Published in: Journal of Cleaner Production (https://doi.org/10.1016/j.jclepro.2021.126954)

Challenges for sustainable lithium supply: a critical review

Amato Alessia[a]*, Becci Alessandro[a], Villen-Guzman Maria[b], Vereda-Alonso Carlos[b], Beolchini Francesca[a]

[a]Department of Life and Environmental Sciences, Università Politecnica delle Marche, Ancona, 60121, Italy

[b] Department of Chemical Engineering, Faculty of Sciences; University of Malaga. 29071-Malaga. Spain.

Abstract

The growing diffusion of green technologies, essential for a low carbon emission economy, has caused an increasingly demand of lithium and its recent inclusion in the list of critical raw material for Europe. Currently, the main resources of lithium include brines and hard rock ores (placed in Chile, Australia, Argentina and China). Nevertheless, the possibility of an integrated supply system, able to include primary and secondary productions (from waste batteries), is becoming more and more interesting to decrease the environmental impacts. In this context, the present review combines a complete overview of lithium supply state-of-art, with an environmental assessment of several scenarios where primary production is integrated with urban mining strategies. The assessment aims at including several market aspects, to obtain results consistent with the evolving European context. Starting from real information, the evaluation proved the possible substitution of about 30% of primary lithium, with a consequent reduction of the environmental impact (>10%). The results represent an important supporting tool for the improvement of lithium recycling value chain. Indeed, several variables characterize the waste batteries management, including the available quantities, the selected recycling process and the relative amount of rechargeable/not rechargeable batteries. These aspects affect the real sustainability of European lithium supply strategy: centralized system vs a decentralized approach with many facilities on the territory. The combination of the carbon footprint

assessment and the Monte Carlo methodology suggested the lowest impact of decentralized facilities for the greatest waste quantities, especially with high contribution of rechargeable batteries. The holistic view of the present review, able to include both the recycling and the strategic choices in the recycling planning, fit perfectly with the circular economy principles to meet the challenge of a sustainable and clean lithium supply system.

Keywords

Lithium supply, circular economy, batteries, carbon footprint, sustainability

Declarations of interest: none

Contents

1. Introduction

The concept of "critical material" was first introduced by a United States (US) federal law in 1939 (Councel, 1939) to identify materials, which play an important role for military, industrial and essential civilian needs, and which were not found or produced in the US in the enough quantities to satisfy such needs. Since then, a wide variety of definitions of "criticality" of a material has been reported: "the combination of high economic importance with a comparatively high risk of supply disruptions" (Buijs et al., 2012), "the quality, state or degree of being of highest importance, of particular interest in the case of metals" (Graedel and Nuss, 2014), "the extent of current and future risks associated with a certain material" (Gleich et al., 2013). According to Frenzel et al. (Frenzel et al., 2017), the imprecise definition for raw material criticality is mainly associated to the variability in definitions and assessment methodologies. However, the authors also concluded that most of studied are based on 1) the importance of the raw material and the consequent impact of supply shortfalls and 2) the supply risk, i.e. the probability for the occurrence of such disruptions (Frenzel et al., 2017).

The European Commission launched the European raw material initiative in 2008 with the aim to favour the raw material market of the European Union (EU), decreasing the primary raw material depletion and promoting the recycling strategy (European Commission, 2008). The identification of critical raw materials (CRM), relevant for the EU, economy, was established as a priority action of the initiative of 2008. As a result, in 2011, after the assessment of 41 non-energy, non-agricultural raw materials, the first CRMs list was published identifying 14 CRMs (European Commission, 2011), based on the comparison of two main parameters: the Economic Importance (in terms of end-use applications and the value added of corresponding EU manufacturing sectors), and the Supply Risk (based on the concentration of primary supply from raw materials producing countries, considering their governance performance and trade data (Blengini et al., 2017; Petranikova et al., 2020; Tkaczyk et al., 2018)). A regular updating of the CRMs list is essential due to market and technological developments and new information on the environmental impact of a material, the EC published the second list of 20 CRMs in 2014 (European Commission, 2014a). The third list of 26 CRMs in 2017 (European Commission, 2017a), was assessed by an improved methodology able to use data from over the last 5 years and to introduce a data resource priority for the identification of the production stages with the highest supply risk for the EU (i.e. extraction or processing) (European Commission, 2017b). Among the selected materials, lithium was considered a borderline to carefully evaluate. As outlined in Figure 1, lithium exceeded the threshold for economic importance, but the supply risk was considered non-critical according to the CRMs list of 2011 and 2014. According to European Commission (European Commission, 2014b), lithium would have been considered critical using the Environmental Performance Index (EPI) to evaluate the supply risk instead of the poor governance indicator. The evolution of the economic importance cannot be evaluated since the criticality threshold value was moved from 5 to 2.8, as a result of the implementation of the aforementioned revised methodology (European Commission, 2017c). The most recent list of 2020 has finally included lithium among the CRM, since the production of vehicle batteries and the necessity of energy storage will increase the lithium demand up to 18 times in 2030 and 60 times in 2050, compared to the current European supply (European Commission, 2020a). The geographical distribution of lithium resources, mainly located in South America, entails a high import European dependence of this materials, which is also an important aspect in the evaluation of risk supply (Oliveira et al., 2015). Hence, selective lithium recovery from all possible resources should be addressed to close the loop for a circular economy. Considering that the current lithium recovered is insignificant, great efforts should be made to avoid criticality (Sun et al., 2019).

2. Aims and scopes of the review

As confirmed by the more recent policies, lithium is essential for the transition towards a low carbon economy (European Commission, 2020a, 2020b, 2019a). Considering the strategic interest for this element, many reviews are present in the scientific literature, focusing on specific aspects, including the best strategies for a cleaner production (intended as reduction of impacts, waste flows, energy demand and raw material consumption). In this regard, Stamp at al. (2012) proposed a critical review about lithium carbonate supply that compares three possibilities of metal supply from primary resources, neglecting the recycling aspects (Stamp et al., 2012). On the other hand, social and political aspects of primary lithium production were discussed by Hancock et al. (2018). Authors described the Bolivia case study and how the challenge of the clean lithium production has been translated into unusual governance arrangements between state enterprises and foreign-owned private corporations (Hancock et al., 2018). Many specific aspects about lithium were discussed in the recent literature, including chemistry, primary production and deposits (Choubey et al., 2016; Li et al., 2019; Olsher et al., 1991; Tadesse et al., 2019), use, recovery (Meshram et al., 2014; Swain, 2017; Talens Peiró et al., 2013) and demand forecast (Gil-Alana and Monge, 2019; Mohr et al., 2012; Tarascon, 2010; Vikström et al., 2013). Further reviews focus on the possible toxicity effects on both human and environmental health (Aral and Vecchio-Sadus, 2008; Livingstone and Rampes, 2006; McKnight et al., 2012; Oruch et al., 2014; Young, 2009). Nevertheless, a critical assessment of the sustainability of an integrated supply system, able to combine primary and secondary resources, still lacks. Considering their adoption in an extensive range of applications, the most discussed topic in the recent literature is the lithium in batteries, both in production and recovery terms. Indeed, the lithium batteries are considered one of the main success of the modern electrochemistry and research focuses on the possible improvements for their manufacturing, considering safety, environmental and energetic aspects (Kavanagh et al., 2018; Scrosati, 2011, 2000; Scrosati and Garche, 2010; Wang et al., 2015). In this regard, the life cycle assessment (LCA) tool was used to compare the environmental burden due to battery life, comparing different technologies and performances (Oliveira et al., 2015). The battery exploitation for lithium recovery was proposed by several authors, which presented an overview of the existing treatments (from pre-treatment to metal recovery) (Liu et al., 2019; Lv et al., 2018; Ordoñez et al., 2016; Xu et al., 2008; Zeng et al., 2014) or deepened specific aspects, like environmental friendly recycling treatments (Golmohammadzadeh et al., 2018) and commercial processes (mainly hydrometallurgical, pyrometallurgical, mechanical treatments) (Georgi-Maschler et al., 2012; Pinegar and Smith, 2019). A battery overview was proposed by both Richa et al. 2017 and Meshram et al. (2020), which took into account different steps of circular economy hierarchy, including production, use, collection, reuse, recycling, incineration, landfilling and transport (Meshram et al., 2020; Richa et al., 2017). Nevertheless, in the first case a sustainability assessment was not included to support the main observations (Richa et al., 2017).

The common conclusion of the current literature is that we are living the lithium revolution and there is an evident metal need for an effective European green economy transaction (Ciez and Whitacre, 2019). Nevertheless, this target can be achieved by the identification of the most sustainable and the cleanest strategy able to implement a holistic view, which includes all the aspects connected to lithium supply and recycling, in agreement with the "New circular economy action plan for a cleaner and more competitive Europe" (European Commission, 2020c).

In this context the present review aims at supplying a complete overview about lithium covering all the aspects of its life: from the supply to the recovery from waste, following a circular economy schema. The hypothesis of complex scenarios, able to combine the traditional primary production with the available recovery options was assessed by the LCA tool. The creations of many scenarios able to include several variables aims to supply a model for the sustainability assessment, suitable for the European context, to meet the challenge of the sustainable lithium production.

3. Lithium use

The properties of lithium and its compounds, clearly different from the other alkali metals, explain its high versatility and low substitutability. Lithium presents the highest sublimation energy, electronegativity and ionization energy and the smallest ionic radius for the alkali groups which entails a high charge density for lithium ions (Hart and Beumel, 1970). Some of the most relevant applications of lithium metal are as a chemical intermediate in many reactions, as a polymerization catalyst, in high strength glass and glass ceramics, as allowing agent and in batteries. Lithium is extracted from primary resources as lithium carbonate (Li_2CO_3) which has been widely used to lower boiling point and increase the resistance to thermal expansion in glass and ceramic applications, as a pharmaceutical (Talens Peiró et al., 2013) and as cathode material for Lithium Ion batteries (LIBs) (Scrosati, 2011). Furthermore, lithium carbonate is applied industrially as a starting reagent for the production of lithium salts, such as lithium chloride (LiCl), used in air-conditioning industry and lithium hydroxide (LiOH), used in the preparation of lithium-based greases and as an effective carbon dioxide absorbent (Hart and Beumel, 1970). Furthermore, the recent literature reports the use of LiOH as innovative catalyst for biodiesel production, thanks to its availability in battery waste (Brito et al., 2020).

According to the U.S. Geological Survey and several thematic sites (Table 1), batteries and ceramic and glass are the most relevant end-use market for lithium (Mining.com, 2020; Statista, 2020; USGS, 2019). From the evolution of percentage shares in the most relevant applications (Figure 2), it can be concluded that the lithium use in batteries is increasing among all the other sectors in recent years. Regarding the use of lithium to produce high energy density batteries, studies started in the 1950s as a consequence of promising results concerning properties of this metal (Brandt, 1994). The production of not rechargeable lithium batteries (also called primary batteries) was launched in the late 1960s with applications in military and industrial systems. Non-rechargeable lithium batteries use lithium metal as anode and different materials, such as manganese dioxide, iron disulphide, sulphur dioxide and carbon monofluoride, for the cathode. Lithium/manganese dioxide cell is currently the most commercialized not-rechargeable lithium battery used in memory backup, cameras, consumer devices and military applications (Linden and Reddy, 2002). The practical application of lithium in rechargeable batteries was introduced by the Japanese Sony manufacturer in 1991, mainly trigged by power necessity in military field (Scrosati, 2011). The current growth of LIB production is due to its increasing demand in transport, connectivity and stationary applications, such as energy storage systems (Lebedeva et al., 2016). European Commission (European Commission, 2019b, 2019c) reports that the demand for LIBs is expected to increase, in terms of capacity, to a maximum of 660 GWh by 2023, 1100 GWh by 2028 and 4000 GWh by 2040 which means an increase of about 50 times respect to the current consumption.

Based on the specific product, a battery may include from one “battery” cell (e.g., smart phones) to more than 1000 cells (e.g., computers, power tools, electric vehicles) (Huo et al., 2017). It should be noted that lithium content in rechargeable batteries is higher than in some primary resources. Furthermore, Sonoc et al. (Sonoc et al., 2015) predicted that the supply of lithium could be only ensured through high LIB recycling rates with a minimum lithium recovery of 90%.

The most typical structure of a LIB consists of a couple of electrodes, an electrolyte separator contained in a stainless steel shell or in a pouch case. The bulk composition of LIBs depends mainly on the type of battery chemistry and the manufacturer. The average composition is about 50% of base metal, 29% of graphite, 7% of binder and plastics, 4% electrolyte and 10 % other components (Wang et al., 2014a). The anode is usually made up of graphite supported on a copper foil collector by polyvinylidene fluoride (PVDF) binder. The carbon structure allows the intercalation of lithium ion between graphene planes during the charging process offering attractive properties such as good mechanical stability, electrical conductivity and lithium transport (Nitta et al., 2015). The amount of lithium that can be stored per mass of anodic material is directly associated with the energy storage density which is around 372 milliamp hours per gram ($mAhg^{-1}$) in the case of graphite anodes (Wang et al., 1998). The relatively low volumetric capacity of commercial graphite electrodes has promoted research to explore alternative anode materials. Some of the most promising materials to replace common carbon based materials as negative electrode are lithium metal alloys (Scrosati and Garche, 2010). Titanium oxides, such as titanium oxide (TiO_2) and lithium titanium oxide ($Li_4Ti_5O_{12}$), are also gaining relevance as attractive anode alternative (Chen et al., 2012). The replacement of graphite anode with the aforementioned materials with a high value would involve a clear increase of concern about the recovery of raw materials from spent LIBs.

The cathode consists of active material supported on both sides of an aluminium foil acting as current collector (Goodenough and Park, 2013). Cathode composition is about 88% of valuable metal oxides containing lithium, 8% of conductive agent and 4% of PVDF binder (An, 2019). Regarding active material, lithium cobalt oxide (LCO), disclosed by Goodenough in 1980 (Mizushima et al., 1980), is still used in the majority of commercial LIBs due to its properties: relatively high theoretical specific capacity, high theoretical volumetric capacity, low self-discharge, high discharge voltage and good cycling performance. The average composition for $LiCoO_2$ cathode LIBs from seven manufacturers is presented in Figure 3 (Wang et al., 2014b). Other cathode materials have been explored to overcome some of the limitations of LCO cathodes, including low thermal stability, fast capacity loss at high current rates or during deep cycling and high cost due to cobalt. Most cathode material research is mainly focused on transition metal oxide and polyanion compounds, such as lithium manganese oxide (LMO), lithium nickel cobalt manganese oxide (NCM), lithium nickel cobalt aluminium oxide (NCA), lithium cobalt phosphate (LCP) and lithium iron phosphate (LFP). The role of lithium as guest ions in these intercalation compounds highlights the relevance of this metal in the technology. The electrolyte usually consists of a lithium salt solution (e.g. $LiPF_6$, $LiClO_4$, $LiBF_4$) dissolved in a mixture of organic solvent (e.g. ethylene carbonate-dimethyl carbonate, EC-DMC) (Nitta et al., 2015).

The study of lithium uses represents the starting point for the identification of the most promising secondary resources (mainly batteries on the quantity basis, Table 1) and the development of effective recovery strategies.

Table 1: The global lithium market, the most relevant applications (Mining.com, 2020; Statista, 2020; USGS, 2019)

Lithium use	Lithium demand (tonnes/year)
Rechargeable batteries	42200
Not rechargeable batteries	1500
Ceramics	7800
Glass-ceramics	7000
Greases	7000
Polymer	3100

Metallurgical powder	2300
Glass	2300
Air treatment	1500
Other	5500

4. Lithium primary production

4.1 Mining

As highlighted by Gil-Alana & Monge (Gil-Alana and Monge, 2019), when discussing about the lithium supply, the distinction between reserves and production is essential. Many estimations of lithium deposits are reported in the literature, identifying the main reserves in Chile, Australia, Argentina and China (Gil-Alana and Monge, 2019; USGS, 2019) (Figure 4a). In this regard, the literature identified two main economic resources of lithium: brines and hard rock ores (Tadesse et al., 2019). The first kind of deposits includes saline water with high content of dissolved salts. They could be identified in the rock pores, where water from lakes or sea water has undergone extreme evaporation. Lacustrine and playas environments are the most common for lithium deposits formation since the brine fills the pores among grains (Gruber et al., 2011). The presence of metal within brines is due to both the rock erosion and the hot springs that feed water in these areas (Gruber et al., 2011; Munk et al., 2011). The brines is extracted from the aquifers and pumped into evaporation ponds for the concentration at controlled conditions, able to eliminate some contained compounds, mainly magnesium and sulphate (Gruber et al., 2011; Kushnir and Sandén, 2012). The process needs low electricity consumption since the initial step uses solar evaporation. Overall, lithium is present with variable concentrations which reach the 0.14% in the Salar Atacama, a salt flat in Chile, where the metal is produced in carbonate form (Gruber et al., 2011). Further brine resources include oil field and geothermal brines (Kushnir and Sandén, 2012). The ore deposits is the alternative for the lithium supply, with the highest impact due to the extraction process (Kushnir and Sandén, 2012). Literature reports 131 minerals of lithium, nevertheless, only six silicate and phosphate based minerals are identified with an economic impact (Choubey et al., 2016; Li et al., 2019; Tadesse et al., 2019;

Webmineral.com, 2020). In this regard, lithium is mainly present in aluminium silicate deposits, known as pegmatites (Bale and May, 1989; Tadesse et al., 2019). Pegmatite ores contain mineral such as spodumene ($LiAl[SiO_3]_2$) (the most important for the market), petalite ($LiAlSi_4O_{10}$), lepidolite ($KLiAl_2Si_3O_{10}(OH,F)_3$) and amblygonite ($LiAl[PO_4][F,OH]$). Furthermore, Tadesse et al. (2019) report zinnwaldite ($K[Li,Al,Fe]_3[Al,Si]_4O_{10}[F,OH]_2$) and eucryptite ($LiAlSiO_4$). Table 2 summarizes the main deposits worldwide.

Table 2. Estimated deposits worldwide (Adapted from (Tadesse et al., 2019; Vikström et al., 2013))

Country	Minerals	Estimated quantities (Mtons)
Afghanistan	Spodumene	n.a.
Australia	Spodumene	0.79
Austria	Spodumene	0.10
Brazil	Spodumene, Petalite	0.92
Canada	Spodumene	2.41
China	Spodumene, Petalite, Lepidolite	2.40
Congo	Spodumene	3.80
Finland	Spodumene	0.68
Mali	Amblygonite	0.03
Portugal	Petalite	0.01
Namibia	Petalite	0.15
Russia	Spodumene, lepidolite	3.69
Serbia	Jadarite	1.00
USA	Spodumene	13.8
Zimbabwe	Spodumene, Petalite	0.73

After mining, a combination of treatments for lithium beneficiation is necessary. Generally, it includes gravity separation, magnetic separation and froth flotation, with the final production of lithium concentrate (Tadesse et al., 2019). Currently, the lithium minerals, mainly pegmatite and spodumene, represent only the 25% of the whole reserves. On the other hand, brines show the highest availability, around 65%, combined with the cheapest exploitation (Swain, 2017). Nevertheless, this resource shows an important weak point due to the long evaporation time (between 1 and 2 years) necessary to achieve the final product (Tadesse et al., 2019). This limit makes the brines not adaptable to the market changes and highlights the necessity of an integrated supply system able to include not only primary resources.

4.2 The primary lithium market and the criticality of worldwide transportation

An interesting date concerning the production decrease is reported in Figure 4b. The reduction of about 20% from 2018 to 2019 is mainly due to China which has scaled back the subsides on the electric vehicles (USGS, 2019). The response of excess of lithium production, also due to the considerable increase of the Australian mining from spodumene pegmatite deposit, has been translated into a carbonate price drop in the last two years (Figure 5) (Fastmarkets, 2020; Metalary, 2020; Tadesse et al., 2019; USGS, 2019). The relevant effect of the decrease of China demand on lithium carbonate price could be better explained by Figure 6a, which includes the country among the three main importers (22% of the whole economic imported value, pairs to approximately 360 Million $), between Korea (28%) and Japan (20%), in 2018 (TrendEconomy, 2019). In addition, the role of USA and Belgium is relevant for the worldwide lithium economy, as deducible form the related economic flows (Chen et al., 2020; TrendEconomy, 2019). Around 70% of carbonate lithium flow (Figure 6b) comes from Chile with a considerable transportation impact from both an environmental and economic point view (Resource Trade Earth.com, 2020). A possible trip of raw material to Europe includes a preliminary transportation from Atacama's brine to Antofagasta port by rail (Kogel et al., 2006) followed by the loading on a transoceanic ship freight for the route Antofagasta-Rotterdam of 11062 km (Marine traffic, 2020; Oliveira et al., 2015; Searoutes.com, 2020). The ship transport cost depends on both the distance and the taxes of countries (Korinek and Sourdin, 2009).

Overall, the share of the shipping cost affects the raw material cost of about 15-25% (Korinek and Sourdin, 2009; Rodrigue, 2020).

5. Lithium secondary production

5.1 Batteries as potential lithium secondary resources

The growing demand of lithium has encouraged the exploration of secondary resources to reduce the amount of primary metal required. In addition to the rise of metal availability, the recycling aims at the increase of both the supply sustainability, and the cleaner metal production compared to the traditional resources. These targets could be reached thanks to a double advantage: the implementation of low impact processes (with lower emissions, resulting waste and consumptions) and the waste accessibility all over the word, with the consequent reduction of the transportation. The recovery of lithium from dissipative applications, including lubricants greases, air treatment and pharmaceuticals, is not viable due to its gradual release into the environment. Recycling technologies for glass and ceramics have not been reported in literature due to the difficulty of separating the target metal to obtain high-quality product (Ziemann et al., 2012). Currently, the main secondary lithium resource for recycling on a large scale are rechargeable and not rechargeable batteries, the most important use of lithium. The identification of sustainable recycling strategies could further solve the issue of waste battery management, currently regulated by the Directive 2006/66/EC (European Commission, 2006). LIBs are classified in the category of "other batteries" which also includes alkaline batteries. The significant growing of lithium batteries consumption since 2006 makes needed to consider its peculiarities in legislation (Lebedeva et al., 2016). The minimum recycling target of 50% stipulated by the EU Batteries Directive does not guarantee the recovery of valuable materials contained in spent batteries, like lithium. On the other hand, the lack of clear regulation to promote the collection of waste industrial lithium batteries together with the growing electric vehicles use corroborates the urge of developing sustainable recycling schemes and establishing clear targets for lithium recovery. According to Mathieux et al. (Mathieux et al., 2017), the current contribution of recovered lithium to materials demand is less than 1%. However, there are high expectations that lithium recovery from spent batteries significantly increases with the improvement of recycling technologies. Some positive impacts related to recycling and recovery of lithium has been predicted by European Commission. For example, collection rate of 65%, together with a recycling efficiency of lithium of 57%, would involve not only an income of € 408 million in 2030 from recovered materials, such as cobalt, nickel, aluminium and lithium but also the creation of more than 2600 jobs (Drabik and Rizos, 2018; European Commission, 2019b). Furthermore, the recovery of lithium from batteries would decrease the demand for raw materials required to produce electric vehicle batteries (European Commission, 2019c). Depending on the metal extraction process, recycling technologies could be divided into hydrometallurgy, pyrometallurgy, biometallurgy and combined. These processes are focused on the transformation of solid metal contained in residues into solution state or alloy. Most of current industrial processes devoted to batteries recycling focuses on the recovery of cobalt and nickel with the exception of TOXCO and Recupyl, hydrometallurgy-dominant processes, and Accurec GmbH, a pyrometallurgical method, which also pursues lithium recovery (Liu et al., 2019). These processes are briefly described in the following sections.

5.2 Pre-treatment processes for recycling lithium batteries

Previous to processing of spent lithium batteries, full discharging is required to prevent explosions, gas emission and fires as a consequence of short-circuiting and self-ignition during the dismantling step (Table 3) (Li et al., 2010a; Sun and Qiu, 2011). To discharge batteries, the most common discharging methods used are chemical discharging (Wang et al., 2018) and cryogenic freezing (Contestabile et al., 1999; Dorella and Mansur, 2007). Chemical methods are based on the soaking of spent batteries in conductor solution. Although the most common salts used are NaCl and Na_2SO_4 (Chen et al., 2017; Pinna et al., 2017), other reagents have been analysed not only to improve the discharging efficiency but also to avoid corrosion, such as electric iron powder (Nan et al., 2005), K^+, NH_4^+ (Shaw-Stewart et al., 2019), $MnSO_4$, $FeSO_4$, Cu or graphite (Yao et al., 2020). During chemical discharging, losses of lithium contained in the electrolyte of LIBs has been reported which could be avoided using physical discharging and cryogenic freezing. Although high recycling cost has restricted the recovery of lithium contained in electrolyte up to now (Liu et al., 2019), different approaches to deal with this aspect has been reported in literature. Grützke et al. (Grützke et al., 2014) proposed supercritical carbon dioxide extraction of electrolytes as a promising method to be implemented in batteries recycling. In the same way, Liu et al. (Liu et al., 2017) developed a method based on the use of supercritical carbon dioxide concluding that reused electrolyte presented electrochemical performance very similar to the commercial electrolyte.

Preliminary treatments are usually applied to improve the recycling efficiency of valuable raw materials. Mechanical pre-treatment, including crushing, shredding, sieving and air and magnetic separation, combined with manual dismantling, are applied to obtain different fractions: plastic, paper, ferrous and non-ferrous metals and electrodic powder (Georgi-Maschler et al., 2012; Hu et al., 2017; Li et al., 2012, 2010a; Zeng et al., 2014). Mechanical separations are often based on the changes of material physical properties (e.g. density, magnetic behavior and conductivity (Golmohammadzadeh et al., 2018; Xu et al., 2008). Thermal processes (150-500°C) are commonly used to remove impurities applying high temperatures to decompose organic binders contained in cathodes; solvent and electrolytes solvent are evaporated and combusted (Diekmann et al., 2017; Georgi-Maschler et al., 2012; Zeng et al., 2014). Alternatively, organic compounds are employed to dissolve the binder in the spent batteries to make more efficient the separation of cathode active material and subsequently, the recovery of copper and aluminium (Li et al., 2011). The combination of mechanical treatments and reagents, i.e. mechanochemical treatments, to improve separation processes is a promising alternative pre-treatment method. The induction of physical and chemical changes of active materials through mechanical energy allows working at room temperature and atmospheric pressure with the subsequent reduction of costs (Wang et al., 2017).

Table 3: State-of-the-art of pre-treatment process for recycling lithium batteries

References	Pre-treatment method	Operative conditions
Contestabile et al. 1999	Cryogenic freezing + mechanical treatment	Liquid nitrogen
Nan et al. 2005	Mechanochemical treatment	Washing with water and electric iron powder for 30 min.
Dorella and Mansur 2007	Manual + cryogenic freezing	Liquid nitrogen
Li et al. 2010a	Manual + cryogenic freezing + thermal treatment	Liquid nitrogen for 4 min, followed by washing with N-methylpyrrolidone (NMP) at 100°C for 1 h and a calcination at 700°C for 5 h
Li et al. 2011	Chemical + thermal treatment	Ultrasonic washing with NMP followed by a thermal treatment at 800°C for 2 h
Georgi-Maschler et al. 2012	Manual + thermal treatment	Pyrolysis at 250°C
Li et al. 2012	Manual + thermal treatment	Ultrasonic washing with NMP solution for 20 min followed by roasting at 450°C for 1 h
Grützke et al. 2014	Electrical + thermal treatment	Constant current of 2.25 A followed by thermal treatment with an electrolytic solution in autoclave
Chen et al. 2017	Manual + thermal/mechanical treatment	Washing with an electrolytic solution (Na_2SO_4 10%w/v) for 24 h, afterwards, thermal treatment at 90°C for 2 h with a NMP solution
Diekmann et al. 2017	Manual + mechanical treatment	Crushing within a nitrogen atmosphere with a flow of 4 l/min
Hu et al. 2017	Chemical + thermal treatment	Washing with a 1.5 M NaOH solution for 3 h followed by a thermal treatment at 650°C for 3 h
Liu et al. 2017	Mechanical treatment	Mechanical dismantling into an extraction vessel in an argon-filled glovebox with moisture and oxygen level lower than 1%
Pinna et al. 2017	Chemical + thermal treatment	Washing with a saturated solution of NaCl followed by a thermal treatment at 300°C
Wang et al. 2017	Mechanochemical treatment	Washing with a 5% (w/v) NaCl solution followed by a washing with a NMP solution
Wang et al. 2018	Mechanochemical treatment	Washing with a 5% (w/v) NaCl solution for 24 h
Shaw-Stewart et al. 2019	Chemical treatment	Washing with aqueous solutions using different salts

Yao et al. 2020	Chemical or physical treatment	(Na^+, K^+ or NH_4^+) at final concentration of 5% (w/v). Chemical treatment: washing with a solution 0.8 M of NaCl, $FeSO_4$ or $MnSO_4$ for 125 min. Physical treatment: copper and graphite powder.

5.3 Processes for recycling lithium batteries

Hydrometallurgical processes are primarily based on leaching, extraction and precipitation as summarized in Table 4. Few studies dealing with the hydrometallurgical treatment exclusively focused on not rechargeable lithium batteries have been reported. Taking Li/MnO_2 as the most representative example system, Contestabile et al. (1999) proposed a recycling process using as a promising extracting reagent, the isobutyl alcohol/H_2O system with the aim of promoting a mild oxidation of lithium metal contained in anodes. Afterward, lithium was recovered as lithium carbonate passing CO_2 gas through the solution. Several inorganic acids, including HCl, HNO_3 and H_2SO_4, were proposed to recover cathodic material. (Contestabile et al., 1999).

Most of the studies are focused on rechargeable lithium batteries as a result of increased importance of this residue as prime secondary resource of lithium. The relative importance of different leaching parameters has been evaluated by Gao et al., concluding that the most influential parameters are, in descending order: acid species and concentration, leaching time, reductant species and content, solid to liquid ratio, reaction temperature and stirring speed (Gao et al., 2018). Overall, the main leaching agents studied could be categorized into inorganic acids, organic acids and alkaline solutions. Inorganic acids including HCl, HNO_3 and H_2SO_4, have been thoroughly studied. Taking as an example $LiCoO_2$ as the most commonly used LIBs cathode, the leaching reactions by HCl, HNO_3 and H_2SO_4 could be represented as:

$$3\ HCl + LiCoO_2 \rightarrow LiCl + CoCl_2 + 1.5\ H_2O + 0.25\ O_2 \qquad (1)$$

$$3\ HNO_3 + LiCoO_2 \rightarrow LiNO_3 + Co(NO_3)_2 + 1.5\ H_2O + 0.25\ O_2 \qquad (2)$$

$$1.5\ H_2SO_4 + LiCoO_2 \rightarrow CoSO_4 + 0.5\ Li_2SO_4 + 0.25\ O_2 + 1.5\ H_2O \quad (3)$$

The recovery of lithium carbonate was performed by Zhang et al. (1998) and Barik et..al (2017), after a leaching with HCl able to reach efficiencies around 99% for Li, Co and Mn (Barik et al., 2017; Zhang et al., 1998). As can be concluded, the relevant action of HCl is a consequence of its relative high reducibility. The lithium leaching performance has been also widely explored using H_2SO_4 as leaching agent, reaching efficiencies higher than 90% (Georgi-Maschler et al., 2012; Sun and Qiu, 2011). The use of reducing agents, typically H_2O_2, has been demonstrated to improve leaching efficiency, with relatively short times (Ferreira et al., 2009; Shin et al., 2005). Furthermore, Jiang et al. (2018) proposed ultrasound-assisted leaching, as alternatives to conventional magnetic stirring, to improve efficiency and decrease the reaction time (Jiang et al., 2018). The use of other reductant, including sodium bisulphite ($NaHSO_3$) (Meshram et al., 2015) and glucose (Pagnanelli et al., 2017), has been explored to avoid limitations associated with the use of H_2O_2, such as stability problems in storage for long time. (Joulié et al., 2014). On the other hand, Lee et al. evaluated the use of HNO_3 concluding that the recovery of lithium and cobalt was poor even using high concentrations (Lee et al., 2007). The use of inorganic acids, as leaching agents, entails some limitations, such as: release of toxic gases, consumption of a large amount of water, corrosion of the equipment and generation of secondary pollutants. With the aim of reducing the footprint of recycling processes, the use of organic acids is explored as alternative leaching agents (Golmohammadzadeh et al., 2018). Organic acids tested for the recovery of lithium and cobalt includes citric acid (Li et al., 2013, 2010a), malic acid (Li et al., 2010b, Li et al., 2013), aspartic acid (Li et al., 2013), oxalic acid (Zeng et al., 2015), ascorbic acid (Li et al., 2012), gluconic and lactic acids, with hydrogen peroxide (Golmohammadzadeh et al., 2018; Roshanfar et al., 2019).

Most of the studies dealing with hydrometallurgical processes focused on the recovery of cobalt, nickel and manganese, without considering lithium. Hence, recent researches explore new approaches, able to be integrated for the recovery of lithium, to be applied at industrial scale. The recovery of lithium from leaching solution has been usually carried out by evaporation and precipitation after the recovery of other metals by solvent extraction. Generally, the lithium rich solution is treated with a saturated carbonate solution, at a temperature of 100 ºC to obtain lithium carbonate after filtration and washing. The recovery efficiency of lithium has been reported to be low (70%-80%) as a consequence of metal losses during all the previous separation phases (Nan et al., 2005; Zhang et al., 1998; Zhu et al., 2012; Zou et al., 2013). With the aim of increasing selective lithium recovery, research efforts have been made. Paulino et al. proposed adding a KF solution after removal of carbon to obtain high purity grade LiF. They achieved recovery of lithium salts, fluoride and phosphate, close to 90% (Paulino et al., 2008). Although lithium recovery was higher than recovery as carbonate and both lithium salts present commercial value, treatment of mixture salts to obtain single final products was required. The synthesis of Li-mixed oxides for manufacturing new cathodic materials from spent lithium batteries has been also recently explored (Gratz et al., 2014; Li et al., 2018). Recovering of Na_2SO_4 before Li_2CO_3 precipitation as an optimized hydrometallurgical route has been confirmed a promising method to remove impurities allowing recovery of lithium carbonate with a purity of 99.8 % (Abo et al., 2019).

Table 4: State-of-the-art of lithium recovery processes

Reference	**Reagents**	**Metal recovery efficiency**
Roshanfar et al. 2019	Lactic acid, H_2O_2	97% for Co and 100% for Li
Abo et al. 2019	H_2SO_4, H_2O_2	98% for Co, 99% for Li and 96% for Mn
Li et al. 2018	Citric acid, H_2O_2	99% for Co and Li and 95% for Mn
Gao et al. 2018	HCl, ascorbic acid	99% for Co and Li
Jiang et al. 2018	H_2SO_4, H_2O_2	95% for Co and 98% for Li
Barik et al. 2017	HCl	99% for Co, Li and Mn
Pagnanelli et al. 2017	H_2SO_4, glucose	99% for Co and Li
Zeng et al. 2014	Oxalic acid	97% for Co and 98% for Li
Meshram et al. 2015	H_2SO_4, $NaHSO_3$	91% for Co, 97% for Li and 88% for Mn
Gratz et al. 2014	H_2SO_4, H_2O_2	94% for Co, 90% for Li and 91% for Mn
Joulié et al., 2014	HCl	80% for Co and Li
Li et al., 2013	Citric acid, malic acid, aspartic acid, H_2O_2	90% for Co and Li
Georgi-Maschler et al., 2012	H_2SO_4	90% for Li

Li et al. 2012	Ascorbic acid	95% for Co and 98% for Li
Zhu et al. 2012	H_2SO_4, H_2O_2	96% for Co and 87% for Li
Sun and Qui 2011	H_2SO_4	99% for Co and Li
Li et al. 2010a	Citric acid, H_2O_2	90% for Co and 99% for Li
Li et al. 2010b	Malic acid, H_2O_2	90% for Co and 99% for Li
Ferreira et al. 2009	H_2SO_4, H_2O_2	97% for Co and 99% for Li
Paulino et al. 2008	H_2SO_4, H_2O_2	99% for Co, 90% for Li and 99% for Mn
Zou, et al. 2007	H_2SO_4, H_2O_2	100% for Co and Mn and 80% for Li
Nan et al. 2005	H_2SO_4	97% for Co and Li
Shin et al. 2005	H_2SO_4, H_2O_2	100% for Co and Li
Contestabile et al. 1999	Iso-butyl alcohol/H_2O, H_2SO_4, HCl, HNO_3	N.A.
McLaughlin and Adams, 1999	H_2SO_4	97% for Li
Zhang et al. 1998	HCl	100% for Co and 99% for Li and Mn

Regarding industrial processes, many companies worldwide recycle batteries (both rechargeable and not rechargeable) with different metal targets as confirmed by Table 5. The most common approaches include pyrometallurgical treatments, where LIBs are smelted, target metals are separated, refined and lithium lost within the resulting slags, combined with other valuable elements (e.g. aluminium, calcium) (Barik et al., 2016; Yazicioglu and Tygat, 2011). The thermal choice allows the quick removal of organic compounds as carbon powder and plastics, with a negative effect due to the production of emissions to air that need stringent air filtration systems (Li et al., 2010b; Zeng et al., 2014). Inmetco facility (USA), with a capacity of 6000 tons/year, involves a rotary earth furnace treatment followed by the refining in an electric arc furnace to produce Co/Ni/Fe alloys. Similarly, SNAM company (France, annual capacity 300 tons/year) focuses on cobalt and nickel oxides from rechargeable batteries and Batrec AG (Switzerland, 200 tons/year) obtains iron, manganese, zinc and mercury from not rechargeable batteries (Barik et al., 2016; Lv et al., 2018; Talens Peiró et al., 2013). The combination of pyrometallurgical approach with hydrometallurgical techniques, has been necessary for lithium enhancement, as reported for Accurec GmBH (Germany, 6000 tons/year) and Umicore (Belgium, 7000 tons/year) facilities (Lebedeva et al., 2016; Lv et al., 2018; Pellow et al., 2020; Raugei and Winfield, 2019; Reuter et al., 2014). Umicore pyrometallurgical process consists of the direct melting of the spent LIBs, combined with slag formers, using a unique ultra-high temperature furnace. The system is divided into three temperature zones: the pre-heating zone (< 300°C for electrolyte removal), the plastic pyrolyzing zone (around 700°C) and the smelting and reaction zone (1200-1450°C), with a substantial energy request (Liu et al., 2019). The resulting slags need additional chemical reactions for lithium extraction. As aforementioned, TOXCO (Canada, with a 4500 tons/year capacity), is one of the most relevant hydrometallurgical method focused on lithium recovery from batteries and cells made of lithium. With the aim of reducing the reactivity of lithium, scraps are firstly placed in liquid nitrogen before crushing. By using lithium hydroxide to control pH in the reaction of lithium containing materials with water, a variety of lithium salts are formed. The salts are subsequently refined using H_2SO_4 and pass through a membrane into a basic solution to form LiOH. Finally, lithium carbonated is formed by the addition of CO_2. This process, characterized by a recovery efficiency of lithium close to 97%, allows the reuse of lithium in the production of new batteries (McLaughlin and Adams, 1999). The Recupyl process (France with an annual capacity of 110 tons/year), also devoted to the recycling of rechargeable and not rechargeable lithium batteries, consists of a hydrometallurgical method to recover lithium. The batteries scraps are submitted to dry crushing, at room temperature, in an inert atmosphere to avoid safety issues related to remaining charge batteries. By magnetic and density separation, a rich fraction in metal is obtained to be submitted to hydrolysis process. Lithium contained in the finest fractions reacts with water producing a rich lithium solution which is subsequently treated with sodium carbonate and phosphoric acid allowing the recovery of lithium as carbonate and phosphate (Tedjar and Foudraz, 2007). Overall, in addition to the economic advantage for both the lithium recovery and the lowest energy demand, the hydrometallurgical processes show the strength of the adaptability to the LIB technology changes (Georgi-Maschler et al., 2012; Pagnanelli et al., 2016; Sun and Qiu, 2011; Wang et al., 2014a).

Table 5: Industrial processes for battery recycling worldwide (Barik et al. 2016, Lv et al. 2018, Liu et al. 2019, Lebedeva et al. 2016, Talens Peiró et al. 2013, Pellow et al. 2020, Reuter et al. 2014)

Company	**Country**	**Process**	**Target metals**
Inmetco	USA	Pyrometallurgy	Co, Fe and Ni

SNAM	France	Pyrometallurgy	Co and Ni
Batrec AG	Switzerland	Pyrometallurgy	Fe, Mn, Zn and Hg
Accurec GmBH	Germany	Pyrometallurgy/ Hydrometallugy	Co and Li
Umicore	Belgium	Pyrometallurgy/ Hydrometallugy	In
TOXCO	Canada	Hydrometallurgy	Co and In
Recupyl	France	Hydrometallurgy	Co and In

6. The sustainability of lithium production

6.1 Description of scenarios and main assumptions

Considering the relevance of lithium for the market, combined with the availability of high quantity of waste batteries, the present section aims at the assessment of the possible environmental advantage resulting from the secondary resources. Figure 7 compares data related to lithium flows on the European territory in 2017 (including, import, production, export and consumption) with a more complex scenario, where the primary lithium production (essential to respond to the market request) has been integrated with a secondary production, through the exploitation of waste batteries (both rechargeable and not) (European Commission, 2020d). The carbon footprint has been assessed for both scenarios using the LCA approach (Figure 7). The study has been carried out by the thinkstep GaBi software-System and Database for Life Cycle Engineering (compilation 7.3.3.153; DB version 6.115), used for the production processes of energy and raw materials and the quantification of the carbon footprint of the treatments, following the recommendation of ISO 14040:2006 and 14044:2006 norms.

Figure 8 shows the system boundaries. It can be observed that current scenario refers to a complete primary production, where brine has been selected as resource, since it is considered the most relevant reserve (Figure 4). The transport has been included within system boundaries considering a first transfer using rail, from Salar de Atacama to Antofagasta, where Sociedad Quimica y Minera de Chile (the largest Chilean producer of lithium carbonate with 99.5% purity) is located (Tran and Luong, 2015). For the consequent transportation from Antofagasta port to Europe, Rotterdam has been chosen since it is the largest European port, through a route of 13,000 km (Marine traffic, 2020; Searates, 2020; Searoutes.com, 2020). The distance within the European territory has been excluded from the system boundaries because it depends on the final metal use and it is comparable for both scenarios. The Li_2CO_3 production process from concentrated lithium brine has been considered following the details described by Dunn et al. (Dunn et al., 2012). An additional assumption is that the brine fed to the plant is concentrated until 60,000 ppm of lithium, by evaporating salty water using solar energy (Talens Peiró et al., 2013). The ideal scenario includes the production of about 20% of the European lithium demand through recycling. More in details, a quantity of about 90 tons has been considered for the not rechargeable batteries and 780 tons for rechargeable, following the data reported in both Raw Materials Information System (RMSI) and Urban mine platform (European Commission, 2020d; Urban Mine Platform.eu, 2020), for the reference year 2017. The waste collection impact has been excluded from the assessment and the hydrometallurgical route has been considered for Li recovery. As not rechargeable system, the Li/MnO_2 has been selected as the most representative chemistry. The most used LIBs cathode, $LiCoO_2$, has been taken as reference to assess the lithium recovery from spent rechargeable lithium batteries. The composition for LCO cathode has been obtained as the average composition for cathodes from seven manufacturers (Wang et al., 2014b). As concerns the hydrometallurgical route, we have considered the technologies developed within two EU funded project: the LIFE ENVIRONMENT Libat project (European Commission CORDIS, 2018) for not rechargeable lithium batteries and the FP7 HydroWEEE project (European Commission, 2009) for rechargeable batteries. Both treatments produce lithium in carbonate form, comparable to the primary metal from brines. This information is important to hypothesize the product use in new battery manufacturing. A lithium content of 4.0% and 2.5% has been assumed for not rechargeable and rechargeable batteries, respectively. As reported in Figure 8, the exploitation of batteries produces further valuable metals, mainly cobalt from LIBs (11 kg of cobalt for each kg of lithium) and manganese from not rechargeable (6 kg of manganese for each kg of lithium). In order to include this relevant added value, the impact of each treatment has been allocated adopting a mass allocation criterion (Iannicelli-Zubiani et al., 2016). Considering the variability of the distances included in the ideal scenarios, from the collection site to the treatment facilities, the transport has been excluded from the first analysis and introduced in the further sensitivity analysis, taking into account the environmental load of an articulated lorry with a payload of 27 tons.

6.2 Carbon footprint assessment

The substitution of about 30% of primary lithium by recycled lithium has the potential to reduce the carbon footprint associated to the lithium supply (Figure 9a). This environmental advantage, around 10%, has been evaluated considering the collected batteries on the European territory in 2017 (European Commission, 2020d; Urban Mine Platform.eu, 2020). Nevertheless, the European Commission estimated an average collection efficiency of batteries around 45%, that should reach at least 57% by 2030 (European Commission, 2019b). This percentage should further increase combining more stringent regulations with technological collection systems. Furthermore, considering the average lifetime of many kinds of equipment which use batteries, the available quantity to manage in Europe could increase in next years. In this regard, data reported by RMSI database (European Commission, 2020d), describe an increase of waste batteries higher than 33% from 2017 to 2020. As a whole, all these aspects will further enhance the potential environmental gain of the ideal scenario, where primary lithium is partially replaced by secondary lithium. Considering the variability of many assumptions included in the ideal scenario, a sensitivity analysis was carried out through a Monte Carlo method implemented in RStudio software: 5000 simulations allowed to evaluate the possible variations of the whole impact of the ideal scenario. More in details, two variables have been selected for the analysis: the potential secondary lithium contribution, compared to the primary lithium (not completely replaceable) and the transportation distance from the collection site to the recycling facility. The secondary lithium contribution was evaluated as reported in Eq. 4:

$$Secondary\ Li\ contribution = \frac{Li\ from\ not\ rechargeable\ batteries + Li\ from\ rechargeable\ batteries}{Li\ demand} * 100 \quad (4)$$

This was varied on the basis of the collected waste batteries (both rechargeable and not rechargeable), from the current value to a possible doubling. This choice has been considered useful to evaluate the real effect on the whole impact. As concerns the distance, the considered interval has been between 0 (to simulate a decentralized management, with several recycling facilities on the European territory) and 1,500 km (to reproduce a centralized facility, able to treat batteries from different European countries). A further variation of the process impact of 20% allowed to consider the possibility of different hydrometallurgical treatments. Each dot reported in Figure 9b represents the carbon footprint of ideal scenario, combining different values of the two selected variables. The results exclude the impact of the current scenario, since it results higher than ideal scenario at all the considered conditions. The point cloud in Fig. 9b suggests that the centralized waste batteries treatment (high distances) represents the most sustainable choice for low percentage of secondary lithium, associated to a relatively low availability of waste. The growth of waste batteries makes the decentralized management system the lowest impact option, thanks to the reduction of the transport emissions. To quantitatively assess when the centralized waste batteries treatment is preferable to the decentralized one, another important aspect to consider is the resource of secondary lithium: either rechargeable or not rechargeable lithium batteries. Indeed, the impact of lithium recycling is different for the two resources. Consequently, a further step in the assessment has been the identification of the critical distance which defines the transition from a centralized to a decentralized management system, as the most sustainable choice. With this aim, further evaluations have been performed, including a third variable: the collected rechargeable and not rechargeable batteries ratio. Figure 10 identifies this critical distance considering three levels of secondary lithium contribution: the highest (light blue), the average (blue), and the lowest (dark blue) assessed. The distance value is variable between 1200 km (for rechargeable/not rechargeable batteries ratio of 3, Figure 10b) and 500 km (for rechargeable/not rechargeable batteries ratio of 12, Figure 10d). The extreme scenarios, evaluated as a whole contribution of not rechargeable batteries (Figure 10a) and rechargeable batteries (Figure 10e), considering the fixed metal demand, allow the evaluation of the critical distance dependency on the recycling process impact. Indeed, the highest impact of the rechargeable lithium batteries treatment, due to the further recovery of cobalt, decreases the critical distance value up to 250 km, compared to a complete not rechargeable batteries exploitation which makes a centralized management the most sustainable strategy in any case. Nevertheless, both the conditions are theoretical scenarios, not feasible in a real context in which the highest rechargeable lithium battery contribution is connected to the increase of sustainable technologies (e.g. electric cars) and a production of not rechargeable batteries will be ensured for the short lifetime of this technology, at least for the next few years.

7. Conclusions

The present review dealt with the main steps of lithium life, including its mining, application and recycling. The preliminary study of lithium applications has been essential to identify the most promising secondary resources, mainly waste batteries (for both compositions and availability). The further environmental sustainability assessment has allowed the evaluation of possible scenarios of lithium supply, implementable on the European territory. The results have aimed at matching the priority of the CRM list of 2020 to identify a sustainable and cleaner production system of lithium for the building of a competing European market.

The assessment has proved the relevance of additional aspects of the lithium recycling value chain, in addition to the production environmental load. In this regard, issues often neglected in the current scientific literature has been addressed, mainly:

- The crucial role of the waste battery collection, to make the integrated production system of primary and secondary lithium a reality. In this regard, the analysis has proved that a complete secondary production of this metal is not possible; nevertheless, a substitution of 30% of primary lithium can be hypothesised, based on the waste batteries production.

- The relevance of the planning of the recycling system. Indeed, the proposed assessment has suggested the greatest environmental sustainability of a decentralized system of small and medium enterprises of recyclers for the greatest waste quantities, especially with high contribution of rechargeable batteries.

The proposed evaluation, which could be farther improved with many market variables, represents an important tool for all the stakeholders involved in the battery recycling value chain, that often neglect the lithium recovery, favoring other elements (e.g. cobalt). Nevertheless, to make this estimation a reality, it is necessary a view evolution, able to make a waste (a problem) a resource. An efficient exploitation of waste battery could be translated in a double advantage: the growth of secondary lithium production and the decrease of waste flows to manage.

Acknowledgments

This work has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 778045. Villen-Guzman acknowledges the postdoctoral fellowship obtained from the University of Malaga.

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

References

Abo, T., Elia, G., Hahn, R., Altimari, P., Pagnanelli, F., 2019. Closed-loop hydrometallurgical treatment of end-of-life lithium ion batteries : Towards zero-waste process and metal recycling in advanced batteries 35, 220–227. https://doi.org/10.1016/j.jechem.2019.03.022.

An, L., 2019. Recycling of spent lithium-ion batteries: Processing methods and environmental impacts. Springer Nat. Switz. AG. https://doi.org/10.1007/978-3-

030-31834-5.

Aral, H., Vecchio-Sadus, A., 2008. Toxicity of lithium to humans and the environment-A literature review. Ecotoxicol. Environ. Saf. 70, 349–356. https://doi.org/10.1016/j.ecoenv.2008.02.026.

Bale, M.D., May, A. V., 1989. Processing of ores to produce tantalum and lithium 2, 299–320.

Barik, S.P., Prabaharan, G., Kumar, L., 2017. Leaching and separation of Co and Mn from electrode materials of spent lithium-ion batteries using hydrochloric acid : Laboratory and pilot scale study. J. Clean. Prod. 147, 37–43. https://doi.org/10.1016/j.jclepro.2017.01.095.

Barik, S.P., Prabaharan, G., Kumar, B., 2016. An innovative approach to recover the metal values from spent lithium-ion batteries. Waste Manag. 51, 222–226. https://doi.org/10.1016/j.wasman.2015.11.004.

Blengini, G.A.A., Blagoeva, D., Dewulf, J., Others, A., Others, 2017. JRC Technical reports - Assessment of the methodology for establishing the EU list of critical Raw Materials - Annexes. https://doi.org/10.2760/73303.

Brandt, K., 1994. Historical development of secondary lithium batteries. Solid State Ionics 69, 173–183.

Brito, G.M., Chicon, M.B., Coelho, E.R.C., Faria, D.N., Freitas, J.C.C., 2020. Eco-green biodiesel production from domestic waste cooking oil by transesterification using LiOH into basic catalysts mixtures. J. Renew. Sustain. Energy 12. https://doi.org/10.1063/5.0005625.

Buijs, B., Sievers, H., Espinoza, T., 2012. Limits to the critical raw materials approach. Waste Resour. Manag. https://doi.org/DOI: 10.1680/warm.12.00010.

Chen, G., Kong, R., Wang, Y., 2020. Research on the evolution of lithium trade communities based on the complex network. Physica A 540, 123002. https://doi.org/10.1016/j.physa.2019.123002.

Chen, X., Ma, H., Luo, C., Zhou, T., 2017. Recovery of valuable metals from waste cathode materials of spent lithium-ion batteries using mild phosphoric acid. J. Hazard. Mater. 326, 77–86. https://doi.org/10.1016/j.jhazmat.2016.12.021.

Chen, Z., Belharouak, I., Sun, Y.-K., Amine, K., 2012. Titanium- based anode materials for safe lithium- ion batteries. Adv. Funct. Mater. https://doi.org/https://doi.org/10.1002/adfm.201200698.

Choubey, P.K., Kim, M., Srivastava, R.R., Lee, J.C., Lee, J.Y., 2016. Advance review on the exploitation of the prominent energy-storage element : Lithium . Part I : From mineral and brine resources. Miner. Eng. 89, 119–137. https://doi.org/10.1016/j.mineng.2016.01.010.

Ciez, R.E., Whitacre, J.F., 2019. Examining different recycling processes for lithium-ion batteries. Nat. Sustain. 2, 148–156. https://doi.org/10.1038/s41893-019-0222-5.

Contestabile, M., Panero, S., Scrosati, B., 1999. A laboratory-scale lithium battery recycling process. J. Power Sources 83, 75–78. https://doi.org/https://doi.org/10.1016/S0378-7753(99)00261-X.

Councel, L., 1939. Strategic and critical materials stock piling act.

Diekmann, J., Hanisch, C., Froböse, L., Schälicke, G., Loellhoeffel, T., Fölster, A.S., Kwade, A., 2017. Ecological Recycling of Lithium-Ion Batteries from Electric Vehicles with Focus on Mechanical Processes. J. Electrochem. Soc. 164, A6184–A6191. https://doi.org/10.1149/2.0271701jes.

Dorella, G., Mansur, M.B., 2007. A study of the separation of cobalt from spent Li-ion battery residues. J. Power Sources 170, 210–215.

https://doi.org/10.1016/j.jpowsour.2007.04.025.

Drabik, E., Rizos, V., 2018. Prospects for electric vehicle batteries in a circular economy, Ceps.

Dunn, J.B., GAines, L., Barnes, M., Sullivan, J., Wang, M., 2012. Material and energy flows in the materials production, assembly, and end-of-life stages of the automotive lithium-ion battery life cycle.

European Commission, 2020a. Critical raw materials resilience: Charting a path towards greater security and sustainability. https://doi.org/10.1007/978-3-030-40268-6_9.

European Commission, 2020b. Changing how we produce and consume: New circular economy action plan shows the way to a climate-neutral, competitive economy of empowered 2020, 11–12.

European Commission, 2020c. A new circular economy action plan for a cleaner and more competitive Europe.

European Commission, 2020d. Raw materials information system (RMSI) [WWW Document].

European Commission, 2019a. The European Green Deal sets out how to make Europe the first climate-neutral continent by 2050, boosting the economy, improving people's health and quality of life, caring for nature, and leaving no one behind. Eur. Comm. Press Room 11–12.

European Commission, 2019b. Commission staff working document on the evaluation of the Directive 2006/66/EC on batteries and accumulators and waste batteries and accumulators and repealing Directive 91/157/EEC. Brussels.

European Commission, 2019c. Report from the Commission to the European Parliament, the Council, the European Economic and Social Committee, the

Committee of the regions and the European Investment Bank on the Implementation of the Strategic Action Plan on Batteries: Building a Strat.

European Commission, 2017a. Communication on the 2017 list of critical raw materials for the EU.

European Commission, 2017b. Methodology for establishing the EU list of critical raw materials. Publ. Off. Eur. Union 1–25. https://doi.org/10.2873/769526

European Commission, 2017c. Study on the review of the list of critical raw materials, European Comissmion. https://doi.org/10.2873/876644

European Commission, 2014a. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the regions on the review of the list of critical raw materials for the EU and the implementation of the raw material initiative. Brussels.

European Commission, 2014b. Report on critical raw materials for the EU. Report of the ad hoc working group on defining critical raw materials.

European Commission, 2011. Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the regions. Tackling the challenges in commodity markets and on raw materials. Brussels.

European Commission, 2009. HydroWEEE Project [WWW Document]. URL https://cordis.europa.eu/project/id/231962/it (accessed 4.10.20).

European Commission, 2008. Directive 2008/98/EC of the European Parliament and of the Counciul of 19 November 2008 on the waste and the repealing certain Directives.

European Commission, 2006. Directive 2006/66/ec of the European Parliament and of the Council of 6 September 2006 on batteries and accumulators and waste batteries and accumulators and repealing Directive 91/157/EEC.

European Commission CORDIS, 2018. LiBAT Project [WWW Document]. URL https://cordis.europa.eu/project/id/821226/it (accessed 4.24.20).

Fastmarkets, 2020. Fastmarkets lithium price [WWW Document]. URL https://www.fastmarkets.com/commodities/industrial-minerals/lithium-price-spotlight.

Ferreira, D.A., Prados, L.M.Z., Majuste, D., Mansur, M.B., 2009. Hydrometallurgical separation of aluminium, cobalt, copper and lithium from spent Li-ion batteries. J. Power Sources 187, 238–246. https://doi.org/10.1016/j.jpowsour.2008.10.077.

Frenzel, M., Kullik, J., Reuter, M.A., Gutzmer, J., 2017. Raw material ‘ criticality ’- sense or nonsense? https://doi.org/10.1088/1361-6463/aa5b64.

Gao, W., Liu, C., Cao, H., Zheng, X., Lin, X., Wang, H., 2018. Comprehensive evaluation on effective leaching of critical metals from spent lithium-ion batteries. Waste Manag. 75, 477–485. https://doi.org/10.1016/j.wasman.2018.02.023.

Georgi-Maschler, T., Friedrich, B., Weyhe, R., Heegn, H., Rutz, M., 2012. Development of a recycling process for Li-ion batteries. J. Power Sources 207, 173–182. https://doi.org/10.1016/j.jpowsour.2012.01.152.

Gil-Alana, L.A., Monge, M., 2019. Lithium: Production and estimated consumption. Evidence of persistence. Resour. Policy 60, 198–202. https://doi.org/10.1016/j.resourpol.2019.01.006.

Gleich, B., Achzet, B., Mayer, H., Rathgeber, A., 2013. An empirical approach to determine specific weights of driving factors for the price of commodities - A contribution to the measurement of the economic scarcity of minerals and metals.

Resour. Policy 38, 350–362. https://doi.org/10.1016/j.resourpol.2013.03.011.

Golmohammadzadeh, R., Faraji, F., Rashchi, F., 2018. Recovery of lithium and cobalt from spent lithium ion batteries (LIBs) using organic acids as leaching reagents: A review. Resour. Conserv. Recycl. 136, 418–435. https://doi.org/10.1016/j.resconrec.2018.04.024.

Goodenough, J.B., Park, K.S., 2013. The Li-ion rechargeable battery: A Perspective. J. Am. Chem. Soc. 135, 1167–1176. https://doi.org/https://doi.org/10.1021/ja3091438.

Graedel, T.E., Nuss, P., 2014. Employing considerations of criticality in product design. Jom. 66, 2360–2366. https://doi.org/10.1007/s11837-014-1188-4

Gratz, E., Sa, Q., Apelian, D., Wang, Y., 2014. A closed loop process for recycling spent lithium ion batteries. J. Power Sources. 262, 255–262. https://doi.org/10.1016/j.jpowsour.2014.03.126.

Gruber, P.W., Medina, P.A., Keoleian, G.A., Kesler, S.E., Everson, M.P., Wallington, T.J., 2011. Global lithium availability: A constraint for electric vehicles? J. Ind. Ecol. 15, 760–775. https://doi.org/10.1111/j.1530-9290.2011.00359.x.

Grützke, M., Kraft, V., Weber, W., Wendt, C., Friesen, A., Klamor, S., Winter, M., Nowak, S., 2014. Supercritical carbon dioxide extraction of lithium-ion battery electrolytes. J. Supercrit. Fluids 94, 216–222. https://doi.org/10.1016/j.supflu.2014.07.014.

Hancock, L., Ralph, N., Ali, S.H., 2018. Bolivia’s lithium frontier: Can public private partnerships deliver a minerals boom for sustainable development? J. Clean. Prod. 178, 551–560. https://doi.org/10.1016/j.jclepro.2017.12.264.

Hart, W.A., Beumel, O.F., 1970. Lithium and its compounds, The Chemistry of lithium, sodium, potassium, rubidium, cesium and francium. Pergamon Press. https://doi.org/10.1016/B978-0-08-018799-0.50005-4

Hu, J., Zhang, J., Li, H., Chen, Y., Wang, C., 2017. A promising approach for the recovery of high value-added metals from spent lithium-ion batteries. J. Power Sources 351, 192–199. https://doi.org/10.1016/j.jpowsour.2017.03.093.

Huo, H., Xing, Y., Pecht, M., Züger, B.J., Khare, N., Vezzini, A., 2017. Safety requirements for transportation of lithium batteries Haibo. Energies 10. https://doi.org/10.3390/en10060793.

Iannicelli-Zubiani, E.M., Giani, M.I., Recanati, F., Dotelli, G., Puricelli, S., Cristiani, C., 2016. Environmental impacts of a hydrometallurgical process for electronic waste treatment: A life cycle assessment case study. J. Clean. Prod. 140, 1204-1216. https://doi.org/10.1016/j.jclepro.2016.10.040.

Jiang, F., Chen, Y., Ju, S., Zhu, Q., Zhang, L., Peng, J., Wang, X., Miller, J.D., 2018. Ultrasound-assisted leaching of cobalt and lithium from spent lithium-ion batteries. Ultrason. Sonochem. 48, 88–95. https://doi.org/10.1016/j.ultsonch.2018.05.019.

Joulié, M., Laucournet, R., Billy, E., 2014. Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries. J. Power Sources. 247, 551–555. https://doi.org/10.1016/j.jpowsour.2013.08.128.

Kavanagh, L., Keohane, J., Cabellos, G.G., Lloyd, A., 2018. Global lithium sources - industrial use and future in the electric vehicle industry : A review. Resources. 7, 53. https://doi.org/10.3390/resources7030057

Kogel, J.E., Trivedi, N.C., Barker, M.J., Krukowski, S.T., 2006. Industrial minerals & rocks: Commodities, markets, and uses. Society for mining, metallurgy, and exploration, inc.

Korinek, J., Sourdin, P., 2009. Maritime transport costs and their impact on trade. Organization for Economic Co-operation and Development TAD/TC/WP, 7.

Kushnir, D., Sandén, B.A., 2012. The time dimension and lithium resource constraints for electric vehicles. Resour. Policy. 37, 93–103. https://doi.org/10.1016/j.resourpol.2011.11.003.

Lebedeva, N., Di Persio, F., Lois, B.B., 2016. Lithium ion battery value chain and related opportunities for Europe. European Commission, Petten

Lee, C.W., Tsai, S.L., Chen, M.J., 2007. Treatment and recycling of scrap lithium battery, in: Warey, P.B. (Ed.), New Research on Hazardous Materials. Nova SCience Publishers, Inc., New York, pp. 209–224.

Li, H., Eksteen, J., Kuang, G., 2019. Recovery of lithium from mineral resources: State-of-the-art and perspectives – A review. Hydrometallurgy. 189, 105129. https://doi.org/10.1016/j.hydromet.2019.105129.

Li, L., Bian, Y., Zhang, X., Guan, Y., Fan, E., Wu, F., Chen, R., 2018. Process for recycling mixed-cathode materials from spent lithium-ion batteries and kinetics of leaching. Waste Manag. 71, 362–371. https://doi.org/10.1016/j.wasman.2017.10.028.

Li, L., Dunn, J.B., Xiao, X., Gaines, L., Jie, R., Wu, F., Amine, K., 2013. Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment. J. Power Sources. 233, 180–189. https://doi.org/10.1016/j.jpowsour.2012.12.089.

Li, L., Lu, J., Ren, Y., Xiao, X., Jie, R., Wu, F., Amine, K., 2012. Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries. J. Power Sources. 218, 21–27. https://doi.org/10.1016/j.jpowsour.2012.06.068.

Li, L., Chen, R., Sun, F., Wu, F., Liu, J., 2011. Preparation of $LiCoO_2$ films from spent lithium-ion batteries by a combined recycling process. Hydrometallurgy. 108, 220–225. https://doi.org/10.1016/j.hydromet.2011.04.013.

Li, L., Ge, J., Wu, F., Chen, R., Chen, S., Wu, B., 2010a. Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant. J. Hazard. Mater. 176, 288–293. https://doi.org/10.1016/j.jhazmat.2009.11.026.

Li, L., Ge, J., Chen, R., Wu, F., Chen, S., Zhang, X., 2010b. Environmental friendly leaching reagent for cobalt and lithium recovery from spent lithium-ion batteries. Waste Manag. 30, 2615–2621. https://doi.org/10.1016/j.wasman.2010.08.008.

Linden, D., Reddy, T.B., 2002. Handbook of batteries. McGraw-Hill.

Liu, C., Lin, J., Cao, H., Zhang, Y., Sun, Z., 2019. Recycling of spent lithium-ion batteries in view of lithium recovery : A critical review. J. Clean. Prod. 228, 801–813. https://doi.org/10.1016/j.jclepro.2019.04.304.

Liu, Y., Mu, D., Li, R., Ma, Q., Zheng, R., Dai, C., 2017. Purification and characterization of reclaimed electrolytes from spent lithium-ion batteries. J. Phys. Chem. C 121, 4181–4187. https://doi.org/https://doi.org/10.1021/acs.jpcc.6b12970.

Livingstone, C., Rampes, H., 2006. Lithium: A review of its metabolic adverse effects. J. Psychopharmacol. 20, 347–355. https://doi.org/10.1177/0269881105057515.

Lv, W., Wang, Z., Cao, H., Sun, Y., Zhang, Y., Sun, Z., 2018. A critical review and analysis on the recycling of spent lithium-ion batteries. ACS Sustain. Chem. Eng. 6, 1504–1521. https://doi.org/10.1021/acssuschemeng.7b03811.

Marine traffic, 2020. Marine Traffic [WWW Document]. URL https://www.marinetraffic.com/en/ais/home/centerx:-70.406/centery:-23.647/zoom:14 (accessed 4.23.20).

Mathieux, F., Ardente, F., Bobba, S., Nuss, P., Blengini, G.A., Dias, P.A., Blagoeva, D., de Matos, C.T., Wittmer, D., Pavel, C., Hamor, T., Saveyn, H., Gawlik, B., Orveillon, G., Huygens, D., Garbarino, E., Tzimas, E., Bouraoui, F., Solar, S., 2017. Critical raw materials and the circular economy Background report. Publications Office of the European Union: Luxembourg. https://doi.org/10.2760/378123.

McKnight, R.F., Adida, M., Budge, K., Stockton, S., Goodwin, G.M., Geddes, J.R., 2012. Lithium toxicity profile: A systematic review and meta-analysis. Lancet 379, 721–728. https://doi.org/10.1016/S0140-6736(11)61516-X.

McLaughlin, W., Adams, T.S., 1999. Li reclamation process. US5888463A.

Meshram, P., Mishra, A., Abhilash, Sahu, R., 2020. Environmental impact of spent lithium ion batteries and green recycling perspectives by organic acids – A review. Chemosphere. 242, 125291. https://doi.org/10.1016/j.chemosphere.2019.125291.

Meshram, P., Pandey, B.D., Mankhand, T.R., 2015. Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching. Chem. Eng. J. 281, 418–427. https://doi.org/10.1016/j.cej.2015.06.071.

Meshram, P., Pandey, B.D., Mankhand, T.R., 2014. Extraction of lithium from primary and secondary sources by pre-treatment, leaching and separation: A comprehensive review. Hydrometallurgy. 150, 192–208. https://doi.org/10.1016/j.hydromet.2014.10.012.

Metalary, 2020. Metalary Lithium price [WWW Document]. URL https://www.metalary.com/lithium-price/.

Mining.com, 2020. Global lithium demand expected to double by 2024 [WWW Document]. URL https://www.mining.com/global-lithium-demand-expected-to-double-by-2024/#:~:text=Lithium demand%2C the mining analyst,in 2020 to 410.5 GWh (accessed 2.2.21).

Mizushima, K., Jones, P.C., Wiseman, P.J., Goodenough, J.B., 1980. Li$xCoO_2$ (0<x<-1): A new cathode material for batteries of high energy density. Mater. Res. Bull. 15, 783–789. https://doi.org/https://doi.org/10.1016/0025-5408(80)90012-4.

Mohr, S.H., Mudd, G.M., Giurco, D., 2012. Lithium resources and production: Critical assessment and global projections. Minerals. 2, 65–84. https://doi.org/10.3390/min2010065.

Munk, L.A., Jochens, H., Jennings, M., Bradley, D.C., Hynek, S.A., Godfrey, L., 2011. Origin and evolution of Li-rich brines at Clayton Valley, Nevada, USA. 11th SGA Bienn. Meet. Let's Talk, 217–219.

Nan, J., Han, D., Zuo, X., 2005. Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction. J. Power Sources. 152, 278–284. https://doi.org/10.1016/j.jpowsour.2005.03.134.

Nitta, N., Wu, F., Lee, J.T., Yushin, G., 2015. Li-ion battery materials : present and future. Biochem. Pharmacol. 18, 252–264. https://doi.org/10.1016/j.mattod.2014.10.040.

Oliveira, L., Messagie, M., Rangaraju, S., Sanfelix, J., Hernandez Rivas, M., Van Mierlo, J., 2015. Key issues of lithium-ion batteries - From resource depletion to environmental performance indicators. J. Clean. Prod. 108, 354–362. https://doi.org/10.1016/j.jclepro.2015.06.021.

Olsher, U., Izatt, R.M., Bradshaw, J.S., Dalley, N.K., 1991. Coordination chemistry of lithium ion: A crystal and molecular structure review. Chem. Rev. 91, 137–164. https://doi.org/10.1021/cr00002a003.

Ordoñez, J., Gago, E.J., Girard, A., 2016. Processes and technologies for the recycling and recovery of spent lithium-ion batteries. Renew. Sustain. Energy Rev. 60, 195–205. https://doi.org/10.1016/j.rser.2015.12.363.

Oruch, R., Elderbi, M.A., Khattab, H.A., Pryme, I.F., Lund, A., 2014. Lithium: A review of pharmacology, clinical uses, and toxicity. Eur. J. Pharmacol. 740, 464–473. https://doi.org/10.1016/j.ejphar.2014.06.042.

Pagnanelli, F., Moscardini, E., Altimari, P., Atia, T.A., Toro, L., 2017. Leaching of electrodic powders from lithium ion batteries : Optimization of operating conditions and effect of physical pretreatment for waste fraction retrieval. Waste Manag. 60, 706–715. https://doi.org/10.1016/j.wasman.2016.11.037.

Pagnanelli, F., Moscardini, E., Altimari, P., Abo Atia, T., Toro, L., 2016. Cobalt products from real waste fractions of end of life lithium ion batteries. Waste Manag. 51, 214–221. https://doi.org/10.1016/j.wasman.2015.11.003.

Paulino, F., Busnardo, G., Afonso, J.C., 2008. Recovery of valuable elements from spent Li-batteries. J. Hazard. Mater. 150, 843–849. https://doi.org/10.1016/j.jhazmat.2007.10.048.

Pellow, M.A., Ambrose, H., Mulvaney, D., Betita, R., Shaw, S., 2020. Research gaps in environmental life cycle assessments of lithium ion batteries for grid-scale stationary energy storage systems : End-of-life options and other issues. Sustain. Mater. Technol. 23, e00120. https://doi.org/10.1016/j.susmat.2019.e00120.

Petranikova, M., Tkaczyk, A.H., Bartl, A., Amato, A., Lapkovskis, V., Tunsu, C., 2020. Vanadium sustainability in the context of innovative recycling and sourcing development. Waste Manag. 113, 521-544. https://doi.org/10.1016/j.wasman.2020.04.007

Pinegar, H., Smith, Y.R., 2019. Recycling of End-of-Life Lithium Ion Batteries, Part I: Commercial Processes. J. Sustain. Metall. 5, 402-416.

https://doi.org/10.1007/s40831-019-00235-9.

Pinna, E.G., Ruiz, M.C., Ojeda, M.W., Rodriguez, M.H., 2017. Cathodes of spent Li-ion batteries: Dissolution with phosphoric acid and recovery of lithium and cobalt from leach liquors. Hydrometallurgy. 167, 66–71. https://doi.org/10.1016/j.hydromet.2016.10.024.

Raugei, M., Winfield, P., 2019. Prospective LCA of the production and EoL recycling of a novel type of Li-ion battery for electric vehicles. J. Clean. Prod. 213, 926–932. https://doi.org/10.1016/j.jclepro.2018.12.237.

Resource Trade Earth.com, 2020. Resource Trade Earth (Lithium carbonate) [WWW Document]. URL https://resourcetrade.earth/data?year=2018&category=1575&units=value (accessed 4.23.20).

Reuter, B., Riedl, J., Hamacher, T., Lienkamp, Ma., Bradshaw, A.M., 2014. Future resource availability for the production of lithium-ion vehicle batteries, in: COFAT 2014. pp. 1–17.

Richa, K., Babbitt, C.W., Gaustad, G., 2017. Eco-efficiency analysis of a lithium-ion battery waste hierarchy inspired by circular economy. J. Ind. Ecol. 21, 715–730. https://doi.org/10.1111/jiec.12607.

Rodrigue, J.P., 2020. Transport Costs, in Rodrigue, J.P., The Geography of Transport Systems FIFTH EDITION. Routledge, New York.

Roshanfar, M., Golmohammadzadeh, R., Rashchi, F., 2019. An environmentally friendly method for recovery of lithium and cobalt from spent lithium-ion batteries using gluconic and lactic acids. J. Environ. Chem. Eng. 7, 102794. https://doi.org/10.1016/j.jece.2018.11.039.

Scrosati, B., 2011. History of lithium batteries. J. Solid State Electrochem. 15, 1623–

1630. https://doi.org/10.1007/s10008-011-1386-8.

Scrosati, B., 2000. Recent advances in lithium ion battery materials. Electrochim. Acta. 45, 2461–2466. https://doi.org/10.1016/S0013-4686(00)00333-9.

Scrosati, B., Garche, J., 2010. Lithium batteries : Status , prospects and future. J. Power Sources. 195, 2419–2430. https://doi.org/10.1016/j.jpowsour.2009.11.048.

Searates, 2020. Searates.com [WWW Document]. URL https://www.searates.com/services/distances-time/ (accessed 4.24.20).

Searoutes.com, 2020. Searoutes.com [WWW Document]. URL https://www.searoutes.com/port-distances-table?name=Antofagasta&locode=CLANF.

Shaw-Stewart, J., Alvarez-Reguera, A., Greszta, A., Marco, J., Masood, M., Sommerville, R., Kendrick, E., 2019. Aqueous solution discharge of cylindrical lithium-ion cells. Sustain. Mater. Technol. 22, e00110. https://doi.org/10.1016/j.susmat.2019.e00110.

Shin, S.M., Kim, N.H., Sohn, J.S., Yang, D.H., Kim, Y.H., 2005. Development of a metal recovery process from Li-ion battery wastes. Hydrometallurgy. 79, 172–181. https://doi.org/10.1016/j.hydromet.2005.06.004.

Sonoc, A., Jeswiet, J., Kie, V., 2015. Opportunities to improve recycling of automotive lithium ion batteries. Procedia CIRP. 29, 752–757. https://doi.org/10.1016/j.procir.2015.02.039.

Stamp, A., Lang, D.J., Wäger, P.A., 2012. Environmental impacts of a transition toward e-mobility: The present and future role of lithium carbonate production. J. Clean. Prod. 23, 104–112. https://doi.org/10.1016/j.jclepro.2011.10.026.

Statista, 2020. Lithium mine production worldwide from 2010 to 2019 [WWW Document]. URL https://www.statista.com/statistics/606684/world-production-of-lithium/ (accessed 2.2.21).

Sun, X., Hao, H., Hartmann, P., Liu, Z., Zhao, F., 2019. Supply risks of lithium-ion battery materials : An entire supply chain estimation. Mater. Today Energy. 14, 100347. https://doi.org/10.1016/j.mtener.2019.100347.

Sun, L., Qiu, K., 2011. Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries. J. Hazard. Mater. 194, 378–384. https://doi.org/10.1016/j.jhazmat.2011.07.114.

Swain, B., 2017. Recovery and recycling of lithium: A review. Sep. Purif. Technol. 172, 388–403. https://doi.org/10.1016/j.seppur.2016.08.031.

Tadesse, B., Makuei, F., Albijanic, B., Dyer, L., 2019. The beneficiation of lithium minerals from hard rock ores: A review. Miner. Eng. 131, 170–184. https://doi.org/10.1016/j.mineng.2018.11.023.

Talens Peiró, L., Villalba Méndez, G., Ayres, R.U., 2013. Lithium: Sources, production, uses, and recovery outlook. Jom. 65, 986–996. https://doi.org/10.1007/s11837-013-0666-4.

Tarascon, J.M., 2010. Is lithium the new gold? Nat. Chem. 2, 510-510. https://doi.org/10.1038/nchem.680.

Tedjar, F., Foudraz, J.C., 2007. Method for the mixed recycling of Lithium-based anode batteries and cells. US 2007/0196725 A1.

Tkaczyk, A.H., Bartl, A., Amato, A., Lapkovskis, V., Petranikova, M., 2018. Sustainability evaluation of essential critical raw materials: cobalt, niobium, tungsten and rare earth elements. J. Phys. D. Appl. Phys. 51, 203001. https://doi.org/10.1088/1361-6463/aaba99.

Tran, T., Luong, V.T., 2015. Lithium production processes, lithium process chemistry. Elsevier Inc. https://doi.org/10.1016/b978-0-12-801417-2.00003-7.

TrendEconomy, 2019. World Merchandise Exports and Imports by Commodity (lithium carbonate) [WWW Document]. URL https://trendeconomy.com/data/commodity_h2?commodity=283691.

Urban Mine Platform.eu [WWW Document], 2020. URL www.urbanmineplatform.eu/urbanmine/batteries/quantity (accessed 4.24.20).

USGS, 2019. Mineral Commodity Summaries 2020 (Lithium).

Vikström, H., Davidsson, S., Höök, M., 2013. Lithium availability and future production outlooks. Appl. Energy. 110, 252–266. https://doi.org/10.1016/j.apenergy.2013.04.005.

Wang, F., Zhang, T., He, Y., Zhao, Y., Wang, S., 2018. Recovery of valuable materials from spent lithium-ion batteries by mechanical separation and thermal treatment. J. Clean. Prod. 185, 646–652. https://doi.org/10.1016/j.jclepro.2018.03.069.

Wang, M., Zhang, C., Zhang, F., 2017. Recycling of spent lithium-ion battery with polyvinyl chloride by mechanochemical process. Waste Manag. 67, 232–239. https://doi.org/10.1016/j.wasman.2017.05.013.

Wang, Y., Liu, B., Li, Q., Cartmell, S., Ferrara, S., Deng, Z.D., Xiao, J., 2015. Lithium and lithium ion batteries for applications in microelectronic devices: A review. J. Power Sources. 286, 330–345. https://doi.org/10.1016/j.jpowsour.2015.03.164.

Wang, X., Gaustad, G., Babbitt, C.W., Bailey, C., Ganter, M.J., Landi, B.J., 2014a. Economic and environmental characterization of an evolving Li-ion battery waste stream. J. Environ. Manage. 135, 126–134. https://doi.org/10.1016/j.jenvman.2014.01.021.

Wang, X., Gaustad, G., Babbitt, C.W., Richa, K., 2014b. Economies of scale for future lithium-ion battery recycling infrastructure. Resour. Conserv. Recycl. 83, 53–62. https://doi.org/10.1016/j.resconrec.2013.11.009.

Wang, C.S., Wu, G.T., Li, W.Z., 1998. Lithium insertion in ball-milled graphite. J. Power Sources. 76, 1–10. https://doi.org/10.1016/S0378-7753(98)00114-1.

Webmineral.com, 2020. Mineral Species containing Lithium (Li) [WWW Document]. URL http://webmineral.com/chem/Chem-Li.shtml#.XoiHCIgzZPb (accessed 4.23.20).

Xu, J., Thomas, H.R., Francis, R.W., Lum, K.R., Wang, J., Liang, B., 2008. A review of processes and technologies for the recycling of lithium-ion secondary batteries. J. Power Sources. 177, 512–527. https://doi.org/10.1016/j.jpowsour.2007.11.074.

Yao, L.P., Zeng, Q., Qi, T., Li, J., 2020. An environmentally friendly discharge technology to pretreat spent lithium-ion batteries. J. Clean. Prod. 245, 118820. https://doi.org/10.1016/j.jclepro.2019.118820.

Yazicioglu, B., Tygat, J., 2011. Life Cycle Assessments involving Umicore's Battery Recycling process, in: DG Environment - Stakeholder Meeting.

Young, W., 2009. Review of lithium effects on brain and blood. Cell Transplant. 18, 951–975. https://doi.org/10.3727/096368909X471251.

Zeng, X., Li, J., Shen, B., 2015. Novel approach to recover cobalt and lithium from spent lithium-ion battery using oxalic acid. J. Hazard. Mater. 295, 112–118. https://doi.org/10.1016/j.jhazmat.2015.02.064.

Zeng, X., Li, J., Singh, N., 2014. Recycling of spent lithium-ion battery: A critical review. Crit. Rev. Environ. Sci. Technol. 44, 1129–1165. https://doi.org/10.1080/10643389.2013.763578.

Zhang, P., Yokoyama, T., Itabashi, O., Wakui, Y., Suzuki, T.M., Inoue, K., 1998.

Hydrometallurgical process for recovery of metal values from spent nickel-metal hydride secondary batteries. Hydrometallurgy. 50, 61–75. https://doi.org/10.1016/S0304-386X(98)00046-2.

Zhu, S.G., He, W.Z., Li, G.M., Zhou, X., Zhang, X.J., Huang, J.W., 2012. Recovery of Co and Li from spent lithium-ion batteries by combination method of acid leaching and chemical precipitation. Trans. Nonferrous Met. Soc. China (English Ed.). 22, 2274–2281. https://doi.org/10.1016/S1003-6326(11)61460-X.

Ziemann, S., Weil, M., Schebek, L., 2012. Tracing the fate of lithium — The development of a material flow model. Resour. Conserv. Recycl. 63, 26–34. https://doi.org/10.1016/j.resconrec.2012.04.002.

Zou, H., Gratz, E., Apelian, D., Wang, Y., 2013. A novel method to recycle mixed cathode materials for lithium ion batteries. Green Chem. 15, 1183-1191. https://doi.org/https://doi.org/10.1039/C3GC40182K.

Highlights (for review)

- The versatility and the low substitutability make lithium essential for the market
- The production of batteries represents the most relevant use of lithium
- Waste batteries represent an important secondary source of lithium
- The substitution of 30% of primary lithium increases the metal supply sustainability
- A decentralized waste management is the lowest impact choice for high battery amounts

Graphical Abstract

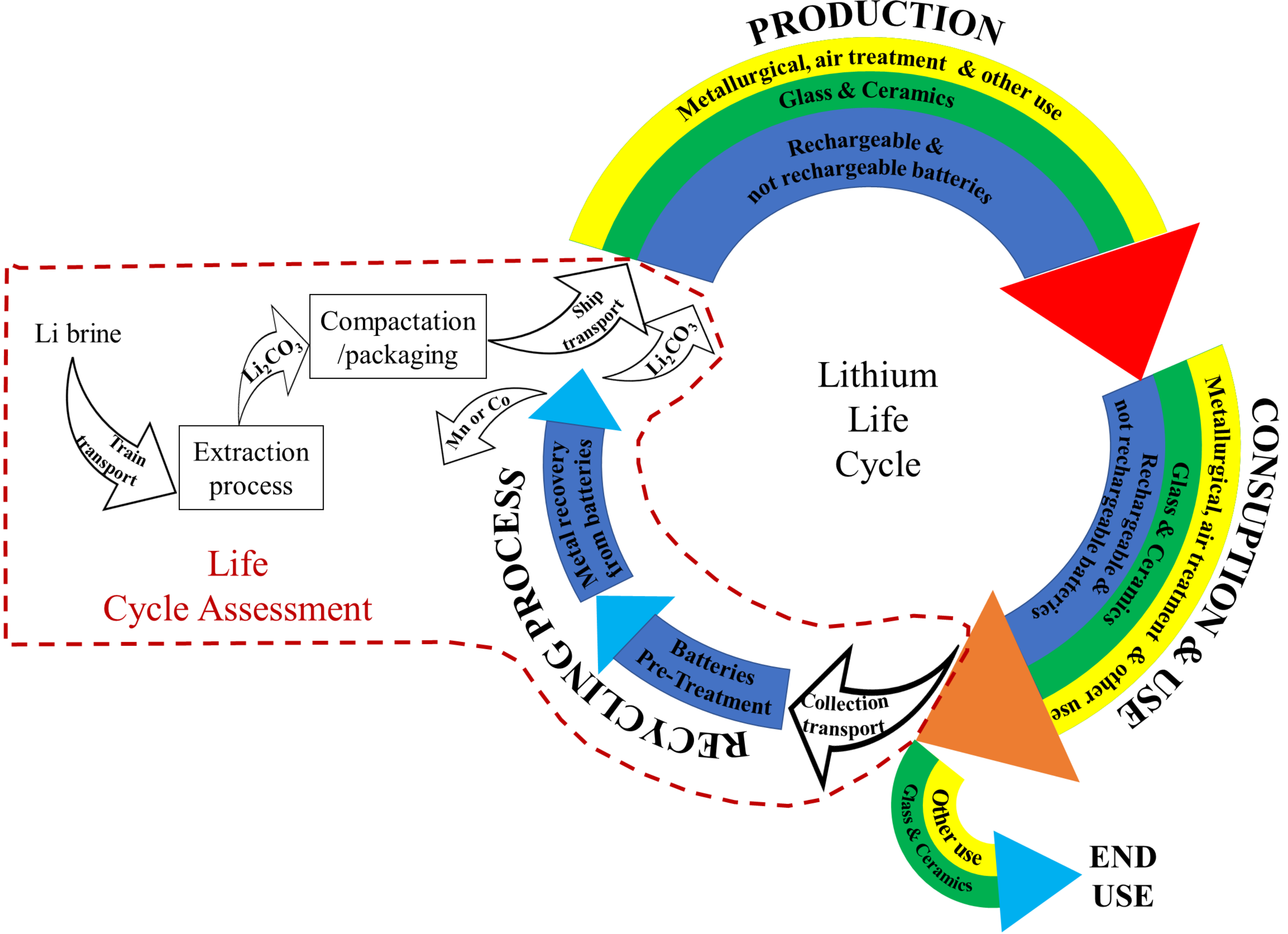


Figure 1

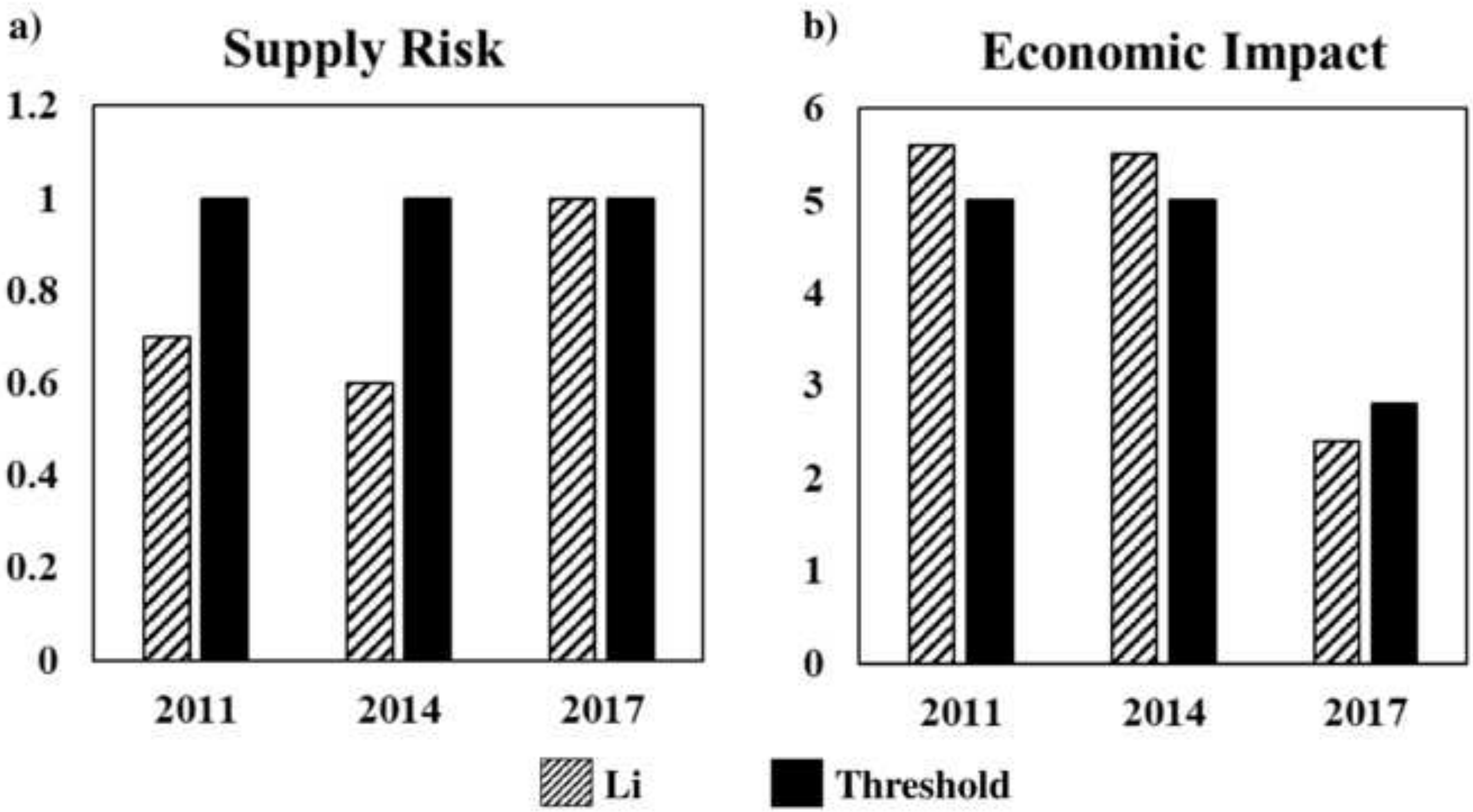
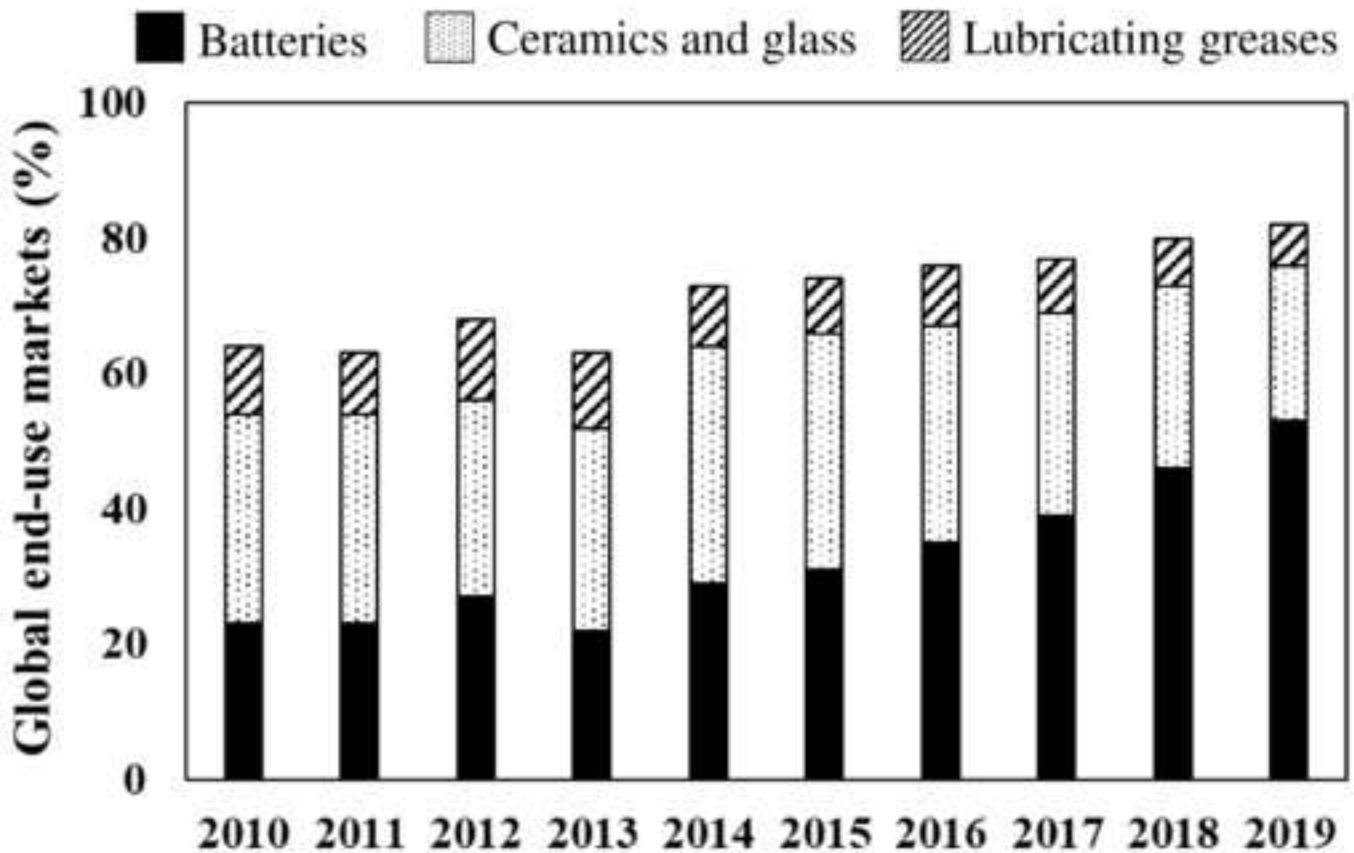
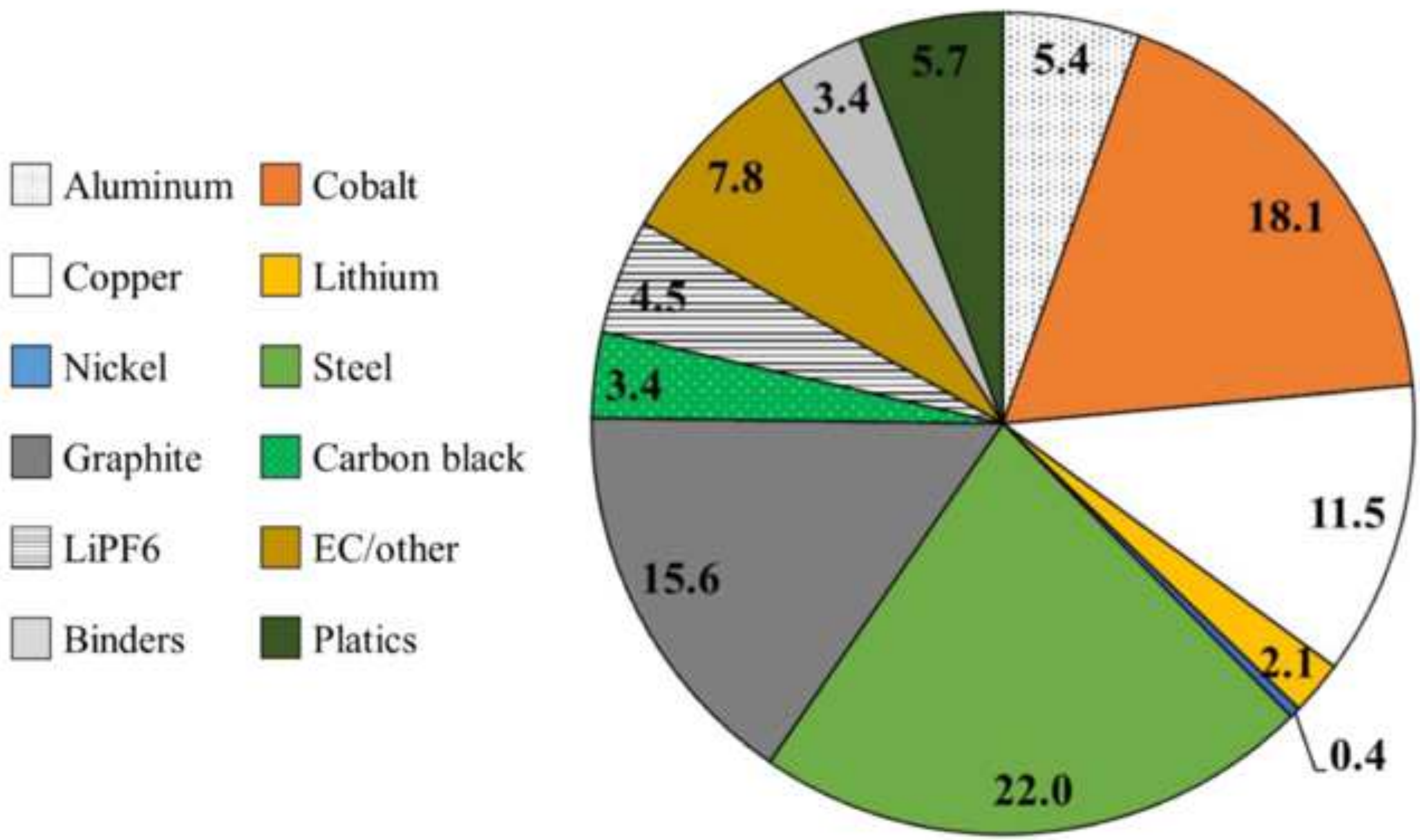

a)
Supply Risk
1.2
1
0.8
0.6
0.4
0.2
0
2011
2014
2017
b)
Economic Impact
6
5
4
3
2
1
0
2011
2014
2017
Li
Threshold

Figure 2



Batteries
Ceramics and glass
Lubricating greases
Global end-use markets (%)
0
20
40
60
80
100
2010
2011
2012
2013
2014
2015
2016
2017
2018
2019

Figure 3



Aluminum
Cobalt
Copper
Lithium
Nickel
Steel
Graphite
Carbon black
LiPF6
EC/other
Binders
Platics
5.4
18.1
11.5
2.1
0.4
22.0
15.6
3.4
4.5
7.8
3.4
5.7

Figure 4

Click here to access/download;Figure;Figure 4.tiff

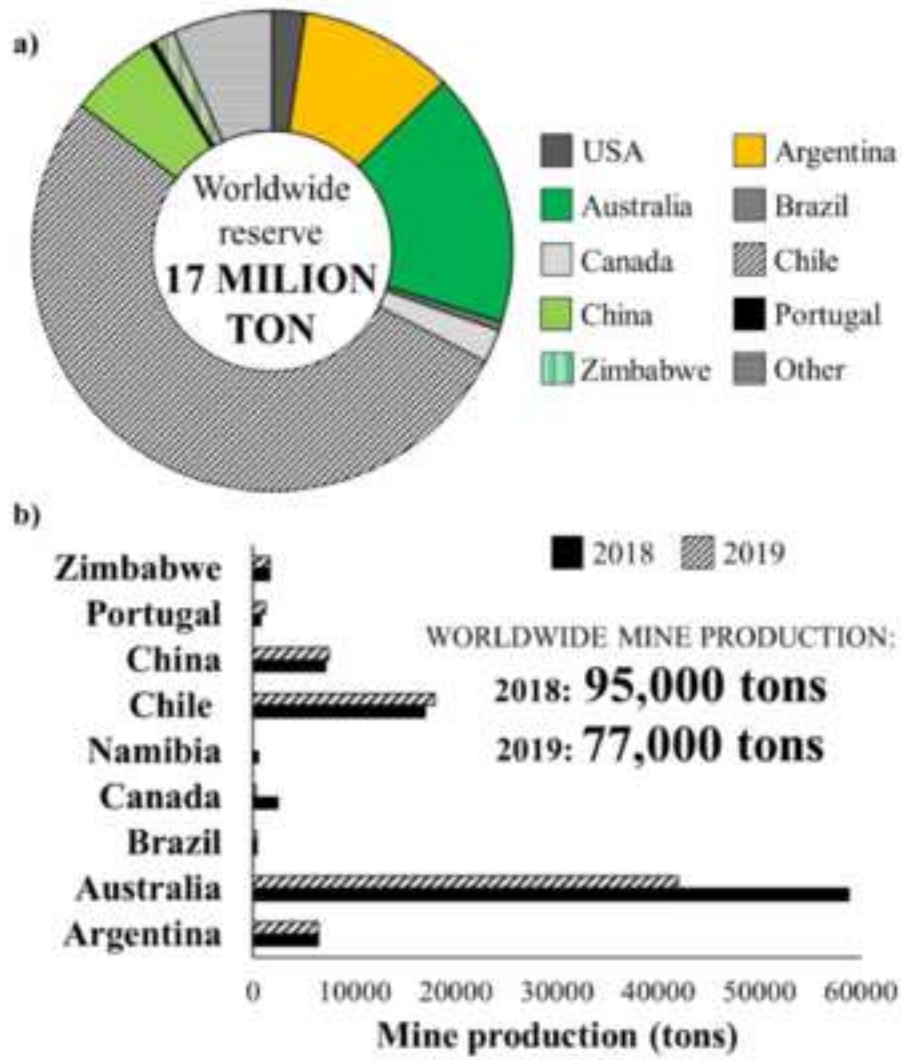


Figure 5

Click here to access/download;Figure;Figure 5.tiff

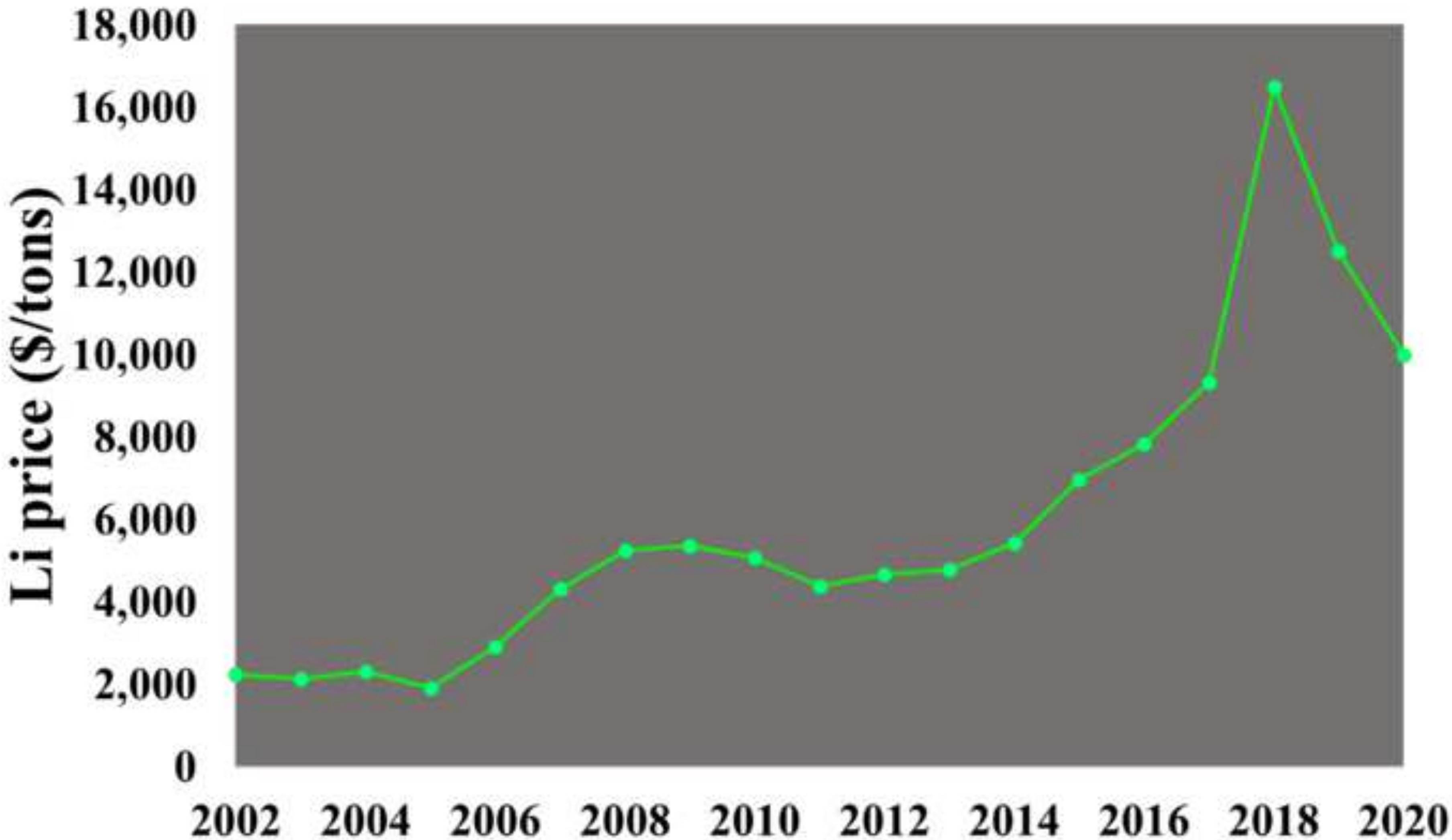


Figure 6

Click here to access/download;Figure;Figure 6.tiff

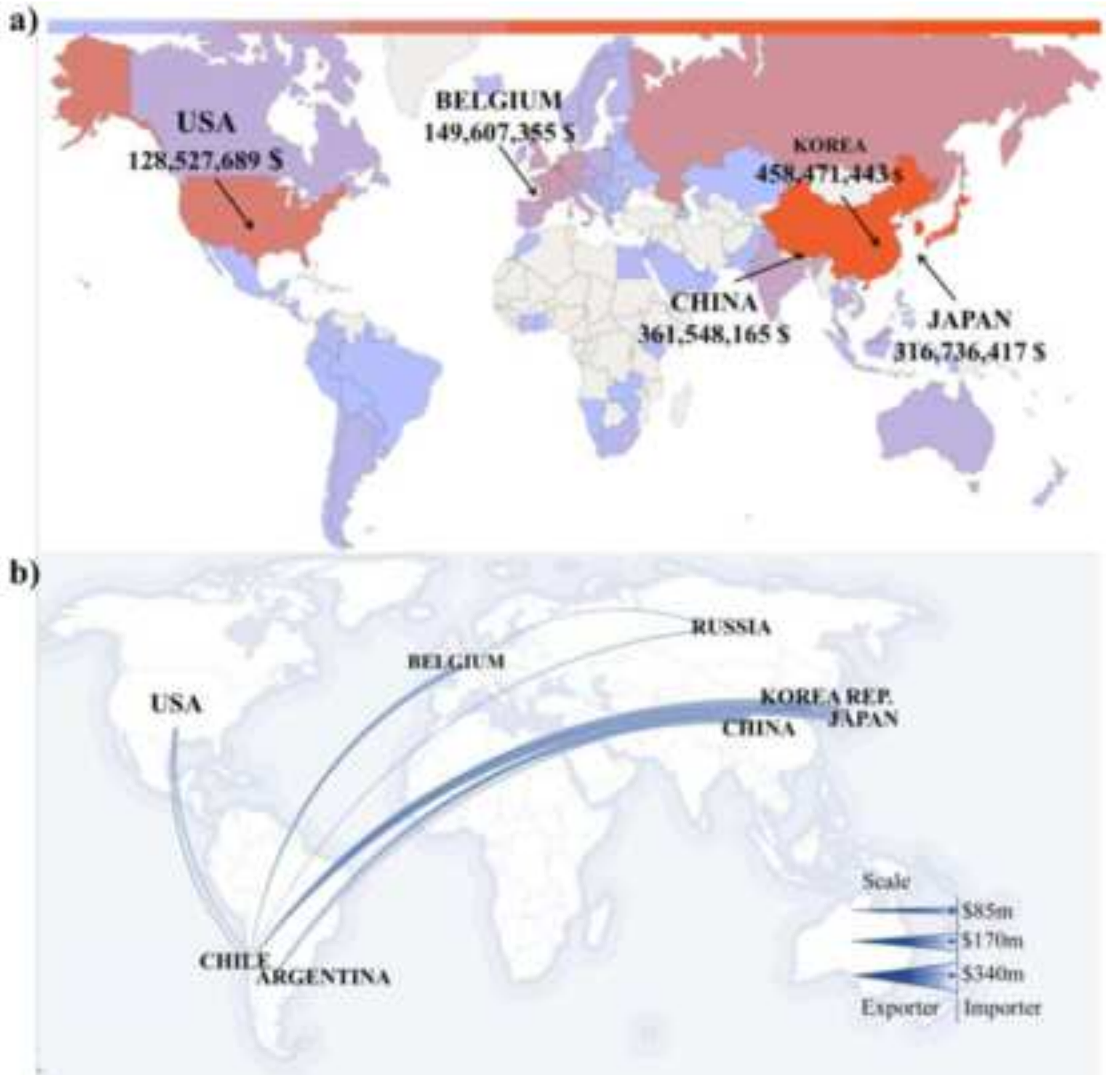


Figure 7

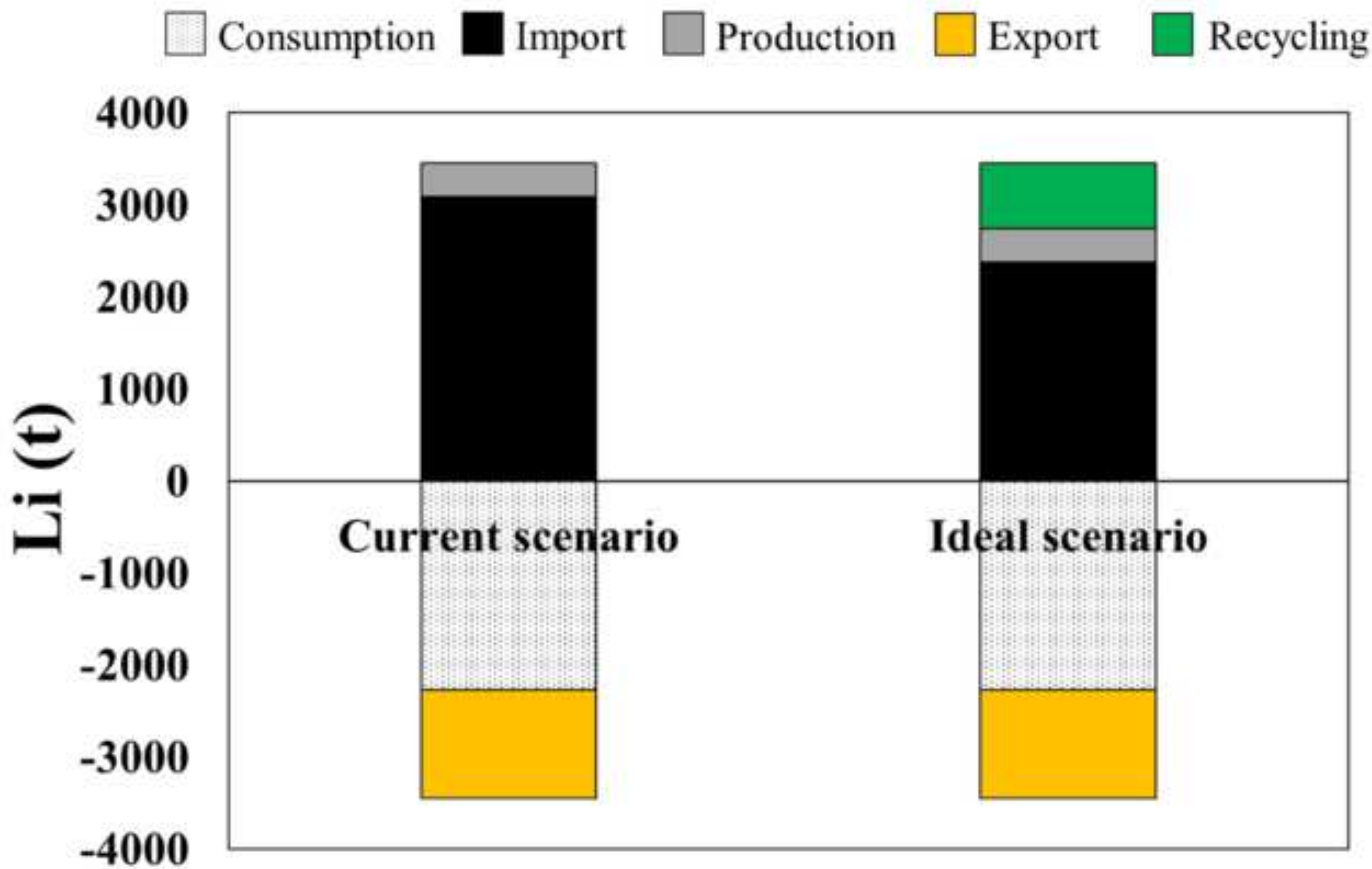
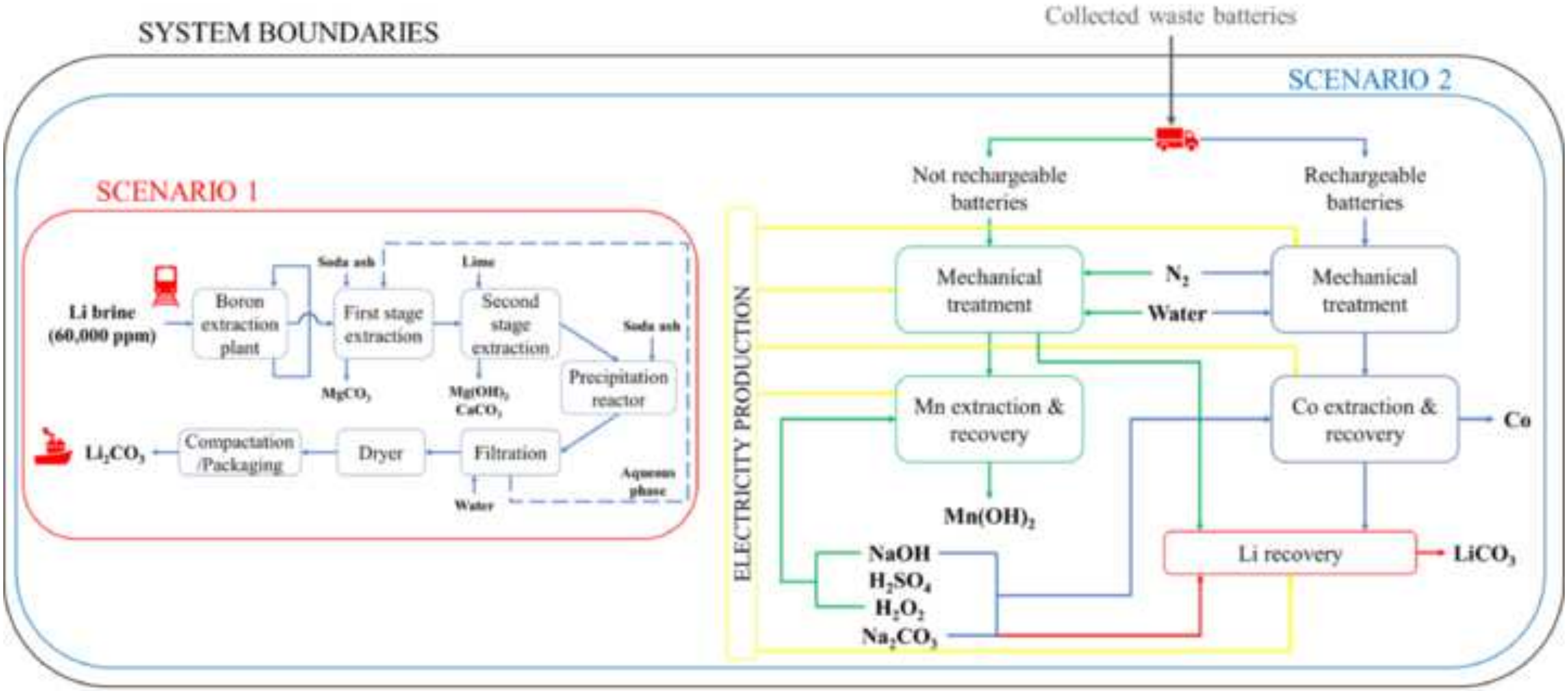

Consumption
Import
Production
Export
Recycling
4000
3000
2000
1000
0
-1000
-2000
-3000
-4000
Li (t)
Current scenario
Ideal scenario

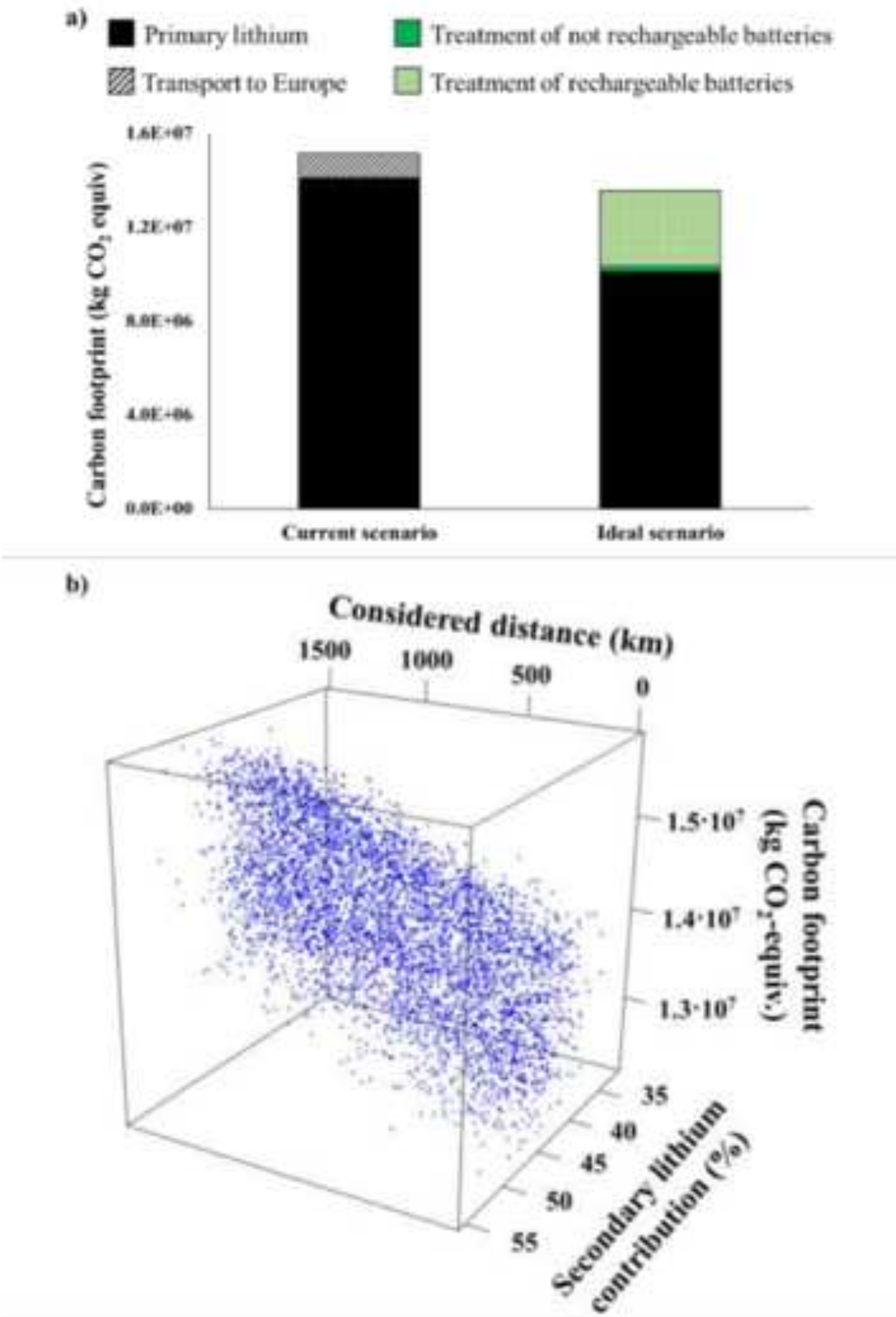
Figure 8



SYSTEM BOUNDARIES
Collected waste batteries
SCENARIO 2
SCENARIO 1
Li brine (60,000 ppm)
Boron extraction plant
Soda ash
First stage extraction
Lime
Second stage extraction
Soda ash
Precipitation reactor
$MgCO_3$
$Mg(OH)_2$ $CaCO_3$
Li_2CO_3
Compactation /Packaging
Dryer
Filtration
Aqueous phase
Water
ELECTRICITY PRODUCTION
Not rechargeable batteries
Rechargeable batteries
Mechanical treatment
N_2
Water
Mechanical treatment
Mn extraction & recovery
Co extraction & recovery
Co
$Mn(OH)_2$
NaOH
H_2SO_4
H_2O_2
Na_2CO_3
Li recovery
$LiCO_3$

Figure 9

Click here to access/download;Figure;Figure 9.tiff



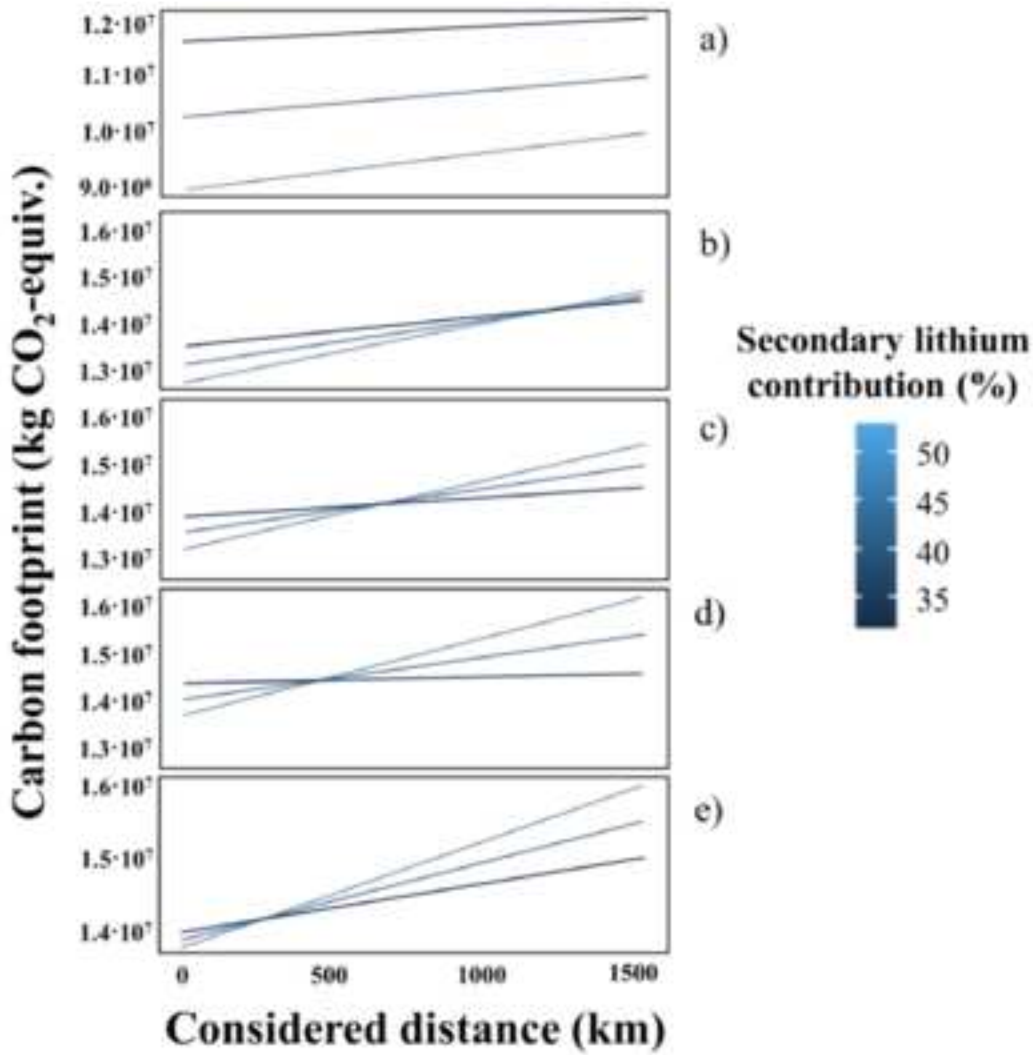

Carbon footprint (kg CO2-equiv.)
1.2·10⁷
1.1·10⁷
1.0·10⁷
9.0·10⁶
1.6·10⁷
1.5·10⁷
1.4·10⁷
1.3·10⁷
a)
b)
c)
d)
e)
0
500
1000
1500
Considered distance (km)
Secondary lithium contribution (%)
50
45
40
35

Figure 1. Assessment of lithium criticality, considering a) the supply risk and b) the economic impact (European Commission, 2017).

Figure 2. The global market for lithium: The evolution of end-use percentage for the global market for lithium from 2010 to 2019 (USGS, 2019).

Figure 3. Average material composition of LCO cathode LIBs from different manufacturers (Wang et al., 2014).

Figure 4. Role of the countries in Li reserves (a) and production (b). USA is not included in the graphic b, since the only production was from a brine production in Nevada and no data were shared (USGS, 2019).

Figure 5. Historical lithium carbonate price (adapted from Metalary.com and Fastmerkets.com (Metalary, 2020; Fastmarkets, 2020).

Figure 6. Worldwide economic flows related to lithium carbonate import (adapted from TrendEconomy.com (TrendEconomy, 2019)) (a) and export (adapted from Resource Trade Earth.com (Resource Trade Earth.com, 2020)) (b).

Figure 7. Lithium balance on the European territory in 2017 (current scenario) vs implementation of an integrated scenario able to combine primary and secondary lithium production (ideal scenario).

Figure 8. Simplified system boundaries considered for the carbon footprint assessment. Functional unit annual lithium production on European territory (reference year 2017).

Figure 9. Carbon footprint assessment: comparison between the primary production and a scenario which includes 30% of lithium secondary production from waste batteries (a), sensitivity analysis on carbon footprint of ideal scenario (impact as a function of the secondary lithium contribution and the transport distance) (b).

Figure 10. Identification of the critical distance which defines the transition from a centralized to a decentralized management system achieved by the treatment of a) not rechargeable batteries, b) LIBs/not rechargeable batteries ratio of 3, c) LIBs/not rechargeable batteries ratio of 7, d) LIBs/not rechargeable batteries ratio of 12, e) rechargeable batteries.