Life-cycle environmental and economic assessment of electric vehicle lithium-ion batteries using different recycling methods in a closed loop supply chain

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Abstract

With growing adoption of the electric vehicles and their lithium-ion batteries (LIBs), increasing urgency has arisen in searching for an environmentally-friendly and commercially-profitable LIB recycling process, and closed-loop recycling has been proved more applicably promising than open-loop recycling as for less virgin material demand. Therefore, this paper addresses environmental influence (carbon emissions and water consumption) and economic impact in closed-loop supply chain. Life-cycle model and process-based cost model are employed to quantify differences among three recycling methods (pyrometallurgical, hydrometallurgical and direct physical recycling processes) for LIBs in aspect of five cathode technologies: three types of lithium nickel manganese cobalt oxide (NCM_{111}, NCM_{622} and NCM_{811}), lithium nickel cobalt aluminum oxide and lithium iron phosphate (LFP). Considering the deficiencies of existing studies, we assess the life cycle environmental impact of LIBs at not only the national level but also the provincial level due to the provincial electricity mix disparity in China. Results demonstrates that direct physical recycling process (DPRP) has the lower environmental burdens and higher economic feasibility over the other methods, excluding LFP cells in which mitigated carbon emissions and higher economic viability are observed but meanwhile direct recycling process water consumption increases. Surprisingly, the majority of high-emission provinces that have relatively higher proportion of thermoelectricity are classified as low water consumption areas. Other contradictory outcome also indicates that the three objectives (carbon emission reduction, water consumption reduction and economic development) may not meet simultaneously, which appeals for further comprehensive research on these policy-making-related indicators.

Keywords: Recycling processes; Closed loop supply chain; Lithium-ion battery; Carbon emissions; Water consumption; Economic benefits.

1 Introduction

As a sustainable transportation alternative [1], electric vehicles (EVs) are increasingly adopted, promising to shift China's transport sector away from conventional fossil fuel and thus addressing the aggravating air pollution and greenhouse emission issues in China[2,3].

China's EV market has rocketed, with over 1.256 million in 2018 [4], 3.8 times growth from 0.331 million in 2015 [5]. In addition, China aims to achieve the cumulative sales of 5 million electric vehicles by 2020 [6]. Due to the rapid adoption of EVs and the limited lifespan of LIB, which would need to be retired before its usage reaching 70-80% of the initial capacity [7], there will be a large-scale battery scrapping in the future, in which raises concerns about waste management of end-of-life batteries. It is forbidden to dispose at will because the LIB contains toxic chemicals. Recycling is an increasingly attractive alternative from the perspectives of environmental benefits, economic feasibility and so forth [8]. In China, a series of policies and regulations have been formulated to establish a sound battery recycling system and ensure the efficient battery recycling [9].

Although battery recycling could decrease the primary raw material production [8], Ziemann et al. [10] reveal that recycled material will be in substantial oversupply and the virgin material demand will be still in an incredible growth if the quality of the recycled materials is not high enough to meet the requirements of battery manufacture. Compared with open-loop recycling that generates recycled materials not reusable for LIB productions, closed-loop recycling is able to save a significant amount of virgin material consumption [10]. It also reduces dependences on imported materials and the corresponding foreign political risks [11,12]. Therefore, this paper evaluates the environmental and economic impact of life cycle LIBs in closed loop supply chain.

LIB recycling is not yet well-established [8] and its infrastructure is limited [13]. Pyrometallurgy and hydrometallurgy processes are two commonly applied recycling methods, while direct physical recycling as a nascent but promising recovery method is also being developed. It is necessary to understand the environmental impact of LIBs [14,15]. Furthermore, the battery recycling process is a crucial factor affecting the batteries life cycle environmental impacts. Dunn et al. [14] analyze the greenhouse gas (GHG) emissions of the life-cycle lithium manganese oxide (LMO) cells considering three recycling processes, i.e. hydrometallurgical, intermediate physical, and direct physical recycling methods. However, due to the instability and low energy density [16], LMO batteries are less suitable for EVs. Ciez, Whitacre [13] conduct a cradle-to-grave analysis to evaluate the GHG emissions and breakeven costs of three type's cathode batteries (NMC_{622}, lithium nickel cobalt aluminum oxide (NCA) and LFP) as well as the environmental impacts of pyrometallurgical, hydrometallurgical and direct physical recovery methods. However, two most commonly used NCM batteries, NCM_{111} and NCM_{811}, were not examined. While most of the literature focused on the US context [13,14], China has by far the largest EV market [17], it is of importance to evaluate the environmental and economic impacts of power batteries in China.

Moreover, water consumption was not included in the LIBs life-cycle environmental impact assessments. Examining the water consumption has its unique significance because the escalation of the water demand is a Gordian knot for many countries [18].

Although Onat et al. [18] and Liao et al. [19] evaluated EV water consumption in the electricity production stage, water consumption in the manufacturing and recycling stages were overlooked. Kim et al. [20] included the battery recycling process for their evaluation of life-cycle water consumption of Ford Focus Battery Electric Vehicles but their calculation is coarse. A detailed analysis into the recycling process life-cycle water consumption comparing different cathode materials needs to be carried out.

This study conducts a life-cycle analysis to evaluate the GHG emissions, water consumption and economic impacts of EV LIBs using different recycling methods in a closed loop supply chain. Using national electricity data, we calculate the life cycle GHG emissions and water consumption of battery using pyrometallurgical, hydrometallurgical and direct physical recycling proesses. A hot spot analysis is conducted to identify the emission-intensive and water-intensive steps. In addition, the sensitivity analysis is applied to assess the influence of recycled material proportion. Secondly, considering GHG emissions and water consumption of the electric power sector differ substantially by province but provincial assessments are lacking in the existing literature, this work evaluates and compares the environmental impacts of battery life cycle based on provincial data. Finally, a process-based cost model is established to calculate the cost of cell manufacturing with the virgin materials and the recycled materials. Lastly, the sensitivity analysis is employed to determine the maximum affordable purchase price of spent batteries at the breakeven recycling cost, and to evaluate the impact of throughput.

2 Methodologies and data

2.1 Life cycle assessment

Life cycle assessment (LCA) has been employed extensively to quantify environmental impacts over the entire life cycles of products and processes [21]. It provides significant insights for the development of circular economy. In addition, LCA could identify "hot spots" of environmental effects and therefore offer substitution suggestions [22].

The main objective of this study is to calculate the life cycle environmental impacts (including energy consumption, GHG emissions and water consumption) and cost of LIB in a closed loop supply chain in China, comparing with battery manufacturing with virgin materials. Three recycling methods (pyrometallurgical, hydrometallurgical and direct physical recycling processes) and five cathode technologies (NMC_{111}, NMC_{622}, NMC_{811}, NCA and LFP) are assessed. In addition, a hot spot analysis is used to determine the emission-intensive and water-intensive steps.

ReCell [23] model developed by Argonne National Laboratory is applied to quantify transportation-related cost and environmental impacts throughout the life cycle of LIB. This model leverages GREET [24] model and BatPaC [25] model, which are applied extensively to analyze the environmental influence (GREET), performance and cost (BatPaC) of LIB.

A process-based attributional LCA is employed in this study, to assess the raw material extraction and processing, manufacturing and recycling of LIBs. It is worth noting that the use phase of LIBs is not the focus of this paper. The BatPaC model develops life cycle inventories for batteries and the characteristics of the five battery technologies are outlined in Table 1. GREET model is employed to analyze the material and energy flows and calculate the energy input, GHG emissions and water consumption. The data are obtained from government reports, literature, GREET model, BatPaC model and ReCell model. Material prices are obtained from market information and default values of BatPaC as shown in Table S1.

Table 1. Battery Parameters Used in this Study [23].

	NMC(111)	NMC(622)	NMC(811)	NCA	LFP
Energy (kwh)	0.143	0.143	0.143	0.143	0.143
Mass (kg)	0.870	0.785	0.803	0.754	1.069
Specific energy (kWh/kg)	0.164	0.182	0.178	0.190	0.134

To calculate carbon emissions and water consumption by GREET model, we use the electricity generation mix and transmission loss data from the National Power Industry Statistics 2016 (China Electricity Council) [26,27]. Due to the huge differences of electricity mix among provinces in China, this paper also conducts provincial assessment. Provincial energy mix of the electric power sector (cf. Fig. 2) and transmission losses in 2018 (c.f. Table S6) are obtained from Wind Database [28].. Note that Hong Kong, Macon and Taiwan are not considered and because the power grids in Beijing, Tianjin and Hebei are closely interconnected, they are regarded as an area called Jingjinji.

2.2 Manufacture and recycling assumptions

Two types of materials, virgin materials and recycled materials recovering from the spent batteries or manufacturing scrap, are considered. Fig 1. shows the recycle module from ReCell model, which is the focus of this study. The recycle module is divided into four parts: collection and transportation, recycling process (via pyrometallurgical/hydrometallurgical/direct physical technology), material conversion and cathode production with the recycled materials [23].

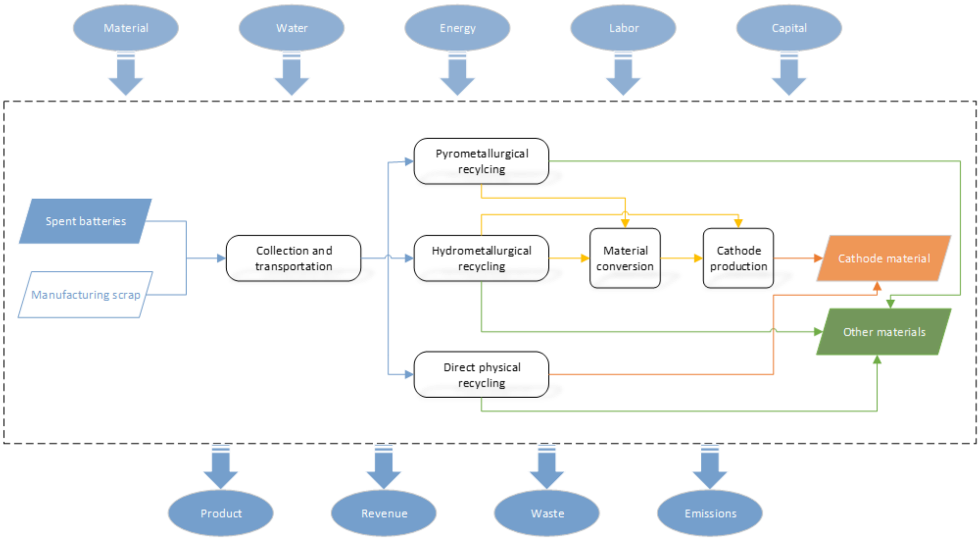

Fig 1. ReCell Model Recycle Module Schematic [23].

In the recycling process, the spent battery packs are recycled via three recycling methods and the recoverable materials are reused to produce new battery cathode, while the other chemicals either are landfilled or burnt (Table 2). Products from the three recycling processes are shown in Table S2-S4. Lithium carbonate is not recovered using a pyrometallurgical process. The major objective of this process is to recover Ni and Co, Li will be eventually in slag because recovering it from slag is an energy-intensive and economically disadvantageous process [14]. Additionally, this paper only considers the direct physical recycling process to recycle LFP batteries since they do not contain precious metals (e.g. cobalt). Besides, due to the high quality requirements of recycled materials [29], it is not economically feasible to construct a closed-loop supply chain for LFP batteries by pyrometallurgical and hydrometallurgical methods.

Table 2. Fate of feed materials during the recycling process.

	Pyrometallurgical	Hydrometallurgical	Direct
Active cathode materials	Recycle	Recycle	Recycle
Graphite	Burn for energy	Landfill	Landfill
Copper	Recycle	Recycle	Recycle
Aluminum	Landfill	Recycle	Recycle
Fe	Recycle	Recycle	Recycle
Plastics	Burn for energy	Landfill	Landfill
Electrolyte	Burn for energy	Burn for energy	Burn for energy
Carbon black	Burn for energy	Burn for energy	Landfill
PVDF	Burn for energy	Burn for energy	Landfill

We assume battery manufacturing with virgin materials and recycled materials have the same energy and material inputs during the entire manufacturing process except the cathode prodution process. The location hypothesis is used to roughly estimate the collection and transportation distance (Table S12), which assume that a city has a complete recycling and manufacturing network (e.g. Shenzhen, China). The production from virgin materials and recycled materials are both 10000 tonne/year.. Furthermore, the proportion of recycled materials is assumed to be 50% for production with recycled materials.

2.3 Recycling methods

Pyrometallurgy promotes oxidation and reduction by high temperature. Slag, limestone, sand and coke are burnt together with the batteries to produce mixed metal alloys. In oxidation and reduction, transition metals such as cobalt and nickel are reduced from oxides to metals and recovered from mixed metal alloys. However, the aluminum and lithium are sent to the slag. Although they could be recovered by leaching [30], it is an energy-intensive and costly process and therefore not normally carried out. Additionally, other materials including graphite, plastics, electrolyte, carbon black and PVDF, are burned in a furnace, to provide energy.

Unlike the pyrometallurgy process, the direct physical and hydrometallurgical processes start with discharging and dismantling battery, so that the external battery hardware can be disassembled and recovered separately. The cathode is dissolved by strong acid in the hydrometallurgical process and a mixture of ionic species is produced in solution. Solvent or precipitation extraction could be used to separate the dissolved constituents from each other and recover the materials, which could eventually reuse to form new cathode materials. Electrolyte, carbon black and PVDF are burned for energy while graphite and plastics are landfilled. In a word, hydrometallurgical process is divided into five parts [31]: discharge and dismantled; removing the current collectors using the process of crushing cathode material; filtering and calcining cathode active materials; grinding; leaching.

Direct physical recycling method generally begins with a discharging and disassembly step. And then super critical CO_2 is used for electrolyte extraction. Granulation process is used to reduce the size of batteries and these will become active material powder (black mass). Black mass is separated by physical processes (e.g. gravity separation). Without causing chemical changes, direct physical recycling method could recover the materials, so that it could make the materials reusable with the least treatment. In the final step, lithium carbonate is added to relithiation and the recycled cathode is made. In the direct physical process, electrolyte is burned for energy while carbon black, PVDF, graphite and plastics are landfilled. The flow diagrams of pyrometallurgy, hydrometallurgical and direct physical processes are shown in the Fig. S1-S3.

2.4 GHG emissions, energy use and water consumption

CO_2e emission with 100-year warming potentials, energy use (MJ) and water consumption (US gallon) per kilogram of LIB batteries and per KWh of energy storage capacity are calculated. The assumptions of process-based GHG emission, energy use and water consumption quantifications are detailed in Supplementary Information [32,33,31,34].

In this paper, we consider the water consumption of battery manufacturing with the virgin and recycled materials. Two parts of water consumption are considered, including material and energy inputs (i.e. the water consumption of electricity and natural gas when manufacturing batteries and recycling the spent batteries). Water consumption of 47 materials are included as shown in Table S5, which is much more refined than previous research [20] . As in Vassolo and Doll [37] and Liao et al. [19], the water consumption for electricity is calculated by the Eqs. (1):

$$W_c = E \cdot I_c \tag{1}$$

where W_c represents the water consumption and E is the electricity use. I_c refers to the water consumption coefficient of electricity generation. In addition, this paper considers the electricity mix at the national and the provincial level to evaluate the spatial differences.

Water consumption factors are shown in Table S9. Data sources include Liao et al. [35], Lin, Chen [36] and the default values in the GREET model. Water consumption of each electricity generation types are detailed in the Supplementary Materials.

Energy consumption avoided, GHG emissions avoided and water consumption avoided are calculate to reflect the environmental impact of various recovery methods. Eq. (2) below shows how these are calculated.

$$A_{m,r} = (T_{m,v} - T_{m,r})/T_{m,v} \tag{2}$$

where m represents energy consumption or GHG emissions or water consumption; r represents different recycling methods; v represents the virgin materials. $A_{m,r}$ is the energy consumption avoided or GHG emissions avoided or water consumption avoided using different recycling methods; T is the total energy consumption or GHG emissions or water consumption.

To evaluate the impact of recycled material proportion on GHG emissions and water consumption, a sensitivity analysis is conducted.

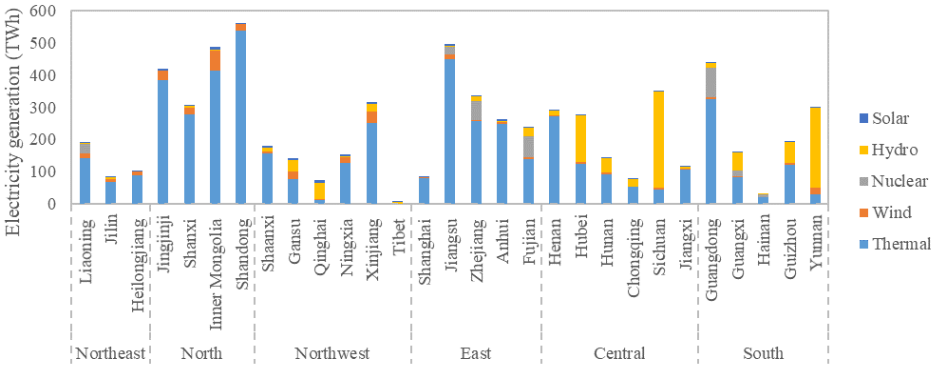

Fig 2. Provincial energy mix in the electric power sector in China in 2018.

2.5 Cost model

This study employs a process-based model (PBCM) to calculate the entire cost of the battery production . In this model, the total cost is determined by many factors including materials, equipment, building, auxiliary equipment, labor, energy, maintenance, and fixed indirect costs [37,38]. The model adds yield losses to adjust production volumes. Table 3 and Table 4 show the assumptions within the facility and the material yield rates for each step. This study calculates the cost and the breakeven cost in the baseline. Meanwhile, in China, the spent battery market is still developing, so the battery recycling market is immature, the price information is not sufficiently transparent and the price is volatile. Therefore, a sensitivity analysis is carried out to determine the maximum affordable purchase price of spent batteries at the breakeven recycling cost. In addition, in order to assess the impact of production, we conduct a sensitivity analysis to analyze the cost changes of production from 1000 to 100000.

Table 3. Facility-wide model parameters.

Input	Data
Actual Processing hours per day (hr)	20
Days per year (days)	320
Plant life (yr)	10
Plant capacity (tonne per yr)	15,000
Throughput (tonne per year)	10,000
Building cost ($/m^2)	$1,500
Direct labor ($/hr)	$2.00
Capital cost adjustment (%)	50%
Discount rate	5%

Table 4. Material yield rates.

	Yield rate
Cell accepted after testing (%)	95.0%
Active cathode material (%)	92.2%
Active anode material (%)	92.2%
Aluminum foil (%)	90.2%
Copper foil (%)	90.2%
Separator (%)	98.0%
Electrolyte (%)	94.0%
NMP recovery rate (%)	99.5%

3 Results

3.1 GHG emissions

Fig. 3 shows the total estimated life-cycle energy consumption, GHG emissions and water consumption to manufacture batteries with virgin materials and recycled materials using pyrometallurgical, hydrometallurgical and direct psychical recycling processes. GHG emissions into those caused by material and energy input.

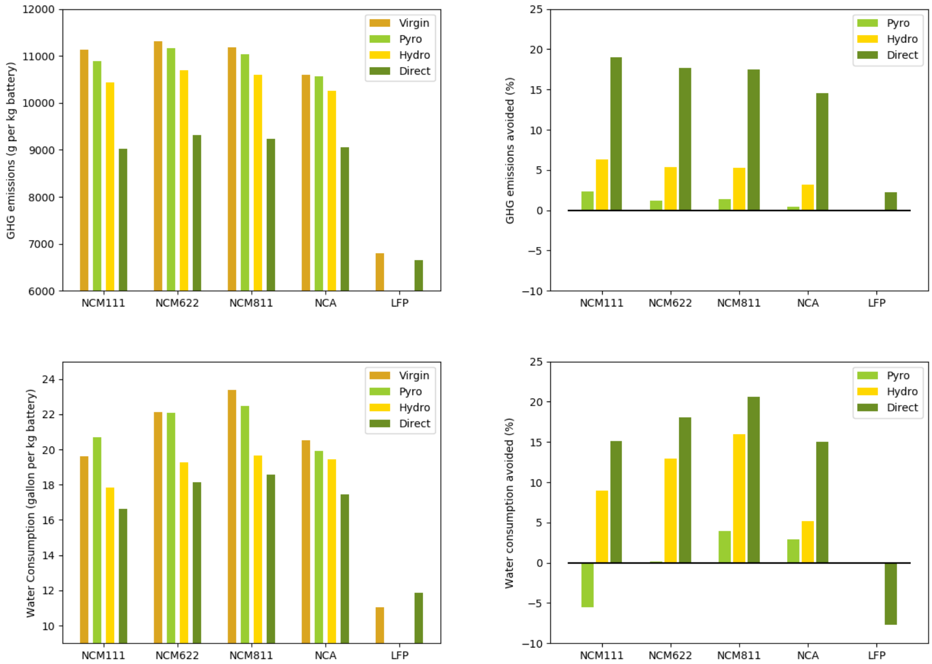

Fig 3. Total estimated GHG emissions (gCO_2e per kg battery), water consumption (gallon per kg battery), GHG emissions avoided (%) and water consumption avoided (%) for NCM111, NCM622, NCM811, NCM and LFP cells. All processes use the national electricity mix data.

All batteries except LFP batteries emit the least GHG when using direct physical recovery method, while having the largest GHG emissions when using the pyrometallurgical recycling process. In addition, for NCM and NCA batteries, these three recycling methods can reduce GHG emissions compared to using virgin material. Nevertheless, Hydrometallurgical and direct physical recycling methods result in significant GHG reductions, with the largest reduction from using direct physical methods and the least with pyrometallurgical process. Direct recovery recycle cathode materials at low temperature with minimal treatment and therefore the least GHG emissions [39].

Breakdown of life-cycle GHG emissions are illustrated in Fig 4. It can be seen that recycling cathode can reduce GHG emissions due to its lower emissions than mining. The direct recovery has the best emission reduction effect, followed by hydrometallurgical and pyrometallurgical recycling methods. Fig 5. shows the breakdowns of GHG emissions for different recycling processes, i.e. pyrometallurgical, hydrometallurgical and direct psychical processes. It can be concluded that the recycling phase emits the largest amount of GHG. For the pyrometallurgical method, the majority (94%) of its GHG emissions is from material combustion including graphite, carbon black, PVDF, plastics and electrolyte organics (c.f. Table 2) and others (6%) from material decomposition, like limestone. The GHG emissions of electricity input accounts for the largest proportion of the total GHG emissions of the hydrometallurgical and direct physical recycling processes. However, the absolute value of direct recovery energy input is far less than that of hydrometallurgical recycling process (Table S8). The calciner has the highest electrical requirements (about 46%) in the hydrometallurgical processes. Hydrochloric acid accounts for significant amounts of GHG emissions for pyrometallurgical and hydrometallurgical recycling processes.

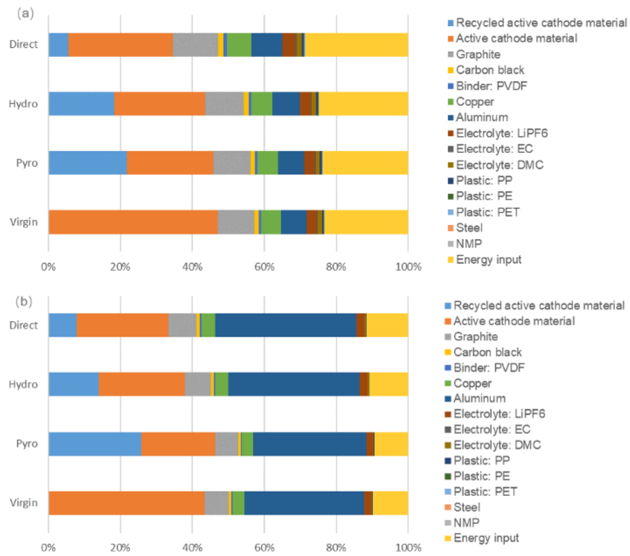

Fig 4. Breakdowns of cradle-to-gate life cycle GHG emissions and water consumption for NCM_{111} battery. (a) life-cycle GHG emissions of battery components and energy input (gCO_2e per kg battery) and (b) life-cycle water consumption of battery components and energy input (gallon per kg battery).

This study finds that direct physical recycling of LFP cells has certain environmental benefits, unlike the research results of Ciez, Whitacre [13], which consider the direct recycling will increase the GHG emissions. However, compared with other types of cathode batteries using direct recycling methods, direct recycle LFP has the minimum effect in reducing GHG emissions, approximately one-eighth of the emission reduction achieved by direct recovery of NCM_{111}. It is because that LFP cathodes use iron precursors, which has the lower GHG emissions level and needs lower energy when mining (c.f. Table S5) than the nickel, cobalt precursors used to produce NCM and NCA cathode, leading to the limited emission reduction effect.

For NCM batteries, we calculate three type of them, with different compositions of lithium, nickel and cobalt. Among them, NCM_{111} has the highest market penetration rate and the most mature manufacturing technology. However, due to the high price in cobalt, which is about 2.7 times the price of nickel (according to the price of April 26 in 2019 given by CCMN [40]), battery manufacturers are striving to develop batteries with higher nickel and lower cobalt content to decrease the costs. Fig. 3 illustrates that producing NCM_{622} with the virgin materials emits the most greenhouse gases. There is insignificant difference in GHG emissions of NCM_{111}, NCM_{622}, NCM_{811} cathode materials. The production process of NCM_{622} is the most energy-intensive (c.f. Table S7) and emits the most GHG in this process while the NCM_{111} using the least energy. Among the three recycling methods, NCM_{111} can avoid the most GHG emissions because the material precursors of NCM_{111} emit highest levels of GHGs per kg so recycling them can avoid excessive carbon emissions during the extraction of raw materials. Although the NCA batteries needs more energy input during manufacturing process (c.f. Table S7), it still has more environmental benefit than the NCM cells because it uses more environmentally friendly materials, which result in the relatively little GHG avoidance after recycling.

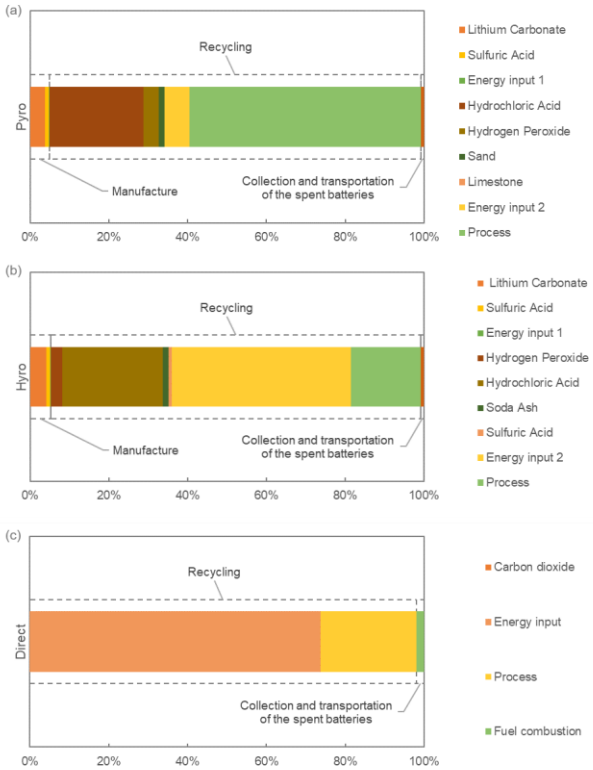

Fig 5. Breakdowns of GHG emissions for recycled cathode materials via pyrometallurgical, hydrometallurgical and direct psychical processes. All batteries are NCM_{111} cells. In the manufacture process, recycled materials will be used to produce NCM_{111} cathode precursor.

3.2 Water consumption

For all batteries except LFP batteries, compared with the other recycling process, direct physical process could result in the greatest reduction in water consumption, followed by hydrometallurgical process. However, the water-saving efficiency of the pyrometallurgy process is less than 5%, even for NCM_{111}, it will bring about a net increase in water consumption. In addition, the reduction effect of the water consumption of NCM_{622} could be neglectable. Among them, the pyrometallurgy process is not a water efficiency recycling process, so its application in water shortage regions should be carefully considered.

As shown in Fig 4, the total active cathode material is the most water-intensive process throughout the battery's life cycle. Besides, aluminum also consumes lots of water. Moreover, recycling cathode material can significantly reduce the water demand of total active cathode material via hydrometallurgical and direct psychical pathways as visualized in Fig 4. However, the water demand will rise when using the pyrometallurgical method. As in Figure 6, regarding the pyrometallurgical process, the recycling process dominates the water consumption. Recycling process and limestone inputs account for 37% and 33% of the total water consumption.The process water use and the limestone are the hot spots of the recycling process, accounting for 37% and 33%, respectively. Unlike pyrometallurgical method, hHydrometallurgy method consumes the most water in the manufacturing process, which is the process of using the recycled materials to produce NCM_{111} cathode precursor. Because hydrometallurgy method consumes less water in the recovery process, which is about one tenth of that in pyrometallurgical method. The process water use and the water consumption of the energy input account for the largest part of manufacturing process. Recycling process also the hot spot of the recycled material, with 49% and 51% of process water use and the water consumption of energy input respectively. The water consumption of carbon dioxide that used in the super critical CO_2 extraction process is insignificant. Meanwhile, it is noted that water demand of the collection and transportation processes for pyrometallurgical, hydrometallurgical and direct psychical processes is also negligible.

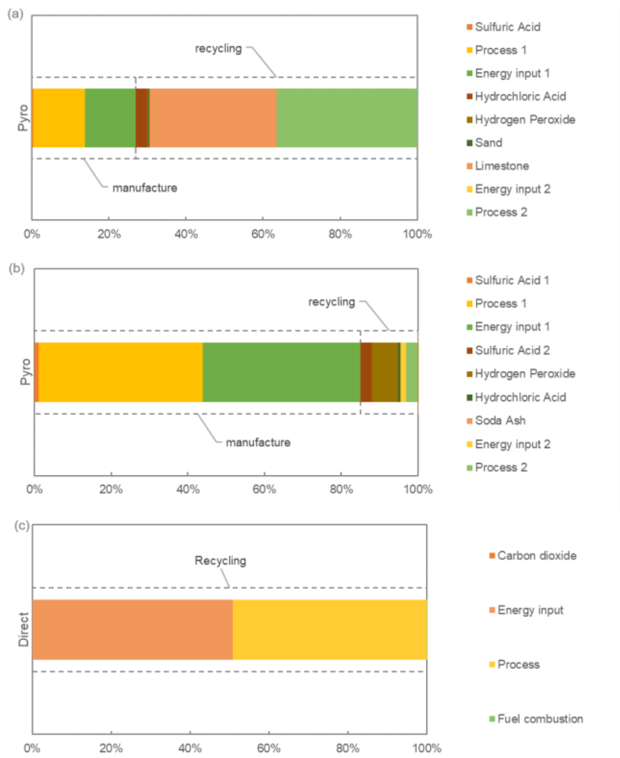

Fig 6. Breakdowns of water consumption for recycled cathode materials via pyrometallurgical, hydrometallurgical and direct psychical processes. All batteries are NCM_{111} cells. In the manufacture process, recycled materials will be used to produce NCM_{111} cathode precursor.

For LFP batteries, this paper finds that although other types of batteries could significantly reduce the water consumption when using the direct physical methods, directly recycle LFP cells would bring about a net augmentation in water consumption. Because LFP cathode precursor materials are the most water-saving materials we consider herein (c.f. Table S5), and it is one-fourteenth of the water consumption of NCM_{811} cathode precursor material. Further tracing the extraction process of raw materials, we could find that compared with the Ni and Co, the iron precursor material used in LFP cells, is most water efficient to mine.

For NCM cells, as the proportion of nickel material in the battery composition increases, the water consumption of life cycle battery rise to a higher point. It is because that the water consumption of nickel sulfate is about 6 times and 33 times that of cobalt and manganese sulfate, respectively. Therefore, although NCM_{811} is more economically viable (see Section 3.3), it may not be suitable to be introduced into water-deficient areas for production. Among the three recycling pathways, NCM_{811} can avoid the most water consumption, even using the pyrometallurgical process that results in the net increase in water consumption of NCM_{111} and the negligible reduction in water consumption of $NCM6_{22}$. This is because the significant nickel content in NCM_{811} cell consumes most water so recycling it can avoid excessive water consumption during the extraction of raw materials. For NCA cells, compared with NCM_{111} cells, NCA batteries consume the similar amount of water when they are manufactured with virgin materials. However, the difference is that when recycled materials original from pyrometallurgical process are used to produce batteries, they will result in a net decrease in water consumption, which cannot be achieved in NCM batteries.

Greenhouse gas emissions and water consumption of NCM111 batteries with varying proportion of recycled materials during their life cycle are shown in Fig 8. It illustrates that with increased proportion of recycled materials, all recycling processes could realize larger GHG reductions, with larger slope of direct recovery curve and faster descending speed, followed by hydrometallurgical and pyrometallurgical methods. Meanwhile, the water consumption have an upward trend via pyrometallurgical process while the water consumption of hydrometallurgical and direct physical methods decline.

3.3 Impact of electricity mix structures

In some regions (e.g. Jingjinji, Shanxi and Shangdong,) more than 90% of the electricity is generated from fossil fuels, which emits the most GHG per MJ electricity while the proportion of thermoelectric can be less than 15% in some provinces like Yunnan and Sichuan. More than 47% of the renewable power (wind power, solar power and hydroelectricity) is generated in the Hubei, Sichuan and Yunnan. For water consumption factors, hydroelectricity has a higher coefficient than other types of electricity power, particularly in the northern and northwestern province because of the local climate and surface conditions, such as strong winds and sparse vegetation [41,35]. In addition, the water consumption coefficient of thermal power plants varies significantly with the different cooling technologies used [42]. In order to evaluate the provincial differences in the GHG emissions and water consumption, provincial electricity mix and provincial water consumption factors are used. The results are shown in Fig 7. We focus on NCM_{111} cells using hydrometallurgical recycling method in this section.

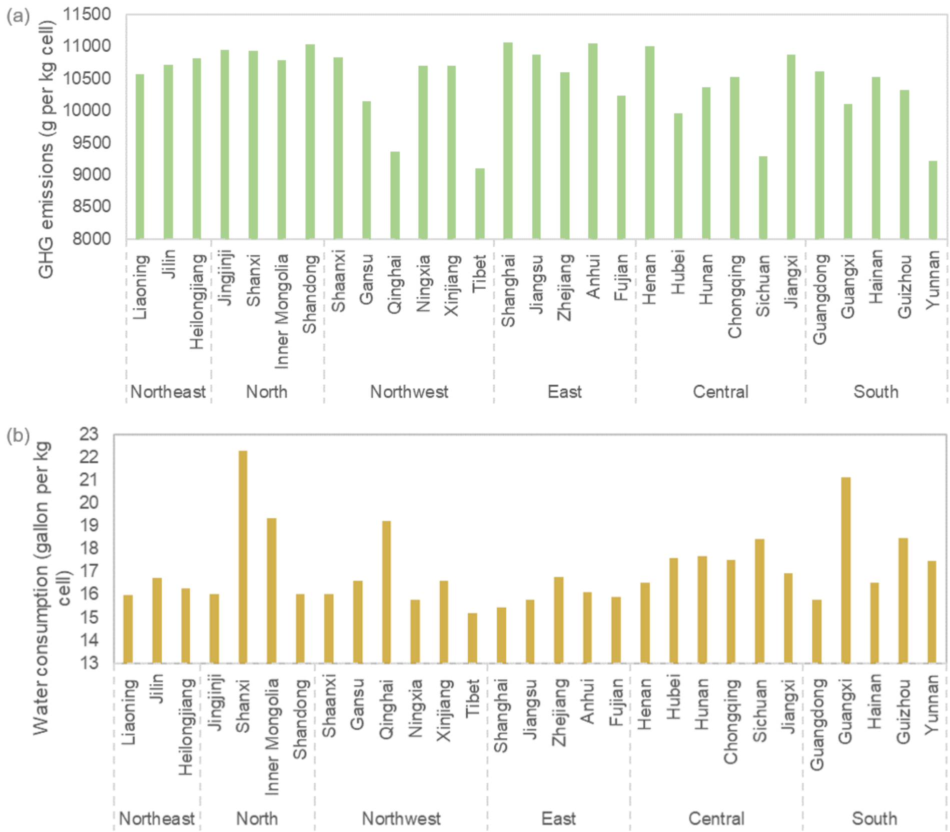

Fig 7. GHG emissions and water consumption in battery life cycle based on the provincial electricity mix. NCM_{111} cells using hydrometallurgical recycling method are assessed.

Provincial GHG emissions of the batteries' life cycle differ significantly as shown in Fig. 7. According to the range of carbon emissions, we divide these regions into three it into three equal parts on average: low-emission provinces (9096-9753g CO2e / kg battery), including Qinghai, Tibet, Sichuan and Yunnan; medium-emission areas (9753-10409g CO2e / kg battery), including Gansu, Fujian, Hubei, Hunan, Guangxi, Guizhou; and high-emission regions (10409-11065g CO2e / kg battery), including all the others. In the low-emission provinces, the thermoelectricity only accounts for 10% to 17% and renewable energy has a high proportion of electricity generation. Their electricity structure determines that they have lower carbon emissions, because renewable energy we considered herein are low carbon electricity. In the high-emission regions, they generate more than 69% electricity by fossil fuel combustion, which cause the higher GHG emissions because coal-based electricity has higher GHG emissions intensity. In the high-emission regions, especially in the northern and northeastern China, the electricity power structure needs to be improved and the development of green electricity should be emphasized.

The results of the water consumption of the battery life cycle are shown in Fig 7, which indicates that the difference of water consumption are statistically significant. Shanxi has the highest water consumption, while Tibet has the lowest. However, because we lack the water consumption of hydropower and thermoelectricity in Tibet, we replace it with the national data. Therefore, in the following analysis, we will not discuss the water consumption in Tibet. Excluding Tibet, Water consumption of battery life cycle also differ by province. Shanghai has the least water consumption, 32% lower than that in Shanxi. It is noted that Shanghai has the highest GHG emission of the battery life cycle, but it consumes the least water in this process. The main reasons are as follows: (1) Except thermoelectricity, hydroelectricity accounts for the most proportion while the percentage of non-hydro renewable power and nuclear power is less than 11%. Therefore, the major of the low carbon electricity generated in the low-emission regions is hydroelectricity. (2) The water consumption factor of hydroelectricity are higher than thermoelectricity, the average of hydroelectricity is about 18 times that of thermoelectricity. Therefore, it is reasonable that Shanghai, which has the highest proportion of thermoelectricity, has the least water consumption in the battery life cycle.

Based on the water consumption, we also divided all regions into three parts in order to compare their GHG emissions, and analyze the impact of power structure on carbon emissions and water consumption. The results of classification are as follows: the first is the high water consumption areas (20.01-22.29 gallon / kg battery), which are Shanxi and Guangxi; the second is the medium water consumption regions (17.72-20.01 gallon / kg battery), which are Inner Mongolia, Qinghai, Sichuan and Guizhou (17.72 – 20.01 gallon / kg). ; the other regions are classified as the low water consumption areas. The major of high-emission area, which have the relatively high proportion of coal-based electricity, are classified as the low water consumption areas. However, Shanxi and Inner Mongolia are two exceptions mainly because of the excessive water consumption factor of hydroelectricity in these two provinces, approximately 46 times than the national average data. These are two typical water-scarce provinces having relatively high water consumption of hydropower because of their local climate and surface conditions. Although 82% of electricity is hydropower in Yunnan, it still belong to the low water consumption areas because its water consumption factor of hydroelectricity is just a quarter of the national average data and just slightly higher than the water consumption factor of thermoelectricity. Therefore, although the hydroelectricity is a kind of low carbon electricity, its development should take into account regional water volume, climate and geographic conditions. Other green electricity, such as wind and solar power, should be considered in water-scarce areas and in some areas where the water consumption factor of hydroelectricity is high. Coastal nuclear power plant consumes relatively little water like the wind and solar power, while inland nuclear power plants consume more water than thermal power, so it is also not appropriate to consider it in water-deficient areas.

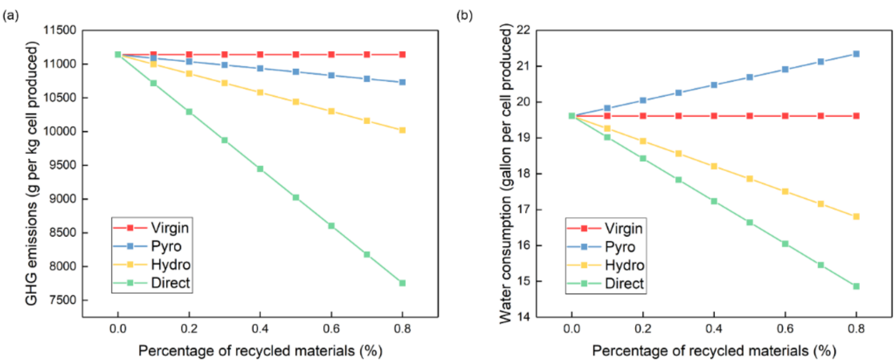

Fig 8. GHG emissions and water consumption of life cycle batteries when the proportion of recycled materials changes. All batteries are NCM_{111} cells.

3.3 Breakeven cost

The results of the cost reduction via three recycling processes are shown in Table 5, which indicates that all recycling processes could reduce the cost. Specifically, direct physical recycling method provides the highest economic benefits, reducing the cost by at least 12%, followed by hydrometallurgical and pyrometallurgical recycling processes. As shown in Fig. 11 and Table S11, material cost accounts for the largest proportion, about 81%, while labor costs account for only 1%.

Table 5. Cost reduction (%).

	Pyro	Hydro	Direct
NCM_{111}	3.81%	7.34%	14.90%
NCM_{622}	1.97%	5.36%	12.97%
NCM_{811}	1.60%	5.02%	12.57%
NCA	8.79%	11.73%	18.10%
LFP			14.67%

Fig 9 illustrates that direct physical recycling method could tolerate the highest purchase price of spent batteries at breakeven point. Taking NCM_{811} battery as an example, the maximum affordable purchase price of spent batteries using the direct physical method is 3 times and 2 times than using the pyrometallurgical and hydrometallurgical process, respectively. Because direct physical method does not require too much recycling process and chemical treatment. NCM_{111} can afford the highest purchase price of spent batteries because precious metals play a substantial role in battery component and those metals could bring a higher recovery value. For NCM cells, although the battery cost declines as the cobalt content decreases, the window for economic feasibility will be narrowed when the batteries are recycled.

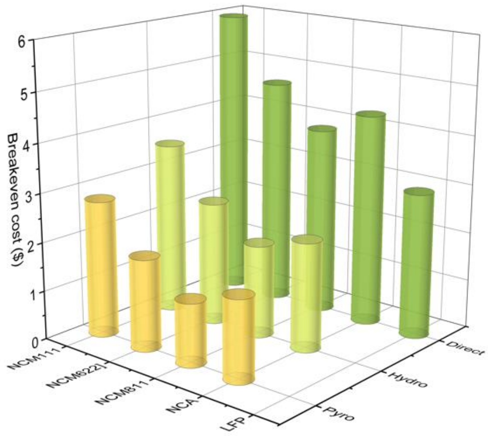

Fig 9. Purchase price of spent batteries at breakeven point.

As shown in Fig 10, with the increase of throughput, the battery cost shows a downward trend, with a large decrease from 1000 to 5000, and then the decrease slows down. Therefore, we suggest that battery manufacturers produce batteries on a relatively large scale to ensure that the manufacturing cost of batteries is within a reasonable and relatively low range.

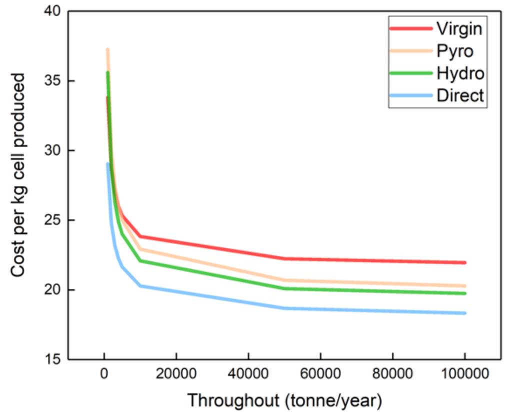

Fig 10. NCM_{111} battery cost varies with throughput. (unit:$)

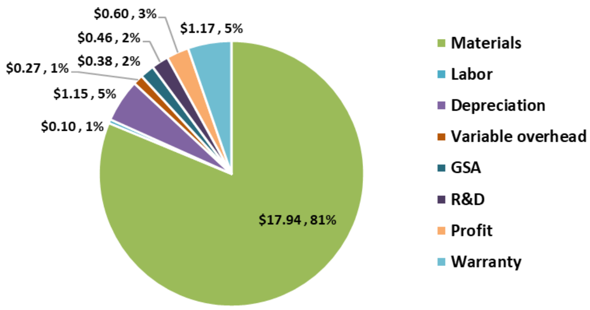

Fig 11. Cell manufacturing cost breakdown. The recycling process is hydrometallurgical recycling method and the battery is NCM_{111} cell. "GSA" represents general, sales and administration. R&D means research and development.

4 Discussion and conclusion

In a closed-loop supply chain system, this study quantifies the energy consumption, GHG emissions and water consumption of LIBs with different recycling methods throughout the life cycle. Excluding LFP cells, direct physical recycling process has the GHG emission and water consumption reduction potential as well as the economic benefits over the pyrometallurgical or hydrometallurgical methods. For LFP batteries, although direct recovery method could decrease the GHG emission and broad the window of the economic viability, it potentially increases the water requirement. The direct recycling method yields greater value final products after recycling [30] and generates the higher environmental benefit compared to the other two recycling methods, but it requires higher quality of spent batteries with relatively high state-of-charge [43] because it recycles materials with still serviceable morphology [8]. Therefore, the detection that determine the battery classification and feasibility of direct recovery method plays a substantial role in the recycling process of LIBs.

In addition, the hot spot analysis is conducted to identify the emission-intensive and water-intensive processes as well as major cost components. Carbon emissions and water consumption in manufacturing and recycling processes generated in the application of materials account for considerable proportions. Therefore, one of the future directions of technology development should be to apply environmentally friendly materials to substitute those materials with negative environmental impact. Direct physical recycling method could tolerate the highest purchase price of spent batteries at breakeven point. In recent years, extend producer responsibility recycling mechanism is developed and implemented in China and this mechanism could ensure the recovery of batteries to a certain extent and ensure the quantity of recovery, and potentially help to form a reasonable price of spent batteries.

However, some contradictions will occur between environmental and economic targets. Although LFP cells could decrease the environmental impacts compared to the other types of batteries, the value of the raw materials that make up the cathode is lower, reducing the potential window of economic feasibility when recycling. In order to reduce manufacturing costs and make electric automobiles more competitive in price, battery manufacturers incline to develop high-Ni and low-Co batteries, but significant nickel content will lead to a greater water burden when they are recycled by pyrometallurgical or hydrometallurgical or direct physical methods.

Moreover, there also have some contradictions between GHG emissions and water consumption reduction objectives. This mismatch potentially occurs in the selection of battery type or electricity mix structure in various provinces. Compared with NCM_{622}, NCM_{811} has more advantages in terms of emission reduction potential, but it simultaneously results in a greater water burden. Taking hydrometallurgical method of NCM_{111} batteries as an example, in China, the major of high-emission provinces, which has the relatively high proportion of thermoelectricity, are classified as the low water consumption areas. This is because that the bulk of non-thermoelectricity electricity used in China is hydropower, which is a water-intensive electricity generation method. However, water consumption coefficients vary greatly in different provinces, depending on the local climate and surface conditions. In some provinces with relatively low water coefficient of hydroelectricity, like Yunnan, hydropower is still a favorable way to reduce GHG emission. Nevertheless, in water-deficient provinces, which generally have a high water consumption of hydropower due to its environmental conditions, promotion of hydroelectricity is not encouraged. Other alternatives, such as wind, solar power and coastal nuclear power, are appropriate for these water-stress areas, because they could bring co-benefits of carbon emissions and water consumption reductions. Notably, those clean energies are the impure public goods and a price premium would be paid when consuming them [44]. Although comprehensive consideration of carbon emissions, water consumption and economic benefits is an arduous task, the government, cell manufacturers and battery recycling enterprises should carefully layout and fully consider those environmental and economic benefits in order to meet the environmental challenges brought about by the booming development of electric vehicle market and LIB recycling market.

There have some certain limitations in this work. Some studies point out that process-based LCA applied in this study have cutoff errors because it overlooks many upstream processes and is affected by system boundary truncation [45-48]. Thus, an integrated hybrid LCA, which integrates the economic input-output system and the process-based LCA, to reduce the uncertainty of the results is recommended for future studies [49]. Furthermore, Ji et al. [50] reveal that replacing the conventional automobiles with the electric vehicles transfers the GHG emissions from city (exhaust pipes) to predominant countryside (electricity power plant), due to the power source of automobiles is provided by electricity instead of fossil fuels. Therefore, evaluating the transferring of other negative environmental impacts, such as water consumption, of using LIBs is also an interesting and worth exploring issue.

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