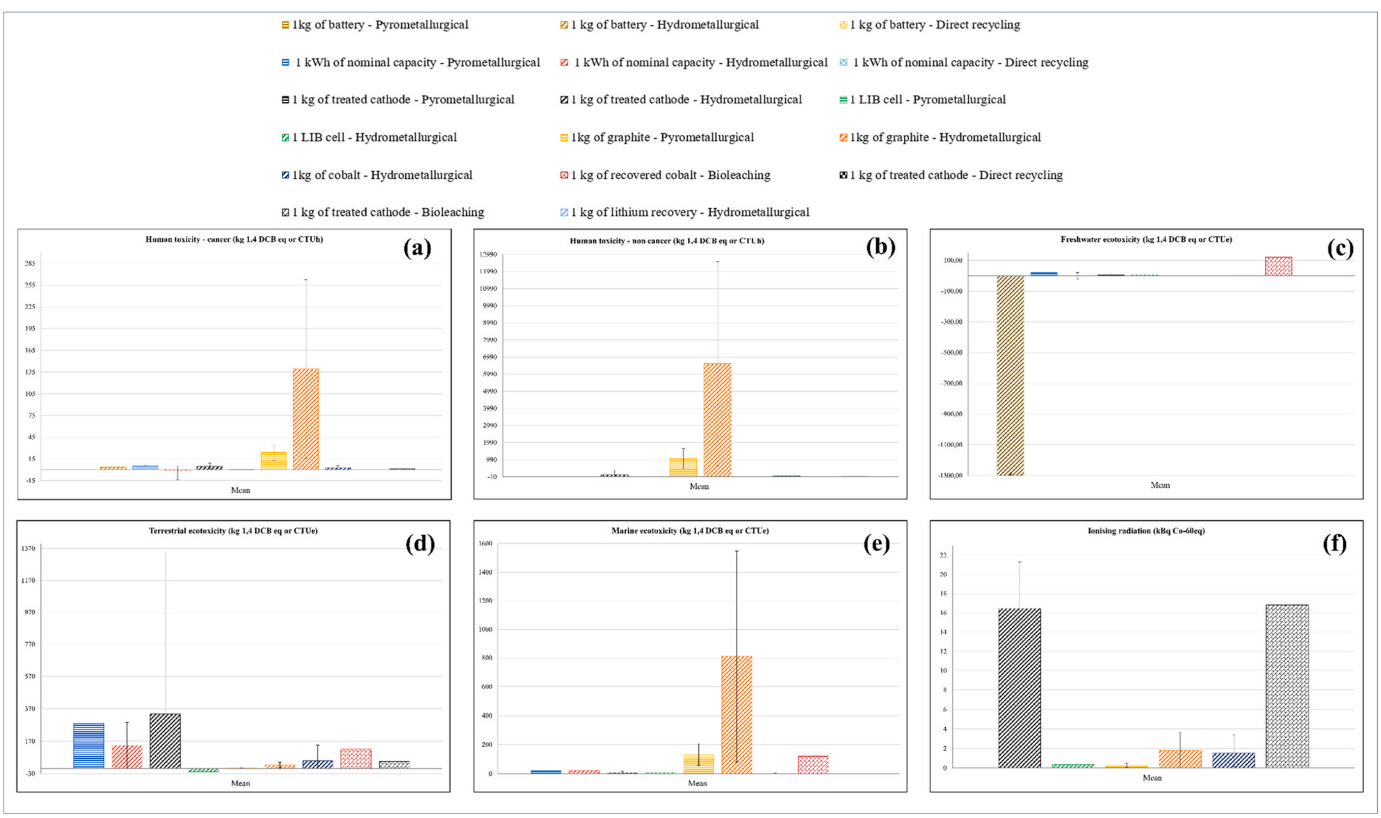

Fig. 18. Environmental impact by category, recycling technology and functional unit.

variation of 0.09 kg CO_2eq [56] at 4 kg CO2eq [132]. When compared to the functional unit of 1kWh of nominal capacity, direct recycling technology has an average of 26% and 28% more impact than hydrometallurgy and pyrometallurgy. This can be explained because the direct recycling process can consume additional materials, energy, and heat to recover more materials [25]. In addition, some results found suggest that depending on the type of functional unit, pyrometallurgy may have lower impacts than hydrometallurgy, contrasting with the current consensus in the literature that pyrometallurgy is always more impactful than hydrometallurgy [61]. When we compare the kg CO_2eq emissions per functional unit that are equal but differ in the recycling technology of pyrometallurgy and hydrometallurgy, we find that pyrometallurgy can have lower average impacts in the recycling of 1 LIB cell, 1 kg of treated cathode and the recovery of 1 kg of regenerated graphite.

The use of electricity is the main contributor of impacts for the Climate change category in the four recycling options. It is noted that the

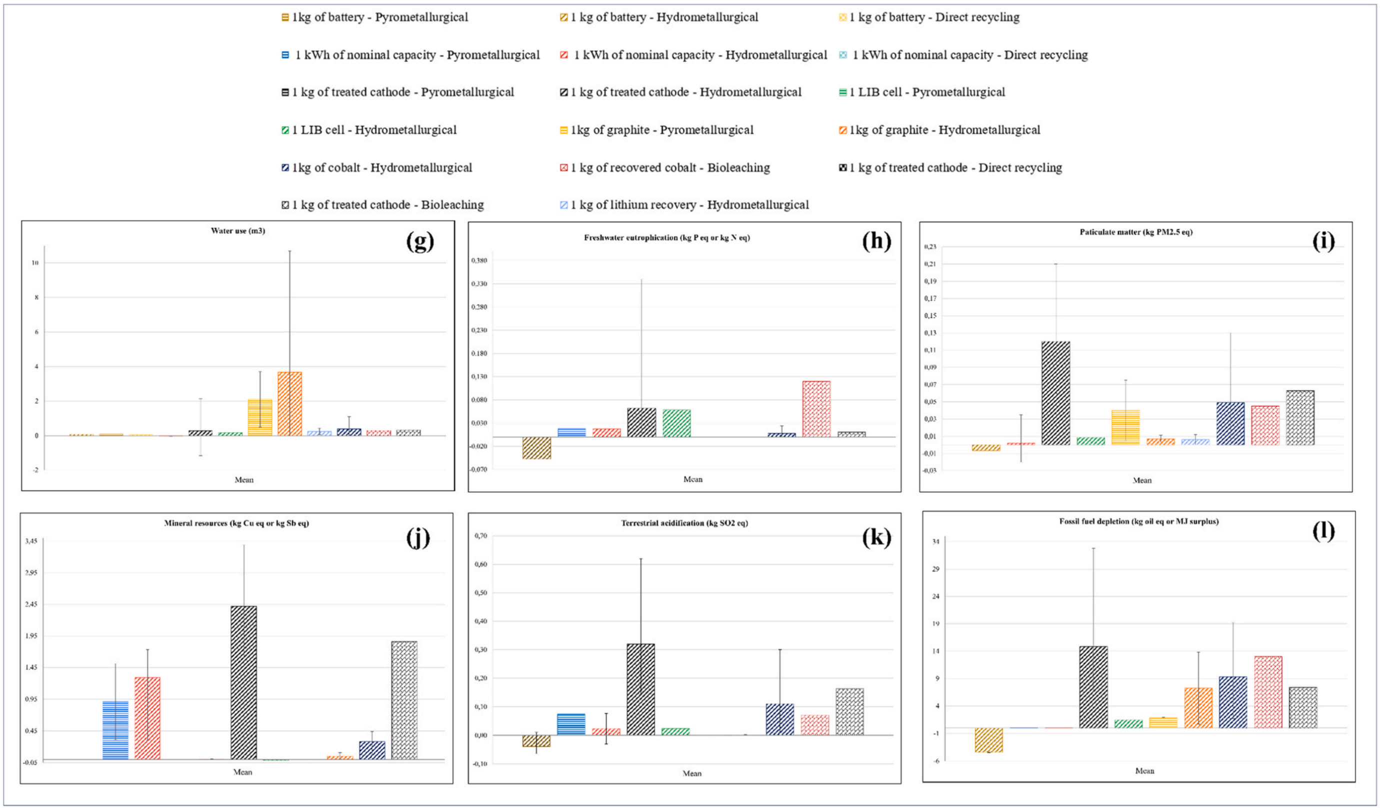

Fig. 19. Environmental impact by category, recycling technology and functional unit (continuation).

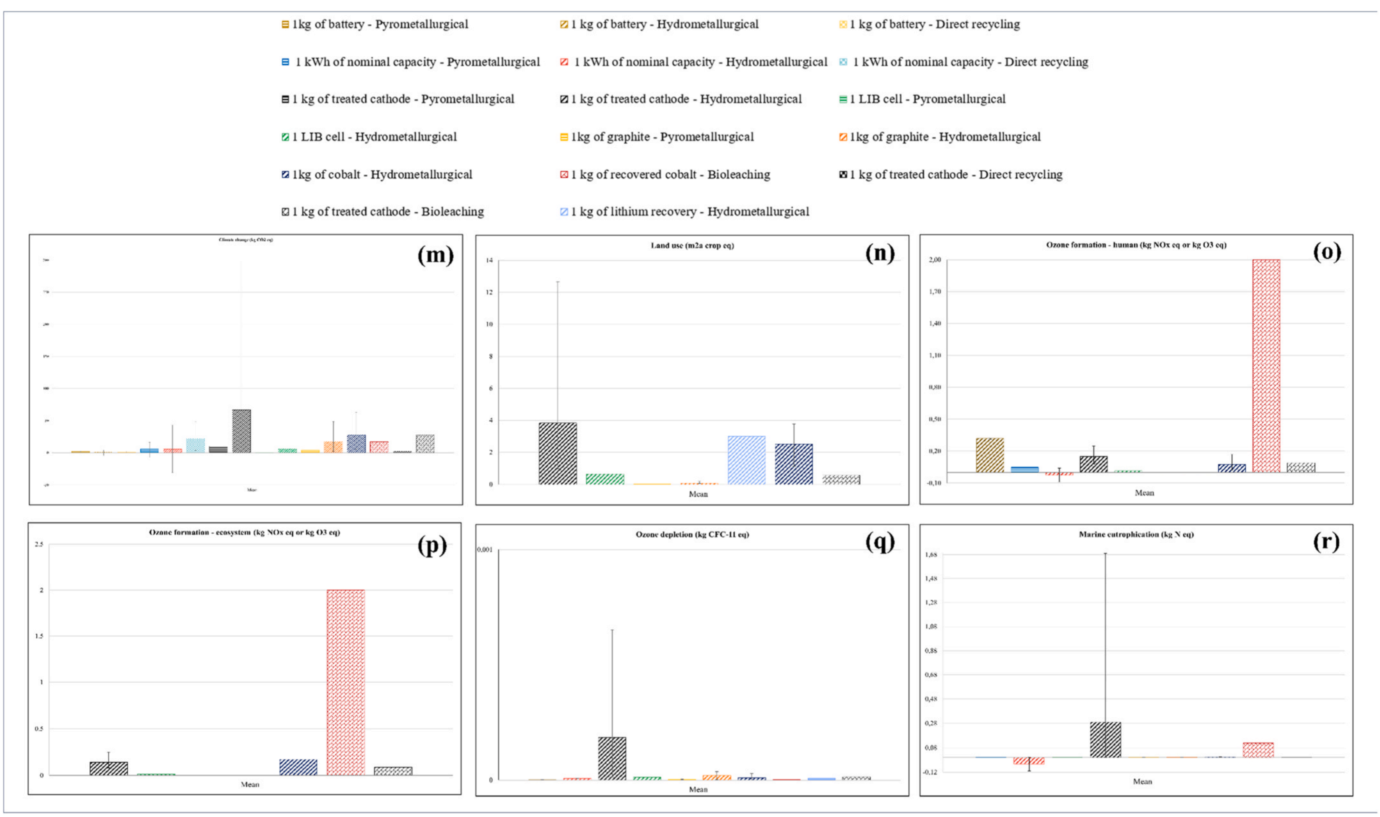

Fig. 20. Environmental impact by category, recycling technology and functional unit (continuation).

greatest impacts are observed in the treatment of the cathode by hydrometallurgical technology, as this is the fraction rich in metals and requires greater inputs of inputs and energy for the recycling of different types of metals. The use of electricity and the production of chemical inputs are also identified as the main contributors to all 17 other categories of impacts.

For the particulate material category, the highest impact values are also associated with the treatment of 1 kg of cathode and 1 kg of batteries by hydrometallurgy and 1 kg of graphite by pyrometallurgy. This result can be explained because the emission of fine particles are mainly associated with electricity production [72] and pyrometallurgical or hydrometallurgical processes are energy intensive. However, detailed explanations on impacts that are direct from the pretreatment and recovery processes of materials or associated with the background system were not found. This differentiation is important, as fine particles can be released from LIBs into the air during disassembly and pretreatment recycling processes [106,40], causing significant environmental impacts.

For the ionizing radiation category, few results were available. The highest impact values are also associated with the 1 kg cathode treatment, with an average of 16.5 kBqCo-60 eq and a maximum of 21.3 kBqCo-60 eq. The impacts of ionizing radiation are associated with the use of electricity in recycling operations and production of chemical inputs [23].

For categories related to ecotoxicity (freshwater, marine and terrestrial ecotoxicity), the main contributors to impacts are the production of chemical inputs [128], the incineration of plastic waste and the treatment of copper sheets [23].

For the marine and freshwater eutrophication categories, the main impacts are related to the production of chemical inputs and direct emissions from the precipitation stages. The direct emission of phosphate into water is the main source of impact on freshwater eutrophication [23]. For terrestrial acidification, the production of chemical inputs and electricity are the main contributors of impacts [23].

Results reveal that coverage of impacts in LIBs recycling assessment is limited. It is essential to conduct more studies and include a greater number of impact categories. From the selected studies it is not possible to reach conclusions about the environmental superiority of a particular technology, which may result in serious secondary environmental problems in the future. Unless all significant environmental impact categories are included, there is no way to attest to the environmental superiority of an alternative [161].

4.5.3. EverBatt model results

The EverBatt model, a parameterized Excel-based model developed within the GREET framework, has been widely utilized in LCA studies on LIBs recycling. It provides detailed data on processes, inputs, and GHG emissions per kg of different types of recycled LIBs through pyrometallurgical, hydrometallurgical, and direct recycling processes in various country contexts. Studies based on the EverBatt model show a consistent trend in impact generation per kg of recycled battery, with some minor differences that could not be traced due to the lack of detail on the chosen parameters in the EverBatt model (e.g., whether transportation was included, which types of unit processes and inputs were selected).

For pyrometallurgical and hydrometallurgical recycling, results from studies using the EverBatt model indicate that recycling in China has a greater impact than recycling in the USA. Regarding direct recycling using the EverBatt model as a reference point, results show greater variability, with some processes using fewer chemicals and less energy yielding better outcomes. For example, direct recycling in China using the chemical lithiation strategy with pyrene [167] shows 299% fewer impacts than direct recycling using high-temperature solid-state sintering [43]. However, when the same direct recycling technologies from EverBatt are compared for recycling in China [43]; M. [177] or the USA, recycling in the USA [43,56,172,173] consistently shows lower impact.

4.6. Interpretation

This section provides the mapping of hotspots identified in studies and key parameters to sensitivity and uncertainty analysis in LIBs recycling.

4.6.1. Hotspots

The analysis of the results of LIBs recycling studies allowed for identifying, for each type of technology, the process hotspots (Table 5).

To Pyrometallurgy the main impacts derive from the high use of energy in the casting stage, as temperatures of up to 1400.00º are required (e.g., [34]. In addition, the pyrometallurgical process has great potential for atmospheric pollution, due to the toxic gases released when the organic components present in LIBs are burned [127]; [177]. Hydrometallurgy: the main contributors to impacts are the production of chemical inputs used for the leaching and recovery of metals [104,132]. Some studies have indicated that hydrometallurgy can also be an energy-intensive process, depending on the types of equipment used in unit operations [55,128]. To Direct recycling, the main hotspots are related to the consumption of primary inputs (lithium carbonate) for annealing the cathodes, and the use of electricity. To bioleaching, electricity, and sodium gluconate were identified as hotspots [3].

Improving the environmental profile for any functional unit and recycling technology relies on increasing energy and material efficiency, coupled with the use of renewable energy sources. Energy and material efficiency have the potential to reduce impacts in all impact categories [23,25]. The use of renewable energy can decrease the impacts of LIBs recycling by up to 75% in categories such as fine particulate matter formation, fossil resource depletion, freshwater eutrophication, ionizing radiation, ozone depletion, and terrestrial acidification [72]. Reducing the use of chemical reagents in the recycling process is a crucial measure to improve the green development of battery recycling [25].

4.6.2. Uncertainty and sensitivity analysis

The uncertainties presented in the reviewed studies can be summarized by the lack of data, lack of data quality assessment and black-box processes. The data in a black-box process do not provide detailed origins of flows, only the total values of inputs and outputs [48]. Almost no study discusses the uncertainty of the used data, even less quantifying the uncertainty magnitude. The implications of the lack of data uncertainty analysis lie mainly in the lack of representativeness, making the analysis of little value for decision-making [155]. For example, process parameters determined in batch laboratory experiments may not be directly applicable on an industrial scale, due to differences in equipment efficiency, such as stirrers [132]. Another relevant issue regarding uncertainty is related to the types of direct and indirect impacts assessed. When evaluating only impacts by the consumption of energy and chemicals, the analysis is limited to indirect impacts. Studies are still needed on the direct impacts of LIBs recycling, such as emissions from crushing processes, characterization of effluents and final solid waste. Particularly with regard to the loads associated with the final waste streams, there is much uncertainty [106].

In the LCAs on LIBs recycling, sensitivity analyses were conducted on several input parameters such as quantity of chemical inputs per functional unit, recycling rate, metal content in the samples, and mix of the energy matrix. An important parameter in sensitivity analysis for waste management with material recovery is the substitution rate [155]. Table 6 summarizes sensitive parameters for LIBs recycling and how to conduct their assessment.

Sensitivity analyses can be conducted using some methods, such as perturbation analysis on input data, either on one parameter at a time or global perturbations, quantitative and qualitative uncertainty analysis, break-even analysis, and scenario analysis. For more details on each method see [85]. It is recommended that future LCA studies in the recycling of LIBs perform uncertainty and sensitivity analysis steps together with completeness and consistency checks.

Table 5
Hotspots by recycling type.

Recycling type	Hotspots (process)	ID studies
Hydrometallurgical recycling	Electricity use (pretreatment, leaching, solvent extraction, precipitation, filtration)	1–2–3–7–8–10–11–12–15–16–19–22–23–24–35–47–50–54–64
	NaClO (Leaching)	1
	NaOH (Precipitation)	2–15–17–21–27–47
	Hydrochloric acid (Leaching)	2–17
	Sodium Gluconate (Bioleaching)	3
	Cyanex272 and D2EPHA (Solvent extraction)	5
	Hydrogen Peroxide (Leaching)	5
	Chemicals (Leaching, precipitation)	7
	Hydrometallurgical processing	8
	Chemical process (bioleaching)	9–12–35
	Citric acid (Leaching)	11
	Phosphoric acid (Leaching)	11
	Sulfuric acid (Leaching)	12–17–21–27
	Ammonia (Leaching)	16
	Acids and solvents (Leaching)	18
	Acid waste leaching)	22
	Inorganic acids (leaching)	22
	Crystallization process (solvent extraction)	24
	Waste treatment (copper treatment)	47
	Hotspots	Study
Pyrometallurgical	Electricity (smelting)	7–8–9–10–15–16–22–23–35–44–47–50–63–64
	Water consumption (smelting and lime production)	23
	Smelting (burn of batteries)	23–35
	Inputs production (furnace operations)	32
	Brine (discharging)	44
	NOx emissions (smelting)	58
	Hotspots	Study
Direct Recycling	Electricity (pretreatment, re-sintering)	9–10–16–19–23–28–31–36–50
	Organic solvent (DME) (chemical re-lithiation)	4
	The nickel and lithium-rich material (re-lithiation of ternary cathode materials)	9–31
	NaOH (water adjust pH)	28

4.7. Recommendations for future LCA studies in LIB recycling systems

Based on the detailed analysis of the LCA studies on the recycling of LIBs and the methodological choices that most affect their results, as well as gaps and key improvement issues identified throughout the review, some recommendations and guidelines for future studies are:

1. There is a knowledge gap regarding the environmental profile of LIBs recycling in different country contexts beyond of China and USA. Further LCA studies in other contexts are needed because with the global dissemination of electric vehicles and the increased consumption of portable electronics, the development of recycling capacities in different countries will be necessary.
2. To increase the quality of studies and guide future decisions, it is recommended to detail important methodological choices, such as the inventory modeling framework, choices to solve the multifunctionality problem and cut-off rules for excluding processes from the system boundary.
3. Detailed inventories per unit processes must be made available, avoiding the black-box presentation of processes. This is important, as process improvements can be made more assertively.
4. A major gap in the data concerns collection, transport, and pretreatment processes. Future studies should direct efforts to these stages. When it is not possible to collect these data, a clear justification for the cut and eventual implications of the lack of these data for the results must be provided.
5. Obtaining primary data from different industrial facilities in worldwide and their operating conditions to achieve technological, geographical, and temporal representation is fundamental.
6. Is necessary to collect primary data and the analysis of environmental implications of final waste disposal of recycling processes (solids and effluents).
7. There is a need to apply the LCA with a consequential approach, as this elucidates which marginal processes are affected and the possible environmental consequences of decisions like the implementation and increase of different LIBs recycling processes.
8. Evaluated the impacts with different impact assessment methods, seeking to capture the sensitivity of the results through their use.
9. Evaluating the consequences and sensitivity of selecting the impact assessment method for different locations/regions.
10. Current studies focusing on the climate change impact category, representing simplified LCA, whose results do not provide a holistic view of the environmental impacts of recycling LIBs options, which can omit serious secondary pollution risks. Limited coverage of impacts cannot support conclusions about the environmental superiority of one technology. Thus, is recommended that studies conduct comprehensive assessments with good coverage of impacts for all four technologies.
11. As LIBs waste management may involve multiple technological, environmental, social, and economic criteria, the integration of multi-criteria decision-making (MCDA) with LCA is recommended. MCDA helps to integrate sustainability requirements to identify technology options for waste treatment, ideal locations for treatment facilities, and the identification of better waste management scenarios [151]. In the case of LIBs waste, future studies should be developed with the integration of MCDA and LCA to improve decision-making on the choice of recycling technologies and recycling facility locations that improve sustainability standards. Future studies can utilize the framework and recommendations developed by Torkayesh et al. [151] to integrate MCDA and LCA in studies of LIBs recycling.
12. Finally, the use of Material Flow Analysis (MFA) should be further explored in conjunction with LCA of LIBs recycling. Synergies between these two methods produce more complete assessments, providing tracking of waste generation sources and flows, material recovery potentials from these flows, as well as potential emissions and benefits of recycling [10]. MFA can

Table 6
Parameters for sensitivity analysis.

Parameters for sensitivity analysis in LIBs recycling		
Sensitive parameters	How to evaluate	Revised study
Energy consumption	Variations in the main influencing factors in the consumption of electric energy in hydrometallurgy: pulp density, temperature and leaching time, acid concentration.	3–26–30–54–59
Material recovery/recycling rate	Variation in the recycling/recovery rate of materials using ranges of values (e.g., 10%; 30%; 50%; 80%; 100%)	5–11–19–30–32–35–47–50
Virgin material substitution rate	Substitution rate of virgin material in proportions other than 1:1 (e.g., 1:0,9; 1:0,8; 1:0,5)	7
Geographic location of recycling	Recycling operations in different countries (e.g., recycling in China, Europe or North America)	8
Quality of recovered materials	Comparison of environmental credits provided when materials are reclaimed battery grade or ore grade	8–35
Country electricity grid mix	Changes in the energy grid mix towards a mix of renewable sources (e.g., Substitution of energy generated by coal and oil with solar and hydropower energy).	9–12–19–23–27–46–62
Inputs variation	Variation on Material Inputs: (e.g., Sulfuric Acid, Sodium Hydroxide, Hydrochloric Acid, and Water): A parameter rate of change (%) can be adjusted one at a time, keeping the other constants, or a global variation can be applied on all parameters simultaneously. Optimization to less use of acids for leaching.	11–17–18–22
Transport distance	Variation in transport distances within a range of +/-50%	19
Input material weight	Variation in weight input material	26
Loss of materials in the pretreatment stages	Loss of material in process	26
Process operation time	Variation in unit process operation time	26
Inclusion of the pretreatment step	Inclusion of pretreatment steps and additional recovered materials such as Cu and Al	27
Amount of metals in LIBs	Decreased cobalt and increased nickel in cells	30–35
	Substitution of cathode chemistries from NCM 622 to NCM 811	37
Database used for background data	Different databases are used and compared for background data collection	46
Market value of recycled materials	Compare the market value of recycled materials considering differences for ore grade and battery grade or discounts due to loss of quality	10

quantify LIBs waste stocks and flows and the potential to recover raw materials for recycling [187]. Especially for LIBs end-of-life, the combination of MFA and LCA is promising to elucidate the amount of waste generated spatially and temporally and for carrying out impact assessments considering different recycling locations and different recycling technologies to improve material efficiency strategies and policies. For example, the use of MFA and LCA can help verify whether the requirements for recycled content and recycling efficiency of LIBs can be achieved and to what extent these initiatives will result in a reduction in the Carbon Footprint due to the use of specific recycling processes.

5. Conclusion

This paper reviewed 64 LCA studies on LIBs recycling, providing a comprehensive understanding of the environmental impacts of LIB recycling in available studies, covering 18 impact categories in different functional units, and identifying research gaps.

The available studies exhibit data gaps and lack consistency with LCA standards and guidelines. The focus is on LIBs from electric vehicles with the NMC chemical composition. Meta-analysis of environmental impacts reveals significant variability in impact results among different functional units, cathode chemistries, and within the same unit and technology. Treatment of the cathode with a hydrometallurgical process shows the highest impacts due to the metal-rich composition requiring substantial inputs and energy for recycling. Electricity production and chemical inputs are major contributors for all technologies and impact categories. Notably, lacks a comprehensive impact assessment for direct recycling and bioleaching in the literature.

While LIBs offer advantages for decarbonizing transportation and renewable energy storage, the environmental benefits of different recycling options remain uncertain. The lack of comprehensive environmental impact assessments and data on reverse chain processes hinders the affirmation of any recycling technology's superiority. Assessing the environmental consequences of LIB recycling involves complex factors such as energy sources, recovery inputs, material recovery rates, waste stream destinations, substitution materials, the scale of operation, location, and transport distances.

The incompleteness, imprecision, and limited representativeness of data and impact categories in the studies raise concerns as they hinder informed decisions regarding environmentally appropriate recycling alternatives for LIBs. The availability of actual operational data and comprehensive impact coverage requires further development. Many studies neglect the collection, transport, pretreatment, and final disposal of waste streams after recycling. The lack of understanding of the impacts of these processes, especially landfill disposal and treatment of effluents from hydrometallurgical processes, results in incomplete assessments. Studies with a limited scope can only inform decisions with limited scope. In the case of LIBs recycling, this is a matter of concern since LIBs waste streams will increase in the future and secondary pollution flows may be neglected.

Considering the environmental implications evaluated by LCA studies, the results of this study can guide the development and implementation of more sustainable recycling technologies for managing and recovering valuable materials from LIBs. For LCA researchers and practitioners, the mapping the methodological characteristics of studies and synthesizing a wide range of environmental impacts can assist in the development of more robust studies and serve as an easily recoverable database for consultation and comparison of results from future studies on LCA in LIBs recycling. Furthermore, systematic mapping by type of recycling technology and impacts by the functional unit can assist other users of LCA data, such as decision-makers in industry and policymakers, by providing access to different environmental impact data on recycling LIBs that can help in decision-making about incentives and investments in sustainable recycling processes.

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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.

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Appendix A. Supporting information

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