Life cycle comparison of industrial-scale lithium-ion battery recycling and mining supply chains

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Abstract

Recycling lithium-ion batteries (LIBs) can supplement existing supplies of critical materials and improve the environmental sustainability of LIB supply chains. In this work, environmental impacts (greenhouse gas emissions, water consumption, energy consumption) of industrial-scale production of battery-grade cathode materials from used LIBs are compared to the environmental impacts of conventional mining supply chains. Refining mixed-stream LIBs into battery-grade materials reduces these environmental impacts by at least 59%. Recycling batteries to mixed metal products instead of discrete salts further reduces environmental impacts. Electricity consumption is identified as the principal contributor to all LIB recycling environmental impacts, and different electricity sources can change greenhouse gas emissions up to eight times. Supply chain steps that precede refinement (material extraction and transport) contribute marginally to the environmental impacts of circular LIB supply chains (<5%), but are more significant in conventional supply chains (31%). This analysis disaggregates conventional and circular steps based on material extraction, transport, and industrial refinement operations; provides important insights for advancing sustainable LIB supply chains; and informs optimization of industrial-scale environmental impacts for emerging battery recycling efforts.

Keywords: circular economy, critical materials, hydrometallurgy, life cycle assessment, pyrometallurgy, reductive calcination

The rise of intermittent renewable energy generation and vehicle electrification has created exponential growth in lithium-ion battery (LIB) production beyond consumer electronics. By 2030, the electric vehicle (EV) sector is projected to dominate LIB growth, accounting for 82% of an estimated 2.4 TWh yr^{-1} of total global LIB production (**Fig. S1, Supplementary Information)**. However, the limited supply of critical materials (e.g., Li, Ni, Co, and Cu[1]) needed for prominent LIB chemistries has exacerbated environmental, economic, national security, and human rights concerns[2,3]. Critical LIB materials are projected to reach major global supply-demand balance deficits before 2030 (**Fig. S1**). Further, both mining of LIB materials and improper disposal of end-of-life LIBs can damage natural and human ecosystems, cause occupational hazards during handling, and result in monetary losses[4].

Recycling critical materials in end-of-life LIBs can help alleviate growing environmental concerns and is essential for the long-term sustainability of electrified transportation. While recycled materials may not contribute substantially to global LIB demand for decades, the establishment of domestic circular supply chains is iterative, requiring multiple learning curves as the dominant supply of end-of-life LIB chemistries and form factors evolve and as supply grows. Factors central to the success of recycling include the ease of collecting products, the cost of recycling processes, and the economic value of recovered materials. The average embodied economic values of representative LIBs between 2018–2021 are shown in **Fig. 1a** (complete references are listed in **Supplementary Information**). In LIBs, between 2018–2021, Li, Ni, and Co comprise the highest embodied economic value, and Al and Cu account for a significant weight percentage of EV battery packs (approximately 25%)[5]. Despite an embodied economic value that is 2–10 times higher compared to the lead in lead-acid batteries, LIBs are only recycled 2–47% globally[6], compared to 99% for lead-acid batteries in the U.S. Regardless, the untapped potential of LIB recycling constitutes a significant economic and environmental opportunity that requires evaluation across several application scales, from numerous small-scale consumer electronic LIBs (e.g., 10–100 Wh) to fewer large-scale transportation and stationary storage LIB packs (e.g., 10–100 kWh)[7]. In addition, the preferred chemistries by automakers have evolved to hedge potential critical mineral shortages and react to market shifts, such as the near tripling of lithium carbonate prices in early 2022. Existing LIB variation and supply chain complexity highlight the need for a methodical and comparative life cycle assessment (LCA) between circular (i.e., recycling used

batteries) and conventional supply chains, which is also necessary for future recycling of the evolving portfolio of battery chemistries.

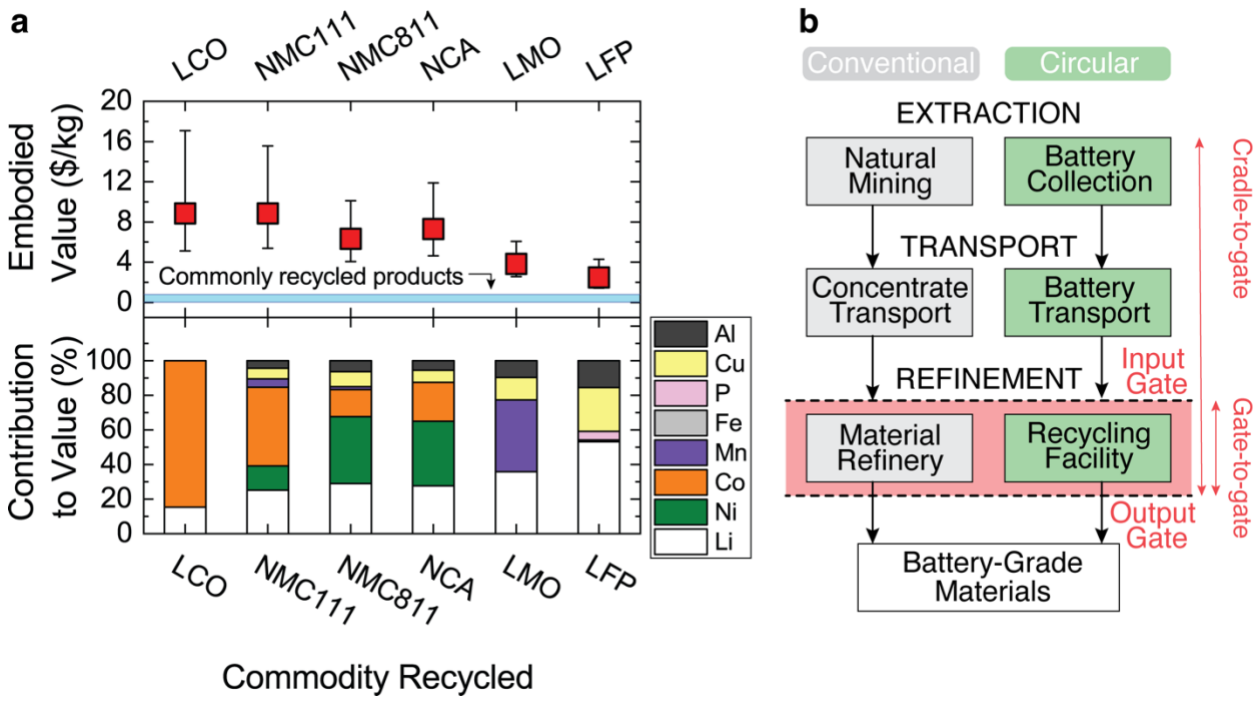


Fig. 1 | Economic drivers of lithium-ion battery (LIB) recycling and supply chain options for producing battery-grade materials. a, Commodity values of representative LIBs (upper panel) and relative contributions of embodied metal elements to the LIB values (lower panel). Representative LIBs are from consumer electronics using lithium cobalt oxide (LCO), and electric vehicle battery packs including lithium nickel manganese cobalt oxide (NMC111 and NMC811), lithium nickel cobalt aluminum oxide (NCA), lithium manganese oxide (LMO), and lithium iron phosphate (LFP). Data are based on market values adjusted for inflation between January 2018 and December 2021 (complete references are listed in **Fig. S1** in **Supplementary Information**), and the uncertainty denotes a 90% confidence interval, which may overlap with the data point in some instances, obscuring their view. The blue shaded area in the upper panel represents the average commodity values of commonly recycled products: glass, paper, plastic, and metal cans (more details are provided in **Fig. S1**). **b**, Cradle-to-gate steps of manufacturing battery-grade LIB materials (i.e., salts) from conventional and circular supply chains, both of which include three steps: extraction, transport, and refinement. Extraction and transport are considered upstream steps relative to gate-to-gate refinement, which is indicated by the red shaded area between "input" and "output" gates. Cradle-to-gate analysis considers the refinement and upstream processes together.

Despite significant progress, current understanding of the relative environmental impacts of recycling LIBs is still incomplete. The most significant environmental differences between LIB production from circular and conventionally mined cathode material lie early in supply chains, comprised of extraction, transport, and refinement steps (together "cradle-to-gate," **Fig. 1b**). While several previous studies have investigated cradle-to-gate environmental impacts, gate-to-gate analyses of circular refinement processes are inconsistent, reporting environmental impacts that differ by >30%[8-10], and are not completely based on industrial-scale LIB recycling operations. The gate-to-gate refinement processes utilized at established and emerging circular refinement facilities may include mechanical separation (Me), pyrometallurgy (Py), and hydrometallurgy (Hy)[8,9]. Specifically, Me physically dismantles LIBs into constituent components, Py leverages elevated temperature to facilitate thermally-driven material transformations, and Hy separates materials in the aqueous phase via leaching, precipitation, and solvent extraction processes. Variations in environmental impacts arise from the specific operational choices at refinement facilities that utilize different processing pathways and from the methods to evaluate them. There is a critical need for transparency and detailed insights into the environmental impacts (e.g., energy consumption, greenhouse gas emission, and water consumption) of LIB refinement pathways and all cradle-to-gate supply chain steps. Previous efforts have worked towards addressing this need[8,11], and this study builds on the comparative methodology of a recent step-by-step study to provide higher resolution and more actionable primary data, insights, and recommendations. Advancing decision-making capabilities to scale sustainable LIB supply chains requires life cycle assessment with more granular data at each step, inclusion of industrial-scale refinement operations with practical mixed-stream battery feedstocks, documentation of operational parameters, and qualification of results in terms of limitations and applicability to real-world scenarios.

In this study we quantify the cradle-to-gate environmental impacts of battery-grade cathode material salts manufactured in conventional and circular supply chains across three major steps: material extraction, transport, and refinement (**Fig. 1b**). First, we quantify and compare the refinement of mined concentrate from natural deposits into battery-grade materials in conventional supply chains with production of these materials by Redwood Materials (a recycling company in Nevada, U.S.). Two LIB feedstocks are explored: non-energized LIB production scrap from

manufacturing facilities and energized end-of-life LIBs collected from consumers. Industrial-scale operational data provided by Redwood Materials are analyzed and compared to conventional LIB supply chain values based on Argonne National Laboratory's Greenhouse Gases, Regulated Emissions, and Energy use in Technologies (GREET 2021) model[12]. Second, influences of the product formats in the refinement pathways on environmental impacts are examined. For both conventional and circular refinement, impacts of producing mixed Ni-Co compounds and discrete salts are analyzed. Third, we assess the environmental impacts of upstream processes before gate-to-gate refinement based on modeling. The upstream assessment includes the extraction of LIB material from conventional (i.e., mined ore) or circular (i.e., collected batteries) sources and the transport of extracted material to relevant refinement facilities for production of battery-grade cathode materials as Li, Co and Ni sulfate or carbonate salts. To the best of our knowledge, this study is the first life cycle assessment with primary industrial-scale circular refinement data that includes stepwise, cradle-to-gate comparison of conventional and circular LIB supply chains. With the methodologies and results reported herein, researchers can prioritize major opportunities to improve process efficiencies, practitioners can benchmark their environmental impacts, and policymakers can incentivize best environmental practices in LIB supply chain management. Insights provided by this study can also help recyclers optimize the environmental impacts of their refinement processes.

Results

In LIB supply chains, the refinement step converts the collected feedstocks into battery-grade salts for further manufacturing (**Fig. 2a**). In both conventional and circular supply chains, the refinement pathways vary significantly depending on multiple factors. Five refinement pathways are compared in this study (**Fig. 2b**). While conventional refinement starts with mined ores/brines (1 and 2), circular refinement starts with either end-of-life batteries (1 and 2) or battery scrap (5). Ni and Co in refinement products for subsequent manufacturing can be discrete salts (1 and 3) or mixed compounds (2, 4, and 5).

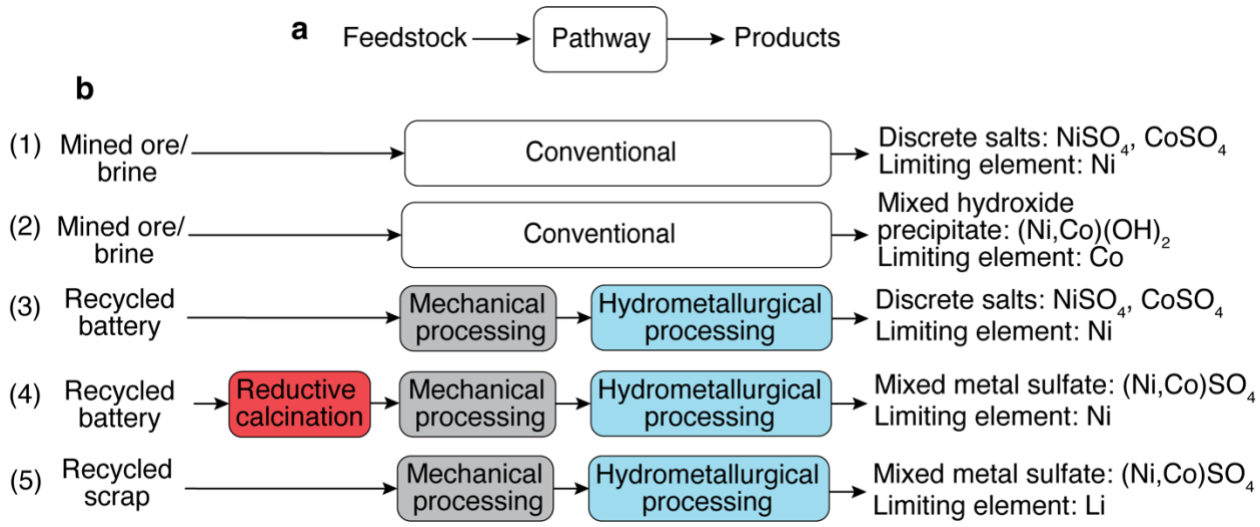


Fig. 2 | Schematic summarizing feedstocks, pathways, and products in refinement analyses. **a,** General schematic showing the feedstock, pathway, and products as a legend for the refinement methods shown below. **b,** Five specific refinement analyses in this study: conventional refining (1 and 2) receives mined ore and brines, and circular refining methods (3−5) recycle from end-of-life batteries or scraps. While all methods produce identical Li_2SO_4 and Al_2O_3, Ni and Co products exist in the form of discrete salts, $NiSO_4$ and $CoSO_4$ (1 and 3), mixed hydroxide $(Ni,Co)(OH)_2$ (2), or mixed metal sulfate $(Ni,Co)SO_4$ (4–5).

Refining lithium-ion batteries into battery-grade materials exhibits lower environmental impacts than production from mined natural materials. The upstream steps of material extraction and transport are considered in later sections. Environmental impacts including energy consumption, greenhouse gas emissions (CO_2-equivalents, CO_2-eq; additional criteria air pollutants are detailed in **Table S1**) and water consumption of refinement pathways in conventional and circular LIB supply chains are compared in **Fig. 3** for the gate-to-gate production of battery-grade cathode materials. State-of-the-art conventional pathways generating discrete salts (Method (1) in **Fig. 2**) are analyzed here. One kg of lithium-nickel-cobalt-aluminum-oxide cathode-equivalent material (NCA-eq) is employed as a functional unit throughout this study for supply chain comparison, accounting for the elemental requirements to produce stoichiometric $LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$. NCA chemistry is selected for the functional unit because it comprises the second-largest category of EV battery chemistries following NMC batteries[7,13], and is projected to utilize less Co compared to NMC[6]. Excluding the environmental impacts of material extraction

and transport steps, the gate-to-gate production of one kg NCA-eq battery-grade material from conventional mined natural materials consumes 193.9 MJ and 77.3 L H_2O while emitting 14.5 kg CO_2-eq (**Fig. 3**). The values of energy consumption and greenhouse gas emissions are comparable with previous studies based on GREET datasets[11,12] (**Fig. S3**). Refinement of mined material concentrate into battery-grade Ni material dominates NCA environmental impacts, representing >57% of total values.

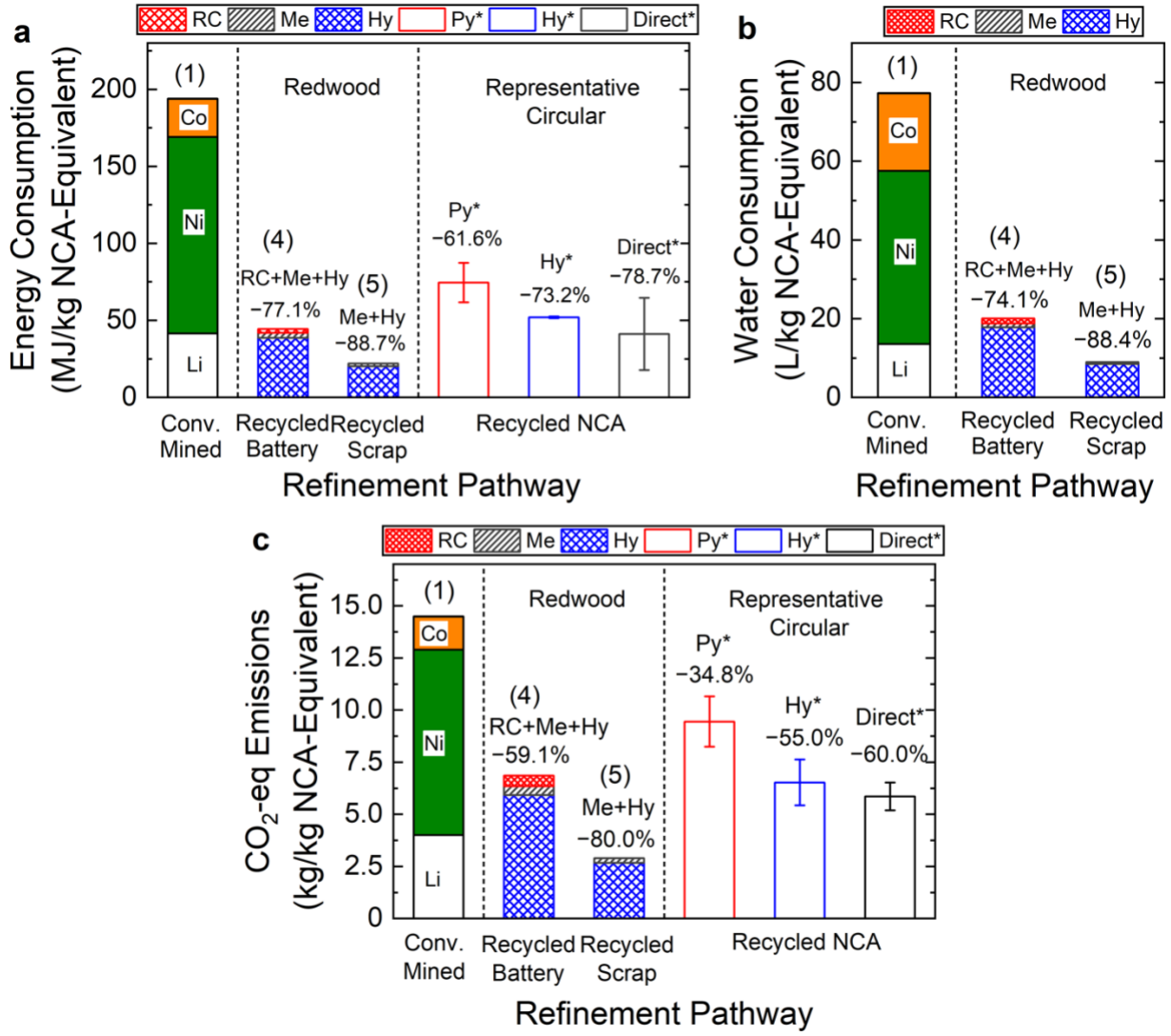


Fig. 3 | Environmental impacts of conventional and circular refining technologies. a, Energy consumption, **b**, CO_2-eq emissions, and **c**, water consumption of gate-to-gate refinement by different pathways for NCA battery-grade salts. Numbers in parentheses labelled on the top of stacked bars denote the refinement methods summarized in **Fig. 2**. The conventional mined

pathway (Conv. Mined) refines natural deposits and produce discrete salts (Method (1) in **Fig. 2**); note that Al is presented on the top of the stacked bars but its contributions are too small to be seen; however specific environmental impacts of each element contributor are detailed in Table S1. Circular supply chains refine from either mixed energized end-of-life lithium-ion batteries collected from consumers (Recycled Battery, Method (4) in **Fig. 2**) or non-energized battery scrap from a production facility (Recycled Scrap, Method (5) in **Fig. 2**), producing mixed metal sulfates. Multi-step circular refinement pathways include mechanical processing (Me, grey), reductive calcination (RC, red), and hydrometallurgy (Hy, blue). RC is an additional processing step for energized batteries and is not used for non-energized recycled scrap. Open bars in the right panels denote environmental impacts of recycling NCA batteries with representative existing pyrometallurgical (Py*), hydrometallurgical (Hy*), and direct recycling (Direct*) methods as comparison, and data are obtained from the literature[8]. Literature data is normalized by the same functional unit in this study, and uncertainties are determined by combining two different battery form factors: pouch and cylindrical (detailed in **Table S14–S15**). The vertical dashed line in each graph demarcates different data types, where the model-based conventional and representative existing pathways are summarized in the left panel, operational data from Redwood Materials are presented in the middle panel, and literature data in the right panel. Note that water consumption has generally not been quantified in previous studies, leading to no literature data panel for **Fig. 3b**. Environmental impacts of material extraction and transport in the supply chains are not included.

The environmental impacts of two circular refinement pathways are presented in each graph in **Fig. 3** for mixed-stream LIB feedstocks processed at Redwood Materials: non-energized production scrap from LIB production facilities (recycled scrap) and energized, end-of-life LIBs collected from consumers (recycled battery). Using a limiting-reagent approach of output products to produce one kg NCA-eq material, energy requirements for processing recycled scrap and recycled battery streams are 22.0 MJ/kg and 44.4 MJ/kg NCA-eq materials, significantly lower than conventional refinement by 88.7% and 77.1%, respectively (**Fig. 3a**). Relatedly, 2.9 and 6.9 kg CO_2-eq/kg NCA-eq materials are generated from scrap and battery streams, respectively, a substantial reduction in CO_2-eq emissions by 80.0% and 59.1% (**Fig. 3b**). Water consumption is also lower by 88.4% for scrap and 74.1% for battery streams relative to the conventional scenario,

resulting from the consumption of 9.0 and 20.0 L H_2O/kg NCA-eq materials, respectively (**Fig. 3c**). Note that while the elemental stoichiometry is identical, the output battery-grade materials vary slightly between conventional (Li_2CO_3, $NiSO_4$, $CoSO_4$) and circular (Li_2SO_4, (Ni, Co)SO_4) supply chains (detailed in **Methods**). Converting the final lithium product to Li_2CO_3 does not substantially change the environmental impacts of the circular supply chains (Supplementary Note 3, **Fig. S3**), and impacts of producing discrete or mixed products are examined in the following section.

To produce battery-grade cathode materials, Redwood Materials uses a combination of reductive calcination (RC), mechanical (Me), and hydrometallurgical (Hy) LIB refinement processes (pathways detailed in **Fig. S2**). The RC process converts energized battery feedstock under certain conditions that leverage heat from exothermic processes and inhibit graphite combustion. This process does not use direct fossil fuel inputs onsite and facilitates subsequent hydrometallurgical refinement into battery-grade materials. Because RC is not required for non-energized LIB production scrap materials, the two feedstock streams (recycled scrap and recycled batteries) are analyzed separately. Energy consumption and CO_2-eq emissions of representative existing recycling pathways from the literature, including pyrometallurgy (Py*), hydrometallurgy (Hy*), and direct recycling (Direct*), are also presented in **Fig. 3** for comparison. In general, the RC+Me+Hy pathway at Redwood exhibits comparable energy consumption and CO_2-eq emissions with Hy and Direct literature values[8], and substantially lower environmental impacts than Py*. Note that traditional pyrometallurgy and Redwood Material's reductive calcination can process energized batteries of varying states of charge, health, and formats with minimal modification, whereas traditional hydrometallurgy may need to discharge energized batteries in salt bath or cryogenically remove electrolyte for safe mechanical processing. While this analysis is focused on Redwood Materials refinement pathways, the methodology can be used to evaluate additional refinement pathways (e.g., hydrometallurgy in **Fig. S3c**), or others that use different material feedstocks, refinement processes, and energy supplies.

Among the few studies that directly compare environmental impacts of circular and conventional NCA refinement using industrial-scale operational data, 35% lower greenhouse gas emissions (**Fig. S3**) are reported for Me+Hy circular refinement compared with the current study[8,11]. However, direct comparison can be inexact due to varying underlying assumptions and data sources. For example, Argonne National Laboratory's GREET and EverBatt models leverage

a combination of technology descriptions from patent applications (the most recent from 2007), literature data on process flow consumptions, industry site visits and surveys, expert advice solicitation, and stated assumptions to form complete pathways. Further, Ciez and Whitacre quantified environmental impacts using output products represented as "metal offsets" for pyrometallurgy or with metals in solution for hydrometallurgy[8] (**Note 3** in **Supplementary Information**), rather than cathode salts in this study. In addition, the previous studies included a portion of recycled metal materials in its conventional supply chain analysis, whereas this work references only mined natural deposits in conventional supply chains to fully deconvolute the environmental impacts[11]. The different conclusions highlight divergent life cycle assessment approaches, processing conditions, and the utility of primary industrial data access over modeling processes from literature sources.

Formats of refinement products influence environmental impacts

Ni and Co are key elements for battery manufacturing, and can be traded in the format of mixed metal salts or discrete salt products between battery refiners and battery manufacturers[14,15]. To examine the influences of the refinement product formats, the environmental impacts of refinement to mixed salt are compared to the refinement to discrete sulfate salts, $NiSO_4$ and $CoSO_4$ (**Fig. 4**). Both conventional and circular refinement pathways are analyzed.

The GREET model is employed to analyze different conventional mining pathways generating different product formats (detailed in **Methods**). In conventional mining, refining Ni-Co ores to mixed hydroxide precipitate, $(Ni,Co)(OH)_2$ (Method (2) in **Fig. 2**), elevates energy consumption and CO_2-eq emissions by 77.% and 89.4%, respectively, over the discrete salts-based pathway (**Fig. 4A** and **4B**, left panels). While the discrete products $NiSO_4$ and $CoSO_4$ are produced from the mixed hydroxide precipitates through additional post-treatment, the very low composition of Co (3.6%) in the latter limits the NCA stoichiometry, thus increasing the total energy cost to generate 1 kg NCA-equivalent materials. On the other hand, water consumption of refining mixed hydroxides is slightly lower (−6.6%) than that in producing discrete salts.

Circular pathways refining batteries to different products are analyzed using the Redwood data by the RC+Me+Hy process and the modeling of a representative battery recycling method combining mechanical and hydrometallurgy (Me+Hy) refinement (Method (3) in **Fig. 2)**. The Redwood process refines recycled batteries to mixed metal sulfate, $(Ni,Co)SO_4$, whereas the

representative Me+Hy produces discrete $NiSO_4$ and $CoSO_4$ as the products. The RC pathway (RC+Me+Hy) exhibits lower energy consumption (−72.3%), CO_2-eq emissions (−39.5%), and water consumption (−12%) relative to the Me+Hy pathway (**Fig. 4**), because it avoids additional treatment separating (Ni,Co)SO_4 to discrete salts. Overall, our results indicate that refining batteries to mixed metal salts instead of discrete salts can substantially save environmental impacts while still satisfying the needs of circular LIB supply chains. Our findings also provide important insights to optimizing plant-scale battery refining operations. In the following sections, mixed salt-based pathways are analyzed for refinement.

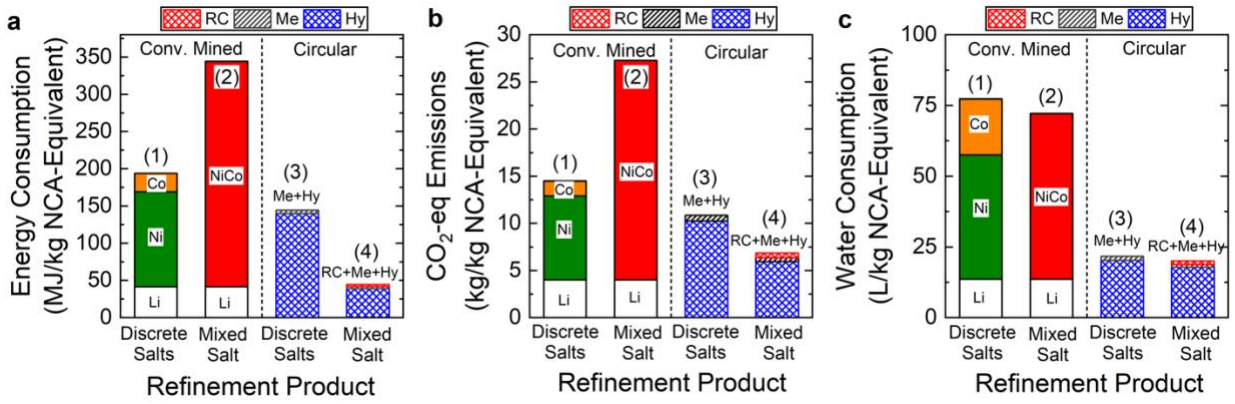
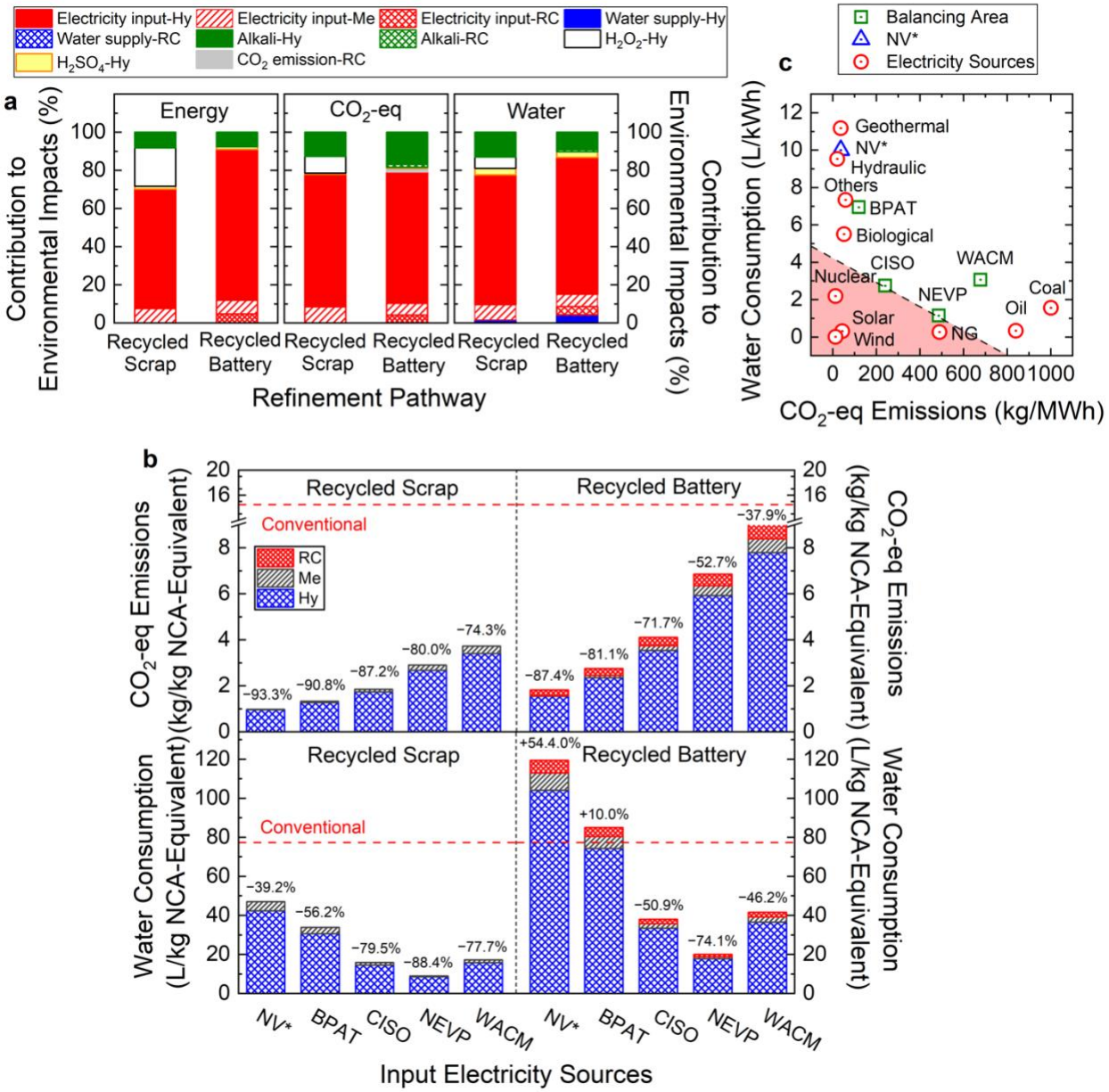


Fig. 4 | Influences of refining products on environmental impacts in circular refining. a, energy consumption, **b,** CO_2-eq emissions, and **c,** water consumption. Left and right panels denote conventional (Conv. Mined) and circular pathways refining end-of-life batteries to discrete Ni and Co salts, or mixed Ni-Co salts. Note that Al is presented on the top of the stack bars of conventional supply chains but its contributions are too small to be seen (detailed values in **Table S1**). Numbers in parentheses labelled on the top of stacked bars denote the refinement methods summarized in **Fig. 2**.

Electricity consumption dominates the environmental impacts of lithium-ion battery circular refinement. The relative environmental impacts of input consumables (e.g., energy, water, commodity chemicals) in the gate-to-gate refinement processes are disaggregated in **Fig. 5** (additional criteria air pollutants in **Tables S2–S3, Figs. S4–S5**). Note that the embodied environmental impacts of electricity consumption in **Fig. 3** are based on the Nevada Power Company (NEVP) at the Redwood Materials location. Electricity consumption is a principal factor

dominating the environmental impacts. For both LIB feedstock pathways (Methods (4) and (5) in **Fig. 2**), electricity accounts for 70.3–91.0% of the total energy consumption, 71.8–79.1% of the total CO_2-eq emissions, and 54.3–63.6% of water consumption (**Fig. 5a**). For both feedstocks, Hy processes comprise the majority of environmental impacts, contributing more than 87.3% to energy consumption, 86.3% to CO_2-eq emission, and 88.8% to water consumption. Notably, the additional RC step required for processing energized batteries only marginally contributes to CO_2-eq emissions (7.4% of total). Unlike conventional pyrometallurgical processes that require external energy sources[8,16], the RC process is primarily autothermic because it leverages process heat released from exothermic reactions of the LIB materials[17,18]. In addition to electricity consumption, chemical reagents used in circular refinement processes also contribute to embodied environmental impacts. Alkali reagents used to precipitate metals contribute between 19.0–21.3% of environmental impacts (largest relative contribution to water consumption). H_2O_2 is used to reduce high-oxidation state metal compounds for hydrometallurgical leaching of scrap material, and accounts for 11.3–20.1% of environmental impacts (largest relative contribution to energy consumption).



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309 **Fig. 5 | Breakdown of environmental impacts of lithium-ion battery (LIB) recycling using**
310 **different input electricity sources.** **a**, Contributions to the environmental impacts of recycling
311 processes using electricity from the Nevada Power Company, including energy consumption, CO_2-
312 eq emission, and water consumption by different input consumables used in circular processes for
313 LIB feedstocks from production scrap (recycled scrap) and used energized batteries (recycled
314 battery) used by Redwood Materials. **b**, Environmental impacts of input electricity sources on
315 CO_2-eq emissions and water consumption in the LIB recycling operations employed by Redwood
316 Materials methods for production scrap and energized batteries. CO_2-eq emissions and water

consumption are based on the resources consumed by unit electricity generated from a Nevada renewable energy tariff (NV*), Bonneville Power Administration (BPAT), California Independent System Operator (CISO), Nevada Power Company (NEVP), and Western Area Power Administration: Colorado-Missouri (WACM). The red dashed lines denote the environmental impacts of the analogous conventional refining process. Note that influences of energy sources on environmental impacts are only presented for the circular supply chains, but not for conventional supply chains. Specific environmental impacts presented in the figures are detailed in **Table S5**. **c,** Tradeoff relationship between embodied water consumption and CO_2-eq emission by different power sources, including electricity grids in different locations (⊙), purely power sources (⊡), and Nevada renewable energy tariff (NV*, ◬). The red dashed line denotes the lower bound of the water-CO_2 performance, i.e., the existing electricity grids that have the lowest water consumption and CO_2-eq emission simultaneously, and the green shaded area covers the power sources that can transcend the current limit of water-CO_2 performance.

Because electricity dominates the environmental impacts of LIB recycling processes, a comparison of electricity grid balancing areas that emit a range of CO_2-eq emissions per MWh (averaged for 2019)[19-21] are examined in **Fig. 5b** (additional criteria air pollutants detailed in **Table S5**). Substituting NEVP electricity with other balancing areas including Bonneville Power Administration Transmission (BPAT), California Independent System Operator (CISO), Western Area Power Administration of Colorado-Missouri (WACM), and a 100% renewable energy tariff in Nevada (NV*), yields a significant reduction in CO_2-eq emissions of up to 93.3% (recycled scrap) and 87.4% (recycled battery) relative to conventional refinement (**Fig. 5b**). Conversely, employing low-carbon electricity grids can increase water consumption compared with NEVP-based operation, following the order of NV* > BPAT > WACM > CISO > NEVP (**Fig. 5b**). Note that NV*- and BPAT-based circular refinement processes exceed the water consumption level of conventional refinement due to significant contributions from hydro- and geothermal power. Further investigation into the grid electricity sources of balancing areas reveals a tradeoff between CO_2-eq emissions and water consumption based on electricity generation type (**Fig. 5c)**; most electricity sources with relatively low CO_2-eq emissions (e.g., those based on bio-, hydro-, or geothermal energy) exhibit high water consumption, and vice versa. This tradeoff also explains the different influences of electricity source on environmental impacts of the Redwood Materials

refinement step and other pathways (**Fig. S3d**). However, the electricity sources for each balancing area will affect both CO_2-eq emissions and water consumption. For example, because NEVP-based electricity includes a relatively large proportion (70%) from CO_2-eq emissions-intensive natural gas with low water consumption, a switch to hydro-intensive (73%) BPAT electricity decreases CO_2-eq emissions while increasing water consumption.

Environmental impacts of material extraction and transport are significantly lower in circular lithium-ion battery supply chains than in conventional supply chains. Upstream of gate-to-gate refinement are material extraction and transport to refinement facilities (**Fig. 1b**). Environmental impacts of these upstream steps are analyzed for two representative LIB chemistries and battery use cases: NCA in EV battery packs, and lithium cobalt oxide ($LiCoO_2$ or LCO) in smartphones. California is chosen to assess circular extraction because it has the largest population and EV market share of any state in the U.S.[22,23]. Smartphones are considered extracted when collected, aggregated, and transported from all California residents (analyzed per census block) to the nearest existing collection facility (CF)[24]. The analytical model for this circular extraction is depicted in **Fig. 6a**, where a shortest-path route for collection from block group to CF is modeled[22]. To quantify conventional material extraction environmental impacts from mining, global supply chain data are adapted from GREET (**Table S6–S7**)[12]. Smartphone extraction in the circular supply chain emits only 0.0189 kg CO_2-eq/kg LCO-eq, significantly lower than conventional mining (1.96 kg CO_2-eq/kg LCO-eq) by 99.0%. Energy and water consumption are similarly lower in the circular supply chain (**Table S9**).

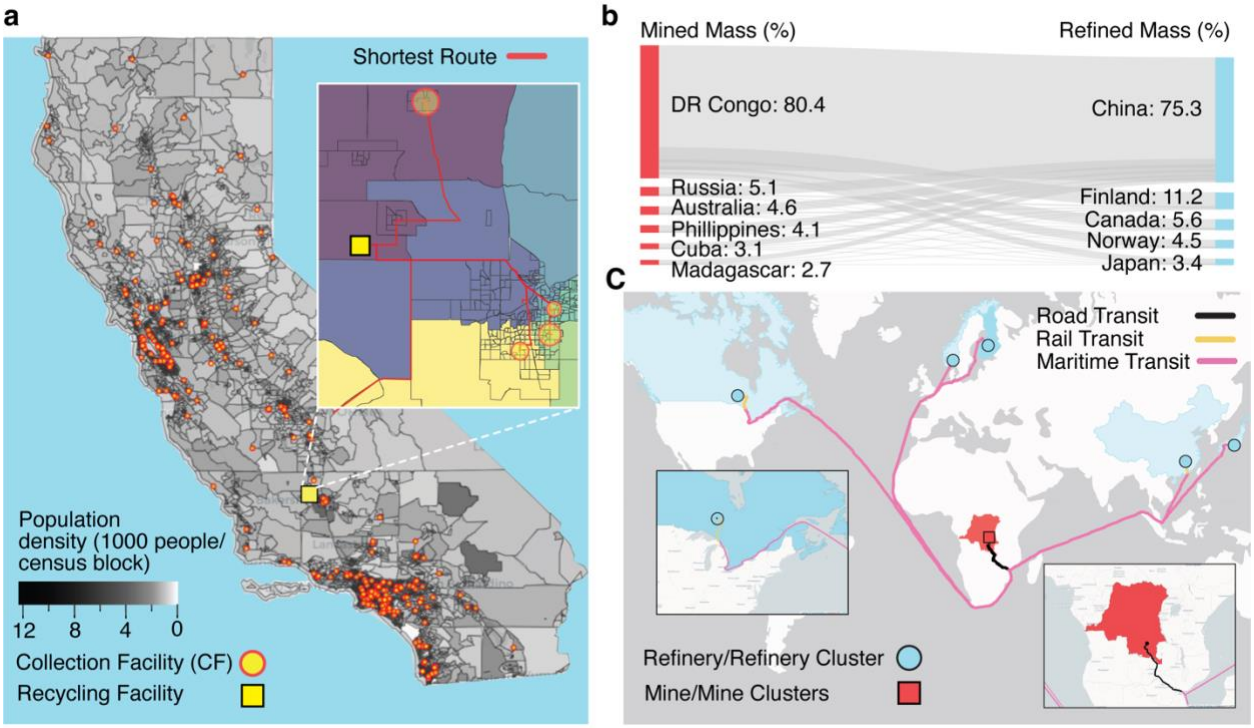

Fig. 6 | A logistics model for assessing upstream environmental impacts of extraction and transport in circular and conventional lithium-ion battery supply chains. a, Modeled circular extraction of LCO-based smartphones from every census-block group based on population to the closest, existing private or municipal collection facility (CF) using a shortest-route algorithm. Inset details modeled circular transport of smartphones aggregated at CFs and then transported to a central recycling facility at the center (gravity point) of the California population by the shortest route (red lines). Colors of block groups indicate the catchment area of a specific CF, where CF size shows the relative number of smartphones collected in 2019. **b**, A weighted distribution estimate of international transport logistics for conventional supply chains between mining and refining countries based on cobalt productivity in the top Sankey diagram. **c**, An example of transport logistics for cobalt mined and aggregated in the Democratic Republic of the Congo (DRC) and then shipped via primary road, train rail, and maritime routes using a shortest-distance path to major refinery locations, with insets showing the degree of detail considered. Similar analyses were performed for Li, Ni, Co, and Al. Inserts present more detailed transit routes in DRC and Canada.

After extraction, LIB material concentrates are transported along domestic and international routes by truck, train rail, and maritime cargo ship to refinery locations (**Fig. S7** and **Table S8–S15**; complete references in **Supplementary Information)**. An algorithm is developed to quantify environmental impacts based on a weighted distribution of participating countries and the shortest distance along major transport routes (the case of cobalt is presented as an example in **Fig. 6b.**) Conventional mine-to-refinery environmental impacts are calculated for one kg of embodied Li, Ni, Co, and Al metal (**Table S7**). While transport emissions for Li, Ni, and Co range from 5.4–6.4 kg CO_2-eq/kg embodied metal, Al is three times lower. For the circular case applied to California, smartphones and EV battery packs collected at CFs are transported to a hypothetical central LIB circular refinement facility at the population-weighted center (i.e., gravity point) of California (near Bakersfield)[22]. In conventional supply chains, transporting mined material concentrates accounts for 3.68 kg CO_2-eq/kg NCA-eq and 4.32 kg CO_2-eq/kg LCO-eq. By comparison, emissions for the transport of aggregated end-of-life NCA EV battery packs (i.e., not disassembled) and LCO smartphone batteries (not separated from phones) to a circular refinement facility are 0.073 kg CO_2-eq/kg NCA-eq and 0.47 kg CO_2-eq/kg LCO-eq, 98.2% and 89.1% lower than transport of mined concentrate, respectively. The reduction in CO_2-eq emissions is attributed to differences in elemental concentrations of transported materials and aggregate transport distance (e.g., a weighted average of 224 km for circular NCA-eq materials, and 57,600 km for conventional NCA-eq materials).

The refinement step dominates environmental impacts of circular and conventional supply chains. Combining material extraction, transport, and refinement steps yields a cradle-to-gate comparison of the most differentiated steps of conventional and circular LIB supply chains for producing battery-grade cathode materials (**Fig. 7**). Here the environmental impacts of the LIB refinement step in California are analyzed for a hypothetical scenario employing the same circular multi-step refinement technologies as Redwood Materials (i.e., RC+Me+Hy) in Nevada, but using California (CISO) electricity to produce battery-grade cathode materials. A circular supply chain in California for NCA EV and LCO smartphone batteries lowers energy and greenhouse gas emissions by at least 47.3% and water consumption by over 37.6%. In the case of recycling NCA EV batteries in California, the entire cradle-to-gate greenhouse gas emissions of the circular supply chain are lower than the transport emissions of mined concentrate in conventional supply chains

(**Fig. 7**, **Table S8)**. Circular production of LCO-grade materials leads to higher environmental impacts than that of NCA-grade materials based on the mixed-stream feedstock composition processed by Redwood Materials. Overall, upstream steps (extraction and transport) contribute marginally to the total environmental impacts of both circular supply chains, accounting for ≤4.9% CO_2-eq emission, ≤8.2% energy consumption, and ≤0.24% water consumption. Accordingly, the refinement process dominates the environmental impacts of the circular supply chain. In contrast, upstream steps in the conventional supply chain play a larger role (still smaller than refinement) in cradle-to-gate environmental impacts, contributing between 7.8–31.0% to the environmental metrics considered (**Table S8**).

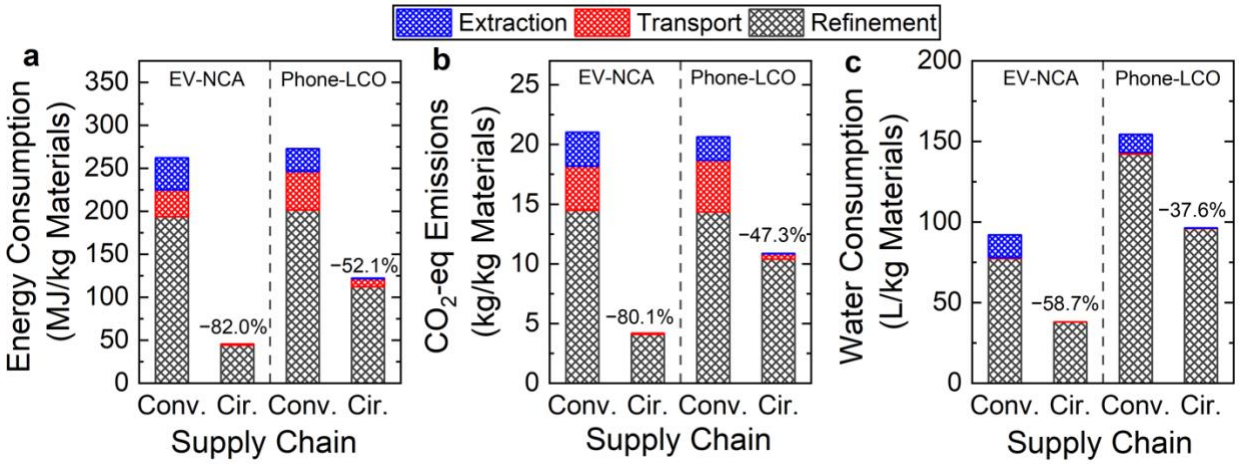

Fig. 7 | **Cradle-to-gate environmental impacts of different supply chains. a,** Energy consumption (left), **b,** CO_2-eq emissions (middle), and **c,** water consumption (right) of conventional (conv.) and circular (cir.) supply chains by step including material extraction, transport, and refinement. NCA-eq cathode used in electric vehicles (EV-NCA, left panels) and LCO-eq cathode material used in smartphones (Phone-LCO, right panels) are provided. Environmental impacts of refinement are analyzed based on electricity generated from balancing grid authority CISO and upstream supply chain steps (extraction and transport) are based on data from GREET and transport models developed in the preceding section and depicted in **Fig. 6**. Specific environmental impacts of each step are detailed in **Tables S5–S7**.

Discussion

This study is the first quantitative cradle-to-gate life cycle assessment of disaggregated conventional and circular LIB supply chains that include primary data from an industrial-scale

recycling facility. Practical LIB feedstock and refinement pathways are analyzed from recycling company (Redwood Materials) and modeling is employed to examine the environmental impacts of upstream material extraction and transport steps. The analysis reveals that refining end-of-life LIBs into battery-grade cathode materials exhibits lower environmental impacts than conventional refinement of mined materials, mixed salts products are more beneficial for circular refinement, and the source of input electricity is the principal factor governing circular refinement environmental impacts. Upstream circular supply chain steps contribute marginally to overall environmental impacts, and the refinement step comprises the largest source of cradle-to-gate environmental impacts.

Disaggregated analysis of LIB refinement pathways at Redwood Materials provides important insights into the performance and potential of different refinement processes. While pyrometallurgical processing is widely considered as more environmentally intensive than hydrometallurgy, Redwood Materials' RC pathway exhibits much lower environmental impacts than current Hy-containing pathways reported in practice and in literature (**Fig. S3**). The optimized conditions of RC processing minimizes the combustion of carbon-containing LIB materials, significantly reducing CO_2-eq emissions while simultaneously generating products that are amenable for hydrometallurgical separation. Because chemical consumables such as H_2O_2 are important contributors to hydrometallurgy, environmental impacts of Hy processes could be reduced through more sustainable (e.g., electrochemical) production methods[25]. Our findings also advocate the refinement products of mixed metal sulfates over the single salts, indicating that the further separations among Ni and Co salts can be avoided. An emerging alternative LIB recycling technology, "direct recycling", recovers functional battery materials without decomposition into substituent elements, and is reported to exhibit comparable environmental impacts to Redwood Materials methods[9]. However, direct recycling is still under development and warrants further assessment after process optimization and industrial-scale implementation.

Electricity greatly influences environmental impacts in LIB circular refinement, and the variability among grid electricity sources elucidates a tradeoff between CO_2-eq emissions and water consumption (**Fig. 5**). Therefore, considering water consumption and CO_2-eq emissions is necessary for selecting recycling facility locations, particularly in water-sensitive or emissions-sensitive scenarios. Further examination suggests that the tradeoff is primarily driven by water-intensive hydroelectric and geothermal electricity in certain locations versus CO_2-intensive coal

and natural gas in others, implying that increasing the proportion of electricity from nuclear, wind, and solar energy sources simultaneously reduces CO_2-eq emissions and water consumption relative to existing balancing areas (**Fig. 5**).

Analyses of upstream environmental impacts inform better operations for future resource-saving extraction and transport. Conventional mining and concentrating of ore or brine is resource-intensive due to the low natural concentrations of critical materials (0.01–1%), while critical material concentrations for transport rise to 3–15% after beneficiation. Further concentrating materials near mine sites or building reinterests closer to sources can efficiently reduce environmental impacts of the conventional mined materials. In contrast, smartphones contain 5% LCO material by mass, with the batteries themselves at 24% LCO[25]. Circular material extraction via LIB collection decreases environmental impacts by 99% versus conventional. A "shortest-route" approach is used in this study to quantify the environmental impacts of battery extraction and transport supply chain steps. Practical battery collection operations will likely vary based on route selection and preprocessing strategy further influencing environmental impacts[26]. For example, the disassembly of collected EV battery packs or removal of smartphone batteries from devices prior to transport to a recycling facility can increase energy usage through extraction but reduce environmental impacts by lowering transportation weight (**Table S10**). Trucks are used as the primary vehicle for transport analysis given regulatory concerns that consider LIBs hazardous material in many transportation scenarios[27]. However, alternative transport like railway can further lower environmental impacts by approximately four times versus trucking (**Tables S12**). Upstream process optimization of environmental impacts warrants further investigation, such as the active area of high-throughput automation of LIB extraction from non-standardized devices and EV battery packs or rapid assessment of LIBS for second life uses.

While the current cradle-to-gate study is focused on Li, Ni, and Co as the major output materials, the potential benefits of extracting additional LIB constitutive elements from ore (e.g., Cu and Co in Cu-Co sulfides) or from LIBs (e.g., Cu or Mn) warrants further investigation. Additionally, the same mixed-stream LIB feedstocks consumed at Redwood Materials are used to quantify NCA- and LCO-equivalent values, and results would vary for single-stream LIB feedstocks. Generally, the incremental benefits of extracting additional critical materials from concentrated sources like LIBs can offset the environmental impacts of both supply chains.

As the prevalence of LIBs grows in the mobility sector and beyond, strategic placement of domestic LIB collection, refinement and manufacturing facilities can further minimize future environmental impacts by considering heterogenous LIB growth by location, collection approach, transportation distance, and electricity source for refinement processes. As LIB production scales, policies informed by consumer surveys, focus groups, pilot testing, and diverse stakeholder engagement will be needed to research and scale battery collection[28]. Business models for collection of all LIB types and sizes will likely vary from manufacturer-led to municipal or private collection programs. In addition to collection costs, the varied scale of collection requires further investigation, particularly for localized environmental impacts. Notably, analogous economic and environmental impacts to local ecosystems of conventional mining are not considered in this analysis, and warrant future studies[29]. Additionally, designing and manufacturing LIBs for recycling in a circular economy can reduce resource usage identified in this study[30]. Future efforts should also focus on optimizing refinement processes for subsequent steps of the circular supply chain in LIB manufacturing, product performance, and economic cost.

Methods

Goal and scope. The goal of this study is to compare stepwise cradle-to-gate environmental impacts (energy consumption, CO_2-eq emission, and water consumption) for two supply chains: a conventional, linear supply chain fed by natural mined material for refinement into battery materials, and a circular supply chain fed by LIBs. Both supply chains produce battery-grade cathode materials. A cradle-to-gate analysis of the whole supply chain considers steps of material extraction, transport, and refinement, and gate-to-gate analysis investigates the refinement step, which is focused on in this study. A gate-to-gate scope is broadly defined as the boundary surrounding processing facility operations. In this analysis, gate-to-gate refinement only considers direct processing (e.g., alteration, concentration, precipitation) of the feedstock material once it is extracted from its original state and transported to the refinement location (shown in **Fig. 1b**). For Redwood Materials, this scope includes mechanical processing, reductive calcination, and hydrometallurgy (**Fig. S2**). The system boundary does not include other operations outside of the direct refinement processes as discussed in study limitations below.

Two LIB feedstock streams are evaluated: (1) battery production scrap and (2) mixed, spent LIBs from consumers (**Fig. S2)**. The study scope upstream of the gate-to-gate supply chain step completes cradle-to-gate analysis, and includes both material extraction and transport steps. For conventional extraction, GREET is used for quantifying the environmental impacts of mining. Transport between supply chain steps and for the circular extraction step are quantified using a logistics transportation model developed in this study, where limitations are summarized below.

Methodology. An attributional life cycle assessment is conducted to quantify and compare conventional and circular LIB supply chains for the production of battery cathode materials. This analysis complies with the International Organization for Standardization (ISO) 14040 standards but omits conversion to environmental impact indicators and external review[31]. Data for conventional material extraction (e.g., mining) and refining are adapted from the Argonne National Laboratory's Greenhouse gases, Regulated Emissions, and Energy use in Transportation (GREET®) 2021 model. GREET and the ecoinvent 3.3 database[32] are employed for life cycle inventory data of chemical consumables for the conventional and circular supply chains.

To assess circular LIB refinement, primary operational data detailing energy, water, on-site emissions, and consumables usage are provided by Redwood Materials and normalized to mass flows of the different elements of interest in input feedstocks and output products. A representative prevailing circular refinement, Method (2) in **Fig. 2**, is modeled with the software HSC Sim[33], based on the technical procedures available in the literature[34-37] and the practical feedstock amount received by Redwood.

Conventional refinement was modeled by aggregating the environmental impacts of the individual refining pathways for each LIB cathode element (**Table S1**), normalizing by the mass of the individual element of interest within the output product (e.g., Li in Li_2CO_3) and then normalizing again by the mass of that element in the functional unit for this life cycle assessment (defined in the next section). For elements where more than one pathway of production exists in the GREET model (i.e., Ni and Li), the overall environmental impacts are calculated by averaging pathways weighted by their respective share of global production (45% Li production from brine and 55% from ore, and 60% Ni production from mixed hydroxide precipitate and 40% from Class 1 Ni). Both discrete and mixed output products are considered. Discrete salts from conventional refinement are Li_2CO_3, $NiSO_4$, $CoSO_4$, and Al_2O_3; alternatively, $(Ni,Co)(OH)_2$ is considered as

the mixed product. Lithium outputs produced by Redwood Materials are Li_2SO_4 (environmental impacts for converting to Li_2CO_3 are detailed in Supplemental Information **Note 3**), and other outputs exist as mixed metal sulfates of (Ni, Co)SO_4 or as Al_2O_3 and $Al(OH)_3$. With additional treatment further transform the mixed metal sulfate into separate Ni and Co compounds, discrete salts as $NiSO_4$ and $CoSO_4$ are analyzed based on modeling of a prevailing Hy+Me refinement pathway. In the cradle-to-gate analysis, material transportation between stages was not included because it was not consistently available in the GREET model. Mixes vary between elements, as well as between pathway stages. For example, crude production of $Co(OH)_2$ uses a distributed electricity source in the Democratic Republic of the Congo, and the refinement of these materials into $CoSO_4$ and $CoCl_2$ uses a distributed electricity source in China. While exploring the sensitivity of environmental impacts for conventional battery material production is important, it is beyond the scope of this paper, and instead, this work focuses on the sensitivity of electricity sources in U.S.-based LIB recycling. See **Supplementary Data File A** for the breakdown of the conventional refining data workflow.

Defining functional units. Functional units standardize comparisons of the resource consumption and emissions in life cycle assessments. In this study, two functional units are considered in this assessment to normalize environmental impacts between conventional and circular supply chains: the battery-grade material required to make one kg of stoichiometric lithium nickel cobalt aluminum oxide ($LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$, NCA-eq) and lithium cobalt oxide ($LiCoO_2$, LCO-eq) cathode material. Mass was selected as the primary normalizing factor because any energy-based functional unit (e.g., per kWh) could vary based on battery manufacturing and cycling characteristics. The NCA chemistry was selected because reports suggest future cathodes may utilize less Co compared to NMC batteries in EVs, and NCA comprised the second-largest category of EV battery chemistries in 2016, following NMC batteries[7]. LCO is a representative chemistry used in handheld rechargeable devices (e.g., cellphones and laptops) which are currently available to recycle in larger quantities than EV LIBs. The environmental impacts of other LIB-relevant materials (Cu and Mn) in conventional supply chains can be found in **Table S7**.

In both conventional and circular supply chains, the extraction, transport, and refinement steps are converted into environmental impacts metrics for the production of battery-grade materials and normalized by NCA and LCO functional units. A limiting reagent approach is used

to quantify the environmental impacts of a functional unit in circular refinement pathways. According to current multi-step pathways using mixed-stream LIB feedstocks (either recycled scrap or recycled battery), the Li output is the limiting element for creating one kg of NCA-eq materials from recycled scrap, where other refined elemental products are produced in excess. Relatedly, Ni is the limiting output element from recycled batteries. For multi-step refinement processes, the recovery rate of Ni and Co is 95% and for Li is 92%. Additionally, a sensitivity analysis of environmental impacts from circular refinement is conducted based on facility location in different grid balancing areas and their associated electricity sources.

Life cycle inventory and assessment. The life cycle inventory (LCI) data for conventional mining pathways are normalized by each critical metal element: Li, Ni, Co, Al, Cu, and Mn (**Table S7**). The LCI for consumables in the Redwood process are adapted from the GREET 2021 model and ecoinvent 3.3 (**Table S2**).[12,32] The LCI for the Redwood processes also lists water consumption and criteria emissions for different electricity sources by grid balancing areas in the Western U.S. (**Table S6**). Three categories of environmental impacts are detailed in this study: energy consumption, air pollutant emissions, and water consumption. Energy consumption includes the input electricity for different applications and the energy required to produce required consumables. Criteria air pollutant emissions include the embodied emissions generated by the production of input electricity and the consumed reagents. CO_2, CH_4, CO, NO_x, N_2O, SO_x, PM_{10}, and $PM_{2.5}$ are the air pollutants provided in the GREET model and considered here. The greenhouse gas emissions are reported as CO_2 equivalents (CO_2-eq) summing CO_2, CH_4, and N_2O weighed by the corresponding 100-year global warming potential (GWP). Water consumption considers the withdrawn water that is not returned to the original source, and both the input city water usage and the embodied water consumption in electricity generation and the manufacturing of consumable materials are included.

Estimating environmental impacts of material extraction. For conventionally mined ore and brine, energy consumption, CO_2-eq emission, and water consumption values are separated for the material extraction processes found in the GREET model. For the circular extraction case, LCO-based smartphones are assumed to be collected and transported to existing private and municipal collection facilities (CFs) from each census block group in CA, assuming every person owned a cell phone and purchased a new phone every three years. A shortest-route algorithm was used for

collection at the closest municipal collection facility determined by *k*-means clustering (**Note 4** in **Supplementary Information**).

Estimating environmental impacts of material transport. In the conventional supply chain, a network model of primary transport routes is established that connects mines to refinery locations for Li, Co, Ni, and Al on a country-level basis (**Tables S8–S15**) because the amount of mined material transported from each mine to each refinery was not known. The distances of the shortest-path routes are calculated between mines and refineries by country, predicated on the closest available modes of transport (including road, rail, and maritime). A major mine cluster or refinery location was selected to represent country-level transport values (**Tables S14–S15**) based on production volumes, and distances are quantified between international destinations. These distances are used to calculate the energy consumption, CO_2-eq emission, and water consumption associated with transportation of critical materials as mined concentrate. Mined concentrate is ore or brine that is concentrated locally beyond natural concentration values to reduce weight for transport to a refinery. By considering the total elemental mass and elemental weight percentage of the mined concentrate transported along a route (**Tables S14–15**), the environmental impacts on a per-element basis are calculated as a global weighted average (**Table S11**) with additional process details in **Supplementary Information**.

For the circular case applied to California, end-of-life EV NCA LIBs are aggregated at one CF per county closest to its centroid, where county-level data was the most granular data available. All smartphones are aggregated at their nearest CFs. Aggregated smartphone and EV batteries are assumed transported via truck to a single recycling facility located at the gravity point of California's population based on census block-level data (detailed in **Note 4** in **Supplementary Information**). The mass-distances traveled are converted to energy consumption, CO_2-eq emission, and water consumption (**Table S8–S9**).

Summary of study limitations. Limitations based on key assumptions of supply chain steps (extraction, transport, refinement) in each supply chain (conventional and circular) are briefly discussed in this section.

Extraction. Mining data in conventional supply chains in GREET often only refer to one mining country per material, meaning the global supply chain is not well captured. Transport required between mining unit processes (e.g., crushing, flotation, and concentration) prior to

refinement is excluded from the analysis due to the lack of information in GREET. In collection of end-of-life batteries in smartphones, inefficient transport to a CF (e.g., driving each smartphone individually or taking longer transport routes to a CF) is not considered. In addition, all end-of-life EV battery packs are assumed to be driven to each CF in their original vehicles, which is attributed to the "product use" stage instead of extraction in life cycle assessment; therefore, zero CO_2-eq emissions are assumed for the extraction step of EV batteries.

Transport. An inter-country LIB material transportation assessment is performed as a weighted distribution between all major mining and refining countries. Results are sensitive to the weight percentage of critical material in transported concentrate found in **Tables S14-S15**. Transport between a domestic mine and refinery is not considered, resulting in net zero use of resources in such cases. The resources required to separate an embedded battery from its device prior to a refinement facility is not considered in a circular supply chains. Similarly, the effect of transporting only LIBs separated from the devices is not considered. Incorporating the domestic transport and separation operations can increase environmental impacts.

Refinement. Refinement data in conventional supply chains are limited to the country scenarios reported in GREET, and transport between refinement unit processes is not included. Ancillary processes (e.g., transport between unit processes) beyond direct refinement unit processes and embodied resources of the capital equipment used for material refinement are not considered for the circular supply chain. The chemical formats of output products differ between the conventional and circular supply chains, but converting them to the same products will not substantially change the results due to the similarity between the cathode salts of the two supply chains (**Note 3** in **Supplementary Information**).

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Supplementary Information

Life cycle comparison of industrial-scale lithium-ion battery recycling and mining supply chains

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Supplementary Note 1. Drivers for recycling lithium-ion batteries

Projected LIB growth and supply

Historic and projected values for global lithium-ion battery production by year are shown in **Fig. S1**. Average values are presented as stacked bars by category and uncertainties represent minimum and maximum reported values that highlight the range in uncertainty of projections. In all projections, electric vehicles comprise the majority of LIB growth in the 2020s. The variation in reported values is due, in part, to data selection and availability, such as including plug-in hybrid electric vehicles in some projections while only considering fully electric vehicles in others.

Projected major supply-demand deficits of critical materials are extracted from industry reports and other sources[1-7]. The years where the supply-demand gap falls below zero and does not recover to a positive value is shown in **Fig. S1** for Li, Ni, Co, and Cu. The range of deficit years shows the supply and demand uncertainty of these critical elements, while the nearness of these years to the publication date of this manuscript highlights the immediacy of supply risk and deficit concerns.

Embodied value of recycled materials

Economic value ranges for commonly recycled commodities (i.e., glass, paper, plastic, and metal cans) in **Fig. 1a** are based on historic monthly values in the U.S. from January 2018 to December 2021 considering inflation[8]. 90% confidence intervals, represented by uncertainty bars, show variation in pricing not only based on time, but also based on commodity variety in each category. Because their constituent elements can be reused indefinitely, metal cans experience recycling rates between 55–70%, whereas single-use plastic products are much lower at 3–30%. Fundamentally, carbon atoms have minimal intrinsic economic value due to abundance and the degradation of carbon-carbon bonds over multiple recycles.

Historic monthly LIB-relevant elemental value data is extracted from online databases[9], and weighted by the composition of EV 27-kWh battery packs provided by GREET[10]. Values are also adjusted for inflation based on Dec 2021 USD. The relative composition of cathode powder in smartphone batteries is taken from literature[11], and used to estimate the composition of an LCO-based smartphone battery. In both cases, the entire mass of the battery or battery pack is not included in reported recoverable value, only those elements shown in the figure. The reported values are weighted by their proportion per kg of battery or battery pack. As such, additional recoverable material (i.e., graphite, battery management system components) may still add additional economic value but are not considered here. Note that, by March 2022, prices for many critical elements were near or exceeding their 90% confidence maximum, while lithium carbonate prices had nearly doubled.

Supplementary Note 2. Emissions and water embodied in electricity consumption

The year 2019 was chosen as the reference year for electricity consumption to remove any major anomalies caused by the SARS-CoV-2 pandemic. For each environmental impact category (energy, criteria air pollutants, and water), environmental impact conversion factors were used to calculate total impacts of electricity in each balancing area based on the composition of different electricity sources (e.g., natural gas, solar, hydroelectric). Total electricity production and consumption in each balancing area, as well as exchanges with other balancing areas, were then used to compute the environmental intensities embodied in electricity consumption for each balancing area. This calculation generalizes an approach that was previously developed for CO_2, SO_2 and NO_x[12-14], and now incorporates additional data sources for electricity-embodied water[15] and criteria air pollutants (SO_x, $PM_{2.5}$, PM_{10}, CO, CH_4, N_2O)[15,16] Results of environmental intensities are summarized as an electricity life cycle inventory in **Table S4**, and the original data sources are detailed in **Supplementary Data Files A** and **B**.

Supplementary Note 3. Environmental Intensity Differences

Compounds: Lithium carbonate versus Lithium sulfate

Because many studies use lithium carbonate (Li_2CO_3) as a reference compound as opposed to lithium sulfate (Li_2SO_4, the primary reference compound in this manuscript), the environmental intensities of both are compared for the reductive calcination pathway (RC+Me+Hy) and the hydrometallurgy pathway (Me+Hy) for energized LIBs. The results of this sensitivity analysis can be found in **Fig. S3a** and show that relative to Li_2SO_4, the production of Li_2CO_3 exhibits higher energy consumption intensity 3.9–28.8%, CO_2-eq emissions intensity 6.4–29.3%, and water consumption intensity 2.1–21.9%.

Pathways: Explaining discrepancies in literature of similar pathways

Few previous studies have quantified the environmental impacts of recycling NCA LIBs. Results from a previous study assessing the CO_2-eq emissions of different prevailing circular refinement pathways are compared to the results from the current study[17] (visualized in **Fig. S3c**). After normalizing the literature data by the same functional unit as in the current study, the previous study reports 7.53 ± 0.30 kg CO_2-eq/ kg NCA-eq, which is 48% lower than our result (14.5 kg/kg NCA-eq). This discrepancy is due to the inclusion of recycled elemental inputs for refinement into their analysis rather than considering only mined material[17].

CO_2-eq emissions of prevailing circular refinement pathways, including pyrometallurgy (Py), hydrometallurgy (Hy), and direct recycling (direct), are summarized in **Fig. S3c**. A mechanical and hydrometallurgy refinement pathway (Me+Hy) used to recycle energized batteries (recycled battery) is analyzed to model the existing circular refinement pathway in industries. In general, the Me+Hy pathway in Redwood exhibits 66.2% higher CO_2-eq emissions than literature values[17]. Because the embodied CO_2-eq emissions are similar to the electricity used in the two studies (483 kg CO_2-eq/MWh in this study, and 510 CO_2-eq/MWh in the previous study), we postulate that the different environmental intensities can be attributed to the different chemical formulations of products in the two studies. While the previous study considered aqueous-phase

metal salts as the final products, subsequent crystallization or precipitation of metal compounds will substantially increase the environmental intensities. For example, alkali consumables used for metal precipitation contribute 24.3% to the overall CO_2-eq emissions of the Me+Hy pathway. The component contributions in **Fig. S3b** indicate that electricity is a principal contributor (59.8%) to CO_2-eq emissions in the Me+Hy pathway, implying that using electricity with lower CO_2-eq intensity can further decarbonize the prevailing circular refinement pathways.

Supplementary Note 4. Environmental intensities of upstream material extraction

Two different methods for extracting battery-grade cathode materials are compared: mining natural ore and brine, and collecting end-of-life LIBs per the comparison of functionally equivalent process steps depicted in **Fig. 1b** of the main manuscript. The environmental intensities of mining are based on GREET 2021 and are summarized in **Table S1**. Note that transportation of material between mining locations and subsequent refinement steps are often excluded in GREET, and production pathways incorporating transportation in between refining stages were modified by subtracting the associated environmental intensities of the transportation steps (**Table S9**). Relevant values can also be found in **Supplementary File A**.

To estimate the emissions of collecting end-of-life energized LIBs from consumers for transport to a recycling facility for refinement, a transportation logistics model is established for the state of California, U.S. As a model system, California (CA) was chosen because it is the most populated US state (39.6 million, 2022) and has the highest number of electric vehicles, representing 42% of the entire country in 2020[18]. Existing Collection Facilities (CFs) are provided by the California Department of Resources Recycling and Recovery (CalRecycle)[18]. Two different LIB products are considered: end-of-life LCO-based smartphones and end-of-life NCA-based EV batteries. The assumed mass of transported smartphones and EV battery packs along with the mass of active cathode material is summarized in **Table S10**.

In the first scenario, transport of smartphones from the census-block level to the nearest CF is used to estimate environmental intensities of extraction from consumers. Population in every census block is obtained from the U.S. Census Bureau[19]. It is assumed that one smartphone is owned by each person in CA and is recycled every three years. To determine the closest CF, *k*-means clustering is used for the allocation of census blocks to CFs based on minimizing the distance between a census block's centroid and a singular CF, for all census blocks. This process produces clusters of block groups whose LIB-embedded smartphones are allocated to the closest CF. The distance from the census blocks to the CFs is determined by the shortest route using Dijkstra's algorithm. The number of clusters, "*k*," used in this algorithm is fixed and equals the total number of CFs. The relative size of a CF in the inset of **Fig. 4** indicates the number of collected smartphones as compared to nearby CF clusters, and the smartphone collection area is color-coded for distinction for each CF. Multiplying the resulting distances with the average mass of a smartphone yields a mass-distance product (i.e., kg × km), which are converted to

environmental intensities using values for trucking (**Table S12**). Note that the shortest-route value represents a lower bound of emissions.

In the second scenario, the number of registered zero-emission EVs in each CA county in 2012 is obtained from the California Energy Commission[20]. All EV batteries are assumed to be NCA-based and to reach end-of-life in 2019 (the reference year of this study). Because there is more than one CF per county in some situations, the CF that was closest to the centroid of that county was selected to represent the entire county. Additionally, because the distribution of EVs within each county was not known beyond the county-level, it is assumed that LIB collection occurred when an EV was driven to the CF for LIB extraction. The driving of an EV is considered a separate step in the use of an EV and is excluded from the recycling lifetime; therefore, the environmental intensities associated with driving to a CF are assumed zero. However, the removal of EV batteries from a vehicle requires additional resources, and additional investigation is required for quantifying these environmental intensities.

Supplementary Note 5. Environmental intensities of upstream material transport

Mined concentrate transport

A network model is used to determine the routes employed for transporting concentrated mined material from mining to refinement locations. The resulting mass-distance values are converted to environmental intensities using cradle-to-wheel fuel consumption data (**Table S12**). For the critical LIB materials Li, Co, Ni, and Al, the contributions of material transported from mines to refineries are associated with and aggregated based on the countries in which they are extracted or refined, respectively (**Tables S14–S15**). The allocation of mined concentrate from mines to refineries is based on a weighted distribution because data detailing the transported amount and destination of material from every mine to every refinery was not accessible. Mined materials from each extraction country are often concentrated domestically and then transported to countries where they are refined, with allocation weighted by the amount of refinement being performed relative to other refinery countries. The locations used to represent where the mines and refineries are situated are determined based on where most extractive and refinement activity takes place (indicated by latitude and longitude coordinates). The locations are incorporated into the network model as the basis of origins and destinations of the routes employed for transportation logistics.

Specific transit routes are chosen for the network model, including road (i.e., by truck), rail (i.e., by train), and maritime (i.e., by shipping vessel) transport, exhaustive of the major routes available worldwide[21-24]. A portion of the network model and routes considered are presented in **Fig. S5**. The ports associated with maritime transport connect all major sea ports to the closest rail junction. Extraction of Co in the Democratic Republic of the Congo (DRC) is a unique case, where logistics are limited to the road network for the transport of ores or brines from mines to a port[25]. The port in Beira, Mozambique is a major port for which mined Co from the DRC is transported for refining. The maritime network connects the port in Beira, Mozambique to the closest road

junction to bridge the activity in the network model[26]. For a particular element, the shortest route utilizing all combinations between mines and refineries is calculated with Dijkstra's algorithm[27].

The normalized mass-based environmental intensities of LIB material concentrate transported, $\overline{e}^{\mathrm{m}}_{\mathrm{Mat},i}$, are calculated by Eq. S1, where $e^{\mathrm{m}}_{\mathrm{Mat},i}$ is mass-based environmental intensities of the material i with a certain transport model (**Table S9**), d_i is the segmental transport distance in the shortest-route model, n_{d} is the total segments along that route, and w_i is the element weight percent from the country of origin.

$$\overline{e}^{\mathrm{m}}_{\mathrm{Mat},i} = \frac{\sum_{i=1}^{n_{\mathrm{d}}} \left(e^{\mathrm{m}}_{\mathrm{Mat},i} d_i \right)}{w_i} \tag{S1}$$

Environmental intensities with weighted distributions, $E_{\mathrm{Mat},i}$, for each element i are calculated to consider all routes for the three environmental intensities:

$$E_{\mathrm{Mat},i} = \sum_{i=1}^{n_{\mathrm{d}}} \left(\overline{e}^{\mathrm{m}}_{\mathrm{Mat},i} m_i \right) \tag{S2}$$

where m_i denotes the global percentage of an element considered for mined production or refinement production in 2019 (values summarized in **Tables S9–S10**). Environmental intensities per kg of embodied element are in **Table S12**. Lastly, these values of $E_{\mathrm{Mat},i}$ are converted to LCO-eq and NCA-eq bases for comparison in **Fig. 5** in the main manuscript.

Formulations for life cycle impacts of fuel use by different transport mode

Life cycle inventories are developed for road, rail, and maritime modes of transport based on their use of fuel. The fuel used for each mode of transport is a composition of gasoline, diesel, and residual fuel oil, where the exact proportions (ϕ_j) are shown in **Table S13**. Volumetric environmental intensities (e_i) for fuel j were acquired for "cradle-to-tank" and "tank-to-wheel" portions of the life cycle regarding energy consumption, CO_2-eq emissions, and water consumption, as shown in **Table S12.** Efficiency factors (ξ_j) are employed to convert these volumetric properties to a mass-length basis (**Table S13**). All aforementioned values are used to develop life cycle inventories for the environmental intensities of fuel for rail, road, and maritime modes of transport on a per-unit-mass-distance basis:

$$E_{\mathrm{Fuel}} = \frac{\sum_{j=1}^{n} \left(e^{\mathrm{v}}_{\mathrm{F},j} \phi_j \right)}{\xi_j} \tag{S3}$$

These results are summed for each fuel type proportionally for each mode of transport, producing the life cycle inventories for the entire life cycle (**Table S12**).

Collected smartphones and EV battery packs

Environmental intensities of transportation of extracted LCO-based smartphones and NCA-based EV battery packs from CFs to the central recycling facility are calculated as a weighted

distribution based on mass-distance. The mass and distance traveled were converted to energy consumption, CO_2-eq emissions, and water consumption using conversion factors from **Table S12** and later normalized per kg of NCA-eq and LCO-eq precursor by the fractional percentage of active material contained in each transported product. Again, note that disassembly and extraction of battery cells from smartphones or EV battery packs prior to transport to a LIB recycling facility will alter calculated values.

Supplementary Data

Table S1. The gate-to-gate environmental intensities of conventional and circular LIB supply chains including energy consumption, criteria air pollutant emissions, and water consumption, for producing one kg of NCA-eq battery-grade materials ($LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$). The conventional supply chain is based on the GREET 2021 model[10]. Circular refinement includes reductive calcination (RC), mechanical (Me), and hydrometallurgical (Hy) steps, dealing with the recycled scrap and energized batteries. Note that the RC step is only required in recycled battery feedstocks. All circular data are based on power sources from Nevada Power Company (NEVP). The output products considered for conventional refinement are Li_2CO_3, $NiSO_4$, $CoSO_4$, and Al_2O_3; the outputs produced by Redwood Materials are Li_2SO_4, (Ni, Co)SO_4 or as Al_2O_3 and $Al(OH)_3$. Li_2CO_3 is also analyzed as a product for Redwood Materials. Data presented here are visualized in **Fig. 2** in the main manuscript, and **Fig. S3** below (note the comparison of Li_2SO_4 and Li_2CO_3 in Fig. S3a).

Supply chain	Element/Step		Energy (MJ/kg)	Criteria Air Pollutant Emission									Water (L/kg)
				CO_2 (kg/kg)	CH_4 (g/kg)	N_2O (mg/kg)	CO (g/kg)	NO_x (g/kg)	SO_x (g/kg)	PM_{10} (g/kg)	$PM_{2.5}$ (g/kg)	CO_2-eq (kg/kg)	
Conventional		Li	41.61	3.85	5.61	23.23	2.84	3.63	4.52	1.03	0.68	4.00	13.61
		Ni	127.53	8.43	16.75	16.92	11.39	16.92	692.41	6.23	3.48	8.90	43.94
		Co	24.56	1.50	3.45	35.60	3.45	1.75	35.60	5.83	0.70	1.59	16.69
		Al	0.21	0.014	0.034	0.35	0.0089	0.016	0.020	0.012	0.0063	0.015	0.080
Circular	Recycled scrap	Me	1.67	0.22	0.02	2.33	0.04	0.09	0.08	0.01	0.01	0.23	0.53
		Hy	20.31	2.62	1.04	96.48	0.63	0.87	4.06	0.17	0.14	2.68	8.46
		Total	21.98	2.85	1.06	98.81	0.67	0.96	4.14	0.18	0.15	2.91	8.99
	Recycled battery	RC	2.45	0.47	0.07	22.05	0.09	0.13	0.11	0.03	0.02	0.48	1.25
		Me	3.18	0.43	0.03	4.44	0.08	0.18	0.15	0.02	0.02	0.43	1.00
		Hy	38.7	5.52	0.83	50.58	1.23	2.13	8.47	0.35	0.30	5.56	17.79
		Total	40.35	6.42	0.93	77.07	1.40	2.44	8.73	0.40	0.34	6.46	20.04

Table S2. Life cycle inventory for consumables used in circular refinement steps. Data for all consumables are generated using the GREET 2021 model and are normalized by 1 kg of each consumable[26]. Data of natural gas are obtained by averaging U.S. natural gas production from shale and conventional methods[28]. Total CO_2-eq are calculated by summing greenhouse gases multiplied by the corresponding global warming potential (GWP), with conversion factor global warming potentials (GWP) for 100 years: CO_2 (GWP = 1), CH_4 (GWP = 25), and N_2O (GWP = 298)[29].

Consumable	Energy (MJ/kg)	Water (L/kg)	CO_2 (kg/kg)	CH_4 (g/kg)	N_2O (g/kg)	CO (g/kg)	NO_x (g/kg)	SO_x (g/kg)	PM_{10} (g/kg)	$PM_{2.5}$ (g/kg)	CO_2-eq (kg/kg)
H_2SO_4	0.19	0.29	0.011	0.026	0.00023	0.0053	0.0088	20.01	0.0011	0.00067	0.011
HCl	31.62	5.20	1.82	4.66	0.39	0.98	1.57	0.86	0.16	0.11	1.94
HNO_3	12.05	5.06	0.67	2.10	4.77	1.51	1.45	0.21	0.027	0.025	2.15
H_2O_2	17.49	2.10	0.99	2.46	0.016	0.35	0.53	0.30	0.063	0.048	1.06
$Ca(OH)_2$	4.93	6.16	1.26	0.68	0.0012	0.46	0.26	0.14	0.14	0.10	1.28
NaOH	32.00	13.88	1.84	4.81	0.044	1.09	1.78	0.95	0.18	0.12	1.98
Natural gas	50.47	0.51	0.20	6.42	0.0014	0.51	0.57	0.48	0.017	0.015	0.36

Table S3. Relative contribution of different inputs and consumables in the circular refinement processes (for scrap and energized battery feedstocks) to the environmental intensities, including energy consumption, CO_2-eq emissions, and water consumption. The data are based on the GREET 2021 model[10] and are visualized in **Fig. 3a** in the main manuscript. "N/A" denotes "not applicable."

Environmental impact	Recycling pathway	Reductive Calcination				Mechanical		Hydrometallurgy			
		Electricity input (%)	Water input (%)	Alkali (%)	CO_2 emission (%)	Primary electricity (%)	Water input (%)	Primary electricity (%)	H_2SO_4 (%)	H_2O_2 (%)	Alkali (%)
Energy consumption	Recycled scrap	N/A	N/A	N/A	N/A	7.60	N/A	62.65	1.42	20.14	8.19
	Recycled battery	4.70	N/A	0.82	N/A	7.16	N/A	79.09	1.44	N/A	6.79
CO_2-eq emissions	Recycled scrap	N/A	N/A	N/A	N/A	1.83	N/A	15.05	2.00	31.13	49.99
	Recycled battery	4.36	N/A	1.56	1.47	6.64	N/A	73.25	0.63	N/A	12.10
Water consumption	Recycled scrap	N/A	N/A	N/A	N/A	9.87	0.53	81.38	1.27	2.18	4.77
	Recycled battery	3.29	0.67	2.26	N/A	5.01	8.27	55.33	6.13	N/A	19.03

Table S4. Environmental intensities of circular refinement steps using different electricity sources for producing one kg of NCA-eq battery-grade materials ($LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$). Circular refinement includes reductive calcination (RC), mechanical (Me), and hydrometallurgical (Hy) steps, dealing with the production scrap and energized batteries. Note that the RC step is only required for energized batteries. Power sources are from Nevada renewable energy tariff (NV*), Bonneville Power Administration (BPAT), California Independent System Operator (CISO), and Western Area Power Administration–Colorado-Missouri (WACM)[12,13]. Data presented here are visualized in **Fig. 3b** in the main manuscript.

Power sources	Recycling pathways	Step	Energy (MJ/kg)	Criteria Air Pollutant Emission									Water (L/kg)
				CO_2 (kg/kg)	CH_4 (g/kg)	N_2O (mg/kg)	CO (g/kg)	NO_x (g/kg)	SO_x (g/kg)	PM_{10} (g/kg)	$PM_{2.5}$ (g/kg)	CO_2-eq (kg/kg)	
NV*	Recycled scrap	Me	1.67	0.02	0	0	0	0	0	0	0	0.02	4.64
		Hy	20.31	0.91	0.91	77.24	0.27	0.11	3.40	0.07	0.05	0.96	42.35
		Total	21.98	0.93	0.91	77.24	0.27	0.11	0	0.07	0.05	0.98	46.99
	Recycled Battery	RC	2.45	0.21	0.05	19.13	0.03	0.02	0.01	0.01	0.008	0.22	6.38
		Me	3.18	0.03	0	0	0	0	0	0	0	0.03	8.83
		Hy	38.73	1.17	0.50	1.55	0.30	0.19	6.80	0.09	0.07	1.18	104.14
		Total	44.36	1.41	0.55	20.69	0.33	0.21	6.81	0.10	0.078	1.43	119.35
BPAT	Recycled scrap	Me	4.30	0.06	0.005	0.70	0.01	0.03	0.03	0.004	0.003	0.06	3.22
		Hy	1.23	1.23	0.95	83.03	0.36	0.33	3.62	0.09	0.07	1.27	30.65
		Total	1.28	1.29	0.95	83.73	0.37	0.36	3.65	0.09	0.08	1.33	33.87
	Recycled scrap	RC	2.45	0.26	0.06	20.01	0.05	0.05	0.05	0.01	0.01	0.26	4.61
		Me	3.18	0.10	0.009	1.34	0.02	0.05	0.05	0.006	0.005	0.10	6.12
		Hy	38.73	1.97	0.60	16.32	0.54	0.75	7.37	0.15	0.12	1.99	74.32
		Total	44.36	2.33	0.66	37.67	0.61	0.85	7.47	0.17	0.14	2.36	85.06
CISO	Recycled scrap	Me	1.67	0.11	0.007	1.44	0.03	0.05	0.03	0.006	0.006	0.11	1.27
		Hy	20.31	1.69	0.96	89.07	0.52	0.50	3.62	0.12	0.10	1.74	14.59
		Total	21.98	1.80	0.97	90.51	0.55	0.55	3.65	0.13	0.11	1.85	15.86

	Recycled scrap	RC	2.45	0.33	0.06	20.93	0.07	0.08	0.04	0.02	0.02	0.33	2.18
		Me	3.18	0.21	0.01	2.73	0.06	0.09	0.05	0.01	0.01	0.21	2.42
		Hy	38.73	3.14	0.64	31.70	0.96	1.19	7.36	0.22	0.19	3.17	33.40
		Total	44.36	3.68	0.71	55.35	1.09	1.36	7.45	0.25	0.21	3.76	37.99
WACM	Recycled scrap	Me	1.67	0.31	0.05	6.90	0.09	0.21	0.28	0.01	0.02	0.31	1.42
		Hy	20.31	3.34	1.30	134.16	1.01	1.88	5.72	0.17	0.20	3.41	15.81
		Total	21.98	3.65	1.35	141.06	1.10	2.09	6.00	0.18	0.22	3.72	17.23
	Recycled scrap	RC	2.45	0.58	0.11	27.76	0.15	0.29	0.36	0.03	0.03	0.59	2.36
		Me	3.18	0.59	0.09	13.14	0.17	0.41	0.54	0.02	0.04	0.60	2.70
		Hy	38.73	7.34	1.49	146.60	2.20	4.70	12.71	0.35	0.45	7.42	36.51
		Total	44.36	8.51	1.69	187.50	2.52	5.39	13.60	0.40	0.52	8.61	41.57

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Table S5. Life cycle inventory of embodied emission of air pollutants and water consumption of different electricity sources. Total CO_2-eq values are calculated by summing greenhouse gases multiplied by the corresponding 100-year global warming potential (GWP), including CO_2 (GWP = 1), CH_4 (GWP = 25), and N_2O (GWP = 298)[29]. Electricity sources are balancing grids from Bonneville Power Administration (BPAT), California Independent System Operator (CISO), Nevada Power Company (NEVP), Western Area Power Administration–Colorado-Missouri (WACM), and a Nevada renewable energy tariff (NV*) comprised of 85% geothermal, 10% solar, and 5% hydropower. Alternative power generated by hydraulic, nuclear, solar, natural gas (Ng), wind, coal, oil, biomass, geothermal, and others are summarized in the lower sections. Data listed are visualized in Fig. S5. "N/A" denotes "not applicable" [12,13].

	Electricity source	Criteria Air Pollutant Emissions									Water (L/kWh)
		CO_2 (kg/MWh)	CH_4 (kg/MWh)	N_2O (kg/MWh)	CO (kg/MWh)	NO_x (kg/MWh)	SO_x (kg/MWh)	PM_{10} (kg/MWh)	$PM_{2.5}$ (kg/MWh)	CO_2-eq (g/MWh)	
Balancing Areas	BPAT	118.57	0.010	0.0015	0.024	0.057	0.059	0.0066	0.0057	119.28	6.94
	CISO	239.30	0.014	0.0031	0.067	0.10	0.058	0.014	0.013	240.58	2.74
	NEVP	483.48	0.034	0.0050	0.095	0.20	0.17	0.027	0.024	485.83	1.14
	WACM	670.34	0.10	0.015	0.20	0.46	0.61	0.050	0.040	677.33	3.06
	NV*	36.56	N/A	N/A	N/A	N/A	N/A	N/A	N/A	36.56	10.00
Electricity generation sources	Hydro	20.50	N/A	N/A	N/A	N/A	N/A	N/A	N/A	20.50	9.53
	Nuclear	13.00	N/A	N/A	N/A	N/A	N/A	N/A	N/A	13.00	2.18
	Solar	43.40	N/A	N/A	N/A	N/A	N/A	N/A	N/A	43.40	0.31
	Nat. gas	489.28	0.010	0.0016	0.067	0.11	0.0071	0.021	0.021	490	0.26
	Wind	13.00	N/A	N/A	N/A	N/A	N/A	N/A	N/A	13.00	0.01
	Coal	990.20	0.16	0.023	0.30	0.71	0.94	0.076	0.060	1001	1.55
	Oil	837.23	0.013	0.0082	0.51	3.70	2.29	0.24	0.217	840	0.33
	Biomass	31.30	0.11	0.060	1.18	0.68	0.049	0.073	0.069	52	5.50
	Geotherm	36.70	N/A	N/A	N/A	N/A	N/A	N/A	N/A	36.70	11.17
	Others	58.96	0.00087	0.00013	0.0057	0.010	0.00060	0.002	0.0018	59	7.34

Table S6. The environmental intensities of the material extraction step for producing one kg of NCA-eq ($LiNi_{0.80}Co_{0.15}Al_{0.05}O_2$) mined natural material in the conventional supply chain. Impacts include energy consumption, criteria air pollutant emissions, and water consumption. The conventional extraction step is based on the GREET 2021 model[10]. Data presented are visualized in **Fig. 5** in the main manuscript.

Element	Energy (MJ/kg)	Criteria Air Pollutant Emissions									Water (L/kg)
		CO_2 (kg/kg)	CH_4 (g/kg)	N_2O (mg/kg)	CO (g/kg)	NO_x (g/kg)	SO_x (g/kg)	PM_{10} (g/kg)	$PM_{2.5}$ (g/kg)	CO_2-eq (kg/kg)	
Li	8.25	0.58	0.74	7.56	0.33	1.22	0.047	0.057	0.048	0.60	5.35
Ni	26.06	1.95	2.59	97.78	1.31	6.53	3.93	0.52	0.40	2.00	7.77
Co	2.76	0.20	0.25	4.91	0.21	4.86	0.011	5.70	0.61	0.21	0.87
Al	2.0×10^{-3}	1.5×10^{-4}	1.8×10^{-4}	3.4×10^{-3}	1.1×10^{-4}	4.3×10^{-4}	8.1×10^{-5}	2.4×10^{-3}	1.2×10^{-3}	1.6×10^{-4}	0.016

Table S7. The environmental intensities of material extraction and refinement steps in conventional supply chains, including energy consumption, criteria air pollutant emissions, and water consumption. The values are normalized by one kg of each element regardless of material type (e.g., ore type or elemental concentration). The conventional material extraction and refinement steps are based on the GREET 2021 model[10]. Note that values of lithium are weight averages of 45% brine- and 55% ore-based lithium production[30]. Refinement materials reference Li_2CO_3, $NiSO_4$, $CoSO_4$, Al_2O_3, and $MnSO_4$.

Step	Element	Energy (MJ/kg)	Criteria Air Pollutant Emissions									Water (L/kg)
			CO_2 (kg/kg)	CH_4 (g/kg)	N_2O (mg/kg)	CO (g/kg)	NO_x (g/kg)	SO_x (g/kg)	PM_{10} (g/kg)	$PM_{2.5}$ (g/kg)	CO_2-eq (kg/kg)	
Extraction	Li	114.78	8.07	10.24	105.16	4.53	17.02	0.66	0.79	0.66	8.40	9.41
	Ni	53.30	3.98	5.31	200.00	2.68	13.36	8.05	1.06	0.81	4.20	15.89
	Co	29.97	2.18	2.68	53.39	2.31	5.28	0.12	61.89	6.66	2.30	9.41
	Al	0.14	0.01	0.01	0.24	0.01	0.03	0.01	0.17	0.09	0.01	1.12
	Mn	23.11	1.34	2.76	23.45	0.86	1.11	0.73	5.29	2.65	1.40	6.73
	Cu	4.06	0.25	0.45	3.32	0.26	0.45	0.10	0.08	0.04	0.26	0.95
Refinement	Li	579.22	53.60	78.09	323.40	39.56	50.48	62.97	14.27	9.48	55.60	189.37
	Ni	260.87	17.24	34.26	350.60	23.30	34.62	1416.30	12.75	7.12	18.20	89.88
	Co	266.79	16.26	37.44	386.77	11.22	19.04	64.16	63.33	7.60	17.30	213.94
	Al	15.18	0.98	2.42	24.63	0.63	1.15	1.41	0.89	0.45	1.00	5.66
	Mn	1.08	0.86	0.11	1.78	7.3E-02	0.57	3.84	4.1×10^{-2}	3.6×10^{-2}	0.86	0.57
	Cu	32.25	1.99	4.68	42.26	1.78	2.23	140.28	0.21	0.14	2.10	5.26

Table S8. Separated cradle-to-gate conventional and circular LIB supply steps showing energy consumption, greenhouse gas emissions, and water consumption for two different functional units: NCA-eq and LCO-eq cathode material salts. Conventional supply chain values reflect dominant global supply chains (excluding recycled feedstocks) extracted from GREET, with transport values modeled in this work. The circular supply chain represents recycling of NCA-based battery packs and LCO-based smartphones in California as described elsewhere in the study.

Supply chain	Step	Energy Consumption				CO_2-eq Emission				Water Consumption			
		NCA		LCO		NCA		LCO		NCA		LCO	
		Value (MJ/kg NCA-eq)	Percentage (%)	Value (kg/kg LCO-eq)	Percentage (%)	Value (kg/kg NCA-eq)	Percentage (%)	Value (kg/kg LCO-eq)	Percentage (%)	Value (L/kg NCA-eq)	Percentage (%)	Value (kg/kg LCO-eq)	Percentage (%)
Conventional	Extraction	37.1	14	26.2	10	2.85	14	1.96	9	14.0	15	10.9	7
	Transport	31.2	12	44.9	16	3.68	17	4.32	21	0.757	1	1.11	1
	Refinement	194	74	202	74	14.5	69	14.4	70	77.3	84	142	92
	Total	262	100	273	100	21.0	100	20.6	100	92.1	100	154	100
Circular	Extraction	0	0	0.385	0	0	0	0.0189	0.2	0	0	0.00920	0
	Transport	1.49	3.5	9.58	8	0.0729	2.2	0.470	5	0.036	0	0.229	0
	Refinement	44.4	97	112	92	3.72	98	9.40	95	38.0	99.9	96.1	99.8
	Total	45.8	100	122	100	3.79	100	9.89	100	38.0	100	96.3	100

Table S9. The estimated environmental intensities of transporting Li, Co, Ni, and Al concentrates of mined material in conventional supply chains normalized by the content of metal in transit (top table). The transportation impacts of aggregated LCO-based smartphones and NCA-based EV batteries are found in the bottom table from collection facilities to a recycling center positioned at the gravity point (i.e., center) of the California population. Data presented are visualized in **Fig. 5** in the main manuscript.

Conventional Material Transport

Material Transported	Energy Consumption (MJ/t metal)	CO_2-eq Emissions (kg/t metal)	Water Consumption (L H_2O/t metal)
Li concentrate	37,808	6,152	964
Co concentrate	70,149	6,445	1,727
Ni concentrate	44,755	5,355	1,073
Al concentrate	10,770	1,809	278

Circular Material Transport

Product Transported	Energy Consumption (MJ/t product)	CO_2-eq Emissions (kg/t product)	Water Consumption (L/t product)
Smartphone Collection*	20.5	1.00	0.490
Smartphone Transport	510	25.0	12.2
EV Battery Transport	515	25.3	12.3

*Smartphone collection is a material extraction step but included here for reference. Collection is modeled by the transportation resources required for battery collection from consumer census block and transport to a collection facility; it does not include other potential steps such as device disassembly for battery extraction from products.

Table S10. Mass of lithium-ion battery embedded products considered in this analysis[10,11,31].

Battery Type	Element	Assumed Mass (kg)
LCO	LCO-battery embedded smartphone	0.118
	Mass of battery per smartphone	0.026
	LCO active material per kg of battery	0.162
NCA	End-of-life 27-kWh EV NCA battery pack	108
	NCA active material per NCA battery pack	37.4

Table S11. Total metal extraction mass of ores or brines from global mining activity for Li, Co, Ni, and Al in 2019[32].

Element	Total Mined & Refined (t)
Li	86,000
Co	144,000
Ni	2,400,000
Al	189,000,000

Table S12. Environmental intensities of fuels use by different modes of transport in the stages of "cradle-to-wheel", "cradle-to-tank", and "tank-to-wheel". Data are from references[10,33,34].

Transport Stage	Mode of Transport	Maritime	Rail	Road	Gasoline	Diesel	Residual Fuel Oil
Cradle-to-wheel	Energy Consumption (MJ/t km)	0.15	0.213	2.3	N/A	N/A	N/A
	CO_2-eq emissions (g/t km)	27.28	33.69	112.8	N/A	N/A	N/A
	Water Consumption (L/t km)	0.00396	0.00319	0.055	N/A	N/A	N/A
Cradle-to-tank	Energy Consumption (MJ/t km)	0.01	0.024	0.3	7.11	4.54	2.80
	CO_2-eq emissions (g/t km)	1.00	1.57	17.9	503	299	193
	Water Consumption (L/t km)	0.00396	0.00319	0.055	4.36	0.608	0.396
Tank-to-wheel	Energy Consumption (MJ/t km)	0.14	0.189	2.0	33.8	36.0	39.7
	CO_2-eq emissions (g/t km)	26.3	32.1	94.9	N/A	N/A	N/A
	Water Consumption (L/t km)	N/A	N/A	N/A	N/A	N/A	N/A

Table S13. Proportions of different fuel use, ϕ, and fuel efficiency ξ, by different transport modes. ξ are obtained from references[28,35-37]. Data presented are employed in eq. S3 for the visualization in **Fig. 5** in the main manuscript, and are quantified in **Table S8**.

Transport Mode	Fuel	Proportion of Fuel Use, ϕ (%)	Fuel Efficiency, ξ (t km/L)
Maritime	Gasoline	15.3	270.8
	Diesel	29.7	
	Residual Fuel Oil	55.1	
Rail	Gasoline	0	181.6
	Diesel	0	
	Residual Fuel Oil	100	
Road	Gasoline	9.9	18.7
	Diesel	89.4	
	Residual Fuel Oil	0	

Table S14. Mine/mine cluster country with latitude and longitude used in analysis, mine capacity, and element weight percentage, for Li[38], Co[39], Ni[40], and Al[41]. Standard quantity is as reported and scaled quantity includes locations considered in the study scaled to 100%. References for all mine locations and assumed weight percentages of mined concentrates are obtained from the public repository[42].

Element Type	Mine Location	Latitude-Longitude Coordinates for Analysis	Mine Capacity (% of total / scaled to 100%)	Mine Capacity (standard quantity / scaled by total)	Element Content (wt. % of element in transported concentrate)
Li	Australia	(−33.85, 116.06)	52.3% / 52.9%	45,000 / 45,529	3.7
	Chile	(−26.23, −69.12)	22.4% / 22.7%	19,300 / 19,527	18.8
	China	(36.62, 101.78)	12.6% / 12.7%	10,800 / 10,927	18.8
	Argentina	(−23.79, −66.76)	7.3% / 7.4%	6,300 / 6,374	18.5
	Brazil	(−20.06, −44.57)	2.8% / 2.8%	2,400 / 2,428	3.7
	Zimbabwe	(−17.37, 31.43)	1.4% / 1.4%	1,200 / 1,214	2.2
Co	Democratic Republic of Congo	(−8.75, 26.41)	69.4% / 80.4%	100,000 / 115,811	9.5 (5.0–14.0)
	Russia	(69.35, 88.21)	4.4% / 5.1%	6,300 / 7,296	12.5
	Australia	(−28.77, 121.88)	4.0% / 4.6%	5,740 / 6,648	3.0
	Philippines	(9.76, 125.51)	3.5% / 4.1%	5,100 / 5,906	4.9
	Cuba	(20.66, −74.95)	2.6% / 3.1%	3,800 / 4,401	5.0
	Madagascar	(−18.95, 48.30)	2.4% / 2.7%	3,400 / 3,938	4.2
Ni	Indonesia	(−1.43, 121.45)	25.3% / 35.1%	606,000 / 843,179	8.45
	Philippines	(9.48, 125.80)	14.4% / 20.0%	344,915 / 479,909	5.11
	Russia	(58.45, 92.19)	11.3% / 15.8%	272,000 / 378,457	9.45
	New Caledonia	(−22.37, 166.87)	9.0% / 12.5%	216,225 / 378,457	7.89
	Canada	(46.83, −71.25)	7.3% / 10.2%	175,761 / 244,551	15.7
	China	(30.66, 104.07)	4.6% / 6.4%	110,000 / 153,052	9.48
Al	Australia	(−31.95, 115.86)	29.3% / 34.1%	105,000 / 122,117	15.0
	China	(23.64, 108.27)	19.6% / 22.7%	70,000 / 81,411	24.3
	Guinea	(10.37, −13.58)	18.7% / 21.8%	67,000 / 77,922	23.8
	Brazil	(−1.46, −48.50)	9.5% / 11.0%	34,000 / 39,543	21.2
	Indonesia	(3.95, 108.14)	4.7% / 5.5%	17,000 / 19,771	21.2

Jamaica	(18.04, −77.51)	2.5% / 2.9%	9,020 / 10,490	23.8
Kazakhstan	(52.27, 77.00)	1.6% / 1.9%	5,800 / 6,746	23.0

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Table S15. Refinery location (latitude and longitude) and refining capacity for Li, Co, Ni, and Al. Standard quantity is as reported by USGS and others; scaled quantity includes locations considered in the study scaled to 100%. References for all refinery locations are itemized and obtained from a public repository[42].

Element	Refining Location	Latitude-Longitude Coordinates for Analysis	Refining Capacity (% of total / scaled to 100%)	Refining Capacity (standard quantity / scaled by total)
Li	China	(28.68, 115.88)	61.0% / 61.0%	52,306 / 52,306
	Australia	(−31.95, −115.86)	18.0% / 18.0%	15,403 / 15,403
	United States	(35.18, −81.34)	11.0% / 11.0%	9,627 / 9,627
	Chile	(−23.65, −70.40)	10.0% / 10.0%	8,664 / 8,664
Co	China	(24.72, 114.95)	67.0% / 75.3%	96,480 / 108,404
	Finland	(63.84, 23.13)	10.0% / 11.2%	14,400 / 16,180
	Canada	(50.00, −85.00)	5.0% / 5.6%	7,200 / 8,090
	Norway	(58.15, 8.00)	4.0% / 4.5%	5,760 / 6,472
	Japan	(33.96, 133.31)	3.0% / 3.4%	4,320 / 4,854
Ni	China	(27.99, 120.70	23.3% / 23.3%	244,900 / 244,900
	Canada	(50.00, −85.00)	15.5% / 15.5%	163,200 / 163,200
	Russia	(69.35, 88.20)	15.0% / 15.0%	157,396 / 157,396
	Japan	(33.96, 133.31)	11.6% / 11.6%	121,750 / 121,750
	Australia	(−27.28, 120.55)	9.9% / 9.9%	103,900 / 103,900
	Norway	(58.15, 8.00)	8.2% / 8.2%	86,500 / 86,500
	Finland	(61.31, 22.14)	5.7% / 5.7%	59,700 / 59,700
	South Africa	(−25.65, 27.26)	4.6% / 4.6%	48,100 / 48,100
	Madagascar	(−18.86, 48.30)	3.4% / 3.4%	35,474 / 35,474
	New Caledonia	(−22.28, 167.02)	2.9% / 2.9%	30,875 / 30,875
Al	China	(36.07, 119.16)	54.5% / 63.1%	195,150 / 225,971
	Australia	(−32.82, 151.71)	15.2% / 17.6%	54,373 / 62,960
	Brazil	(−2.53, −4.30)	6.5% / 7.6%	23,418 / 27,116
	India	(21.85, 84.03)	6.5% / 7.6%	18,008 / 20,852
	Russia	(61.67, 50.82)	5.0% / 5.8%	7,429 / 8,602
	Jamaica	(17.96, −77.60)	2.1% / 1.9%	5,841 / 6,764

	Saudi Arabia	(27.49, 49.14)	1.4% / 1.6%	4,953 / 5,735

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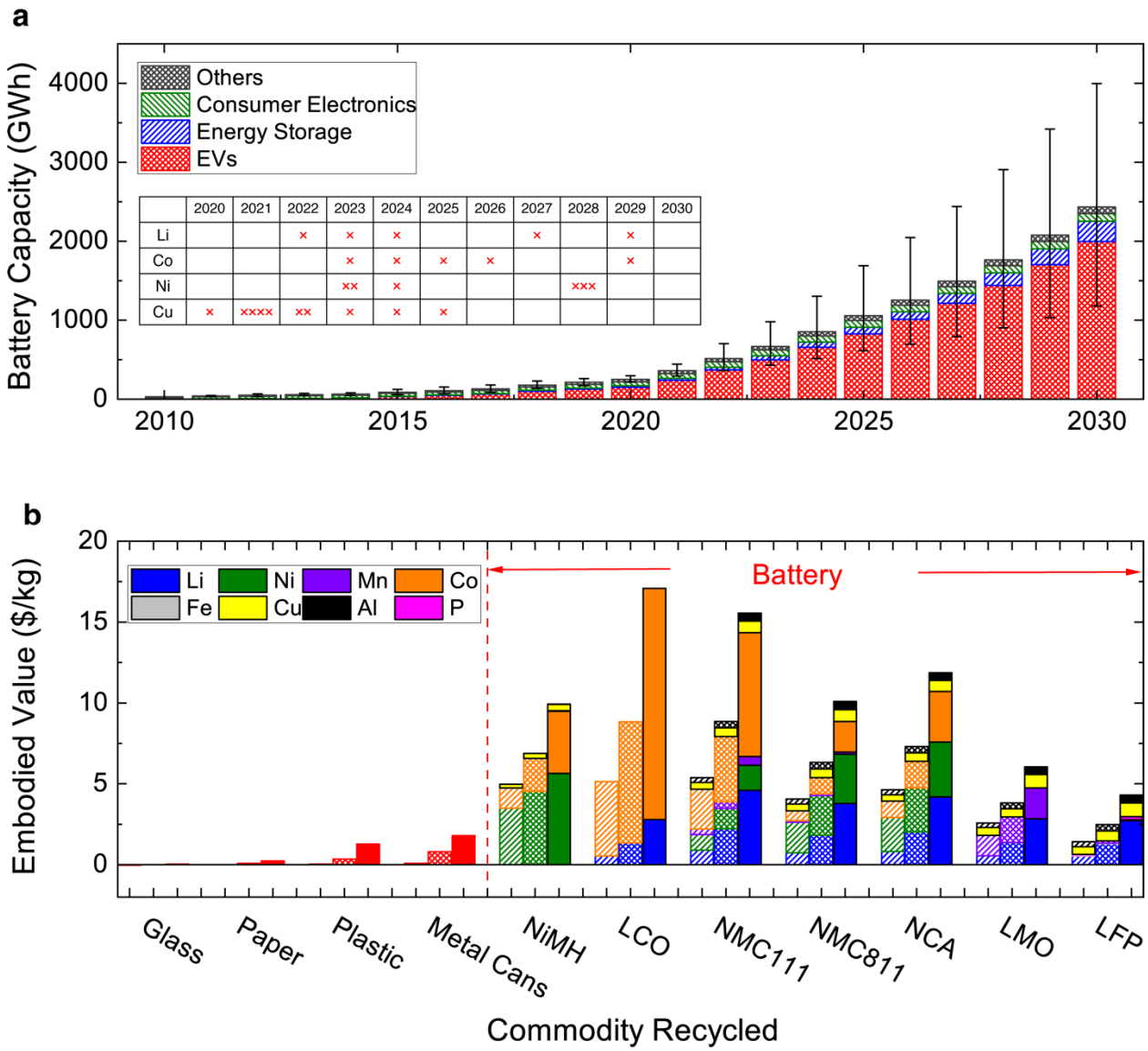

Fig. S1 | a, Projected global demand of lithium-ion batteries in representative industries including electric vehicles (EVs), energy storage, consumers electronics, and others. The uncertainty bars indicate the range of projections by different reports, highlighting varying optimism in supply and production capacity, along with methods of quantification, where hybrid or plug-in EVs may not be included in some analyses. Inset presents a compilation of industry report projections of when global supply-demand gaps will become negative and will not recover to positive values, running a supply deficit. Each "×" denotes a literature estimate[43-46]. **b,** composition breakdown of the embodied value of each commodity. Glass, paper, plastic, and metal cans are an average of three to five major sub-categories. The center bar for each commodity is the average value adjusted for inflation between January 2018 and December 2021 and in December 2021 USD[1-7,47]. The side bars represent the 90% confidence interval of those commodity values. Notably, all batteries chemistries represent 27-kWh battery packs with breakdowns provided by GREET 2021. LCO is

included because it is a common battery in consumer electronics, although it is not a common EV battery material[48,49].

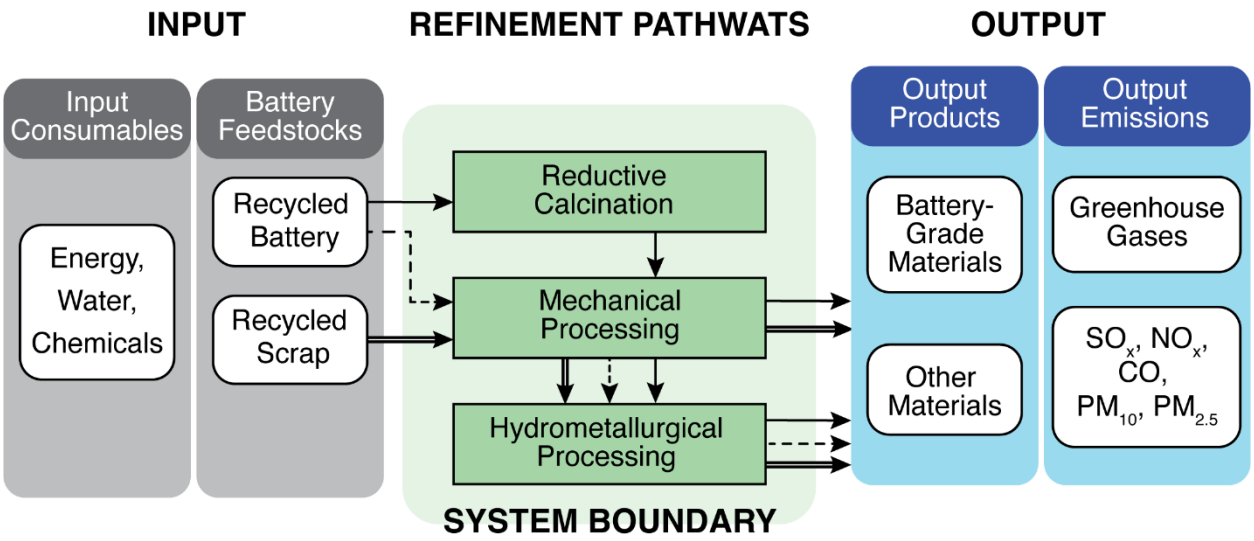

Fig. S2 | Gate-to-gate refinement pathways at Redwood Materials for processing two lithium-ion battery (LIB) feedstocks: LIB production scrap (recycled scrap), and LIBs collected from consumers (recycled battery). Consumables including energy, water, and chemicals are input to the refinement pathways. Three refinement processes are employed in Redwood Materials including reductive calcination (RC), mechanical processing (Me), and hydrometallurgical processing/refinement (Hy). Recycled scrap is refined by a multi-step pathway employing Me and Hy (denoted by double-line arrows), while recycled battery are refined by an RC→Me→Hy multi-step pathway (denoted by solid single arrows) and a Hy-only pathway (denoted by dashed arrows). Output products are battery-grade materials and other materials (e.g., graphite), and output emissions include greenhouse gases and SO_x, NO_x, CO, PM_{10}, and $PM_{2.5}$. Note that, in the system boundary, only direct processes involved in refining pathways are analyzed and no other site-wide operations (e.g., running office computers, lights) are considered.

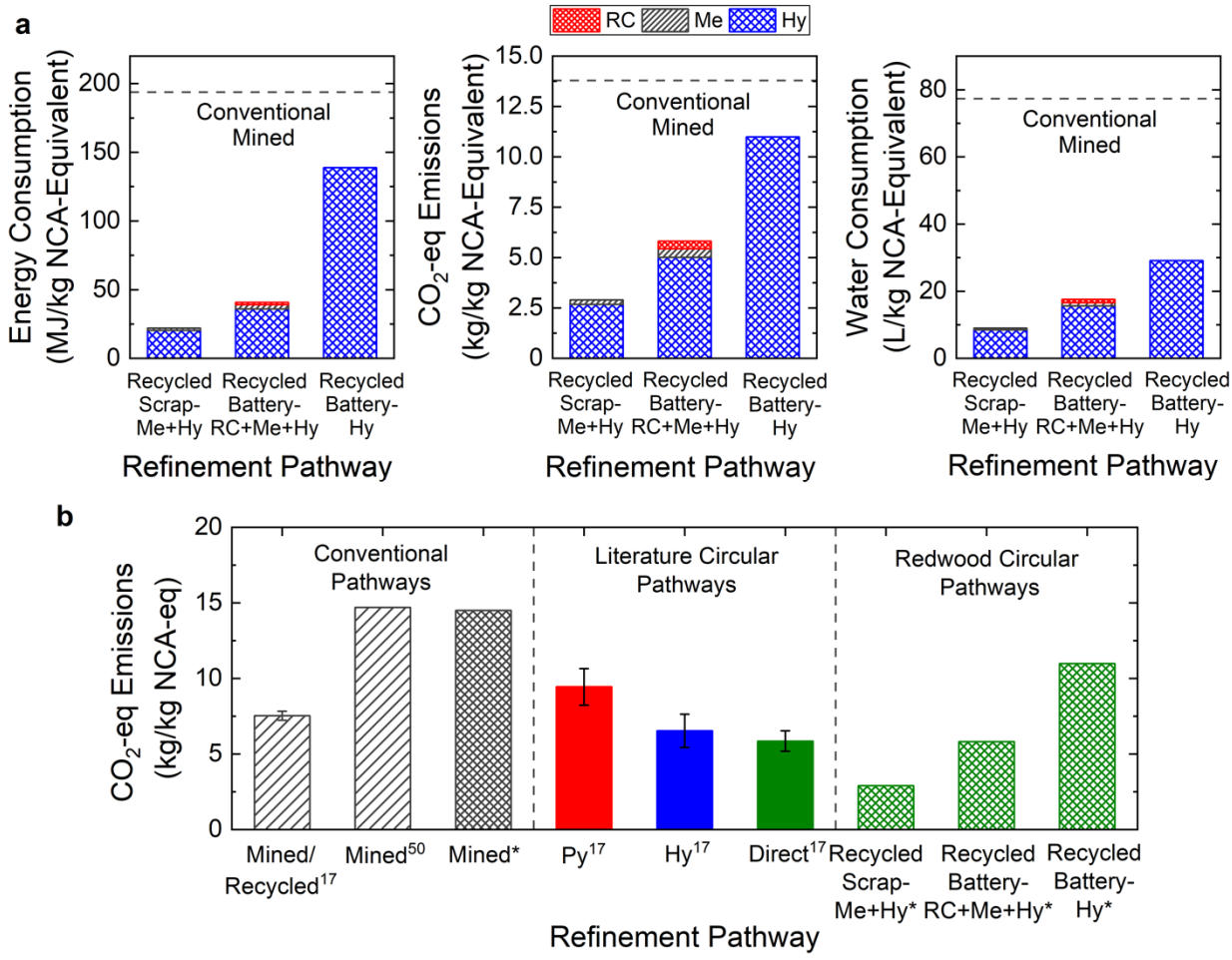

Fig. S3 | a, Comparisons of the environmental metrics (i.e., energy consumption, CO_2-eq emission, and water consumption) of different refinement lithium products, Li_2SO_4 and Li_2CO_3, using Redwood (RC+Me+Hy) pathway and hydrometallurgy (Me+Hy) pathway. Both pathways refine energized batteries (recycled battery). Horizontal dash lines denote conventional mined pathways. **b,** Relative contributions of input energy, water, and consumables to the environmental metrics refining energized batteries by the hydrometallurgy (Me+Hy) pathway. **c,** Comparison of environmental impacts between refinement pathways in this study and data in literature[17,50]. In conventional pathways, data reported in literature are based on a combined sources including mined and recycled metals, and purely mined sources. Circular pathways in literature all employ a single refinement technology, including pyrometallurgical (Py), hydrometallurgical (Hy), and direct recycling (direct). Data in this study and literature are denoted by "*" and superscripts of reference numbers, respectively. Data from the literature is normalized by the same functional unit in this study, and uncertainties are determined by combining two different battery form factors: pouch and cylindrical (detailed in **Table S14–S15**).

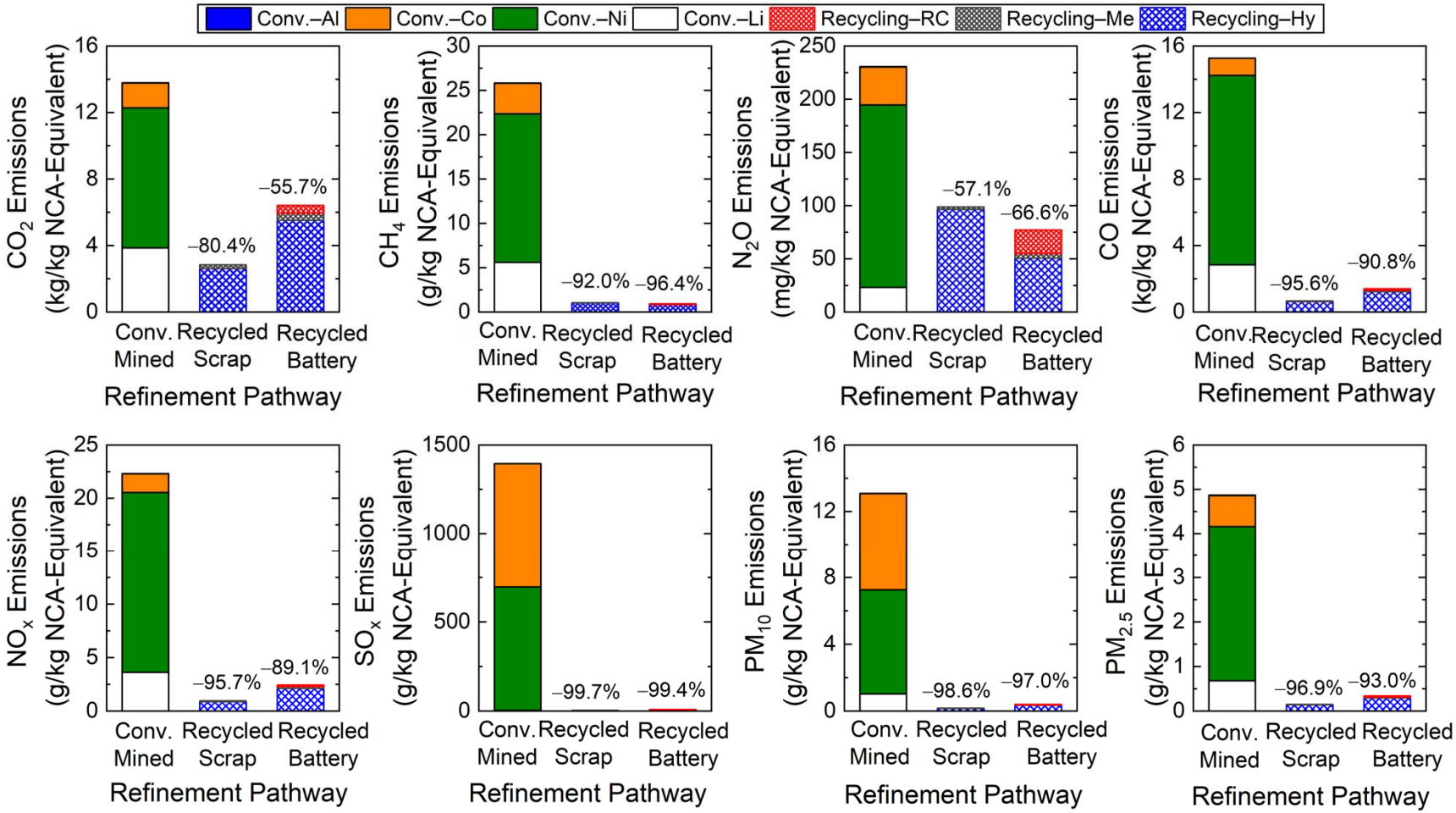

Fig. S4 | Emissions of air pollutants, including CO_2, CH_4, N_2O, CO, NO_x, SO_x, PM_{10}, and $PM_{2.5}$, by the mined conventional refinement and recycling processes from the scrap and energized batteries using Nevada energy (NEVP). Note that CO_2-eq emission presented in Fig. 2 is calculated by summing the greenhouse gases multiplied by the corresponding 100-year global warming potential (GWP), including CO_2 (GWP = 1), CH_4 (GWP = 25), and N_2O (GWP = 298)[17].

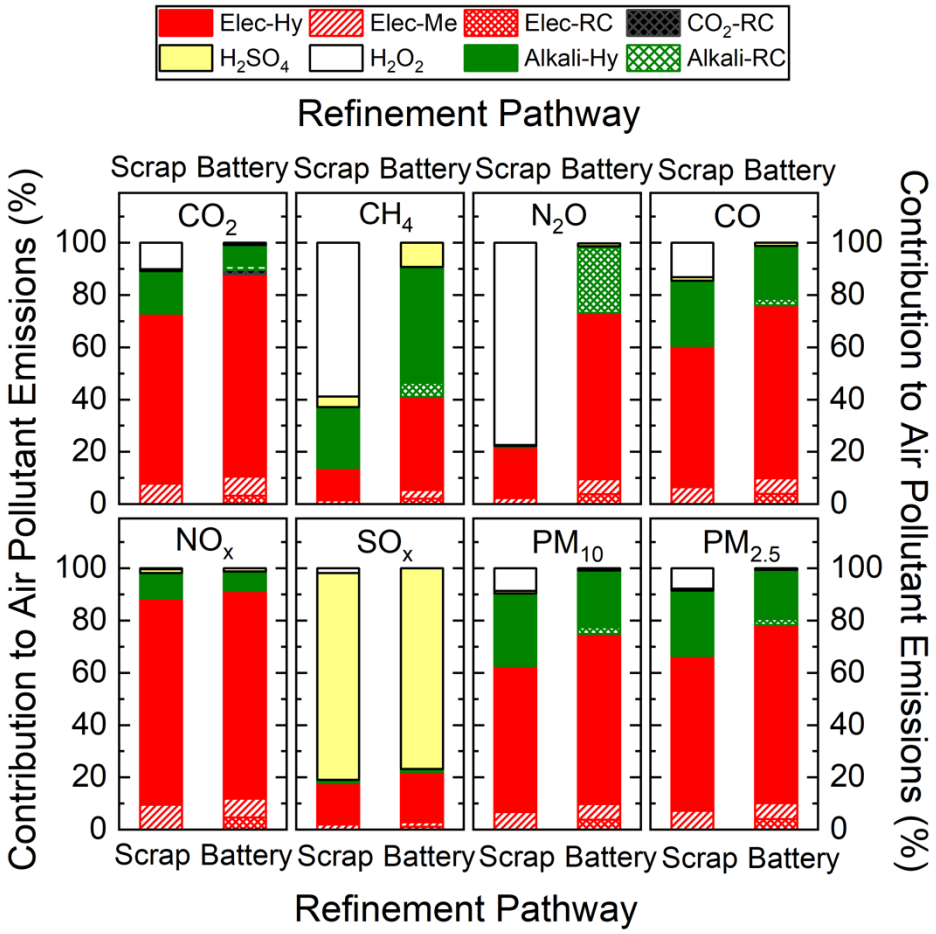

Fig. S5 | Relative contributions to air pollutant emissions, including CO_2, CH_4, N_2O, CO, NO_x, SO_x, PM_{10}, and $PM_{2.5}$, by lithium-ion battery circular refinement processes from production scraps and energized batteries based on Nevada electricity (NEVP). Note that CO_2-eq emissions presented in Fig. 2 are calculated by summing the greenhouse gases multiplied by the corresponding 100-year global warming potential (GWP), including CO_2 (GWP = 1), CH_4 (GWP = 25), and N_2O (GWP = 298)[17].

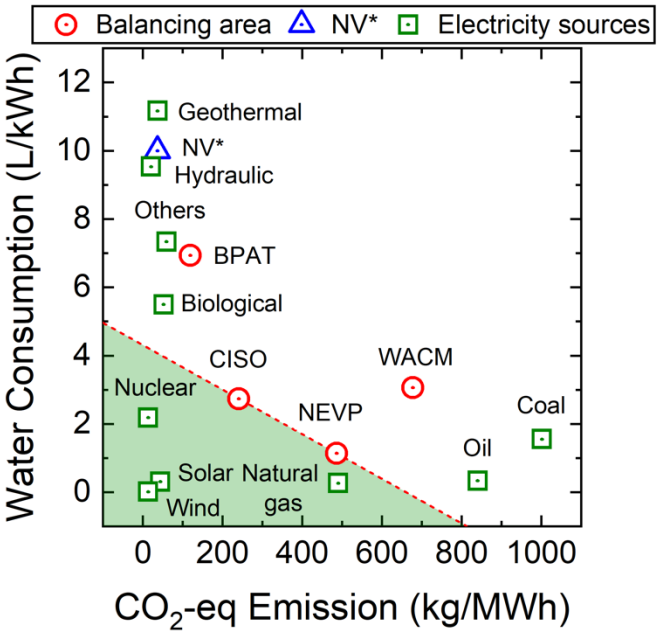

Fig. S6 | Tradeoff relationship between embodied water consumption and CO_2-eq emission by different power sources, including electricity grids in different locations (⊙), purely power sources (⊡), and Nevada renewable energy tariff (NV*, △). The red dashed line denotes the lower bound of the water-CO_2 performance, i.e., the existing electricity grids that have the lowest water consumption and CO_2-eq emission simultaneously, and the green shaded area covers the power sources that can transcend the current limit of water-CO_2 performance. Electricity grids, including Bonneville Power Administration (BPAT), California Independent System Operator (CISO), Nevada Power Company (NEVP), and Western Area Power Administration–Colorado-Missouri (WACM), are combinations of different power sources.

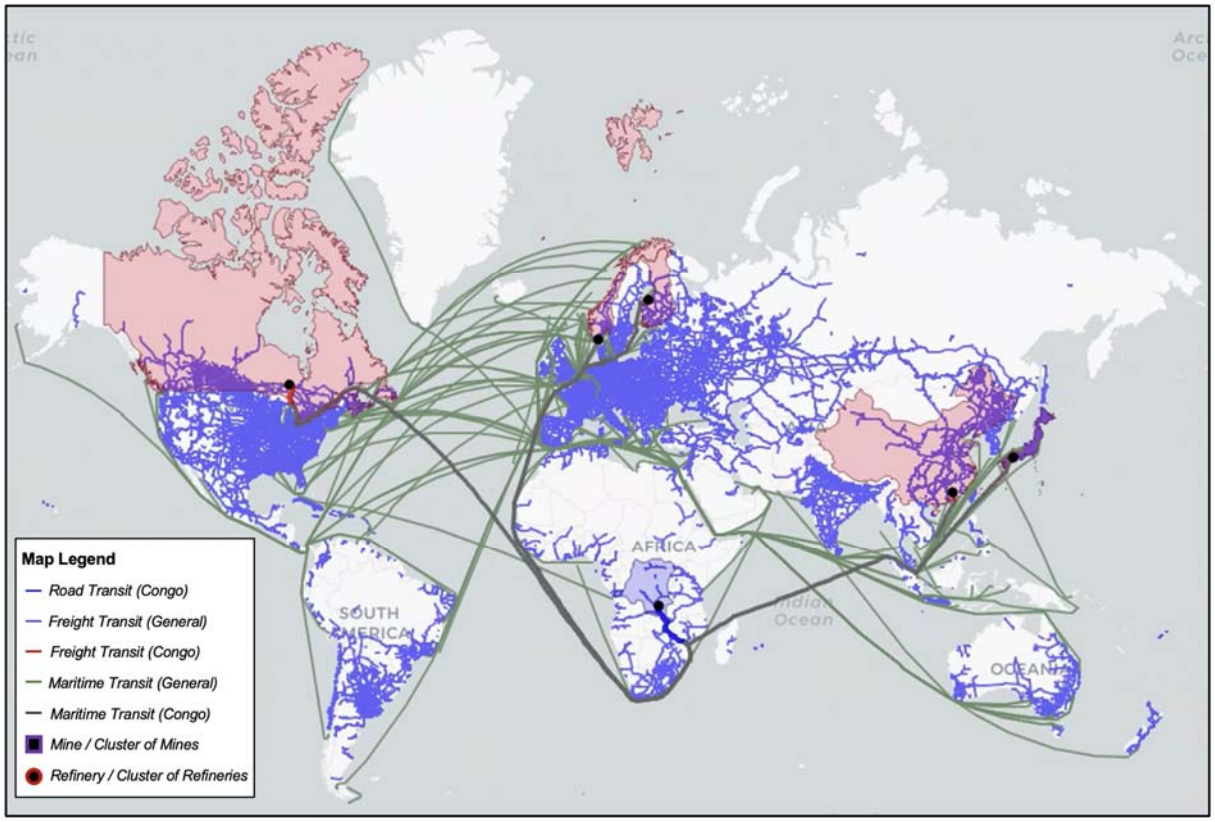

Fig. S7 | A portion of the network model used for transportation logistics showing major road, rail, and maritime routes. The overlapping transport of cobalt concentrate from the Democratic Republic of the Congo to major global refineries is also shown[23,24,51,52].

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