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LIFE CYCLE ASSESSMENT OF SCENARIOS FOR END-OF-LIFE MANAGEMENT OF LITHIUM-ION BATTERIES FROM SMARTPHONES AND LAPTOPS

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ABSTRACT

Recycling lithium-ion batteries (LIBs) is a solution to minimise the environmental problems caused by the consumption of natural resources and the generation of hazardous waste. This paper aims to assess the potential environmental impacts and benefits of four scenarios for recycling LIBs from smartphones and laptops using Life Cycle Assessment (LCA). The methodological approach followed four steps: i) scenario modelling representing the current and future situations of LIBs End-of-Life (EoL) management from smartphones and laptops; ii) estimating smartphones, laptops and respective LIBs waste generation; iii) mapping representative recycling options; and iv) assessment of potential environmental impacts using LCA with 16 ILCD midpoint categories. The results revealed that hydrometallurgical recycling in Brazil could be less harmful than pyrohydrometallurgical recycling in Europe in 12 impact categories. The benefits of recycling are mainly of Co and Ni recovery. Results of scenarios indicate that the more optimistic scenario, which includes expanding Reverse Logistics to 50% of collection, internal recycling to 75%, and reducing of LIBs waste sent to landfills in 44%, had the best environmental performance in all 13 impacts categories. For the Climate change category, scenario 4 presents net environmental benefits of -1.83E+05 kgCO_2eq while scenarios 1, 2 and 3 do not present a net environmental benefit. Scenarios assessment shows that more significant environmental benefits are achieved when the formal collection rate is increased, and the less impactful technology option makes the recovery of materials. These results can help decision-makers promote the management and recycling more sustainable of LIBs waste.

1. INTRODUCTION

The increasing use of Lithium-ion batteries (LIBs) generates thousands of tons of LIBs waste per year (Larouche et al., 2020; Meshram et al., 2020). LIBs waste is one of the fastest-growing segments in global solid waste streams (Song et al., 2017). The flow of Waste Electric and Electronic Equipment (WEEE) contains large amounts of LIBs because portable devices, such as smartphones, tablets and laptops, attach a battery to supply energy (Song et al., 2019; Mejame et al., 2020). Estimates indicate a 30% growth in demand for LIBs annually by 2030 (Hanicke et al., 2023). This increase is driven by rising demand for portable electronics and other applications that utilize LIBs, such as Electric Vehicles (EV) and energy storage (Bauer et al., 2022; Fahimi et al., 2022). As sales increase, larger volumes of LIBs waste will enter the solid waste stream (Boyden et al., 2016; Mejame et al., 2020). This scenario raises concerns about the environmental impacts of consuming natural resources to produce LIBs and and the management of these hazardous wastes at the End-of-Life (EoL) stage (Or et al., 2020; Xu et al., 2020; Lybbert et al., 2021).

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The production of LIBs requires various metallic raw materials and chemicals (Zubi et al., 2018), including cobalt, lithium, nickel and graphite. Improper EoL disposal of LIBs must be avoided to prevent adverse effects on human health and the environment (Winslow et al., 2018; Mrozik et al., 2021). LIBs discarded with Municipal Solid Waste (MSW) can cause fires in collection vehicles and landfills (Herreras-Martínez et al., 2021). Electrolytes can lead to contamination with hazardous substances such as arsenic and phosphorus (Jin et al., 2022). Improper treatment results in the loss of valuable resources (Meshram et al., 2020). Beyond direct environmental impacts, the LIBs supply chain also has serious social consequences. Cobalt mining in the Democratic Republic of Congo involves human rights violations, including forced labor and child labor (Amnesty International, 2017; Harper et al., 2019). Lithium production in Latin America negatively affects local communities' access to drinking water, agriculture, and fishing, leading to human and indigenous rights violations (Henríquez, 2018; Schüler et al., 2018).

LIBs recycling offers significant potential for generating economic, environmental, and social value (Dai et al., 2019). Environmentally, recycling reduces waste and pollution, conserves natural resources and ecosystems while increasing resource efficiency (Harper et al., 2019). Socially and economically, recycling strengthens the economy by creating new jobs, wages, and tax income (EPA, 2020), and it also presents profitable business opportunities (Latini et al., 2022; Lima et al., 2022). The recovery of raw materials from LIBs prevents environmental pollution and resource loss, provides a secondary supply of materials, and contributes to Circular Economy and Sustainable Development (Ducoli et al., 2022; Fahimi et al., 2022).

The technologies for recycling LIBs are classified into three types (Harper et al., 2019; Jin et al., 2022): hydrometallurgical, pyrometallurgical and direct recycling. Pyrometallurgical recycling uses high-temperature thermal processing, including smelting, pyrolysis, incineration or torrefaction/calcination to reduce metal oxides in metal alloys (Or et al., 2020; Makuza et al., 2021). Hydrometallurgical recycling involves a chemical process that uses aqueous solutions with chemical reagents to leach metals from the cathode material (Yun et al., 2018; Raj et al., 2022). Direct, or functional, recycling reconditions and recovers negative and positive electrode materials for reuse in LIBs, avoiding the separation and dissolution of multiple metallic ions in their elemental form (Harper et al., 2019; Kim et al., 2021). While some studies indicate an environmental and economic advantage of hydrometallurgy compared to pyrometallurgy (e.g., Dunn et al., 2022; Jin et al., 2022; Chen et al., 2023), there is still no definitive consensus on the environmentally preferred recycling method (Rey et al., 2021; Rajaeifar et al., 2022).

Recycling technologies for LIBs waste are in the early stages of development and commercialization (Rhee, Jang & Kim, 2021; Rajaeifar et al., 2022). The LIBs recycling chain encounters political, technical, and socioeconomic challenges globally, particularly in developing countries, such as a lack of commercial recycling infrastructure, disposal points, and adequate collection vehicles (Slattery et al., 2021). Specific guidelines and legislation for EoL LIBs management are lacking (Duarte Castro et al., 2021; Cabral-Neto et al., 2022). Many LIBs present in WEEE are poorly managed, improperly disposed of with MSW, or stored in homes (Duarte Castro et al., 2022). There is a need for more assessments of the impacts of LIBs recycling processes, considering a broad set of environmental impact categories, to avoid damage to the environment and human health (Rajaeifar et al., 2022).

Life Cycle Assessment (LCA) is an efficient technique to analysing potential environmental impacts from a system perspective (Souza et al., 2016; Zhao et al., 2021). LCA has been widely used to assess the environmental implications of waste management systems, including WEEE management systems (Torkayesh et al., 2022; Xavier et al., 2023). LCA allows for assessing the level of impact of the waste management system, from the generation stage to final disposal (Wang et al., 2022). By modelling scenarios in LCA, future implications of waste systems can be assessed, describing the impacts of different system options in advance (Villares et al., 2017; Tsoy et al., 2020). LCA plays a crucial role in supporting the development of a Circular Economy by providing a robust assessment of the impacts of circular solutions and enabling the understanding and minimization of impacts from a holistic view (Ingemarsdotter & Dumont, 2022). However, there are limited investigations of the environmental impacts of recycling LIBs using LCA (Ferrara et al., 2021; Arshad et al., 2022; Rajaeifar et al., 2022). Supplementary material 1 provides a summary of LCA studies on LIBs recycling. In summary analysis, existing studies that evaluate the recycling of LIBs through LCA are more focused on LIBs waste from EV, using pyrometallurgical and hydrometallurgical technologies in the context of China and the USA, lacking LCA studies in developing countries. Besides, data from inventories are lacking, and the studies have limited coverage of emissions and potential environmental impacts of LIBs recycling (Rajaeifar et al., 2022).

Recent LCA studies have been conducted to assess the impacts and benefits of various LIBs recycling processes. For instance, there are studies on hydrometallurgical (Dunn et al., 2022; Cao et al., 2023), pyrometallurgical (Rajaeifar et al., 2021; Koroma et al., 2022), and direct recycling (Jiang et al., 2022). However, these studies focus primarily on the direct impacts of unit processes and avoided impacts of materials recovered by each recycling technology, considering only processes for recycling and avoided production within of the system boundary. There is a notable absence of studies evaluating the EoL of LIBs in WEEE from a systemic perspective. Specifically, there is a lack of assessment regarding the impacts of implementing Reverse Logistics (RL) policies with different collection and recycling routes, as well as the effects of disposal in landfills using a functional unit based on generation of LIBs waste per year. There is a lack of LCA studies focused on the number of LIBs present in WEEE streams and the respective impacts of recycling or disposal. Data from the amount of LIBs waste generated per year in WEEE streams and inventories of LIBs EoL beyond of recycling process are lacking. These gaps in research underscore the need for more comprehensive LCA studies that adopt a holistic approach to assess the environmental impacts of various LIBs recycling and disposal methods.

This article aims to evaluate the potential impacts and environmental benefits (avoided impacts) of different scenarios for the recycling of LIBs, adopting the EoL context of smartphones and laptops. For this purpose, this article estimates the generation of LIBs waste from these WEEE streams in the period of 2022-2026; models a potential LIBs recycling process to be implemented in the country; assesses four progressive scenarios of RL implementation, according to annual targets in policies; identifies environmental hotspots to be tackled by the implementation of LIBs recycling capacities. No study has evaluated the EoL of LIBs in WEEE considering a systemic view of the impacts of implementing Reverse Logistics (RL) policies with different collection and recycling routes, disposal in landfills and using a functional unit based on generation of LIBs waste per year. There is a lack of LCA studies focused on the number of LIBs present in WEEE streams and the respective impacts of recycling or disposal. Data from the amount of LIBs waste generated per year in WEEE streams and inventories of LIBs EoL beyond of recycling process are lacking.

2. MATERIALS AND METHODS

Four macro methodological steps were performed in this study:

1) Scenario modelling to represent the current and future situation of LIBs EoL management in WEEE streams. Data from Brazil is used for building scenarios and exemplifiying methodological steps. Scenario modelling is building according to WEEE RL implementation targets defined by Law (Brasil, 2020, which establishes the WEEE Reverse Logistics Sectorial Agreement). This WEEE RL implementation targets policy is used to capture the amount (in the percentage) of WEEE collection per year in the annual implementation plans to verify the potential amount of collection through formal channels. It is important to note that due to a lack of reliable data, internal logistics (transporting the collection and sending it to the pre-treatment recycling plant in Brazil) was not included in the main assessment, but to address possible transport impacts, intermediate transport from each city included in the RL plan to the pretreatment city was analysed through a sensitivity analysis. WEEE RL implementation was used to capture the amount (rates) collected by RL that could be destined for recycling. In addition, data on landfill distances in Brazil and Europe were also not included in the model due to a lack of data.
2) Estimate of smartphones, laptops and LIBs waste generation for 2022 to 2026 using trend analysis in time series and lifespan distribution;
3) Mapping and modelling of a hydrometallurgical recycling process. The hydrometallurgical process was chosen for three main reasons: i) it is the type of process in which pilot tests are being carried out for implementation in Brazil; ii) the hydrometallurgical process has the potential to generate less environmental impact because it uses less energy and recovers a greater number of materials (Dunn et al., 2022; Jin et al., 2022); iii) hydrometallurgical recycling plants are less complex in terms of costs to installation and processing compared to pyrometallurgical recycling (Gaines et al., 2018; Jin et al., 2022; Chen et al., 2023).
4) Assessment of potential environmental impacts of recycling processes and scenarios using LCA.

2.1 Scenario modelling for LIBs recycling in Brazil

Four progressive scenarios for recycling LIBs in Brazil were modelled in line with the Brazilian National Policy on Solid Waste (Brasil, 2010), which determines the progressive increase in WEEE RL (including LIBs) and the reduction in landfilling. The construction of the scenarios aimed to provide a more in-depth view of the paths that WEEE takes to obtain secondary raw materials, quantifying the impacts and benefits (avoided impacts of raw materials production) of different routes and recovery rates in Brazil. The RL collection rates for the first 3 scenarios were derived from the Implementation Schedule of the WEEE RL Sectoral Agreement (Brasil, 2020), which defines percentage targets for collection and destination for each year: 2023-6%, 2024-12% and 2025-17%. As the collection targets for the year 2026 and subsequent years are not defined in the implementation plan, for scenario 4 (2026), which is considered more optimistic, the collection rate by the LR was increased to 50% of the WEEE generated (laptops and smartphones) to verify the environmental implications of a more efficient collection system. The progressive approach increases RL and decreases LIBs waste collected with MSW and informally and destined for landfills, a common practice in WEEE management in Brazil (Souza, 2020; Xavier et al., 2021). The scenarios present variations in the progressive increase in the collection rate by LR and in the recycling that takes place in Brazil, progressively reducing the sending of materials to landfill and the quantity sent to be recycled in Europe.

Scenario 1 - Pessimistic: reflects the baseline (current) scenario, in which the fraction collected by the RL is lower than that collected informally or mixed with MSW. In this scenario, there are no formal recycling operations for LIBs within Brazil, where recycling activities are limited to disassembly and separation, and the more complex components are exported for recycling abroad, mainly in Europe and Asia (Dias et al., 2018; Duarte Castro et al., 2021). Of the fraction of LIBs collected in smartphones and laptops by formal RL in 2023 (6% according to the Brazilian law) 50% are assumed to be destined for pyrohydrometallurgical recycling abroad (Europe), and the other 50% are destined for hazardous waste landfill in Brazil. Reminder LIBs wastes (94%) are collected informally (50%) or mixed with MSW (50%) and are fully sent to an MSW landfill.

Scenario 2 - Intermediate: the formal RL increases the collection rate to 12% of waste LIBs generated in 2024 (Brazil, 2020), reducing the amount collected informally or discarded with MSW. This scenario assumes that there are LIBs recycling operations in Brazil (hydrometallurgical) in their first steps. 75% of LIBs waste formally collected by RL is sent to recycling abroad, 12.5% to recycling in Brazil and 12.5% to hazardous waste landfill. The assumptions for fractions collected by the informal sector or mixed with MSW are the same as in Scenario 1, with 50% collected by each route and a full destination to the MSW landfill.

Scenario 3 - Optimistic: RL increases the collection to 17% of LIBS waste in 2025 (Brasil, 2020) and the fraction recycled in Brazil increases. 50% of LIBs waste formally collected by the RL is destined for recycling abroad, 25% for recycling in Brazil, and 25% for the hazardous waste landfill. The assumptions are the same for fractions collected by the informal sector or mixed with MSW (50% collected by each channel and full destination to the MSW landfill). Table 1 summarises the rates of collection and recycling for the four scenarios. Although the scenarios designed in this study followed a progressive approach, some assumptions can be considered limitations, such as the amount of waste collected in each collection channel, and the amounts sent outside, can be over or underestimated, due to the lack of consolidated data.

2.2 Estimation of LIBs waste generation from laptops and smartphones

The annual flow of LIBs on smartphones and laptops was estimated based on consolidated historical sales data in Brazil and sales forecasting. For the period from 2012 to 2021, historical sales data provided by the Brazilian Electrical and Electronics Industry Association (ABINEE) (ABINEE, 2022) were used for both types of products (smartphones and laptops). From 2022 to 2025, there were no sales forecast data for these products in Brazil. To fill this gap, sales forecasting methods and projected global sales rates were applied to project sales in this period. Sales forecasting for Notebooks from 2022 to 2025 was performed using projected global sales rates provided by Statista (2022); the annual percentage change was calculated and assumed to project sales for this period in Brazil. Sales forecasts for smartphones from 2022 to 2025 were made using trend analysis in time series (Montgomery et al., 2015). Trend analysis uses linear regression to examine patterns and trends in historical data and predict the future values of a serie (Nokeri, 2022). The use of the global rate of change in sales of laptops for the years 2022 to 2025 was necessary because due to the increase in sales in the period 2020 and 2021 (due to the covid-2019 pandemic) the use of the projection using time series could be overestimated - in this way the use of variations considering the global rate of sales each year was considered more appropriate, however, it can be considered an important limitation, as it can also cause underestimation of the quantity sold in a year.

The estimate of yearly smartphones and laptops wastes generation was calculated using the lifespan distribution of the products. The choice of lifespan distribution instead of average lifespan seeks to model the waste generation system as closely as possible to the reality of consumers. The WEEE lifespan distribution in the Brazilian context was extracted from a previous study (Abbondanza & Souza, 2019). The lifespan distribution for smartphones each year is calculated considering 8 years, being: 1 - 35.90% of smartphones discarded; 2 - 52.65%; 3 - 6.09%; 4 - 2.33%; 5 - 2.43%; 6, 7 and 8 years - 0.20% (Abbondanza & Souza, 2019). For laptops, the lifespan distribution is calculated considering 9 years, being: 1 - 7.0% of laptops discarded; 2 - 14.0%; 3- 25.0%; 4 - 12.0%; 5 - 16.0%; 6 - 9.0%; 7 - 3.0%; 8 and 9 years - 7.0% (Abbondanza & Souza, 2019). Supplementary material 3 provides the calculation details for each projection of the quantity of products sold and the waste generated each year.

The mass of waste generated for each product by year was calculated by multiplying the amount (in units) by the average weight values of smartphones and laptops. The average weight of smartphones was 154.54 g by product unit (Ercan, 2016; Babbitt et al., 2020; Cordella et al., 2021). Notebooks' average weight was 2.711 kg by product unit (Abbondanza & Souza, 2019; Babbitt et al., 2020; Kasulaits et al., 2015).

The estimate of the generation of LIBs waste (in mass) was calculated by multiplying the total mass value of the products by the percentage referring to the mass of the LIBs: 27.3% weight for LIBs in smartphones and 9.34% weight for LIBs in laptops (e.g., Wang et al., 2016; Cordella et al., 2021).

2.3 LCA phases

LCA was applied in accordance with the guidelines of ISO 14040 (ISO, 2006a), ISO 14044 (ISO, 2006b) and best practices indicated in the reference manual of the International Reference Life Cycle Data System - Handbook (ILCD) (EC-JRC, 2010).

2.3.1 Definition of the objective, scope and system boundaries

The objective is to evaluate the potential impacts and environmental benefits related to different and progressive recycling scenarios for LIBs from two types of WEEE products widely used and discarded in worldwide, smartphones and laptops. The geographical scope of the study is Bra-

TABLE 1: Collection and recycling rate for scenarios.

Scenario	Collection rate by RL	Collection rate by informal	Collection rate by MSW	Recycling rate in Brazil	Recycling rate ex-ported to Europe
Scenario 1	6%	47%*	47%*	0	50%
Scenario 2	12%	44%*	44%*	12.5%**	75%
Scenario 3	17%	41.5%*	41.5%*	25%**	50%
Scenario 4	50%	25%*	25%*	75%**	25%

** Full LIBs collected in these routes are sent to MSW landfill; **The difference is sent to hazardous waste landfill.*

zil. The intended application of the study is to identify the hotspots and the main environmental indicators related to waste LIBs recycling in the Brazilian context. The structure used for modelling the inventory is attributional.

The evaluated function of the system is the environmentally sound destination of LIBs waste from the mix of laptops and smartphones waste generated in Brazil. The functional unit to assess unit processes of hydrometallurgical recycling carried out in Brazil and pyrohydrometallurgical processes abroad (Europe) was defined as treating 1 t of LIBs waste in Brazil and abroad. To evaluate scenarios, the functional unit was defined as the treatment of LIBs from the mix of smartphones and laptops discarded in Brazil in 1 year. Supplementary material 4 summarises the function, functional unit, and reference flow of the four scenarios.

The hydrometallurgical process was chosen for Brazil because the hydrometallurgical process is one of the most promising methods for recycling of LIBs waste (Zhou et al., 2021).The hydrometallurgical process is highly effective for all types of battery chemical recovery (Zhang et al., 2022) and has the potential to generate less environmental impact because it uses less energy and recovers a greater number of materials (Dunn et al., 2022; Jin et al., 2022). Hydrometallurgical recycling plants are less complex in terms of costs to install and process compared to pyrometallurgical recycling, which seems more suitable for developing country contexts (Gaines et al., 2018; Jin et al., 2022; Chen et al., 2023). Finally, a company that has been operating the recycling of LIBs in Brazil (Energy Source) uses a hydrometallurgical process (Energy Source, 2022).

The pyrohydrometallurgical process was chosen for Europe because evidence states that this is the type of process most commonly used in recycling plants in European countries (Chen et al., 2019; Mayyas et al., 2019; Yu et al., 2022; Jin et al., 2022). There are well-established LIB recycling plants in Europe that use pyrometallurgy and hydrometallurgy processes together (Mayyas et al., 2019). Pyrometallurgy in conjunction with hydrometallurgy is the dominant recycling method, widely prevalent in European recyclers such as Umicore, Batrec, Accurec, Glencore (Jin et al., 2022).

Multifunctionality is addressed with the substitution approach, including the avoided production of recovered metals and graphite in the system boundary. Assumptions for substitution, such as the amount of each material present in the LIBs waste stream, the recovery efficiency materials in each recycling process, the quality of the recovered material, and the types of processes avoided in the upstream chain were derived from literature (Supplementary material 5 details assumptions for substitution). The substitution was modeled considering open-loop recycling. There are two ways to model the substitution of materials by recycling, Open loop or Closed loop. Closed-loop recycling considers that recycled material can replace the original virgin material and be used in the same type of products. Open-loop recycling, the properties of the recycled material differ from those of the material used in the original product, which is why it is used in other applications of the product, replacing other materials (Larrain et al., 2020). In this LIBs recycling case, recovered metals displace equivalent amounts of raw materials (assumed recycling substitution ratio 1:1) in the respective primary supply chains (mining and ore beneficiation processes). Closed-loop recycling, in which the recovered materials would return to the supply chain to produce new LIBs, cannot be evaluated because of uncertainty about the purity of the recovered metallic salts and their suitability for manufacturing new LIBs (Rajaeifar et al., 2021).

The system is modelled considering the limit of the grave-to-gate system, which includes the steps from the waste entry in the recycling process, recovery of materials and avoided processes (Hao et al., 2017; Bobba et al., 2018; Qiao et al., 2019). LIBs waste is modelled as post-consumer waste, entering the system without environmental charges from the production and use of LIBs (Allacker et al., 2014; Rinne et al., 2021). Due to the lack of data on WEEE collection within Brazil, the transport for collection, sorting and disassembly of smartphones and laptops was excluded from the system boundary. It can be considered a limitation because significant environmental burdens can arise from these steps. Infrastructure is also not considered in the model.

Recycling of LIBs through hydrometallurgy in Brazil begins with the reception of LIBs after the smartphones and laptops products have been disassembled. Then, a resistor discharged LIBs cells to avoid combustion and explosions. Next, three sequential mechanical processes (crushing, shredding and sieving) are used to separate fine fractions rich in metals (black mass) of the medium and coarse fractions, including plastics, current collectors and papers from LIBs. The medium and coarse fractions are sent for final disposal in landfills according to the characteristics of each material. The fine fraction (approximately 50% of the weight of the starting material) is sent to the hydrometallurgical processing, where the black mass undergoes a leaching step with sulfuric acid and glucose. After the leaching, sequential processes separate the graphite and precipitate the metals, which are filtered and oven-dried. The product system and system boundary for recycling technology in Brazil are shown in Figure 1.

The pyrohydrometallurgical process for recycling in Europe is modelled using Ecoinvent inventories and updated data from the literature. The European pyrohydrometallurgical process begins with the national and international transports of sorted cells and the receipt of LIBs at the recycling plant. Next, the cells are heat-treated through a blast furnace casting process. Furnace products consist of alloys and slag that are sent for further treatment. The alloys are sent to a hydrometallurgical process that uses inorganic chemical reagents. Supplementary material S6 provide flowchart the pyrohydrometallurgical recycling in Europe.

Figure 2, Figure 3, Figure 4 and Figure 5 shows flowcharts of scenarios 1 at scenario 4.

2.3.2 Inventory analysis

Data for the recycling process of LIBs in Brazil (hydrometallurgical technology) were collected from the literature and a research institute (Center for Information Technolo-

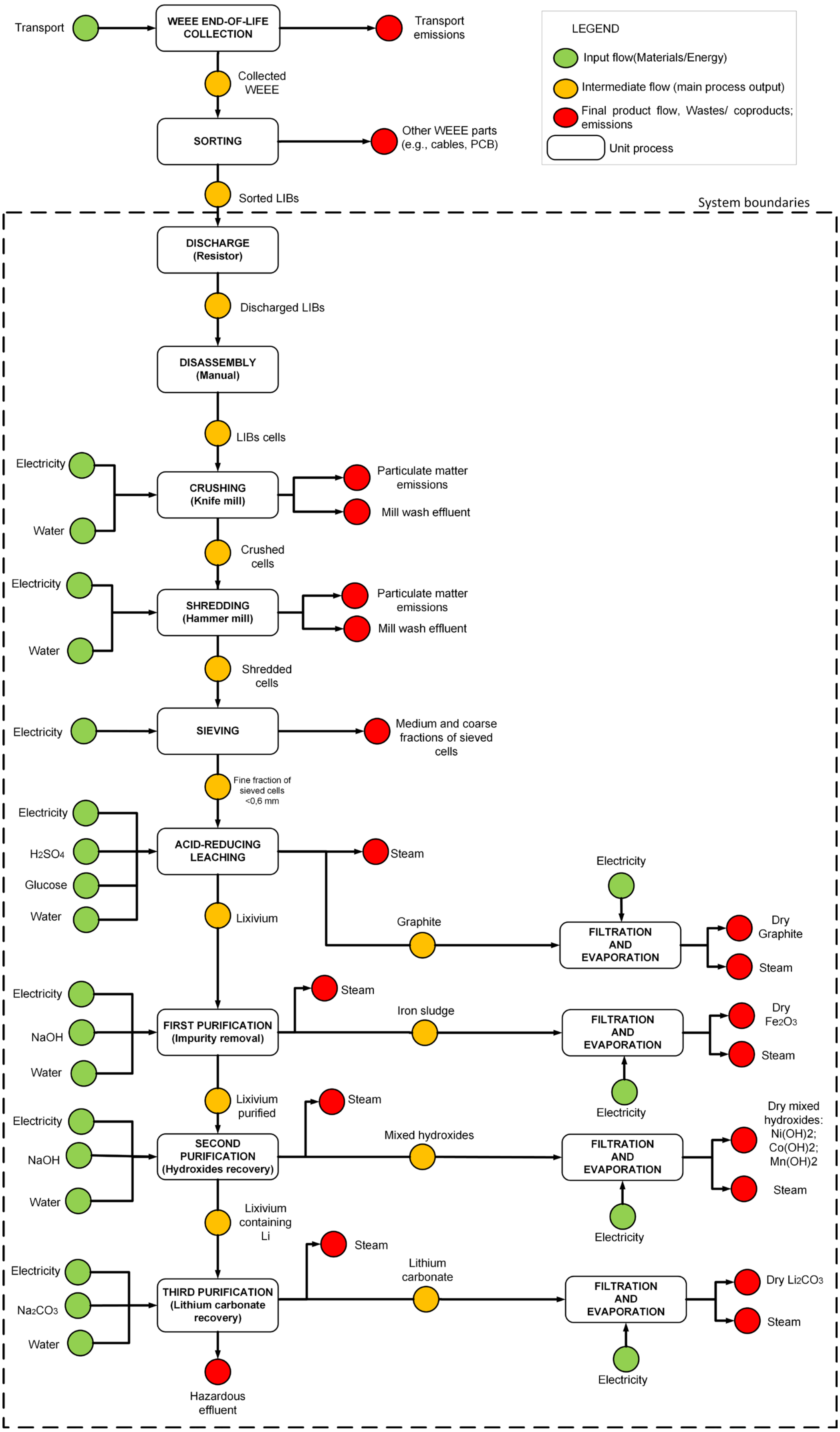

FIGURE 1: Product system and system boundary for hydrometallurgical LIBs recycling in Brazil.

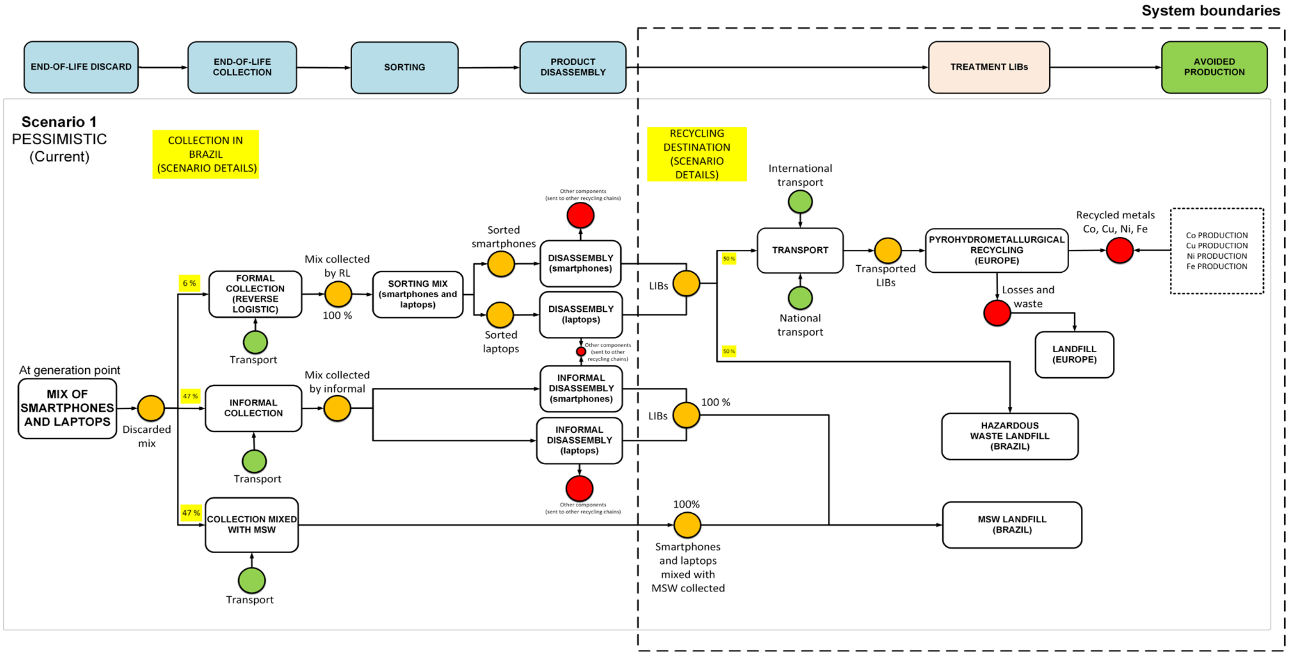

FIGURE 2: Flowchart Scenario 1 for LIBs recycling.

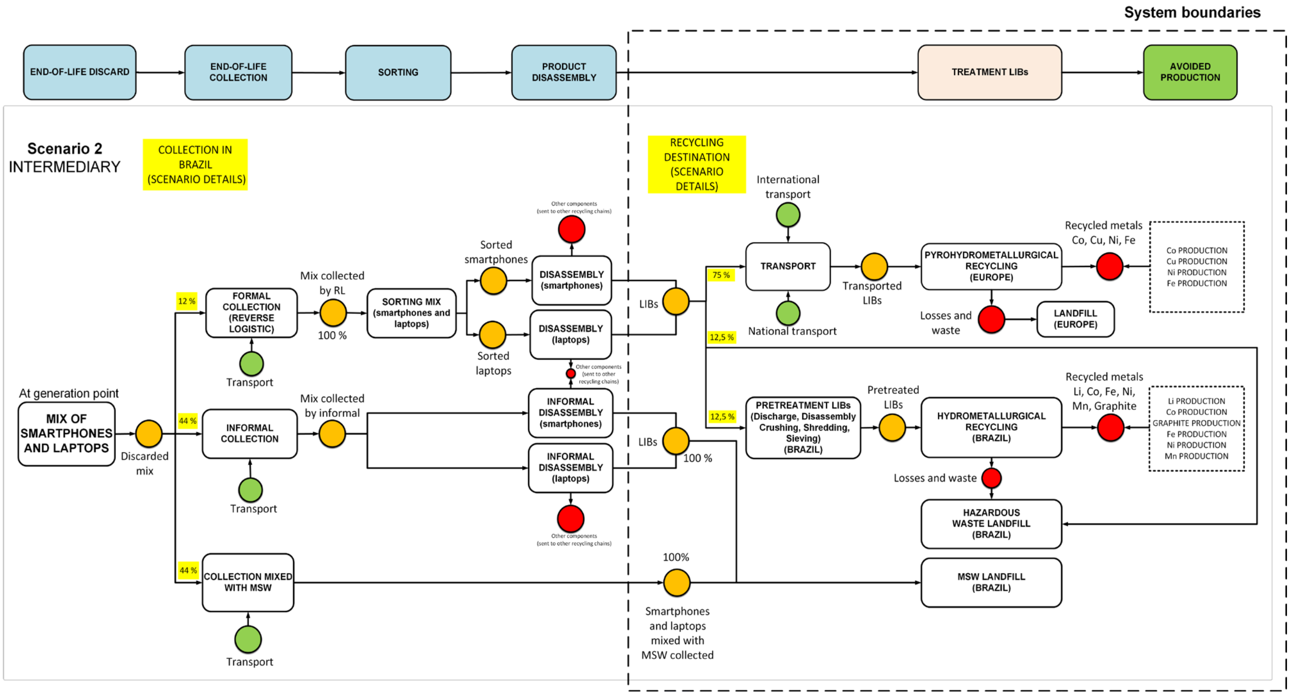

FIGURE 3: Flowchart Scenario 2 for LIBs recycling.

gy Renato Archer - CTI) located in the Campinas//Brazil. This institute provided experimental data and a patent containing the main data on mass and energy flows of unit processes (European Patent EP 2 450 991 B1-2011). The characterisation of the electrolytic powder and recovery rate of metals from leaching using glucose was obtained from the literature (Granata et al., 2012; Pagnanelli et al., 2017), as well as air emissions of fine particulate material from crushing and shredding processes (Costa, 2010). Emissions to water were estimated based on stoichiometric calculations (Supplementary material 7 provides stoichiometric details).

Data for LIBs waste recycling processes abroad were collected from the Ecoinvent v.3.6 database and literature. The process of recycling LIBs abroad was modelled with the processes available in the Ecoinvent database; the available inventories use data from Fisher et al. (2006). The pyrometallurgical process has been edited to include the

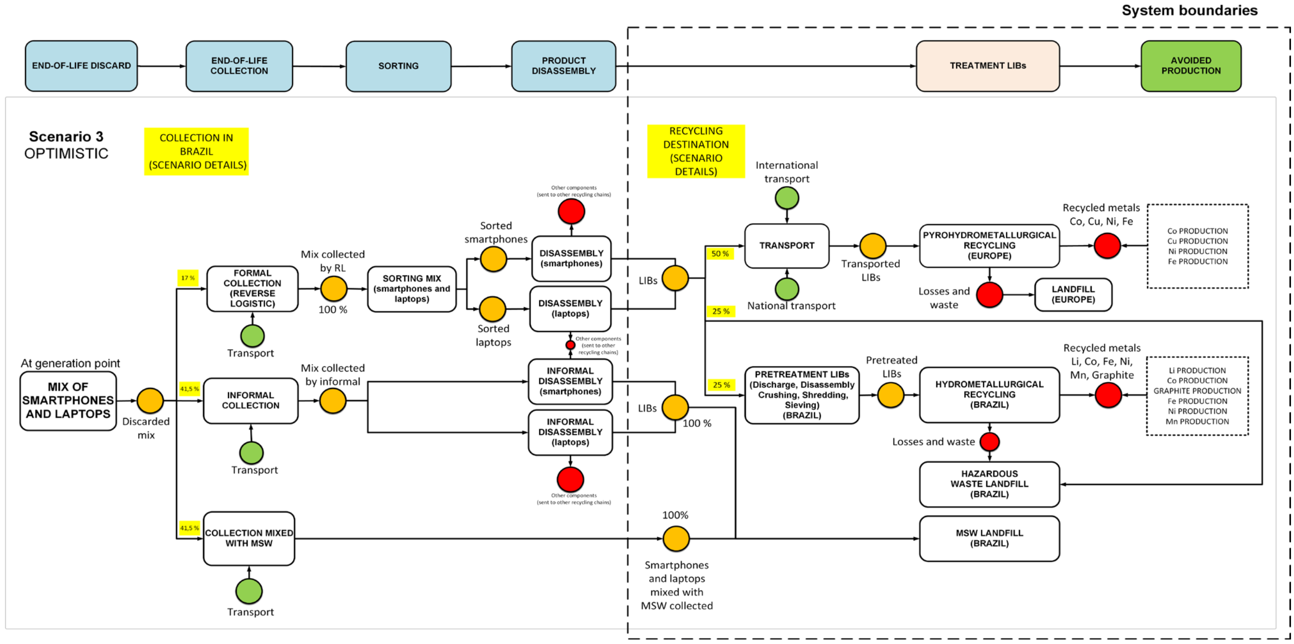

FIGURE 4: Flowchart Scenario 3 for LIBs recycling.

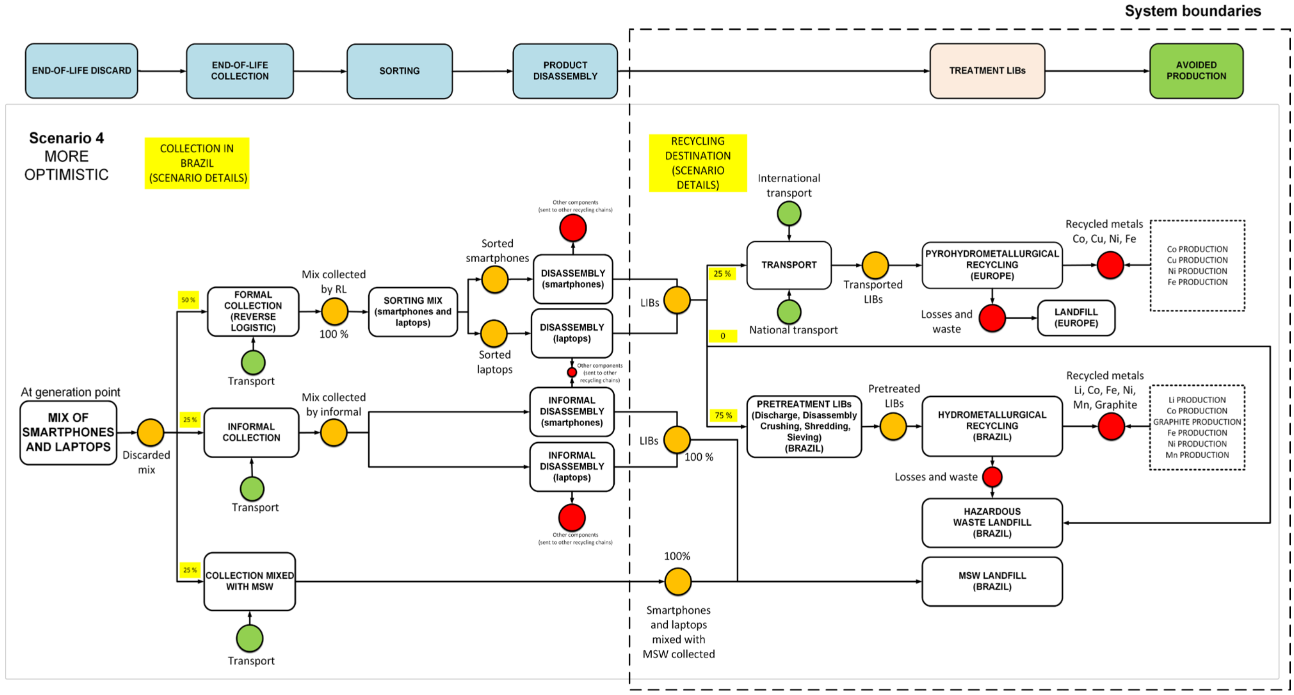

FIGURE 5: Flowchart Scenario 4 for LIBs recycling.

subsequent hydrometallurgy step. In addition, materials recovered, and recovery rates were adapted from Rajaeifar et al. (2021). The adaptation was necessary, as the process available in Ecoinvent is modelled with older data (2006), where manganese, instead of nickel, was recovered, which does not reflect current operations (Harper et al., 2019; Jin et al. al., 2022).

Data on the production of chemical inputs, energy generation, landfilling, water treatment and transport are collected in the Ecoinvent v.3.6 database. It is important to note that medium landfill inventories used within the system boundary of each recycling process to model the shipment of fractions that are destined for final disposal, such as plastics and other materials, to both Brazil and Europe. However, due to the lack of specific local landfill data, the landfills used may not be representative enough to capture the differences between Brazilian and European landfills, which is a limitation of this study.

International transport distances (maritime transport) are estimated using geospatial tools available on the internet (9.361 km – distance from the port of Santos – Brazil for the port of Le Havre - France) (Google Maps, 2022). The average road transport distance from cities to the main port in Brazil (200 km) is collected from the literature (Rocha & Penteado, 2021). This average transport distance considers a pretreatment centre in the state of São Paulo to the Port of Santos. This distance was considered due to the scope of the study considering the transport from the consolidation of WEEE collected in Brazil. Supplementary material 8 provides the life cycle inventory normalised to 1 t and sources data types to hydrometallurgical recycling in Brazil and pyrohydrometallurgical recycling in Europe.

SimaPro Faculty software v. 9.1.1.1 was used for modelling and assessing environmental impacts.

LIBs landfill limitations

Regarding the landfill inventory used to model the disposal of LIBs in the scenarios, one limitation should be considered the use of an average landfill inventory available on Ecoinvent for the disposal of LIBs, which may not adequately reflect the direct emissions of LIBs. Although this strategy of using average landfills is widely used in the literature, there are clear limitations on the representation of specific wastes. To try to address the uncertainty involved in this limitation, we followed guidelines from the literature (Doka, 2023) and inventoried the substances at the elemental level of each constituent material of the LIBs. This is a suitable strategy to make transparent the limitations of landfill modelling for specific wastes where no data is available (Doka, 2023). The inventory at the elemental level shows that LIBs have the potential to emit different substances, such as CH_4, CO_2 and hydrofluorocarbon (HFC), due to the presence of hydrogen, fluorine, and carbon in LIB materials. In addition, the amount of different metals present in LIBs can pose risks of potential impacts, mainly related to toxicity.

As LIBs have a complex composition, with different materials that can interact in different ways in landfill environments (Kilgo et al., 2022), it was not possible to model all the possible emissions that can occur. However, we performed a simulation using parameters from some recent studies that determine the daily emission of CH_4, CO_2 and C_2H_4 from discarded PP, PE and PET plastics due to photodegradation (Royer et al., 2018; Shaw et al., 2023) to model possible emissions of these gases from these plastic fractions that are present in LIBs sent to landfill. Following guidelines from the literature on landfill modelling, we considered a time horizon of 100 years (Kirkeby et al., 2007). In addition, we calculated the potential for metals to be leached from the fraction of LIBs sent to landfill by considering parameters from a study that carried out laboratory tests on metal leaching from LIBs under landfill conditions (Kilgo et al., 2022). The results of this landfill-specific assessment were not considered in the evaluation of the results but were used to compare the sensitivity of how possible emissions may change due to waste specific modelling. The inventory at the elemental level and the details for the calculations of possible emissions can be found in supplementary material S16.

2.3.3 Impact assessment

The assessment of potential environmental impacts was carried out using the impact assessment method ILCD 2011 Midpoint + version 1.0.9 (EC-JRC, 2012), including the 16 impact categories: Climate change (kgCO_2eq), Ozone depletion (kgCFC-11 eq), Human toxicity – non-cancer effects (CTUh), Human toxicity-cancer effects (CTUh), Particulate matter (kgPM2.5eq), Ionising radiation HH (kBqU235eq), Ionising radiation E -interim (CTUe), Photochemical ozone formation (kgNMVOCeq), Acidification (molcH+eq), Terrestrial eutrophication (molcNeq), Freshwater eutrophication (kgPeq), Marine eutrophication (kgNeq), Freshwater ecotoxicity (CTUe), Land Use (kgCdeficit), Water resource depletion (m3 water eq), Mineral, fossil & renewable resources (kgSbeq). The midpoint level assessment was chosen so that the cause-and-effect chains of environmental impacts could be better understood (EC-JRC, 2010). The assessment of impacts at the midpoint level has fewer uncertainties associated with environmental mechanisms when compared to the endpoint analysis (Huijbregts et al., 2016; Mulya et al., 2022).

2.3.4 Interpretation

The interpretation phase presents a critical evaluation of the results, drawing conclusions and recommendations on the main points of environmental impacts per process, the benefits arising from the avoided production, and the identification of the best and worst scenarios for managing the LIBs waste generated in Brazil. Possible alternatives for improving the environmental profile are recommended, and a sensitivity analysis was conducted.

Sensitivity analysis

A perturbation analysis was performed to assess the robustness of the results: first, one perturbation of one input at a time and then a global perturbation analysis, with all inputs being varied together (Laurent et al., 2020). A range of -/+5% and -/+ 10% were used for the evaluation. A second sensitivity analysis was conducted to assess how sensitive the results are to the recycling efficiency ratio. The recycling efficiency rate is an important parameter, as different technologies, input material quality and subsequent use of recovered materials interfere with the result of the recycling system (Rocha & Penteado, 2021). A Third sensitivity analysis of the results was performed using another impact assessment method, the Recipe Midpoint H (Huijbregts et al., 2016), to verify whether the results could change due to the impact assessment method.

Due to the lack of complete data on the stages of RL, such as transport distances for collection and to dismantling and recycling facilities within Brazil, a simplified RL model that only considers transport from consolidation sites in different cities, after collection, to pre-treatment in São Paulo was tested to check the sensitivity in evaluating the scenarios due to the incorporation of intermediate transport in the model. The calculation took into account the coverage of cities served each year as defined in the sector agreement, the population of the cities and the distances to São Paulo. As the WEEE RL implementation targets defined in the Law (Brazil, 2020) establish the gradual implementation of RL in the states, but do not provide the specific cities for each year, only the number of cities served each year, the cities were added to the RL according to population, with the most populous cities being served first until a total of 400 cities were served. The cities with the most inhabitants are served first until the 400 cities are reached in 2025. For 2026, as there was no data on the cities served, the same number of cities was assumed as in 2025 (400 cities). Point-to-point collection transport within the cities was not included due to the lack of data on the average distances travelled in each of the cities. Therefore, what is included in the calculations is the intermediate transport, after collection, from each generating city to the pre-treatment centre (here considered to be the city of São Paulo). All the data and details of the calculations are provided in supplementary material S15.

3. RESULTS AND DISCUSSION

This section presents the results and discussion of the assessment of unit processes and scenarios, and potential revenues of LIBs hydrometallurgical recycling in Brazil.

3.1 Contribution analysis of recycling technologies

Figure 6 presents the results of the contribution analysis for the overall and unit process of hydrometallurgical recycling in Brazil. Results are presented for a functional unit of 1 t of LIBs waste treated. Results are detailed for the supply of key inputs and direct impacts on processes. Direct impact refers to emissions released directly by a unit process to the foreground system (EC-JRC, 2010), for example, particulate matter emissions from crushing.

The overall contribution analysis for the characterised impacts of the hydrometallurgical technology shows that the main contributors to the impacts are the leaching and material recovery (UP 4) and sieving (UP 3) unit processes. The UP 4, referring to the leaching, precipitation and final recovery of metals, is the main contributor to 13 of the 16 impact categories evaluated (Climate Change, Ozone depletion, Human Toxicity - cancer effects, Ionising radiation HH, Ionising radiation E (interim), Photochemical ozone formation, Acidification, Terrestrial eutrophication, Freshwater eutrophication, Marine eutrophication, Land Use, Water resource depletion, Mineral, fossil & ren resource depletion). The UP 3 is the main contributor to 2 categories (Human Toxicity – non-cancer, Freshwater ecotoxicity). Due to direct process emissions, crushing (UP 1) and shredding (UP 2) unit processes are the main contributors to the Particulate Matter category. The leaching and recovery of materials in UP 4 is the step that consumes a larger amount of chemicals and glucose, with significant impacts on its production. The production of sulfuric acid emits sulphur dioxide and nitrogen oxide, contributing to terrestrial acidification, ozone formation and particulate matter emissions. Using glucose as a reducing agent implies emissions of ammonia, nitrate and nitrous oxide derived from maise cultivation to produce maise starch. These substances contribute to terrestrial acidification, marine eutrophication and stratospheric ozone depletion. Sending materials such as plastics and ferrous metals for final disposal in landfills contributes mainly to freshwater ecotoxicity, Marine eutrophication, Human toxicity- non cancer and Climate change categories due to landfill emissions. The use of sodium carbonate, sulfuric acid and glucose contributes to the reduction of mineral resources. CO_2, methane and nitrous oxide emissions from electricity production, maise production and direct process emissions are the main contributors to the Climate change category. These results align with previous studies that found that the impacts of the hydrometallurgical process are mainly derived from indirect emissions from the production of chemical inputs and electricity (Cusenza et al., 2019; Mohr et al., 2020; Rinne et al., 2021). For the unitary process UP 1, the production of the electricity mix used for crushing is the largest contributor in 14 of the 16 categories. Water production, used to wash away particles that adhere to the mill surface, is the second largest contributor to 12 of the 16 impact categories. For the Particulate matter category, the crushing process is the largest contributor, with 100% of the impact derived from direct emissions of fine particles (<2.5 microns). Water production impacts are mainly for the Mineral, fossil & renewable resources category, due to the use of copper and aluminium sulphate in water treatment at the municipal plant. The water production process provides credits towards the Water resource depletion category due to the return of unpolluted water to the environment during water treatment.

The profile is similar to the previous process for the shredding process (UP 2). Electricity consumption for hammer mill operation is the main contributor to impacts in 15 out of 16 categories. The total Particulate matter category emission is the direct emissions of fine particulates <2.5 microns from the process. The impacts of the crushing and shredding process are mainly derived from the auxiliary systems, which supply energy and water to the system. The control of Particulate matter emissions is a priority for developing the industrial recycling plants for LIBs. Using mechanisms that filter particulate emissions and equipment with increased energy efficiency and higher t/h yield should improve the environmental profile of the crushing operation.

The sieving process contribution analysis (UP 3) shows that the destination of the medium and coarse fractions to the landfill (paper, plastics, and metals of current collectors) is mainly responsible for the impacts in the 16 impact categories. Recovery of medium and coarse fraction materials, such as plastics of casing and metals of current collectors (steel, copper, aluminium) can reduce the environmental impacts associated with the sieving process in the future, as it will reduce the amount of material sent to landfills and production of virgin materials (Hua et al., 2020; Sun et al., 2020; Mohr et al., 2020, Windisch-Kern et al., 2022). For example, recycling of aluminium from waste LIBs has the potential to reduce emissions of 0.03 kg of C_2H_4 eq and 41.6 kg of 1,4-DCB equivalent, avoiding impacts on the Photochemical oxidant formation e Human toxicity categories (Sun et al., 2020). Furthermore, aluminium and copper are easily recovered in the pretreatment and do not require further hydrometallurgical processes (Cao et al., 2023).

The contribution analysis for the inputs of the leaching and material recovery process (UP 4) shows that the main contributors are the production of chemical inputs: sulfuric acid, sodium hydroxide, sodium carbonate and glucose. Chemical inputs were also the main contributors to impacts in the literature, mainly sulfuric acid and sodium hydroxide (Rinne et al., 2021). The use of glucose reduces the general impacts on the Climate change categories, as in the cultivation of maise for production of maise, there is CO_2 sequestration. However, using glucose increases direct CO_2 emissions from the leaching process, as glucose breaks down into water and CO_2 by oxidation. The oxidation of glucose into CO_2 is responsible for 22.5% of CO_2 emissions from hydrometallurgy. Although CO_2 emitted from glucose is considered biogenic, in this study it was treated with a characterisation factor of 1 in the emissions count, since there is still no consensus among scientists on the neutrality of biogenic CO_2 (Cusenza et al., 2021). Glucose has a negative impact reduction in the Human Toxicity non-cancer category, as maise cultivation also captures metals, such as zinc and copper, from the soil, providing an environmental credit. Regarding the credits provides from absorption of metals in crop of maise, it is important to point out that the ILCD method does not include the consumption of these metals within the system boundaries and so the plants serve as a sink (Nessi et al., 2021). The fate of heavy metal emissions taken up by crops are not further considered within the system boundary is a limitation of the potential environmental impacts and should be viewed with caution. Supplementary material 9 provides detailed numerical results of a contribution analysis for the hydrometallurgical technology.

The contribution analysis of the impacts of the modelled pyrohydrometallurgical process for recycling in Europe shows that the impacts of the pyrohydrometallurgical process derive mainly from indirect emissions from producing electricity and chemical inputs and from direct emissions from burning materials in the smelting stage. The main emissions are CO_2, methane, nitrous oxide, sulphur dioxide and heavy metals. The pyrometallurgical smelting process generates many greenhouse gas emissions due to the combustion of materials. The water consumption category is also significant, as both the pyrometallurgical and hydrometallurgical processes consume high volumes of water. It induces the highest amount of water consumption because it uses limestone, which requires a large amount of indirect water consumption (Yu et al., 2021). Transport, road and maritime contribute up to 30% of the impacts for the Photochemical ozone formation and Terrestrial eutrophication categories and 20% for Marine eutrophication and Acidification. Detailed results for contribution analysis of pyrohydrometallurgical recycling to Europe are presented in the supplementary material 10. Additionally, it is important to point out that the energy mix used to evaluate the process in Europe refers to a European mix that considers a proportion of energy generated by different European countries, therefore a comparison with an energy mix from a specific country, such as France, which has an energy matrix with more nuclear sources can change the result of the amount of emissions derived from energy.

A direct comparison of the categories for the two technologies is presented in Figure 7. Recycling 1t of LIBs with the pyrohydrometallurgical option of the Europe has greater environmental impacts in 12 categories. For example, for the Climate Change category, the external option emits 1240.00 kgCO_2eq/ton of recycled LIBs, which is 143% more impactful than the hydrometallurgical technology in Brazil, emits 510.1 kgCO_2eq/ton of recycled LIBs. This result is explained by the fact that recycling with pyrohydrometallurgical technologies consumes more resources to be performed, mainly energy. Electricity production emits large amounts of CO_2 and methane, the main greenhouse gases. The difference can also be explained by the fact that the Brazilian energy mix is composed of more renewable energy than the global average (Lima et al., 2019). Contributions to the Human toxicity – non-cancer category are derived from zinc, arsenic and lead emissions from the treatment of waste coal used in European electricity production. Recycling in Brazil has greater impacts in 4 categories (Particulate matter, Acidification, Land Use, and Water resource depletion). For the Particulate matter, Acidification, and Water resource depletion categories, the greatest impacts are derived from using sulfuric acid and the direct emission of particulates from crushing and shredding processes. The production of sulfuric acid consumes large amounts of water and emits substances that contribute to terrestrial acidification and the formation of particulates, such as SO_2 and NOx. For the Land use category, maise cultivation for glucose production is the main cause of impacts.

3.1.1 Avoided impacts of material recovery

The results shown that the greatest environmental benefits (avoided impacts) are derived from the avoided production of Co and Ni. The recovery of Co and Ni is responsible for more than 93% of the avoided impacts, mainly in the categories of Climate Change, Freshwater ecotoxicity and Land Use. The production of Co and Ni is intensive in energy resources and CO_2 emissions (Chen et al., 2023). The production of Cobalt emits 8.8 kg of CO_2 eq/kg and Nickel production 13.2 kg of CO_2 eq/kg. In addition, they occupy large areas of soil and release toxic pollutants into the water. Nickel production incurs high toxicity impacts, and its recovery avoids them (Cao et al., 2023). Although Graphite recovered is significant per ton of LIBs recycled (252 kg/ton of LIBs waste in this study), the recovery benefit is small compared to other metals because the considered process of mining natural graphite is less energy and emissions intensive than metals, around 0.037 kgCO_2eq/kg of natural graphite production. Comparatively, even though 44% more graphite is recovered by mass than cobalt, the avoided impact of graphite is less than 1% of the avoided impact of cobalt. Greater benefits would be captured if the quality of the recovered graphite was battery grade. A recent study shows that battery-grade graphite production emits 9.66 kg of CO_2eq/kg of battery-grade graphite produced (Engels et al., 2022), but further studies on additional treatments

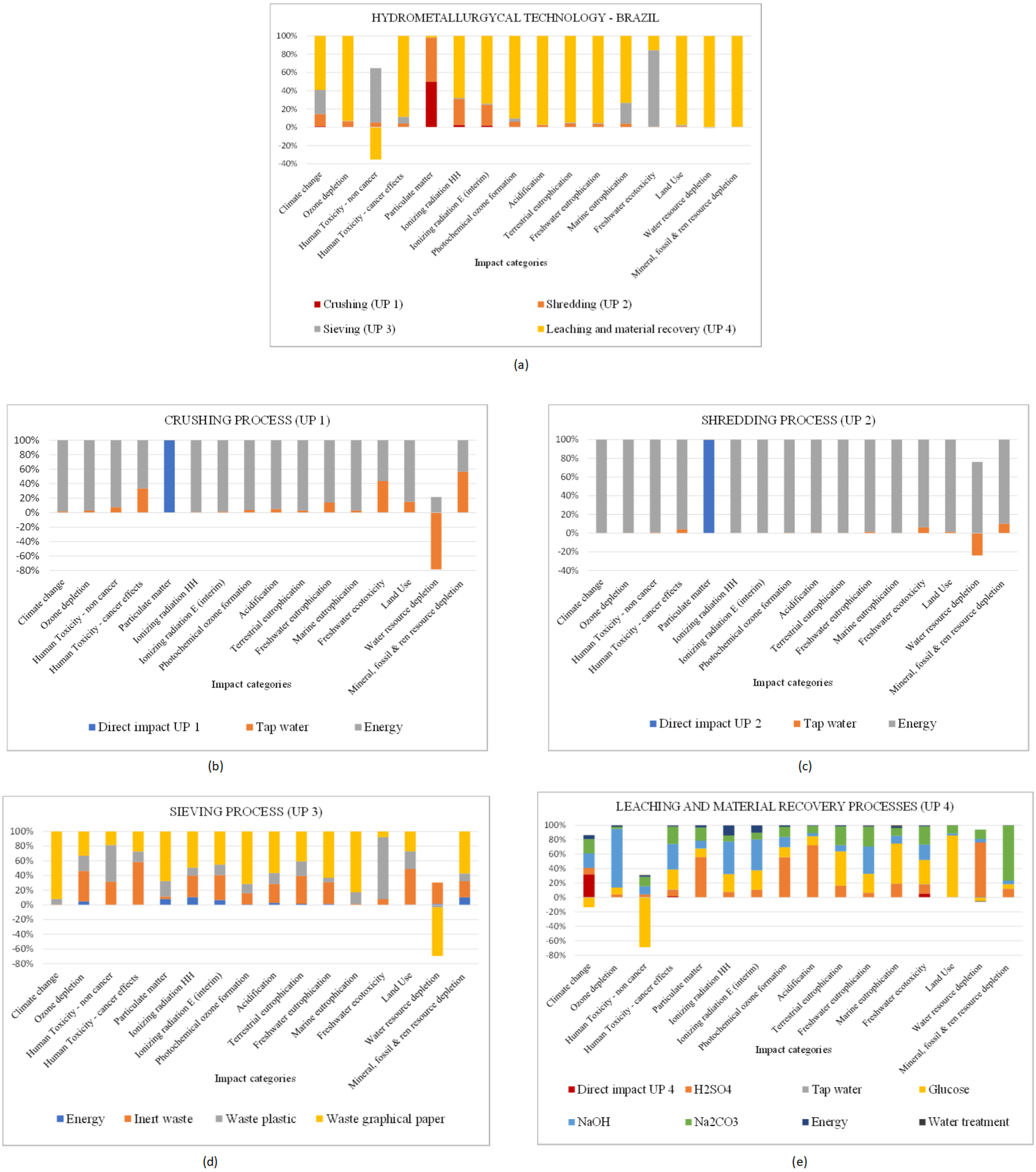

FIGURE 6: Contribution analysis of the unit processes of LIBs hydrometallurgical recycling in Brazil (and supply of key inputs). Note: (a) Overall contribution analysis of the hydrometallurgical technology – Brazil; (b) Contribution analysis of the crushing process (UP 1); (c) Contribution analysis of the shredding process (UP 2); (d) Contribution analysis of the sieving process (UP 3); (e) Contribution analysis of the leaching and material recovery process (UP 4).

are needed to verify the actual benefit. The detailed results for avoided impacts of material recovery to all impact categories are shown in Supplementary material 11.

3.1.2 Comparison with other LCAs

Direct comparison of LCA results from LIBs is challenging, as modelling choices, such as functional unit, system boundary, types of the treatment process, chemical inputs, number of evaluated impact categories and recovered materials interfere with study results and cannot provide a basis for direct comparison (Arshad et al., 2022). However, in order to provide a comparison of the modelled hydrometallurgical recycling process, Table 2 presents a general comparison with other LCA studies of the recycling of LIBs

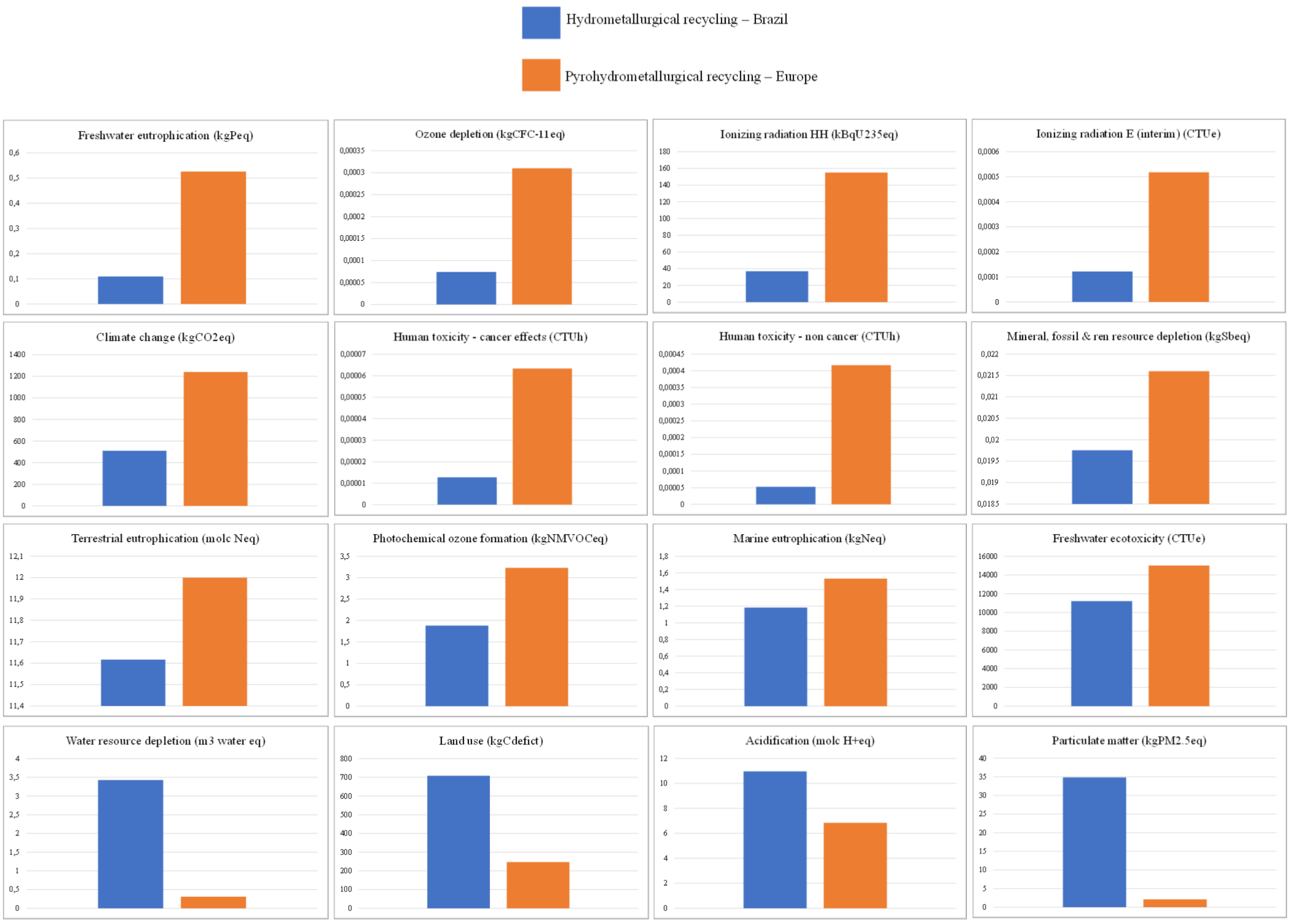

FIGURE 7: Comparison between recycling of 1t of LIBs waste with hydrometallurgical process in Brazil and pyrohydrometallurgical in Europe.

in the Global Warming category (kg of CO_2 eq), which is the most evaluated category in studies of recycling on LIBs (Rajaeifar et al., 2022). Analysing the normalised results for the treatment of 1 kg and 1 ton of LIBs, the impacts of the hydrometallurgical treatment proposed for implementation in Brazil have less impact in the Global Warming category than all other LCAs analysed.

3.2 Scenario assessment

The estimation of LIBs waste generation indicates that more than 3.000.00 tons of LIBs waste can be discarded each year (from 2023 to 2026) in Brazil's smartphones and laptops waste streams. Considering that these two WEEE products have a wide market penetration worldwide and a short lifespan, this result confirms the great potential of LIBs waste generation and resource recovery in the WEEE streams in all countries. Supplementary material 3 provides the results of estimating the generation of LIBs waste.

The results of the environmental assessment of the four scenarios with the functional unit of annual LIBs waste generation are summarised in Figure 8 and Figure 9. Scenario 1 shows the worst performance due to greater disposal of LIBs waste in landfills (2.925.00 t), lower collection rate by RL (6%) and recycling in Europe, which is more impactful. Scenario 4 presents the best performance due to a reduction of 44% in the disposal of LIBs waste in landfills (compared to the baseline scenario), an increase in RL collection rate to 50% and 75% of LIBs waste recycled in Brazil, using the technology less impactful. The results indicate that RL progression and landfill rate reduction significantly improve the environmental profile of LIBs management in Brazil. In previous evaluations of scenarios for waste management, scenarios in which the largest share of waste is destined for landfills result in greater environmental impacts (Turner et al., 2016; Souza et al., 2016; Coelho & Lange, 2018). Landfilling mainly contributes to large greenhouse gas emissions and has a greater impact on global warming (Turner et al., 2016; Jaafarzadeh et al., 2021). To detailed results of each scenario, see supplementary material 12. These landfill results should be interpreted with caution due to the use of an medium landfill to calculate the potential impacts of the fraction of LIBs destined for landfill. However, the analysis of the inventory at the elemental level of LIBs, the modelling of CO_2 and CH_4 emissions from the plastic fraction of LIBs shows that LIBs have the potential for GHG emissions directly through plastics but also through other interactions, such as the formation of Hydrofluorocarbon (HFC), since hydrogen, fluorine, and carbon are present in the composition of LIBs.

TABLE 2: Comparison with Other LCAs.

Wet methods			
Authors	**Treatment type**	**kg CO_2eq / kg of battery**	**kg CO_2eq / ton of battery**
This study	Hydrometallurgical	0.51	510.00
Souza et al. (2022)	Hydrometallurgical	1.13 to 1.99	1130.00 to 1990.00
Dunn et al. (2022)	Hydrometallurgical	1.8	1800.00
Xu et al. (2021)	Hydrometallurgical	2.27	2270.00
Xu et al. (2020)	Hydrometallurgical	2.3	2300.00
Jiang et al. (2022)	Hydrometallurgical	2.7	2700.00
Rinne et al. (2021)	Hydrometallurgical	3.8	3800.00
Yu et al. (2021)	Hydrometallurgical	10	10000.00
Dry methods			
Authors	**Treatment type**	**kg CO_2eq / kg of battery**	**kg CO_2eq / ton of battery**
This study	Pyrometallurgical	1.24	1240.00
Rajaeifar et al. (2021)	Pyrometallurgical	1.4	1400.00
Dunn et al. (2022)	Pyrometallurgical	2.0	2000.00
Xu et al. (2021)	Pyrometallurgical	2.16	2160.00
Xu et al. (2020)	Pyrometallurgical	2.4	2400.00
Yu et al. (2021)	Pyrometallurgical	10.9	10900.00

In addition, the metal leaching potential of LIBs can also contribute significantly to the toxicity categories. The comparison between the results of the fraction sent to landfill using the limited simulated emissions data shows that modelling the sending of LIBs with limited data is not adequate, as several emissions could not be accounted for, as well as the destination of such emissions. Figure S1 in supplementary material S16 shows that compared to the medium landfill used with the limited LIBs landfill that was simulated, the results for the climate change category is much lower than the medium landfill. In contrast, for the Freshwater ecotoxicity and Human toxicity - cancer effects categories, the amount of metals leached from the LIBs causes much more impact compared to the medium landfill. Therefore, the landfill results in the evaluation of LIB waste management scenarios should be interpreted with caution, since not all the potential toxicity or GHG emissions can be modelled, and the average landfill does not capture all the potential toxicity from metals. Further studies into the disposal of LIBs in landfill is urgent.

The results of net environmental impacts indicate that recycling more LIBs waste by Brazil's hydrometallurgical technology results in net environmental benefits across 13 impact categories. For example, in Climate change, scenario 4 presents net environmental benefits of -183.355.00 kgCO_2eq. In contrast, scenarios 1, 2 and 3 do not present net benefits because recycling credits do not outweigh the burdens derived from the greater amount of LIBs waste sent to landfills and a higher recycling rate with pyrohydrometallurgical technology.

The impact categories in which scenario 4 does not present the best net impact results, that is, the material recovery credits do not exceed the total impacts of the scenario, are Ozone depletion, Particulate matter and Water resource depletion categories. This result is explained by the increase in the consumption of inputs such as NaOH, glucose, Na_2CO_3, limestone and particulate matter emissions due to the increase in the number of waste LIBs that are sent for recycling. Therefore, improvements in particulate matter emissions in the crushing and shredding processes and the increase in yield of the leaching processes and material recovery can help reduce the overall impacts and result in net environmental benefits (avoided impacts) far superior to the impacts generated by recycling.

In summary, the results indicate that it is beneficial for the environment to recycle LIBs in Brazil, avoiding flows to landfills. Suppose additional improvements are made to the hydrometallurgical process, such as improving recycling efficiency, reducing the use of chemicals, controlling particulate matter, and using glucose sources from agro-industrial waste, such as molasses or whey (Pagnanelli et al., 2017). In that case, the environmental profile of recycling can significantly improve.

3.3 Sensitivity analysis

The sensitivity analysis considering a variation in inputs in the hydrometallurgical process, both individually and globally, indicates that a variation of 5% to 10% in the inputs results in insignificant changes in the results. The sensitivity analysis to the recycling efficiency ratio indicated that it would take a reduction of more than 70% in material recovery rates or in the replacement rate of all materials for the impacts of the recycling system to outweigh the benefits. The results of this sensitivity analysis indicate that for LIBs recycling systems that allow high efficiency and purity of metals is a critical factor in achieving net benefits. Previous studies have also highlighted that the yield of metal recovery and the consumption of chemical inputs are sensitive parameters for the environmental and economic viability of recycling LIBs (Cao et al., 2023). The nu-

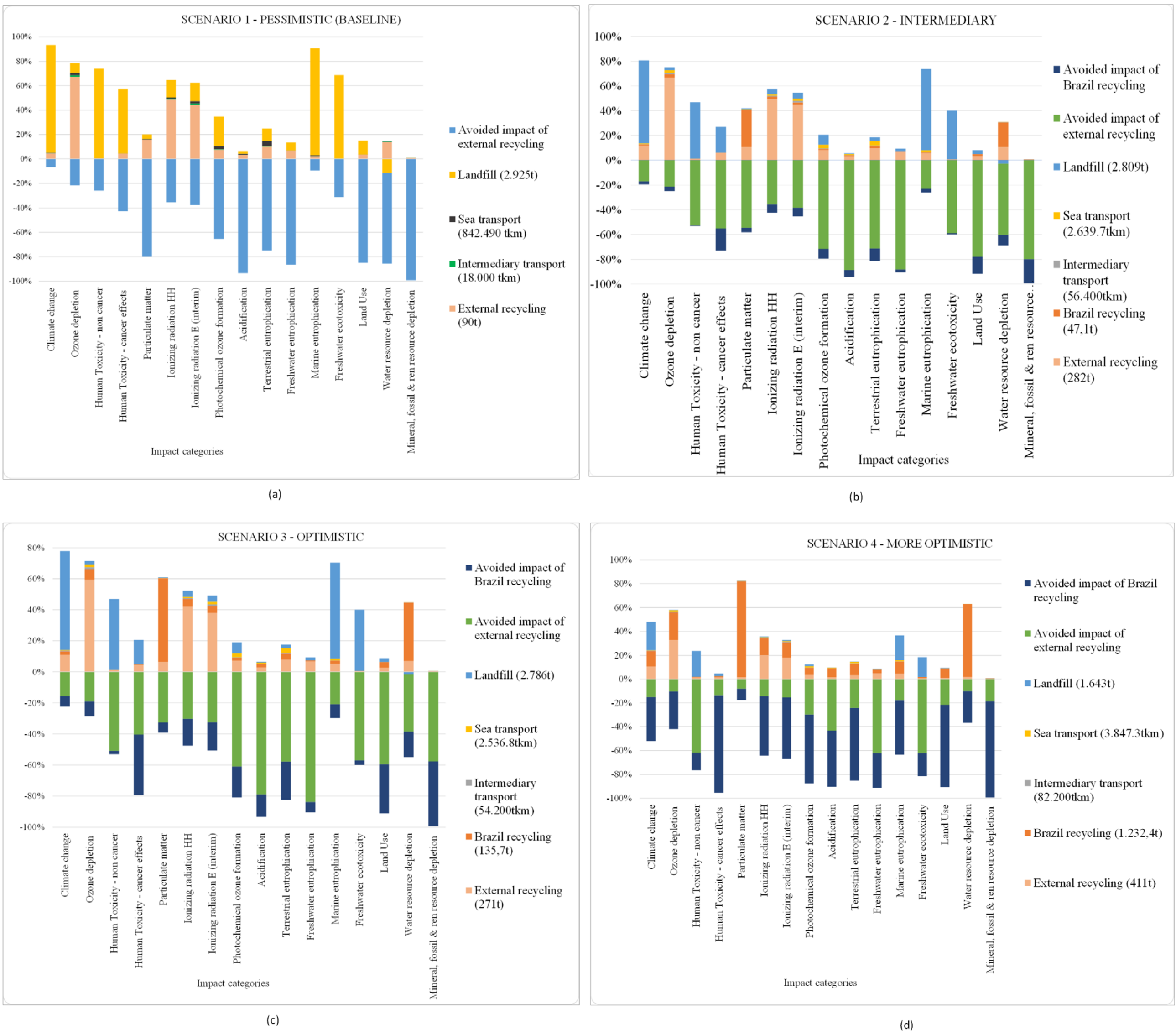

FIGURE 8: Scenarios assessment based on the amount of LIBs waste generated per year. Note: Scenarios assumptions: (a) Scenario 1 - Pessimistic (baseline): Collection: 6% collected by RL and 94% collected by informal and MSW / Recycling: 100% collected by RL sent for pyrohydrometallurgical recycling in Europe; (b) Scenario 2 - Intermediate: Collection: 12% collected by RL and 88% collected by informal and MSW/Recycling: 75% collected by RL sent for pyrohydrometallurgical recycling in Europe and 12,5% sent for hydrometallurgical recycling in Brazil; (c) Scenario 3 - Optimistic: Collection: 17% collected by RL and 83% collected by informal and MSW /Recycling: 50% collected by RL sent for pyrohydrometallurgical recycling in Europe and 25% sent for hydrometallurgical recycling in Brazil; (d) Scenario 4 - More Optimistic: Collection: 50% collected by RL and 50% collected by informal and MSW/Recycling: 25% collected by RL sent for pyrohydrometallurgical recycling in Europe and 75% sent for hydrometallurgical recycling in Brazil.

merical results of these sensitivity analyses can be found in Supplementary material 13.

Figure 10 shows the results of sensitivity analysis using another impact assessment method (ReCiPe midpoint H). Net performance analysis of the scenarios in the six comparable categories revealed that there are significant differences in the results. In this study, the difference is seen mainly in the Global Warming category due to the use of glucose. In the Recipe 2016 method, maise cultivation causes additional impacts on CO_2 emissions. Unlike the ILCD 2011 method, maise cultivation has negative impacts because it sequesters CO_2 from the atmosphere. In the Recipe method, carbon sequestration is not included. Therefore, depending on the method used, the results of the recycling process may differ slightly. Other studies that compare the Recipe 2016 midpoint and ILCD 2011 midpoint + also observed differences in results, mainly in the Global warming category (Borghesi et al., 2022). For numerical results of the comparison in each method, see Supplementary material 14.

As the methods have different measurement units for some categories, it is impossible to directly compare the results for all categories. It was only possible to directly compare the results for 6 categories in which the measurement units were the same in both methods: Climate Change ($kgCO_2eq$), Ozone Depletion (kgCFC-11eq), Marine Eutrophication (kgNeq), Freshwater Eutrophication (kgPeq), Particulate Matter (kgPM2.5 eq), Water Depletion

FIGURE 9: Numerical results of total impact, total avoided impact and total net impact for each scenario.

(m3). The net results of the scenarios, that is, the total impact of the scenario minus the total recycling credits in the two methods, are comparable in the Freshwater Eutrophication (kgPeq) category, presenting practically matching results. However, in the other 5 categories, there were discrepancies in the net result of the scenario. For example, for the Ozone Depletion category (kgCFC-11eq), the Recipe method has a negative net benefit, while the impacts of the ILCD method are still positive. Recipe 2016 results have lower impacts in all scenarios in the Particulate matter category. However, even with the discrepancies in the final net performance of the categories, the methods agreed with the classification of the scenarios' performance: scenario 1 was the worst scenario, and scenario 4 was the best in both. Therefore, the sensitivity analysis results indicate that the results of the LCA conducted for recycling scenarios in Brazil are robust.

The sensitivity analysis showed that the recovery rates of the materials and the impact assessment method used mainly influenced the results of net impacts. Greater recovery of materials in the hydrometallurgical process is essential to obtain net negative environmental benefits because it uses many processes and chemical inputs that bring an environmental burden to its production. In addition, the disposal of materials such as plastics, paper and nonferrous metals by the cases and current collectors can influence the overall impacts. For Brazil, which still does not have LIBs collection networks and recycling systems developed, the results of this sensitivity analysis help to clarify the importance of implementing efficient systems for material recovery, to increase the benefits from avoiding the production of virgin raw materials and the disposal of large amounts of valuable materials in landfills.

3.3.1 Sensitivity analysis from inclusion of RL transport

The sensitivity analysis that considers the inclusion of intermediate transport in the evaluation of the scenarios shows that the inclusion of intermediate transport to the pretreatment point in each scenario can increase the overall impacts of each scenario. The generation of impacts is directly related to the increase in the amount collected by RL and the number of cities served, as more weight is collected and transported over more distances once more cities are included in the plan. In scenario 1 where less waste is collected (6%) and fewer cities are served (186 cities) the impacts of transport for the Climate change category increase the total impacts by only 1.48% while in scenarios 2, 3 and 4 the impacts increase by 13.67%, 10.44% and 32.21% respectively. If intermediate transport is included, the magnitude of the impacts can increase from less than 0.01% to 1000% depending on the impact category analysed and the scenario analysed. For details of the results of the scenario evaluation considering RL, see supplementary material S15.

The results show that the categories Terrestrial eutrophication, Water resource depletion, Land Use, Acidification, Photochemical ozone formation, Ionising radiation and Ozone depletion are the most sensitive when transport

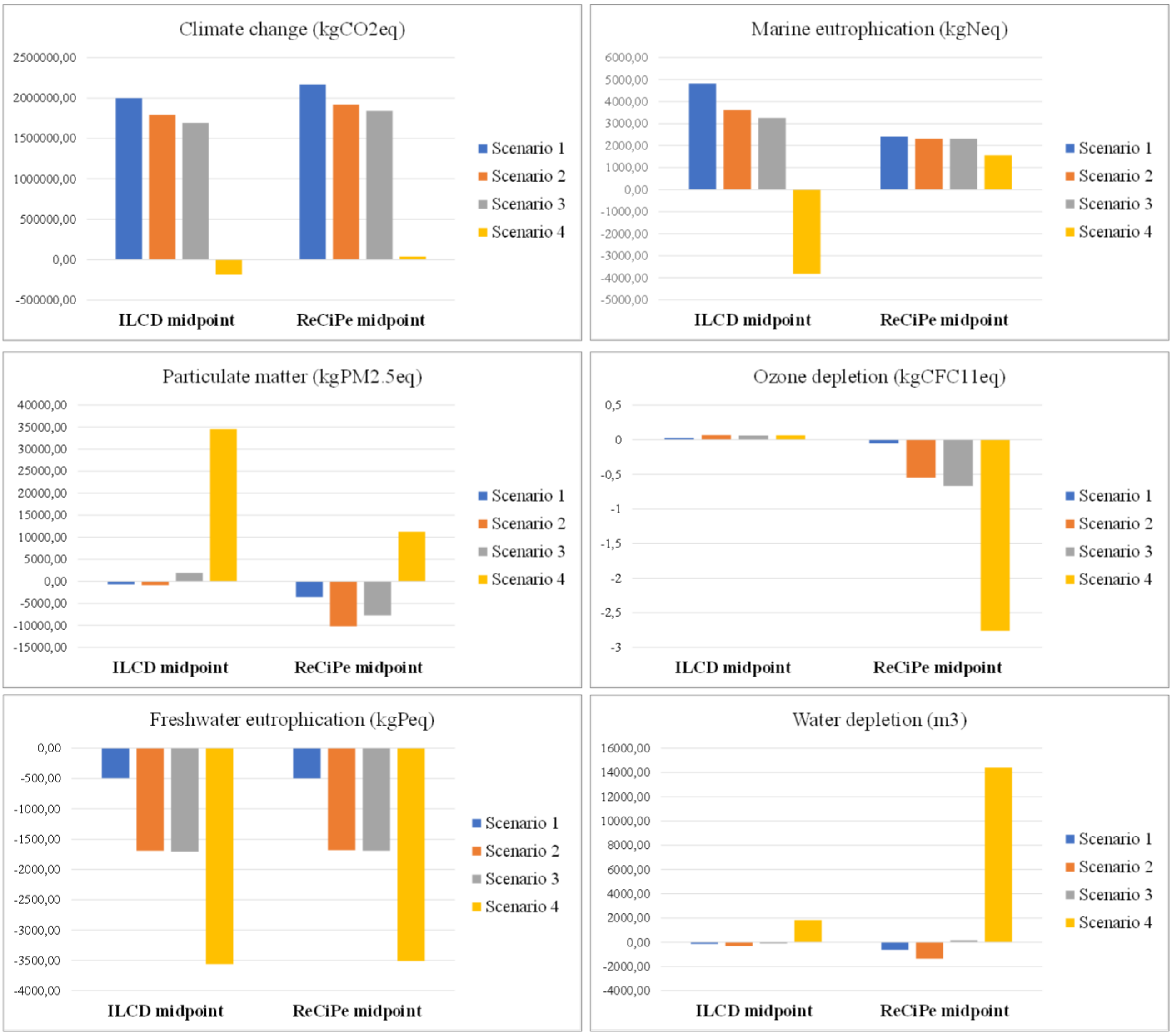

FIGURE 10: Sensitivity analysis comparing results of ILCD midpoint and ReCiPe midpoint methods in net result of scenarios.

is included. This makes sense, since road transport LCAs point out that transport exhaust emissions such as Nox, SOx and NMVOC are the main contributors to the acidification, eutrophication, photochemical ozone formation and ionising radiation categories, the use of diesel fuel contributes to increased impacts in the ozone depletion category (Merchan et al., 2020) and the use of roads contributes mainly to increased impacts in the land use categories (Osorio-Tejada et al., 2023). For the water depletion category, water consumption in the production of diesel and trucks is the main contributor to the increase in impacts.

However, due to the progressive approach of the scenarios, even with the inclusion of transport the ranking of the scenarios does not change, although the environmental benefits arising from material recovery are no longer sufficient to reduce emissions to a net negative state in scenario 4, even so the total net impacts are largely reduced. Therefore, the results of the sensitivity analysis show that depending on the amount of transport required for RL operations, the net impact results can be largely affected.

Finally, it should be noted that the lack of data on the average distance of collection routes in each city and to landfills in Brazil and Europe meant that the transport of these operations could not be included in the system limit, which is a limitation and can interfere with the total amount of impacts generated by the RL of LIBs.

4. CONCLUSIONS

This study assesses potential environmental impacts for prospective LIBs waste recycling from smartphones and laptops. The estimate of LIBS waste generation revealed that a large amount of LIBs waste can be discarded each year in the smartphones and laptops waste stream in Brazil (3,000 tons/year from 2023 to 2026), confirming the great potential of LIBs waste generation and resource recovery in the WEEE streams.

Through a hydrometallurgical route, recycling brings net environmental benefits, reducing environmental impacts in all assessed categories. Ni and Co recovery provides the highest environmental benefits across all impact categories. The pretreatment processes implemented enriched the electrolytic material fraction of LIBs can be enriched, increasing the potential material recovery and providing greater environmental benefits. The use of an organic reducing agent (glucose) resulted in negative impacts (benefits) due to carbon sequestration accounted in the maise cultivation phase. However, the glucose produced from maise can still be replaced by agro-industrial waste, such as molasses and whey, which can further improve the overall impacts of the process. In addition, recovery of other types of materials, such as plastics, steel, aluminium and copper, from casings and current collectors can improve the performance of the hydrometallurgical process.

The LCA results for scenario assessment show that more significant environmental benefits are achieved when the RL collection rate is increased, and the less impactful technology option makes the recovery of materials. The results of net environmental impacts indicate that recycling a greater amount of LIBs waste by Brazil's hydrometallurgical technology results in net environmental benefits across 13 impact categories. The scenario 4 presents the best performance, with an increase in the collection by RL to 50% of the waste generated and a reduction in the landfill rate by 44%. Three impact categories (Ozone depletion category, Particulate matter category and Water resource depletion category) in scenario 4 do not present the best net impact results due to the increase in the consumption of inputs such as NaOH, glucose, Na_2CO_3, limestone and particulate matter emissions for recycling a greater amount of LIBs waste. Therefore, further improvements in the hydrometallurgical process, such as recycling efficiency, reducing the use of chemicals, controlling particulate matter, and using glucose sources from agro-industrial waste, can improve the environmental profile of hydrometallurgical recycling and LIBs waste management in general.

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