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The Environmental Burdens of Lead-Acid Batteries in China: Insights from an Integrated Material Flow Analysis and Life Cycle Assessment of Lead

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Received: 17 October 2017; Accepted: 22 November 2017; Published: 26 November 2017

Abstract: Lead-acid batteries (LABs), a widely used energy storage equipment in cars and electric vehicles, are becoming serious problems due to their high environmental impact. In this study, an integrated method, combining material flow analysis with life cycle assessment, was developed to analyze the environmental emissions and burdens of lead in LABs. The environmental burdens from other materials in LABs were not included. The results indicated that the amount of primary lead used in LABs accounted for 77% of the total lead production in 2014 in China. The amount of discharged lead into the environment was 8.54 × 10⁵ tonnes, which was mainly from raw material extraction (57.2%). The largest environmental burden was from the raw materials extraction and processing, which accounted for 81.7% of the total environmental burdens. The environmental burdens of the environmental toxicity potential, human toxicity potential-cancer, human toxicity potential-non-cancer, water footprint and land use accounted for more than 90% at this stage. Moreover, the environmental burdens from primary lead was much more serious than regenerated lead. On the basis of the results, main practical measures and policies were proposed to reduce the lead emissions and environmental burdens of LABs in China, namely establishing an effective LABs recycling system, enlarging the market share of the legal regenerated lead, regulating the production of regenerated lead, and avoiding the long-distance transportation of the waste LABs.

Keywords: lead-acid battery; material flow analysis; life cycle assessment; primary lead; regenerated lead

1. Introduction

Lead-acid batteries (LABs), an important kind of energy storage equipment, are widely used because they are low-cost, reliable, easily available, and suitable for a wide range of current discharge and temperature conditions [1,2]. LABs are becoming indispensable in transportation, communications, electric power, navigation, aviation and other fields [3]. In China, the production of LABs increased rapidly from 120 million KVAh in 2009 to 210 million KVAh in 2015 [4]. Nevertheless, because of the high toxicity of lead, the environmental pollution, especially the lead emissions from the LABs, is growing serious concern with the development of the LAB industry [5–8]. The life cycle toxicity potential of LABs is mainly from lead, especially the total lead emissions to the environment



[9]. In 2011, there were more than 60 reported incidents involving lead pollution, 24 of which were caused by lead smelting and battery plants in mainland China [10]. It is important to reduce lead emissions and lessen the environmental burdens through implement of practical and efficient policies in China.

Material flow analysis (MFA) and life cycle assessment (LCA), as systematic assessment methods [11-13], have been applied to analyze the lead material flow and to evaluate the environmental impacts of LABs, respectively. Several studies introduced the lead flows in different aspects [14-18]. Mao built the lead flow diagram of lead acid battery system and analyzed the relationship between a lead-acid battery system and its exterior environment [17]. Guo analyzed the lead stocks and flows in China using MFA [15]. In addition, a framework for applying MFA in a typical lead smelting system was presented. Meanwhile, a material flow chart model of a single process and the entire system were developed [14]. Meanwhile, some of papers focused on comparing the environmental impacts of LABs with the other kinds of batteries [19–27]. The environmental impacts of different type of batteries for electrochemical storage systems were investigated which indicated that LABs had a relatively higher environmental impact [22]. It also found that LABs had higher environmental burdens compared to the other batteries in terms of cumulative energy demand (CED) and global warming potential (GWP) [24,25]. Moreover, Rydh compared vanadium redox with LABs for stationary application by accounting for five environmental impact categories and indicated that LABs had similarly significant environmental burdens [26]. Several LCA studies for LABs aimed to distinguish the serious environmental impact categories and their stages. Liu undertook a LCA method for LABs used in e-bikes by accounting for twelve environmental impact categories and claimed that 95% of total lead emissions were released at the end-of-life stage [9]. According to the study of Davidson, lead production is the dominant contributor to environmental impacts in LABs and lead sheet [28]. Zhang also evaluated the life cycle environmental impacts of LABs in terms of global warming, acidification and respiratory effect, which showed that the proportion of lead in the LABs was 73.26% of the total mass and the environmental burdens were mainly caused by the consumption of lead resources [3].

However, to the best of our knowledge, there was still a significant lack of detailed LCA studies on lead, which is the worst pollutant in LABs. There are limited temporal and spatial data in existing lead material flow studies. There are also few comprehensive works which integrate MFA and LCA which was relevant to the efficient and practical reduction of the lead emissions and environmental burden of LABs.

In order to reduce the environmental burden of lead in LABs and bring some enlightenment on green development of the LAB industry in China, an integrated method of combining MFA with LCA was used to quantify the lead material flow and its life cycle environmental burdens, which covered 14 kinds of environmental impact categories. The key process or stages of the lead environmental emissions and its environmental impacts were quantitatively investigated with the collected data from the national level and the typical production companies in 2014 in China. As a result, several efficient and practical policies and measurements are proposed in order to reduce the lead emissions and their life cycle environmental burden.

2. Methodology

2.1. Material Flow Analysis (MFA)

MFA is a method based on the principle of the conservation of matter. It connects the sources, the pathways, and the intermediate and final sinks of a material [11,29]. In this study, the lead flow of LABs was confined to five stages: raw material extraction stage, production stage, use stage, end of life stage, and recycled or disposal of waste LABs. Primary lead and regenerated lead are the main raw materials for the production of LABs. Some LABs are imported or exported. Because of the limited data and little emitted lead at the use stage, the loss of lead during this stage was not taken into account. As for end of life stage, some LAB wastes, including lead slag and lead dust, are recycled for the production of regenerated lead and the others go to landfilling. The time margin was the year

2014. The data (including the imported and exported LABs, LAB waste, the total primary lead used in LABs, lead emissions in the process of LABs production) were provided by China Battery Industry Association (CUBA) and China Nonferrous Metals Industry Association Recycling Metal Branch (CMRA). The lead flow model is illustrated in Figure 1.



Figure 1. The model for the lead flows of LABs (note: solid lines represent input and output flows; dotted lines represent circular flows).

2.2. Life Cycle Assessment (LCA)

LCA is used to assess the overall environmental impacts of a product or service by including all direct and indirect emissions. The LCA study was conducted consistent with the ISO 14040 series, including four phases: goal and scope definition, life-cycle inventory (LCI), life-cycle impact assessment (LCIA), and interpretation [30,31].

2.2.1. Goal and Scope

Definition of the goal and scope is important as the primary phase of an LCA study [13]. Firstly, this study aimed to reduce the lead environmental burdens through analysis of lead flow in LABs. Secondly, it tried to provide the scientific and technical support for the production of LABs by using regenerated lead instead of primary lead.

A graphical representation of the life cycle process related lead of LABs is shown in Figure 2. The life cycle environmental burdens from lead in LABs were quantified, including raw material extraction, LABs production, transportation, use and recycling. The inputs included the raw materials, such as primary lead, regenerated lead and lead alloy. In the LABs production process, the positive and negative lead plates were manufactured, in which a part of the materials were recovered. At the stage of recovery, the waste lead (lead dust, lead paste) was recycled [32].

All the processes related to the lead in LABs were considered and the system boundaries are shown in Figure 3. The use stage of the batteries was not included in the investigation because in this stage, there was little lead environmental emissions. The life cycle stages included raw materials extraction and processing, manufacture, transportation, and recycling.



Figure 2. The life cycle process-related lead flow of LABs.



Figure 3. System boundary of the life cycle assessment for lead-acid batteries based on lead material flow.

Raw materials extraction and processing refers to the acquisition of primary lead and regenerated lead, as well as their environment burdens. Manufacture includes the manufacture of the lead plates and lead paste. The transportation stage only covered the transportation of LABs from manufacturers to the users. Recycling stage applied to the recovery of LABs at the end of their useful life, including reuse of batteries after refurbishment and lead containing materials recycling. According to the characteristics of LABs, 1 KVAh of electricity stored was defined as the functional unit in this study.

2.2.2. Data Source and LCI Analysis

The data of primary and regenerated lead production came from the investigation of the enterprises and companies in China. The Ecoinvent database was used as the secondary data source for the processes of producing the batteries components in the LCA model [33].

(1) Raw material extraction and processing

This stage is the upstream phase of the LAB manufacturing stage. In this stage, the environmental burdens from the primary and regenerated lead production were taken account,

which include ore mining, smelting, and the smelting of recycled lead used for the production of LABs. The inventory of the most of the materials was obtained from the typical LABs manufacturers in China, but the unavailable data of unit processes and the mean value of environmental burdens were taken from the Ecoinvent database.

(2) Manufacture

The manufacture stage is the process of the LABs production related to lead, mainly referring the manufacture of the battery lead components (such as lead paste and plate). The bills of materials of this stage were from the survey of typical enterprises in China, which included the input of raw materials (primary lead and regenerated lead), output of waste lead and the final products.

(3) Transportation

The transportation stage of lead was considered in this study in order to optimize the industrial layout of LABs. In previous studies, transportation distance was usually estimated [9]. In this paper, we fulfilled a part of the knowledge gap via a survey. The transportation of lead in battery (from manufactures to user) was separated as a single stage and other transportations (for materials and waste lead) were incorporated into their respective stages.

(4) End of life

Almost all of the LABs in China were recycled because of their high-value as recyclable items [9]. This indicated that 98% of all recovered waste LABs were used to produce regenerated lead through hydrometallurgical or pyro-metallurgical processes. In addition, a number of lead wastes (including lead slag, lead dust, etc.) were also recycled to produce regenerated lead. In this stage, the total amount of lead released into the environment came from the process of regenerated lead smelting. The data were obtained from typical enterprises in China. The lead emissions from the smelting process were managed as hazardous waste and the mean value of those environmental burdens data were from the Ecoinvent database. It was assumed that 1 KVAh capacity of LABs contained 15 kg lead [34]. Through the analysis of the collected data from the typical enterprises, the LCI analysis results of 1 t regenerated lead and 1 kWh capacity of LABs were shown in Tables 1 and 2, respectively.

In put or out put	Material	Mass/kg
Input	Waste LABs	1084.13
	Lead alloy	188.80
	Pure lead	124.31
Output	Regenerated lead	1000
	Lead sludge	0.35
	Lead smoke/lead dust	0.02
	Waste lead residue	2.20

Table 1. The input and output for 1 t of regenerated lead.

Table 2. The input and output of lead for 1 KVAh lead-acid batteries.

In put or out put	Material	Mass/kg
Input	Positive plate lead-regenerated	3.15
	Negative plate lead-regenerated	2.47
	Lead powder-primary	7.48
	Cast lead-regenerated	1.12
Output	Lead in waste water	4.64×10^{-6}
	Lead sludge	9.71× 10⁻³
	Lead smoke	6.17× 10 ⁻⁵
	Waste lead residue	2.97×10^{-1}
	Waste positive and negative plates	2.84×10^{-1}

Lead sludge (workshop)	9.19 × 10⁻₃
Lead dust	7.49×10^{-4}
Waste positive and negative plate lead	3.86×10^{-2}

As shown in the Table 1, the main material input of the regeneration lead production was waste LABs. The output of the waste lead mainly existed in waste residues due to the lead emissions in pyro-metallurgical process. In Table 2, lead powder was the greatest input in the production of LABs because of the lead plate production. Besides, in the output of wastes, the waste lead residue and waste positive and negative plates accounted for the largest proportion. In addition, transportation distance correlates positively with the energy consumption. Therefore, improving the utilization rate of raw materials, increasing the utilization of waste lead, and shortening the transportation distance can reduce the energy consumption and environmental emissions.

2.2.3. LCIA Impact Categories

In this study, we built three accounting models to quantify the environmental burdens. (1) Based on environmental factors, an accounting model was applied to 12 types of environment impacts, the basis of LCIA was shown in Table 3 [35–46]; (2) A model of the water footprint (WF) was applied in the WF accounting, including blue WF, green WF and grey WF [47,48]; (3) An accounting model of land use (LU) was used to calculate the environmental impact of LU, including land occupation and land conversion [49].

LCIA Impact Categories	Abbreviation	Unit	Reference [35]
Global warming potential	GWP	Kg CO2eq.	CML(100 year) from Forster et al. (2007)
Ozone depletion potential	ODP	Kg CFC-11eq.	CML from WMO(2003)
Respiratory inorganics	RI	Kg PM2.5eq.	Rispoll
Ionizing radiation	IR	Kg U235eq.	ReCiPe 1.08 Midpoint from Frischknecht et al. (2000)
Photochemical ozone creation	POCP	Ka Calling	CML from Jenkin & Hayman(1999) and
potential	FOCF	Kg C2H4eq.	Derwent et al. (1998)
Acidification potential	ΔP	Ka SOpea	CML baseline factors from Huijbregts
Actumention potential	AI	Kg 502eq.	(1999)
Eutrophication Potential-land	EP-land	Kg PO ₄ 3-	CML from Heijungs et al. (1992)
Eutrophication Potential-water	EP-water	Kg PO ₄ 3-	CML from Heijungs et al. (1992)
Abiotic resource depletion	ADP	Ka Shoa	CML from Guinee et al. (2002) and van
potential	ADI	Kg Sbeq.	Oers et al. (2002)
Environmental toxicity potential	ETP	CTUeco	USETox (Rosenbaum et al., 2008)
Human toxicity potential-cancer	HTP-CA	CTUh	USETox (Rosenbaum et al., 2008)
Human toxicity potential-non-	HTP NCA	CTUb	LISETox (Recordsum et al. 2008)
cancer	IIII-NCA	CIOII	00E10X (Noschbauiit et al., 2000)

Table 3. The environmental categories on the basis of environmental factors.

3. Results and Discussion

3.1. Lead Flow Analysis of Lead-Acid Batteries in Mainland China

As shown in Figure 4, the production of LABs increased dramatically from 2004 to 2014 in China [4] and the annual average increasing rate was approximately 15.9%, due to the increased use of electric vehicles. The production of LABs has declined since 2014 due to the application of new batteries, especially lithium ion batteries. By 2015, there were about 1800 LAB manufacturing enterprises and 240 regenerated lead enterprises in China respectively. Most of them were mainly located the Yangtze River the Pearl River deltas.

The LAB lead emission results are illustrated in Figure 5. It is shown that the lead contained in primary lead smelting that was used for LABs production was around 2.16×10^6 tonnes in 2014, and 3.38×10^6 tonnes lead were used in LABs through domestic production, of which 7.77×10^5 tonnes of lead was regenerated lead. The amount of lead in exported and imported LABs was 5.48×10^5 and 7.10×10^4 tonnes, respectively.



Figure 4. (a)The amount of the production of LABs from 1998 to 2015 in China; and (b) The production layout of LABs in China in 2015.



Figure 5. Lead flow of LABs in China in 2014 (×10⁴ t) (note: solid lines represent input and output flows; dotted lines represent circular flows).

The quantities of lead used in LABs was 2.63×10^6 tonnes and 1.71×10^6 tonnes of it went to the end of life stage. At the end of life stage, 1.54×10^6 tonnes of lead were recycled, the rest was disposed as the household waste. The amount of regenerated lead through the regeneration smelting process was 1.62×10^6 tonnes and part of them was reused in the production of LABs.

3.1.1. Raw material extraction stage

This stage included the production of primary lead and regenerated lead. In the process of primary lead production, lead concentrates are obtained through the mining and dressing process, then lead concentrates are smelted into primary lead. In 2014, the lead production in China was 4.22 \times 10⁶ tonnes, and 80% of them were used in the production of LABs, including 2.60 \times 10⁶ tonnes of primary lead and 7.77 \times 10⁵ tonnes of regenerated lead. During the process of lead ore mining and dressing, it was estimated that around 3.31 \times 10⁵ tonnes of lead went to the tailings if its efficiency was assumed as 84.7% [15]. This could be discharged into the environment due to their low lead content. In the smelting process, the amount of imported lead used for LABs was 9.28 \times 10⁵ tonnes

and smelting efficiency was 94.27% [15], so 1.58×10^5 tonnes of lead was discharged into the environment.

3.1.2. LABs Production stage

In the LABs production stage, 3.38×10^6 tonnes of lead was used. However, there were about 2.68×10^5 tonnes of lead slag produced. 27.10 tonnes of the lead dust and about 0.93 tonnes of lead released into water. Then, about 80% of the lead slag could be reused for regenerated lead production [50]. In addition, the amount of lead in exported and imported LABs was 5.48×10^5 and 7.10×10^4 tonnes, respectively. The exported was significantly more than the imported, which indicated that China is one of the main producing countries of LABs.

3.1.3. LABs use stage

The total amount of lead in the use stage was 2.63 × 10⁶ tonnes. Because the charge and discharge of the battery in the use stage only consumed electricity power, the lead emissions in this stage could be ignored.

3.1.4. End of life stage

Waste LABs were one of the fast growing waste streams in China [34]. In 2014, 9.18×10^5 tonnes of lead entered in-use stock and the amount of lead in waste LABs was about 1.71×10^6 tonnes. According to the investigation, 1.54×10^6 tonnes of lead from their use stage was recycled in 2014. The rest was discarded as the municipal solid wastes or regenerated lead in the next year.

3.1.5. Regenerated lead production stage

This stage was the production of regenerated lead from the waste LABs and the lead slag. As shown in Figure 5, the amount of waste LABs and the lead slag used in the regenerated lead production was about 1.54×10^6 and 2.14×10^5 tonnes in 2014, respectively. Because the recovery rate of regenerated lead was reported as 92% [34], about 1.67×10^6 tonnes of the regenerated lead was obtained while about 1.41×10^5 tonnes of lead released into the environment. Above all the total amount of lead discharged into the environment was about 8.54×10^5 tonnes. The proportion in the stages of raw material extraction, LABs production, end-of-life and regenerated lead production was 57.2%, 6.3%, 20.1% and 16.4% respectively.

3.2. The Environmental Burdens of LABs Based on Lead Material Flow

In order to control the lead pollution and reduce its environmental burdens, 14 kinds of environmental impacts of primary and regeneration lead were quantitatively evaluated through the life cycle of LABs on the basis of the lead material flow analysis.

3.2.1. Environmental Burdens of LABs

Based on the model shown in Table 3 and the data from the enterprises, the results of the environmental burdens of 1 KVAh capacity of LABs are shown in Table 4. For a better understanding of the effects in each impact category, all impact categories were scaled to 100%. As shown in Figure 6, each column represents the impact arising from the different stages.

It can be figured out that the life cycle environmental burdens of the raw materials extraction and processing stage was the largest, mainly resulted from the production of primary lead. It accounted for about 99.96% of LU, because the type of land used was transformed. The results showed that almost all of the life-cycle toxicity potential was from the raw materials extraction and processing stage due to the heavy lead toxicity.

The environmental burdens of recycling stage contributed the second largest environmental burdens due to the lead emissions in the smelting of regenerated lead, and its ODP accounted for about 29.5% of those from all stages.

Furthermore, the environmental burdens of the transportation stage ranked the third during the whole life of LABs. In term of ODP, POCP, and ADP, the environmental burdens were 14–18% in the life cycle of LABs. The main reason could be the consumption of gas, because the transportation of lead used in LABs was considered in this paper. Therefore, the transportation distance was also one of the concerns.

LCIA		Percentage by Stages				
Impact	Value	Materials Extraction and Processing	Manufacture	Transportation	Recycling	
GWP	3.78 Kg CO2eq.	80.33%	0.22%	7.18%	12.27%	
ODP	2.03 × 10 ⁻⁶ Kg CFC-11eq.	52.68%	0.54%	17.31%	29.47%	
RI	8.98 × 10 ⁻³ Kg PM2.5eq.	87.91%	0.13%	4.35%	7.61%	
IR	2.83 Kg U235eq.	92.74%	0.08%	2.65%	4.52%	
POCP	3.17 × 10 ⁻³ Kg C₂H₄eq.	58.94%	0.47%	15.02%	25.58%	
AP	3.45 × 10 ⁻² Kg SO₂eq.	81.10%	0.21%	6.90%	11.79%	
EP-land	2.01 × 10 ⁻³ Kg PO ₄ ³⁻	81.10%	0.21%	6.88%	11.80%	
EP-water	3.40 × 10 ⁻³ Kg PO ₄ ³⁻	71.42%	0.32%	10.44%	17.82%	
ADP	5.81 Kg Sbeq.	60.82%	0.44%	14.32%	24.41%	
ETP	9.78 CTUeco	96.40%	0.06%	1.31%	2.23%	
HTP-CA	1.68 × 10 ⁻⁸ CTUh	97.66%	0.04%	0.85%	1.45%	
HTP-NCA	1.11 × 10⁻⁵ CTUh	99.47%	0.01%	0.19%	0.33%	
WF	9.01 m ³	82.59%	0.00%	0.00%	17.41%	
LU	12.72 KgC	99.96%	0.02%	0.00%	0.01%	
Average		81.65%	0.20%	6.24%	11.91%	

Table 4. The life cycle burdens for 1 KVAh capacity of LABs.

As shown in Table 4, the environmental burdens of the manufacturing LABs stage were the least, with less than 1% of all the impact categories. The main reason was that almost all of waste lead generated in this stage was recycled.



Figure 6. Contribution of the life cycle stages of the lead in LABs packs to each of the impact categories.

3.2.2. Environmental Burdens of Primary and Regenerated Lead

Because lead is the major material for LABs, the regenerated lead is the main industrial product of the regeneration process of waste LABs and waste lead. The regenerated lead industry has made enormous contributions to reduce the environmental pollution and relieve the pressures of the resource shortage [51,52]. The environmental burdens of 14 categories of the primary lead and the regenerated lead used as the battery raw material are shown in Table 5 and Figure 7. The results show that the life cycle environmental burdens of the regenerated lead were much less than those of the primary lead, accounting for 5.52% of those from the primary lead on average, because it avoided the pollution from the lead-zinc ore smelting process.

LCIA Primary Lead		Regenerated Lead	Difference	T Les 1	
Impact	Value	Value	Difference	Unit	
GWP	6.77	5.99× 10 ⁻¹	6.17	Kg CO2eq.	
ODP	1.88×10^{-6}	7.74× 10 ⁻⁷	1.11×10^{-6}	Kg CFC-11eq.	
RI	1.82× 10 ⁻²	8.83×10-7	1.73× 10-2	Kg PM2.5eq.	
IR	6.16	1.65×10^{-1}	6.00	Kg U235eq.	
POCP	3.55× 10 ⁻³	1.05×10^{-3}	2.51× 10 ⁻³	Kg C ₂ H ₄ eq.	
AP	6.27× 10 ⁻²	5.26× 10 ⁻³	5.75× 10 ⁻²	Kg SO2eq.	
EP-land	3.66× 10 ⁻³	3.07×10^{-4}	3.35× 10 ⁻³	Kg PO ₄ ³⁻	
EP-water	5.13× 10 ⁻³	7.82×10^{-4}	4.35× 10 ⁻³	Kg PO4 ³⁻	
ADP	6.85	1.83	5.02	Kg Sbeq.	
ETP	22.5	2.82× 10 ⁻¹	22.2	CTUeco	
HTP-CA	3.92× 10 ⁻⁸	3.15× 10 ⁻¹⁰	3.89× 10 ⁻⁸	CTUh	
HTP-NCA	2.66× 10 ⁻⁵	4.75× 10 ⁻⁸	2.66× 10 ⁻⁵	CTUh	
WF	16.1	2.03	14.1	m ³	
LU	30.6	1.68× 10 ⁻³	30.6	kgC	

Table 5. The environmental burdens of primary and regenerated lead of 1 KVAh capacity in LABs.

The toxic potential resulted from lead emissions during the lead-zinc ore smelting process, so the environmental burdens of ETP, ATP-NCA, HTP-NCA from the regenerated lead accounted for about 1.3%, 0.2%, 0.8% of those from the primary lead (Figure 7). This could explain why LU from the regenerated lead accounted for only 0.005% of that from the primary lead. As for ODP, the environmental burdens of regenerated lead accounted for 41.2% of primary lead, mainly caused by the consumption of gas for transportation.



Figure 7. Comparative results of environmental burdens for primary lead and regenerated lead.

3.3. Pathways to Reduce the Environmental Burdens of LABs

3.3.1. Increase the Utilization Efficiency of Lead

In China, the utilization efficiency of lead wais lower than that in developed countries [16], which was about 80% in the raw material extraction stage [15]. Based on the above lead material flow analysis, if the utilization efficiency was enhanced up to 90% by improving primary ore mining

technology and using more advanced and environmentally friendly mining equipment, the amount of lead emissions into the environment would have been reduced by nearly 2.44 × 10⁵ tonnes in 2014.

In addition, it is important for manufacturers to improve the fabrication efficiency in LABs production. For example, in 2014, the LAB output was 2.21×10^8 KVAh. If all produced LABs in 2014 were in accordance with the level of the typical enterprise production technology, the amount of waste lead in the manufacturing stage was about 1.41×10^5 tonnes, which could be 47.3% less compared to the waste lead in the LAB industry in 2014 (2.68 × 10⁵ tonnes). The result indicated it is urgent to improve the average level of LABs production technology and lead utilization in China.

3.3.2. Increase the Recycling Rate of LABs

Because the lead contained in the waste LABs could seriously pollute water and soil [53], enhancing the recycling rate of LABs could reduce the environmental pollution and resource depletion. Until now, a nationwide recycling network for waste LABs recycling is still lacking in China [32], and the recycling rate of LABs was only about 25% [54]. Non-recovered waste LABs causes a large amount of heavy metals into the soil and underground water. In the United States, levying environmental taxes, prepaid recycling fees when buying a car and other ways are practical policies to fund the recycling and help regenerated lead manufacturers, so the waste lead recovery rate is nearly 100% in the United States [55]. Accordingly, recycling programs should be implemented, and more collection points and infrastructures should be built. The government should propose extending the producer responsibility of battery manufacturing enterprises with the obligation to recycle their products. Overall, reducing lead emissions requires the joint efforts of the policy makers, product manufacturers and active public participation.

3.3.3. Increase the Proportion of Regenerated Lead in the Production of LABs

Currently, the proportion of regenerated lead production in the total lead production in China is low, while the production of regenerated lead accounts for more than 60% of the total lead production in European and the United States [55,56]. Based on the investigation, the proportion of regenerated lead in raw material extraction and processing stage was 48%. This suggests that the contribution of the raw materials extraction and processing stage to the life cycle impacts was the largest and regenerated lead therefore becomes more competitive from a life cycle perspective.

In a previous study, it was estimated that the total lead oversupply stocks will be 3–5 times as big as the lead in-use stocks in 2030 [34], so it is essential to increase the proportion of regenerated lead as raw material in the production of LABs.

As shown in Table 6, if all primary lead was replaced instead by regenerated lead to produce LABs, it could reduce the environmental burden by 69.5% on average. In terms of the impact categories of ETP, HTP-CA, HTP-NCA and LU, the reduction of environmental burdens could be higher than 90%. Therefore, it is necessary to increase the proportion of regenerated lead in the LABs production process and to use advanced technology decrease the lead emission in the stage of raw material extraction.

LCIA Impact	Unit	Impact (48% Regenerated Lead)	Impact (100% Regenerated Lead)	Difference	Reduced Percentage
GWP	Kg CO2eq.	3.78	1.22	2.56	67.82%
ODP	Kg CFC-11eq.	2.03× 10-6	1.57× 10 ⁻⁶	4.59× 10-7	22.60%
RI	Kg PM2.5eq.	8.98× 10 ⁻³	1.78× 10 ⁻³	7.19× 10⁻³	80.14%
IR	Kg U235eq.	2.83	3.35× 10 ⁻¹	2.49	88.13%
POCP	Kg C2H4eq.	3.17× 10⁻₃	2.13× 10-3	1.04× 10-3	32.83%
AP	Kg SO2eq.	3.45× 10 ⁻²	1.07× 10 ⁻²	2.39× 10 ⁻²	69.07%
EP-land	Kg PO ₄ 3-	2.01× 10-3	6.24× 10-4	1.39× 10⁻³	69.06%
EP-water	Kg PO ₄ ³⁻	3.40× 10 ⁻³	1.5× 10 ⁻³	1.80×10^{-3}	53.23%
ADP	Kg Sbeq.	5.81	3.72	2.09	35.91%
ETP	CTUeco	9.78	5.7× 10 ⁻¹	9.21	94.13%
HTP-CA	CTUh	1.68×10^{-8}	6.41× 10 ⁻¹⁰	1.61× 10 ⁻⁸	96.18%

Table 6. The LCIA impacts of LABs in two different scenarios.

HTP-NCA	CTUh	1.11× 10 ⁻⁵	9.70× 10 ⁻⁸	1.10× 10 ⁻⁵	99.13%
WF	m ³	9.01	3.17	5.84	64.82%
LU	kgC	12.72	5.78× 10 ⁻³	12.71	99.95%

3.3.4. Regulating the Production of Regenerated Lead

In developed countries, like Japan and the USA, comprehensive laws have been enacted to reduce lead emissions. However, in China, the largest regenerated lead producer cannot collect enough waste LABs, because 60% of waste LABs are recycled by peddlers [57], and building a regular LAB recycling plant requires more money than nonstandard recycling plants. Many small plants use direct fired reverberatory furnaces, cupola and other outdated technologies, and some even use original smelting kilns with no environmental protection facilities [58], so a business-oriented, market-oriented and university-industry collaboration technological innovation system should be built in China. In 2013, Ministry of Environmental Protection (MEP) and Ministry of Industry and Information Technology (MIIT) launched the "Opinions on promoting standardized development of LABs and regenerated lead industry". Firstly, the government should accelerate industrial restructuring and eliminate the outdated and illegal plants. Secondly, the government should strengthen environmental inspection action in the regenerated lead industry. Moreover, the government should set regenerated lead industry standards combined with the country's overall requirements and industry's demands. Finally, a standardized recovery system for waste LABs should be established in the future.

3.3.5. Optimize LABs Industrial Layout and Reduce Transportation Distance

In this investigation, transportation distance is one of the key factors of environmental burdens in this stage, especially in the categories of ODP, POCP, ADP, EP-land and EP-water. In China, regenerated lead distribution and production areas basically occurs in some cities, with more than 80% scaled lead plants being mostly located in the middle and eastern regions [56], so waste LABs from other cities need to be transported for a relatively long distance to the designated destination. This causes more environmental burdens and economic cost. Thus, the government should optimize LABs industrial layout in China based on the development of the LAB industry in the future.

4. Conclusions

In this paper, we quantified the flow of lead in LABs in mainland China in 2014 using the material flow analysis (MFA) approach. The total lead discharged into the environment was 8.54 × 10⁵ tonnes. The proportions in the stages of raw material extraction, LABs production, end-of-life and regenerated lead production were 57.2%, 6.3%, 20.1% and 16.4%, respectively. The lead emissions from the raw material extraction stage accounted for the largest proportion in the lead flow of LABs in China in 2014, which was consistent with previous study results [16–18]. In addition, the qualitative LCA, which covering 14 impact categories of the primary lead and regenerated lead showed that the regenerated lead was more competitive, especially in the impacts categories of ETP, HTP-CA, HTP-NCA and LU. As for the environmental impacts of the lead in LABs, the life cycle environmental burden of the raw materials extraction and processing stage was the largest, with an average share as high as 81.7%, especially in the impact categories of ETP, HTP-CA, HTP-NCA and LU. The recycling stage was the second most important stage due to the large number of lead emissions during the smelting of regenerated lead.

Base on the results, Several pathways for further reducing the overall environmental impacts of LABs were provided and their efficiencies were quantitatively evaluated, such as increasing the utilization efficiency of lead and the recycling rate of LABs, increasing the proportion of regenerated lead as raw material in the production of LABs, encouraging the use of regenerated lead to substitute primary lead and comprehensively plan the location of LAB enterprises to shorten the transportation distances.

Acknowledgments: We would like to acknowledge the financial support of the National key research and development program (2017YFF0211801). We would also like to acknowledge all of the enterprises and experts for their assistance in interviews and data collection.

Author Contributions: Sha Chen and Zunwen Liu designed the conceptual approach and finalized the manuscript; Zhenyue Lian and Sumei Li performed the research and drafted the manuscript; Junbeum Kim provided feedback on the paper and underlying methodology; Yipei Li and Lei Cao collected the data from enterprises.

Conflicts of Interest: The authors declare no conflict of interest.

References

- 1. Martha, S.K.; Hariprakash, B.; Gaffoor, S.A.; Shukla, A.K. Performance characteristics of a gelled-electrolyte valve-regulated lead-acid battery. *Bull. Mater. Sci.* **2003**, *26*, 465–469.
- 2. Newnham, R.H. Advantages and disadvantages of valve-regulated, lead-acid-batteries. *J. Power Sources* **1994**, *52*, 149–153.
- Zhang, H.; Wang, H.M.; Ma, C.L.; Xu, Y. Life cycle assessment of lead-acid battery. Chin. J. Environ. Manag. 2013, 5, 39–48.
- 4. National Bureau of Statistics (NBS). *China Statistical Yearbook 2015;* China Statistics Press: Beijing, China, 2015.
- Chen, K.; Huang, L.; Yan, B.Z.; Li, H.B.; Sun, H.; Bi, J. Effect of Lead Pollution Control on Environmental and Childhood Blood Lead Level in Nantong, China: An Interventional Study. *Environ. Sci. Technol.* 2014, 48, 12930–12936.
- Wang, M.; Zhang, C.; Zhang, Z.; Li, F.S.; Guo, G.L. Distribution and integrated assessment of lead in an abandoned lead-acid battery site in Southwest China before redevelopment. *Ecotoxicol. Environ. Saf.* 2016, 128, 126–132.
- 7. Cheng, H.F.; Hu, Y.A. Lead (Pb) isotopic fingerprinting and its applications in lead pollution studies in China: A review. *Environ. Pollut.* **2010**, *158*, 1134–1146.
- He, K.M.; Wang, S.Q.; Zhang, J.L. Blood Lead Levels of Children and Its Trend in China. *Sci. Total Environ*. 2009, 407, 3986–3993.
- 9. Liu, W.; Sang, J.; Chen, L.; Tian, J.; Zhang, H.; Olvera Palma, G. Life cycle assessment of lead-acid batteries used in electric bicycles in China. *J. Clean. Prod.* **2015**, *108*, 1149–1156.
- 10. Wang, D. Develop circular economy for heavy metal pollution prevention and control. *Renew. Resour. Circ. Econ.* **2012**, *5*, 1–3.
- 11. Brunner, P.H.; Rechberger, H. *Practical Handbook of Material Flow Analysis*; Lewis Publishers: Boca Raton, FL, USA, 2004.
- 12. Zhang, L.; Yuan, Z.W.; Bi, J. Substance flow analysis (SFA): A critical review. *China Acta Ecol. Sin.* **2009**, *29*, 6189–6198.
- 13. Ling-Chin, J.; Heidrich, O.; Roskilly, A.P. Life cycle assessment (LCA)—From analysing methodology development to introducing an LCA framework for marine photovoltaic (PV) systems. *Renew. Sustain. Energy Rev.* **2016**, *59*, 352–378.
- 14. Bai, L.; Qiao, Q.; Li, Y.P.; Xie, M.H.; Wan, S.; Zhong, Q.D. Substance flow analysis of production process: A case study of a lead smelting process. *J. Clean. Prod.* **2015**, *104*, 502–512.
- 15. Guo, X.Y.; Zhong, J.Y.; Song, Y.; Tian, Q.H. Lead flow analysis in mainland China. *J. Beijing Univ. Technol.* **2009**, *35*, 1554–1561.
- 16. Liang, J.; Mao, J.S. A dynamic analysis of environmental losses from anthropogenic lead flow and their accumulation in China. *Trans. Nonferr. Met. Soc.* **2014**, *24*, 1125–1133.
- 17. Mao, J.S.; Lu, Z.W.; Yang, Z.F. Lead Flow Analysis for Lead-Acid Battery System. *China J. Environ. Sci.* 2006, 27, 3442–3447.
- 18. Mao, J.S.; Yang, Z.F.; Lu, Z.W. Industrial flow of lead in China. Trans. Nonferr. Met. Soc. 2007, 17, 400-411.
- 19. Matheys, J.; Timmermans, J.M.; Van Mierlo, J.; Meyer, S.; Van den Bossche, P. Comparison of the environmental impact of five electric vehicle battery technologies using LCA. *Int. J. Sustain. Manuf.* **2009**, *1*, 318–329.
- Sullivan, J.L.; Gaines, L. Status of life cycle inventories for batteries. *Energy Convers. Manag.* 2012, 58, 134–148.

- 21. Van den Bossche, P.; Vergels, F.; Van Mierlo, J.; Matheys, J.; Van Autenboer, W. SUBAT: An assessment of sustainable battery technology. *J. Power Sources* **2006**, *162*, 913–919.
- 22. Oliveira, L.; Messagie, M.; Mertens, J.; Laget, H.; Coosemans, T.; Van Mierlo, J. Environmental performance of electricity storage systems for grid applications, a life cycle approach. *Energy Convers Manag.* **2015**, *101*, 326–335.
- 23. Premrudee, K.; Jantima, U.; Kittinan, A.; Naruetep, L.; Kittiwan, K.; Sudkla, B. Life cycle assessment of lead acid battery: Case study for Thailand. *Environ. Prot. Eng.* **2013**, *39*, 101–114.
- 24. Hiremath, M.; Derendorf, K.; Vogt, T. Comparative life cycle assessment of battery storage systems for stationary applications. *Environ. Sci. Technol.* **2015**, *49*, 4825–4833.
- Spanos, C.; Turney, D.E.; Fthenakis, V. Life-cycle analysis of flow-assisted nickel zinc-, manganese dioxide-, and valve-regulated lead-acid batteries designed for demand-charge reduction. *Renew. Sustain. Energy Rev.* 2015, 43, 478–494.
- 26. Rydh, C.J. Environmental assessment of vanadium redox and lead-acid batteries for stationary energy storage. *J. Power Sources* **1999**, *80*, 21–29.
- 27. Unterreiner, L.; Jülch, V.; Reith, S. Recycling of Battery Technologies—Ecological Impact Analysis Using Life Cycle Assessment (LCA). *Energy Procedia* **2016**, *99*, 229–234.
- 28. Davidson, A.J.; Binks, S.P.; Gediga, J. Lead industry life cycle studies: Environmental impact and life cycle assessment of lead battery and architectural sheet production. *Int. J. Life Cycle Assess.* **2016**, *21*, 1624–1636.
- 29. Hendriks, C.; Obernosterer, R.; Muller, D.; Kytzia, S.; Baccini, P.; Brunner, P.H. Material flow analysis: A tool to support environmental policy decision making, Case-studies on the city of Vienna and the Swiss lowlands. *Local Environ*. 2000, *5*, 238–311.
- 30. International Organization for Standardization (ISO). *Environmental Management-Life Cycle Assessment Requirements and Guidelines;* ISO Copyright Office: Geneva, Switzerland, 2006.
- 31. International Organization for Standardization (ISO). *Environmental Management-Life Cycle Assessment Principles and Framework;* ISO Copyright Office: Geneva, Switzerland, 2006.
- 32. Zhang, Z.J.; Li, J.L. Management of waste lead acid batteries to collect the best possible model to explore. *China Renew. Resour.* **2013**, *2*, 67–69.
- 33. Ecoinvent. Ecoinvent Data v2.2. Swiss Center for Life Cycle Inventories; Ecoinvent: Zurich, Switzerland, 2012.
- 34. Liu, W.; Chen, L.; Tian, J. Uncovering the Evolution of Lead In-Use Stocks in Lead-Acid Batteries and the Impact on Future Lead Metabolism in China. *Environ. Sci. Technol.* **2016**, *50*, 5412–5419.
- 35. Building Research Association of New Zealand (BRANZ). New Zealand Whole Building Whole of Life Framework: Life Cycle Assessment-Based Indicators; BRANZ: Judgeford, New Zealand, 2014.
- 36. Huang, X.H.; Fang, X.W.; Chen, J.X.; Gu, X.J. Review of product environmental. *China J. Mech. Electr. Eng.* **2014**, *31*, 1554–1561.
- 37. Lawrence, P. Technology-transfer funds and the Law—Recent amendments to the Monteeal protocol on substances that deplete the ozone-layer. *J. Environ. Law* **1992**, *4*, 15–27.
- 38. Ferreira, J.G.; Andersen, J.H.; Borja, A.; Bricker, S.B.; Camp, J.; da Silva, M.C.; Garces, E.; Heiskanen, A.S.; Humborg, C.; Ignatiades, L.; et al. Overview of eutrophication indicators to assess environmental status within the European Marine Strategy Framework Directive. *Estuar. Coast. Shelf Sci.* 2011, 93, 117–131.
- 39. Hunter, K.A.; Liss, P.S.; Surapipith, V.; Dentener, F.; Duce, R.; Kanakidou, M.; Kubilay, N.; Mahowald, N.; Okin, G.; Sarin, M.; et al. Impacts of anthropogenic SO*x*, NO*x* and NH₃ on acidification of coastal waters and shipping lanes. *Geophys. Res. Lett.* **2011**, *38*, 142–154.
- 40. Intergovernmental Panel on Climate Change (IPCC). Contribution of Working Group III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. In *Climate Change 2014: Mitigation of Climate Change*; Cambridge University Press: Cambridge, UK, 2014.
- 41. Jensen, N.R.; Putaud, J.P.; Borowiak, A. *Literature Review on ODS (Ozone Depleting Substances) Measurement Methods and Data*; Publications Office of the European Union: Ispra, Italy, 2015.
- 42. Joint Research Centre (JRC). *ILCD Handbook: Framework and Requirements for LCIA Models and Indicators;* Publications Office of the European Union: Luxembourg, 2010.
- 43. Morales-Mora, M.A.; Rodriguez-Perez, B.; Martinez-Delgadillo, S.A.; Rosa-Dominguez, E.; Nolasco-Hipolito, C. Human and ecotoxicological impacts assessment from the Mexican oil industry in the Coatzacoalcos region, as revealed by the USEtox (TM) model. *Environ. Sci. Pollut. Res.* **2014**, *21*, 9819–9831.
- 44. Rosenbaum, R.K.; Huijbregts, M.A.J.; Henderson, A.D.; Margni, M.; McKone, T.E.; van de Meent, D.; Hauschild, M.Z.; Shaked, S.; Li, D.S.; Gold, L.S.; et al. USEtox human exposure and toxicity factors for

comparative assessment of toxic emissions in life cycle analysis: Sensitivity to key chemical properties. *Int. J. Life Cycle Assess.* **2011**, *16*, 710–727.

- 45. Seppala, J.; Posch, M.; Johansson, M.; Hettelingh, J.P. Country-dependent characterisation factors for acidification and terrestrial eutrophication based on accumulated exceedance as an impact category indicator. *Int. J. Life Cycle Assess.* **2006**, *11*, 403–416.
- 46. Van Zelm, R.; Huijbregts, M.A.J.; Van Jaarsveld, H.A.; Reinds, G.J.; De Zwart, D.; Struijs, J.; Van de Meent, D. Time horizon dependent characterization factors for acidification in life-cycle assessment based on forest plant species occurrence in Europe. *Environ. Sci. Technol.* 2007, *41*, 922–927.
- 47. Lovarelli, D.; Bacenetti, J.; Fiala, M. Water Footprint of crop productions: A review. *Sci. Total Environ.* **2016**, 548, 236–251.
- 48. Xu, C.C.; Huang, J.; Ridoutt, B.G.; Liu, J.J.; Chen, F. Calculation method and case analysis of product water footprint based on life cycle assessment. *China J. Nat. Resour.* **2013**, *28*, 873–880.
- 49. Canals, L.M.I. *LCA Methodology and Modelling Considerations for Vegetable Production and Consumption;* Centre for Environmental Strategy, University of Surrey: Surrey, UK, 2007.
- 50. Mao, J.S.; Cao, J.; Graedel, T.E. Losses to the environment from the multilevel cycle of anthropogenic lead. *Environ. Pollut.* **2009**, 157, 2670–2677.
- 51. Tian, X.; Gong, Y.; Wu, Y.; Agyeiwaa, A.; Zuo, T. Management of used lead acid battery in China: Secondary lead industry progress, policies and problems. *Resour. Conserv. Recycl.* **2014**, 93, 75–84.
- 52. Hu, G.F. Ministry of Industry and Information Technology Encourages Secondary Lead Enterprise Mergers and Acquisitions and There Is a Lot of Room for Industrial Upgrading. Available online: http://finance.eastday.com/stock/m3/20120321/u1a6438439.html (accessed on 21 March 2012).
- 53. Haefliger, P.; Mathieu-Nolf, M.; Lociciro, S.; Ndiaye, C.; Coly, M.; Diouf, A.; Faye, A.L.; Sow, A.; Tempowski, J.; Pronczuk, J.; et al. Mass Lead Intoxication from Informal Used Lead-Acid Battery Recycling in Dakar, Senegal. *Environ. Health Perspect.* **2009**, *117*, 1535–1540.
- 54. An, S.Y. Present development situation and recovery of lead-acid battery. *J. Heilongjiang Sci. Technol.* **2012**, *13*, 83–83.
- 55. Xiao, X.K. Development of American secondary lead industry. Enterp. Technol. Dev. 2012, 12, 18–20.
- 56. China Nonferrous Metals Industry Association (CNIA). *China Nonferrous Metals Industry Yearbook;* China Industry Press: Beijing, China, 2014.
- 57. Ge, R.J.; Jiang, G. Most the material of secondary lead comes from illegal recycling. *China Econ. Inf.* **2011**, *3*, 287–288.
- 58. Raghupathy, L.; Chaturvedi, A. Secondary resources and recycling in developing economies. *Sci. Total Environ.* **2013**, *461–462*, 830–834.



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