Life-cycle Assessment and Energy Systems Analysis of Second-life Li-ion Batteries

Master's thesis in

Marcus Brunström

Department of Space, Earth and Environment
Division of Energy technology

Chalmers University of Technology
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Abstract

With a large increase in electric vehicles, a subsequent increase in the retirement of Li-ion batteries (LIBs) will follow. The environmental performance of second-life LIBs in Swedish context has so far only been assessed in a LCA by Janssen et al. (2019), which this study intends to build upon. Energy system modelling is used as a mean to further understand the second-life use case of LIBs in Swedish context. Hence, what flows are needed for energy systems analysis to be successfully integrated with LCA and how it can enrich the assessment on the environmental performance of second-life LIBs is also explored. Using energy systems modeling, more than 2000 Swedish households with energy storages are modeled for economic efficiency for the homeowners maximizing the value of PV self-consumption. How the utilization of the storage differs between households, installation sizes and relative remaining storage capacities of the LIBs (as they initiate the second life) is also investigated. The results include estimated degradation, which is used to estimate the duration of the LIB's second life and consequently the second lifetime performance. The environmental burden from manufacturing, can be allocated between the LIB's first and second life by their respective energy throughput. Hence, the utilization of the storage in the second-life use case will directly effect this allocation. Furthermore, this study investigates if there is any environmental benefit in extending a LIB's lifetime in Sweden from a marginal and average perspective and will identify the main processes contributing to the impact.

The specific utilization of the storages was found to be similar between households and the different installation sizes modeled, resulting in similar environmental performances. This is likely because the utilization of the storages are mostly limited by storage volume rather than availability of PV. The environmental performance of a second-life LIB was found mainly to depend on the energy throughput during the second life, which is directly linked to the allocated burden of manufacturing, emissions from charging and avoided electricity consumption. With similar utilization between storages, and thus degradation rates, the state of health (SoH) of the LIB as it enters the second life was determining for the second-life duration and total energy throughput.
A residential second-life LIB was found to charge 0.5 to 1 $\mathrm{MWh_{el}/kWh_{NSC}}$ (nominal storage capacity) over a second lifetime of 4 to 9 years, depending on the initial SoH. Thus, a second life was estimated to relieve the first life from 15 to 29% of the burden of manufacturing by energy allocation. Under the assumption that the LIBs give rise to 250$\mathrm{kg\ CO_2eq/kWh}$ battery storage (Janssen et al., 2019), then 17 to 31$\mathrm{g\ CO_2eq/kWh_{el}}$ supplied from the second-life LIB will originate from allocated burden of manufacturing. A high energy throughput results in less impact per kWh electricity supplied. It is considered that emissions are related to charging PV electricity to the storage. For the chosen average perspective, the emission intensity of the PV electricity charged was assumed to be an attributional LCA value of 41$\mathrm{g\ CO_2eq/kWh_{el}}$ (Schlömer et al., 2014). From the marginal perspective a consequential LCA value was assumed of 76.7$\mathrm{g\ CO_2eq/kWh_{el}}$ (Jones & Gilbert, 2018). By allocated manufacturing impacts and emissions related to charging the storage, the second-life LIB investigated in this thesis was found to be able to supply electricity at 62 to 76$\mathrm{g\ CO_2eq/kWh_{el}}$ from the average perspective and 102 to 116g $\mathrm{CO_2eq/kWh_{el}}$ from the marginal. By both perspectives, emissions related to charging the storage with PV electricity was the main contribution to the environmental impact while impacts allocated from processes prior to the second-life were mainly constituted by battery pack manufacturing.

As a storage is introduced to the system, some electricity and its related emissions were considered avoided by the storage. by average accounting, the avoided emissions were considered to come from the energy mix while by marginal accounting, from the marginal technology (waste incineration). With positive and negative impacts, a net impact of the storage can be calculated. From the average perspective, extending the performing lifetime of a LIB with a second-life in Sweden resulted in an environmental burden with a net impact of 22 to 37$\mathrm{kg}\ \mathrm{CO_2eq/kWh}$ storage. From the marginal perspective, an environmental benefit was found at net impact -88 to -33$\mathrm{kg}\ \mathrm{CO_2eq/kWh}$ storage. The environmental burden is caused by the storage replacing grid electricity with lower emission intensity than it is able to supply electricity at. Similarly, the environmental benefit comes from replacing electricity, on the margin, with higher emission intensity than that supplied by the storage.

In a static system, typically depicted in LCA, avoided emissions would have been calculated with the total electricity consumption avoided and the average emission intensity of this electricity. By using average accounting for avoided emissions and hourly resolution in the energy systems model, the resulting emissions were 7 to 8% lower than when calculated for a static system, using the same data. This indicates that incorporating energy systems modeling into the LCA added value to the assessment.

Content

Abbreviations

ENTSO-E	European Network of Transmission System Operators
EV	Electric vehicle
HEV	Hybrid electric vehicle
IPCC	Intergovernmental panel on climate change
LIB	Lithium-ion battery
NMC	Ni, Mn and Co
NSC	Nominal storage capacity
PHEV	Plug-in hybrid electric vehicle
PV	Photovoltaic
SoC	State of charge
SoH	State of health
VRES	Variable renewable energy sources

1 Introduction

Electro-mobility is developing rapidly and chargeable vehicles are taking an increasing share in the market (IEA, 2019a). The European Union has set a goal of reaching carbon neutrality by 2050. Sweden has committed to reach this goal by 2045 (Government Sweden, 2017). In order to reach the 2050 goals of the European union, Berggren and Kågesson (2017) conclude that a large share of the car fleet will need to be fossil-free. With respect to current and announced policies, the International Energy Agency (IEA) estimates that 130 million electric vehicles (EVs) globally (excluding two- and three wheelers) will be on the market by 2030 (IEA, 2019a). In a scenario that includes policies set to reach climate goals and sustainability targets as in the Clean Energy Ministerial's (a global forum for promoting policies, programs and best practices towards clean energy) EV30@30 campaign, a 30% market share for EVs by 2030 is targeted (Clean Energy Ministerial, 2019), and the estimations exceed 250 million vehicles (IEA, 2019a).

Car manufacturers also see electric propulsion as the future. More than half of the manufacturers selling cars in Sweden have communicated a complete transition to electric models (Andersson & Kulin, 2018). By the end of 2019 there were in Sweden 214464 registered EVs, HEVs and PHEVs of which 73825 were newly registered during 2019 (Statistics Sweden, 2020). These numbers are also predicted to be increasing throughout the coming years (Andersson & Kulin, 2018). PHEVs and EVs are predicted to dominate the Swedish market around 2030, and by then there could be 2,5 million chargeable vehicles on Swedish roads, based on sales forecasts (Andersson & Kulin, 2018).

As a result of a significant increase in EVs on the roads there will be a subsequent increase in the retirement of lithium-ion batteries (LIBs). Tadaros (2019) estimates that the sales of EVs in Sweden between 2010-2018 will accumulate around 16 thousand tons of LIBs. Furthermore, Tadaros (2019) estimates that until 2030 Sweden will have accumulated between 207 and 727 thousand tons of LIBs in demand for recycling, based on actual and predicted sales numbers. As large quantities of batteries are predicted to be retired from their first automotive life throughout the coming years, it is important to manage these.

LIBs contribute to a large share of total cost and initial material and energy investment for EVs (Bobba, 2018; Madlener & Kirmas, 2017). When retired from their first-life automotive use, the LIBs still retain around 70-80% of their original capacity (Wood, Alexander & Bradley, 2011; Jiao & Evans, 2016; Madlener & Kirmas, 2017; Bobba, 2018; Casals, Barbero & Corchero, 2019), whereas a definitive point of retirement can be considered to be at 60% of original capacity (Casals et al., 2015; Lacey et al., 2013; Oliveira, 2017). After automotive retirement, the LIBs can be used in a less demanding second-life application, like a stationary energy storage system. Extending the life of the battery has potential to increase its total lifetime value. Furthermore, second use of EV LIBs could reduce the demand for new batteries intended for other energy storage applications, potentially reducing manufacturing related emissions through avoided production.

The technical viability of a second-use battery depends on its aging history and the requirements of the intended application (Swierczynski, 2016). Thus, when retired from their first life, the LIBs should be tested to identify their state of health (SoH) and remaining capacity so that a suitable second-life application can be chosen (Ahmadi, 2014b). However, due to both economic and technical reasons, dismantling of the cells or the battery pack is not a preferable option (Ahmadi, 2014a). For economic and technical feasibility, keeping the battery pack intact is advised for the intent of re-use (Ahmadi, 2014b; Mudgal, 2014). If the batteries prove unfit for any further use, they are retired and recycled to recover materials they are made of.

Energy storages can mitigate the variability of renewable energy sources (Beaudin, 2010) and batteries are often combined with solar photovoltaic (solar PV) due to their synergistic properties (Göransson & Johnsson, 2018). If paired with solar PV, a battery energy storage can be used to increase a household's PV self-consumption and self-sufficiency. By increasing the self-consumption of PV electricity in a household, less electricity needs to be bought from the grid to supply the household's load. Hence, any related emissions and costs of the grid electricity that is replaced by the PV are avoided. Sweden, has a very clean energy mix (Swedish energy agency, 2018; OECD, 2013; IEA, 2019b) priced slightly below European-28 average for households (including taxes) (Eurostat, 2019). Thus, the potential economic and environmental gains of avoiding electricity supplied by the Swedish grid should be relatively low compared to other national systems.

Appropriate sizing of an energy storage using batteries is crucial for maximizing revenue (DEA, 2018). In a study investigating the optimal storage size for dwellings with solar panels, Mulder et al. (2010) found the optimal storage size would be in the range of 0.4-1.5 kWh per annually produced MWh PV electricity. In a techno-economic study, Madlener and Kirmas (2017) investigated a dwelling with 5 kWp of installed PV capacity and found maximum net present value of the storage to be reached at 5.5 kWh of installed storage capacity, rising to 7 kWh as battery prices decrease from 117 €/kWh to 34 €/kWh installed storage capacity (Madlener & Kirmas, 2017). The average price of a new LIB pack for EVs in 2020 was reported by Bloomberg New Energy Finance (2020) to be 135 $/kWh. Like Naumann et al. (2015), Madlener and Kirmas (2017) estimate that the optimal PV and storage dimensions are roughly 1 kWh storage per kWp of solar PV. However, to compensate for the initial capacity loss of the batteries, 1.2 kWh/kWp is suggested (Madlener & Kirmas, 2017). It is stated that each kWp of PV capacity produces 980 kWh of electricity annually (Madlener & Kirmas, 2017). The 1.2 kWh/kWp provided in Madlener & Kirmas study (2017) can be recalculated to 1.22 kWh storage capacity per annually produced MWh PV electricity, which is in line with Mulder's (2010) assessment.
Nyholm et al. (2016a) also states that the initial capacity loss of the batteries should be compensated for. However, due to lack of consistent values for battery degradation, Nyholm et al. (2016a) neglect it and are instead using effective energy storage volume as the basis for calculation. The approach to dimensioning of the PV and storage used by Nyholm et al. (2016a), and in this study, includes the consumption of the house at which the storage system is installed. The dimensioning is based on the array-to-load ratio (ALR), which describes the relation between installed PV capacity and average annual load, and the relative battery capacity (RBC), which describes the battery storage capacity relative to the installed PV capacity (see section 3.1). Nyholm et al. (2016a) found the additionally gained degree of self-sufficiency and self-consumption of the house to decline gradually at higher RBCs. As benefits gained from adding storage capacity gradually declines it is deemed unlikely that RBCs over 2-3 would be employed for PV related purposes, in terms of both economic efficiency and increasing self-sufficiency (Nyholm, 2016b). As a reference, a RBC of 1 and the household

load data used by Nyholm (2016b) produces roughly 1.1kWh/kWp. The same load data is used for this study.

Previously, second-life LIBs have been investigated for places like Germany (Madlener & Kirmas, 2017), Spain (Casals, 2019) and the Netherlands (Bobba, 2018), providing results regarding the utilization of the residential storage and economic performance. Studies that investigate the performance of PV and storage in the Swedish electricity system have also been conducted (Nyholm et al., 2016a; Thygesen & Karlsson, 2014; Widén & Munkhammar, 2013). These studies provide results mainly on the utilization of storages, e.g. by the terms of degree of self-consumption and self-sufficiency. Furthermore, a difference in potential for these parameters depending on geographic location has been identified (Nyholm, 2016b). The supply- and value chain has also been explored in Swedish studies (Tadaros, 2019; Olsson et al., 2018). However, the environmental performance and degradation of batteries in a Swedish use case remains relatively unexplored. The environmental performance of second-life LIBs in Swedish context has so far only been assessed in a LCA by Janssen et al. (2019). Battery degradation was not included in studies, due to a lack of information regarding aging and degradation for second-life LIBs (Bobba, 2018; Nyholm et al., 2016a; Martinez-Laserna, 2018). Although LIBs aging performance is still in need of further understanding and available information may be inadequate for estimating the performance beyond its measurement timeframe (Neubauer, 2015), degradation remains important to the viability of second-life LIBs (Martinez-Laserna, 2018).

There is room for further investigation of the environmental performance of second-life LIBs used in Sweden. LCA is a good method of investigating environmental performance, but it typically describes a static system. As a consequence, information about activities in the energy system are lost in average values, which could lead to an inaccurate result. For example, using a yearly average emission intensity of the electricity system may not be representative when investigating avoided emissions by using solar PV, which mainly produces electricity during summer in Sweden and only during the day. Knowing the electricity mix and its emission intensity when the PV produces would be better for investigating avoided emissions than assuming the emission intensity of the yearly average mix, if such a method is chosen. Time dependencies like this and other interactions between units in an electricity system can be investigated with energy systems modeling. Thus, incorporating an energy systems analysis into a LCA could be a way of including valuable information otherwise lost in an assessment of environmental performance. In the case of LIBs, energy systems modelling can be used to further understand a second-life use case in Sweden through results describing its utilization, like energy throughput and interaction with PV and household load. By applying degradation values to the utilization, an estimate on the duration of the LIB's second life would be possible. As a result, the second lifetime performance of the LIBs could also be estimated. Adding degradation values to the utilization, and thus connecting use case and life duration, should be beneficial when learning about a specific use case. However, as LIB degradation is still in need of further understanding, the accuracy and quality of a life duration estimate based on it will be uncertain. Here benefits are set against uncertainties.

There are previous LCAs on LIBs which can vary in scope and goal but mostly have some common ground in cradle-to-gate impacts. This impact can for NMC (Ni, Mn & Co) LIBs range 73 to 250 $\mathrm{kg\,CO_2eq/kWh}$ storage capacity for varying cathode chemistries, cell and module count as well as total storage capacity (Ellingsen, 2014; Dai et al., 2019; Janssen et al., 2019;

Accardo et al., 2021). The impact of LIB production depends heavily on the supply chain as well as the bill of materials (BOM), which can vary with battery size, configuration and desired performance characteristics (Dai et al., 2019). For the production of a LIB, Ellingsen (2014) found the cell manufacture, positive electrode paste and negative current collector to be the main contributors to all impact categories. Towards the CO_2eq impact found by Dai et al. (2019), the cathode material, aluminum and energy for cell production were the largest contributors. Ellingsen (2014) identifies a broad range of reported energy requirements for battery cell production. Furthermore, performed sensitivity analysis showed that the most effective means of reducing cradle-to-gate emissions was to use a cleaner energy mix for battery cell production (Ellingsen, 2014). Studies that assess the environmental performance of batteries for their performing lifetime can reach very different conclusions depending on which assumptions are made regarding cycle numbers (Ellingsen, 2014). Casals et al. (2017) found that the environmental impact per unit of storage capacity decreases with increased utilization. Thus, a battery chemistry that has a longer lifetime as a result of less degradation, than what the common graphite and manganese-based batteries can offer, is favorable (Casals et al, 2017). A longer lifetime would benefit the second life LIB as well. Wilson et al. (2021) showed that a repurposed LIB can achieve carbon reductions if the second lifetime exceeds 4.25 years in an Australian home energy system.

1.1 Aim

This thesis will make an interdisciplinary analysis, by integrating energy systems analysis with life-cycle assessment, to investigate the related emissions and utilization of second-life LIBs from EVs used as residential energy storage in Sweden.

Research questions for the thesis are as follows:

1. What flows and parameters are needed to be exchanged between an LCA and an energy system model used to define and understand the second-life use case of a LIB?

2. If a 2nd life LIB is used as a residential energy storage paired with solar PV and operated for economic efficiency for a Swedish homeowner:
    - Will the LIB provide any environmental benefit in terms of CO_2eq emissions?
    - What are the main processes contributing to the environmental impact?
    - To which degree is the environmental impact affected by the size of the installation and relative remaining storage capacity of the LIB (SoH)?

2 Theory and Background

2.1 The Lithium-ion Battery

The Lithium-ion battery (LIB) technology is commonly used for EVs. It has characteristics like high specific energy and capacity, low internal resistance and self-discharge rate as well as good coulombic efficiency (Buchmann, 2018). In addition, it also has a long cycle- and shelf-life (Buchmann, 2018). Limitations of the LIB include lithium being inherently instable in metallic form, potential of degradation during certain conditions and requirements of protection to ensure safety (Buchmann, 2018).

The main components of a LIB cell are the negative and positive electrodes, i.e. the anode and cathode, as well as the electrolyte and separator. The chemistries of these components can be altered to modify the desired performance of the battery. The anode in most commercial LIBs is graphite based (Li et al., 2018; Bruce, 2008). The cathode material is an alloy in which Li, Ni, Mn and Co are the most frequently used metals (Gopalakrishnan, 2016). A commonly used electrolyte is $LiPF_6$ (Xu, 2012).

Several connected battery cells along with a battery management system and an encasing constitutes a battery pack. A battery pack is sized and designed to meet the requirements of its intended application.

2.2 Battery Degradation

The degradation and lifespan of a LIB is hard to predict but depends on the state of health (SoH), load levels, calendar aging, charge and discharge rates and operating temperature (Tadaros, 2019; Buchmann, 2020a; DEA, 2018). Furthermore, it also depends on the quality of the battery i.e., chemistry and manufacturing (DEA, 2018). Generally, a battery's aging process leads to an increase in internal resistance and self-discharge rate and reduced capacity (Broussley, 2001; Barré, 2013). However, under specific circumstances the condition and performance of a battery can be caused great harm in complex ways that are hard to account for and measure. If deeply discharged, graphite exfoliation of the anode and electrolyte degradation can occur which heavily degrades the battery performance (DEA, 2018). High state of charge (SoC) at low temperatures can cause capacity loss through lithium plating on the anode (Gopalakrishnan, 2016; Buchmann, 2020a). Cycling at high SoC and temperature can cause capacity loss and increased internal resistance as a result of a solid electrolyte interface forming on the anode (Andersson, 2003; Vetter, 2005; Buchmann, 2014; Gopalakrishnan, 2016). High cell voltage and temperature can result in lost capacity caused by electrolyte oxidation on the cathode (Buchmann, 2014). Fast charging at low temperatures can impose safety issues as it promotes dendrite growth on the anode, being a solid formation that can penetrate the separator and cause a short circuit (Buchmann, 2018; Buchmann, 2020b). Post automotive retirement, an accelerated degradation can come into effect, called the aging knee, which requires immediate retirement (Spotnitz, 2003; Bobba, 2018; Martinez-Laserna, 2018).

In general terms, SoC above 80% hastens cathode degradation while discharging below 20% increases internal resistance (Buchmann, 2020a). Operating the battery in a manner that avoids circumstances where impactful degradation mechanisms are present will effectively extend the life length and cycle count. Hence, approaching the upper and lower limits of SoC, maintaining high voltage, operating under high or low temperatures as well as using fast charge and discharge rates, should be avoided.

Several studies identify the lack of data regarding degradation and aging performance of second-life LIBs (Bobba, 2018; Nyholm et al., 2016a; Martinez-Laserna, 2018). These parameters were identified by Martinez-Laserna (2018) as pivotal to the economic and technical viability of the second-life batteries. Neubauer (2015) states that predicting the relative remaining performance of a battery requires understanding of the degradation, which still needs further research. Models that attempt to estimate the lifetime and remaining relative performance often require extensive amounts of data, which is usually inadequate for extrapolating beyond its timeframe (Neubauer, 2015). Neubauer (2015) also identifies physics-based degradation models present in the literature, although these are limited by complexity, narrow scope and operating conditions.

3 Method

In this study, a use case for second-life LIBs is investigated, focusing on its life from the cradle to the end of performing life, meaning end-of-life management such as battery material recycling is not accounted for. To investigate the environmental performance of a LIB from the cradle to the point where the battery is ready to initiate a second performing lifetime, LCA is applied. The LCA of this study is based on a comparative attributional LCA by Janssen et al. (2019) which investigates the environmental performance of the battery pack from a Mitsubishi Outlander PHEV. The manufacturing process for these batteries was modeled (Janssen et al., 2019) based on the inventories provided by Bobba (2018) and Ellingsen (2014). Similarly, the refurbishment process preparing the batteries for a second performing lifetime were based on the inventory by Bobba (2018). In this study, the assessment Janssen et al. (2019) made on the production and refurbishment process is used and the same first life is also assumed. However, for the purposes of this study, the assessment on the second life is replaced with information gathered from applying energy systems analysis. The refurbishment process would take place in a facility in Halmstad, after which the LIBs would be transported by truck to the households to serve a second life.
The tool used for the LCA is the open source LCA software OpenLCA 1.10.2 (OpenLCA, 2019) with the ecoinvent 3.4 database (Wernet, 2016; ecoinvent, 2017). The impact assessment method used to calculate the climate impact of the product system is the IPCC 2013 GWP 100a method (Stocker et al., 2014).

To investigate the second-life stage of a LIB, a linear programming model of a residential energy system with PV and energy storage was created that minimizes the cost of electricity for the homeowner. The connection layout for the residential energy systems modeled can be seen in figure 1. A battery and PV installation operate with DC while a household’s load is AC. This is not physically represented in the model, only an inverter is added to separate the two sides. The tool used for linear programming in this study is GAMS release 30.3 (GAMS, 2020) and perfect foresight was assumed during optimization.

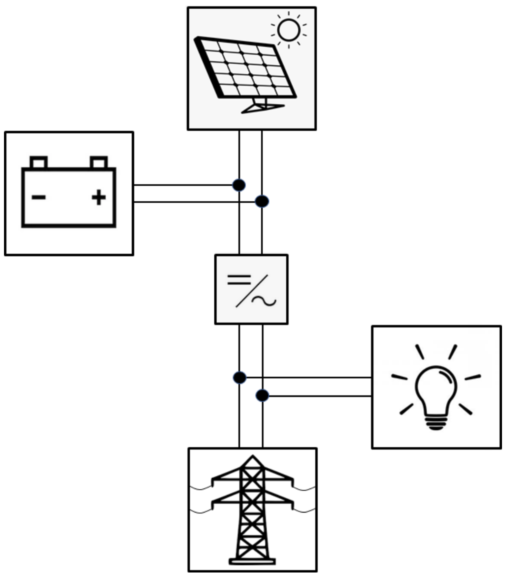

Figure 1: The connection layout for the household used in the energy systems model.

For the energy systems analysis to be successfully integrated with the LCA, the flows and processes of the two models need to be understood, as well as their intended purpose. The goal of using the energy systems analysis is to learn more about the second-life use case in Sweden. The goal of incorporating it into the LCA is to capture important information, which might have been lost if only LCA would have been used. A visualization of how the flows are exchanged between the methods can be seen in figure 2.

The energy systems model enables investigating the utilization of the LIB storage over time. In this case calculated once per hour over one year. Modeling the storage is done through optimizing its use for economic efficiency. For this to be optimized, data on the overtime change in electricity price, PV production and household load is needed. If the storage is assumed to only be charging PV produced electricity and the utilization is known, the emissions related to charging the storage can be calculated and added to the LCA.

As illustrated in figure 2, adding battery degradation to the modeled utilization of the storage allows the duration of the second lifetime to be estimated. As a result, the second lifetime performance of the LIBs can also be estimated. Furthermore, it connects the second-life duration with the modeled use case.

A battery's performance is measured by parameters such as available capacity and power, energy density, charge acceptance, self-discharge and cycle life. The degree to which a battery has retained its original performance parameters describes the general condition of the battery and is referred to as its state of health (SoH). In this study, the aging and degradation of the LIBs is measured by its SoH only. The degradation of the batteries limits the remaining available capacity. Hence, it also limits the utilization of the storage, which is being optimized. In the context of energy modeling, adding degradation as a constraint in the optimization process would make the model non-linear, which means the model may not produce an optimal solution. To this end, the degradation of the batteries is calculated post-optimization, maintaining a linear model and ensuring an optimal solution.

By discharging an energy storage to supply electricity to the load, less electricity needs to be bought from the grid. The electricity that does not need to be bought from the grid can then be considered avoided consumption. Any related emissions and costs of this electricity are then also avoided. Which plants actively generate electricity in the system depend on their cost and availability. Preferably, the cheapest available electricity is used until the load of the system is supplied. Since both the availability of electricity generation and the load changes over the day and over seasons, the electricity mix does too. Furthermore, the different technologies present in the mix supply electricity at different emission intensities. Thus, related emissions of the grid electricity depends on the hourly mix. If data on how the emission intensity of grid electricity changes over time is fed into the energy systems model, the avoided emissions could be calculated for every hour modeled. Moving calculations on environmental impact for the second life from the LCA to the energy systems analysis should then be beneficial since these changes over time can be captured into the assessment. The results can then be fed into the LCA for the overall assessment.

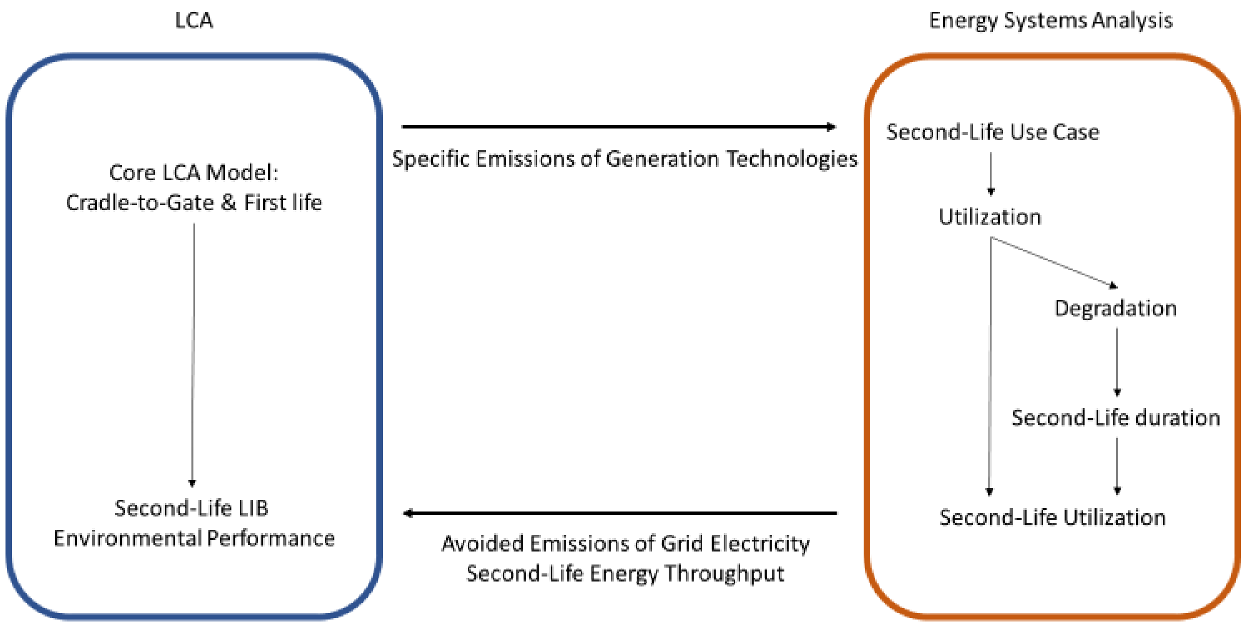

Figure 2: Mapping of the main functions of the methods used and the flows exchanged between them. The LCA model by Janssen et al. (2019) investigates the manufacturing and first life of the LIB. The energy systems model is used to find the LIB's second-life utilization and avoided emissions. The results can then be compiled into the LIB's second-life performance. Specific emissions refer to the emission intensity of supplied electricity.

When a new unit like a LIB energy storage enters the energy system, the load of the system is affected. In this case, the total load would decrease since the household needs to buy less grid electricity. When less electricity is needed, the most expensive active unit in the grid i.e. the marginal technology will need to reduce its output. By marginal accounting, the storage will be considered responsible for the reduced output of the marginal technology since this is the immediate effect on the system when the storage is added. The emissions associated with the electricity avoided are then also avoided by the storage. By average accounting, avoided electricity and its related emissions will be considered to come from the energy mix rather than the marginal technology alone. The accounting methods are applied to the second life only while the LCA on the first life is kept attributional. However, these accounting methods will be considered to affect the emissions related to PV production as well, and as a result the emissions related to charging the battery storage during its second life. For average accounting, PV emisisons from attributional LCA approach will be used while for marginal accounting results from a consequential approach, seeking to capture the changes in a system from an activity, will be used.

The system boundary chosen for the electricity system surrounding the PV and storage installations is in this study drawn at the Swedish border while Imported and exported electricity to surrounding electricity systems is not included. Therefore, electricity supplied to the houses by the grid is limited to Swedish production.

To keep simplicity in the model, it is chosen to limit the storage to charging PV generated electricity. This means that arbitrage trade and charging low-cost grid electricity for later use are not included functions. Since perfect foresight is assumed for optimization, utilization of the storage related to these functions would likely be overestimated. With known electricity prices and abundancy of PV, the model may utilize the storage for gains toward economic efficiency that are very small. When the utilization is not directly connected to lost storage capacity, the gains from increased utilization may not make up for the lost capacity.

3.1 Energy Systems Analysis - Model Description

The energy systems model created seeks to minimize the total electricity costs for the homeowner, which is defined as:

$$\text{Total cost} = \sum_h \sum_t \text{v\_Grid to Load}_{t,h} \times (\text{Spot price}_t + \text{Add}^{bought}) - \text{v\_PV to Grid}_{t,h} \times (\text{Spot price}_t + \text{Add}^{sold}) \quad (1)$$

Where the variable $\text{v\_Grid to Load}_{t,h}$ is the energy bought from the grid to supply the household's load and the variable $\text{v\_PV to Grid}_{t,h}$ is the excess PV electricity which is sold to the grid. Spot price_t is the spot price of electricity in the Swedish electricity system while Add^{bought} and Add^{sold} are additions to the spot price set to represent the cost of bought and sold electricity. The denotations t and h are indexes representing time and household number.

The dimensioning of PV and storage for the home installations are set by the array-to-load ratio (ALR), as defined by Widén & Wäckelgård (2009):

$$\text{ALR} = \frac{\text{kWp PV}_h}{\text{Average annual load}_h} \quad (2)$$

and the relative battery capacity (RBC), as defined by Nyholm et al. (2016a):

$$\text{RBC} = \frac{\text{kWh storage}_h \times 1000}{\sum_t \text{kWp PV}_h \times \text{PV profile}_{t,h}} \quad (3)$$

where kWp PV_h is the peak power production capacity of the solar panels and kWh storage_h is the nominal storage capacity (NSC) of the battery. $\text{PV profile}_{t,h}$ is the production profile of the solar panels, declaring how much of the installed capacity can be produced for each time step. These PV profiles were created from typical meteorological year data for their respective household by Nyholm et al. (2016a) based on the model framework by Norwood et al. (2014).

The PV electricity generation is limited by the installed capacity and the PV profile:

$$\text{kWp PV}_h \times \text{PV profile}_{t,h} \geq \text{v\_Charge}_{t,h} + \text{v\_PV to load}_{t,h} + \text{v\_PV to Grid}_{t,h} \quad (4)$$

Where the variable $\text{v\_Charge}_{t,h}$ is the energy volume used to charge the battery storage, the variable $\text{v\_PV to load}_{t,h}$ is the PV power used to supply the load directly and the variable $\text{v\_PV to Grid}_{t,h}$ is the excess energy which is sold to the grid.

The maximum amount of energy that can be stored in the batteries is limited by the installed storage capacity, the state of health of the batteries and the state of charge range:

$$\text{v\_SoC}_{t,h} \leq \text{kWh storage}_h \times \text{SoH}_{t=1} \times (\text{SoC limit}^{upper} - \text{SoC limit}^{lower}) \quad (5)$$

Here, the variable $\text{v\_SoC}_{t,h}$ is the state of charge of the storage in kWh, $\text{SoH}_{t=1}$ is the initial SoH of the LIB when it starts its second life. The parameters SoC limit^{upper} and SoC limit^{lower} are the upper and lower state of charge limits in percent.

The following equation, defines the amount of energy present in the storage:

$$\text{v\_SoC}_{t,h} = \text{v\_SoC}_{t-1,h} \times (1 - \text{Self discharge}) + \text{v\_Charge}_{t,h} \times \eta_{Charge} - \frac{\text{v\_Discharge}_{t,h}}{\eta_{Discharge}} \quad (6)$$

Where $\mathrm{v\_SoC}_{t-1,h}$ is the state of charge of the storage at the previous time step in kWh and Self discharge is the rate at which the storage self-discharges stores energy. The variable $\mathrm{v\_Discharge}_{t,h}$ is the amount of energy discharged from the battery. η_{Charge} and $\eta_{\mathrm{Discharge}}$ are the charge and discharge efficiencies of the battery.

A household's load is supplied by the grid and PV and storage system and must always be satisfied:

$$\mathrm{Load}_{t,h} \leq \mathrm{v\_Grid\ to\ load}_{t,h} + (\mathrm{v\_Discharge}_{t,h} + \mathrm{v\_PV\ to\ Load}_{t,h}) \times \eta_{\mathrm{Inverter}} \tag{7}$$

Where η_{Inverter} is the inverter efficiency.

The following equations 8 to 12 are not constraints and are calculated post model optimization. The equations including variables for energy flows use the output values of the already optimized model.

The battery's SoH degradation is calculated according to:

$$\mathrm{SoH}_{t,h} = \mathrm{SoH}_{t-1,h} - \mathrm{Calendar\ aging} - \mathrm{Cycling\ aging} \times \frac{\mathrm{v\_Discharge}_{t,h}}{\mathrm{kWh\ storage}_h \times \mathrm{SoH}_{t-1,h}} \tag{8}$$

Where $\mathrm{SoH}_{t-1,h}$ is the battery's SoH for the previous time step. The Calendar aging defines how much the battery ages over time while Cycling aging defines how much the battery degrades on a full cycle.

The LIB's second lifetime is linearly estimated according to:

$$\mathrm{Linear\ estimation\ of\ second\ lifetime}_h = \frac{\mathrm{SoH}_{t=1,h} - \mathrm{SoH}^{retire}}{\mathrm{SoH}_{t=1,h} - \mathrm{SoH}_{t=n,h}} \tag{9}$$

Where SoH^{retire} is the SoH value for which the LIB has reached a point of definitive retirement and $\mathrm{SoH}_{t=n,h}$ is the SoH of the battery at the last time step of the year modeled.

The $\mathrm{CO_2eq}$ intensity of grid electricity by average accounting is calculated by weighted average:

$$\mathrm{Grid\ CO}_{2\,t}^{\mathrm{Average}} = \frac{\sum_{\mathrm{plant}} \mathrm{Generation}_{t,\mathrm{plant}} \times \mathrm{LCA\ emissions}_{\mathrm{plant}}}{\sum_{\mathrm{plant}} \mathrm{Generation}_{t,\mathrm{plant}}} \tag{10}$$

where $\mathrm{Generation}_{t,\mathrm{plant}}$ is the energy output per energy technology and $\mathrm{LCA\ emissions}_{\mathrm{plant}}$ is the specific lifecycle emissions per energy technology. The index plant represents the type of energy technology. Assumptions on which technologies are present in the energy systemmodeled and their specific emissions can be found in section 3.2.

The emissions avoided through avoided consumption of grid electricity which can be attributed to the battery alone is calculated according to:

$$\mathrm{Avoided\ Grid\ CO}_2^{\mathrm{Accounting}} = \frac{\sum_t \mathrm{v\_Disharge}_{t,h} \times \eta_{\mathrm{inverter}} \times \mathrm{Grid\ CO}_{2\,t}^{\mathrm{Accounting}}}{\mathrm{kWh\ storage}_h} \tag{11}$$

Where the index Accounting represents which accounting method is chosen for avoided emissions, average or marginal.

The PV emissions related to charging the battery are calculated as follows:

$$\text{PV emissions}_{h}^{\textit{Battery charging}} = \frac{\sum_{\text{t}} \text{v\_Charge}_{\text{t,h}} \times \text{LCA emissions}_{\text{PV}}^{\text{Accounting}}}{\text{kWh storage}_{\text{h}}} \tag{12}$$

Here $\text{LCA emissions}_{\text{PV}}^{\text{Accounting}}$ is the emissions from PV production that is allocated to the PV electricity produced per accounting type.

3.2 Inputs, Assumptions and Allocation

The energy systems model of this study uses historic and generated data. The time resolution of the data used is hourly over one year, i.e. 8760 hours/time steps. For the data types to be compatible and the degradation of the storage to be simplified, some assumptions were also necessary. These are described below. The complete list of inputs to the energy systems model are compiled in table 1. A similar compilation for the LCA can be found in in table 2.

The model only accounts for degradation and aging by SoH. However, the battery storages are modeled to operate between 80% and 20% SoC to simulate realistic conditions and avoid impactful degradation mechanisms. This way, something other than the general SoH is less likely to be the cause for retirement of the batteries. Furthermore, it is assumed that all batteries in a pack have the same initial SoH and age at an equal pace in their second lives. The degradation of the batteries is not added as a constraint but is instead calculated post model to maintain a linear model and ensure a reliable optimal solution is found. However, this means that the initial SoH of the batteries determines the available storage capacity throughout the whole year modeled. As a result of this, the SoC limits are also constant throughout the modeled year since they are relative to the maximum storage volume, affected by the SoH. No specific constraints for charge and discharge rates are added to the model. This assumes that the battery level can be fully charged or discharged in one timestep (one hour).

The demand data used in this study are load profiles with an hourly resolution for 2221 Swedish households from 2012 measured in a campaign by E.ON (2013). These households do not originally have solar PV or battery storages installed. It is possible that homeowners become more conscious about their electricity consumption after they have installed solar panels and shift their load slightly towards PV production hours. If these households would install a PV and storage system, the measured load data and the actual load might not be identical. However, for this study it is assumed that the load remains unchanged after a PV and storage system is installed. This assumption makes it easy to investigate how much consumption of grid electricity is avoided by the addition of PV and energy storage system.

The percentage of the installed solar PV capacity that is produced for each hour is represented in PV profiles for each household. These profiles were created for their respective household from typical meteorological year data by Nyholm et al. (2016a) based on the modeling framework by Norwood et al. (2014). For the model, it is assumed that the excess electricity produced by the installed solar panels can be sold to the grid at any point in time and that the sold electricity does not affect the production of other units in the system. It is also assumed that the residential PV and storage installations of the households modeled do not affect each other.

The historic data for spot price of electricity used in the model is retrieved from Nord Pool (2012). Apart from spot price, additions to bought and sold electricity are used. These additions are based on Borg’s (2015) assessment for customers in the Gothenburg area and include grid fee/revenue (Göteborg Energi, 2015), energy tax and VAT (Ekonomifakta, 2015), renewable energy certificate (Swedish Energy Agency Cesar, 2015) and an assumed surcharge (Borg, 2015). The additions can be found in table 1. Since arbitrage trade and charging of grid electricity is not modeled in this study, the values are deemed unlikely to have a large effect on the utilization of the storage. The utilization of the storage should mainly be governed by the availability of PV, spot price and household loads. Hence, the values used are considered acceptable for the purposes of this study.

To account for emissions of grid electricity in the Swedish electricity system, generation per production type data from the European Network of Transmission System Operators (ENTSO-E, 2018) are used to represent the power mix. This information can be combined with the specific emissions (emissions per unit of energy produced) of each generation type to estimate the specific emissions of Swedish grid electricity.
The ENTSO-E transparency platform launched in 2014, and furthermore, no complete production data is provided for Sweden before 2016. Since electricity production follows the load, using production data for another year would create a mismatch between the data sets and as a result inaccurate amounts of avoided emissions by the PV and energy storage. In the absence of 2012 power mix data to match the load data, an average production year data set was created using production data from 2016, 2017 and 2018 (ENTSO-E, 2018). The data from these years were scaled and moved to fit the weekday and weekend pattern of 2012. However, holidays were not adjusted for. A datapoint in the average production year set is then constituted by the average value of corresponding datapoints from 2016, 2017 and 2018. The created average production year dataset is shown in figure 3 below.

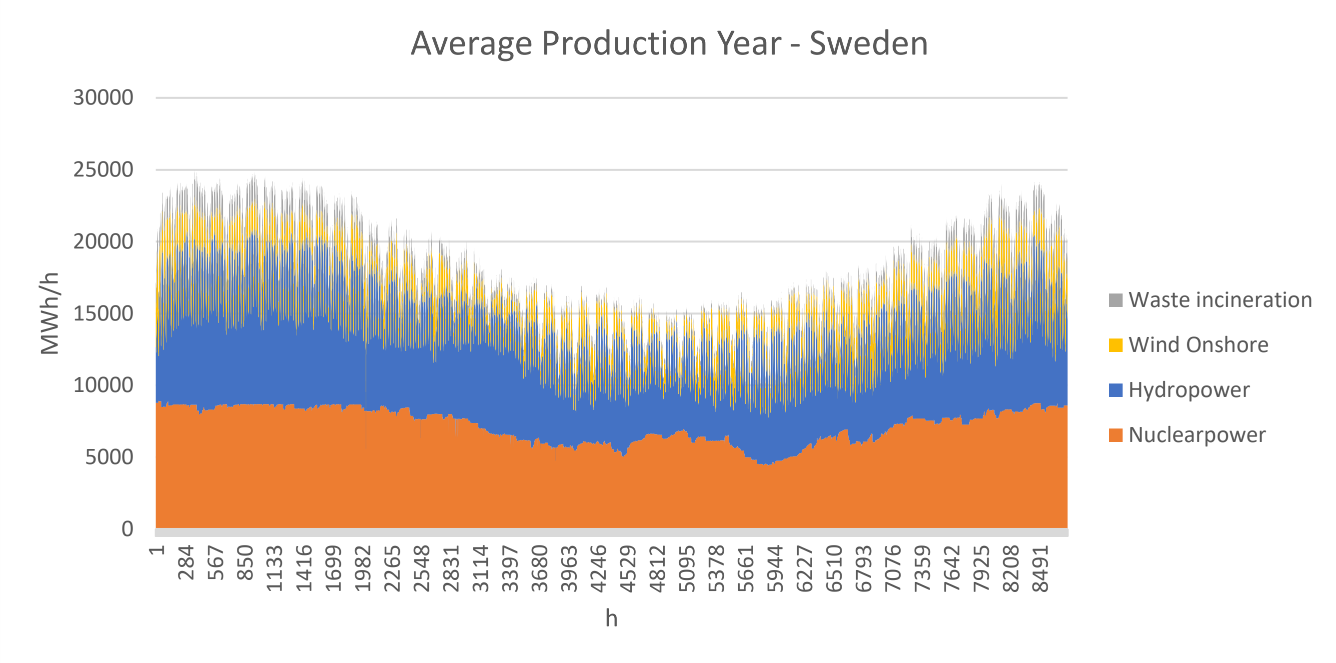

Figure 3: Average electricity production year created with production data from 2016, 2017 and 2018 (ENTSO-E, 2018).

The specific emission values used for most generation technologies are median lifecycle emissions provided in the IPCCs climate change report (Schlömer et al., 2014) and can be found in table 1. By marginal accounting the immediate effect of adding a storage to the system is considered. This approach is considered to also extend to the PV manufacturing. Thus, to account for PV emissions from the marginal perspective, results from a consequential LCA on PV by Jones and Gilbert (2018) is used. This value on specific emission intensity can also found in table 1. In the Swedish energy system, waste incineration plants supply electricity. A process from the Ecoinvent database (2017) based on the inventory by Doka (2013) was chosen to represent the specific emissions of these plants. The process chosen is named *"treatment of municipal solid waste, incineration | electricity, for re-use in municipal waste incineration only | APOS, U"* which emits 604.93g $\mathrm{CO_2/kWh_{el}}$. In the chosen process, the biogenic share of carbon in the waste is 61.1%, which is in line with the results of a study performed by Avfall Sverige, who found the fossil share in solid waste to be around one third in Sweden (Blomqvist, 2012). It is also stated that the products of the incinerating one kg of waste is 1.39MJ/kg electric energy and 2.85MJ/kg thermal energy. From the raw output of the process it is chosen to distribute the impact between the products by their energy value. Thus, 32.8% of the emissions are allocated to the electricity, resulting in the specific emission value 198.31g $\mathrm{CO_2eq/kWh_{el}}$. It is stated that in the process chosen 0.24 kg of slag and residue per kg of waste are generated. However, as the slag can be considered an unusable resource, no emissions are allocated to it. From waste incineration, the waste reduction service and the products of incineration, heat and electricity, can be considered the source of the impact rather than the waste itself. Thus, all emissions are allocated to the products of incineration and none to the fuel. Furthermore, there is no commonly acceptable way of making the allocation between the waste and the products of incineration (Harmelink & Bosselaar, 2013).

The emissions of electricity produced by the energy technologies seen in table 1 is combined with the average production year data set created from figure 3. By weighted average of production volume the emissions of grid electricity with hourly resolution is calculated, which can be seen in figure 4

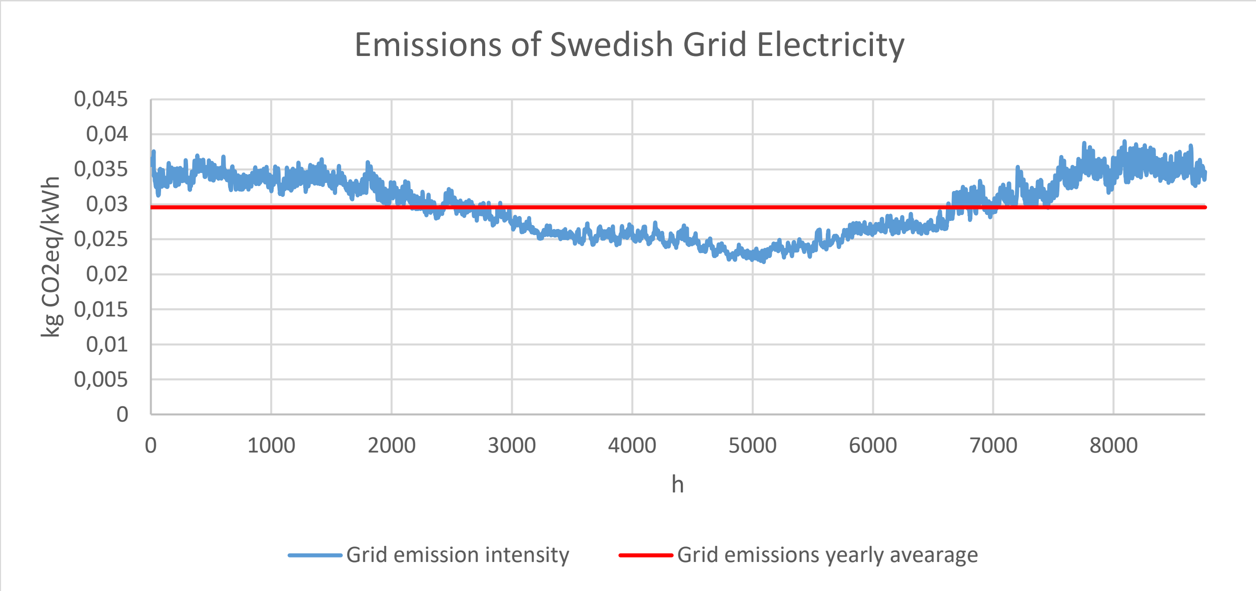

Figure 4: Emissions of Swedish grid electricity with hourly resolution, limited to national production (excluding imports and exports). The yearly average emission intensity is 29.6g CO_2eq/kWh_{el}.

By average accounting, the introduction of a residential PV and storage systems is considered not to cause a specific change in the system but rather impact it as a whole. The avoided electricity and emissions is then assumed to come from the electricity mix, in this case the created data set shown in figure 4.

By marginal accounting, the immediate effect on the system from the addition of a battery storage is considered to be less required output of the most expensive active unit i.e. the marginal technology, which will then constitute the avoided emissions. The merit order of production technologies in the system is assumed to be: Wind<Hydro<Nuclear<Waste (Energy Markets Inspectorate, 2006). Waste incineration plants reduce waste, and get paid to do so, but can also produce electricity for revenue. As the Swedish energy system develops to have higher shares of renewable energy, it is assumed that electricity produced beyond the waste reduction service will increase to manage variability. As the products of the incineration are considered responsible for the impact in this study, generated electricity and heat will carry the cost and environmental impact. Electricity from waste incineration is present at all hours in the load data used and the introduced storage creates a small load reduction. Furthermore, it is assumed that the load reduction does not impair the required waste reduction service. So, the addition of the storage will not imply an accumulation of waste since the development of the energy system will provide opportunity for waste incineration. Thus, waste incineration for revenue constitutes the marginal technology throughout the whole time period modeled.

Table 1: Inputs to energy systems model

Input	Value	Source
Swedish household loads	Data set	E.ON, 2013
PV production profiles for the households	Data set	By Nyholm et al., 2016a based on Norwood et al., 2014
Swedish electricity spot price	Data set	Nord pool, 2012
Addtion to spot price to represent bought grid electricity	$+0.61 \mathrm{SEK/kWh_{el}}$	Borg, 2015; Ekonomifakta, 2015; Swedish Energy Agency, 2015; Göteborg Energi, 2015
Addition to spot price to represent sold PV electricity	$+0.23 \mathrm{SEK/kWh_{el}}$	Borg, 2015; Ekonomifakta, 2015; Swedish Energy Agency, 2015; Göteborg Energi, 2015
Swedish electricity generation per production type	Data set	ENTSO-E, 2018
Specific lifecycle emissions - Nuclear	$3.7\mathrm{g\ CO_2eq/kWh_{el}}$	Schlömer et al., 2014
Specific lifecycle emissions – Hydro	$24\mathrm{g\ CO_2eq/kWh_{el}}$	Schlömer et al., 2014
Specific lifecycle emissions – Onshore wind	$11\mathrm{g\ CO_2eq/kWh_{el}}$	Schlömer et al., 2014
Specific lifecycle emissions – Rooftop solar PV	$41\mathrm{g\ CO_2eq/kWh_{el}}$	Schlömer et al., 2014
Specific consequential lifecycle emissions – Rooftop solar PV	$76.7\mathrm{g\ CO_2eq/kWh_{el}}$	Jones & Gilbert, 2018
Specific Lifecycle emissions – Waste incineration	$198.31\mathrm{g\ CO_2eq/kWh_{el}}$ (modified from 604.93)	Doka, 2013
Array-to-load ratio	2-4	Assumed values
Relative battery capacity	1	Assumed value
Initial SoH for second-life LIBs	70-80% SoH	Wood, Alexander & Bradley, 2011; Jiao & Evans, 2016; Madlener & Kirmas, 2017; Bobba, 2018; Casals, Barbero & Corchero, 2019

LIB - Point of retirement	60% SoH	Casals et al., 2015; Lacey et al., 2013; Oliveira, 2017
Cycling aging	0,0125 %/cycle	Faria et al.,2014
Calendar aging	0,00114 %/day	Bobba, 2018
Self-discharge rate	0,1 %/day	DEA, 2018
Upper state of charge limit	80% SoC	Assumed value
Lower state of charge limit	20% SoC	Assumed value
Charge efficiency	95%	Battke et al., 2013; Faria et al., 2013
Discharge efficiency	95%	Battke et al., 2013; Faria et al., 2013
Inverter efficiency	95%	Notton et al., 2010; Faria et al., 2013

For the first performing lifetime, investigated with LCA, the emissions related to the manufacturing of the battery pack are partitioned between the battery pack and the vehicle by mass allocation. So, 9.2% of the emissions of the battery manufacturing process is allocated to the battery pack itself while the remaining 90.8% is allocated to all other parts of the vehicle (Janssen et al., 2019). During the battery's first life, no emissions from the electricity consumed propelling the vehicle are allocated to the battery, all is allocated to the vehicle (Janssen et al., 2019).

When extending the performing lifetime of the battery, the allocation issue arises for the burden of manufacturing between the first and second life. The emissions from the manufacturing process may be distributed over the whole performing lifetime of the batteries, not just the first life. Thus, the burden of manufacturing is allocated between the first and second life by their respective energy throughputs. During the first lifetime of the batteries, it was assumed that the batteries had an energy throughput corresponding to 1 performed cycle per day with 75% Depth of discharge over a period of 10 years (Janssen et al., 2019). Value can be seen in table 2. The energy throughput of the second life is investigated in the energy systems analysis. Its results will then determine the allocation factor for burden of manufacturing. The emissions of the PV electricity charged to the battery and the emissions of the avoided electricity are calculated with the energy systems model but is fed into the LCA for the complete assessment of second-life LIBs. It is assumed that all grid emissions avoided by using electricity supplied to the load by the storage can be attributed to the battery since the storage is responsible for the change in the system.

The refurbishment of the batteries takes place in a facility in Halmstad. Once refurbished, the batteries are assumed to be transported to the household where they will serve their second performing life. The average distance to the household where the LIBs are installed is assumed to be 456km, which is the distance from Halmstad to Södertälje (most households are located south of Uppsala). Once transported to the household, the LIBs initiate their second performing life.

Table 2: Inputs to LCA

Input	Value	Source
Burden of battery manufacturing allocated to vehicle	90.8%	Janssen et al., 2019
Burden of battery manufacturing allocated to battery	9.2%	Janssen et al., 2019
First-life energy throughput	2737.5$\mathrm{kWh_{el}/kWh_{NSC}}$	Janssen et al., 2019
Emissions of electricity charged in automotive life allocated to vehicle	100%	Janssen et al., 2019
Transport distance – Refurbishment to usecase location	456km	Assumed value – Halmstad to Södertälje
Second-life energy throughput	Model output	Energy systems model
Related emissions of charged PV electricity	Model output	Energy systems model
Hourly emissions avoided by avoiding consumption of grid electricity	Model output	Energy systems model
LIB second-life duration	Model output	Energy systems model

3.3 Scenarios

To investigate the impact that the initial SoH and the size of the residential PV and storage system have on the energy flows and lifetime, different scenarios were employed. A description of these is given in table 3 below. Scenario 2 can be considered the base scenario of this study while the other scenarios can be seen as sensitivity analyses on installation size and relative remaining storage capacity. Since the RBC is relative to the annual production of PV electricity, see equation 3, a change in PV peak capacity will also change the storage capacity. For the load data used, RBC of 1 is roughly equal to $1.1\mathrm{kWh_{NSC}/kWp}$.

Table 3: Scenario configurations used in the energy systems model. Impact of the initial state of the batteries are investigated in scenarios 1, 2 and 3 while relative system size is investigated in scenarios 2, 4 and 5.

	ALR	RBC	Initial SoH
Scenario 1	3	1	70%
Scenario 2	3	1	75%
Scenario 3	3	1	80%
Scenario 4	2	1	75%
Scenario 5	4	1	75%

4 Results

The second-life emissions found with the energy systems analysis are added to the life-cycle assessment, that was used to find the impacts from the first life, to receive the total emissions of a second-life LIB. These results as well as the net-impact for the different scenarios and emission accounting methods modeled are compiled in figure 5 below.

The allocated emissions between scenarios 2, 4 and 5 where ALR was changed are very similar. The minor increase in utilization of the storage seen for increasing ALR results in slightly larger contributions from the charging and avoided electricity. The difference between scenarios 1, 2 and 3 where initial SoH was altered is considerably larger. This could be expected since the contributions are all related to the energy throughput of the second life, which was found to be mostly dependent on its duration. By average accounting the PV electricity charged has higher emission intensity than the avoided grid electricity. With longer lifetime and higher total charge, this creates a positive trend on the net impact. By marginal accounting, the opposite is true and the high emission intensity of avoided electricity from the marginal technology creates a negatively trending net impact with increasing total charge. These effects can be seen clearly in scenarios 1 to 3. In base scenario 2, the processes prior to second life constitute 30% of positive contributions allocated to a second-life LIB by average accounting and 18.7% by marginal.

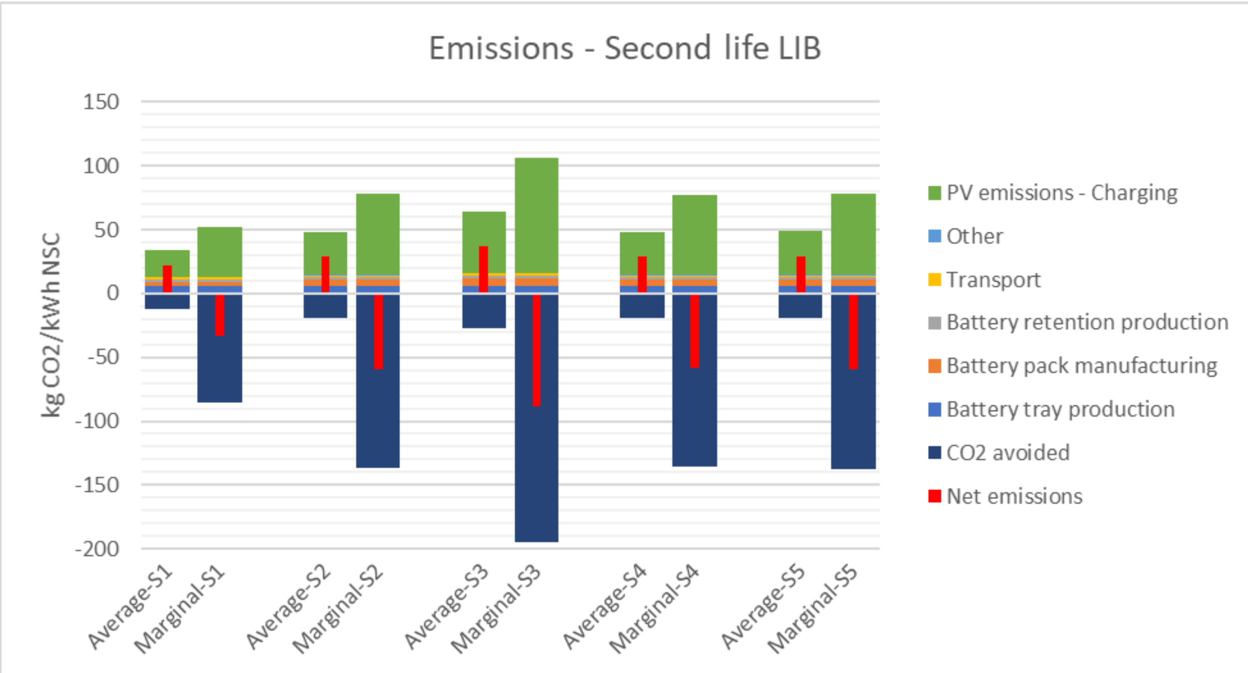

Figure 5: Allocated emissions and net contributions of a second-life LIB for the different scenarios and emission accounting methods employed. The different scenarios are labeled with "S" and their respective number. In scenarios 1, 2 & 3 the initial SoH was altered while in scenarios 2, 4 & 5 the ALR was altered.

After finding the total emissions allocated to the second-life LIB, they can be distributed over the total energy supplied to see which emission intensity the battery storage can supply electricity at. The specific emission intensity which the LIB storage supplies electricity at is the sum of the positive contributions seen in figure 6 below. While charging PV electricity at 41g $\mathrm{CO_2eq/kWh_{el}}$, a second-life LIB storage is found to supply electricity at 62.2-76.2g $\mathrm{CO_2eq/kWh_{el}}$ by average accounting, depending on the scenario. Respectively, by marginal accounting a second-life LIB storage charging PV electricity at 76.7g $\mathrm{CO_2eq/kWh_{el}}$ is found to supply electricity at 101.8-115.8 g $\mathrm{CO_2eq/kWh_{el}}$. From these intensities 16.8-30.8g $\mathrm{CO_2eq/kWh_{el}}$ originates from manufacturing of the LIB which is allocated towards the second life as well as refurbishment and transport. The allocated manufacturing emissions are the same for both accounting methods since it depends on the energy throughput. In figure 5, the total allocated emissions of the second-life LIB are largest for scenarios with a long second-life duration. But as total emissions of the second-life LIB are distributed over its supplied electricity, the emission intensity of supplied electricity is reduced by longer lifetimes.

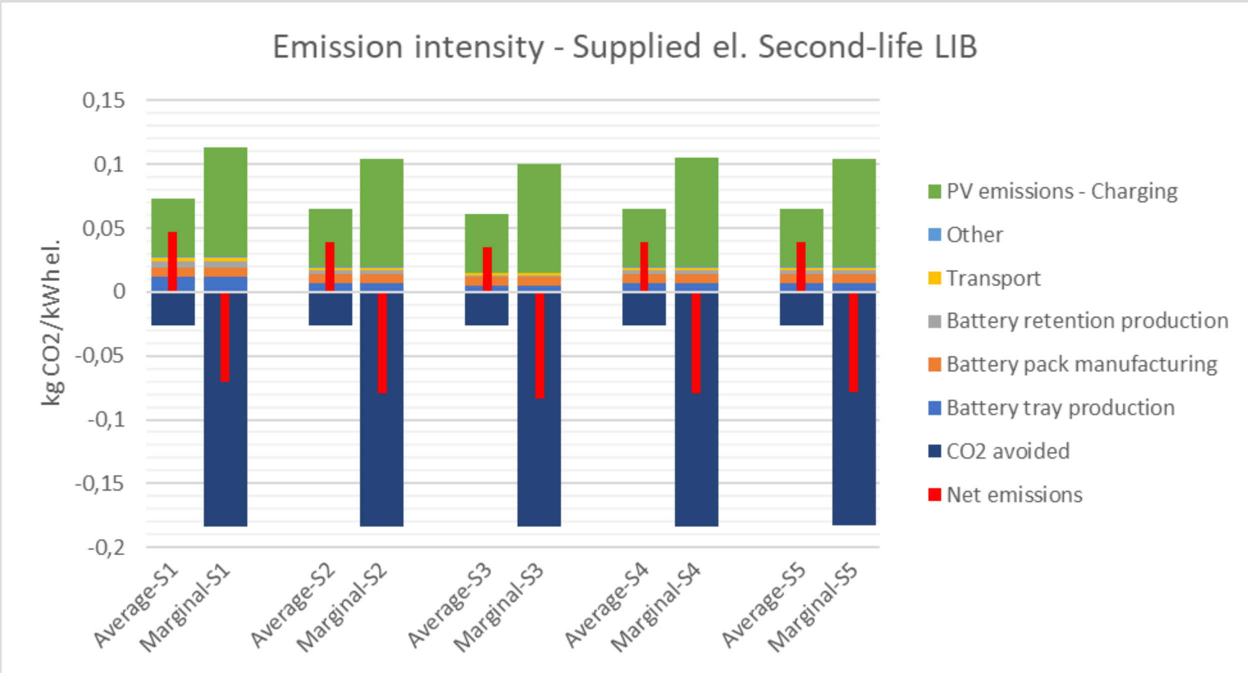

Figure 6: Allocated emissions and net contributions of supplied electricity by a second-life LIB for the different scenarios and emission accounting methods employed. The different scenarios are labeled with "S" and their respective number. In scenarios 1, 2 & 3 the initial SoH was altered while in scenarios 2, 4 & 5 the ALR was altered.

The performance of a second-life LIB storage was found to be largely dependent on the lifetime of the battery. This was in turn mostly dependent on the initial SoH of the LIB as it begins its second performing life and less so on the size of the PV and storage installation it is part of. In figure 7, the second-life total charge is plotted against the duration of the second lifetime. Here, the performance and lifetime dependency can be seen clearly.

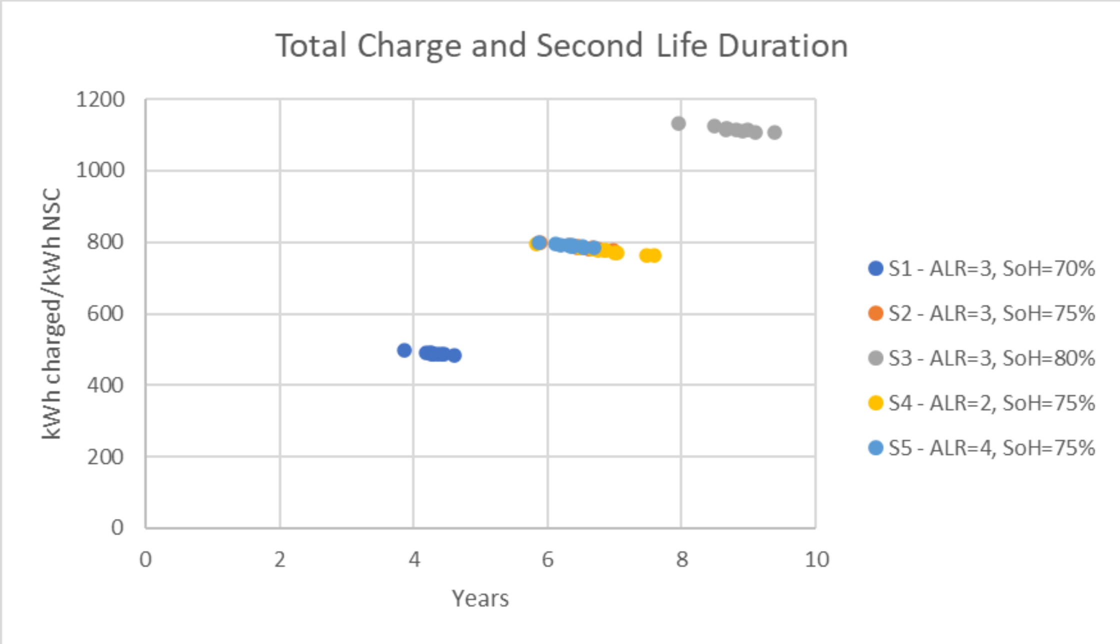

Figure 7: Total charge per kWh_{NSC} in the second lifetime plotted against the duration of the second lifetime. The different scenarios are labeled with "S" and their respective number.

The energy systems analysis was applied in attempt to gather information about the second life use case which may have been lost if only LCA was applied. The utilization of the storage was therefore investigated hourly in the model. By accounting for avoided emissions on an hourly basis, the total was 6.9 to 8.4% lower than if assuming the yearly average emission intensity of the grid for the charged electricity. In figure 8, the PV production, enabling charging of the storage, can be seen to be higher in the summer while the emission intensity of the grid is at its lowest. In winter, the emission intensity of the grid is high while the PV production is low. A majority of the annually stored electricity would then be discharged when the emission intensity of the grid is below its yearly average, causing the difference.

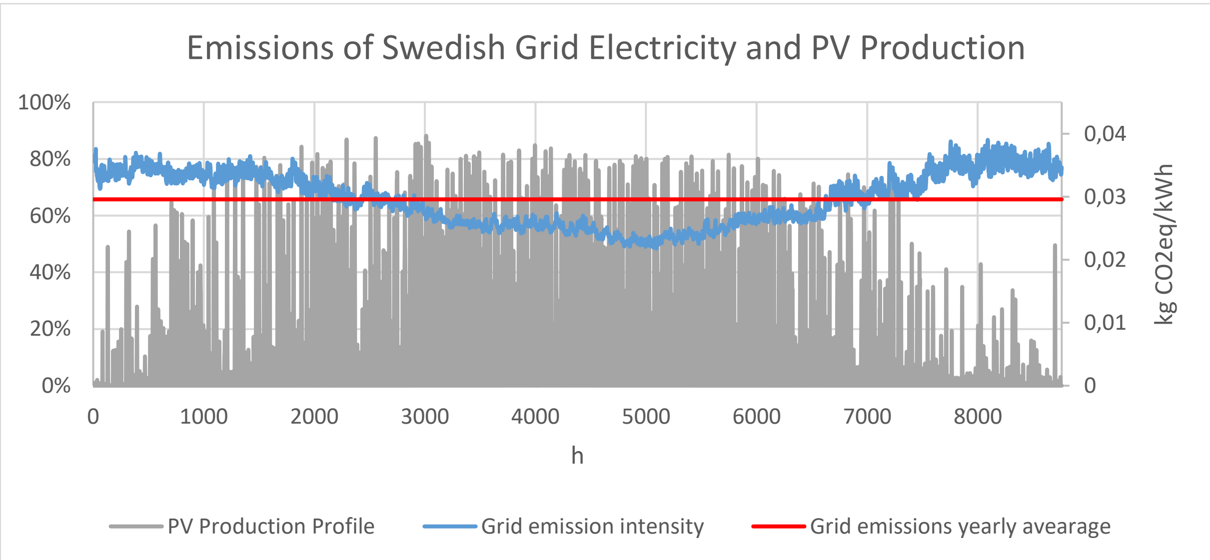

Figure 8: The hourly emission intensity of the Swedish grid ($kg\ CO_2eq/kWh_{el}$) shown with the PV production profile of household 1 (in %) over one year.

4.1 Results - Energy Systems Model

The calculated results from the energy systems analysis are presented in boxplots showing the differences between the 2221 households modeled for the scenarios employed.

To identify the full second-life performance of the LIBs, the second-life duration is needed. These were obtained by modeling the aging and degradation of the LIBs. In figure 9, the LIB's degradation of SoH in the first year modeled can be seen. The degradation can be seen to vary between 1.98-2.60%p/year, depending on which scenario is employed. The annual degradation can in figure 9 be seen to increase with lower initial SoH. This is because the SoH limits the maximum available storage. Thus, a full cycle will occur more frequently at lower SoH leading to more prominent cycling degradation, which can be seen in the increased degradation rising from about 2.25%p/year in Scenario 3 in Figure 9 to 2.35%p/year in Scenario 1. Between scenarios 2, 4 and 5 where installation size was changed, the annual degradation can in figure 9 generally be seen to increase with ALR while the spread decreases. Meaning, as installation size is increased the storages are utilized more frequently, which causes more cycling degradation, and the operation pattern becomes more similar between households. PV production increases with ALR, producing more surplus of PV electricity available for charging the storage. Since the load is fixed, a lesser share of what is produced needs to supply the load when ALR is increased. Therefore, the share of surplus PV electricity increases with ALR. This leads to higher utilization of the storage even though the storage capacity also increases with ALR. Furthermore, PV production surplus increases, the storage approaches its maximum possible utilization where the amount charged is increasingly limited by the storage capacity rather than the available PV production. This would then lead to similar operation patterns for the storages between the households which would explain the decrease in spread with larger installation size seen from scenario 4 to 5 in figure 9. The degradation can for some households be seen to decrease with larger ALR. A possible explanation for this could be that the consumption pattern of these households to a large extent occurs where there is no PV production. So, the surplus of PV electricity would be relatively high compared to the storage capacity at lower ALR. This would cause a smaller storage to go through a high amount of cycles in a year, causing high cycling degradation effects. When increasing the installation size for the households which have a very high share of surplus PV at lower ALR, the relative increase in storage capacity is larger than the relative increase in surplus PV electricity. As a result, the storage goes through less cycles per year at a larger installation size and suffers from less degradation.
From the annual degradation in figure 9, the second lifetimes could be estimated linearly. These can be seen in figure 10.

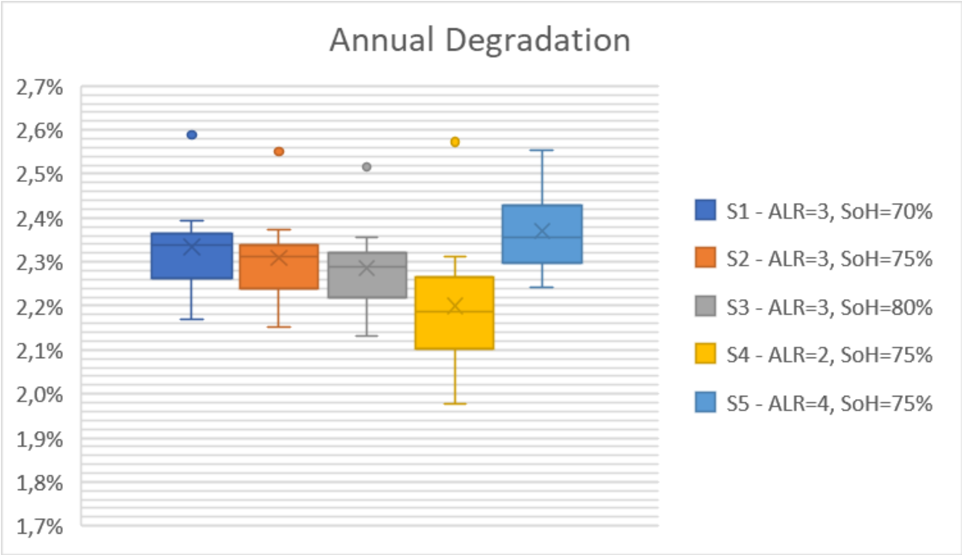

Figure 9: The degradation of nominal capacity during the first year modeled. The different scenarios are labeled with "S" and their respective number.

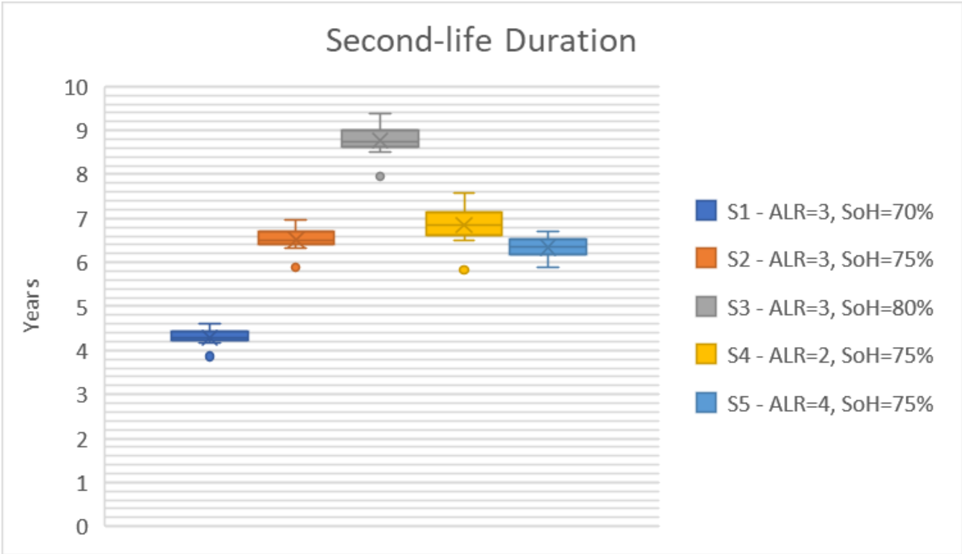

Figure 10: The expected length of a LIB's second performing lifetime. The different scenarios are labeled with "S" and their respective number.

For the scenarios employed, the total specific charge during the second life is found to change mostly with SoH and less with installation size. This is evident in figure 11 where a large increase in total specific charge can be seen from scenario 1 to 3 as initial SoH increases from 70% to 80%. Between scenarios 2, 4 and 5 where installation size was changed, the resulting total charge over the second life is more similar and around 800 $\mathrm{kWh/kWh_{NSC}}$ at 75% initial SoH. As could be expected, batteries with higher initial SoH have a significantly higher total charge during their second lifetime. This is mainly because of longer second lifetimes, allowing them to perform more cycles. Although, higher available storage volumes also have an impact. For the scenarios where the ALR is changed, the total charge increases slightly with ALR. It can also be seen to flatten out and decrease in spread as the installation

size increases. This, again, is because the storages approach their maximum utilization when ALR increases. A similar trend was seen in the annual degradation for these scenarios.
With the results on total charge during the second lifetime, the energy allocation method chosen to distribute the burden of battery manufacturing between first and second life can be made in the LCA. For this allocation, more second-life energy throughput means manufacturing impacts are distributed over more energy and the share allocated to the second life increases. A high initial SoH will then have significant impact on the allocation factor. The effects of ALR on allocation will be smaller, but a higher ALR will lead to a slightly larger allocation to the second life. For the methods chosen, these results indicate that the allocation of burden of manufacturing between first and second life is more determined by the battery's longevity than its second-life use case. The total charge value is also combined with the specific emissions for the solar PV to find the total emissions related to charging the batteries.

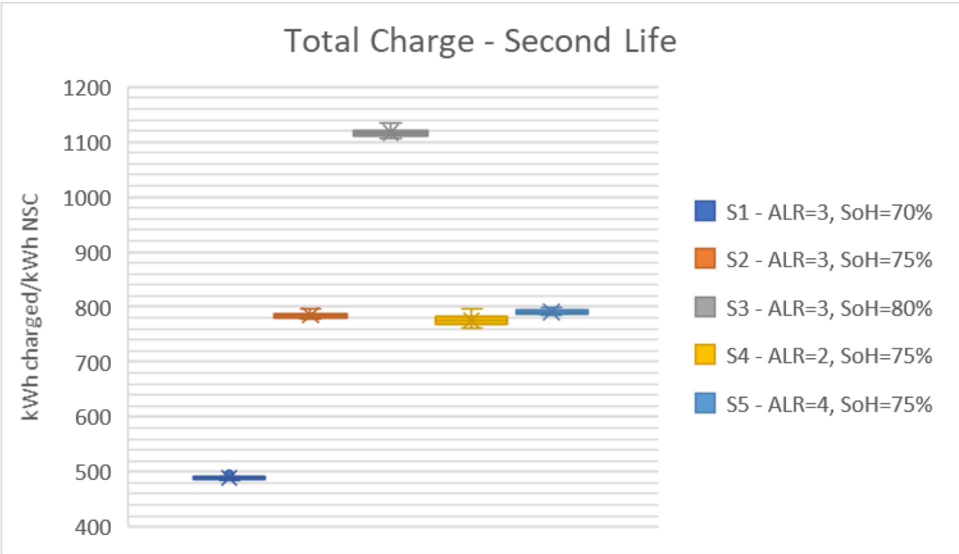

Figure 11: Total amount of electricity charged per kWh of NSC during the second performing lifetime. The different scenarios are labeled with "S" and their respective number.

To assess environmental performance of the second-life LIBs, emissions avoided by marginal and average accounting were investigated hourly in the model. These results were then added to the LCA. The avoided emissions attributed to the battery by average accounting can be seen in figure 12 and by marginal accounting in figure 13. Because the avoided emissions attributed to the battery are calculated by its energy supplied, very similar trends can be seen for avoided emissions by both average and marginal accounting as to what was seen for total charge in figure 11, a high dependency on the second-life durations. Similarly to the total charge, the avoided emissions by both accounting methods trend to flatten out with higher ALR due to the storage approaching its maximum utilization while surplus PV increases. For some household's storages, the avoided emissions by marginal accounting decrease with ALR, whereas this is not the case by average accounting. This trend can be explained by the same reasoning as for the degradation and second lifetime of these households. Consumption hours for these households are mainly in the evening causing higher cycle count for smaller storages. The effect this has on degradation can be seen clearly in figure 9 between scenarios 4 and 5. Because the specific emission intensity is constant by marginal accounting in this study, the reduced utilization per unit of storage capacity seen for these few households when ALR increases causes the avoided emissions decrease with ALR too. The same trend not being seen for average accounting can be explained by the fact that most of the PV production and avoided consumption of electricity occurs in the summer when the emissions of the grid are the lowest in Sweden. So, most of the avoided electricity has relatively little related emissions as opposed to a fixed value as used for marginal accounting. The abundancy of PV and the emission intensity of the grid affecting this result can be seen in figure 8. The reduced emission intensity of the grid during summer is due to less demand, partly because there is no need for electrical heating, and low specific emissions of the cheaper production units.

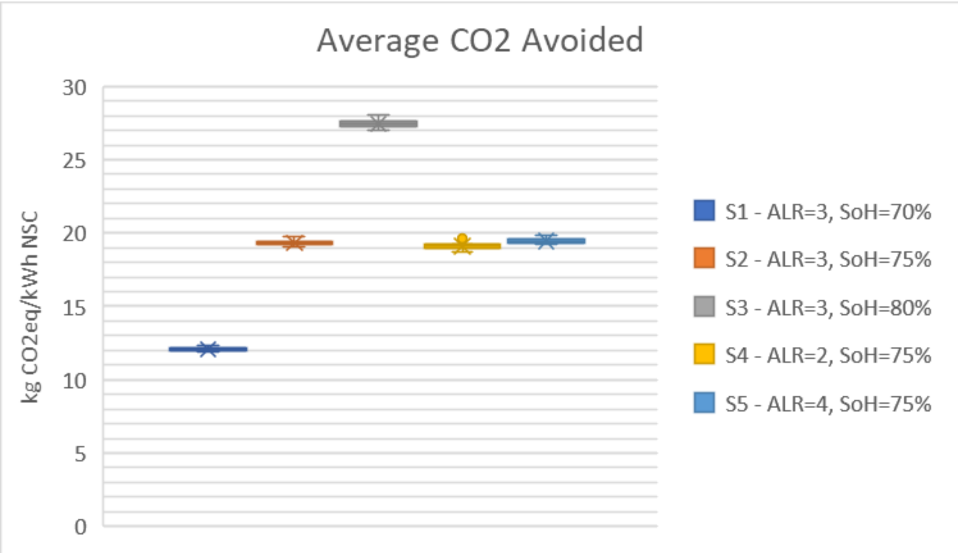

Figure 12: Avoided emissions attributed to the battery alone by average accounting. The different scenarios are labeled with “S” and their respective number.

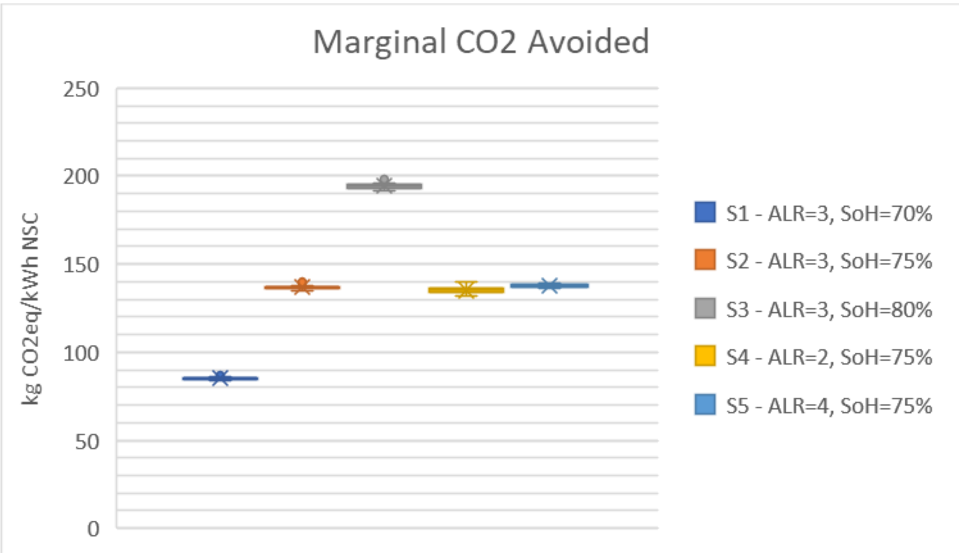

Figure 13: Avoided emissions attributed to the battery alone by marginal accounting. The different scenarios are labeled with “S” and their respective number.

4.2 Results - LCA

After performing the energy systems analysis, the energy throughput of the second life was extracted, enabling energy allocation of the burden of manufacturing between the first and second life. The second-life energy throughput for the storage depends on the initial SoH assumed, installation size, habits of electricity consumption and PV production. The average energy throughput of the storages in the base scenario 2 is 785.3$\mathrm{kWh_{el}/kWh_{NSC}}$ which can then be compared to the throughput 2737.5$\mathrm{kWh_{el}/kWh_{NSC}}$ assumed for the first life (Janssen et al., 2019). Using energy allocation, the resulting share of the burden of manufacturing that should be attributed to the second life can then be calculated to 22.3% for base scenario 2. For all scenarios this value ranges 15-29%, which can also be seen as the share which a second life can relieve the first life from burden of manufacturing. Apart from burden of manufacturing some additional inputs were needed for the LIB to initiate its second performing life. These constitute the second-life LIB's impacts which comes from the production and management of the battery itself and could for base scenario 2 be calculated to 14.54$\mathrm{kg\ CO_2eq/kWh_{NSC}}$. Since the storage operates differently for each scenario, the allocation factor for the burden of manufacturing is also different for each scenario. So, the average total charge of each scenario is used to find the different resulting allocation factors. The contributions for the different scenarios can be seen in figure 14 below.

Tracing the upstream processes beyond surface level, it is estimated that roughly 17.6% of emissions coming from the battery itself are related to electricity consumption and 12.5% to transportation in base scenario 2.

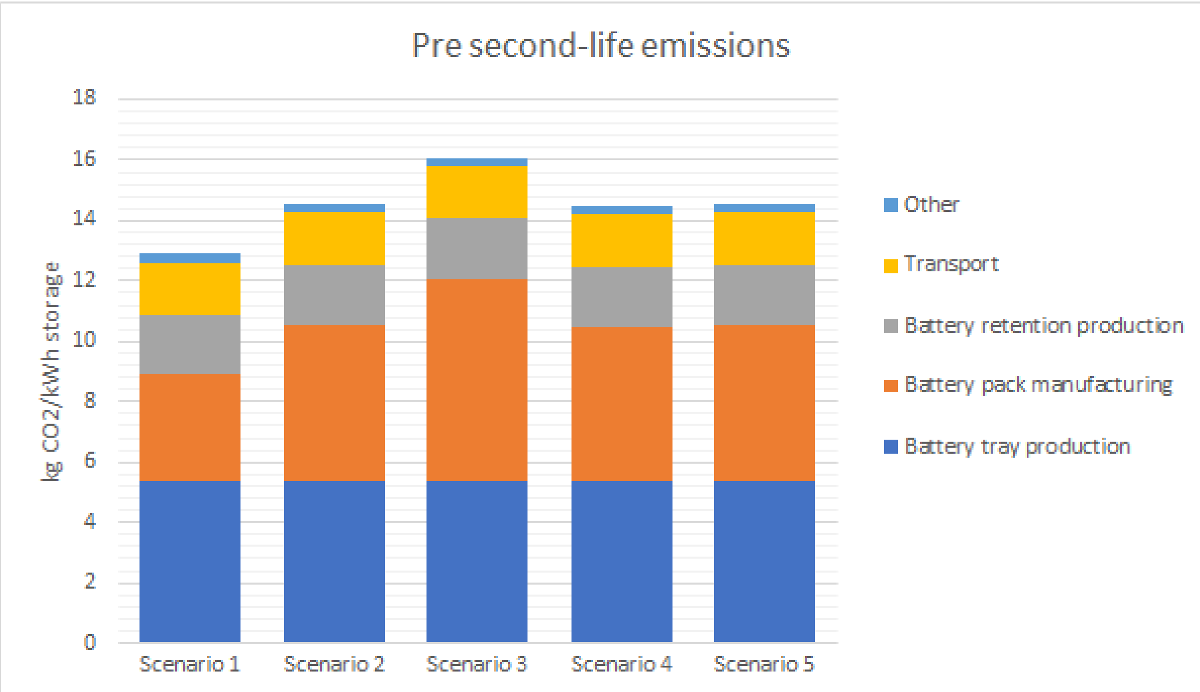

Figure 14: Allocated emissions for a LIB ready to initiate a second performing lifetime in a Swedish household with solar PV. In scenarios 1, 2 & 3 the initial SoH was altered while in scenarios 2, 4 & 5 the ALR was altered.

5 Discussion

The LCA of this study builds upon the LCA performed by Janssen et al. (2019). The same throughput is used for the first life, however, the allocation factors used differ significantly even though results and assumptions on second life are largely similar (52.0% compared to 22.3% for base scenario 2). This is because Janssen et al. (2019) bases the allocation on the energy throughput of the second-life application, which uses 5 batteries with a lifetime of 4 years. In this study, only the second-life energy throughput of the battery determines the allocation. However, Janssen et al. (2019) uses second-life energy throughput for these LIBs as provided by Bobba (2018), which is comparable to the results of this study.

Compared to Bobba (2018), the second-life LIBs modeled in this study stored 24 to 31% less energy per year, resulting in less cycling degradation. A part of the difference can possibly be explained by the latitude difference between Netherlands and Sweden, where these studies were conducted. Nyholm (2016b) identified considerable differences in the potential for PV self-consumption and self-sufficiency for different geographical locations and climates and considers further research regarding this as warranted.

Furthermore, different cultures and climate will also cause different utilization of the storage due to different heating/cooling needs and consumption patterns. The load of household modeled in Bobba's study (2018) is in the range of the modeled households of this study. However, the installation is considerably larger and would by the terms used in this study translate to ALR=11.36 and RBC=0.75. The difference in storage performance between the installation sizes modeled, spanning between ALR 2 to 4, was small. However, since there are considerable differences in results between the studies there is no guarantee that the minor dependence on installation size is true beyond the range modeled.

By the degradation model of this study, the expected second lifetime of Bobba's LIB would have been 4.74 years as opposed to the provided 3.6 years (Bobba, 2018). Hence, there is also a possibility that the second life durations are overestimated in this study. One reason for this could be that the operating conditions chosen for the model does not actually prevent other performance characteristics, like cell-to-cell heterogeneity and internal resistance, from determining the point of end-of-life.

In this study the median value $41\mathrm{g}\ \mathrm{CO_2eq/kWh_{el}}$ (Schlömer et al., 2014) was used for PV by average accounting and $76.7\mathrm{g}\ \mathrm{CO_2eq/kWh_{el}}$ (Jones & Gilbert, 2018) by marginal. The emissions related to c-Si PV manufacture is highly dependent on the emission intensity of the utility used (Alsemma, 2000). This is later reflected in the PV's specific emissions since production emissions are then carried by the total electricity produced. As a result of different assumptions on these parameters, the specific emissions of PV can be seen to vary anywhere from 10 to 100+$\mathrm{g}\ \mathrm{CO_2/kWh_{el}}$ (Schlömer et al., 2014; Alsemma, 2000; Malmström & Olsson Tedin, 2016). Similar effect can be seen for the LIB in figure 6 where the emission intensity of the electricity supplied is reduced from scenario 1 to 3 as the energy throughput increases. PV production in Sweden is very seasonal dependent compared to an EU average, which also means less total production. Hence, the PV values used may be low for a Swedish use case. However, as electricity systems develop, it should be possible to produce PV without fossil resources. In a longer perspective, the PV value used could then be considered high. The same would apply for LIB production. Roughly 18% of pre second-life emissions came from electricity consumption and 13% from transportation. Both having good potential of becoming cleaner with the development of electricity systems and increased electrification. Considering this, the performance of second-life LIBs has good potential of improving in the long-term.

The energy system modeled uses measured historical data which represents the electricity system of 2012. Thus, the development of the electricity system, electricity price, meteorological differences between years and the progress of climate change, affecting the production of VRES, is not accounted for. Sweden has since 2012 been putting efforts into installing more wind power and phasing out nuclear power. An effort which has been increasing in intensity in the recent past. Therefore, these technologies are likely misrepresented in the model. Although the emission intensity used for these technologies are very similar, the change in production mix may have a significant impact. As more wind power is installed, the variability of this technology becomes more apparent in the energy system, which may lead to increased use of the marginal technology or increased need for imported electricity. If the system limitations of the model in this study were expanded beyond the Swedish electricity system and allowed cross border trade of electricity, the marginal emissions could change drastically due to the change of marginal technology. Kristinsdóttir et al. (2013) assessed the emission intensity for consumption of electricity in the Swedish system 2010 and found the average 39g $\mathrm{CO_2/kWh_{el}}$. This was compared to the reported production average 25g $\mathrm{CO_2/kWh_{el}}$ from the same year. It is stated that the difference comes from values used for imported electricity and scope (Kristinsdóttir et al., 2013).

The marginal technology for the model of this study is waste incineration. The main function of these plants is usually the waste reduction service it provides. The products of this service are heat and electricity which can be sold. However, the plants may increase their output for revenue. It is not unlikely that this will occur more frequently in the future since as the share of VRES increases in the grid while nuclear is being phased out. Furthermore, the annual output of hydropower is limited by precipitation. So, the alternative to increased waste output during shortage of renewable energy in Sweden and in Europe could be imported coal energy, which has a larger environmental impact (820g $\mathrm{CO_2eq/kWh_{el}}$ (Schlömer et al., 2014)). Thus, the assumption could be made that electricity from waste incineration produced beyond the waste reduction service is likely to increase in the future. This could then also be assumed to be the first technology to reduce its output when the net load is reduced i.e. the marginal technology.

Nuclear power has frequency stabilizing effects on the grid. As this is currently being phased out, the need for this service may come to increase. Thien et al. (2017) means that frequency regulating services are of high interest to the TSO and could potentially provide high revenue for a battery storage owner. As the shares of VRES, providing electricity at low cost, grow in the energy system, the economic performance of a LIB storage could become more reliant on grid services in the future. However, it is uncertain to what degree the LIB storages can act as relief to the energy system unless a strategy regarding this is employed. Nyholm et al. (2016a) found energy storages to show diminishing returns for increasing storage capacity. Therefore, homeowners are as of now deemed unlikely to invest in storages which exceeds their own needs. However, the utilization of the modeled storages is very low during winter, which could mean there is potential for additional functions like grid services and arbitrage trading. If the events where any considerable revenue can be gained through arbitrage trading is defined as when the price of sold electricity is larger than average spot price+$\mathrm{1SEK/kWh_{el}}$, this is true for only 69 hours in the modeled year. So, a small potential for arbitrage trading exists. Although, since these are all winter hours, where PV production is low, arbitrage trade would likely not interfere much with PV self-consumption.

6 Conclusions

The environmental performance of a second-life LIB operated for economic efficiency for a Swedish homeowner was in this study found to depend largely on its energy throughput, mainly effected by the duration of the second life and less by the installation size.

The emissions from LIB production which were allocated to the second life was done so by energy allocation. A higher cycle count during the second life resulted in a lower emission intensity at which a LIB could supply electricity due to manufacturing impacts being distributed over more energy. By this allocation, a second life was estimated to be able to relieve the first life from 15 to 29% of the burden of manufacturing, which was considered 250$\mathrm{kg\ CO_2eq/kWh}$ nominal storage capacity (Janssen et al., 2019). The difference in storage performance between households were found to be very small. Even though PV production was seen to vary considerably between households the performance of the storages, charging PV electricity, remained relatively indifferent. Thus, the annual performance of LIB storages installed in Sweden can likely be predicted.

By Average accounting the emission intensity of electricity supplied by a second-life LIB storage was calculated to 62 to 76$\mathrm{g\ CO_2eq/kWh_{el}}$. Out of this impact, 17 to 31$\mathrm{g\ CO_2eq/kWh_{el}}$ are allocated manufacturing emissions while remaining emissions comes from charging PV electricity at 41$\mathrm{g\ CO_2eq/kWh_{el}}$. By marginal accounting the LIB could supply electricity at 102-116$\mathrm{g\ CO_2eq/kWh_{el}}$. Out of this impact, PV electricity was assumed to be charged at 76.7$\mathrm{g\ CO_2eq/kWh_{el}}$. With similar energy throughputs, the same amount of manufacturing emissions are allocated towards the second life by this method.

As the storage enters the system, some electricity and its related emissions were considered avoided by the storage. From an average perspective, the avoided emissions were considered to come from the energy mix while from a marginal perspective, from the marginal technology. In this study, waste incineration was considered to be on the margin.

The avoided emissions gave rise to a net impact of the storage. From the average perspective, the introduction of a second-life LIB produced an environmental burden with net impact 22 to 37$\mathrm{kg\ CO_2eq/kWh_{NSC}}$. From the marginal perspective, an environmental benefit was found with net impact -88 to -33$\mathrm{kg\ CO_2eq/kWh_{NSC}}$. The emission intensity the storage is able to supply electricity at is higher than that of the Swedish grid electricity it replaces, causing the net environmental burden. Similarly, replacing electricity on the margin with higher emission intensity causes the net environmental benefit.

In conclusion, the addition of a second-life LIB created an environmental burden from an average perspective while an environmental benefit from a marginal perspective. The environmental burden of a second-life LIB were by both accounting methods mainly constituted by the emissions of electricity charged to the storage. The impacts allocated from processes prior to the second-life were mainly constituted by battery pack manufacturing.

In attempt to better understand the second-life use case of LIBs in Sweden and to capture time dependencies in the energy system, storages were modeled with an hourly resolution using energy systems modeling. Furthermore, grid emissions avoided by the storage were also calculated hourly in the model. By accounting for avoided emissions with average accounting on an hourly basis, the resulting emissions were 6.9 to 8.4% lower than when calculated with total energy throughput and average emission intensity of the grid electricity. This indicates that incorporating energy systems modeling into the LCA added value to the assessment.

The residential storages modeled were utilized relatively little during the winter due to the seasonal dependency of PV production in Sweden. This leaves potential for the storage to have additional functions, like grid services, to provide revenue for the homeowner and relief to the grid, even if this is not the primary intended use and storages are sized only for the energy needs of the homeowner. Thus, models which consider interaction between grid and battery through arbitrage trade and grid services is suggested as further research. So is the interaction between different home energy systems and the value and requirements of supplying grid services

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Appendix

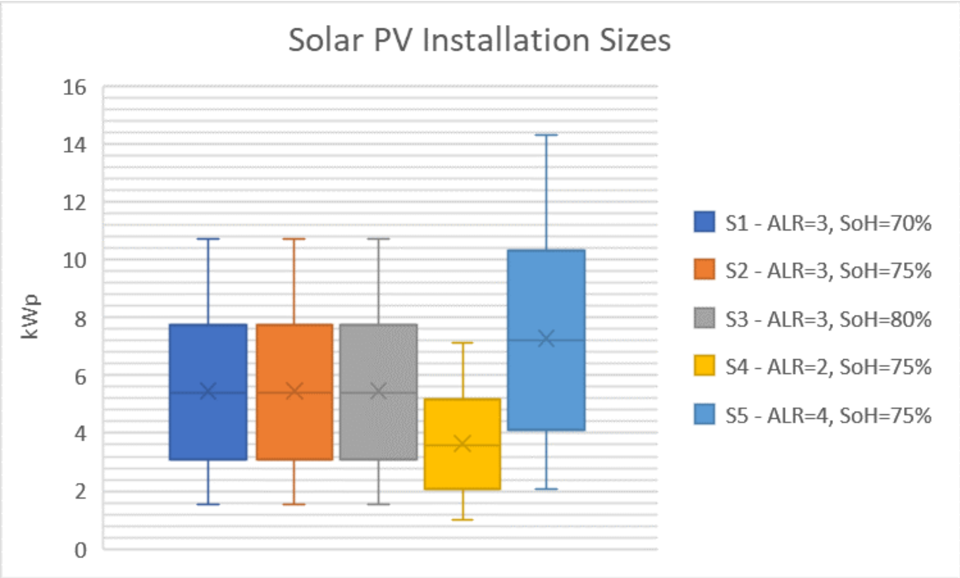

Figure 15: The installed solar PV capacities for the different scenarios.

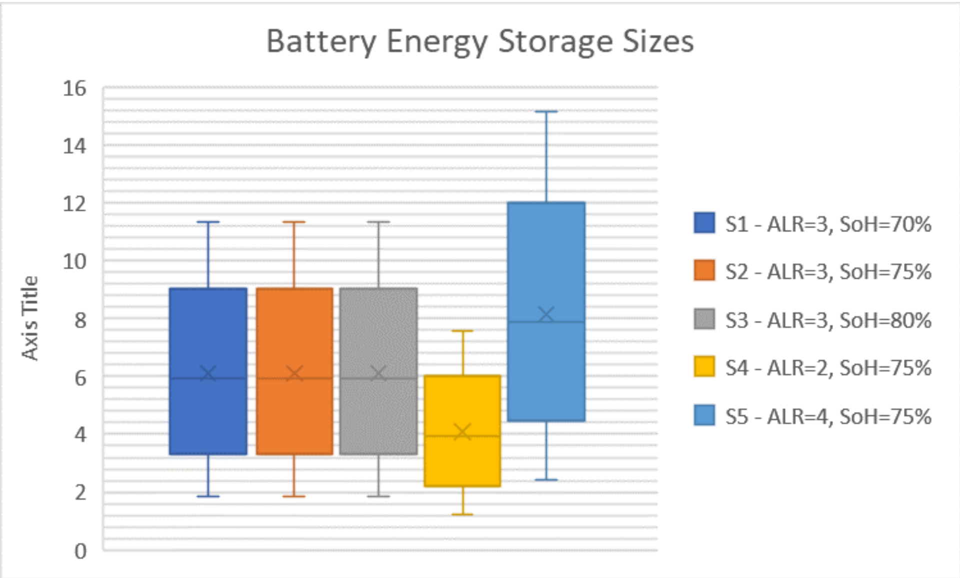

Figure 16: The installed battery storage capacities for the different Scenarios. All households have RBC=1.

For a reference of the installation's effect on the homeowner consumption, the terms self-consumption and self-sufficiency is used. Self-consumption and self-sufficiency are as defined by Luthander et al. (2015). Self-consumption is the share of total PV produced electricity which is used to supply the load of the house and self-sufficiency is the share of the household's total load which is supplied by PV produced electricity. These definitions are visualised in figure 17. Adding batteries to complement the solar panels allows excess PV electricity (produced in region C of figure 17) to be shifted forward in time where it can be used to supply the load. This can effectively increase the self-consumption and self-sufficiency since the consumption of grid electricity can be reduced.

$$\varphi_{sc} = \frac{B}{B+C} \qquad \varphi_{ss} = \frac{B}{A+B}$$

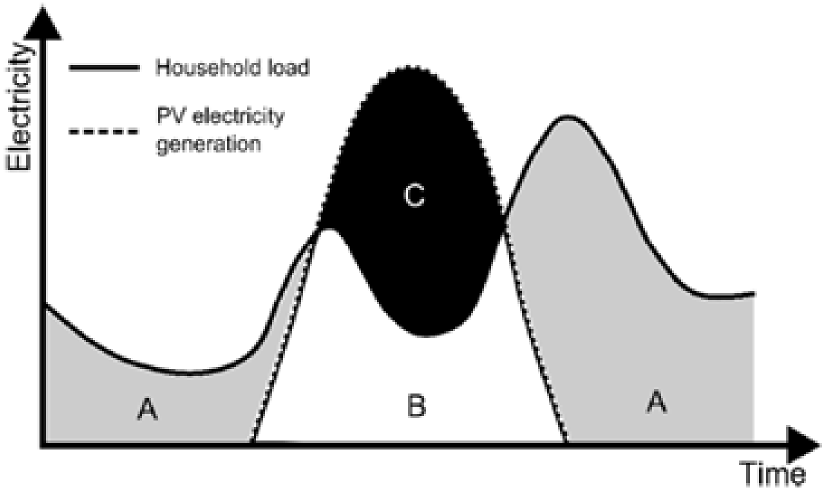

Figure 17: Definitions of self-consumption (SC) and self-sufficiency (SS) as defined by Luthander et al. (2015), illustration by Nyholm (2016b).

The self-sufficiencies and self-consumptions generated in the model can be seen in the following figures 18 and 19 respectively. At smaller installation sizes, a relatively small amount of electricity can be supplied by the PV and storage. So, most of it is used to supply the load and little is sold to the grid, causing the self-consumption to be high and the self-sufficiency low for smaller installation sizes. The opposite can be seen for larger installation sizes. As ALR is increased, the amount of electricity from the PV and storage that can be used to satisfy the load increases, which can be seen as an increase in self-sufficiency. However, as the size of the PV and storage system increases, more electricity needs to be sold to the grid due to higher amounts of surplus production of PV electricity and limited storage capacity. This causes the self-consumption to decrease with higher ALR.

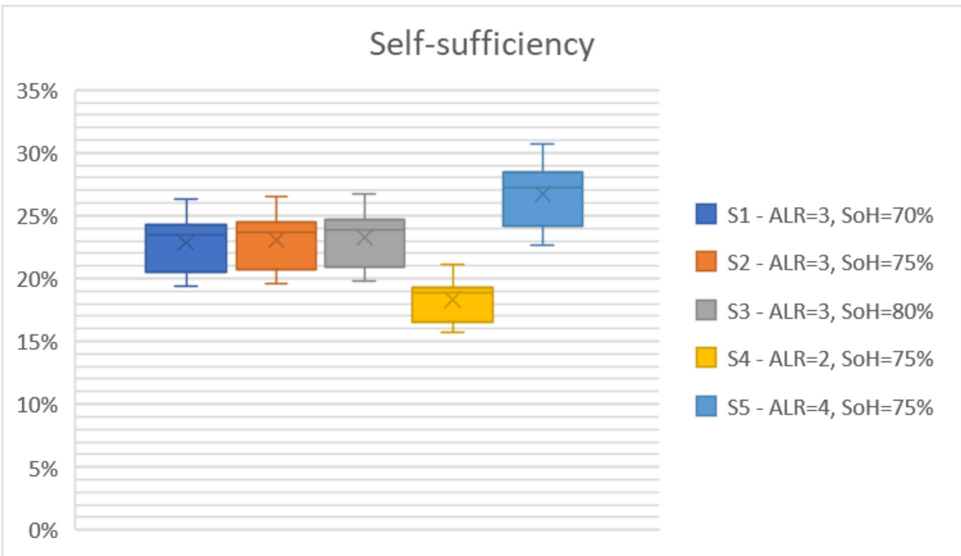

Figure 18: The degree of self-sufficiency of the households. The different scenarios are labeled with "S" and their respective number.

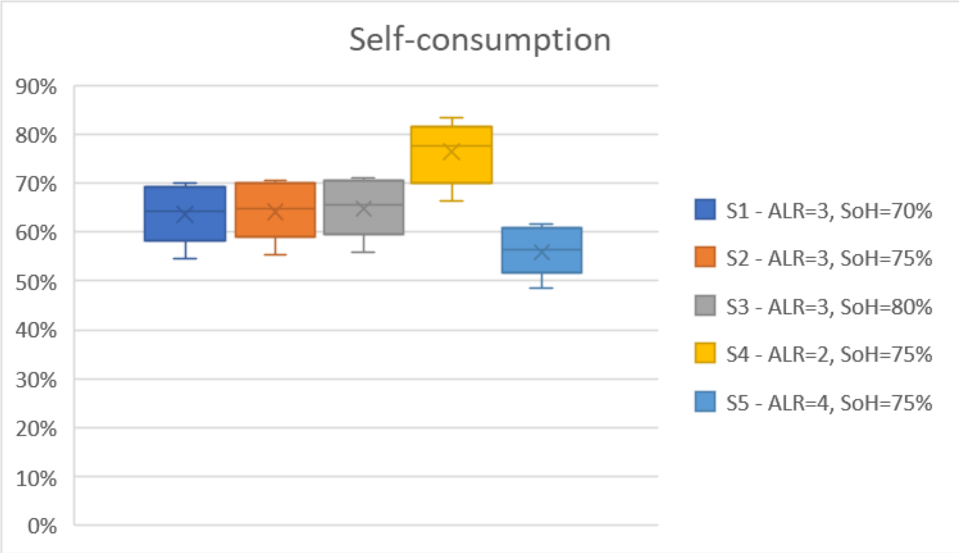

Figure 19: The degree of self-consumption of the households. The different scenarios are labeled with "S" and their respective number.

To investigate the quality of the linear estimations of the second lifetimes, ten households were modeled for several consecutive years. After a model run, each household's final SoH value could be used as its input value for the next model run, starting at the different initial SoH values used in scenarios 1,2 and 3 (70%, 75% & 80%). After the consecutive runs, the point where the batteries reach their end of performing life, i.e. 60% SoH, could be found and compared to what was generated by the linear estimations. The degradation profiles generated for ten households using an initial SoH of 80% (scenario 3) can be seen in figure 20. The model uses the same available storage volume throughout the whole year modeled to keep the model linear and ensure an optimal solution. Since the SoH has an impact on the available storage volume, the cycling degradation is relative to a constant storage volume for the whole year modeled. The degradation profile of household 4, seen in figure 20, deviates slightly from the other profiles with a steeper degradation profile. All households have a similar installation size, defined by the ALR and RBC, relative to their annual consumption. So, household 4 likely has an electricity consumption pattern which causes the storage to be used very frequently, inducing cycling degradation. A possible scenario would be that the consumption to a large extent occurs at hours where there is no PV production, causing large amounts of surplus energy to be stored in the batteries for later use. As a result, the battery works through a large amount of cycles per year.

In the degradation profiles, the increase in spread over time can be seen clearly. This effect was also seen in the linear estimations of the lifetimes, in figure 10.

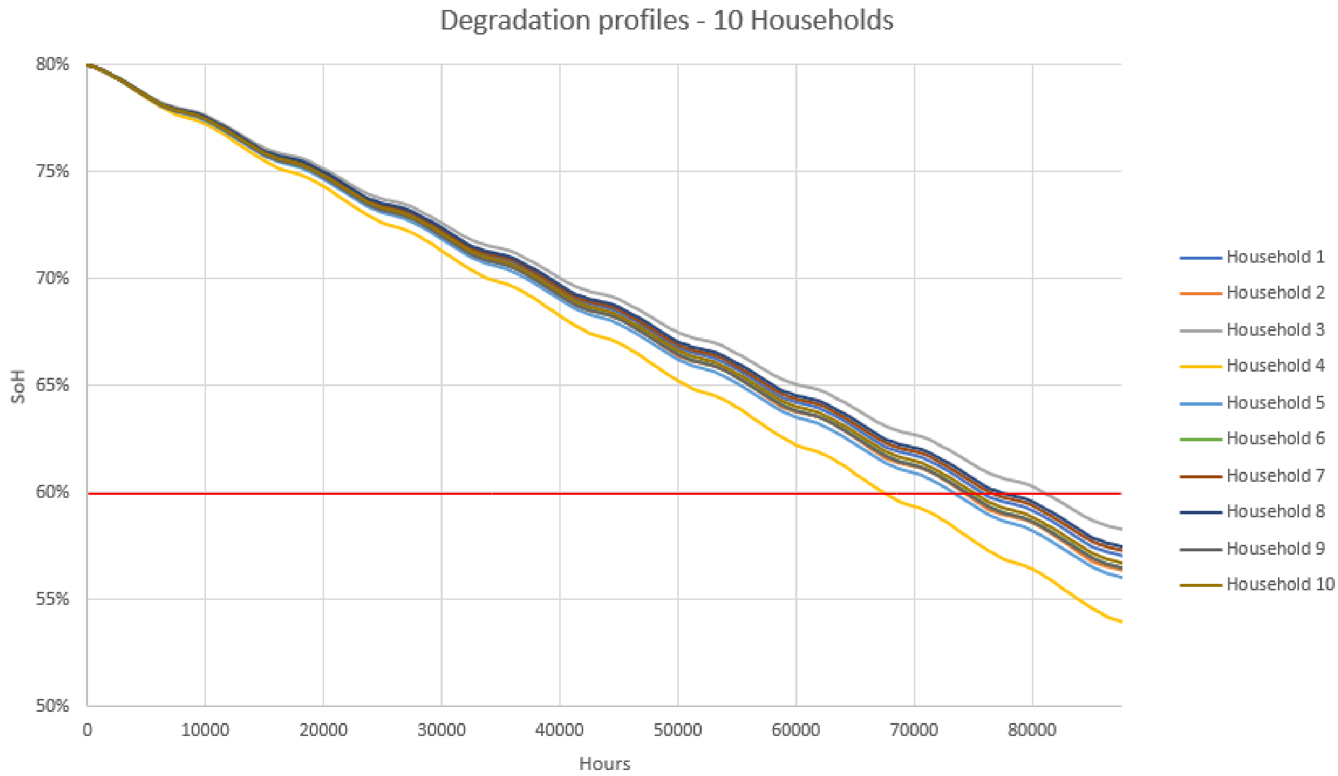

Figure 20: Degradation profiles for the battery storages of 10 households in scenario 3. 60% SoH marks the end of life and is a shown as a red horizontal line at this value.

In figure 21, the offset of the linear estimation of the second lifetime, compared to the results of the consecutive runs, can be seen. The cycling degradation is more prominent at lower SoH due to a larger number of cycles performed as a result of lower available storage volume. This is not accounted for in the model when making the linear estimation. Therefore, the offset can be seen to be larger for higher initial SoH used. Furthermore, it is also the higher frequency of performed cycles which causes the spread to increase for lower initial SoH. The difference in operation of the storages between the households become more evident as the effects of cycling degradation increases, causing larger variation in the quality of the linear estimation. The offset of the linear estimation was found to be 0-3.3% depending on initial SoH. Since a fixed available storage volume is used for each year modeled rather than for each hour, it is not unlikely that these results are slightly overestimated. However, the benefits of accounting for degradation and available storage volume on an hourly basis would likely make a relatively small difference. Furthermore, changing the accounting of SoH to an hourly basis would make the model non-linear. The benefit of making the model non-linear for this purpose is not considered to outweigh the uncertainty of an optimal solution. So, the model is kept linear. However, it should be noted that the found offset is not accounted for in the results of the energy systems analysis. The consecutive runs investigating the offset of the linear estimations were made for validation purposes only.

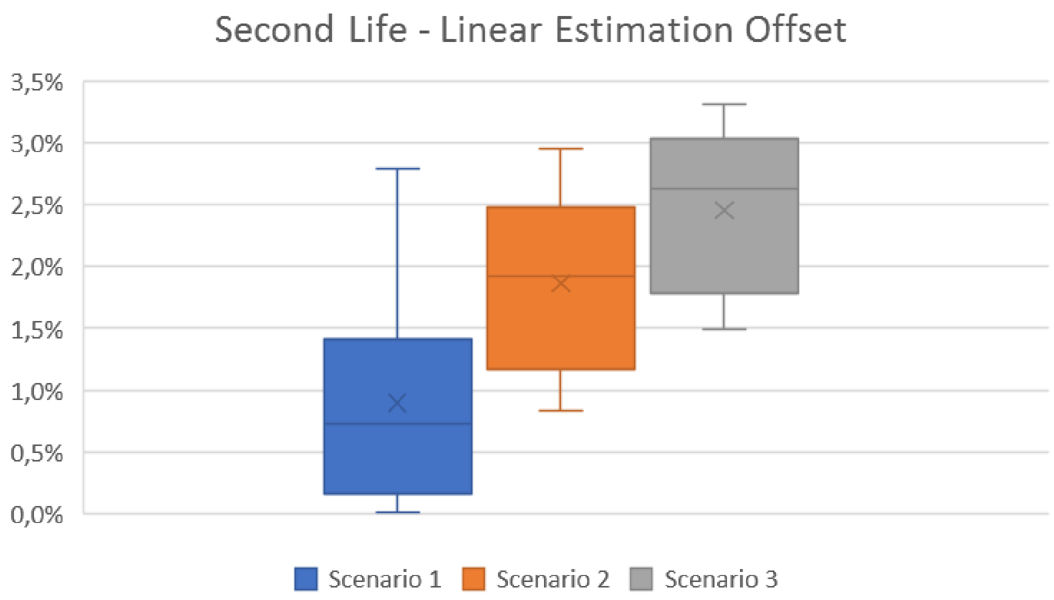

Figure 21: Offset of the linear estimation of the second lifetime compared to their respective degradation profiles derived from the consecutive model runs. Results shown have different assumed initial states of health and represent 10 modeled households.