Eco-Efficiency Analysis of a Lithium-Ion Battery Waste Hierarchy Inspired by Circular Economy

Kirti Richa, Callie W. Babbitt, and Gabrielle Gaustad

Golisano Institute for Sustainability, Rochester Institute of Technology, Rochester, NY, USA

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Summary

A circular economy (CE)-inspired waste management hierarchy was proposed for end-of-life (EOL) lithium-ion batteries (LIBs) from electric vehicles (EVs). Life cycle eco-efficiency metrics were then applied to evaluate potential environmental and economic trade-offs that may result from managing 1,000 end-of-life EV battery packs in the United States according to this CE hierarchy. Results indicate that if technology and markets support reuse of LIBs in used EVs, the net benefit would be 200,000 megajoules of recouped cumulative energy demand, which is equivalent to avoiding the production of 11 new EV battery packs (18 kilowatt-hours each). However, these benefits are magnified almost tenfold when retired EV LIBs are cascaded in a second use for stationary energy storage, thereby replacing the need to produce and use less-efficient lead-acid batteries. Reuse and cascaded use can also provide EV owners and the utility sector with cost savings, although the magnitude of future economic benefits is uncertain, given that future prices of battery systems are still unknown. In spite of these benefits, waste policies do not currently emphasize CE strategies like reuse and cascaded use for batteries. Though loop-closing LIB recycling provides valuable metal recovery, it can prove nonprofitable if high recycling costs persist. Although much attention has been placed on landfill disposal bans for batteries, results actually indicate that direct and cascaded reuse, followed by recycling, can together reduce eco-toxicity burdens to a much greater degree than landfill bans alone. Findings underscore the importance of life cycle and eco-efficiency analysis to understand at what point in a CE hierarchy the greatest environmental benefits are accrued and identify policies and mechanisms to increase feasibility of the proposed system.

Introduction

While use of electric vehicles (EVs) can reduce dependence on fossil-based transportation fuels and may ultimately curb carbon dioxide emissions, a major concern is the management of potential wastes generated when the lithium-ion batteries (LIBs) from EVs reach their end-of-life (EOL) (Richa et al. 2014; Wang et al. 2014). Considering the projected scale of LIB deployment in EVs, a well-defined, proactive EOL management strategy is needed for these batteries. Such a strategy can be informed by *circular economy* (CE) principles such as reuse and recycling (Ramoni and Zhang 2013). A circular or closed-loop economy aims to eliminate waste by cycling

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Address correspondence to: Callie W. Babbitt, Golisano Institute for Sustainability, Rochester Institute of Technology, 190 Lomb Memorial Drive, Rochester, NY 14618, USA. *Email*: cwbgis@rit.edu; *Web*: www.rit.edu/gis

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materials and products within the system to achieve resource and energy efficiency as well as profitability (EMF 2013; McKinsey & Company 2014; Allwood et al. 2011, 2012; Gregson et al. 2015; Ghisellini et al. 2016). Hence, both reuse and recycling propagate this concept by enabling a resilient infrastructure of LIB materials by avoiding primary metal production and reducing disposal of potentially hazardous materials.

Beyond the few companies (e.g., Retriev, Chemetall, Umicore, and Recupyl) that commercially recycle LIBs, a significant body of knowledge has explored novel technologies for recovering constituent metals at bench scale using hydrometallurgical and pyrometallurgical technologies (Espinosa et al. 2004; Xu et al. 2008; Dunn et al. 2012; Georgi-Mascheler et al. 2012; Ramoni and Zhang 2013; Hendrickson et al. 2015; Nan et al. 2005; Dorella and Mansur 2007; Swain et al. 2007; Ferreira et al. 2009; Li et al. 2009; Chen et al. 2011; Li et al. 2013, etc.). While life cycle assessment (LCA) studies confirm the environmental benefit from recovery of LIB materials (Dewulf et al. 2010; Hendrickson et al. 2015; Dunn et al. 2012, 2015; Amarakoon et al. 2013), recycling will never be 100% efficient and can only recoup a fraction of the embodied energy for materials—it does not even address energy input to battery assembly and manufacturing. Though LIB recycling could serve as an enormous source of high-value materials (Richa et al. 2014), major economic barriers to commercial EV LIB recycling also exist, particularly potential transitions away from cobalt-rich battery chemistries (where cobalt drives the economic revenue of recycling) to lower-cost chemistries (containing manganese or iron) by EV manufacturers (Wang et al. 2014; Richa et al. 2014) and use of energy-intensive pyrometallurgical recovery processes (Fisher et al. 2006).

Despite wide commercial and research attention on recycling, it may not be the first priority for LIB waste management, at least without previous consideration of battery reuse. While conventional solid waste hierarchies place reuse above recycling in order of preference, LIB reuse has been far less studied. Yet, clear benefits are promised: Studies suggest that retired EV LIBs would still have 80% of their initial capacity intact (Nagpure et al. 2011; Marano et al. 2009; Zhang et al. 2011; Hoffart 2008; Cready et al. 2003; Neubauer and Pesaran 2011), and directly recycling them without any consideration for reuse can forgo the benefit obtained from taking advantage of this remaining capacity.

Reuse of EV LIBs can theoretically have two forms—direct reuse in the application from which the battery was obtained (i.e., in EVs) and cascaded use in a different and less-demanding stationary application. LCA studies suggest that reuse of EV LIBs in stationary applications can provide environmental credits by avoiding the burden of manufacturing new battery packs for these end uses (Richa et al. 2015; Cicconi et al. 2012; Genikomsakis et al. 2013). Additionally, reuse pathways would provide economic advantages by potentially reducing upfront costs of EV adoption, providing resale value for used batteries (Viswanathan and Kintner-Meyer 2011; Neubauer and Pesaran 2011; Neubauer et al. 2012; Williams and Lipman 2010), avoiding the cost of purchasing a new battery for the reuse application, generating revenue for the utility sector (Neubauer and Pesaran 2011; Narula et al. 2011; Heymans et al. 2014; Neubauer et al. 2012; Williams and Lipman 2010; Viswanathan and Kintner-Meyer 2011; Peterson et al. 2010, etc.).

While recent literature has focused on cascaded use of LIBs in stationary applications, reuse of these batteries in automobile application has not received much attention. A recent study by Saxena and colleagues (2015) suggests that LIBs can be used well below the 80% remaining energy capacity limit for less-demanding daily travel needs of EV users or extensive battery charging infrastructure, thus hinting at automotive reuse possibilities. Even with an 80% capacity fade limit, early vehicle failure (such as significant repairs or collisions) and EVs with battery replacement later in their useful life are likely to yield LIBs with high reuse potential (Richa et al. 2014) that could hypothetically be used as replacement batteries for used EVs if technology and markets exist to support this system.

The last option for EV LIB waste management would be disposal, which is expected to create additional environmental and economic impacts. The U.S. Environmental Protection Agency (US EPA) does not consider EV LIBs to be a major threat to environmental health because they do not usually contain toxic elements like lead, mercury, or cadmium (Gaines and Cuenca 2000). However, these batteries contain metals like lithium, aluminum, cobalt, manganese, nickel, and copper, which do have the potential to leach slowly into the soil, groundwater, and surface water if not disposed properly (Kang et al. 2013; Vimmerstedt et al. 1995). Similar to the case of electronic waste (Williams et al. 2008), risk of material leaching in well-managed sanitary landfills may be negligible, but the greater risk is loss of valuable materials, and the economic benefits of the reuse and recycling sectors should still avoid landfill of EV LIBs to promote a CE.

To delay or avoid disposal flows and tap into potential benefits of reuse and recycling routes, the priority for managing the EV LIB waste stream is likely to follow a *waste management hierarchy*. The European Union (EU) Waste Framework Directive and the US EPA advocate this circular thinking with a waste management hierarchy of prevention, reuse, recycling, other recovery (i.e., energy recovery), and disposal (from increasing to decreasing preference) (European Parliament 2008; US EPA 2015b). In general, such a hierarchy depicts priorities from an environmental, and sometimes economic, perspective for a variety of waste streams such as EOL electronics (Brandstotter et al. 2004), food waste (US EPA 2016; Papargyropoulou et al. 2014; Glew et al. 2013), municipal solid waste (MSW) (Cleary 2009), packaging (Rossi et al. 2015), and construction waste (Batayneh et al. 2007), etc., but its validity is yet to be analyzed for LIBs.

Using concepts from the CE, a theoretical waste management hierarchy has been proposed here for EOL EV LIBs (figure 1) that includes reuse in EVs, cascaded use in stationary application, recycling, and, finally, landfill. A case study was developed to examine the feasibility of this framework and to determine whether the proposed approach, within expected technical limits, will lead to improved environmental and

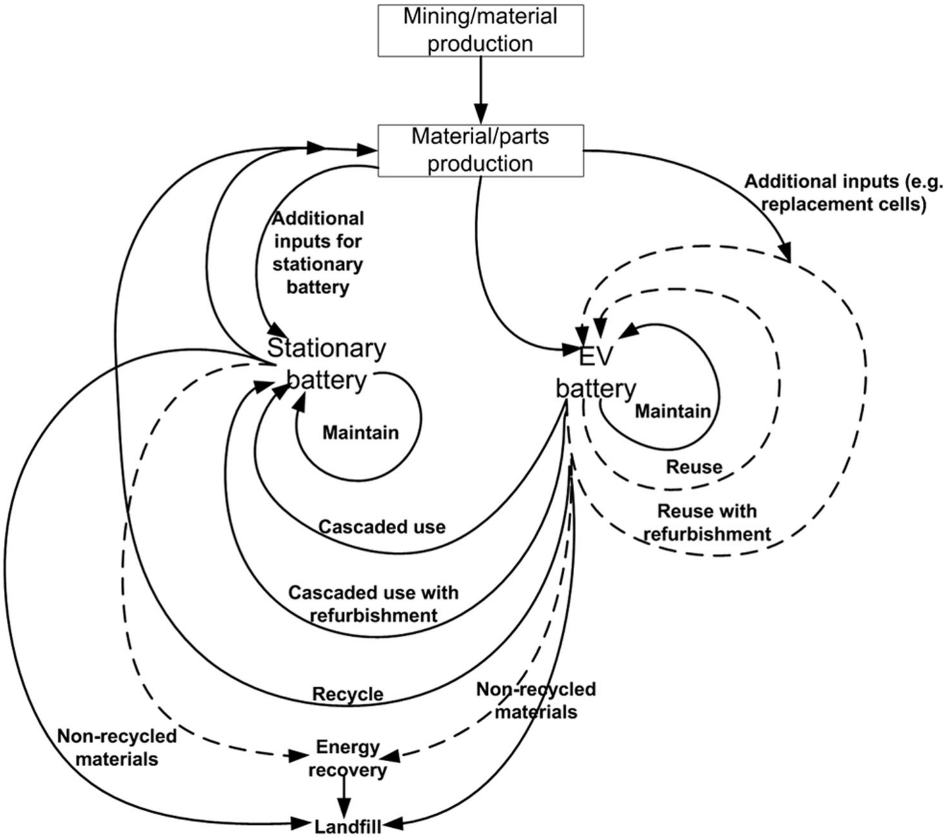

Figure 1 Theoretical waste management hierarchy for retired EV LIBs. EV = electric vehicle; LIBs = lithium-ion batteries.

economic benefits. This case is hypothetical and idealized, because the goal is not to evaluate the impact of current practices for EV LIB waste management, but rather to analyze whether the proposed circular waste management framework would be feasible from an eco-efficiency standpoint. Incineration was not included in the case in spite of high incineration rates (>40%) in some U.S. states like Massachusetts and Connecticut (Michaels 2014), given that landfill is still the predominate waste disposal method in the United States, with barely 13% of MSW combusted for energy recovery (US EPA 2015a). EOL management pathways for EV LIBs along the proposed hierarchy must also consider the existing and emerging waste policy landscape. Hence, a policy analysis was also conducted to review the current mechanisms for battery waste management and identify gaps in current policies. Based on gaps identified, the results of the case study were used to set forth a roadmap for EV battery EOL management research and policy that considers issues specific to these batteries as well as goals of the CE (EMF 2013; Geiser 2001; Gregson et al. 2015; Ghisellini et al. 2016).

Methods

The circular waste management hierarchy analyzed included multiple interacting routes, through which waste batteries would flow in series or parallel, depending on technical feasibility and material composition: (1) closed-loop direct reuse (in EV); (2) open-loop cascaded use (in stationary applications); (3) recycling; and (4) ultimate disposal of materials not reused or recycled (in landfills) (figure 1). Both reuse and cascaded use pathways would include some level of testing and refurbishment to bring batteries back to a usable condition or prepare packs for new applications (Standridge and Corneal 2014; Richa et al. 2014).

For each pathway, the environmental metrics quantified by a life cycle approach were metal input, cumulative energy demand (CED), and eco-toxicity. These metrics represent common environmental concerns about batteries and EVs, namely scarcity or toxicity of contained materials and comparative energy use in production or use of batteries and EVs. The net metal input for different EOL pathways was estimated from the bill of materials of LIB cell and pack components, additional inputs and avoided battery systems in case of reuse/cascaded use of EV LIBs, and recovery efficiencies in recycling. The CED in megajoules (MJ) was calculated based on characterization factors and primary energy sources in the SimaPro CED calculation methodology. Eco-toxicity impacts were based on empirical and database estimates (see below) and calculated with the USEtox method (Rosenbaum et al. 2008). U.S.-based electricity grid mix was used for electricity input data (ecoinvent Center 2010).

The case study was applied to a theoretical stream of 1,000 LIB packs coming out of EV application. This functional unit

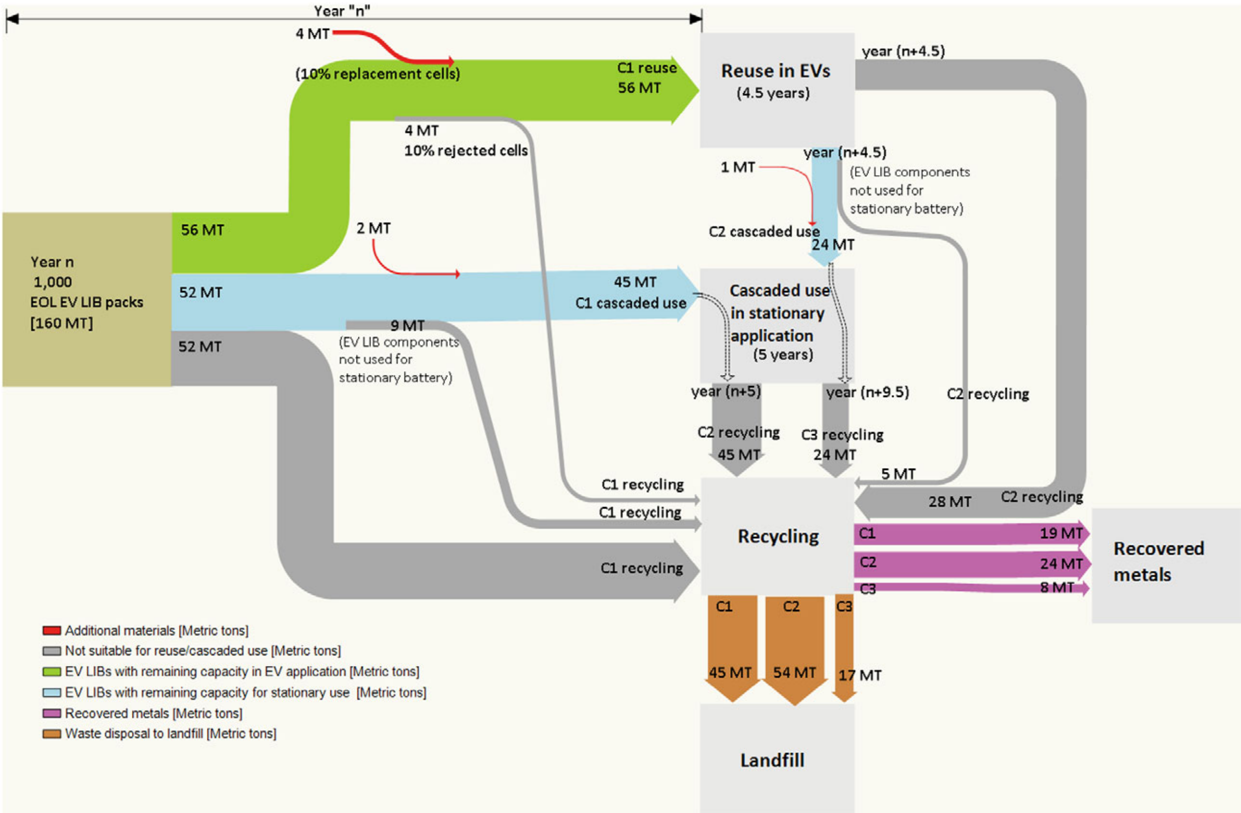

Figure 2 Diagrammatic flows of 1,000 EOL EV LIBs across different waste management routes. C1 denotes a given year "n" in which the battery waste flows out from first use in EVs and into the waste stream, C2 denotes year (n + 4.5), to correspond with maximum expected life span for reuse in EVs, or (n + 5), the maximum expected life span for cascaded use in stationary applications, and C3 denotes year (n + 9.5), for batteries that were technically feasible to cycle through both direct and cascaded reuse. EOL = end-of-life; EV = electric vehicle; LIBs = lithium-ion batteries; MT = metric tonnes.

was selected to create results that could easily be scaled to an actual volume of waste packs, regardless of the time frame or total waste stream. Considering a conservative baseline of EV adoption, material flow analysis results for year 2030 from Richa and colleagues (2014) were normalized to 1,000 LIB packs for a given year "n" (refer to section S1 of the supporting information available on the Journal's website). This waste stream was comprised of 25% battery-electric vehicle (BEV), 36% long-range plug-in hybrid electric vehicle (PHEV), and 39% short-range PHEV LIB packs (Richa et al. 2014).

The baseline battery technology analyzed was lithium manganese oxide (LMO) cathode chemistry, commonly used in blended cathode materials of leading BEV and PHEV batteries (Lu et al. 2013) and previously characterized by LCAs (e.g., Notter et al. 2010; Dunn et al. 2012; Amarakoon et al. 2013; Richa et al. 2015). Additional comparisons were made to determine sensitivity of results to alternate battery technology, specifically cathodes currently used or previously studied (lithium nickel manganese cobalt oxide [NMC] or lithium ferrous phosphate [LFP]; For details, refer to the Supporting Information on the Web).

Figure 2 illustrates the mass flow of 1,000 batteries through the proposed hierarchy. Note that batteries entering into waste management in any given year "n" will cycle through multiple systems at various times in the future. These "cycles" (denoted by C1, C2, and C3) will each be separated by a time lag during which the battery is in its reuse or cascaded use application. For example, for year "n", 40% (by weight) of the EOL EV LIBs would not meet technical criteria required for direct or cascaded reuse and would therefore be recycled in the same year (C1 recycling), whereas the rest of the waste would be recycled in later years after reuse and/or cascaded use, denoted by C2 and C3 recycling (figure 2). Following sections discuss the approach used for the eco-efficiency analysis of each of these EOL pathways.

Reuse in Electric Vehicle Application

A maximum of 35% of the EV LIB outflows in year n may have remaining capacity for use in EVs, having not yet reached 80% capacity due to early vehicle failure, crashes, or cases when an older EV received a new battery replacement and then reached the end of its life before the second battery capacity is used ("life span mismatch"; figure 2). An average of 9 years is considered the "design life span" of LIBs for EV application (Standridge and Corneal 2014; Richa et al. 2014) corresponding to 3,285 cycles if cycled daily. Half of this design life span is considered to be already spent in the first life EV use, leaving 4.5 years for reuse in EVs. This is a theoretical assumption, given that direct reuse is not occurring in practice, to our knowledge, but is a conceivable future scenario as EVs become more widely adopted. An underlying assumption for this pathway is that 10% of the LIB cells would be replaced during refurbishment (Standridge and Corneal 2014). Electrical performance is analyzed using a conservative approach of testing whole battery packs by charging once to 70% battery capacity (Notter et al. 2010).

The net environmental impact of directly reusing EV LIBs, $E_{reuse,\ EV}$ accounted for Avoided environmental impact of manufacturing a new LIB pack (E_m), Design life span of EV LIB (l_d), Life span of EV LIB in reuse application (l_r), Impact of manufacturing replacement cells ($E_{m,cells}$), Impact of battery pack (or cells) testing (E_t), Impact of charge-discharge efficiency losses of refurbished LIB in EV application ($E_{c\text{-}d,\ reuse}$), and the Avoided impact of charge-discharge efficiency losses of new LIB in EV application ($E_{c\text{-}d,new}$) (equation 1):

$$E_{reuse,EV} = -(E_m/l_d)^*(l_r)^*E_{m,cells} + E_t + E_{c-d,reuse} - E_{c-d,new} \quad (1)$$

Life cycle inventory (LCI) data for LIB pack production was obtained from Richa and colleagues (2015), and efficiency loss calculations were based on Zackrisson and colleagues (2010). A direct correlation between capacity decay and battery charge-discharge efficiency was applied, and after reuse in EVs, the capacity and efficiency was reduced to 80% (Ahmadi et al. 2014; Richa et al. 2015) (refer to section S4 of the supporting information on the Web).

The economic cost or benefit (in U.S. dollars [USD]) of directly reusing EV batteries ($V_{reuse,\ EV}$) was determined from Avoided cost of buying a new replacement LIB ($B_{LIB,new}$), Avoided resale value of the new EV battery at vehicle EOL ($S_{LIBused,new}$), Cost of buying a refurbished EV LIB ($B_{LIB,\ refurb}$), and Resale value of refurbished LIB at vehicle EOL($S_{LIBused,\ refurb}$) most likely for stationary applications (equation 2):

$$V_{reuse,EV} = (B_{LIB,refurb} - S_{LIBused,refurb}) - (B_{LIB,new} - S_{LIBused,new}) \quad (2)$$

Future new EV LIB cost (US$125/kWh [kilowatt-hour]), refurbished battery purchase price (US$38/kWh), and used battery selling price (US$20/kWh) were obtained from Neubauer and colleagues (2012) based on similar cost estimates by the U.S. Department of Energy (Howell 2012; Neubauer and Pesaran 2011).

Cascaded Use in Stationary Applications

Used LIB packs in year n could be used for cascaded use in stationary application over two cycles, C1 (immediately on entering the waste stream) and C2 (after reuse in EVs). Some additional component input is required for refurbishing and assembling these systems (wiring, battery management system, etc.) whereas some components may be discarded (Richa et al. 2015). Based on expected cell failures during testing and technical limits of cascaded reuse, only 50% of the LIBs that outlived their usage capacity in EVs were assumed to be feasible for stationary use, with a 5-year life span in stationary application (Richa et al. 2015).

The environmental benefit of cascaded reuse of retired EV LIBs was based on avoiding production and use of lead-acid (PbA) batteries (450 kWh each), the latter being a widely used technology for certain stationary and industrial purposes (Soloveichik 2011), such as grid storage and off-grid renewable systems (Albright et al. 2012). The net environmental benefit from cascaded reuse in stationary energy storage ($E_{reuse,\ stat}$) accounted for environmental impact of a refurbished EV LIB-based stationary energy storage system ($E_{refurb,LIB}$) and the avoided impact of an equivalent functionality PbA battery system, E_{PbA} (equation 3):

$$E_{reuse,stat} = E_{refurb,LIB} - E_{PbA} \quad (3)$$

LCA data for production and use of PbA and cascaded use EV battery systems was obtained from Richa and colleagues (2015). The economic cost or benefit of the second use pathway ($V_{reuse,stat}$) was calculated from Refurbished battery purchase price, $B_{LIB,refurb}$ (Neubauer et al. 2012) and valve-regulated PbA (VRLA) battery purchase price, $B_{PbA\ new}$ (Albright et al. 2012), for the utility sector (equation 4):

$$V_{reuse,stat} = B_{LIB,refurb} - B_{PbA\ new} \quad (4)$$

Recycling

Recycling EOL EV LIBs generated in year n and any additional material input used in reuse and cascaded use stages is likely to occur in three cycles (C1, C2, and C3) separated by time lags (figure 2). Net environmental impact of EV LIB recycling (E_{rec}) along these cycles was calculated assuming equal proportions of the waste stream were processed by either hydrometallurgical or pyrometallurgical processes, considering Environmental impact of recycling process ($E_{rec\ process}$) and Avoided environmental impact due to material recovery ($E_{material\ recovery}$) (equation 5):

$$E_{rec} = E_{rec\ process} - E_{material\ recovery} \quad (5)$$

All metals in EV and stationary LIB packs were assumed to be recycled according to known or estimated recycling rates. $E_{rec\ process}$ and $E_{material\ recovery}$ were calculated from LIB cell recycling and secondary metal production LCA data (Hischier et al. 2007; ecoinvent Center 2010; Fisher et al. 2006; Richa et al. 2015) and recycling efficiencies of metals contained in LIBs (Graedel et al. 2011; Sibley 2011; Mantuano et al. 2006). Speculative recycling rates for lithium and manganese were used because these metals are not currently recovered from LIBs commercially (Gaines 2014). Therefore, results represent the maximum theoretical value achievable from recycling.

Table 1 Average landfill leaching potential of specific metals from the LIB waste stream

LIB metal	*Average leachate concentration (mg/L leachant)*	*Average leaching potential (mg/kg LIB waste)*
Aluminum	131	11,000
Copper	1.61	200
Lithium	273	420,000
Manganese	335	110,000
Steel	13.7	3,100
Cobalt	15	36,000
Nickel	160	21,000

Note: LIB = lithium-ion battery; mg/L = milligrams per liter; mg/kg = milligrams per kilogram.

The economic cost or benefit of EV LIB recycling (V_{rec}) was calculated from the Total cost of recycling operations (TC_{rec}) and Value of recovered materials ($V_{material\ recovery}$) (equation 6).

$$V_{rec} = TC_{rec} - V_{material\ recovery} \tag{6}$$

$V_{material\ recovery}$ was calculated using yearly average metals prices (USGS 2015; Infomine 2015) and recycling efficiencies. TC_{rec} was calculated based on methodology from Wang and colleagues (2014) using fixed cost (1,000,000 US$/year), maximum recycling capacity (34,000 tonnes [t] annually), and variable cost (1,100 $/t) for a recycling facility (refer to section S8 of the supporting information on the Web).

Landfill

Some LIB materials, such as mixed plastics, graphite, and electrolyte, cannot be recycled due to lack of economic motivation or infrastructure (Richa et al. 2014). In addition, some metals will be lost from the value stream due to recycling inefficiencies. Thus, ultimate disposal options must still be considered for nonrecoverable materials generated over the three temporal cycles. CED and indirect eco-toxicity impact of processes associated with landfilling LIB materials were estimated using battery waste transportation and landfill operation LCI data (ecoinvent Center 2010). Eco-toxicity associated with direct releases from battery materials in landfills was estimated from "average leaching potential" shown in table 1 and determined by empirical toxicity characteristic leaching procedure (TCLP) (see section S13 of the supporting information on the Web).

TCLP represents an average leachate concentration over a moderate period of 3 to 10 years for specific landfill design, waste composition, and landfill water percolation characteristics (Frampton 1998). These releases represent environmental loads, and their impacts on ecosystem quality were calculated by multiplying total mass of a metal leached with the USEtox eco-toxicity characterization factor (comparative toxic units eco-toxicity per kilogram; CTUe/kg) for that metal. Currently, the eco-toxicity impacts of copper, manganese, iron, cobalt, and nickel are characterized by USEtox.

The implicit assumption is that nonrecoverable materials from recycling operations will be sent to a landfill, based on similar assumption for batteries in e-waste in recent studies (Wang et al. 2014; Köhler et al. 2008; Espinoza et al. 2014; Asari and Sakai 2013). However, it cannot be over-ruled that in many jurisdictions, EV LIB recycling residue may be reclaimed by as yet developed technology or disposed by other systems (e.g., waste-to-energy), all of which would change loads, pathways, and ultimate impacts of the contained materials. An average landfill disposal cost of US$1,170/t of LIB waste was used, which included a collection fee of US$1,120/t (Wang et al. 2014) and average landfill tipping fee for MSW in the United States at US$49.78/t (US EPA 2015a).

Results

(A) Eco-Efficiency Analysis of Proposed Lithium-Ion Battery End-of-Life Hierarchy

The eco-efficiency analysis of the proposed EV LIB waste management hierarchy as a whole, including reuse, cascaded use, recycling, and landfill, is discussed in the next sections. The goal was to determine whether a CE-inspired system will generate net environmental benefits or introduce unforeseen trade-offs. Therefore, results are presented as net costs or benefits for the circular management of the entire 1,000 battery pack waste stream and are not intended to make comparisons between each pathway. Additionally, areas of uncertainties, which may cause deviations from these results, are discussed for specific cases.

Reuse in Electric Vehicles

Even though a traditional closed-loop circular economy philosophy would suggest that product reuse would be at the top of the waste hierarchy, results actually show minimal benefit from direct reuse of LIBs in EVs. LCA results have consistently pointed out the importance of the use phase in the LIB life cycle, particularly associated with declining round-trip efficiency and increasing energy losses as the battery ages (Richa et al. 2015; Ahmadi et al. 2014). For reuse of BEV and high-range PHEV battery packs in EVs, the avoided CED impact of LIB production exceeded the CED of charge-discharge losses, replacement cells, and LIB testing, resulting in a net benefit of 3,200 MJ and 73 MJ per pack, respectively. However, no CED benefit was observed for short-range PHEV packs due to lower avoided CED value for these smaller packs, compared to the CED impacts of electricity use and losses. Overall, the CED benefit of reusing the maximum feasible number of EV LIBs packs in automotive application (37.2% of the 1,000 packs entering the waste stream) only offsets around 1% of the original energy input required to produce all 1,000 battery packs (figure 3a). Because these benefits are primarily due to avoiding

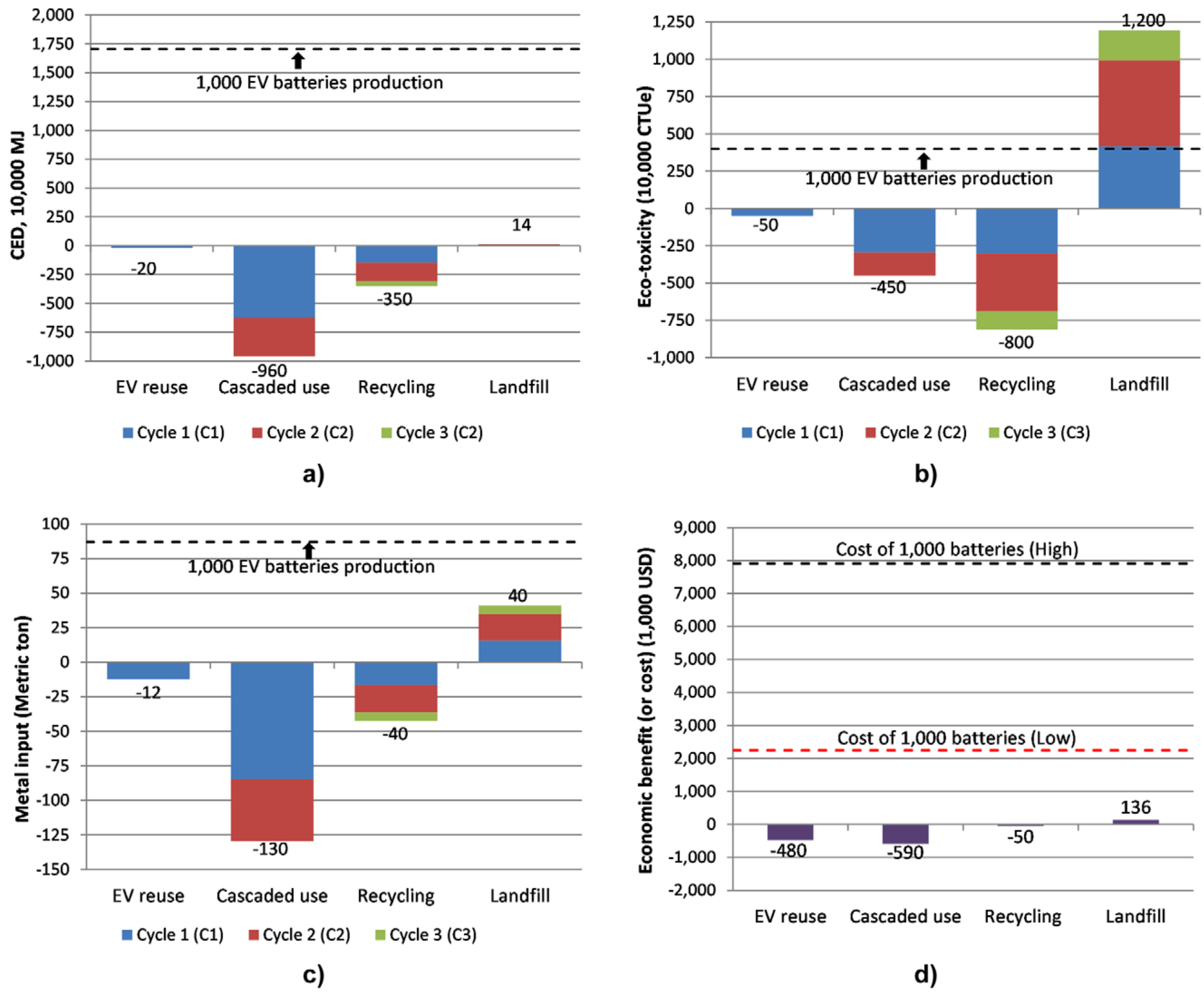

Figure 3 Eco-efficiency of circular management of 1,000 EOL EV LIB packs in terms of net (a) cumulative energy demand (CED), (b) eco-toxicity, (c) metal input, and (d) economic benefit or cost. Results showcase a circular economy–inspired holistic approach of handling this waste stream and are not meant to be a comparison of different waste management routes. For Figure 3a, 3b, and 3c, a negative value denotes environmental benefit due to avoided production of new battery systems or avoided energy use in less-efficient displaced systems. The dotted lines represent impact due to initial production of the 1,000 LIB packs. These environmental impacts are showcased for specific temporal cycles C1, C2, and C3. Figures reporting disaggregated contributors to these results are provided in the Supporting Information on the Web. In Figure 3d, a negative value denotes economic savings whereas a positive value denotes economic cost. The dotted lines represent the "beginning of life" cost of the 1,000 EV LIBs for both high (upper line) and low (lower line) battery cost scenarios. EOL = end-of-life; EV = electric vehicle; LIBs = lithium-ion batteries; MJ = megajoules.

new battery packs, LIB chemistries such as LFP, which have already low manufacturing CED impact, would not gain additional CED benefit from this pathway (refer to the Supporting Information on the Web).

If an aggressive testing procedure was used during refurbishing, wherein individual LIB cells are tested for reuse capability, no CED benefit is expected in any reuse pathway. On the other hand, if energy efficiency of EV LIB does not decline significantly with aging, the net CED benefit could be as high as 2.6 million MJ (around 10% of the initial CED associated with producing the original 1,000 packs). This represents a "hypothetical best case" given that efficiency degradation in reused products often makes them environmentally unfavorable when compared to a new product for the same application (Gutowski et al. 2011).

Reuse of EV LIBs in vehicles can provide an eco-toxicity benefit per pack of 430 to 3,100 CTUe for short-range PHEV to BEV battery (figure 3b). This benefit is due to avoiding the production of new LIB packs (as replacement EV batteries), which outweighs the potential eco-toxicity during the use phase, even with aggressive battery testing. This pathway has the capability of avoiding mining of approximately 12 t of metals: primarily aluminum; manganese; and copper (figure 3c). The estimated economic benefit from LIB reuse in vehicles ranges from USD

330 to USD 3,000 per pack for short-range PHEV up to BEV battery (resulting in a net benefit of 480,000 USD for the functional unit of 1,000 battery packs [figure 3d]). These economic gains could increase more than 3 times if high values of new EV LIB cost (US$440/kWh), refurbished battery buying price (US$132/kWh), and used battery selling price (US$100/kWh) are considered (Neubauer et al. 2012).

Cascaded Use in Stationary Applications

For a stationary energy storage system operating for 5 years, a net CED and eco-toxicity benefit of 1,330 MJ/kWh and 626 CTUe/kWh can be obtained through cascaded use of retired EV LIBs and by avoiding the production and use of PbA battery systems. Scaled to the entire 1,000 EOL battery stream, a potential net CED and eco-toxicity benefit of 9.6 million MJ and 4.5 million CTUe was estimated. Reuse in stationary battery systems would potentially avoid 130 t of metal inputs, primarily by avoiding primary and secondary lead production.

Approximately 600,000 USD cost savings for the utility sector is estimated from installing these stationary energy storage systems with combined storage of 7,200 kWh (figure 3d). However, these estimates are based on low future LIB cost (Neubauer et al. 2012) and comparisons made with VRLA battery systems. There is immense uncertainty in future LIB prices and the type of PbA battery systems that would be replaced by these retired LIBs. When compared to flooded PbA battery systems, these cost savings would be reduced by 67%, and in case of high future LIB cost scenario, where the refurbished battery buying price could be as high as US$132/kWh, no economic benefit is expected from cascaded use.

Recycling

Recovery by recycling is applicable both for metals contained in the original 1,000 EV LIB waste stream as well as for any additional material input necessary for repair or refurbishment. Recycling provided CED and eco-toxicity credits of approximately 3.5 million MJ and 8 million CTUe, respectively. Using hydrometallurgical recycling provided 25% higher CED credits when compared to the pyrometallurgical process, primarily due to 4 times higher energy input for the latter (Fisher et al. 2006). For a mixed waste stream of equal fraction of LMO, NMC, and LFP batteries, the CED savings can be as high as 4 million MJ owing to slightly higher energy saving from recycling the latter two chemistries.

Pyrometallurgical recycling resulted in appproximately 50 t of avoided metals, whereas the metal recovery from the hydrometallurgical process was 29% less, leading to 40 t of net avoided metals by both routes (figure 3b). This is because the latter yields lithium, which constitutes 1% to 2% of the cells, whereas the pyrometallurgical process yields manganese, comprising of 22% to 24% of the cells, with both metals exhibiting similar theoretical recovery efficiencies of around 50%. Direct recovery of LIB cathode through chemical relithiation to regain electrochemical performance at EOL has been demonstrated at small scales for LFP and lithium cobalt oxide LIB cathodes and can provide even greater energy savings (Ganter et al. 2014; Dunn et al. 2012). However, we have used the more conservative and currently commercialized hydrometallurgical and pyrometallurgical recycling processes for the LMO chemistry, as demonstrated by Fisher and colleagues (2006), for a European recycling facility, considering the uncertainty in future commercial recycling procedures.

A conservative cost (USD 1,100/t variable cost) of recycling LIB pack materials results in overall recycling cost of 190,000 USD, for processing a total of 167 t of materials. Approximately 240,000 USD in material value can be obtained across the three recycling cycles (~50 t), resulting in net economic benefit of 50,000 USD (figure 3d). Around 65% of this material value is derived from recycling LIB cells; the remaining fraction comes from recycling other material inputs. However, recycling costs for LIBs are uncertain: An average (USD 2,800/t) vs. high-end (USD 4,500/t) cost of recycling operations (Wang et al. 2014) change the net economic cost between 235,000 and 520,000 USD. For the conservative recycling cost scenario, a net benefit of as large as 250,000 USD could be expected if the waste stream was comprised entirely of NMC cathode batteries, due to high commodity value of cobalt (USGS 2015). However, a mixed waste stream of LMO, NMC, and LFP LIB packs would result in negligible material value (refer to Supporting Information on the Web).

Landfill

For all three cycles combined, the quantity of waste materials eventually entering the landfill is expected to account for 70% of the of the total waste stream (1,000 EV LIB packs plus additional material input during refurbishment). This stream would contain 115 t of nonrecyclable materials, such as mixed plastic, electrolyte, or graphite and recycling residues, containing unrecovered metals. The cost of disposing this fraction of the waste LIB material was estimated at 136,000 USD (figure 3d). Approximately 40 t (35%) of this landfill stream was comprised of metals, with up to 5% (~2 t) of these metals potentially leaching in the landfill (figure 4). These releases depend on numerous factors, such as landfill age, control mechanisms, waste composition, water percolation, time dimension of leaching, and metal degradation, which are all uncertain parameters (Olivetti et al. 2011; Rydh and Karlström 2002). Slack and colleagues (2005) estimated 0.02% metal releases in landfill for nonbattery waste, Fisher and colleagues (2006) assumed 5% leaching potential of heavy metals from spent batteries, whereas Rydh and Karlström (2002) assumed all metals from batteries to be released over an infinite time period, hence our results suggest a moderate leaching potential for LIBs.

The US EPA has not set regulatory limits for these metals, though the State of California has total threshold leaching concentration limits for cobalt, nickel, and copper (Eurofins 2012), which are generally present in LIBs with mixed metal chemistries like NMC (Nelson et al. 2011). The CED and "indirect" eco-toxicity impacts of landfill operation would be approximately 140,000 MJ and 14,000 CTUe, respectively. These impacts are primarily (>90%) due to waste transport to landfill, whereas impacts of energy input and land use for landfill

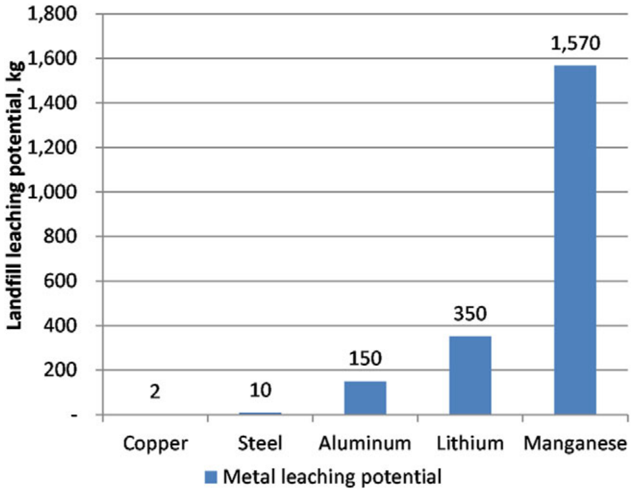

Figure 4 Landfill leaching potential of unrecovered metals from the total (115 t) LIB waste potentially entering landfills. kg = kilograms; LIBs = lithium-ion batteries; t = tonne.

operation are negligible. "Direct" eco-toxicity due to leaching of metals from the analyzed landfill waste (figure 3b) would result in 12 million CTUe of eco-toxicity, which far exceeds the upstream, indirect eco-toxicity contributions. Use of nickel- and cobalt-based LIBs could aggravate the eco-toxicity impacts by an additional 13%, whereas a waste stream composed of only LFP batteries would reduce these impacts by 47% (refer to the Supporting Information on the Web). Nevertheless, despite the widespread attention on eliminating battery landfill, these results underscore that a greater degree of eco-toxicity impact can be avoided by reusing and recycling batteries, to make up for the impact created if materials leach from landfills.

Consolidated Results

The consolidated results of this case study across various waste management routes were compared to the environmental impact of production and "beginning of life" cost of the 1,000 EV LIBs in the analyzed waste stream (figure 3) based on a previous LCA study (Richa et al. 2015). Results suggest that the circular waste management hierarchy as a whole would be able to recoup around 77% of the CED and 30% of the eco-toxicity impact associated with the original production of these batteries. In terms of metal inputs, the proposed system can avoid approximately 1.6 times the net mass that is used in LIB production, primarily because the cascaded use pathway can theoretically displace much larger PbA battery systems. The proposed system can recover 12% to 44% of the initial cost of ownership of these batteries; however, there is immense uncertainty in this domain. All results also hinge on future technological advances necessary for realizing widespread battery reuse in EVs or cascading applications.

(B) Policy Analysis

The results of the case study indicate that a CE-inspired waste management hierarchy for EV LIBs would generally create environmental benefits—with the exception of EV reuse, which is still technically unexplored and environmentally uncertain. However, effective policy mechanisms are needed to encourage development of infrastructure to support this hierarchy, particularly when the economics may not favor implementation. Table 2 analyzes current policy mechanisms at the U.S. and EU level that can impact EV LIB waste management in the future.

The EU Battery Directive (BD), which regulates disposal of all battery types, categorizes EV batteries as "industrial batteries" (EC 2006) and is expected to include special provisions for EV LIB EOL management, unless an independent EV battery regulation is introduced. The EU EOL vehicle (ELV) directive provides guidelines for collection and EOL management of vehicles and their components (EU 2000) and can be further expanded to include specific provisions for EOL EV LIBs. In the United States, the Mercury-Containing and Rechargeable Battery Management Act of 1996 (US GPO 1996) (Battery Act [BA]) mandates all recognized batteries (mercury-based, nickel-cadmium, and PbA) be considered hazardous waste and fall under the standards for Universal Waste Management. The BA mandates guidelines for disposal of PHEV batteries, but because the US EPA Universal Waste Rule does not consider LIB materials to be hazardous, these batteries are excluded. Only three U.S. states have rechargeable battery waste management regulations that incorporate LIBs—California's Rechargeable Battery Recycling Act of 2006 (CA Code 2006), New York State Rechargeable Battery Law (2010), and Minnesota Rechargeable Battery and Products Law of 1994 (MN PCA 2015).

While the current policy landscape governing EOL batteries spans the entire battery life cycle (table 2), considerations specific to EV LIBs would be required to handle the volume and complexity of this waste. Table 2 highlights future regulatory mechanisms that integrate the proposed waste management hierarchy with a life cycle approach, considering that product design and manufacturing are also integral stages in planning a circular system (EMF 2013; Allwood et al. 2011, 2012; Gregson et al. 2015; Bakker et al. 2014).

Battery/Vehicle Production

Initiatives for environmentally safe and convenient EOL management can be introduced at the point of LIB and vehicle production. Some examples include elimination of hazardous substances in EV LIBs, labeling requirements for easy identification and sorting of LIBs at EOL, as well as design principles that enable easy removal of LIBs from EVs, and facilitate battery disassembly, refurbishment, and material recovery (Arbabzadeh et al. 2016). These design strategies can include modular battery pack design, use of compatible materials such as consistent grades of plastics, chemistry standardization, avoiding welded or soldered connections, and minimizing the overall number of pack components (Chiodo 2005). In this direction, The U.S. Advanced Battery Consortium (USABC) is developing design-for-recycling guidelines for automakers (USABC 2013).

Table 2 Current policies governing LIB EOL management and future policy mechanisms

Life cycle stage	*Initiative*	*Current policy that may be extended for EV LIBs*	*Future mechanisms*
Battery/vehicle production	Material selection	**BD**: No restriction on LIB materials (i.e., considered nonhazardous) **ELV**: Limits use of hazardous substances in vehicles (does not explicitly address EV LIBs) **CA state** classifies LIBs hazardous due to excessive levels of cobalt, copper, and nickel	• Can possibly regulate some LIB materials such as Co, Ni, and nano materials • Chemistry standardization to avoid EOL sorting
	Design for EOL	**BD, ELV**: Appliances or vehicles to be designed to facilitate battery/component removal **BA**: Mandates ease of battery removal (policy does not cover LIBs)	• Design principles for ease of disassembly, repair, refurbishment, and recycling of EV LIBs as part of regulations
	Labeling or identification	**BD**: Labeling restricted to heavy material content (mercury, lead, cadmium) and landfill ban (crossed-out wheeled bin); capacity label nonmandatory **ELV**: Material and component coding standards for identification **CA,MN**: Battery type (e.g., LIB, NiCd, etc.) and note on recycling and safe disposal **BA**: Mandates labeling for battery type and recycling and safe disposal (but not LIBs)	• Specific labeling guidelines for LIBs: chemistry, capacity, and LIB materials to facilitate sorting, remanufacturing, and recycling • Bar codes, RFID chips, specialized coloring for LIBs
Use phase	Repair or maintenance	**EU Waste directive** defines "waste hierarchy" wherein waste prevention through product life span extension precedes other EOL management routes.	No current EOL battery policy mandates repair/battery maintenance to extend life.
Collection	Extended producer responsibility	**BD**: Collection financed by battery producers or third parties acting on their behalf **ELV**: Extended producer responsibility collection scheme applied to battery when collected with vehicle **CA,NY, MN**: Retailer or battery manufacturer to provide for collection **Call2Recycle**: Product stewardship program providing no-cost battery collection across United States and Canada funded by battery and product manufacturers	• Regulations to specify transfer of collection responsibility or liability in case of reuse/cascaded use • In U.S. states (NY, CA), collection programs are only for small, nonvehicular rechargeable batteries and may be expanded.
Reuse/cascaded use	Reuse/cascaded use provision	**BD**: Reuse or cascaded use not defined **ELV**: Reuse of vehicle component defined as use in same application; cascaded use in another application not defined; mandates safe stripping operations, storage, and testing to ensure suitability of vehicle component reuse (can include EV LIBs) **CA**: mentions "reuse"; not explicitly defined for LIBs	• Battery testing guidelines • Prioritize second applications based on techno-economic analysis from national labs in the United States • Economic incentives for reuse/cascaded use • Safety laws for large cascaded use installations
Recycling	Targets and process guidelines	**BD**: 50% recycling efficiency and rules for calculating the efficiency **ELV, BD**: Very brief guidelines for dismantling, storage, and handling of batteries (e.g., electrolyte removal, removal of metals and plastic, sorting, etc.) •No recycling or process efficiency targets in United States	• Rules related to worker safety and exposure • More specific dismantling manuals for EV LIBs • Developing mixed stream LIB recycling techniques • Economic incentives to promote recycling • Recycling efficiency improvement

(*Continued*)

Table 2 Continued

Life cycle stage	*Initiative*	*Current policy that may be extended for EV LIBs*	*Future mechanisms*
Incineration and landfill	Prohibition	**BD**: Landfill and incineration prohibited **ELV**: Waste-to-energy and landfill of nonrecycled vehicle components allowed **CA,NY,MN**: Landfill ban only (with ineffective or no penalty for noncompliance)	• Landfill ban to be extended to other U.S. states • Awareness on landfill toxicity of LIB materials • Landfill tax, deposit-refund schemes, recycling incentives, differential taxation, etc.
Transport	Shipping guidelines (listed as Class 9 Miscellaneous hazardous material)	**BD**: Waste batteries exported for recycling to comply with waste shipment laws •LIB transport regulated by U.S. Department of Transportation, U.S. Hazardous Materials Regulations, International Civil Aviation Organization, International Air Transport Association, and International Maritime Dangerous Goods, which provide packaging, labeling, shipping, and fire hazard prevention instructions	• National and international regulations governing LIB transportation can be extended to waste LIBs, and EOL battery management laws would mandate compliance. • Specific guidelines for large-size EOL EV LIBs.

Note: **BD**: EU Battery Directive; **ELV**: EU end-of-life vehicle directive; **NYS**: New York Rechargeable Battery Recycling Act (2010); **CA**: California's Rechargeable Battery Recycling Act (2006); **MN**: Minnesota Rechargeable Batteries and Products law (1994); **BA**: Federal Mercury-Containing and Rechargeable Battery Management Act of 1996.
LIB = lithium-ion battery; EOL = end-of-life; EVs = electric vehicles; NiCd = nickel cadmium; EU = European Union; EPR = extended producer responsibility; Co = cobalt; Ni = nickel; RFID = radiofrequency identification.

Use Phase

Currently, none of the battery waste policies at the United States or EU level emphasize maintenance or repair of batteries to extend their life span during the first use phase. Only the EU Waste Framework Directive has outlined a waste management hierarchy wherein waste prevention through product life span extension precedes reuse, recycling, energy recovery, and disposal (European Parliament 2008). Future efforts can include specific strategies toward ensuring a longer life span of LIBs in EV use, for example, through consumer awareness and education on how driving behavior impacts battery aging.

Collection

In the EU, extended producer responsibility (EPR) mechanisms are in place to ensure collection of waste batteries. The EU BD requires battery producers, or designated third parties, to finance collection, treatment, and recycling of EOL batteries (EC 2006). Under the ELV directive, EPR collection schemes apply only to vehicle components such as batteries and accumulators collected along with scrapped vehicles (EU 2000). In the United States, EPR mechanisms for LIB collection exist only in California, New York, and Minnesota wherein battery manufacturers or retailers must provide consumers with a free system for returning EOL batteries (CA Code 2006; New York State Rechargeable Battery Law 2010; MN PCA 2015). Only Minnesota has set collection targets (90%, nonmandatory) for EOL rechargeable batteries and mandates that EV and battery manufacturers jointly manage EOL batteries (MN PCA 2015). New York and California laws and voluntary collection schemes in the United States, such as Call2Recycle, are restricted to batteries from consumer electronics and exclude larger vehicle batteries (Call2Recycle 2015). New regulations would be required to specify transfer of collection responsibility in case of reuse or cascaded use of EV LIBs.

Transport

LIBs are listed as Class 9 Miscellaneous hazardous material (Mikolajczak et al. 2011), and there are specific shipping, packaging, and labeling guidelines for these batteries for transporting them domestically or internationally (table 2). One concern is the "thermal runaway" of LIB cells causing self-ignition leading to safety hazards (Webster 2010). It is likely that different waste regulations governing EOL LIB management would mandate compliance to these guidelines and associated restrictions while transporting these batteries. For example, the EU BD mandates that waste batteries, when exported for recycling, should comply with waste shipment laws (EC 2006). Additional safety requirements are expected for transporting retired EV LIBs due to their large size, which may increase cost.

Reuse and Cascaded Use

The case study indicated that while direct reuse of LIBs in EVs may not support CE principles for the CED metric, it could still provide minor environmental benefits by avoiding eco-toxicity impacts and metal inputs, along with cost savings for EV users. Future policies governing the reuse pathway can depend on the metrics that stakeholders deem important for their analysis and decision making. However, considering the large environmental benefits and possible cost savings for the utility sector, cascaded use of retired EV LIBs in stationary applications should be prioritized over lower levels of the waste hierarchy. Currently, both reuse and cascaded use are severely under-represented in the policies analyzed. While the ELV directive defines "reuse" of vehicle components for use in the same application, the more specific BD is only focused on safe disposal and recycling of batteries (EC 2006). Waste management policies in the United States do not explicitly address battery reuse or cascaded use, and second-life EV LIBs are currently not part of incentive programs or tax credits for grid or on-site energy storage systems in the United States. However, cascaded use offers significant benefit to augmenting renewable energy deployment efforts: Around 50% of the PHEV batteries in use in California could store up to 850 megawatt-hours of energy after use in EVs (Elkind 2014). To promote such applications, future regulations can mandate battery testing protocols for specific stationary end uses and create economic incentives (such as tax rebates) for cascaded use pathways. It is, however, likely that additional regulatory barrier may govern siting of large stationary energy storage systems due to safety and environmental health concerns of second-life batteries (Elkind 2014).

Recycling

Results suggest that recycling should be considered after cascaded use options for EV LIBs. A well-established EPR network can incentivize an integrated reuse-recycling approach, given that some of the revenue from utility scale operations could bear the burden of downstream EOL management. The major focus of commercialized LIB recycling operations has been on cobalt recovery from consumer electronics batteries (Wang et al. 2014). However, high costs of LIB recycling may reduce the economic incentive from recycling noncobalt chemistries, such as LMO batteries. If the cells in this case study were composed of NMC cathode chemistry instead of LMO, the gross estimated value obtained from recycling increases twofold due to high market value of cobalt and nickel (USGS 2015). Hence, technical advancements to improve recycling processes, regulations to encourage EV LIB collection to promote economies of scale (Wang et al. 2014), and incentives and rebates are needed to encourage recycling of low-material-value LIBs. The EU BD has recycling efficiency mandates of 50% for batteries including LIBs, but no legislation within the United States provides process targets. Owing to expected variability in chemistry and composition of EV LIBs, their recycling will need to avoid cross-contamination or develop recycling procedures to process different LIB chemistries simultaneously. The Society of Automotive Engineers in the United States and EUROBAT in the EU have established active working groups to develop solutions for battery labeling and to prevent cross-contamination in LIB recycling streams (Gaines 2014).

LIB recycling can also face regulatory hurdles due to workplace exposure to LIB materials during battery disassembly and shredding (Wang et al. 2016). The ELV directive provides generic guidelines for dismantling, storing, and handling vehicle batteries, but does not address issues specific to LIBs. A majority of LIB cathode and electrolyte materials have Occupational Safety and Health Administration hazards associated with them (Vimmerstedt et al. 1995), which may require workplace regulations for LIB recycling facilities, further raising the cost of recycling operations.

Landfill

While the share of EV LIB waste ultimately reaching the landfill may be as high as 70% of the entire waste stream, the cost of landfill was estimated to be very low. Preliminary TCLP analysis of bulk of LIB metals (table 1) suggests that metal concentration in LIB leachate could exceed the U.S. Primary and Secondary Drinking Water Standards (US EPA 2009), as well as the EU Drinking Water Directive and World Health Organization's (WHO) guideline limits (EU 1998; WHO 2008). If the landfill leachate from LIBs were to contaminate the groundwater, it could pose a potential threat to human health and environment.

Hence, appropriate landfill tax and widespread landfill bans can help minimize these risks and increase management by reuse and recycling. Based on the regional tipping fee and the expected EV LIB waste disposal for specific states, regional variability is expected in LIB landfill disposal expenses. Differential landfill tax can be introduced based on increasing LIB waste tonnage for a given region. Economic instruments to encourage LIB recycling, such as deposit-refund schemes (Walls 2006), or incentivizing recycling can prevent noncompliance to landfill bans. Additionally, improved recycling efficiencies of LIB metals and recycling currently nonrecovered LIB components, such as electrolyte, graphite, and plastics (Richa et al. 2014), is needed to reduce landfill disposal.

Conclusions

A CE-centric waste management hierarchy can be effective in managing the EV LIB waste stream in the future, but uncertainties exist as to the eco-efficiency of specific EOL routes comprising the overall system. Both EV LIB reuse and cascaded use have potential for providing environmental and economic benefits. However, such benefits rely significantly on LIB size, testing procedures, the incumbent battery systems that used LIBs would displace, future prices of new and old EV LIBs, and regulatory barriers due to environmental health and safety concerns. Case-study results underscore the need for holistic eco-efficiency assessment of systems designed based on circular economy concepts. In this case, a *closed* circularity, in which LIBs are reused directly in their original application (EV), was less desirable than a more *open* loop option of cascading use into a different application (energy storage), which may be nonintuitive to a traditional CE mindset.

Achieving the expected benefit from cascaded use of EV LIBs requires policies and economic incentives to promote cascaded use before recycling. However, metal recovery by recycling is essential to closing the loop on battery materials, and growing this nascent industry will still require policy and technology development to create collection programs, improve recovery efficiencies, and add economic incentives. Additional policy may be needed to ensure safe transport and promote worker health and safety. While traditional policy approaches might intuitively look to battery landfill bans as a strategy to minimize eco-toxicity, this case study demonstrated that reuse, cascaded use, and recycling combined would negate the impact of toxic releases to the environment, due to potential direct releases from a landfill. Given the variety of solid waste disposal practices globally, such an analysis should be repeated for technology specific to each region in which battery waste management is emerging as a sustainability concern. The combination of CE insights and life cycle eco-efficiency analysis offer a holistic approach to designing, validating, and identifying policy barriers and opportunities for a sustainable waste management hierarchy.

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Supporting Information

Supporting information is linked to this article on the *JIE* website:

Supporting Information S1: This supporting information consists of 18 sections labeled S1 to S18. These sections include information on electric vehicle lithium-ion battery waste flows, battery bill of materials, additional material input during reuse in EV, battery charge-discharge efficiency losses, battery testing for refurbishment and reuse, future new, used, and refurbished battery prices, environmental impact of LIB recycling, LIB recycling cost, and LIB metal recycling efficiency and commodity value. The supporting information also presents detailed recycling flows and net CED savings across C1, C2, and C3 phases. Further, this section examines additional material input for cascaded use, a cascaded use life cycle assessment, comparison with BEV vehicle and battery production, a landfill leaching analysis, a TCLP experiment, the environmental impact of landfill pathway, the bill of materials of lithium nickel manganese cobalt oxide (NMC) cells and pack, the bill of materials of lithium ferrous phosphate (LFP) cells and pack, contributions to net results (environmental impacts), and drinking water standards.