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Life cycle assessment of recycling options for automotive Li-ion battery packs

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ABSTRACT

Ramping up automotive lithium-ion battery (LIB) production volumes creates an imperative need for the establishment of end-of-life treatment chains for spent automotive traction battery packs. Life Cycle Assessment (LCA) is an essential tool in evaluating the environmental performance of such chains and options. This work synthesises publicly-available data to expand upon previously reported LCA studies for LIB recycling and holistically model end-of-life treatment chains for spent automotive traction battery packs with lithium nickel cobalt manganese oxide positive electrodes. The study provides an in-depth analysis of unit process contributions to the environmental benefits and burdens of battery recycling options and integrates these with the battery production impacts to estimate the net environmental benefit achieved by the introduction of recycling in the value chain. The attributional LCA model accounts for the whole recycling chain, from the point of end-of-life LIB collection to the provision of secondary materials for battery manufacturing. Pyrometallurgical processing of spent automotive traction battery cells is predicted to have a larger Global Warming Potential (GWP), due to its higher energy intensity, while hydrometallurgical processing is shown to be more environmentally beneficial, due to the additional recovery of lithium as hydroxide. The majority of the environmental benefits arise from the recovery of aluminium and copper fractions of battery packs, with important contributions also arising from the recovery of nickel and cobalt from the battery cells. Overall, the LCA model presented estimates a net benefit in 11 out of 13 environmental impact categories based on the ReCiPe characterisation method, as compared to battery production without recycling. An investigation of the effect of geographic specificity on the combined production and recycling indicates that it is as a key source of GWP impact variability and that the more climate burdening chains offer a significantly higher potential for GWP reductions through battery recycling. The sensitivity analysis carried out shows that impacts related to air quality are higher when recovering lower grade materials. This study provides a quantitative and replicable inventory model which highlights the significance of the environmental benefits achieved through the establishment of circular automotive battery value chains.

1. Introduction

Rapidly falling costs, a series of technological developments and the urgent need for decarbonisation have positioned electric vehicles (EVs) as the dominant choice for establishing widely accessible sustainable transport systems (Günther et al., 2015; Knobloch et al., 2020). Transforming the transport system requires an unprecedented increase on battery production capacity, with demand for EV batteries projected to increase twenty-fold by 2030 (International Energy Agency, 2021). This growth trajectory of the battery market creates an imperative need for effective end-of-life (EoL) management of automotive lithium-ion batteries (LIBs) within the next decade. Battery recycling can divert batteries away from minor EoL treatment options, towards industrial-scale hydrometallurgical and pyrometallurgical processes, which are already being deployed around the world (Chen et al., 2019; Harper et al., 2019). Additionally, battery recycling offers a stream of raw materials, which is crucial in regions with limited availability of primary resources, such as Europe, and provides efficient means of handling production scrap from battery Gigafactories (Peel and Sanderson, 2020).

The environmental impact reduction potential of recycling technologies is known to depend both on the specific processes used and the particulars of the value chain where recycling is implemented (Arshad

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Table of abbreviations

BG	Battery-grade
BMS	Battery Management System
EC	Ethylene Carbonate
EoL	End-of-life
EV	Electric Vehicle
FDP	Fossil Depletion Potential
FEP	Freshwater Eutrophication Potential
FETP	Freshwater Ecotoxicity Potential
FPMFP	Fine Particulate Matter Formation Potential
GHG	Greenhouse-gas
Gr	Graphite
GWP	Global Warming Potential
HTP	Human Toxicity Potential, cancer
ISO	International Organization for Standardization
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LFP	Lithium Iron Phosphate ($LiFePO_4$)
LMO	Lithium Manganese Oxide ($LiMn_2O_4$)
LIB	Lithium-ion Battery
MDP	Metal Depletion Potential
MEP	Marine Eutrophication Potential
METP	Marine Ecotoxicity Potential
NMC	Lithium nickel manganese cobalt oxide ($LiNi_{1-x-y}Mn_xCo_yO_2$)
OG	Ore-grade
POFP	Photochemical Oxidant Formation Potential, human health
SM	Supplementary Material
SODP	Stratospheric Ozone Depletion Potential
TAP	Terrestrial Acidification Potential
TETP	Terrestrial Ecotoxicity Potential
TMS	Thermal Management System

et al., 2022; Rajaeifar et al., 2022). This has triggered a growing interest in the scientific community of using Life Cycle Assessment (LCA) to investigate the environmental benefits that battery recycling can achieve in different settings. Pyrometallurgical and hydrometallurgical treatments of lithium nickel manganese cobalt oxide (NMC) and lithium iron phosphate (LFP) batteries are the most frequently assessed process options amongst LCA studies (Mohr et al., 2020). Such commercial processes have been discussed by recent reviews from Sommerville et al. (2021) and Neumann et al. (2022). As NMC is by far the most common positive electrode (hereby referred to as cathode) chemistry for automotive applications (Kallitsis et al., 2022; World Economic Forum, 2019), its materials recovery processes have attracted increased attention.

Reported Global Warming Potentials (GWPs) of LCA studies focusing on NMC battery recycling, alongside the respective battery production GWP, are shown in Table 1. Cusenza et al. (2019) performed a cradle-to-grave assessment of a LIB pack for hybrid electric vehicles utilising a lithium manganese oxide (LMO)-NMC333 composite cathode material, demonstrating that utilising a pyrometallurgical-hydrometallurgical EoL treatment scheme reduces the production GWP by 5%. Ciez and Whitacre (2019) quantified, the net benefit of pyrometallurgical and hydrometallurgical recycling of NMC battery cells, amongst other battery chemistries, concluding that they do not reduce life cycle greenhouse-gas (GHG) emissions significantly. Dunn et al. (2015) calculated that producing a NMC battery with recycled material coming from pyrometallurgical processing, decreases its production GHG emissions by 20%, compared to production with primary material. Hao et al. (2017) modelled an EV recycling chain in China and estimated that the most important source of environmental benefit from recycling an NMC battery comes from the recovery of cathode materials. Sun et al. (2020) performed a cradle-to-grave assessment of NMC333 battery packs in China, calculating that the battery production footprint of 124.5 kg CO_2-eq $(kW\,h)^{-1}$ can be reduced by 30.9 kg CO_2-eq $(kW\,h)^{-1}$ through the recovery of battery pack materials. More recently, Accardo et al. (2021) studied the production, use and recycling of an NMC333 battery pack, reporting that the introduction of EoL treatment results on a net 4.5% decrease in the LIB production GWP. A recent cradle-to-grave LCA for NMC333 battery packs by Quan et al. (2022) showed that recycling resulted in a small reduction of the production GWP (likely less than 5% judging qualitatively based on graphical illustrations in this work). Another recent study by Du et al. (2022) provides valuable insights on the regeneration of NMC cathode material with a focus on the active material level, which explains why it is not included in Table 1. Jiang et al. (2022) predicted a −16.6 kg CO_2-eq $(kW\,h)^{-1}$ net benefit from hydrometallurgical recycling of NMC333 battery cells. Rajaeifar et al. (2021) performed an LCA of pyrometallurgical cell recycling using a mass functional unit, which is excluded from Table 1 as this and the functional unit of other studies are not directly comparable. An earlier comprehensive review by Mohr et al. (2020) highlighted the varying system boundaries and functional units reported by different authors as a reason for the reported variability and the need for transparency amongst LCAs on LIB recycling. This study confirms this finding (Table 1). Only Ciez and Whitacre (2019) disclosed the unit process and background data used in their recycling inventory. While Quan et al. (2022) reported the foreground inventories used in their study in sufficient detail, the background data used in their work are hard to trace.

As evident from the information presented in Table 1, the reported achievable GWP reduction through battery recycling is highly variable

Table 1
Reported GWP of NMC battery production and recycling across published studies.

Reference	Region	Cathode chemistry	Energy density [$kW\,h\,kg^{-1}$]	Focus	GWP [kg CO_2-eq $(kW\,h)^{-1}$]		
					Production	Recycling (net)[1]	
						Hydromet.	Pyromet.
Cusenza et al. (2019)	Europe	LMO-NMC333	0.065	Pack level	312.4	−15.4 (combination)	
Ciez and Whitacre (2019)	USA	NMC622	0.27[2]	Cell level	36.3	−1.3	6.4
			0.21[3]		42.4	−4.4	2.7
Sun et al. (Sun et al., 2020)	China	NMC622	0.115	Pack level	124.5	−30.9	n/a
Mohr et al. (2020)	n/a	NMC333	0.174	Cell level	75.5	−16.4	−13.8
Accardo et al. (2021)	Europe	NMC333	0.111	Pack level	136[4]	−6.2 (combination)	
Quan et al. (2022)	China	NMC333	0.142	Pack level	87.1	n/a	n/a
Jiang et al. (2022)	China	NMC333	0.150	Cell level	n/a	−16.6	n/a

[1]Additional burden introduced for battery recycling inputs minus benefits through the displacement of primary material, [2]pouch and [3]cylindrical cells, [4]battery production assumed to take place in China.

in published studies, which presents a difficulty for industry and decision makers; this can also be extended to other impact categories. Part of this variability can be attributed to differences in the system boundaries considered. Specifically, literature reports focusing on the recovery of the battery electrode materials (Ciez and Whitacre, 2019; Mohr et al., 2020), which constitute less than half of a battery pack's mass, neglect significant fractions of aluminium, copper and steel that could be recovered from the battery pack's packaging, thermal management system (TMS) and battery management system (BMS). Therefore, an open question arises regarding the environmental benefit of a circular battery value chain when also accounting for the recovery of materials at pack level and for a series of processes involved including collection, transportation, sorting and dismantling, which are usually neglected. Additionally, part of the variability in the results shown in Table 1 can be attributed to the different geographic locations of focus, given that different local electrical energy and material footprints influence the environmental burden of recycling chains, but also its benefit, as displaced material footprints differ significantly across locations (Kelly et al., 2019), with their relative differences remaining to be benchmarked under the same model and assumptions. In addition, evaluating other environmental impacts and benefits associated with LIB recycling, in addition to GHG emissions, is crucial (Ciez and Whitacre, 2019; Dunn et al., 2015) as water pollution from leachate, solid waste such as metal-rich ash, and air pollution impacts (Winslow et al., 2018) need to be considered when comparing potential management strategies for EoL LIBs. The majority of non-energy-related environmental impacts in the life cycle of LIBs are linked with mining and materials processing operations (Porzio and Scown, 2021). The mining and materials processing impacts can be avoided to a certain extent when recycling is implemented, as suggested in recent studies aiming at extending the analysis beyond GHGs for a series of recycling processes (Jiang et al., 2022; Quan et al., 2022).

The above confirm the need for a LCA study that combines transparency, reproducibility of inventory, offers sufficient resolution to better understand the burdens and benefits of battery recycling options and evaluates a wide range of environmental categories. To this end, results from an attributional LCA model for materials recovery from spent automotive traction NMC battery packs are reported below, accounting for all the necessary steps to recover secondary materials from the point of collection of EoL LIBs, to recovery, processing and displacement of primary material. The inventory developed in this study expands the system boundaries commonly used in battery recycling by compiling state-of-the art literature sources, original materials, and information sourced in the Ecoinvent database (Wernet et al., 2016). Dominant and mature hydrometallurgical and pyrometallurgical cell recycling schemes were modelled and complemented by pre-treatment and processing to recover the metal fractions of the battery pack in order to establish a realistic representation of the circularity in the LIB value chain that can be achieved through recycling. The direct recycling of LIBs, which aims at recovering active materials in the cathode and chemically upgrading these for new cathode manufacturing is also an important emerging opportunity (Gastol et al., 2022; Ji et al., 2021). However, the challenge of how to process different cathode chemistries together in direct recycling still needs to be solved and it is for this reason that direct recycling is not considered in this study.

The environmental burdens and benefits of the hydrometallurgical and pyrometallurgical recycling schemes were aggregated with the battery's production impact, to estimate the role recycling plays in reducing the overall environmental impact of the battery pack. The potential of battery recycling in reducing the GWP production impact was specifically explored for China, United States and Europe, enabling to account for the different electrical energy mixes and material production footprints relevant for each region. Moreover, an open issue regarding the role of recovered materials' quality on the overall environmental performance of battery recycling was explored through a sensitivity analysis on the 13 environmental impact categories under study. Finally, an in-depth analysis of unit process contributions to the environmental benefits and burdens of battery recycling options is provided herein, which contributes towards better understanding the environmental aspects of EoL treatment chains for LIBs.

2. Methods

2.1. LCA methodology

The LCA reporting guidelines according to the ISO 14040 standard series (International Organization for Standardization, 2014) were used in order to create a replicable, transparent and reliable inventory and results. The overarching objective was to develop a detailed Life Cycle Inventory (LCI) and assessment to assess the environmental burdens and benefits associated with recycling of spent automotive traction battery packs. The functional unit was chosen as 1 kW h of nominal battery pack capacity, which consists a standard across literature studies on LIB production (Dai et al., 2019a; Kallitsis et al., 2020) and recycling (Ciez and Whitacre, 2019; Mohr et al., 2020). The technology boundaries were defined based on the current industrial practice, including either hydrometallurgical or pyrometallurgical processes for battery cell recycling (Mohr et al., 2020) and well-established aluminium (The Aluminumn Association, 2013), steel (Burchart-Korol, 2013) and copper recycling processes (Wernet et al., 2016). The battery technology was focused on a typical 253 kg NMC333 battery pack for automotive applications utilising an anode made of graphite (Gr) with a storage capacity of 26.6 kW h, as this technology has been one of the first to be commercialised at scale, thus expected to constitute a large fraction of the first wave of EoL batteries (McKinsey&Company, 2018). Battery packs of similar type have been extensively studied in preceding literature studies on battery production (Ellingsen et al., 2014; Kallitsis et al., 2020), use (Lander et al., 2021b) and recycling (Mohr et al., 2020). While other battery chemistries such as lithium iron phosphate (LFP) have been gaining increased attention for automotive traction applications, most commercial EVs currently include NMC formulations (Lima, 2022) which are projected to dominate the automotive sector (Walton et al., 2021; Xu et al., 2020). Within the NMC family of chemistries, we chose to focus on NMC333 as it has been commercialised widely in EVs, and excluded higher nickel chemistries, such as NMC622, as they are currently being deployed at scale and their availability for recycling is expected in more than a decade's time.

The baseline geographical boundary was limited to China, with materials and energy inputs, production processes and other LCI parameters reflecting the Chinese industrial averages. The environmental burdens and benefits of North American and European recycling chains were also investigated by adjusting key background inventories for Al, Cu and electricity production. A detailed depiction of the studied product system is given in Fig. 1, which includes the (a) transportation of the spent battery pack from its end-user to the recycling facility, (b) dismantling of the battery pack to separate the battery modules from the TMS, BMS and packaging. The battery modules are then disassembled to battery cells, while the supporting structures, including the module and pack housing, which are considered part of the packaging, BMS and TMS are sent for further treatment. (c) Next, the battery cells are discharged and fed to the recycling process, including either hydrometallurgical or pyrometallurgical processing. The former includes a mechanical treatment step (crushing), neutralisation and metallurgical processing, while the latter includes a mechanical treatment step to separate Al and Cu with consequent metallurgical processing. The supporting structures are sent to a metals recycling facility, where the mixed scrap is shredded and sorted, and sent to metal recovery processes. Hence, this work analyses hydrometallurgical and pyrometallurgical processing of the spent battery cells, referred to as the hydrometallurgical and pyrometallurgical cases, with the remaining unit processes unchanged.

The EoL recycling approach was followed (Nordelöf et al., 2019), which considers that the recovered materials displace primary material

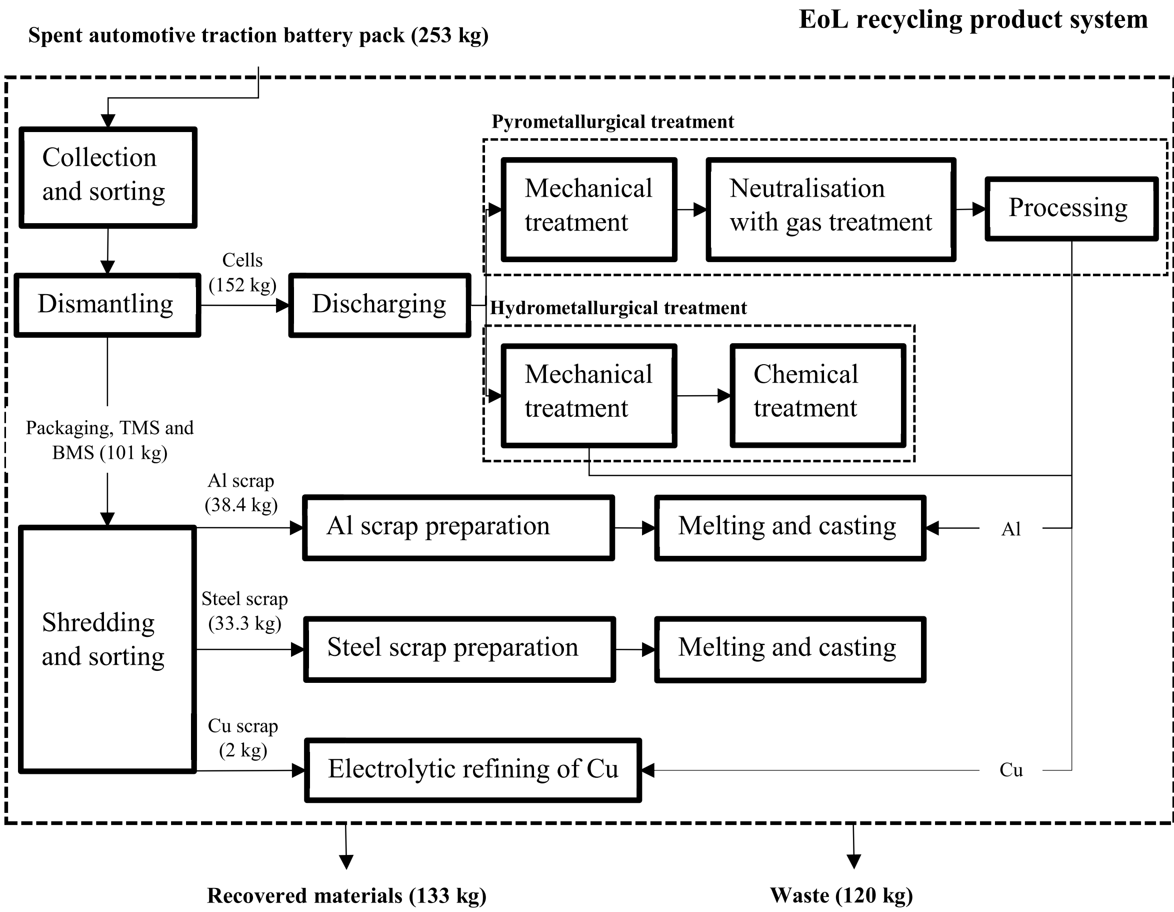


Fig. 1. EoL recycling product system studied for the recovery of valuable metals from a spent automotive traction battery pack (collection of spent battery packs to generation of secondary materials).

upstream in the product system. The EoL treatment burden and benefit was calculated and then added to the production burden to yield the total environmental impact of production and recycling, which was then compared to the battery production impact to calculate the net environmental benefit or burden achieved through the introduction of LIB recycling across all impact categories of interest. The impact assessment was performed based on the ReCiPe 2016 Midpoint characterisation method (Huijbregts et al., 2017), given that it consists of one of the most widely used methods in battery production and recycling LCA studies (Ellingsen et al., 2014; Kallitsis et al., 2020; Oliveira et al., 2015; Sun et al., 2020). The environmental scores for GWP, Fossil Depletion Potential (FDP), Stratospheric Ozone Depletion Potential (SODP), Photochemical Oxidant Formation Potential, human health (POFP), Fine Particulate Matter Formation Potential (FPMFP), Terrestrial Acidification Potential (TAP), Freshwater Eutrophication Potential (FEP), Marine Eutrophication Potential (MEP), Freshwater Ecotoxicity Potential (FETP), Marine Ecotoxicity Potential (METP), Terrestrial Ecotoxicity Potential (TETP), Human Toxicity Potential, cancer (HTP) and Metal Depletion Potential (MDP) were calculated. The LCA modelling was performed using the GaBi software with the background processes mainly being retrieved from the Ecoinvent version 3.5 database (Wernet et al., 2016).

2.2. Inventory analysis

For the collection and sorting inventory, it was assumed that the battery pack is transported by first by freight rail and then by freight lorry to the sorting facility for 688 km and 174 km respectively, based on China's average transport distances (Jiang et al., 2022). Energy inputs associated with the sorting process were taken into account for powering a forklift and conveyor belt based on Fuc et al. (2016) and Fisher et al. (2006); the detailed calculation procedure is described in the supplementary material (SM). The dismantling process was modelled based on the studies of Choux et al. (2021) and Baazouzi et al. (2021). Specifically, we modelled the power consumption of the robotic disassembly machinery used by Choux et al. (2021) based on the task planner of Choux et al. (2021); the detailed calculation procedure is described in the SM. Facility inputs associated with the collection and sorting, and dismantling processes were assumed to be insignificant due to their long lifetime. The LCI for the discharging process was inspired by Yao et al. (2020), who presented a quick discharge device based on the immersion of the battery in $FeSO_4$ solution. Salt-water discharge processes consist the currently dominant option across industrial operations with a series of reagent options being reported across literature studies (Neumann et al., 2022; Shaw-Stewart et al., 2019; Zhong et al., 2020). The discharged battery cells are fed to cell recycling by hydrometallurgical or pyrometallurgical processes. The chemistry-specific foreground LCIs for those processes were taken from Mohr et al. (2020), who modified the corresponding Ecoinvent inventories in order to accurately account for the metal constituents included in the cathode material and the cell housing of NMC battery cells. The LCI for the hydrometallurgical process can be traced back to data from the company Recupyl and for the pyrometallurgical process to the company Batrec. Both processes are currently commercial and have been further discussed by Sommerville et al. (2021) and Neumann et al. (2022). The shredding and sorting of the mixed metal waste of the packaging, BMS and TMS was modelled through an Ecoinvent inventory. Consequently, the aluminium, copper and steel fractions are sent to materials recovery operations, while plastics, electronics, the electrolyte and the coolant are modelled as waste for further treatment. The pre-treatment of aluminium scrap was modelled using an Ecoinvent inventory, while the melting and casting LCI was based on the Aluminium Association's LCA report (The Aluminumn Association, 2013). Steel scrap is sorted and pressed and then fed to an electric arc furnace, with the latter being modelled based on the LCI of Burchart-Korol (2013). Copper scrap is treated by electrolytic refining, based on the corresponding Ecoinvent LCI. The main data sources, assumptions and inventory tables of the LCI are presented and further discussed in the SM.

2.3. Materials recovery and crediting

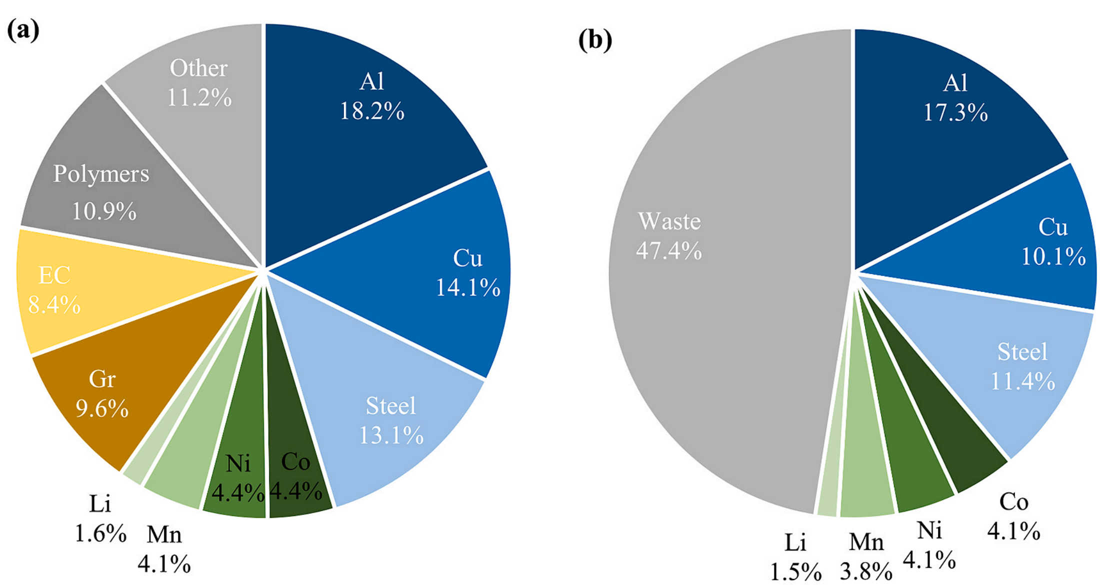
The 253 kg NMC333-Gr battery pack is broken down to its material composition in Fig. 2a, which is in line with a series of preceding literature reports (Cusenza et al., 2019; Ellingsen et al., 2014; Kallitsis et al., 2020). The EoL treatment chain shown in Fig. 1 recovers Al, steel and Cu from the battery's supporting structures, Li^{I} (as hydroxide), Co^{II}, Ni^{II} and Mn^{II} (all as sulfates) from the battery electrodes and elemental Cu, Al from the current collectors, with the recovery for the electrode materials being set to 93.6% and for Al, Cu to 93.8% for the hydrometallurgical process (Mohr et al., 2020). Similarly, the pyrometallurgical process recoveries were set to 93.6% for all materials (Mohr et al., 2020). Similar recovery rates for the electrode materials are considered realistic and have been adopted by a series of preceding studies, with the EverBatt model, for example, including recovery efficiencies of 98% for the key cathode materials (Dai et al., 2019b). Additionally, the recoveries for Al, steel and Cu from the Packaging, BMS and TMS were set to 95.5%, 86.8% and 76.3%, respectively. The recovered Al and Cu from the battery cells was fed to the corresponding melting and casting, and electrolytic refining processes in order to yield secondary materials with high purity. Materials available for recycling comprised 59.5% of the battery pack's mass (Fig. 2a), with the remaining share modelled as waste for further treatment. Fig. 2b presents the recovery of materials from hydrometallurgical recycling of the battery cells, with the combined recovery for each material resulting in the recovery of 52% of the battery pack's mass. Of those, 13.5% comprise cathode materials (Co^{II}, Ni^{II}, Mn^{II}, Li^{I}), while the highest share comes from the recovery of Al, steel and Cu, which are used widely in the packaging, BMS, TMS and the current collectors. For the utilisation of a pyrometallurgical processing scheme, the recovery of Al and Cu from the current collectors are reduced by absolute 0.2%, having a negligible effect on the overall recovery for those metals, and Li^{I} is not recovered. The amounts of recovered materials for both cases are shown in Table A13 of the SM. The displaced materials shown in Fig. 2c were modelled through corresponding Ecoinvent 3.5 datasets, with the exception of cobalt sulfate, which was modelled through the more recent Ecoinvent 3.8 dataset.

Table c in Fig. 2 illustrates the displaced primary material from the recovery of metal fractions from the battery pack. For Al, Cu and steel, their recovery is followed by melting and casting or electrolytic refining as shown in Fig. 1, which transform scrap to secondary material of sufficient quality to displace primary material. For battery electrode materials, recovered material quality might not meet the requirements for battery production. This issue was addressed by assuming that recovered electrode materials are battery-grade as a baseline, in line with literature studies (Mohr et al., 2020), and further exploring the effect of recovered materials quality within a sensitivity analysis.

3. Results and analysis

3.1. Environmental impact of recycling

The environmental impact of battery recycling is presented in Fig. 3 for the pyrometallurgical and hydrometallurgical cases and broken down to the key contributing processes. Hydrometallurgical processing of the battery cells contributed 51% towards the GWP with an important contribution also arising from the electrolytic refining of copper (19%). Similarly, 55% and 18% of the FDP arose from cell recycling and



(c)

Recovered material	Al	Cu	Steel	Co^{II}	Ni^{II}	Mn^{II}	Li^{I}
Avoided material	Primary Al ingot	Primary Cu	Low-alloyed steel	$CoSO_4$	$NiSO_4$	$MnSO_4$	LiOH

Fig. 2. (a) Mass composition of the 253 kg NMC333-Gr automotive traction battery pack, (b) recoverable fractions and waste occurring from the hydrometallurgical case and (c) displaced primary materials from the recovered metal fractions of the battery pack; stoichiometric elemental contributions are displayed for the cathode materials.

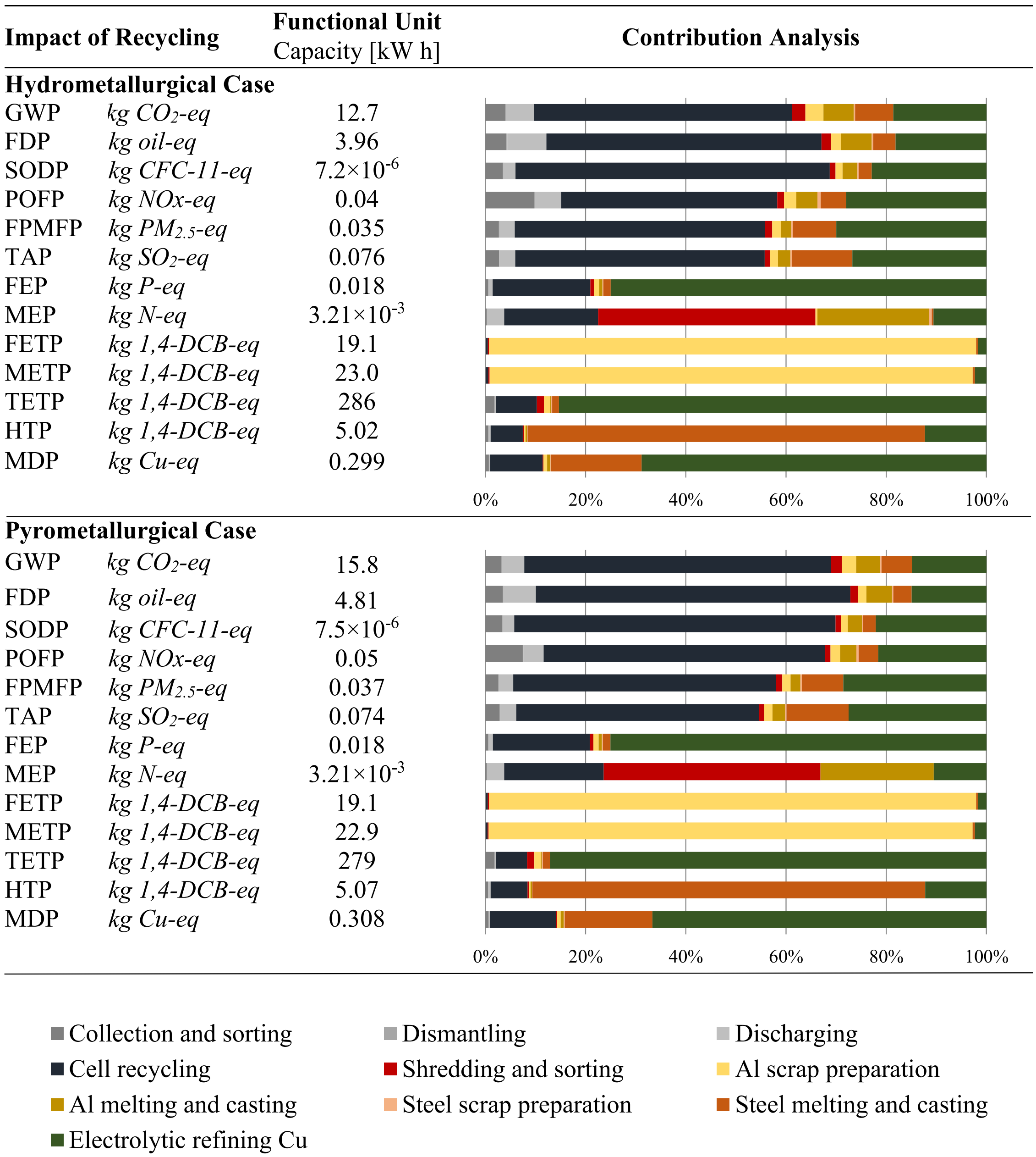


Fig. 3. Environmental impacts of an NMC333-Gr battery pack recycling in China, disaggregated to key contributing processes. Battery cells are treated with either hydrometallurgical or pyrometallurgical processing in the two cases.

electrolytic refining of copper, respectively. The SODP was also dominated by the cell recycling (63%) and electrolytic refining of copper (23%) processes. Similarly, cell recycling and electrolytic refining of copper constituted the dominant contributions towards POFP, FPMFP, TAP and FEP. In the pyrometallurgical case, the GWP, FDP and POFP were increased by 25%, 21% and 30%, respectively, mainly attributed to the almost six times higher electrical energy consumption compared to the hydrometallurgical case; with the remaining impact categories variating by less than 5%.

The FEP was dominated by the electrolytic refining of copper, which contributed more than 75% in both cases. The MEP was built up from the shredding and sorting, aluminium melting and casting, and the cell recycling processes. More than 96% of the FETP and the METP arose from the preparation of aluminium scrap in both cases. The electrolytic refining of copper comprised the primary contribution towards TETP and MDP, while the steel melting and casting process dominated the HTP.

3.2. Environmental benefit of recycling

The recycling chain modelled in this study recovers Al, Cu and steel from the battery pack's supporting structures and Al, Cu, Ni^{II}, Co^{II}, Mn^{II} and Li^{I} from the battery cells, with the latter being recovered only in the case of hydrometallurgical processing. Specifically, the hydrometallurgical case recovered 26.1 kg of $NiSO_4$, 26.3 kg of $CoSO_4$, 25.5 kg of $MnSO_4$ and 12.4 kg of LiOH from the battery cells. Additionally, 44.5 kg of Al, 29 kg of steel and 26.2 kg of Cu were recovered from the 253 kg battery pack's supporting structures and the current collectors. The fraction of materials recovered in the pyrometallurgical case were very similar, as presented in section 2.3, excluding LiOH, which was not recovered. More information on the background datasets used to model materials displacement is provided in the SM.

The environmental benefit arising from the recovery of the battery pack's materials is illustrated in Fig. 4. The hydrometallurgical case resulted in a larger benefit by a maximum of 3% across all impact categories, with the exception of MEP (9%) and MDP (8%), due to the additional recovery of lithium as hydroxide. For both cases, approximately half of the GWP benefit was due to the recovery of aluminium, with the next most important contribution coming from the recovery of Co^{II}. Similarly, approximately half of the FDP benefit was due to the recovery of cobalt, with the next most important contribution coming from aluminium. Recovered copper, Co^{II} and aluminium provided the dominant contributions towards the POFP and SODP benefits. Recovered Ni^{II} provided the primary contribution towards the FPMFP and TAP benefits. The eutrophication and ecotoxicity impact categories were dominated by the recovery of copper and Co^{II}, with 9% of the MEP benefit arising from the recovery of Li^{I} in the hydrometallurgical case. The MDP benefit was dominated by the recovery of Co^{II}, with Ni^{II}, copper and Li^{I} having also important contributions. The percentage contribution of lithium recovery in MDP and MEP explains the larger benefit achieved in the hydrometallurgical case.

4. Discussion

4.1. Reducing the environmental impact of battery production

The net benefit of battery recycling is commonly accounted together with the environmental burden of LIB production to investigate to what extent the use of recycled material in the battery value chain reduces the production burden. The LIB production model of Kallitsis et al. (2020) was used here to calculate the environmental burden of battery production across the 13 impact categories of interest; the inventory model was modified to describe a giga-scale battery production plant (Kallitsis, 2022), to account only for primary material inputs and update the cobalt production inventory based on Ecoinvent 3.8. The combined production and recycling burden is shown in Table A17 of the SM, including all the individual contributions. For both cases, accounting for battery pack recycling resulted in a net environmental benefit across 11 impact categories, with the exception of FETP and METP. The treatment of Al scrap introduced a significant burden to the latter two impact categories, resulting in a net increase of 61% and 35%. For the hydrometallurgical case, the GWP, FDP, SODP, POFP, FPMFP and TAP were reduced by 39%, 41%, 55%, 48%, 70% and 74%, respectively. As these impact

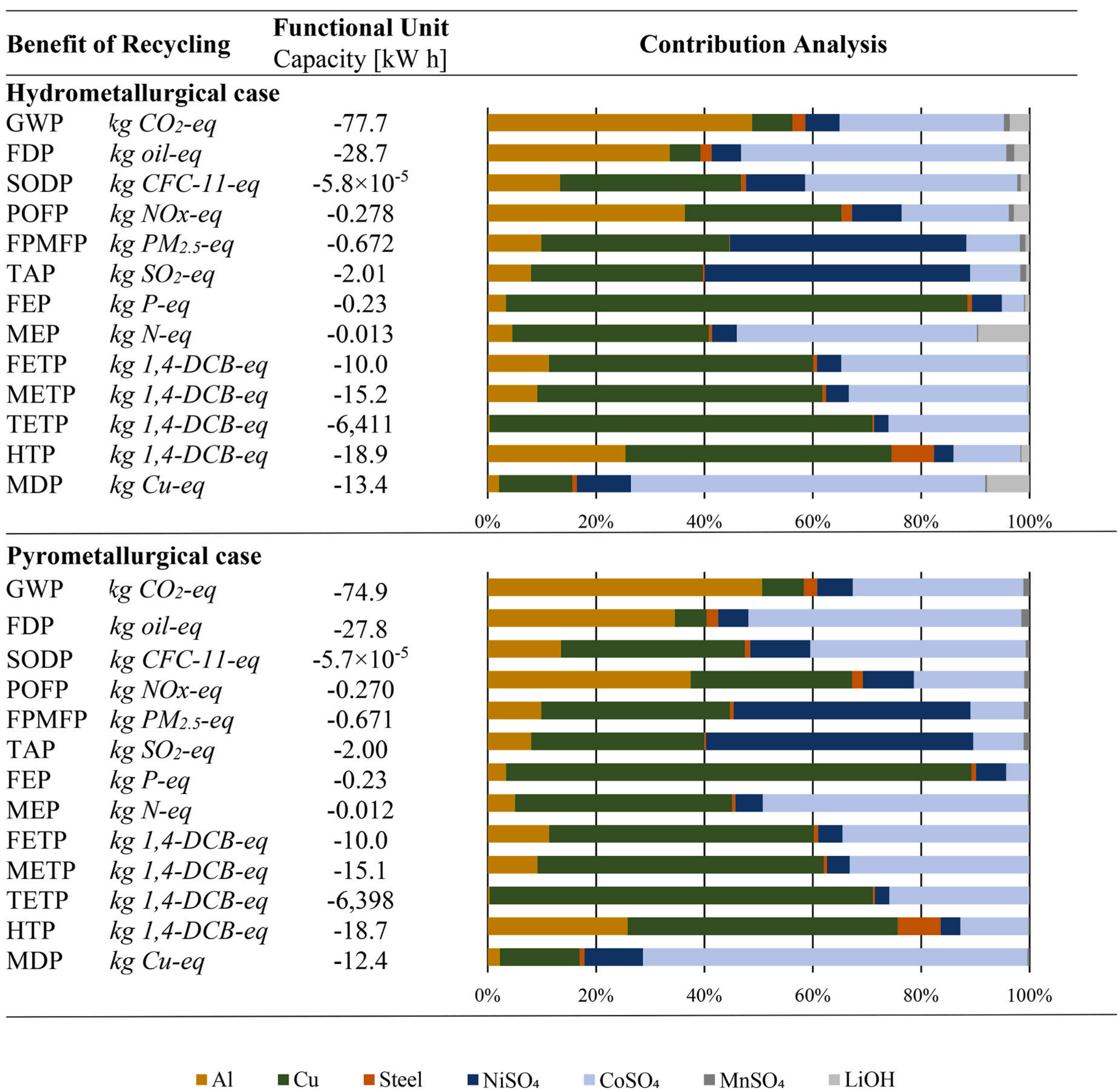


Fig. 4. Environmental benefits of an automotive NMC333-Gr battery pack recycling in China, disaggregated to key contributing processes. Environmental benefits arose from the displacement of primary material based on the EoL recycling approach.

categories had their most important contributions from the cell recycling process, the benefits were of slightly lower magnitude in the pyrometallurgical case, due to the increased energy consumption and the loss of Li^{I}. Benefits of more than 50% were achieved for FEP and TETP, with hydrometallurgical and pyrometallurgical cases resulting in quantitatively similar improvements. The MEP benefit of the hydrometallurgical case was 52%, absolute 7% higher than the pyrometallurgical case, with the variation in the former attributed solely to the recovery of Li^{I}, which also caused a higher MDP benefit of 84%, compared to 77% in the pyrometallurgical case. Finally, the HTP was approximately halved in both cases, with a slightly larger benefit predicted in the hydrometallurgical case, due to the additional recovery of lithium(I).

NMC battery packs have been shown to have significant GWP footprints, resulting from processing of energy intensive materials and the high energy demand of cathode preparation and cell manufacturing (Dai et al., 2019a; Ellingsen et al., 2014; Kallitsis, 2022; Kelly et al., 2019; Majeau-Bettez et al., 2011). This work has shown that establishing a circular battery value chain can reduce LIBs' production GWP footprint by more than 35%, with this benefit mostly due to the recovery of aluminium, which comes from recycling the battery packaging, as its energy intensive primary production in China comes with a high GWP. Nickel and copper production have been identified as hotspots for sulfur dioxide emissions (Kallitsis et al., 2022; Kelly et al., 2019) resulting from the battery production chain; the recovery of those materials was predicted to reduce the TAP by more than 70%. While earlier studies have raised concerns for the increased HTP occurring from the production of battery packs (Brennan and Barder, 2016; Hawkins et al., 2013), HTP was shown here to reduce by approximately 50% when including battery recycling, compared to Kallitsis et al.'s (2020) battery production model. LIB recycling was also shown to significantly avoid depletion of valuable metals, with the MDP reducing by 84% and 77% in the hydrometallurgical and pyrometallurgical case, respectively. Overall, EoL treatment of spent automotive traction battery packs was shown to mitigate the environmental impact across the majority of environmental categories. It is noted that although a few of the impact categories discussed are not as accurate as GWP, a full assessment on the accuracy of environmental scores is not necessary for comparative evaluations. Specifically, most of the background system used is retrieved from the Ecoinvent database, thus inventories fit the necessary accuracy and completeness criteria to assess the full range of environmental scores. Moreover, the foreground system was established to accurately describe resource flows within the EoL treatment chain, with the majority of environmental impacts arising from the background inventories linked to those flows.

4.2. Sensitivity analyses

The geographic specificity of the battery value chain is an important factor when estimating the environmental benefit and burden of battery recycling, as it affects emissions at local level, depends on a varying intensity of embodied emissions from local energy use, and also dictates the effective footprint adjustment required for the displaced material. Moreover, the effect of recovered materials quality on the overall environmental benefit of battery pack recycling remains to be characterised. Here, two sensitivity analyses were performed concerning (a) the effect of geographic specificity on the combined production and recycling GWP and (b) the effect of recovered material's quality on all environmental impact categories studied. For the geographic specificity, GWP was chosen for the analysis as it reflects clearly the energy mix related benefits and impacts, which vary substantially across geographic locations.

The LIB production impact was combined with the recycling burden and benefit of this study to estimate the combined GWP of LIBs in China, North America and Europe, excluding the use phase. In order to calculate the LIB production impact, representative Ecoinvent inventories for each location were selected for electricity, aluminium and copper production, which are shown in the SM. The burden and benefit of battery recycling was calculated for each case by modifying the input electricity and the displaced primary material sourcing for Al and Cu, with the remaining materials being modelled through global Ecoinvent inventories across all scenarios. Al and Cu are considered specifically as their footprints vary highly across locations and they contribute substantially to the GWP. By modifying the background datasets for Al and Cu it is possible to provide a realistic comparison, as more than 55% of the GWP benefit arose from their recovery (see Fig. 4). Remaining battery cell materials were produced through globally-distributed supply chains. This is appropriate as active material mining and processing ($NiSO_4$ and $CoSO_4$) is geographically concentrated in only a few countries around the world; therefore, capturing any differences from sourcing of the active materials used in the US, China and Europe would only be due to transportation distance impacts, which are much smaller and also less variable than the footprint of production of the active materials. On the production side, approximately 60% of the NMC LIB production GWP was due to the energy requirement, Cu and Al inputs, based on the modified model of Kallitsis et al. (2020). This analysis assumes that only input materials and energy vary across locations, with the manufacturing and recycling technologies being practically the same. This assumption also allows to explore the effect of establishing regionalised battery production and recycling chains, considering that secondary material displaces the equivalent amount of primary material in the same region. The modifications for each scenario are described in detail in Table A14 of the SM.

Fig. 5 illustrates the GWP of battery production compared to the combined GWP of production and EoL recycling in China, North America and Europe. Including recycling resulted in a net benefit of 39% and 35% for the hydrometallurgical and pyrometallurgical cases in China, which was reduced to 34% and 30% for North America, and 30% and 27% for Europe, respectively. Evidently, larger GWPs of battery production offer a bigger opportunity for improvement, with a battery produced in Europe shown here to come with a 27% lower GWP compared to a battery produced in China, with the relative difference reduced to approximately 16% when recycling is considered. This demonstrates how regions with high battery production emissions such as China can utilise battery recycling as a method to catch up on the race to reduce the GWP of battery production.

The effect of recovered materials quality on the environmental benefit of LIB recycling was explored by displacing the equivalent amount of ore-grade (OG) materials instead of battery-grade (BG) for Ni^{II}, Co^{II}, Mn and Li^{I}. The percentages of each metal included in the concentrated OG form was taken into account in order to convert the recovered materials in elemental form into ore grade materials as shown in Table A15 of the SM. Fig. 6 presents the hydrometallurgical case, with the pyrometallurgical case not recovering lithium but being quantitatively similar for the recovery of the remaining materials.

The POFP, FEP and HTP benefits were reduced by less than 15% when accounting for the recovery of OG material, due to the fact that the majority of the environmental benefit in those impact categories arose from the recovery of Al and/or Cu from the battery pack. The benefit in the ecotoxicity impact categories, SODP and GWP was reduced by 20%–40%, which is reasonable given that electrode materials contribute to these impact categories significantly but are not the primary contributors. However, for those impact categories dominated by the recovery of cobalt and nickel (namely FDP, FPMFP, TAP, MEP and MDP) the benefit in the OG case was decreased by 40%–60%. Specifically, the FPMFP and TAP benefits were reduced by 52% and 58%, respectively, in the OG case mainly due to the recovery of nickel of ore-grade quality, as the further processing of nickel ore is known to have a detrimental effect in those environmental impact categories (Norgate et al., 2007; Paulikas et al., 2020). Similarly, cobalt refining is known to cause detrimental eutrophication impacts and consume fossil energy, which explains the large variability between the FDP and MEP benefits of the OG and BG

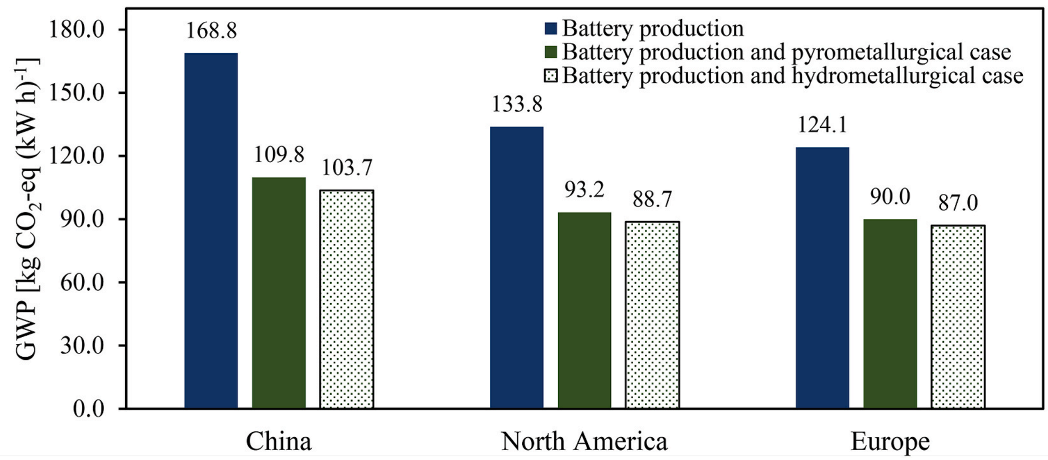


Fig. 5. GWP of LIB production compared to production with EoL recycling for the hydrometallurgical and pyrometallurgical cases in China, North America and Europe.

cases (Paulikas et al., 2020). A recent study by Rajaeifar et al. (2021) employed such multi-functionality approach in quantifying the impact of recovering OG materials versus BG materials. However, their system boundary was limited to pyrometallurgical cell recycling and only quantified implications associated with the GWP. Their findings showed that the GWP benefit reduces more than 50% when recovering lower grade materials in comparison to 28% decrease reported here. The difference can be attributed to the narrower system boundary of Rajaeifar et al. (2021); our study showed that recovering cell materials is only a fraction of the benefit of recycling a battery pack. This work has also shown that the effect of recovering lower grade materials is much more evident in impact categories such as FDP, FPMFP, TAP, MEP and MDP, which was not captured by Rajaeifar et al. (2021). Overall, this sensitivity analysis has shown that the positive impact of recycling is more significant when considering the effective displacement of BG quality materials in the value chain, especially in the FDP, FPMFP, TAP, MEP and MDP impact categories, thus supporting strongly the case for integration of regionalised battery production and recycling facilities. The quantitative evidence provided here offers a valuable methodological resource for LCA practitioners and supports decision making for recyclers and policy-makers interested in establishing sustainable battery recycling operations.

4.3. LIB recycling environmental performance and comparison with preceding studies

Two key observations are made based on the findings of this study, which help explain the variability on preceding literature studies that consider recycling, as shown in Table 1. Firstly, the majority of the environmental benefits of battery recycling arise from the recovery of Al and Cu fractions of the battery pack (Fig. 4), with important contributions also arising from the recovery of nickel and cobalt from the electrodes. Therefore, studies focusing on the cell level only are expected to report lower benefits, as compared to those focusing on pack level recovery. Secondly, geographic specificity plays an important role on the calculated net benefit of battery recycling, with more environmentally burdening chains expected to come with higher net benefits when recycling is introduced (Fig. 5). Ciez and Whitacre (2019) reported insignificant GWP intensity reductions achieved through hydrometallurgical and pyrometallurgical processing of NMC battery cells; the latter resulted in a net increase. More recently, Mohr et al. (2020) reported a −13.8 and −16.4 kg CO_2-eq $(kW\ h)^{-1}$ net benefit achieved through hydrometallurgical and pyrometallurgical processing of NMC battery cells. These observations are reasonable, given that these studies focused on cell level and modelled less environmentally burdening chains outside of China. Cusenza et al. (2019) reported a −15.4 kg CO_2-eq $(kW\ h)^{-1}$ net benefit for pack level recycling of a LMO-NMC333, while Sun et al. (2020) reported a −30.9 kg CO_2-eq $(kW\ h)^{-1}$ net benefit for hydrometallurgical recovery of NMC622. While both studies focused on pack level, the former modelled a European and the latter a Chinese recycling chain, so partly explaining the large variability together with the different chemistries modelled in each study. Additionally, Cusenza et al. (2019) modelled a combined hydrometallurgical-pyrometallurgical recycling scheme, which was also modelled by Zhou et al. (2021), and was more environmentally burdening. A similar scheme was modelled by Accardo et al. (2021), reporting recycling to come with a net benefit of −6.2 kg CO_2-eq $(kW\ h)^{-1}$ in Europe. This study's net benefit for pyrometallurgical recovery through a European recycling chain was −34.1 kg CO_2-eq $(kW\ h)^{-1}$, with the difference attributed to the more environmentally burdening hydrome tallurgical-pyrometallurgical recycling scheme modelled by Accardo et al. (2021). Moreover, in this study the displaced cobalt sulfate (based on the latest Ecoinvent 3.8database) was shown to comprise the second most important contribution towards GWP reductions. The net recycling benefit for the hydrometallurgical case in China was −65.1 kg CO_2-eq $(kW\ h)^{-1}$ here, while Sun et al. (2020) reported a net benefit of −30.9 kg CO_2-eq $(kW\ h)^{-1}$ for similar parameters. Although the contributions of the recycling burden and benefit to the net benefit in Sun et al. (2020) were not reported, the difference can be mainly attributed to the updated cobalt sulfate inventory utilised herein, which was found to contribute 30% towards the GWP benefit. Another key source of variability is associated with varying NMC compositions resulting in different energy densities. This becomes more evident when looking at the sensitivity analysis performed by Mohr et al. (2020), who reported that increasing the energy density reduces the combined production and recycling burden given that under the same mass a higher capacity is achieved when shifting to higher nickel chemistries primarily affecting the kW h functional unit.

4.4. Recovered materials value

This study highlighted the need to recover the total set of battery pack materials in order to achieve maximum environmental benefits from LIB recycling. From an environmental perspective, benefits arise primarily from the recovery of Al and Cu from the battery pack. Here, the economic value for each recovered material in the hydrometallurgical and pyrometallurgical cases is calculated as shown in Fig. 7. Elemental prices were extracted from United States Geological Survey (2020) for 2019 and multiplied by the amount of each recovered material to estimate the economic benefit achieved from the recovery of

Impact category	Units		Benefit per capacity [kW h]
GWP	*kg CO_2-eq*	BG	-77.7
		OG	-52.5
FDP	*kg oil-eq*	BG	-28.7
		OG	-13.9
SODP	*kg CFC-11-eq*	BG	-5.8×10^{-5}
		OG	-3.9×10^{-5}
POFP	*kg NOx-eq*	BG	-0.278
		OG	-0.256
FPMFP	*kg $PM_{2.5}$-eq*	BG	-0.672
		OG	-0.322
TAP	*kg SO_2-eq*	BG	-2.01
		OG	-0.85
FEP	*kg P-eq*	BG	-0.23
		OG	-0.22
MEP	*kg N-eq*	BG	-0.013
		OG	-0.006
FETP	*kg 1,4-DCB-eq*	BG	-10.0
		OG	-6.4
METP	*kg 1,4-DCB-eq*	BG	-15.2
		OG	-9.8
TETP	*kg 1,4-DCB-eq*	BG	-6,411
		OG	-4,598
HTP	*kg 1,4-DCB-eq*	BG	-19.0
		OG	-16.2
MDP	*kg Cu-eq*	BG	-13.5
		OG	-7.97

Contribution Analysis

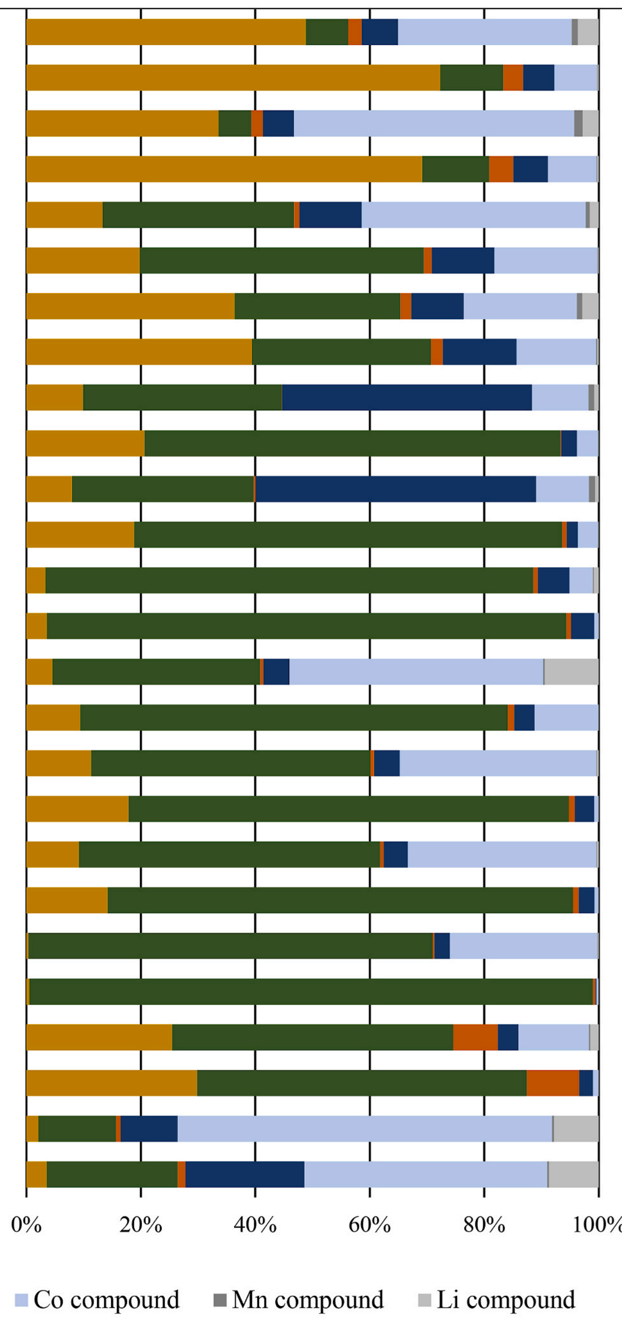


Fig. 6. Environmental benefit for battery-grade (BG) vs ore-grade (OG) material recovery in the case of hydrometallurgical treatment.

each battery material. As currently recovered materials may not be of sufficient quality to displace primary material of the same price, Fig. 7 should be seen as an indication of what is possible, when recycled material quality issues are resolved. The details of the calculation procedure implemented are shown in the SI.

It should be noted that price volatility is typical in raw materials and is affected by many short- and longer-term geopolitical factors, thus the analysis of recovered materials value presented here should be taken as indicative snapshot in time, for the purpose of comparing the hydrometallurgical and pyrometallurgical options.

The value of recovered material in the hydrometallurgical case was USD 1 079, 32% higher than in the pyrometallurgical case. This is connected directly to the additional recovery of Li^{I} through hydrometallurgical processing of the battery cells, which accounted for a quarter of the total (Fig. 7a). The recovery of Ni^{II}, Co^{II}, Mn (as sulfates) and Li^{I} (as hydroxide) accounted for three quarters of the total economic value

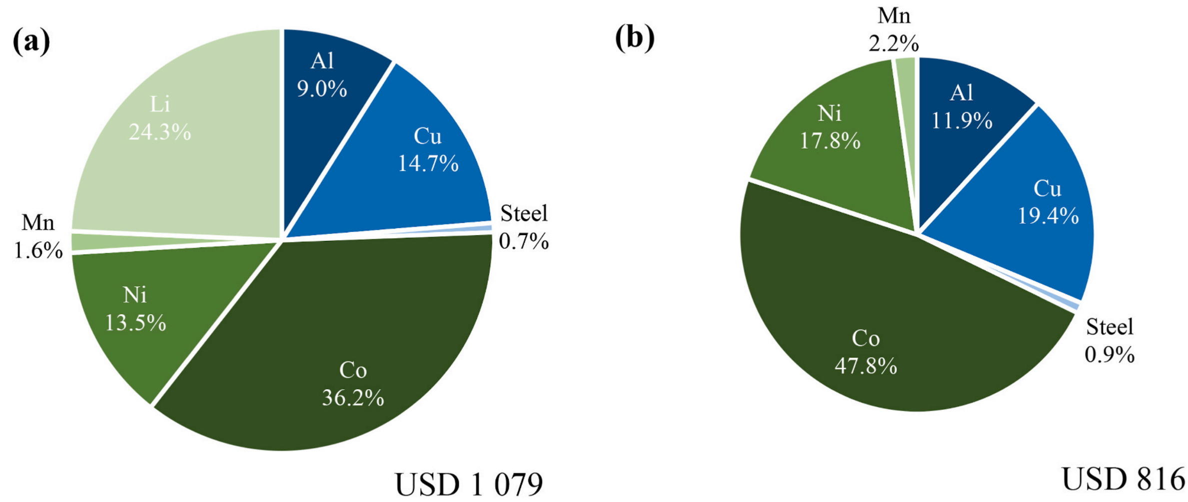


Fig. 7. Economic value of recovered material for one battery pack using the (a) Hydrometallurgical (total: USD 1 079) and (b) Pyrometallurgical (total: USD 816) processes.

of recovered materials in the hydrometallurgical case (Fig. 7a) and the recovery of Ni^{II}, Co^{II} and Mn^{II} (sulfates) accounted for 67.8% of the total in the pyrometallurgical case (Fig. 7b). While the recovery of lithium (hydroxide) contributed less than 6% in 11 out of 13 environmental impact categories studied (Fig. 4), it increased the economic value of recovered materials by 32% in the hydrometallurgical case. The two cases have different capital and operational cost patterns, which were not examined in this study, but the recovery of Li (hydroxide) is indeed an important factor on the economic viability of recycling operations. Finally, while environmental benefits primarily arose from the recovery of Al and Cu, economic benefits primarily arose from the recovery of cell materials, Ni^{II}, Co^{II} (sulfates) and Li^{I} (hydroxide), which highlights the need to target circularity at a pack level to unlock both the environmental and economic benefits of battery recycling.

4.5. Data gaps, limitations and future perspectives

This study aimed to expand the description of the EoL treatment of automotive LIBs, from the point of collection to displacement of primary material, and to fill data gaps by combining a series of publicly available data sources. However, a series of improvements in the LCI could be made through the collection of process specific data. This study utilised Chinese average transport distances to model the collection of spent LIB packs and showed that its contribution to the overall environmental burden of battery recycling is insignificant, in line with Jiang et al. (2022). However, the collection process has been reported to be significant from an economic and safety perspective (Lander et al., 2021a; Slattery et al., 2021). As automotive LIB recycling is under development, average transportation distances could be confirmed through a more thorough investigation of EoL LIB collection and transportation networks. A first attempt to compile an inventory robotic disassembly was made in this study by calculating the energy inputs for robotic dismantling. As robotic dismantling is under development, with manual dismantling being the currently dominant practice (Sommerville et al., 2021) more complete inventories should be compiled as the technology matures. The dismantling process was found here have to have an insignificant contribution to the overall environmental burden of battery recycling. Rajaeifar et al. (2022) also indicate that the significance of dismantling is most evident with regard to the cost and safety perspectives. The discharging inventory was inspired from the quick discharge device presented by Yao et al. (2020). Different physical and chemical discharging techniques are being developed, which might dominate in the future and come with a different environmental profile. The battery cell recycling inventory was inspired from Mohr et al. (2020), with the original data traced back to the Ecoinvent database. Currently, there is a lack of primary data regarding hydrometallurgical and pyrometallurgical cell recycling techniques which could be improved as more LIB recycling plants reach commercial scale. Also, the fate of the anode material in cell recycling operations is still uncertain, which explains why it has been treated as waste in this work. Current pyrometallurgical process usually burn graphite for energy, together with plastics and the electrolyte, while little is known regarding the fate of graphite in hydrometallurgical processes (Sommerville et al., 2021). More advanced options, which include the recovery of graphite and have been shown to be favourable from an environmental perspective have been discussed in recent studies (Mohr et al., 2020; Rey et al., 2021), however, whether these can be sustained at scale is unclear and would depend on the economics. Graphite has been recognised as a valuable waste stream from recycling processes (Rinne et al., 2021) but its recovery usually requires further process steps, which is indeed worth of further investigation as it consists a large contribution to the overall mass of the battery pack (Porzio and Scown, 2021). Also, the quality of recycled material is important, as it determines whether recovered cell materials can be recirculated to LIB production in a closed-loop system. Regarding the LIB pack's packaging, TMS and BMS, the process chains to recover Al, Cu and steel from them are well-established. The Ecoinvent inventory was used here to model shredding of this mixed metal waste, which is notably different from shredding of complete LIB modules (Rajaeifar et al., 2021). The different environmental profile achieved through the displacement of ore-grade materials was explored in the sensitivity analysis of section 4.2. The examination of locality effects on the environmental burdens and benefits of LIB recycling was found to be an important source of variability, mainly due to different footprints of displaced materials. As upstream battery material production chains emerge outside of China, the environmental benefit of battery recycling is expected to come with even higher variability across different locations.

5. Conclusions

Establishing a circular battery value chain was shown to mitigate the environmental impacts significantly across the majority of environmental impact categories considered. Specifically, employing hydrometallurgical or pyrometallurgical battery cell recycling schemes, together with the recovery of metal fractions from battery packs' supporting structures was shown to reduce the environmental burden of battery production by more than 35% across 11 out of 13 environmental impact categories considered. Pyrometallurgical processing of battery cells was associated with a greater GWP burden, so a slightly smaller benefit, mainly due to the almost six times higher electrical energy consumption. The hydrometallurgical case resulted in a larger benefit by a maximum of 9% for the MEP, due to the additional recovery of lithium hydroxide. The majority of the environmental burdens in the toxicity and ecotoxicity categories arose from the recovery chains for aluminium, copper and steel. Half of the GWP benefits were achieved from the recovery of aluminium, mainly included in the battery pack's packaging. Across the 13 impact categories considered, the recovery of Al and Cu provided more than half of the environmental benefit in 7 of them, highlighting their significance in reducing the total environmental impact of a battery pack. However, the recovery of battery cathode materials, especially nickel and cobalt, was shown to make an important contribution to the air quality, toxicity and resource depletion categories. These results highlight the importance of targeting circularity at the pack level to establish sustainable LIB recycling chains. The combination with the LIB production burden revealed that LIB recycling approximately mitigates the TAP by 74%, MDP by 77%, HTP by 50% and GWP by 35%. The investigation of geographic specificity effects on the combined production and recycling GWP revealed that more environmentally burdening chains offer a significantly higher potential for GWP reductions through battery recycling and that geographic specificity accounts for significant variability amongst LCA studies on LIB recycling. As a significant fraction of the environmental benefits arose from the recovery of Al and Cu from the battery pack, together with geographic specificity were found to constitute key sources of variability amongst literature studies reporting results for varying system boundaries and locations of focus.

Decision makers and other stakeholders concerned about the environmental impacts of the LIB value chain can take steps towards their reduction. Firstly, they should target circularity on a pack level, which was shown to unlock the environmental benefits potential of battery recycling, as compared to the cell level. Starting a process of collecting, sorting and separating battery pack components and feeding the fractions in battery cell recycling and well-established metals recovery processes is a hugely important strategy towards the realisation of the environmental benefits of battery recycling; such business models are currently being explored (Berger et al., 2021; Wrålsen et al., 2021). The recovery of Al and Cu from the battery pack, together with the recovery of valuable battery cell materials Ni^{II}, Co^{II} (sulfates) and Li^{I} (hydroxide) can lead to economically beneficial and sustainable battery recycling. However, as recycling can be beneficial only upstream of the value chain, these efforts should be combined with sustainable battery production practices, the utilisation of renewable energy during battery production and the target to ensure longevity of battery systems. These, together with developments in battery technology, could lead to substantial reduction of the cradle-to-grave environmental footprint of LIBs in the near future.

CRediT authorship contribution statement

Evangelos Kallitsis: Conceptualization, Methodology, Software, Investigation, Formal analysis, Writing – original draft. **Anna Korre:** Conceptualization, Supervision, Methodology, Writing - review & editing. **Geoff Kelsall:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

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