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

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19 **Abstract**

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

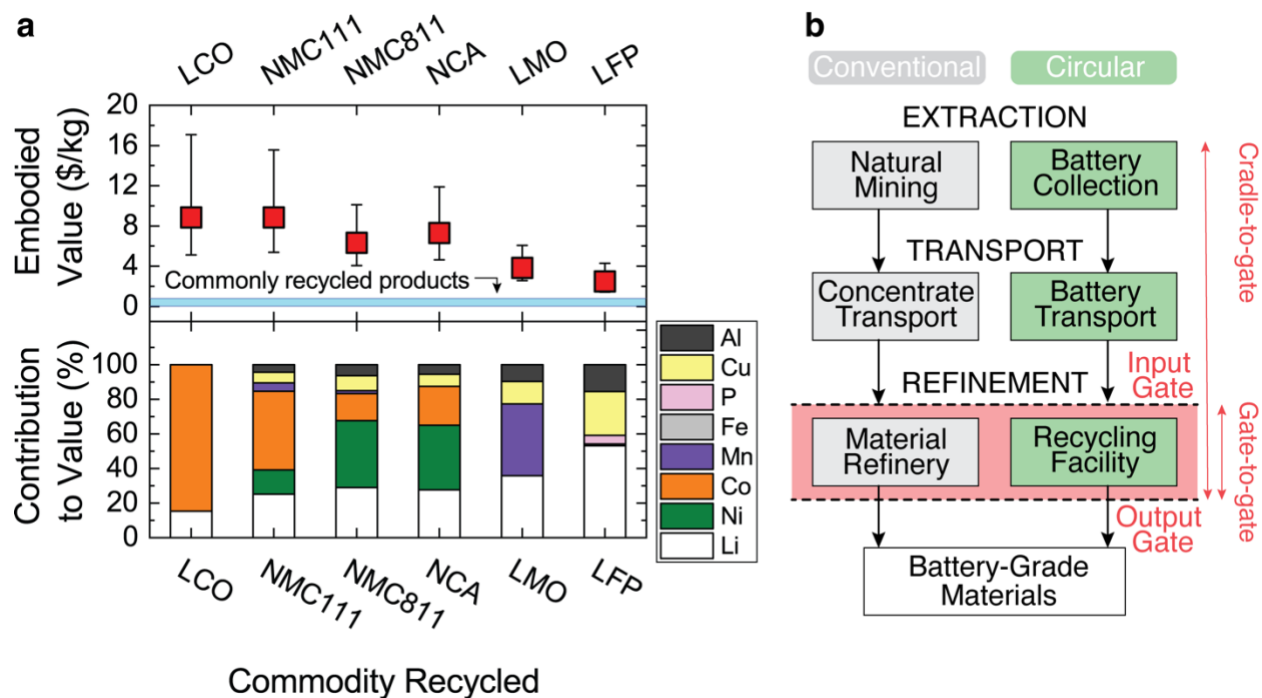
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36 **Keywords:** circular economy, critical materials, hydrometallurgy, life cycle assessment,
37 pyrometallurgy, reductive calcination

38 The rise of intermittent renewable energy generation and vehicle electrification has created
39 exponential growth in lithium-ion battery (LIB) production beyond consumer electronics. By
40 2030, the electric vehicle (EV) sector is projected to dominate LIB growth, accounting for 82% of
41 an estimated 2.4 TWh yr⁻¹ of total global LIB production (**Fig. S1, Supplementary Information**).
42 However, the limited supply of critical materials (e.g., Li, Ni, Co, and Cu¹) needed for prominent
43 LIB chemistries has exacerbated environmental, economic, national security, and human rights
44 concerns^{2,3}. Critical LIB materials are projected to reach major global supply-demand balance
45 deficits before 2030 (**Fig. S1**). Further, both mining of LIB materials and improper disposal of
46 end-of-life LIBs can damage natural and human ecosystems, cause occupational hazards during
47 handling, and result in monetary losses⁴.

48 Recycling critical materials in end-of-life LIBs can help alleviate growing environmental
49 concerns and is essential for the long-term sustainability of electrified transportation. While
50 recycled materials may not contribute substantially to global LIB demand for decades, the
51 establishment of domestic circular supply chains is iterative, requiring multiple learning curves as
52 the dominant supply of end-of-life LIB chemistries and form factors evolve and as supply grows.
53 Factors central to the success of recycling include the ease of collecting products, the cost of
54 recycling processes, and the economic value of recovered materials. The average embodied
55 economic values of representative LIBs between 2018–2021 are shown in **Fig. 1a** (complete
56 references are listed in **Supplementary Information**). In LIBs, between 2018–2021, Li, Ni, and
57 Co comprise the highest embodied economic value, and Al and Cu account for a significant weight
58 percentage of EV battery packs (approximately 25%)⁵. Despite an embodied economic value that
59 is 2–10 times higher compared to the lead in lead-acid batteries, LIBs are only recycled 2–47%
60 globally⁶, compared to 99% for lead-acid batteries in the U.S. Regardless, the untapped potential
61 of LIB recycling constitutes a significant economic and environmental opportunity that requires
62 evaluation across several application scales, from numerous small-scale consumer electronic LIBs
63 (e.g., 10–100 Wh) to fewer large-scale transportation and stationary storage LIB packs (e.g., 10–
64 100 kWh)⁷. In addition, the preferred chemistries by automakers have evolved to hedge potential
65 critical mineral shortages and react to market shifts, such as the near tripling of lithium carbonate
66 prices in early 2022. Existing LIB variation and supply chain complexity highlight the need for a
67 methodical and comparative life cycle assessment (LCA) between circular (i.e., recycling used

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



70
 71 **Fig. 1 | Economic drivers of lithium-ion battery (LIB) recycling and supply chain options for**
 72 **producing battery-grade materials. a**, Commodity values of representative LIBs (upper panel)
 73 and relative contributions of embodied metal elements to the LIB values (lower panel).
 74 Representative LIBs are from consumer electronics using lithium cobalt oxide (LCO), and electric
 75 vehicle battery packs including lithium nickel manganese cobalt oxide (NMC111 and NMC811),
 76 lithium nickel cobalt aluminum oxide (NCA), lithium manganese oxide (LMO), and lithium iron
 77 phosphate (LFP). Data are based on market values adjusted for inflation between January 2018
 78 and December 2021 (complete references are listed in **Fig. S1** in **Supplementary Information**),
 79 and the uncertainty denotes a 90% confidence interval, which may overlap with the data point in
 80 some instances, obscuring their view. The blue shaded area in the upper panel represents the
 81 average commodity values of commonly recycled products: glass, paper, plastic, and metal cans
 82 (more details are provided in **Fig. S1**). **b**, Cradle-to-gate steps of manufacturing battery-grade LIB
 83 materials (i.e., salts) from conventional and circular supply chains, both of which include three
 84 steps: extraction, transport, and refinement. Extraction and transport are considered upstream steps
 85 relative to gate-to-gate refinement, which is indicated by the red shaded area between “input” and
 86 “output” gates. Cradle-to-gate analysis considers the refinement and upstream processes together.

87
88 Despite significant progress, current understanding of the relative environmental impacts
89 of recycling LIBs is still incomplete. The most significant environmental differences between LIB
90 production from circular and conventionally mined cathode material lie early in supply chains,
91 comprised of extraction, transport, and refinement steps (together “cradle-to-gate,” **Fig. 1b**). While
92 several previous studies have investigated cradle-to-gate environmental impacts, gate-to-gate
93 analyses of circular refinement processes are inconsistent, reporting environmental impacts that
94 differ by >30%⁸⁻¹⁰, and are not completely based on industrial-scale LIB recycling operations. The
95 gate-to-gate refinement processes utilized at established and emerging circular refinement
96 facilities may include mechanical separation (Me), pyrometallurgy (Py), and hydrometallurgy
97 (Hy)^{8,9}. Specifically, Me physically dismantles LIBs into constituent components, Py leverages
98 elevated temperature to facilitate thermally-driven material transformations, and Hy separates
99 materials in the aqueous phase via leaching, precipitation, and solvent extraction processes.
100 Variations in environmental impacts arise from the specific operational choices at refinement
101 facilities that utilize different processing pathways and from the methods to evaluate them. There
102 is a critical need for transparency and detailed insights into the environmental impacts (e.g., energy
103 consumption, greenhouse gas emission, and water consumption) of LIB refinement pathways and
104 all cradle-to-gate supply chain steps. Previous efforts have worked towards addressing this
105 need^{8,11}, and this study builds on the comparative methodology of a recent step-by-step study to
106 provide higher resolution and more actionable primary data, insights, and recommendations.
107 Advancing decision-making capabilities to scale sustainable LIB supply chains requires life cycle
108 assessment with more granular data at each step, inclusion of industrial-scale refinement
109 operations with practical mixed-stream battery feedstocks, documentation of operational
110 parameters, and qualification of results in terms of limitations and applicability to real-world
111 scenarios.

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

118 manufacturing facilities and energized end-of-life LIBs collected from consumers. Industrial-scale
119 operational data provided by Redwood Materials are analyzed and compared to conventional LIB
120 supply chain values based on Argonne National Laboratory's Greenhouse Gases, Regulated
121 Emissions, and Energy use in Technologies (GREET 2021) model¹². Second, influences of the
122 product formats in the refinement pathways on environmental impacts are examined. For both
123 conventional and circular refinement, impacts of producing mixed Ni-Co compounds and discrete
124 salts are analyzed. Third, we assess the environmental impacts of upstream processes before gate-
125 to-gate refinement based on modeling. The upstream assessment includes the extraction of LIB
126 material from conventional (i.e., mined ore) or circular (i.e., collected batteries) sources and the
127 transport of extracted material to relevant refinement facilities for production of battery-grade
128 cathode materials as Li, Co and Ni sulfate or carbonate salts. To the best of our knowledge, this
129 study is the first life cycle assessment with primary industrial-scale circular refinement data that
130 includes stepwise, cradle-to-gate comparison of conventional and circular LIB supply chains. With
131 the methodologies and results reported herein, researchers can prioritize major opportunities to
132 improve process efficiencies, practitioners can benchmark their environmental impacts, and
133 policymakers can incentivize best environmental practices in LIB supply chain management.
134 Insights provided by this study can also help recyclers optimize the environmental impacts of their
135 refinement processes.

136 **Results**

137 In LIB supply chains, the refinement step converts the collected feedstocks into battery-grade salts
138 for further manufacturing (**Fig. 2a**). In both conventional and circular supply chains, the
139 refinement pathways vary significantly depending on multiple factors. Five refinement pathways
140 are compared in this study (**Fig. 2b**). While conventional refinement starts with mined ores/brines
141 (1 and 2), circular refinement starts with either end-of-life batteries (1 and 2) or battery scrap (5).
142 Ni and Co in refinement products for subsequent manufacturing can be discrete salts (1 and 3) or
143 mixed compounds (2, 4, and 5).

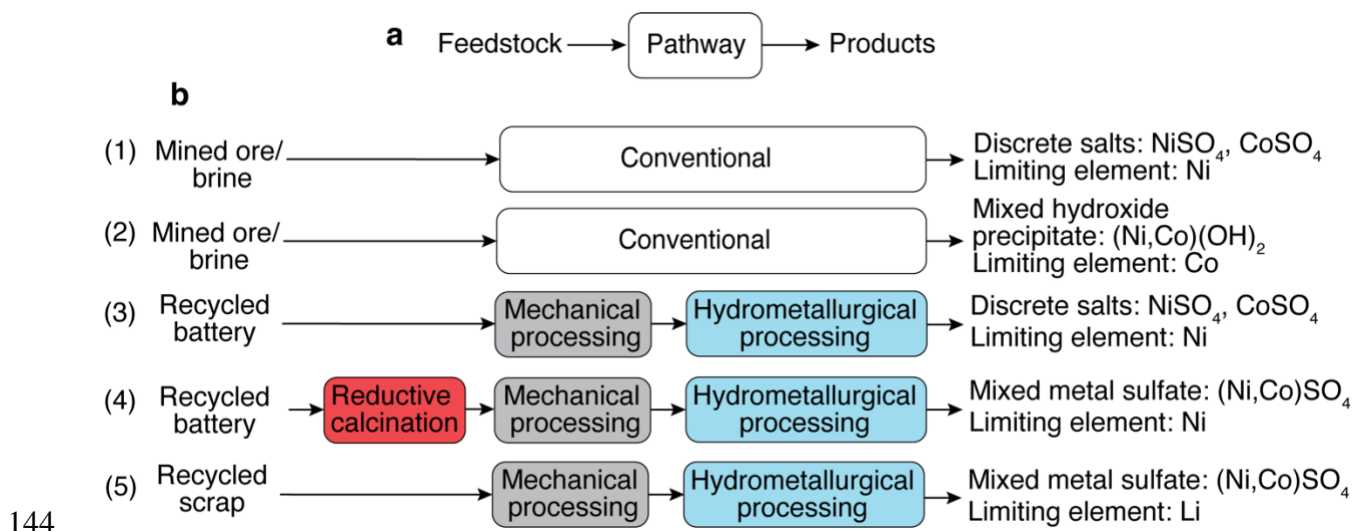
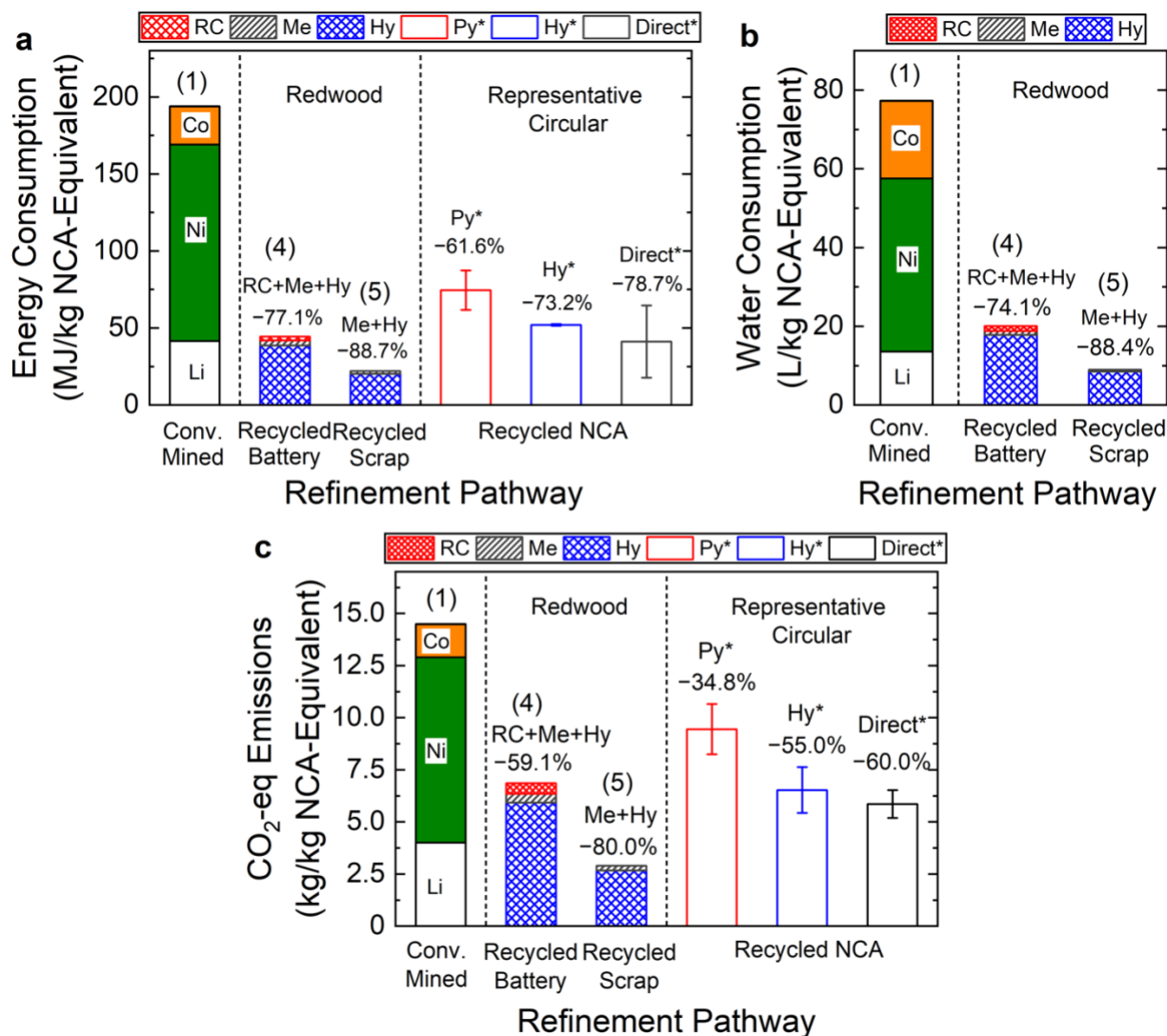


Fig. 2 | Schematic summarizing feedstocks, pathways, and products in refinement analyses.
a, General schematic showing the feedstock, pathway, and products as a legend for the refinement methods shown below. **b**, Five specific refinement analyses in this study: conventional refining (1 and 2) receives mined ore and brines, and circular refining methods (3–5) recycle from end-of-life batteries or scraps. While all methods produce identical Li₂SO₄ and Al₂O₃, Ni and Co products exist in the form of discrete salts, NiSO₄ and CoSO₄ (1 and 3), mixed hydroxide (Ni,Co)(OH)₂ (2), or mixed metal sulfate (Ni,Co)SO₄ (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 (CO₂-equivalents, CO₂-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}O₂. 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 NMC⁶. Excluding the environmental impacts of material extraction

166 and transport steps, the gate-to-gate production of one kg NCA-eq battery-grade material from
 167 conventional mined natural materials consumes 193.9 MJ and 77.3 L H₂O while emitting 14.5 kg
 168 CO₂-eq (**Fig. 3**). The values of energy consumption and greenhouse gas emissions are comparable
 169 with previous studies based on GREET datasets^{11,12} (**Fig. S3**). Refinement of mined material
 170 concentrate into battery-grade Ni material dominates NCA environmental impacts,
 171 representing >57% of total values.



172
 173 **Fig. 3 | Environmental impacts of conventional and circular refining technologies.** a, Energy
 174 consumption, b, CO₂-eq emissions, and c, water consumption of gate-to-gate refinement by
 175 different pathways for NCA battery-grade salts. Numbers in parentheses labelled on the top of
 176 stacked bars denote the refinement methods summarized in **Fig. 2**. The conventional mined

177 pathway (Conv. Mined) refines natural deposits and produce discrete salts (Method (1) in **Fig. 2**);
178 note that Al is presented on the top of the stacked bars but its contributions are too small to be seen;
179 however specific environmental impacts of each element contributor are detailed in Table S1.
180 Circular supply chains refine from either mixed energized end-of-life lithium-ion batteries
181 collected from consumers (Recycled Battery, Method (4) in **Fig. 2**) or non-energized battery scrap
182 from a production facility (Recycled Scrap, Method (5) in **Fig. 2**), producing mixed metal sulfates.
183 Multi-step circular refinement pathways include mechanical processing (Me, grey), reductive
184 calcination (RC, red), and hydrometallurgy (Hy, blue). RC is an additional processing step for
185 energized batteries and is not used for non-energized recycled scrap. Open bars in the right panels
186 denote environmental impacts of recycling NCA batteries with representative existing
187 pyrometallurgical (Py*), hydrometallurgical (Hy*), and direct recycling (Direct*) methods as
188 comparison, and data are obtained from the literature⁸. Literature data is normalized by the same
189 functional unit in this study, and uncertainties are determined by combining two different battery
190 form factors: pouch and cylindrical (detailed in **Table S14–S15**). The vertical dashed line in each
191 graph demarcates different data types, where the model-based conventional and representative
192 existing pathways are summarized in the left panel, operational data from Redwood Materials are
193 presented in the middle panel, and literature data in the right panel. Note that water consumption
194 has generally not been quantified in previous studies, leading to no literature data panel for **Fig.**
195 **3b**. Environmental impacts of material extraction and transport in the supply chains are not
196 included.

197

198 The environmental impacts of two circular refinement pathways are presented in each
199 graph in **Fig. 3** for mixed-stream LIB feedstocks processed at Redwood Materials: non-energized
200 production scrap from LIB production facilities (recycled scrap) and energized, end-of-life LIBs
201 collected from consumers (recycled battery). Using a limiting-reagent approach of output products
202 to produce one kg NCA-eq material, energy requirements for processing recycled scrap and
203 recycled battery streams are 22.0 MJ/kg and 44.4 MJ/kg NCA-eq materials, significantly lower
204 than conventional refinement by 88.7% and 77.1%, respectively (**Fig. 3a**). Relatedly, 2.9 and 6.9
205 kg CO₂-eq/kg NCA-eq materials are generated from scrap and battery streams, respectively, a
206 substantial reduction in CO₂-eq emissions by 80.0% and 59.1% (**Fig. 3b**). Water consumption is
207 also lower by 88.4% for scrap and 74.1% for battery streams relative to the conventional scenario,

208 resulting from the consumption of 9.0 and 20.0 L H₂O/kg NCA-eq materials, respectively (**Fig.**
209 **3c**). Note that while the elemental stoichiometry is identical, the output battery-grade materials
210 vary slightly between conventional (Li₂CO₃, NiSO₄, CoSO₄) and circular (Li₂SO₄, (Ni, Co)SO₄)
211 supply chains (detailed in **Methods**). Converting the final lithium product to Li₂CO₃ does not
212 substantially change the environmental impacts of the circular supply chains (Supplementary Note
213 3, **Fig. S3**), and impacts of producing discrete or mixed products are examined in the following
214 section.

215 To produce battery-grade cathode materials, Redwood Materials uses a combination of
216 reductive calcination (RC), mechanical (Me), and hydrometallurgical (Hy) LIB refinement
217 processes (pathways detailed in **Fig. S2**). The RC process converts energized battery feedstock
218 under certain conditions that leverage heat from exothermic processes and inhibit graphite
219 combustion. This process does not use direct fossil fuel inputs onsite and facilitates subsequent
220 hydrometallurgical refinement into battery-grade materials. Because RC is not required for non-
221 energized LIB production scrap materials, the two feedstock streams (recycled scrap and recycled
222 batteries) are analyzed separately. Energy consumption and CO₂-eq emissions of representative
223 existing recycling pathways from the literature, including pyrometallurgy (Py*), hydrometallurgy
224 (Hy*), and direct recycling (Direct*), are also presented in **Fig. 3** for comparison. In general, the
225 RC+Me+Hy pathway at Redwood exhibits comparable energy consumption and CO₂-eq emissions
226 with Hy and Direct literature values⁸, and substantially lower environmental impacts than Py*.
227 Note that traditional pyrometallurgy and Redwood Material's reductive calcination can process
228 energized batteries of varying states of charge, health, and formats with minimal modification,
229 whereas traditional hydrometallurgy may need to discharge energized batteries in salt bath or
230 cryogenically remove electrolyte for safe mechanical processing. While this analysis is focused on
231 Redwood Materials refinement pathways, the methodology can be used to evaluate additional
232 refinement pathways (e.g., hydrometallurgy in **Fig. S3c**), or others that use different material
233 feedstocks, refinement processes, and energy supplies.

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

239 a combination of technology descriptions from patent applications (the most recent from 2007),
240 literature data on process flow consumptions, industry site visits and surveys, expert advice
241 solicitation, and stated assumptions to form complete pathways. Further, Ciez and Whitacre
242 quantified environmental impacts using output products represented as “metal offsets” for
243 pyrometallurgy or with metals in solution for hydrometallurgy⁸ (**Note 3 in Supplementary**
244 **Information**), rather than cathode salts in this study. In addition, the previous studies included a
245 portion of recycled metal materials in its conventional supply chain analysis, whereas this work
246 references only mined natural deposits in conventional supply chains to fully deconvolute the
247 environmental impacts¹¹. The different conclusions highlight divergent life cycle assessment
248 approaches, processing conditions, and the utility of primary industrial data access over modeling
249 processes from literature sources.

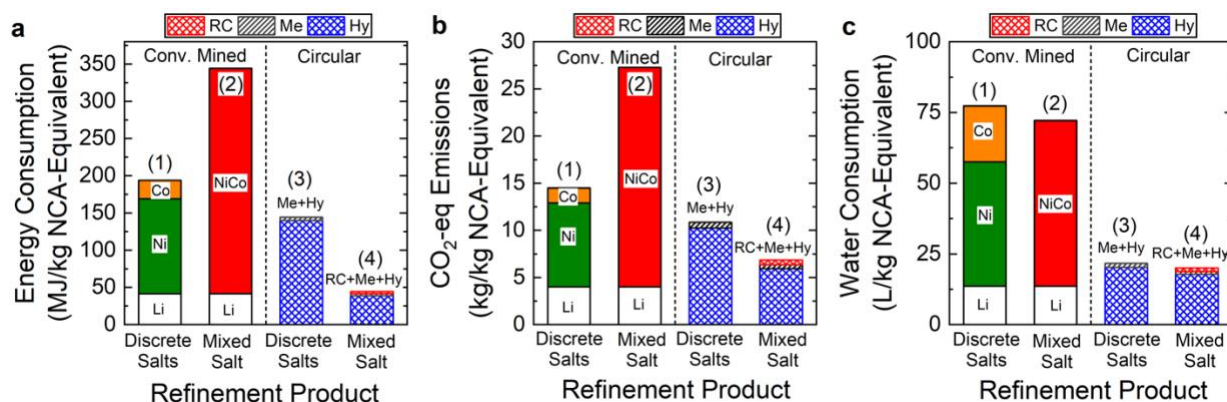
250 **Formats of refinement products influence environmental impacts**

251 Ni and Co are key elements for battery manufacturing, and can be traded in the format of mixed
252 metal salts or discrete salt products between battery refiners and battery manufacturers^{14,15}. To
253 examine the influences of the refinement product formats, the environmental impacts of refinement
254 to mixed salt are compared to the refinement to discrete sulfate salts, NiSO₄ and CoSO₄ (**Fig. 4**).
255 Both conventional and circular refinement pathways are analyzed.

256 The GREET model is employed to analyze different conventional mining pathways
257 generating different product formats (detailed in **Methods**). In conventional mining, refining Ni-
258 Co ores to mixed hydroxide precipitate, (Ni,Co)(OH)₂ (Method (2) in **Fig. 2**), elevates energy
259 consumption and CO₂-eq emissions by 77.% and 89.4%, respectively, over the discrete salts-based
260 pathway (**Fig. 4A** and **4B**, left panels). While the discrete products NiSO₄ and CoSO₄ are produced
261 from the mixed hydroxide precipitates through additional post-treatment, the very low composition
262 of Co (3.6%) in the latter limits the NCA stoichiometry, thus increasing the total energy cost to
263 generate 1 kg NCA-equivalent materials. On the other hand, water consumption of refining mixed
264 hydroxides is slightly lower (−6.6%) than that in producing discrete salts.

265 Circular pathways refining batteries to different products are analyzed using the Redwood
266 data by the RC+Me+Hy process and the modeling of a representative battery recycling method
267 combining mechanical and hydrometallurgy (Me+Hy) refinement (Method (3) in **Fig. 2**). The
268 Redwood process refines recycled batteries to mixed metal sulfate, (Ni,Co)SO₄, whereas the

269 representative Me+Hy produces discrete NiSO₄ and CoSO₄ as the products. The RC pathway
 270 (RC+Me+Hy) exhibits lower energy consumption (−72.3%), CO₂-eq emissions (−39.5%), and
 271 water consumption (−12%) relative to the Me+Hy pathway (**Fig. 4**), because it avoids additional
 272 treatment separating (Ni,Co)SO₄ to discrete salts. Overall, our results indicate that refining
 273 batteries to mixed metal salts instead of discrete salts can substantially save environmental impacts
 274 while still satisfying the needs of circular LIB supply chains. Our findings also provide important
 275 insights to optimizing plant-scale battery refining operations. In the following sections, mixed salt-
 276 based pathways are analyzed for refinement.

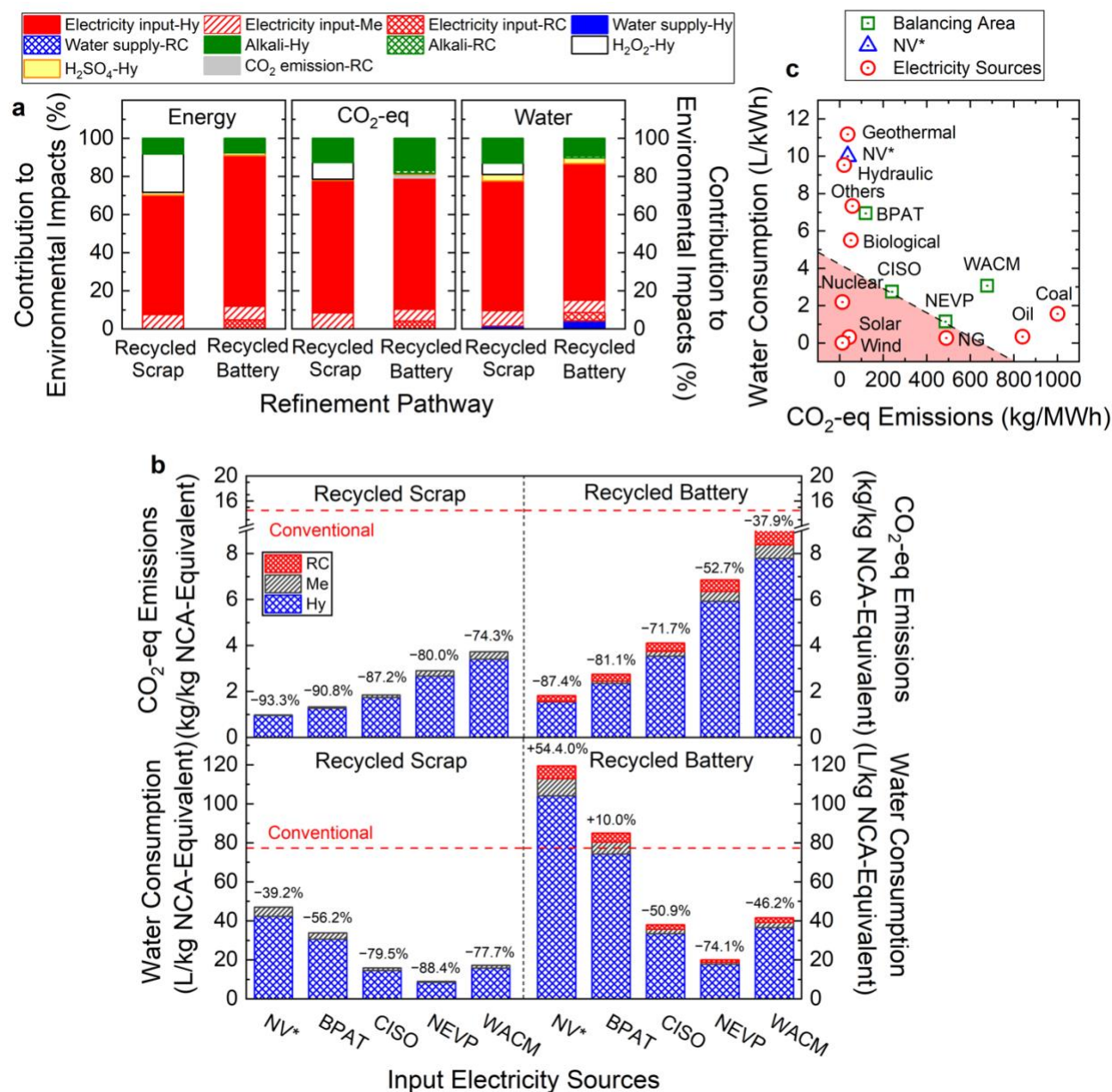


277
 278 **Fig. 4 | Influences of refining products on environmental impacts in circular refining. a,**
 279 **energy consumption, b, CO₂-eq emissions, and c, water consumption.** Left and right panels denote
 280 conventional (Conv. Mined) and circular pathways refining end-of-life batteries to discrete Ni and
 281 Co salts, or mixed Ni-Co salts. Note that Al is presented on the top of the stack bars of conventional
 282 supply chains but its contributions are too small to be seen (detailed values in **Table S1**). Numbers
 283 in parentheses labelled on the top of stacked bars denote the refining methods summarized in
 284 **Fig. 2**.

285
 286 **Electricity consumption dominates the environmental impacts of lithium-ion battery**
 287 **circular refinement.** The relative environmental impacts of input consumables (e.g., energy,
 288 water, commodity chemicals) in the gate-to-gate refinement processes are disaggregated in **Fig. 5**
 289 (additional criteria air pollutants in **Tables S2–S3, Figs. S4–S5**). Note that the embodied
 290 environmental impacts of electricity consumption in **Fig. 3** are based on the Nevada Power
 291 Company (NEVP) at the Redwood Materials location. Electricity consumption is a principal factor

292 dominating the environmental impacts. For both LIB feedstock pathways (Methods (4) and (5) in
293 **Fig. 2**), electricity accounts for 70.3–91.0% of the total energy consumption, 71.8–79.1% of the
294 total CO₂-eq emissions, and 54.3–63.6% of water consumption (**Fig. 5a**). For both feedstocks, Hy
295 processes comprise the majority of environmental impacts, contributing more than 87.3% to
296 energy consumption, 86.3% to CO₂-eq emission, and 88.8% to water consumption. Notably, the
297 additional RC step required for processing energized batteries only marginally contributes to CO₂-
298 eq emissions (7.4% of total). Unlike conventional pyrometallurgical processes that require external
299 energy sources^{8,16}, the RC process is primarily autothermic because it leverages process heat
300 released from exothermic reactions of the LIB materials^{17,18}. In addition to electricity consumption,
301 chemical reagents used in circular refinement processes also contribute to embodied environmental
302 impacts. Alkali reagents used to precipitate metals contribute between 19.0–21.3% of
303 environmental impacts (largest relative contribution to water consumption). H₂O₂ is used to reduce
304 high-oxidation state metal compounds for hydrometallurgical leaching of scrap material, and
305 accounts for 11.3–20.1% of environmental impacts (largest relative contribution to energy
306 consumption).

307



308

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, CO₂-

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 CO₂-eq emissions and water consumption in the LIB recycling operations employed by Redwood

316 Materials methods for production scrap and energized batteries. CO₂-eq emissions and water

317 consumption are based on the resources consumed by unit electricity generated from a Nevada
318 renewable energy tariff (NV*), Bonneville Power Administration (BPAT), California Independent
319 System Operator (CISO), Nevada Power Company (NEVP), and Western Area Power
320 Administration: Colorado-Missouri (WACM). The red dashed lines denote the environmental
321 impacts of the analogous conventional refining process. Note that influences of energy sources on
322 environmental impacts are only presented for the circular supply chains, but not for conventional
323 supply chains. Specific environmental impacts presented in the figures are detailed in **Table S5. c**,
324 Tradeoff relationship between embodied water consumption and CO₂-eq emission by different
325 power sources, including electricity grids in different locations (☉), purely power sources (☐),
326 and Nevada renewable energy tariff (NV*, △). The red dashed line denotes the lower bound of
327 the water-CO₂ performance, i.e., the existing electricity grids that have the lowest water
328 consumption and CO₂-eq emission simultaneously, and the green shaded area covers the power
329 sources that can transcend the current limit of water-CO₂ performance.

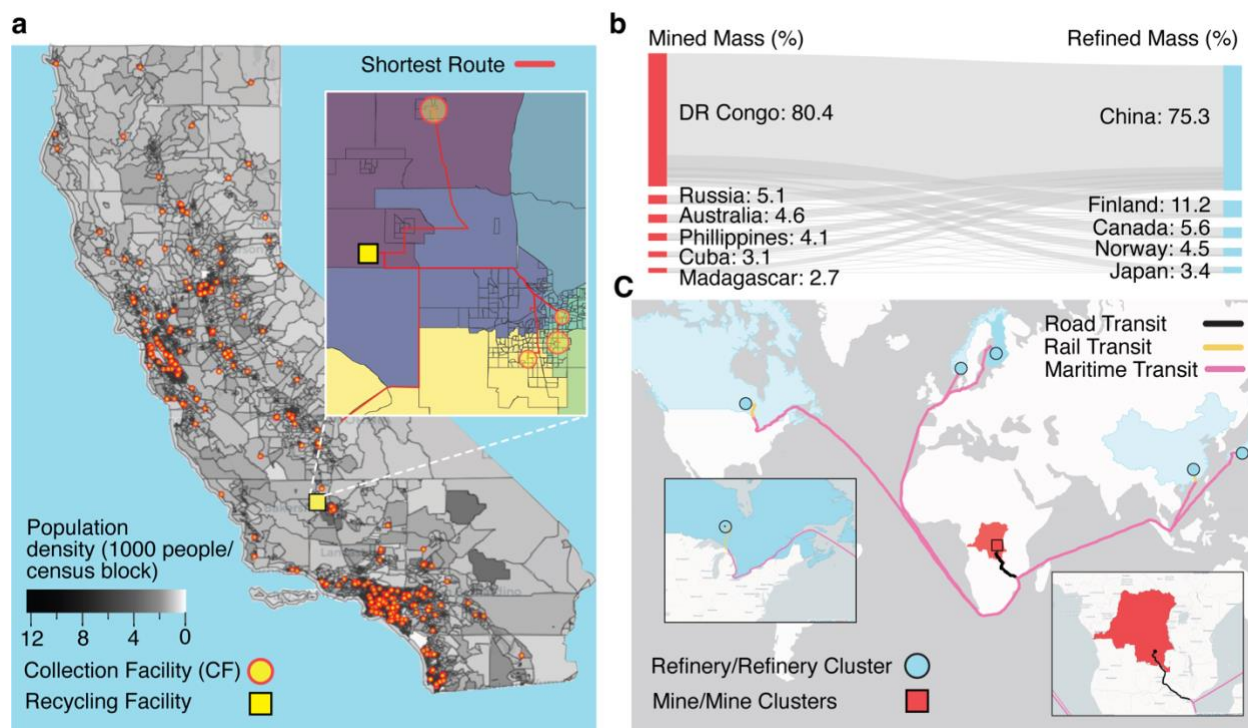
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331 Because electricity dominates the environmental impacts of LIB recycling processes, a
332 comparison of electricity grid balancing areas that emit a range of CO₂-eq emissions per MWh
333 (averaged for 2019)¹⁹⁻²¹ are examined in **Fig. 5b** (additional criteria air pollutants detailed in **Table**
334 **S5**). Substituting NEVP electricity with other balancing areas including Bonneville Power
335 Administration Transmission (BPAT), California Independent System Operator (CISO), Western
336 Area Power Administration of Colorado-Missouri (WACM), and a 100% renewable energy tariff
337 in Nevada (NV*), yields a significant reduction in CO₂-eq emissions of up to 93.3% (recycled
338 scrap) and 87.4% (recycled battery) relative to conventional refinement (**Fig. 5b**). Conversely,
339 employing low-carbon electricity grids can increase water consumption compared with NEVP-
340 based operation, following the order of NV* > BPAT > WACM > CISO > NEVP (**Fig. 5b**). Note
341 that NV*- and BPAT-based circular refinement processes exceed the water consumption level of
342 conventional refinement due to significant contributions from hydro- and geothermal power.
343 Further investigation into the grid electricity sources of balancing areas reveals a tradeoff between
344 CO₂-eq emissions and water consumption based on electricity generation type (**Fig. 5c**); most
345 electricity sources with relatively low CO₂-eq emissions (e.g., those based on bio-, hydro-, or
346 geothermal energy) exhibit high water consumption, and vice versa. This tradeoff also explains
347 the different influences of electricity source on environmental impacts of the Redwood Materials

348 refinement step and other pathways (**Fig. S3d**). However, the electricity sources for each balancing
349 area will affect both CO₂-eq emissions and water consumption. For example, because NEVP-based
350 electricity includes a relatively large proportion (70%) from CO₂-eq emissions-intensive natural
351 gas with low water consumption, a switch to hydro-intensive (73%) BPAT electricity decreases
352 CO₂-eq emissions while increasing water consumption.

353

354 **Environmental impacts of material extraction and transport are significantly lower in**
355 **circular lithium-ion battery supply chains than in conventional supply chains.** Upstream of
356 gate-to-gate refinement are material extraction and transport to refinement facilities (**Fig. 1b**).
357 Environmental impacts of these upstream steps are analyzed for two representative LIB
358 chemistries and battery use cases: NCA in EV battery packs, and lithium cobalt oxide (LiCoO₂ or
359 LCO) in smartphones. California is chosen to assess circular extraction because it has the largest
360 population and EV market share of any state in the U.S.^{22,23}. Smartphones are considered extracted
361 when collected, aggregated, and transported from all California residents (analyzed per census
362 block) to the nearest existing collection facility (CF)²⁴. The analytical model for this circular
363 extraction is depicted in **Fig. 6a**, where a shortest-path route for collection from block group to CF
364 is modeled²². To quantify conventional material extraction environmental impacts from mining,
365 global supply chain data are adapted from GREET (**Table S6–S7**)¹². Smartphone extraction in the
366 circular supply chain emits only 0.0189 kg CO₂-eq/kg LCO-eq, significantly lower than
367 conventional mining (1.96 kg CO₂-eq/kg LCO-eq) by 99.0%. Energy and water consumption are
368 similarly lower in the circular supply chain (**Table S9**).



369

370 **Fig. 6 | A logistics model for assessing upstream environmental impacts of extraction and**

371 **transport in circular and conventional lithium-ion battery supply chains. a,** Modeled circular

372 extraction of LCO-based smartphones from every census-block group based on population to the

373 closest, existing private or municipal collection facility (CF) using a shortest-route algorithm. Inset

374 details modeled circular transport of smartphones aggregated at CFs and then transported to a

375 central recycling facility at the center (gravity point) of the California population by the shortest

376 route (red lines). Colors of block groups indicate the catchment area of a specific CF, where CF

377 size shows the relative number of smartphones collected in 2019. **b,** A weighted distribution

378 estimate of international transport logistics for conventional supply chains between mining and

379 refining countries based on cobalt productivity in the top Sankey diagram. **c,** An example of

380 transport logistics for cobalt mined and aggregated in the Democratic Republic of the Congo (DRC)

381 and then shipped via primary road, train rail, and maritime routes using a shortest-distance path to

382 major refinery locations, with insets showing the degree of detail considered. Similar analyses

383 were performed for Li, Ni, Co, and Al. Inserts present more detailed transit routes in DRC and

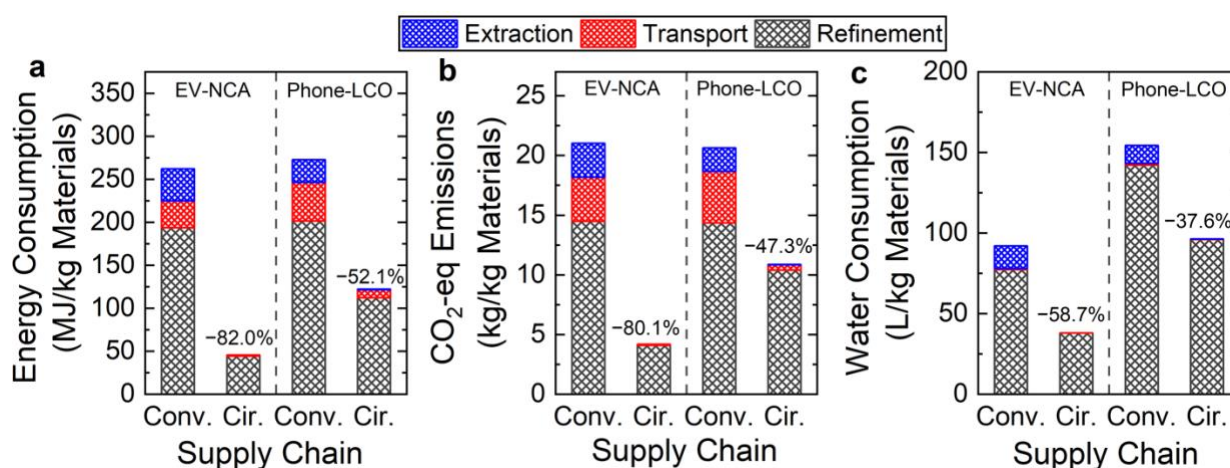
384 Canada.

385

386 After extraction, LIB material concentrates are transported along domestic and
387 international routes by truck, train rail, and maritime cargo ship to refinery locations (**Fig. S7** and
388 **Table S8–S15**; complete references in **Supplementary Information**). An algorithm is developed
389 to quantify environmental impacts based on a weighted distribution of participating countries and
390 the shortest distance along major transport routes (the case of cobalt is presented as an example in
391 **Fig. 6b**.) Conventional mine-to-refinery environmental impacts are calculated for one kg of
392 embodied Li, Ni, Co, and Al metal (**Table S7**). While transport emissions for Li, Ni, and Co range
393 from 5.4–6.4 kg CO₂-eq/kg embodied metal, Al is three times lower. For the circular case applied
394 to California, smartphones and EV battery packs collected at CFs are transported to a hypothetical
395 central LIB circular refinement facility at the population-weighted center (i.e., gravity point) of
396 California (near Bakersfield)²². In conventional supply chains, transporting mined material
397 concentrates accounts for 3.68 kg CO₂-eq/kg NCA-eq and 4.32 kg CO₂-eq/kg LCO-eq. By
398 comparison, emissions for the transport of aggregated end-of-life NCA EV battery packs (i.e., not
399 disassembled) and LCO smartphone batteries (not separated from phones) to a circular refinement
400 facility are 0.073 kg CO₂-eq/kg NCA-eq and 0.47 kg CO₂-eq/kg LCO-eq, 98.2% and 89.1% lower
401 than transport of mined concentrate, respectively. The reduction in CO₂-eq emissions is attributed
402 to differences in elemental concentrations of transported materials and aggregate transport distance
403 (e.g., a weighted average of 224 km for circular NCA-eq materials, and 57,600 km for conventional
404 NCA-eq materials).

405
406 **The refinement step dominates environmental impacts of circular and conventional supply**
407 **chains.** Combining material extraction, transport, and refinement steps yields a cradle-to-gate
408 comparison of the most differentiated steps of conventional and circular LIB supply chains for
409 producing battery-grade cathode materials (**Fig. 7**). Here the environmental impacts of the LIB
410 refinement step in California are analyzed for a hypothetical scenario employing the same circular
411 multi-step refinement technologies as Redwood Materials (i.e., RC+Me+Hy) in Nevada, but using
412 California (CISO) electricity to produce battery-grade cathode materials. A circular supply chain
413 in California for NCA EV and LCO smartphone batteries lowers energy and greenhouse gas
414 emissions by at least 47.3% and water consumption by over 37.6%. In the case of recycling NCA
415 EV batteries in California, the entire cradle-to-gate greenhouse gas emissions of the circular supply
416 chain are lower than the transport emissions of mined concentrate in conventional supply chains

417 (Fig. 7, Table S8). Circular production of LCO-grade materials leads to higher environmental
 418 impacts than that of NCA-grade materials based on the mixed-stream feedstock composition
 419 processed by Redwood Materials. Overall, upstream steps (extraction and transport) contribute
 420 marginally to the total environmental impacts of both circular supply chains, accounting for $\leq 4.9\%$
 421 CO₂-eq emission, $\leq 8.2\%$ energy consumption, and $\leq 0.24\%$ water consumption. Accordingly, the
 422 refinement process dominates the environmental impacts of the circular supply chain. In contrast,
 423 upstream steps in the conventional supply chain play a larger role (still smaller than refinement)
 424 in cradle-to-gate environmental impacts, contributing between 7.8–31.0% to the environmental
 425 metrics considered (Table S8).



426
 427 **Fig. 7 | Cradle-to-gate environmental impacts of different supply chains. a,** Energy
 428 consumption (left), **b,** CO₂-eq emissions (middle), and **c,** water consumption (right) of
 429 conventional (conv.) and circular (cir.) supply chains by step including material extraction,
 430 transport, and refinement. NCA-eq cathode used in electric vehicles (EV-NCA, left panels) and
 431 LCO-eq cathode material used in smartphones (Phone-LCO, right panels) are provided.
 432 Environmental impacts of refinement are analyzed based on electricity generated from balancing
 433 grid authority CISO and upstream supply chain steps (extraction and transport) are based on data
 434 from GREET and transport models developed in the preceding section and depicted in Fig. 6.
 435 Specific environmental impacts of each step are detailed in Tables S5–S7.

436 437 Discussion

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

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

449 Disaggregated analysis of LIB refinement pathways at Redwood Materials provides
450 important insights into the performance and potential of different refinement processes. While
451 pyrometallurgical processing is widely considered as more environmentally intensive than
452 hydrometallurgy, Redwood Materials' RC pathway exhibits much lower environmental impacts
453 than current Hy-containing pathways reported in practice and in literature (**Fig. S3**). The optimized
454 conditions of RC processing minimizes the combustion of carbon-containing LIB materials,
455 significantly reducing CO₂-eq emissions while simultaneously generating products that are
456 amenable for hydrometallurgical separation. Because chemical consumables such as H₂O₂ are
457 important contributors to hydrometallurgy, environmental impacts of Hy processes could be
458 reduced through more sustainable (e.g., electrochemical) production methods²⁵. Our findings also
459 advocate the refinement products of mixed metal sulfates over the single salts, indicating that the
460 further separations among Ni and Co salts can be avoided. An emerging alternative LIB recycling
461 technology, "direct recycling", recovers functional battery materials without decomposition into
462 substituent elements, and is reported to exhibit comparable environmental impacts to Redwood
463 Materials methods⁹. However, direct recycling is still under development and warrants further
464 assessment after process optimization and industrial-scale implementation.

465 Electricity greatly influences environmental impacts in LIB circular refinement, and the
466 variability among grid electricity sources elucidates a tradeoff between CO₂-eq emissions and
467 water consumption (**Fig. 5**). Therefore, considering water consumption and CO₂-eq emissions is
468 necessary for selecting recycling facility locations, particularly in water-sensitive or emissions-
469 sensitive scenarios. Further examination suggests that the tradeoff is primarily driven by water-
470 intensive hydroelectric and geothermal electricity in certain locations versus CO₂-intensive coal

471 and natural gas in others, implying that increasing the proportion of electricity from nuclear, wind,
472 and solar energy sources simultaneously reduces CO₂-eq emissions and water consumption
473 relative to existing balancing areas (**Fig. 5**).

474 Analyses of upstream environmental impacts inform better operations for future resource-
475 saving extraction and transport. Conventional mining and concentrating of ore or brine is resource-
476 intensive due to the low natural concentrations of critical materials (0.01–1%), while critical
477 material concentrations for transport rise to 3–15% after beneficiation. Further concentrating
478 materials near mine sites or building reinterests closer to sources can efficiently reduce
479 environmental impacts of the conventional mined materials. In contrast, smartphones contain 5%
480 LCO material by mass, with the batteries themselves at 24% LCO²⁵. Circular material extraction
481 via LIB collection decreases environmental impacts by 99% versus conventional. A “shortest-
482 route” approach is used in this study to quantify the environmental impacts of battery extraction
483 and transport supply chain steps. Practical battery collection operations will likely vary based on
484 route selection and preprocessing strategy further influencing environmental impacts²⁶. For
485 example, the disassembly of collected EV battery packs or removal of smartphone batteries from
486 devices prior to transport to a recycling facility can increase energy usage through extraction but
487 reduce environmental impacts by lowering transportation weight (**Table S10**). Trucks are used as
488 the primary vehicle for transport analysis given regulatory concerns that consider LIBs hazardous
489 material in many transportation scenarios²⁷. However, alternative transport like railway can further
490 lower environmental impacts by approximately four times versus trucking (**Tables S12**). Upstream
491 process optimization of environmental impacts warrants further investigation, such as the active
492 area of high-throughput automation of LIB extraction from non-standardized devices and EV
493 battery packs or rapid assessment of LIBS for second life uses.

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

501 As the prevalence of LIBs grows in the mobility sector and beyond, strategic placement of
502 domestic LIB collection, refinement and manufacturing facilities can further minimize future
503 environmental impacts by considering heterogenous LIB growth by location, collection approach,
504 transportation distance, and electricity source for refinement processes. As LIB production scales,
505 policies informed by consumer surveys, focus groups, pilot testing, and diverse stakeholder
506 engagement will be needed to research and scale battery collection²⁸. Business models for
507 collection of all LIB types and sizes will likely vary from manufacturer-led to municipal or private
508 collection programs. In addition to collection costs, the varied scale of collection requires further
509 investigation, particularly for localized environmental impacts. Notably, analogous economic and
510 environmental impacts to local ecosystems of conventional mining are not considered in this
511 analysis, and warrant future studies²⁹. Additionally, designing and manufacturing LIBs for
512 recycling in a circular economy can reduce resource usage identified in this study³⁰. Future efforts
513 should also focus on optimizing refinement processes for subsequent steps of the circular supply
514 chain in LIB manufacturing, product performance, and economic cost.

515

516 **Methods**

517 **Goal and scope.** The goal of this study is to compare stepwise cradle-to-gate environmental
518 impacts (energy consumption, CO₂-eq emission, and water consumption) for two supply chains: a
519 conventional, linear supply chain fed by natural mined material for refinement into battery
520 materials, and a circular supply chain fed by LIBs. Both supply chains produce battery-grade
521 cathode materials. A cradle-to-gate analysis of the whole supply chain considers steps of material
522 extraction, transport, and refinement, and gate-to-gate analysis investigates the refinement step,
523 which is focused on in this study. A gate-to-gate scope is broadly defined as the boundary
524 surrounding processing facility operations. In this analysis, gate-to-gate refinement only considers
525 direct processing (e.g., alteration, concentration, precipitation) of the feedstock material once it is
526 extracted from its original state and transported to the refinement location (shown in **Fig. 1b**). For
527 Redwood Materials, this scope includes mechanical processing, reductive calcination, and
528 hydrometallurgy (**Fig. S2**). The system boundary does not include other operations outside of the
529 direct refinement processes as discussed in study limitations below.

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

536 **Methodology.** An attributional life cycle assessment is conducted to quantify and compare
537 conventional and circular LIB supply chains for the production of battery cathode materials. This
538 analysis complies with the International Organization for Standardization (ISO) 14040 standards
539 but omits conversion to environmental impact indicators and external review³¹. Data for
540 conventional material extraction (e.g., mining) and refining are adapted from the Argonne National
541 Laboratory's Greenhouse gases, Regulated Emissions, and Energy use in Transportation
542 (GREET®) 2021 model. GREET and the ecoinvent 3.3 database³² are employed for life cycle
543 inventory data of chemical consumables for the conventional and circular supply chains.

544 To assess circular LIB refinement, primary operational data detailing energy, water, on-
545 site emissions, and consumables usage are provided by Redwood Materials and normalized to
546 mass flows of the different elements of interest in input feedstocks and output products. A
547 representative prevailing circular refinement, Method (2) in **Fig. 2**, is modeled with the software
548 HSC Sim³³, based on the technical procedures available in the literature³⁴⁻³⁷ and the practical
549 feedstock amount received by Redwood.

550 Conventional refinement was modeled by aggregating the environmental impacts of the
551 individual refining pathways for each LIB cathode element (**Table S1**), normalizing by the mass
552 of the individual element of interest within the output product (e.g., Li in Li₂CO₃) and then
553 normalizing again by the mass of that element in the functional unit for this life cycle assessment
554 (defined in the next section). For elements where more than one pathway of production exists in
555 the GREET model (i.e., Ni and Li), the overall environmental impacts are calculated by averaging
556 pathways weighted by their respective share of global production (45% Li production from brine
557 and 55% from ore, and 60% Ni production from mixed hydroxide precipitate and 40% from Class
558 1 Ni). Both discrete and mixed output products are considered. Discrete salts from conventional
559 refinement are Li₂CO₃, NiSO₄, CoSO₄, and Al₂O₃; alternatively, (Ni,Co)(OH)₂ is considered as

560 the mixed product. Lithium outputs produced by Redwood Materials are Li_2SO_4 (environmental
561 impacts for converting to Li_2CO_3 are detailed in Supplemental Information **Note 3**), and other
562 outputs exist as mixed metal sulfates of $(\text{Ni}, \text{Co})\text{SO}_4$ or as Al_2O_3 and $\text{Al}(\text{OH})_3$. With additional
563 treatment further transform the mixed metal sulfate into separate Ni and Co compounds, discrete
564 salts as NiSO_4 and CoSO_4 are analyzed based on modeling of a prevailing Hy+Me refinement
565 pathway. In the cradle-to-gate analysis, material transportation between stages was not included
566 because it was not consistently available in the GREET model. Mixes vary between elements, as
567 well as between pathway stages. For example, crude production of $\text{Co}(\text{OH})_2$ uses a distributed
568 electricity source in the Democratic Republic of the Congo, and the refinement of these materials
569 into CoSO_4 and CoCl_2 uses a distributed electricity source in China. While exploring the sensitivity
570 of environmental impacts for conventional battery material production is important, it is beyond
571 the scope of this paper, and instead, this work focuses on the sensitivity of electricity sources in
572 U.S.-based LIB recycling. See **Supplementary Data File A** for the breakdown of the conventional
573 refining data workflow.

574 **Defining functional units.** Functional units standardize comparisons of the resource consumption
575 and emissions in life cycle assessments. In this study, two functional units are considered in this
576 assessment to normalize environmental impacts between conventional and circular supply chains:
577 the battery-grade material required to make one kg of stoichiometric lithium nickel cobalt
578 aluminum oxide ($\text{LiNi}_{0.80}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$, NCA-eq) and lithium cobalt oxide (LiCoO_2 , LCO-eq)
579 cathode material. Mass was selected as the primary normalizing factor because any energy-based
580 functional unit (e.g., per kWh) could vary based on battery manufacturing and cycling
581 characteristics. The NCA chemistry was selected because reports suggest future cathodes may
582 utilize less Co compared to NMC batteries in EVs, and NCA comprised the second-largest
583 category of EV battery chemistries in 2016, following NMC batteries⁷. LCO is a representative
584 chemistry used in handheld rechargeable devices (e.g., cellphones and laptops) which are currently
585 available to recycle in larger quantities than EV LIBs. The environmental impacts of other LIB-
586 relevant materials (Cu and Mn) in conventional supply chains can be found in **Table S7**.

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

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

598 **Life cycle inventory and assessment.** The life cycle inventory (LCI) data for conventional mining
599 pathways are normalized by each critical metal element: Li, Ni, Co, Al, Cu, and Mn (**Table S7**).
600 The LCI for consumables in the Redwood process are adapted from the GREET 2021 model and
601 ecoinvent 3.3 (**Table S2**).^{12,32} The LCI for the Redwood processes also lists water consumption
602 and criteria emissions for different electricity sources by grid balancing areas in the Western U.S.
603 (**Table S6**). Three categories of environmental impacts are detailed in this study: energy
604 consumption, air pollutant emissions, and water consumption. Energy consumption includes the
605 input electricity for different applications and the energy required to produce required
606 consumables. Criteria air pollutant emissions include the embodied emissions generated by the
607 production of input electricity and the consumed reagents. CO₂, CH₄, CO, NO_x, N₂O, SO_x, PM₁₀,
608 and PM_{2.5} are the air pollutants provided in the GREET model and considered here. The
609 greenhouse gas emissions are reported as CO₂ equivalents (CO₂-eq) summing CO₂, CH₄, and N₂O
610 weighed by the corresponding 100-year global warming potential (GWP). Water consumption
611 considers the withdrawn water that is not returned to the original source, and both the input city
612 water usage and the embodied water consumption in electricity generation and the manufacturing
613 of consumable materials are included.

614 **Estimating environmental impacts of material extraction.** For conventionally mined ore and
615 brine, energy consumption, CO₂-eq emission, and water consumption values are separated for the
616 material extraction processes found in the GREET model. For the circular extraction case, LCO-
617 based smartphones are assumed to be collected and transported to existing private and municipal
618 collection facilities (CFs) from each census block group in CA, assuming every person owned a
619 cell phone and purchased a new phone every three years. A shortest-route algorithm was used for

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

622 **Estimating environmental impacts of material transport.** In the conventional supply chain, a
623 network model of primary transport routes is established that connects mines to refinery locations
624 for Li, Co, Ni, and Al on a country-level basis (**Tables S8–S15**) because the amount of mined
625 material transported from each mine to each refinery was not known. The distances of the shortest-
626 path routes are calculated between mines and refineries by country, predicated on the closest
627 available modes of transport (including road, rail, and maritime). A major mine cluster or refinery
628 location was selected to represent country-level transport values (**Tables S14–S15**) based on
629 production volumes, and distances are quantified between international destinations. These
630 distances are used to calculate the energy consumption, CO₂-eq emission, and water consumption
631 associated with transportation of critical materials as mined concentrate. Mined concentrate is ore
632 or brine that is concentrated locally beyond natural concentration values to reduce weight for
633 transport to a refinery. By considering the total elemental mass and elemental weight percentage
634 of the mined concentrate transported along a route (**Tables S14–15**), the environmental impacts
635 on a per-element basis are calculated as a global weighted average (**Table S11**) with additional
636 process details in **Supplementary Information**.

637 For the circular case applied to California, end-of-life EV NCA LIBs are aggregated at one
638 CF per county closest to its centroid, where county-level data was the most granular data available.
639 All smartphones are aggregated at their nearest CFs. Aggregated smartphone and EV batteries are
640 assumed transported via truck to a single recycling facility located at the gravity point of
641 California's population based on census block-level data (detailed in **Note 4** in **Supplementary**
642 **Information**). The mass-distances traveled are converted to energy consumption, CO₂-eq
643 emission, and water consumption (**Table S8–S9**).

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

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

650 refinement is excluded from the analysis due to the lack of information in GREET. In collection
651 of end-of-life batteries in smartphones, inefficient transport to a CF (e.g., driving each smartphone
652 individually or taking longer transport routes to a CF) is not considered. In addition, all end-of-life
653 EV battery packs are assumed to be driven to each CF in their original vehicles, which is attributed
654 to the “product use” stage instead of extraction in life cycle assessment; therefore, zero CO₂-eq
655 emissions are assumed for the extraction step of EV batteries.

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

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

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