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Towards a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes

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Abstract:	In spite of their success, recycling of lithium-ion batteries (LIB) is still in an early stage. This is reflected by the limited amount of publications on the potential environmental benefits of LIB recycling. These focus on evaluating individual recycling processes for a single cell type or a generic mix of waste batteries, and little information is available about the difference between individual LIB chemistries in terms of recyclability and corresponding environmental benefits. Based on a review of existing LCA studies on LIB recycling, we develop parametrized models of different recycling processes for their application to different cell chemistries, complemented by primary data obtained from a recycling company. These are used for assessing recycling pathways of different cell chemistries, including beyond-lithium batteries like sodium-ion (SIB). Depending on the cell chemistry, recycling can reduce the potential environmental impacts of battery production significantly. Hghest benefit is obtained via advanced hydrometallurgical treatment for NCM- and NCA-type batteries. Especially under resource depletion aspects, recycling of these cells can reduce their impact to an extent that even leads to a lower "net impact" than that of cells made from majorly abundant materials like LFP or SIB, which show a more favourable performance when disregarding recycling. For these cells, recycling can even cause additional environmental impacts. Thus, maximum material recovery might not always be favourable under environmental aspects

	and that especially for the final hydrometallurgical treatment, the process would need to be adapted to the specific cell chemistry, if maximum environmental benefit wants to be obtained.

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Title: Towards a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes

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Keywords: LCA, LIB, environmental impact, battery recycling, , industrial ecology, literature review

Abstract:
In spite of the growing amounts of lithium-ion batteries (LIB) being brought into the market, their recycling is still in a very early stage. This is reflected by the limited amount of publications available on the potential environmental impacts and benefits of LIB recycling. Existing studies and life cycle assessments (LCA) of battery recycling primarily focus on evaluating or comparing individual recycling processes for a single cell type or a generic mix of waste batteries. Since the influence of varying cell chemistries on the recycling process is usually not considered, little information is available about the difference between individual LIB chemistries in terms of recyclability and corresponding environmental benefits.
Based on a review of existing LCA studies on LIB recycling, we develop parametrized models of three different recycling processes for their application to different cell chemistries, including beyond-lithium batteries like sodium-ion (SIB). The models are complemented by primary data obtained from a recycling company and are used for quantifying the potential reduction of environmental impacts that can be achieved by the recycling of different cell chemistries. Depending on the cell chemistry, recycling can reduce the potential environmental impacts of battery production significantly. The highest benefit is obtained via advanced hydrometallurgical treatment for NCM- and NCA-type batteries, mainly due to the recovery of cobalt and nickel. Especially under resource depletion aspects, recycling of these cells can reduce their impact to an extent that even leads to a lower "net impact" than that of cells made from majorly abundant and cheap materials like LFP or the SIB, which show a more favourable performance when recycling is disregarded. For these cells, recycling does not necessarily provide benefits, but can rather cause additional environmental impacts. This indicates that maximum material recovery might not always be favourable under environmental aspects and that especially for the final hydrometallurgical treatment, the process would need to be adapted to the specific cell chemistry, if maximum environmental benefit wants to be obtained.

1. INTRODUCTION

The rapidly growing demand for batteries in automotive, stationary and mobile applications leads to increasing amounts of returned waste batteries expected in future. Lithium-ion batteries (LIB) are the currently dominating and fastest developing battery technology (disregarding lead-acid for automotive starter batteries) (Pillot, 2017). However, concerns associated with resource availability and environmental impacts of LIB production represent a potential limiting factor for their future deployment (Vaalma, Buchholz, Weil, & Passerini, 2018; Weil, Ziemann, & Peters, 2018). For dealing with the expected waste battery streams and for minimising environmental impacts associated with LIB production and the corresponding potential resource limitations, recycling of waste batteries is fundamental. On the other hand, recycling of LIBs is complex and associated with significant inputs of energy and / or chemicals, raising the question about its actual environmental net benefits. Numerous life cycle assessment (LCA) studies on manufacturing and use phase of LIBs have been carried out, but their end-of-life (EoL) phase is often disregarded (J. F. Peters, Baumann, Zimmermann, Braun, & Weil, 2017). Yet, EoL handling can be decisive for the environmental competitiveness of a certain battery type (Weber, Peters, Baumann, & Weil, 2018). Existing LCA studies that include recycling often use rough estimations such as an unspecified mixed battery waste stream or unspecific recycling processes. First-hand life cycle inventory (LCI) data is rarely disclosed, impeding an adaption of the underlying process models to different or novel cell chemistries. Moreover, the focus lies mainly on environmental effectiveness of recycling processes or the difference between varying processes and not on the comparison of different cell chemistries and therefore, often only one or two cell chemistries are assessed. This limits the meaningfulness of these studies, since a cell-specific assessment can be highly relevant for the choice of a certain battery type for a given application. Creating a cell-specific model for LIB recycling also enables the assessment of emerging or future battery types like e.g., sodium-ion or magnesium-ion batteries.

This study therefore presents a parametrized model of LIB recycling processes for the application to different battery compositions. Based on an overview of existing literature, existing inventory data are combined with first-hand data from company visits, developing cell-specific life cycle inventories. With these, the potential environmental impacts or benefits of recycling different types of LIB and of a sodium-ion battery (SIB) are calculated and analysed.

2. METHODS

2.1. Literature review

2.1.1. Methodology

In order to identify all published studies that include quantitative data on the environmental impact of LIB recycling processes, an extensive literature review is carried out. Google Scholar, university libraries (KIT and LiU) and Scopus are searched, using the search strings “LCA Li-Ion recycling”, “environmental impact Li-Ion recycling”, “environmental assessment Li-Ion recycling” and similar terms. Resulting studies are either complete LCAs of LIBs that quantify the impacts or benefits of the EoLphase explicitly or studies that focus specifically on recycling. Only studies that separately indicate the environmental impact of the recycling phase are considered. Studies related to products in which LIBs are embedded, such as electric vehicles, are only included if separate results are given for the battery or battery cell. Moreover, only

studies that specifically examine a recycling process as the EoL treatment are considered. If landfilling, incineration, or the simple transport to recycling facilities is regarded as EoL treatment, the respective study is excluded. Studies with too generic assumptions for the EoL phase are disregarded as well. The underlying recycling process is required to be specified and results for the specific type of treatment must be indicated.

2.1.2. Review Results

Key parameters of studies that fulfil these prerequisites are extracted and displayed in Table 1. Several studies provide results for a wide range of impact categories. However, for better overview and comparability, the impact categories shown are limited to the ones that are the most widely used within all studies (Cumulative Energy Demand (CED), Global Warming Potential (GWP), Abiotic Resource Depletion Potential (ADP), Acidification Potential (AP)). A total number of 27 studies that meet the defined criteria are identified, of which 16 in particular express the impact of recycling as their analysis goal. The remaining 11 consider the whole life cycle of LIBs and include recycling as part of the life cycle. However, there are a few examples of these full LCA studies that show a very detailed analysis of the included recycling stage (Jennifer B. Dunn, Gaines, Linda, Barnes, M., Sullivan, & Wang, 2014; Hawkins, Singh, Majeau-Bettez, & Strømman, 2013; Olofsson & Romare, 2013).

Table 1: Results of literature review, LCA studies that include quantified impact of LIB recycling. Negative values indicate env. benefit, positive values indicate env. burden.

Author and Year	Focus on Recycling	Impact Categories & LCIA method	FU	LCI Source (EoL)	Recycling process	Cell type	CED (MJ)	GWP (kg CO_2-Eq)	ADP (kg Sb-Eq)	AP (kg SO_2-Eq)	Comment
Boyden, Soo, & Doolan, 2016	✓	GWP, HTTP, TETP	1 t bat.	Fisher et al., 2006	hydromet.	mixed		1,320			
					pyromet.	mixed		681			
Buchert, Jenseit, Merz, & Schüler, 2011	✓	CED, GWP, ADP, AP, EP, POCP (CML)	1 t bat.	own (n.d.)	pyromet.	mixed	15,000	1,200	-0.21	-88	LiBRi-process
Buchert, Jenseit, Merz, & Schüler, 2011b	✓	CED, GWP, ADP, AP, EP, POCP (CML)	1 t bat.	own (n.d.)	hydromet.	NMC	-16,000	-1,000	-0.21	-92	LithoRec I-process
						LFP	-29,000	-1,700	-0.26	-32	
Buchert & Sutter, 2015	✓	CED, GWP, ADP, AP, EP, POCP (CML)	1 t bat.	own (n.d.)	hydromet.	NMC	-41,115	-1,835	-0.4	-69	LithoRec II-process
						LFP	-59,487	-2,638	-0.4	-42.8	
Buchert & Sutter, 2016.	✓		1 t bat.	own (n.d.)	hydromet.	NMC	-47,014	-2,747	-0.31	-67	LithoRec II-process (update
Buchert & Sutter, 2015a	✓	CED, GWP, ADP, AP, EP, POCP (CML)	1 t bat.	own (n.d.)	pyromet.	NMC	-58,089	-2,954	-0.4	-77	EcoBatRec-process
						LFP	-66,472	-3,219	-0.4	-49	
Buchert & Sutter, 2016a	✓	CED, GWP, ADP, AP, EP, POCP (CML)	1 t bat.	own (n.d.)	pyromet.	NMC	-55,089	-2,841	-0.3	-62	EcoBatRec-process (update
Ciez & Whitacre, 2019	✓	GWP (GREET)	1 kg cell	Own, Dunn et al., 2014, Grützke et al., 2015, Greet	hydromet.	NMC cylind.		-0.93			Monte Carlo Simulation, pr are medians
						NMC pouch		-0.32			
						NCA cylind.		-0.59			
						NCA pouch		-0.11			
						LFP cylind.		0.83			
						LFP pouch		1.6			
					pyromet.	NMC cyclind.		0.53			
						NMC pouch		1.65			
						NCA cyclind.		0.27			
						NCA pouch		1.09			
						LFP cylind.		1.07			
						LFP pouch		1.92			
					direct	NMC cyclind.		-0.53			
						NMC pouch		-1.33			
						NCA cyclind.		-0.27			
						NCA pouch		-1.07			
						LFP cylind.		1.63			
						LFP pouch		0.53			

Cusenza, Bobba, Ardente, Cellura, & Di Persio, 2019.		PEFCR, CED	1 battery pack (11.4 kWh, 175 kg (pack), 105 kg (cells))	Hischier et al., 2007, modified	pyromet.	LMO-NCM	-5,850	-360	-0.0127	-6.32 $molH^+$-Eq	
Deng, Ma, Li, Li, & Yuan, 2019		ReCiPe	1 km	Hawkins et al. 2013	hydromet.	NMC-SiNT		0.00151			No recovered material credi 6% improvement when cons recycling credits
Dewulf et al., 2010.	✓	CEENE	1 kg cathode mat. rec.	own	pyromet.	NMC	CEENE: -51% (incl. mineral resources)				387,4 instead of 795,4 MJ/k material from virgin materials
Dunn et al., 2014 (first version 2012)		CED, GWP		own	hydromet.	LMO	9,83 MJ/kg Li rec.	1,6 kg CO_2/kg Co rec.			
					direct	LMO, LCO	2,53 MJ/kg LiMnO4 rec.				
					pyromet.	LCO	10,45 MJ/kg Co rec.	1,6 kg CO_2/kg Co rec.			
					intermediate	LCO	4,64 MJ/kg Li_2CO_3 rec.				
Dunn, Gaines, Sullivan, & Wang, 2012	✓	CED, GWP (GREET)	1 kg $LiMn_2O_4$	Dunn et al., 2014	hydromet.	LMO	29				total energy consumption fo kg of bat. reduced by 39%, t producing 1 kg of bat. reduc
					intermediate	LMO	25				total energy consumption fo kg of bat. reduced by 40%, t producing 1 kg of bat. reduc
					direct	LMO	7				total energy consumption fo kg of bat. reduced by 57%, t producing 1 kg of bat. reduc
Dunn, Gaines, Kelly, James, & Gallagher, 2015	✓	GWP, AP (GREET)		Dunn et al., 2014	pyromet.	LCO (SS)		-60%		-99%	Percentage indicates impact compared to production of c from virgin material.
						LCO		-75%		-99%	
					intermediate	NMC (SS)		-20%		-67%	
						LMR-NCM (SS)		-42%		-52%	
						LCO (SS)		-91%		-100%	
						LCO		-54%		-95%	
						LFP (SS)		-18%		-2%	
						LMO (SS)		-11%		-26%	
					direct	NMC (SS)		-96%		-100%	
						LMR-NCM (SS)		-95%		-100%	
						LCO (SS)		-97%		-100%	
						LCO		-98%		-100%	
						LFP (SS)		-81%		-98%	
						LMO (SS)		-87%		-73%	

Ellingsen, Singh, & Strømman, 2016		GWP (ReCiPe)	vehicle lifetime 180,000 km; 24.4 kWh (battery for "medium car")	Dewulf et al., 2010	pyromet.	NMC		100			No recovered material credi Results for "medium car", st different car (and battery) si
Faria et al., 2014		ADP, AP, EP, GWP (CML)	24 kWh, 300 kg bat.pack	Hischier et al., 2007, Van den Bossche et al., 2006	hydromet.	LMO		389.1	2.4	5.7	No recovered material credi
Fisher et al., 2006	✓	ADP, GWP, ODP, HTP, ETP, AP, EP (CML)	621 t bat.	own (industry data)	hydromet., pyromet.	mixed					CO_2 savings between 198 kg CO_2/t of battery waste, with average 35% rec. eff.
Gaines, Sullivan, Burnham, & Belharouak, 2011		CED	1 mile, 75.9 kg	own (industry data, n.d.)		NCA	-30%				
Hawkins et al., 2013		ReCiPe	24 kWh (214kg NMC, 273 kg LFP)	own	dismantling, cryogenic shattering	NMC, LFP		193		1.44	No recovered material credi
Hendrickson et al., 2015	✓	CED	1 kg bat.	own, based on Cheret and Santen 2007	pyromet.	LMO, LPF	-6.5				
					pyromet.	NMC	-12.2				
Hao et al., 2017	✓	CED, GWP	1 traction battery	own (industry data)	hydromet.	NMC	-4,065.4	-1,150.3			
Li et al., 2013	✓	GWP (GREET)	1 kg Co	Dunn et al. 2014		LCO	9.3	3.9			
Messagie, Oliveira, Rangaraju, Forner, & Rivas, 2015		GWP, MD, HTTP, PMF (ReCiPe)	1 kWh	Hadjipaschalis et al., 2009	hydromet.	LFP		0.097			Given LCI source not tracea be based on Fisher et al. 200 methods and results can be f Oliveira et al., 2015.
					hydromet.	LMO		0.029			
Olofsson & Romare, 2013		CED, ADP, AP, EP, GWP, HTP, POF (EI99)	1 kg cell	Dunn et al., 2012	pyromet.	LFP	-40				
				Kosaraju, 2012	hydromet.	NMC	-13				
Raugei & Winfield, 2019		CED, GWP	1 kWh	own (n.d.)	hydromet.	LCP	-20	-5.4			
Sanfélix et al.		GWP, AP, POF, PMF (ReCiPe)	1 km		hydromet.	LFP		-0.00000179		-5.52E-10	Exact numbers calculated ba sector percentages.
Yazicioglu & Tytgat, 2011	✓	CED, GWP	1 Saft MP 176065 cell	own (n.d.)	pyromet.	LFP	-0.0727	-0.00657	-0.000025		
		CEENE	1 kg of cathode material		pyromet.	NMC	-51% (CExD; incl. mineral resources)				

Among the studies that use own inventory data for the considered recycling processes, only 7 disclose these data for verification and further use (Ciez & Whitacre, 2019; Dewulf et al., 2010; Dunn, Gaines, Linda, Barnes, M., Sullivan, & Wang, 2014; Fisher, Wallén, & Paul, 2006; Hao, Qiao, Liu, & Zhao, 2017; Hawkins, Singh, Majeau-Bettez, & Strømman, 2013; Hendrickson, Kavvada, Shah, Sathre, & D Scown, 2015). Dewulf et al. (2010) compare the cumulative exergy requirements (CEENE) of virgin material for LIB production with that of recycled material and base production data for recycled material on obtained information from several Umicore facilities. Dunn et al. (2014) conduct a material and energy flow analysis for four different recycling processes, using e.g. average energy values for industrial process steps. The LCI by Fisher et al. (2006) is based on industry data (Recupyl, Batrec Industrie AG) and also represents the basis for Hischier, Classen, Lehmann, & Scharnhorst (2007) whose work is used for the respective processes in the widely used Ecoinvent database (Wernet et al., 2016). Hao et al. (2017) refer to industry data not further specified, while Hawkins et al. (2013) model their own process (shredding and cryogenic treatment) with auxiliary processes from Ecoinvent. Hendrickson et al. (2015) base their dataset on the Umicore Patent No. US 7,169,206 B2 (2007), and (Ciez & Whitacre, 2019) re-use data from previous studies (Dunn, Gaines, Linda, Barnes, M., Sullivan, & Wang, 2014) and the GREET database. The remaining studies either use inventories from these studies or are based on own obtained industrial data, but do not disclose the inventories for further use.

As can be observed in Table 1, existing LCAs on LIB recycling use varying system boundaries and functional units (FU) like energy, driven distance, or recovered content, with either absolute or relative results. Moreover, the authors use different ways of quantifying the benefit, or generally the impact of recycling. Some indicate the total recycling impact, including process impacts and benefits of material recovery, while others do not account for credits of recovered material outputs and thus obtain environmental burdens for the recycling processes. Many of the studies concentrate on differences between different recycling processes but not on the influence of different cell chemistries on the results. At this point, the study by Ciez & Whitacre (2019) must be highlighted as they present results for three different cell chemistries (Lithium Nickel Manganese Cobalt Oxide (NMC), Lithium Nickel Cobalt Aluminum Oxide (NCA), Lithium Iron Phosphate (LFP)) each in pyrometallurgical, hydrometallurgical, and direct recycling.

The very heterogeneous approaches impede the comparison of results for common existing LIB chemistries such as NMC, NCA, LFP or LTO (Lithium Titanate Oxide) across studies. Despite the difficulties in comparing the results, a few studies that are based on the same FU and system boundaries and assess the same impact categories can be contrasted. NMC and LFP in hydrometallurgical and pyrometallurgical recycling are identified to be the most frequently assessed combination, mostly assessed on a battery mass basis. Table 2 shows the range of results for GWP and CED. It becomes obvious that, in spite of the identical FUs, the results still differ to a large extent.

Table 2: Range of results of comparable studies (referring to same FU and compatible system boundaries). Maximum and minimum impact reduction. Considered: (Buchert et al., 2011b; Buchert & Sutter, 2015a, 2015b, 2016b, 2016a; Ciez & Whitacre, 2019; Hendrickson et al., 2015; Messagie et al., 2015 (adapted from original FU 1kWh))

Impact category (FU=1kg)	Process	NMC max	NMC min	LFP max	LFP min

GWP (kg CO_2-eq/kg bat.)	hydromet.	-2.747	-0.32	-2.638	1.6
	pyromet.	-2.954	1.65	-3.219	1.92
CED (MJ/kg bat.)	hydromet.	-47.014	-16	-58.089	-12.2
	pyromet.	-58.089	-12.2	-66.472	-6.5

To analyse the different recycling results for different cell types more in detail, and especially with a comparable scope and base, a cell-specific recycling model is required, relating the recycling outputs to the various cell inputs. This would allow drawing more reliable conclusions on the environmental effectiveness of current and future battery recycling and the influence of different battery chemistries within.

2.2. LCA

2.2.1. LCA Framework

The system boundaries include cell production and EoL-phase. The FU is defined as 1 kWh of storage capacity provided by the battery, calculated on the basis of given energy densities (Wh/kg) of the different cell types (see Supporting Information - SI). Due to the specific focus on the EoL-phase, the use-phase is excluded, thus highlighting the potential of recycling for reducing the impacts associated with battery production. As a result, the potential for mitigating the production impact by proper EoL handling is obtained, independently of the possible use of the battery cell.

Four different cell types are compared: LIB cell types NCA (lithium nickel cobalt aluminum oxide), NMC (lithium nickel manganese cobalt oxide) and LFP (lithium iron phosphate), and the emerging technology Sodium-Ion battery (SIB). The layered oxides NCA and NMC offer high working voltages and thus high energy densities which makes them favourable for the use in electric powertrains. Yet, they contain scarce metals like cobalt and nickel that are also bound to high prices. LFP, although offering a lower energy density, represents a cheaper and safer alternative, requiring no critical materials (Woehrle, 2018). SIBs are chosen as an example for future cell technologies. Cell compositions are retrieved from Peters & Weil (2018) and Peters, Buchholz, Passerini, & Weil (2016) and provided in the SI.

ILCD midpoint is applied as impact assessment method, considering two impact categories: climate change (“GWP 100a”) and resource depletion (“resources - mineral, fossils and renewables”). Climate change is considered to be a highly relevant impact category due to the energy intensity of the production and recycling processes, whereas resource depletion is an obvious issue because of the scarce metals contained in the battery cells. The results for the remaining impact categories will not be further discussed but shown in tabular form in the SI. OpenLCA 1.7.4 in combination with underlying process data from Ecoinvent 3.4 (Wernet et al., 2016) is used as a software for the implementation and calculation of results.

2.2.2. Inventory

Current industrial recycling processes for LIB cells usually involve either pyrometallurgical (high temperature) or hydrometallurgical (chemical) separation methods for the contained metals. Therefore, the current state of the art of LIB recycling is represented by a pyrometallurgical process and a basic hydrometallurgical process based on secondary inventory

data from Fisher, Wallén, & Paul (2006). In order to consider future developments, first hand data for an advanced hydrometallurgical process is obtained from industry (Duesenfeld GmbH, 2018). Hence, three different recycling processes are considered:

i. current pyrometallurgical treatment
ii. current hydrometallurgical treatment
iii. advanced hydrometallurgical treatment

The datasets for (i) and (ii) in Fisher et al. (2006) also form the basis for respective processes in Ecoinvent (Hischier et al., 2007) but the implemented processes do not differentiate between different LIB cell types. Therefore, the provided inventory data is reviewed and adapted to distinct cell chemistries: The aggregated inventory for the *current pyrometallurgical process (i)* is based on LIB treatment by the company Batrec. The precise process flow is not disclosed, it is only known that the process involves a crushing step before "neutralization" and further "processing" (Fisher et al., 2006). Further details on the exact pyrometallurgical treatment are not revealed. A difference to hydrometallurgy is the loss of lithium to the slag during typical pyrometallurgical treatment. Moreover, components that can possibly be recycled in future, such as electrolyte, are burnt in the smelter (Rothermel, Winter, & Nowak, 2018). Aggregated process data for *current hydrometallurgy (ii)* originates from the Validat process by the company Recupyl, representing their recycling activities in 2004. In this process, waste batteries are first shredded under inert gas and then chemically treated. A more detailed process description can be found in Ekberg & Petranikova (2015). Resulting process outputs are the metal constituents contained in the cathode material (lithium salts and respective other metals) as well as separated parts of the cell housing (aluminum, copper, and plastic).

In addition to the product outputs of current hydrometallurgical processes, the *advanced hydrometallurgical process (iii)* from Duesenfeld GmbH includes the recovery of electrolyte and graphite at battery grade. A previous mechanical treatment comprises initial crushing, air classification and sieving. Off-gas is cleaned through condensing and an activated carbon filter. The subsequent hydrometallurgical treatment includes leaching, solvent extraction and precipitation. Inventory data is available separately for mechanical and hydrometallurgical treatment. The company emphasizes that the data provided for their hydrometallurgy does not represent a suitable treatment for LFP or SIB cells. In the modelled process, the hydrometallurgical step is nevertheless applied to all cell types and the effect on LFP and SIB is particularly analysed.

For every data set, the available inventory data is parametrized for cell-specific assessment. This is done based on recovery rates for different cell components that are retrieved from the given process data and the cell-chemistry specific compositions of the respective waste batteries. Recovered material from recycling is considered avoided primary material and thus gives environmental credits that are rested from the overall impact, assuming closed-loop recycling. Thus, the recovered product quality must comply with battery grade requirements and for each battery input, the recovered material is qualitatively equivalent to the virgin material needed to produce the respective battery cell type. Figure 1 shows the three process flows, indicating all considered recovered products. A detailed description of the parametrization approach,

underlying assumptions for recovered material, and resulting LCI tables for all three processes are included in the SI.

Figure 1

3. RESULTS

Production. As a basis for assessing the impacts of recycling, Figure 2 shows the impacts of production for each battery type broken down to cell components. NCA production causes the lowest GWP per 1 kWh capacity (75.50 CO_2-Eq), followed by NMC, LFP, and SIB. Here, a high cell energy density plays an important role, since less battery is required for providing a certain capacity. The cell manufacturing energy (electricity and heat) represents the largest part of the production GWPs for each cell type. This is important to note since recycling cannot retrospectively reduce GWP caused by the manufacturing energy demand. Cathode material is responsible for another significant share of the total GWP of NCA and NMC cells, while anode material makes up for the second highest GWP share for SIB production (the anode is made of hard carbon for this battery type, unlike graphite for the LIBs). Regarding the ADP, cathode material (especially when containing cobalt and nickel) and anode current collector (copper) influence the total impacts significantly which is why NCA and NMC production show the highest ADP (0.24 and 0.35 kg Sb-Eq/kWh). LFP and SIB rely on less critical materials, namely iron phosphate and sodium, and the SIB also avoids copper for the anode current collector (J. Peters et al., 2016).

Figure 2

Recycling. Figure 3 presents the impacts and benefits of the different battery recycling processes for the considered cell types. Impacts are caused majorly by the required process inputs and emissions (e.g., energy and chemicals, off-gases), while the benefits are obtained from the recovered materials, avoiding primary materials. Resting the process impact (positive value in the diagram) from the credit for recovered materials on the negative side gives the environmental benefit for all assessed cell chemistries in both impact categories.

Figure 3

The highest recycling credits are obtained for those cell components that also cause high impacts during primary production. For the considered cells, these materials are copper, nickel, cobalt, and, under GWP aspects, also aluminium. Lithium also plays a minor role. As a consequence, the regarded recycling processes offer the highest impact reduction potential for NCA and NMC cells, while especially for LFP cells the possible recycling benefit is significantly lower. The advanced hydrometallurgical process shows the highest benefit in all cases due to the additionally recovered graphite and electrolyte. However, also the process inputs and thus the corresponding impacts increase, especially under ADP aspects.

Net impact. Figure 4 shows the "net" impacts obtained for the different recycling processes and cell type i.e., the final impact after subtracting the recycling benefits from the production impacts. For the advanced hydrometallurgical recycling process, the benefit is further broken

down into mechanical and hydrometallurgical treatment. Under GWP aspects, the advanced hydrometallurgical recycling shows the best result in all cases, reducing the impacts of the batteries by between 12 and 25% (in comparison to no recycling). For ADP, the same tendency can be observed for the high-energy LIB NCA and NCM, while for the LFP cells and the SIB, the hydrometallurgical treatment does not obtain further benefits, but rather adds burden. The comparably high process inputs for this step and the low benefit from the recovered materials make this process unfavourable for recycling these cell chemistries.

Figure 4

Under GWP aspects, recycling generally shows a lower relative reduction potential than for ADP. This outcome is influenced by the high share that cell manufacturing contributes to the total GWP, which cannot be reduced by recycling. NCA and NMC cells show the lowest production impacts and thus also lowest net GWP. This is due to their higher energy densities compared to LFP and SIB; less cell mass needs to be produced for providing the same capacity and thus, less production energy is required. Pyrometallurgical recycling shows lower benefits and thus higher net impacts when compared to the current hydrometallurgical process, mainly due to the higher energy consumption of the high temperature processing and the loss of lithium in the slag. Regarding the LFP cell, pyrometallurgical treatment even adds GWP, requiring significant process inputs for recovering a comparable small share of the contained materials. The additional recovery of electrolyte and graphite contribute to the lower net impacts of the advanced hydrometallurgical process, in spite of the highest process inputs. Depending on the recycling process, relative impact reduction potentials for NCA and NMC cells range between 18% and 29% leading to net impacts between 69.58 and 61.00 kg CO_2-Eq (NCA) respectively 61.69 and 54.45 kg CO_2-Eq (NMC). While the GWP reduction potential of recycling for SIB constantly lies around 13%, it varies widely for LFP cells, with a maximum of 11%.

The cells' net ADP is highly influenced by their recyclability, and the results strongly depend on the specific cell chemistries. NCA and NMC cells benefit greatly from recycling, resulting in a significantly lower net impact. Overall, LFP and SIB score worse, in spite of lower initial ADP impacts from their production phase. In advanced hydrometallurgy, especially the recovery of electrolyte adds to a further reduced net impact. It becomes clear that hydrometallurgy (processing of the black mass) for LFP and SIB in the advanced process adds environmental burden. This underlines the company's previous statement that the hydrometallurgy represented in the dataset is not adapted to LFP or SIB type cells. The displayed net impact of LFP and SIB therefore only considers the benefits of mechanical treatment – and is significantly lower than with current hydrometallurgical recycling. Net impacts might still be reduced if adding an adapted, cell-specific hydrometallurgy with e.g., lower input of chemicals. However, possible benefits of additionally recovered cell components must be balanced with environmental impacts of the additional process efforts. Depending on the recycling method, the ADP of NCA cells is reduced by 61%-76% to a net impact between 0.0059 and 0.0094 kg Sb-Eq (NMC: 67%-77% reduction potential, 0.0080-0.0115 kg Sb-Eq net impact). In advanced hydrometallurgical recycling, LFP and SIB cells reach reduction potentials of 57% respectively 35%, reducing their ADP to 0.0083 (0.0088) kg Sb-Eq.

4. DISCUSSION

The results of the present cell-chemistry specific assessment indicate highest recycling benefits and lowest net impacts for NCM-type LIB under GWP aspects, while for ADP, NCA cells obtain the best results. Benefits for LFB and SIB cells are comparably low in both considered impact categories, and hydrometallurgical treatment of the black mass without a specifically tailored process potentially even increases environmental burdens. This is in line with the findings by Ciez & Whitacre (2019), who assess the GWP of NCA, NMC, and LFP pouch cells on a mass basis. In their study, NMC cells show the highest GPW reduction, while in the present work NCA cells perform slightly better. This can be attributed to varying assumptions regarding the cell compositions and different underlying inventory databases. In both studies, LFP cells show the least benefit (respectively the highest added burden) in both of the two assessed recycling processes. However, in their work pyrometallurgical recycling adds burden for all three cell chemistries, while in the present study it performs unfavourable only for LFP cells.

When comparing the results with those by Buchert et al.(2011b) and Buchert & Sutter (2015b, 2015a), who provide results for NMC and LFP type cells on a mass basis, it is noteworthy that they obtain a higher GWP reduction for the recycling of LFP than for NMC cells. A closer look, however, reveals that the difference results from the high benefit from the battery pack disassembling step (recovered steel and aluminium from the housing, which contributes a higher relative share for low energy density batteries) which is excluded in the present study focusing on single cells.

Dunn et al. (2015) also provide results for different recycling routes of different cathode material, and indicate a relative impact reduction potential of recycling for GWP. Their considered intermediate process (comparable to a mechanical-hydrometallurgical route) shows 20 % GWP reduction for NMC and 18 % for LFP recycling. The current hydrometallurgical process in the present study process offers roughly 22 % GWP reduction potential for NMCs and 3.5 % for LFPs. However, these results can hardly be compared since Dunn et al. (2015) only consider cathode material manufacturing and not cell production. Cell manufacturing energy, which makes up large parts of the cell production impact, is not taken into account. Moreover, the modelling of the production phase also plays a significant role for the outcome since the authors refer to the solid-state preparation technique of the cathode material.

In any case, it has to be pointed out that the general lack of detailed inventory data for the recycling processes in combination with a cell chemistry specific assessment makes a comparison with previous works difficult. This highlights the relevance of a transparent and parametrizable inventory for the different recycling processes as provided in the present work

5. CONCLUSION

This work provides a cell specific assessment of LIB and post-LIB cell recycling by means of different recycling processes based partly on original primary data. Two main findings can be pointed out in this regard:

First, recycling leads to a significant reduction of the environmental impact of LIB cells, especially for NMC and NCA, and is therefore crucial for a future LIB industry. Particularly when regarding the resource depletion potential of NMC and NCA cells, closed-loop recycling can reduce their impact to an extent that even leads to a lower “net impact” than that of LFP or

SIB, which show a more favourable performance in their production phase. However, the benefits highly depend on the cell composition and layout. The results are thus only valid for specific battery cells as modelled in this work. For other cell layouts, results may vary. The extent of the recycling benefit also depends on which impact category is considered. Benefits for ADP are much higher than those for GWP, since recycling can only reduce impacts from mining, not from cell manufacturing or active material synthesis. The latter are also major contributors to the GWP of LIB cells, and the potential for reducing GWP of LIB by recycling is therefore limited. However, recycling benefits for GWP are likely to increase in future with a growing share of green electricity used in the manufacturing process or, generally, less energy intensive LIB cell production.

Second, due to substantial differences between the compositions of battery cells, a cell chemistry specific approach for recycling is necessary. Recycling processes need to be tailored and adapted to different cell chemistries in order to obtain the best possible output quality and highest environmental benefit. It is thus of great importance to enable a reliable identification of cell chemistries, e.g. by labelling the cells, allowing recyclers to tailor the recycling processes cell chemistry specifically for minimum input of chemicals and process energy. Otherwise, processes cannot be optimized, and environmental impacts might increase as pointed out for hydrometallurgical treatment of LFP and SIB cells. In fact, current recycling activities focus on the recovery of the cathode active material components cobalt and nickel which offer high economic incentives. However, the development of new battery technologies is going towards minimizing the share of these metals or by using less scarce and environmentally critical materials such as e.g. phosphorus or sodium. Consequently, there might be a lack of economic incentives to develop advanced recycling methods for these cell chemistries. On the other hand, improved or more stringent legal regulations such as the European battery directive, might force the development of efficient recycling technologies also for these cell chemistries. It remains to be seen whether emerging future technologies that are based on cheaper and more abundant materials provide enough incentives to develop appropriate recycling technologies. Here, it is important to be aware of the fact that maximum recycling depth does not automatically mean maximum environmental benefit. Depending on cell chemistry and -composition, the best balance between recycling process impacts (or inputs and emissions, respectively) and benefits has to be found. Direct recycling that recovers cathode material maintaining its crystal structure is often mentioned as an option for low-cost cells such as LFP, offering significantly higher potential benefits. However, this method is not commercial yet and there are still quality concerns regarding the recovered cathode material (J. F. Peters, Baumann, & Weil, 2018).

When evaluating the named findings, some limitations must also be taken into account. The results are overall bound to uncertainties due to often weak or insufficiently modelled background data. For example, the used literature data for pyrometallurgical recycling contains several ambiguities since the given output data – unlike other known pyrometallurgical processes – still contains plastics and aluminium. Furthermore, a closed-loop recycling is assumed. If the recovered material did not meet the same quality criteria as virgin materials, the positive effects of recycling would decrease. Future research on more detailed process flows and cell-chemistry specific input of chemicals and energy for the regarded recycling processes would further help to improve the quality of the inventory data and thus the robustness of the results. This would allow for even better adaption to different cell chemistries and especially emerging battery

technologies. Finally, to ensure the recyclability of future low-cost cells, it would be helpful to pursue design for recycling or design for sustainability already in the earliest stage of technology development.

ACKNOWLEDGEMENTS
The authors would like to thank the Duesenfeld GmbH for their cooperation, time, fruitful discussion and effort.

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SUPPORTING INFORMATION
The supporting information provides a more detailed description of the modelling approach. Moreover, underlying data such as the energy densities of the assessed battery cells as well as cell compositions are displayed. Inventory tables and numerical results can be found in the additional Excel sheet (SI2).

Figure Legends

Figure 1: Process flows, including all considered product outputs. Inputs, waste and emissions not displayed. (Diekmann et al., 2017; Duesenfeld GmbH, 2018; Ekberg & Petranikova, 2015; Fisher et al., 2006)

Figure 2: GWP and ADP of cell production, component-wise

Figure 3: Component-wise share of GWP and ADP, including process efforts (“Input”)

Figure 4: Production and “net” impacts after subtracting recycling benefits

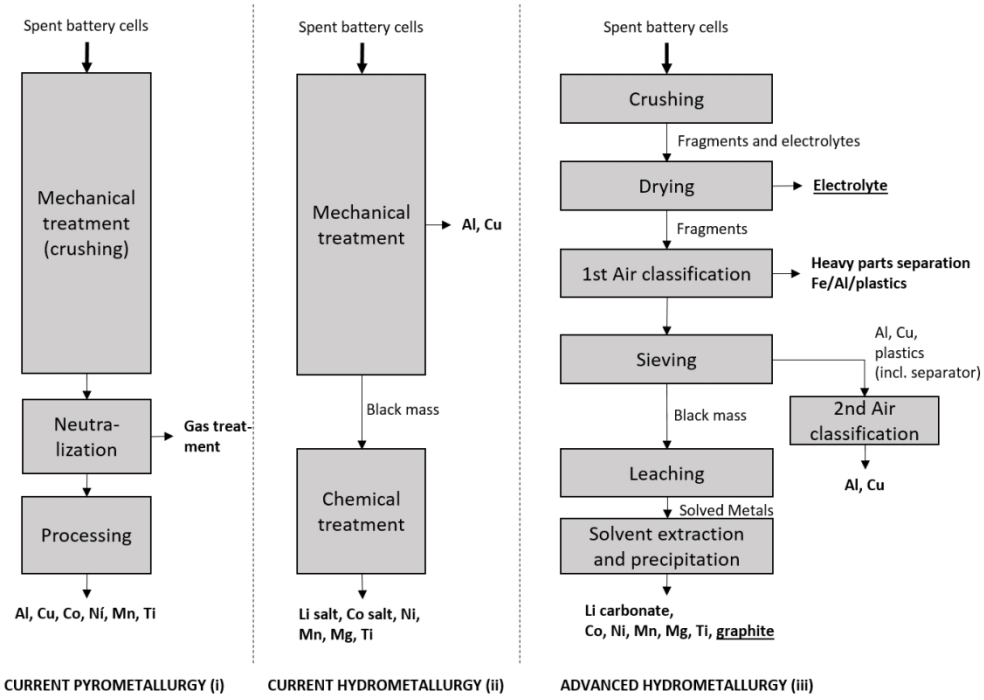

Figure 1. Process flows, including all considered product outputs. Inputs, waste and emissions not displayed. (Diekmann et al., 2017; Duesenfeld GmbH, 2018; Ekberg & Petranikova, 2015; Fisher et al., 2006)

233x163mm (150 x 150 DPI)

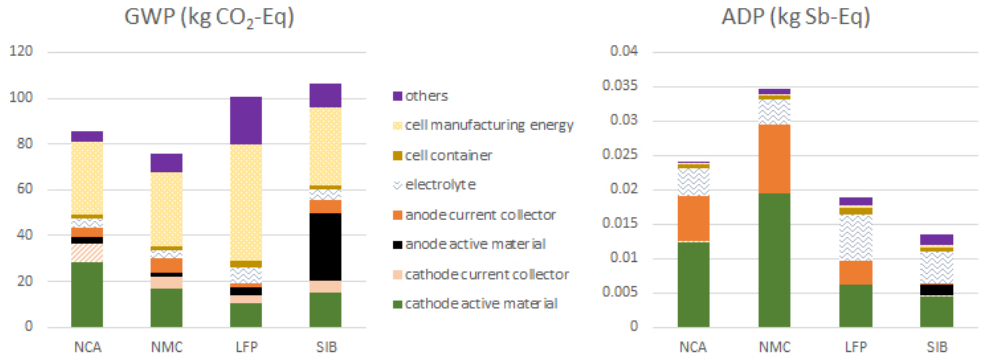

Figure 2. GWP and ADP of cell production, component-wise

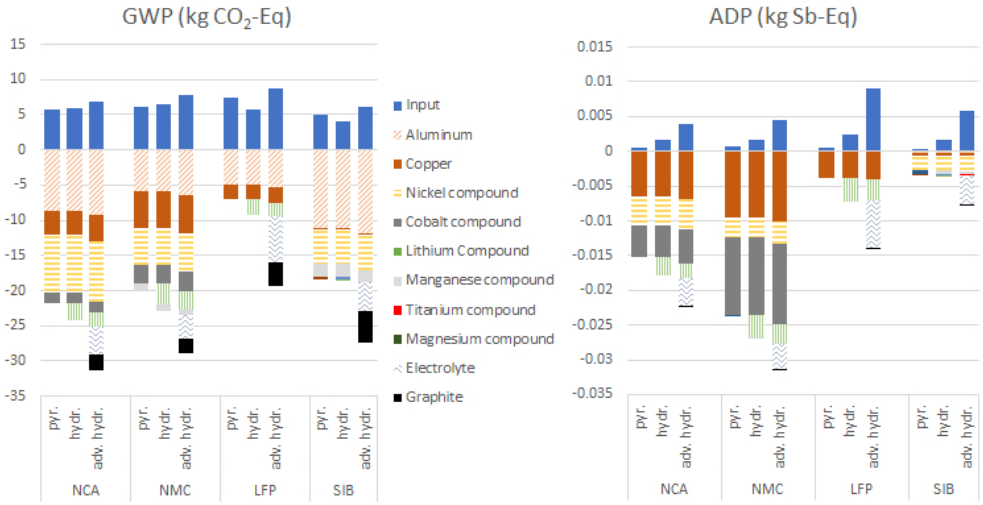

Figure 3. Impacts and benefits of the different battery recycling processes broken down to components, including process efforts ("Input")

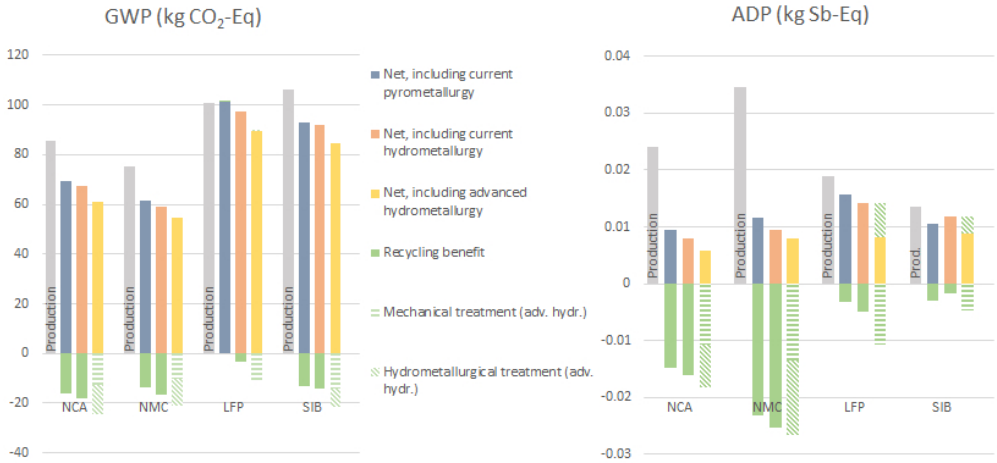

Figure 4. Production and "net" impacts after subtracting recycling benefits

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SUPPORTING INFORMATION FOR:

Mohr, M., Peters, J. F., Weil, M. & Baumann, M. (2019.) Article title: Towards a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes. *Journal of Industrial Ecology.*

Summary

The supporting information provides a more detailed description of the modelling approach. Moreover, underlying data such as the energy densities of the assessed battery cells as well as cell compositions are displayed. Inventory tables and numerical results can be found in the additional Excel sheet (SI2).

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1. Modelling Approach

Parametrized process models are created that can be fed with different battery chemistry inputs and calculate different product outputs regarding the varying inputs. Other process parameters that influence the environmental performance, such as the process energy and emissions, are difficult to allocate to the processing of a distinct battery type. Therefore, the process inputs are to a large extent independent of the cell chemistries.
To obtain parametrized data, it is proceeded as follows: First, a process-specific recovery rate is derived from the ratio of the original output amount to the assumed input amount. The input amount is based on the ratio of the corresponding material in a generic battery input. If the output product is a chemical compound of the respective input, their stoichiometric ratio is considered for the calculation of the recovery rate. In some cases, a recovery rate is directly provided in the dataset. Second, based on the recovery rate and the previously introduced underlying cell compositions, the cell specific process output is calculated as follows for every output component:

cell input [kg] × share of material in cell composition [%] × process-specific recovery rate [%] = material output [kg]

If the output material is a chemical compound of the contained material (e.g. lithium to lithium carbonate), the stoichiometric ratio is taken into account as well.
Following the existing Ecoinvent processes for LIB recycling, the recycling plant infrastructure for all processes involving hydrometallurgy is considered by including the proxy "chemical factory, organics" that is available in the database. Even if these process routes consist of a mechanical and a hydrometallurgical treatment, only the chemical plant is included, and the mechanical treatment infrastructure is not separately considered. This is done to ensure consistency with existing Ecoinvent dataset for hydrometallurgical treatment. Infrastructure thus might be underestimated. For the pyrometallurgical process, the infrastructure is represented by a "blister-copper conversion factory", again following the existing Ecoinvent datasets. Electricity consumption is modelled with a European market mix. The advanced hydrometallurgical process, however, as emphasized by the Duesenfeld GmbH, uses green electricity (Greenpeace energy). The Greenpeace standard energy mix consists of 72 % hydro power and 28 % wind power (Statista, 2018). A corresponding mix is simplified and created with available hydro and wind power datasets in Ecoinvent, using the sets "hydro, run-of-river" and "wind, 1-3 MW turbine, onshore".

The waste treatment flows for plastic and wastewater are included in the system and approximated by the Ecoinvent flows "market for waste plastic, mixture" and "market for wastewater, average". If not stated differently, the European standard electricity mix is assumed. For the implementation in openLCA, the datasets are modelled with the quantitative reference of 1 kg treated battery cells to provide datasets for broader applications. The results of the calculations are finally adjusted according to the above introduced energy densities to obtain results for the defined FU of 1 kWh.

2. Battery Compositions

Inventories for LIB cells are provided by Peters & Weil (2018) who analyze inventories for different LIB chemistries by several authors and build a common base for comparing these. This source is chosen since the battery models are especially built for the comparison of different cell types and were available as ready-built Ecoinvent datasets. For the defined chemistries, the authors' analysis of Bauer (2010) is the basis for the NCA inventory, of Ellingsen et al. (2014) for the NMC inventory and of Zackrisson, Avellán, & Orlenius (2010) for the LFP inventory. Bauer (2010) models the NCA as a $Li(Ni_{0.8}Co_{0.15}Al_{0.05)}O_2$ cell composition; Ellingsen et al. (2014) take a $Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ as a base for their LCI. The common base by Peters & Weil (2018) is built upon unifying assumptions concerning common components that do not alter the battery models significantly in order to provide a better base for comparing the active material compositions. These components are the manufacturing energy, the cell package, the electrolyte and the binder. The cell packaging is assumed to be a pouch cell type with inventory data taken from Ellingsen et al. (2014) since their housing inventory is considered the most comprehensive. The electrolyte is, by all underlying inventories, assumed to be lithium hexafluorophosphate ($LiPF_6$) in an organic solvent and only modelled with different approximations. The best approximation and therefore the base for the unified inventory is decided to be the electrolyte LCI data by Notter et al. (2010). Yet, the amount of electrolyte still differs for the different cell types. As for the binder, a water based anode binder based on Peters, Buchholz, Passerini, & Weil (2016) and an organic cathode binder based on Bauer (2010) are assumed (J. F. Peters & Weil, 2018).

Regarding the transfer to future cell technologies, it is decided to choose one example with an already existing, and available dataset. Peters et al. (2016) carried out an LCA on Sodium-Ion batteries (SIB) and provide the underlying inventory data as readily modelled Ecoinvent datasets. The modelled SIB contains layered oxide (NiMnMgTi) as cathode active material on an aluminum current collector and hard carbon as anode active material on an aluminum current collector. For consistency, the initially modelled cell container (nickel plated steel case for an 18650 cell) is replaced by the pouch cell container that is also used for the Li-Ion cell types.
Detailed cell compositions (metals shares and component-wise) are displayed below in Tables S1.1 and S1.2.
For the impact calculation on the basis of 1 kWh, the energy densities presented in Table S1.3 are considered. These energy densities are linked to the specific cell modifications of the respectively modelled cell types. Within one cell type, energy densities still vary, depending on the exact composition. Therefore, results are not generally valid for *all* NCA or *all* LFP type cells.

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Table S1.1: Mass composition of battery cell, metal requirements (Peters and Weil 2018; Weil et al. 2018; Peters et al. 2016)

Item	NCA (Bauer 2010)	NMC (Ellingsen et al. 2014)	LFP (Zackrisson et al. 2010)	SIB (Peters et al. 2016)
Li	2.1 %	2.6 %	2.0 %	
Al	7.4 %	4.6 %	1.9 %	9.1 %
Cu	14.6 %	21.7 %	4.7 %	
Ni	13.6 %	7.0 %		4.6 %
Co	2.5 %	7.0 %		
Mn		6.5 %		7.2 %
Fe			15.2 %	8.0 %
P			8.4 %	
Mg				0.3 %
Ti				0.7 %
Total	40. 2%	49.4 %	32.2 %	29.9 %

Table S1.2: Mass composition of battery cell, component-wise (Peters and Weil 2018; Peters et al. 2016)

Item	Parameter	NCA (Bauer 2010)	NMC (Ellingsen et al. 2014)	LFP (Zackrisson et al. 2010)	SIB (Peters et al. 2016)
Anode					
	Active material	19.34 %	15.64 %	17.59 %	35.42 %
	Current collector	14.67 %	21.59 %	4.79 %	6.10 %
	Binder	0.49 %	0.65 %	2.19 %	1.53 %
	Cond. carbon	0.58 %	0.00 %	0.00 %	1.15 %
Cathode					
	Active material	27.14 %	34.94 %	43.93 %	28.00 %
	Current collector	7.35 %	4.59 %	1.98 %	2.98 %
	Binder	0.49 %	1.49 %	2.91 %	1.19 %
	Cond. carbon	0.58 %	0.74 %	2.81 %	0.60 %
Separator		8.22 %	2.14 %	1.87 %	2.57 %
Electrolyte		18.34 %	15.54 %	19.26 %	17.80 %
Cell housing					
	Plastic	0.53 %	0.53 %	0.53 %	0.53 %
	Aluminum	1.12 %	1.12 %	1.12 %	1.12 %
	Copper	1.02 %	1.02 %	1.02 %	1.02 %
Total		99.87 %	100.00 %	100.00 %	97.33 %

Table S1.3: Cell energy densities (Wh/kg) (Peters and Weil 2018; Peters et al. 2016)

NCA (Bauer)	NMC (Ellingsen)	LFP (Zackrisson)	SIB (Peters)
173.42	169.86	108.08	162.48

3. Assumptions for Recovered Material

In the performed LCA, recovered material is considered avoided primary material and thus gives positive environmental credits to the overall impact. This approach, which is based on the crediting approach of several battery LCA studies of the *Ökoinstitut e.V.* (e.g. Buchert, Jenseit, Merz, & Schüler, 2011), is outlined in more detail in the following. A closed-loop recycling process is assumed. Based on this assumption, the recovered product quality must comply with the battery grade. That means, for each battery input, the recovered material is equivalent to the material needed to produce the respective battery cell type. The recycling processes mostly do not return pure metal fractions but metallic salts or other chemical compounds. As a simplification and due to missing alternative information, the output metal compounds of the recycling processes are considered to be the same type and quality as the needed corresponding production input for every considered cell chemistry. Potential further transformation requirements are not regarded. The resulting mass for each output is only adapted by taking into consideration the according stoichiometric ratio. Other elements like phosphorus or sodium are neglected in this approach. The current recycling processes do not aim at recovering them due to their abundance and environmental harmlessness.

The output stream with the mixed aluminum-copper fraction ("non-ferrous metals" in the process inventories below) needs further treatment to obtain separated re-usable copper and aluminum. Hischier, Classen, Lehmann, & Scharnhorst (2007) have approximated the subsequent refining in the Ecoinvent-datasets "treatment of non-Fe-Co-metals, from used Li-Ion battery, hydrometallurgical processing", and "treatment of non-Fe-Co-metals, from used Li-Ion battery, pyrometallurgical processing". Although these treatment processes are only roughly modelled and based on auxiliary processes, these datasets are utilized to adumbrate an approximate process effort for the recovery of aluminum and copper from the mixed fraction.

As for the resulting iron or steel fraction that is indicated in the previously shown process flows and in the original datasets in the following modelling approaches, it is assumed to result from a different cell design than the assumed pouch and from the fact that the datasets consider whole battery packs as process inputs. E.g. cylindrical cells' housing is made from steel (Rothermel, Winter, & Nowak, 2018). On cell level, of the analyzed cell chemistries, only the LFP cell contains iron with an iron content of 15.2 %. It is, nevertheless, decided to neglect iron as a recovered product due to underlying uncertainties in the modelling of the iron compounds in the LFP cell production. It is expected that iron recovery would not add relevant positive impacts, being an abundantly available material. I.e. iron is not considered as a product output or avoided primary material.
Table S1.4 shows the recycling process outputs and the respective metal compound that is counted as an avoided product for re-use in cell production.

The respective needed production input (e.g. cobalt for NCA production and cobalt sulfate for NMC production) is taken from the underlying datasets of the production phase by Peters & Weil (2018). The terms in the column "process outputs" represent

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generic terms for various resulting compounds of the corresponding metals since the different processes return varying chemical compounds. All metals that are contained in the regarded cell types and potentially recoverable are listed in this column, even if not found in the previously described process flows. Magnesium and titanium are contained in SIB cells and not yet targeted by existing recycling processes. However, to be able to hypothetically assess the treatment of SIB cells, these materials will be included in the process models that are described in the following.

Table S1.4: Process outputs and respective assumed avoided products for re-use in cell production

Process output (compound of...)	Assumed avoided material for cell production of respective processed cells			
	NCA	**NMC**	**LFP**	**SIB**
Cobalt	Co	$CoSO_4$	-	-
Nickel	Ni	$NiSO_4$	-	$NiCO_3$
Manganese	-	$MnSO_4$	-	MnO_2
Lithium carbonate	LiOH	LiOH	Li_2CO_3	-
Magnesium	-	-	-	$Mg(OH)_2$
Titanium	-	-	-	TiO_2
Copper	Cu	Cu	Cu	Cu
Aluminum	Al	Al	Al	Al
Iron	-	-	Not considered	-

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SUPPORTING INFORMATION FOR:

Mohr, M., Peters, J. F., Weil, M. & Baumann, M. (2019.) Article title: Towards chemistry specific life cycle assessment of lithium-ion battery recycling proces Journal of Industrial Ecology

This additional supporting information provides inventory tables for the three recycling processes, numerical results as underlying data for the plottet figures, and additional LCIA results.

Contents

a cell-
sses.

Tables S2.1, S2.2 and S2.3 show the resulting LCI tables for all three recycling processes.

Table S2.1: Current Pyrometallurgical Process (Fisher), process inventories per 1 kg treated cells

Item	Amount for respective cell input					
	NCA		NMC		LFP	
INPUTS						
Waste battery cel	1.00E+00					
NaOH	2.10E-01					
Electricity	8.00E-01					
Water	1.00E+00					
Infrastructure	5.00E-10					
OUTPUTS						
Product output						
Al+Cu to refining	2.26E-01		2.66E-01		8.18E-02	
Aluminum	7.93E-02		5.37E-02		2.86E-02	
Copper	1.46E-01		2.13E-01		5.32E-02	
Cobalt compound	2.35E-02	Co	1.73E-01	$CoSO_4$		
Nickel compound	1.27E-01	Ni	1.72E-01	$NiSO_4$		
Manganese compound			1.68E-01	$MnSO_4$		
Titanium compound						
Emissions to air						
SO_2	4.80E-05					
	1.00E-04					
dust	1.04E-05					
	9.36E-05					
Emissions to water						
SO_2	4.00E-02					
Cl	4.00E-02					
Water to sewer	1.00E+00					
Solid waste						
Plastic to refining	9.73E-02		4.81E-02		7.51E-02	

Table S2.2: Current hydrometallurgical Process (Fisher), process inventories per 1 kg treated cells

Item	Amount for respective cell input					
	NCA		NMC		LFP	
INPUTS						
Waste battery cel	1.00E+00					
Reagent	2.50E-02					
Electricity	1.40E-01					
Water	7.20E-01					
H2SO4	2.13E-01					
Lime	1.16E-01					
Infrastructure	4.00E-10					
OUTPUTS						
Product output						
Lithium compour	6.64E-02	LiOH	8.20E-02	LiOH	9.84E-02	Li_2CO_3
Al+Cu to refining	2.26E-01		2.67E-01		8.19E-02	

Aluminum	7.95E-02		5.38E-02		2.86E-02	
Copper	1.47E-01		2.13E-01		5.33E-02	
Cobalt compound	2.35E-02	Co	1.73E-01	$CoSO_4$		
Nickel compound	1.27E-01	Ni	1.72E-01	$NiSO_4$		
Manganese compound			1.68E-01	$MnSO_4$		
Magnesium compound						
Titanium compound						
Emissions to air						
SO_2			4.50E-06			
VOC			2.50E-06			
Emissions to water						
Solid suspension			1.20E-05			
Chemical oxygen			3.00E-05			
Total hydrocarbo			1.00E-08			
Cu			1.67E-05			
Co			1.67E-05			
Ni			1.67E-05			
Fluoride			3.00E-08			
Water to sewer			3.37E-01			
Solid waste						
Plastic to refining	9.73E-02		4.81E-02		7.51E-02	
Residue to landfil			2.02E-01			
Gypsum			3.39E-01			

Table S2.3: Advanced Hydrometallurgical Process (Duesenfeld), process inventories per 1 kg treated cells

Item	**Amount for respective cell input**					
	NCA		**NMC**		**LFP**	
INPUTS						
Waste battery cel			1.00E+00			
Inert gas			1.00E+00			
Infrastructure			5.00E-10			
Electricity	1.28E+00		1.37E+00		1.66E+00	
Activated carbon	6.12E-02		6.48E-02		7.67E-02	
Lime	7.24E-02		7.96E-02		1.03E-01	
Silica sand	7.24E-02		7.96E-02		1.03E-01	
H_2SO_4	7.24E-01		7.96E-01		1.03E+00	
Oxygen Liquid	8.69E-02		9.56E-02		1.24E-01	
NaOH 50% in H_2	1.74E-01		1.91E-01		2.48E-01	
Na_2CO_3	4.35E-01		4.78E-01		6.21E-01	
OUTPUTS						
Product output						
Al+Cu to refining	2.41E-01		2.85E-01		8.74E-02	
Aluminum	8.48E-02		5.74E-02		3.05E-02	
Copper	1.56E-01		2.27E-01		5.69E-02	
Cobalt compound	2.46E-02	Co	1.81E-01	$CoSO_4$		
Nickel compound	1.33E-01	Ni	1.80E-01	$NiSO_4$		
Manganese compound			1.44E-01	$MnSO_4$		
Titanium compound						

Lithium compour	5.68E-02	LiOH	7.01E-02	LiOH	8.41E-02	Li_2CO_3
Electrolyte	1.82E-01		1.55E-01		1.93E-01	
Graphite	1.93E-01		1.56E-01		1.76E-01	
Plastic to refining	9.73E-02		4.81E-02		7.51E-02	

			Unit	Ecoinvent dataset
SIB				
…..			kg	
…..			kg	sodium hydroxide, without water, in 50% solution state
…..			kWh	electricity, medium voltage
…..			l	Water, unspecified natural origin
…..			Items	blister-copper conversion facility
	1.05E-01		kg	non-Fe-Co-metals, from Li-ion battery, pyrometallurgical processing
	9.57E-02		kg	aluminium, wrought alloy
	9.36E-03		kg	copper
			kg	cobalt / Cobalt sulfate - GLO
	8.71E-02	$NiCO_3$	kg	nickel, 99.5% / nickel sulfate / nickel carbonate
	1.07E-01	MnO_2	kg	manganese sulfate / manganese dioxide
	1.09E-02	TiO_2	kg	titanium dioxide
…..			kg	Sulfur dioxide
…..			kg	Particulates, < 2.5 um
…..			kg	Particulates, > 10 um
…..			kg	Particulates, > 2.5, and > 10 um
…..			kg	Sulfur dioxide
…..			kg	Chloride
…..			kg	wastewater, average
	5.82E-02		kg	waste plastic, mixture

			Unit	Ecoinvent dataset
SIB				
…..			kg	
…..			kg	chemical, inorganic
…..			kWh	electricity, medium voltage
…..			l	Water, unspecified natural origin
…..			kg	sulfuric acid
…..			kg	lime, hydrated, packed
…..			Items	chemical factory
			kg	lithium hydroxide / lithium carbonate
	1.05E-01		kg	non-Fe-Co-metals, from Li-ion battery, hydrometallurgical processing

9.58E-02		kg	aluminium, wrought alloy
9.38E-03		kg	copper
		kg	cobalt / Cobalt sulfate - GLO
8.71E-02	$NiCO_3$	kg	nickel, 99.5% / nickel sulfate / nickel carbonate
1.07E-01	MnO_2	kg	manganese sulfate / manganese dioxide
6.74E-03	$Mg(OH)_2$	kg	magnesium hydroxide
1.09E-02	TiO_2	kg	titanium dioxide
…..		kg	Sulfur dioxide
…..		kg	NMVOC, non-methane volatile organic compounds, unspecified origin
…..		kg	Suspended solids, unspecified
…..		kg	COD, Chemical Oxygen Demand
…..		kg	Hydrocarbons, unspecified
…..		kg	Copper, ion
…..		kg	Cobalt
…..		kg	Nickel, ion
…..		kg	Fluoride
…..		l	wastewater, average
5.82E-02		kg	waste plastic, mixture
…..		kg	inert waste, for final disposal
…..		kg	waste gypsum

		Unit	**Ecoinvent dataset**
SIB			
…..		kg	
…..		l	Water, unspecified natural origin
…..		Items	chemical factory
1.63E+00		kWh	Greenpeace electricity mix
7.55E-02		kg	treatment of spent activated carbon (90%), activated carbon (10%)
1.01E-01		kg	lime, hydrated, packed
1.01E-01		kg	silica sand
1.01E+00		kg	sulfuric acid
1.21E-01		kg	oxygen, liquid
2.43E-01		kg	sodium hydroxide, without water, in 50% solution state
6.07E-01		kg	sodium carbonate
1.12E-01		kg	non-Fe-Co-metals, from Li-ion battery, hydrometallurgical processing
1.02E-01		kg	aluminium, wrought alloy
1.00E-02		kg	copper
		kg	cobalt / Cobalt sulfate - GLO
9.12E-02	$NiCO_3$	kg	nickel, 99.5% / nickel sulfate / nickel carbonate
9.12E-02	MnO_2	kg	manganese sulfate / manganese dioxide
9.34E-03	TiO_2	kg	titanium dioxide

	kg	lithium hydroxide / lithium carbonate
1.78E-01	kg	electrolyte for Li-ion battery
3.54E-01	kg	graphite, battery grade
5.82E-02	kg	waste plastic, mixture

Tables S2.4, S2.5 and S2.6 show the underlying data plotted in figures 2, 3 and 4 of the main t

Table S2.4: Data plotted in figure 2 of the main text. GWP (kg CO_2-Eq) and ADP (kg Sb-Eq) of cell produ

		cathode active m	cathode current	anode active mat	anode current co	electrolyte
GWP	NCA	2.86E+01	7.86E+00	2.91E+00	3.88E+00	3.98E+00
	NMC	1.67E+01	5.32E+00	1.91E+00	5.98E+00	3.47E+00
	LFP	1.03E+01	3.57E+00	3.35E+00	2.03E+00	6.71E+00
	SIB	1.53E+01	5.09E+00	2.91E+01	5.72E+00	4.75E+00
ADP	NCA	1.24E-02	1.11E-04	2.59E-05	6.54E-03	4.04E-03
	NMC	1.94E-02	7.64E-05	8.25E-06	1.01E-02	3.53E-03
	LFP	6.20E-03	5.04E-05	1.45E-05	3.46E-03	6.75E-03
	SIB	4.49E-03	7.20E-05	1.66E-03	8.09E-05	4.74E-03

Table S2.5: Data plotted in figure 3 of the main text. Component-wise share of GWP (kg CO_2-Eq) and AD

			Input	Aluminum	Copper	Nickel compoun
GWP	NCA	pyr.	5.83E+00	-8.60E+00	-3.50E+00	-8.22E+00
		hydr.	5.89E+00	-8.62E+00	-3.50E+00	-8.22E+00
		adv. hydr.	6.88E+00	-9.19E+00	-3.73E+00	-8.61E+00
	NMC	pyr.	6.06E+00	-5.95E+00	-5.18E+00	-5.22E+00
		hydr.	6.47E+00	-5.96E+00	-5.19E+00	-5.22E+00
		adv. hydr.	7.77E+00	-6.36E+00	-5.54E+00	-5.46E+00
	LFP	pyr.	7.46E+00	-4.97E+00	-2.04E+00	
		hydr.	5.67E+00	-4.98E+00	-2.04E+00	
		adv. hydr.	8.70E+00	-5.31E+00	-2.18E+00	
	SIB	pyr.	5.08E+00	-1.11E+01	-2.38E-01	-4.79E+00
		hydr.	4.07E+00	-1.11E+01	-2.39E-01	-4.79E+00
		adv. hydr.	6.05E+00	-1.18E+01	-2.55E-01	-5.01E+00
ADP	NCA	pyr.	4.70E-04	-1.24E-04	-6.29E-03	-4.21E-03
		hydr.	1.57E-03	-1.24E-04	-6.34E-03	-4.21E-03
		adv. hydr.	3.95E-03	-1.32E-04	-6.75E-03	-4.44E-03
	NMC	pyr.	6.33E-04	-8.56E-05	-9.36E-03	-3.00E-03
		hydr.	1.63E-03	-8.57E-05	-9.36E-03	-3.00E-03
		adv. hydr.	4.55E-03	-9.14E-05	-1.00E-02	-3.12E-03
	LFP	pyr.	4.42E-04	-7.15E-05	-3.70E-03	
		hydr.	2.38E-03	-7.16E-05	-3.70E-03	
		adv. hydr.	9.07E-03	-7.64E-05	-3.89E-03	
	SIB	pyr.	3.64E-04	-1.59E-04	-4.30E-04	-2.15E-03
		hydr.	1.66E-03	-1.60E-04	-4.31E-04	-2.15E-03
		adv. hydr.	5.86E-03	-1.70E-04	-4.60E-04	-2.22E-03

Table S2.6: Data plotted in figure 4 of the main text. Production and "net" impacts (GWP (kg CO_2-Eq) an

		Production	Current pyrometallurgical treatr		Current hydrometallurgical treat	
			Net, incl. curren	Recycling benefi	Net, incl. curren	Recycling benefi
GWP	NCA	8.55E+01	6.96E+01	-1.59E+01	6.72E+01	-1.83E+01

	NMC	7.55E+01	6.17E+01	-1.38E+01	5.91E+01	-1.64E+01
	LFP	1.01E+02	1.01E+02	4.54E-01	9.73E+01	-3.52E+00
	SIB	1.06E+02	9.31E+01	-1.33E+01	9.20E+01	-1.44E+01
ADP	NCA	2.42E-02	9.41E-03	-1.48E-02	7.96E-03	-1.62E-02
	NMC	3.47E-02	1.15E-02	-2.32E-02	9.39E-03	-2.53E-02
	LFP	1.90E-02	1.57E-02	-3.33E-03	1.41E-02	-4.90E-03
	SIB	1.36E-02	1.05E-02	-3.08E-03	1.18E-02	-1.78E-03

text

ction, component-wise.

cell container	cell manufacturi	others
1.90E+00	3.18E+01	4.64E+00
1.94E+00	3.24E+01	7.78E+00
3.05E+00	5.10E+01	2.08E+01
2.13E+00	3.39E+01	1.03E+01
5.44E-04	2.41E-04	2.70E-04
5.55E-04	2.46E-04	7.91E-04
8.73E-04	3.86E-04	1.25E-03
6.10E-04	2.57E-04	1.64E-03

P (kg Sb-Eq), including process efforts ("Input").

Cobalt compoun	Lithium Compo	Manganese com	Titanium compo	Mag-nesium	Electrolyte	Graphite
-1.41E+00						
-1.41E+00	-2.40E+00					
-1.48E+00	-2.05E+00				-3.99E+00	-2.30E+00
-2.62E+00		-8.99E-01				
-2.62E+00	-3.03E+00	-8.99E-01				
-2.74E+00	-2.59E+00	-7.69E-01			-3.45E+00	-1.90E+00
	-2.17E+00					
	-1.85E+00				-6.73E+00	-3.36E+00
		-1.87E+00	-3.74E-01			
		-1.87E+00	-3.74E-01	-7.88E-02		
		-1.60E+00	-5.20E-02	-1.09E-02	-4.14E+00	-4.50E+00
-4.61E-03						
-4.61E-03	-2.48E-03					
-4.79E-03	-2.08E-03				-4.04E-03	-9.94E-06
-1.11E-02		-2.53E-04				
-1.11E-02	-3.12E-03	-2.53E-04				
-1.17E-02	-2.65E-03	-2.16E-04			-3.53E-03	-8.20E-06
	-3.52E-03					
	-3.05E-03				-6.85E-03	-1.45E-05
		-4.97E-04	-2.01E-04			
		-4.97E-04	-2.01E-04	-4.15E-07		
		-4.24E-04	-1.72E-04	-3.54E-07	-4.19E-03	-1.94E-05

d ADP (kg Sb-Eq)) after subtracting recycling benefits.

	Adv. hydrometallurgical treatme	
Net, incl. adv. hy	Mechanical trea	Hydrometallur-gical treatment (adv. hydr.)
6.10E+01	-1.25E+01	-1.19E+01

5.45E+01	-1.04E+01	-1.06E+01
8.93E+01	-1.15E+01	7.67E-01
8.47E+01	-1.40E+01	-7.71E+00
5.89E-03	-1.08E-02	-7.44E-03
7.98E-03	-1.35E-02	-1.32E-02
8.25E-03	-1.07E-02	5.92E-03
8.75E-03	-4.80E-03	3.02E-03

Table S2.7 shows complete results for all available impact categories in ILCD midpoint

Table S2.7: LCIA results, ILCD 1.0.8 2016 midpoint, all available impact categories (FU = 1 kWh)

		climate change – GWP 100a	ecosystem quality – freshwater and terrestrial acidification	ecosystem quality – freshwater ecotoxicity	ecosystem quality – freshwater eutrophication	ecosystem quality – ionising radiation
		kg CO2-Eq	mol H+-Eq	CTUh.m3.yr	kg P-Eq	mol N-Eq
Production	NCA	8.55E+01	3.40E+00	4.68E+03	1.40E-01	4.25E-05
	NMC	7.55E+01	2.45E+00	5.75E+03	1.65E-01	3.90E-05
	LFP	1.01E+02	2.06E+00	3.01E+03	1.00E-01	5.49E-05
	SIB	1.06E+02	2.40E+00	1.62E+03	6.49E-02	4.69E-05
Current pyrometallurgi	NCA	-1.59E+01	-2.81E+00	-3.77E+03	-9.21E-02	-5.65E-07
	NMC	-1.38E+01	-1.94E+00	-4.73E+03	-1.16E-01	-1.08E-06
	LFP	4.54E-01	-1.93E-01	-1.59E+03	-3.70E-02	4.51E-06
	SIB	-1.33E+01	-1.02E+00	-6.81E+02	-1.55E-02	1.85E-07
Current hydrometallur	NCA	-1.83E+01	-2.78E+00	-3.77E+03	-9.42E-02	-2.61E-06
	NMC	-1.64E+01	-1.91E+00	-4.73E+03	-1.18E-01	-3.16E-06
	LFP	-3.52E+00	-1.44E-01	-1.59E+03	-4.09E-02	7.19E-07
	SIB	-1.44E+01	-9.71E-01	-6.63E+02	-1.66E-02	-1.87E-06
Advanced hydrometall	NCA	-2.45E+01	-2.98E+00	-4.04E+03	-1.03E-01	-3.81E-06
	NMC	-2.10E+01	-2.05E+00	-5.05E+03	-1.28E-01	-3.88E-06
	LFP	-1.07E+01	-2.25E-01	-1.72E+03	-4.69E-02	-1.85E-07
	SIB	-2.17E+01	-1.08E+00	-7.27E+02	-2.18E-02	-2.84E-06

ecosystem quality – marine eutrophication	ecosystem quality – terrestrial eutrophication	human health – carcinogenic effects	human health – ionising radiation	human health – non-carcinogenic effects	human health – ozone layer depletion	human health –photochemical ozone creation
kg N-Eq	mol N-Eq	CTUh	kg U235-Eq	CTUh	kg CFC-11-Eq	kg ethylene-Eq
1.35E-01	1.36E+00	1.26E-05	1.68E+01	2.07E-04	7.61E-05	4.44E-01
1.22E-01	1.26E+00	1.35E-05	1.54E+01	2.59E-04	1.87E-04	3.93E-01
1.32E-01	7.00E+00	4.17E-05	2.24E+01	1.14E-04	5.57E-04	2.42E-01
2.53E-01	3.16E+00	8.98E-06	1.76E+01	5.31E-05	2.20E-04	3.76E-01
-5.28E-02	-1.77E+01	-7.43E-06	2.93E-01	-1.68E-04	1.29E-07	-2.78E-01
-6.39E-02	-6.15E-01	-8.47E-06	4.35E-02	-2.17E-04	1.04E-07	-2.36E-01
-1.27E-02	-8.70E-02	-2.97E-06	1.90E+00	-7.53E-05	1.56E-06	-3.03E-02
-1.98E-02	-1.94E-01	-3.20E-06	3.87E-01	-2.53E-05	3.66E-07	-1.06E-01
-5.89E-02	-5.83E-01	-7.86E-06	-5.74E-01	-1.69E-04	-8.90E-07	-2.82E-01
-7.13E-02	-6.75E-01	-8.98E-06	-8.29E-01	-2.18E-04	-9.67E-07	-2.41E-01
-2.24E-02	-1.66E-01	-3.53E-06	3.24E-01	-7.56E-05	-4.28E-08	-3.71E-02
-2.10E-02	-2.11E-01	-3.28E-06	-5.51E-01	-2.44E-05	-5.77E-07	-1.04E-01
-6.67E-02	-6.56E-01	-8.68E-06	-9.44E-01	-1.80E-04	-1.16E-06	-3.24E-01
-7.79E-02	-7.36E-01	-9.77E-06	-1.06E+00	-2.32E-04	-1.05E-06	-2.75E-01
-2.88E-02	-2.18E-01	-4.14E-06	3.00E-02	-8.03E-05	-3.84E-08	-7.52E-02
-2.81E-02	-2.73E-01	-3.95E-06	-8.34E-01	-2.61E-05	-8.36E-07	-1.52E-01

human health – respiratory effects, inorganics	resources – land use	resources - mineral, fossils and renewables
kg PM2.5-Eq	kg Soil Organic Carbo	kg Sb-Eq
1.89E-01	5.55E+01	2.42E-02
1.45E-01	6.31E+01	3.47E-02
9.33E-02	6.42E+01	1.90E-02
1.35E-01	1.53E+02	1.35E-02
-1.45E-01	-2.13E+01	-1.47E-02
-1.03E-01	-2.80E+01	-2.32E-02
-9.91E-03	-3.63E+00	-3.34E-03
-5.52E-02	-8.97E+00	-3.10E-03
-1.46E-01	-2.81E+01	-1.62E-02
-1.04E-01	-3.69E+01	-2.53E-02
-1.23E-02	-1.36E+01	-4.90E-03
-5.51E-02	-1.62E+01	-1.81E-03
-1.61E-01	-3.16E+01	-1.83E-02
-1.15E-01	-3.90E+01	-2.67E-02
-2.22E-02	-1.56E+01	-4.83E-03
-6.78E-02	-1.86E+01	-1.78E-03