TECHNICAL UNIVERSITY OF CRETE

SCHOOL OF CHEMICAL AND ENVIRONMENTAL ENGINEERING

PhD Thesis

«Recovery of valuable materials from electronic waste»

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Chemical Engineer, MSc

Chania 2023

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ΠΟΛΥΤΕΧΝΕΙΟ ΚΡΗΤΗΣ

ΣΧΟΛΗ ΧΗΜΙΚΩΝ ΜΗΧΑΝΙΚΏΝ ΚΑΙ ΜΗΧΑΝΙΚΩΝ ΠΕΡΙΒΑΛΛΟΝΤΟΣ

ΔΙΔΑΚΤΟΡΙΚΗ ΔΙΑΤΡΙΒΗ

«Ανάκτηση πολύτιμων υλικών από ηλεκτρονικά απόβλητα»

Ελένη Καστανάκη

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ABSTRACT

As the e-waste generation increases continuously, the estimation of future quantities and the key metals contained in them is of primary importance to policy makers, producers and manufacturers, so as to promote actions and legislations related to their sustainable management. In order to cope with the increasing demand for supply resources, the recycling of discarded devices can ensure the way to sustainability. To assess the feasibility of urban mining, three questions must be answered: What quantities of key metals will potentially be available in discarded devices in the future? What will the economic revenue of recycling be and for which metals is the process financially viable? How can we improve the recycling efficiency of the various key metals? This thesis focuses on estimating future quantities of 4 e-waste categories: **laptops** and **mobile phones** due to their rapid-increasing waste stream, **lithium-ion batteries** from electric vehicles, and **photovoltaic** waste as emerging e-waste. Among the four e-waste categories, PV waste is also chosen to experimentally recover Ag, which can offer significant economic revenue.

This thesis accomplishes a detailed estimation of future End-of-Life (EoL) tablet/laptops amounts in Greece by considering various parameters and clarifying their effect on the estimated e-waste amounts. The content of precious metals (PMs) and critical raw materials (CRMs) in these obsolete e-wastes both for average and dynamic changing parameters is estimated, which revealed the great potential recovery of PMs and CRMs.

Moreover, the waste mobile phone generation and their embedded key metals (CRMs and PMs) in Greece until 2035 (2010-2035), differentiating between feature phones and smartphones are estimated. The quantities of precious and critical raw materials in obsolete smart and feature phones in Greece until 2035 are calculated and it is revealed that effective recycling of obsolete phones (1995-2020) can cover the demand for key metals for new smartphones in Greece for more than a decade.

Considering the rapid promotion of electric vehicles (EV) in the European Union (EU), a new e-waste category is emerging, the lithium-ion batteries (LIBs). The generation of future electric vehicles (EV) battery waste in the EU-27 countries is estimated. LIBs require proper management through circular economy business models. These include Remanufacturing, Reuse and Recycling of LIBs to extend their life before valuable materials are recovered. Material and substance flow analysis with a 3-parameter Weibull distribution function are employed to quantify all battery waste flows and their embedded materials. The available LIBs for remanufacturing and the capacity of Second Life LIBs are calculated. The recovered metals Li, Co, Ni and Cu in the waste LIB are calculated considering the recycling efficiencies of the 2020 EU Battery Directive.

Furthermore, the estimation of future photovoltaic (PV) waste amounts in EU-27 countries considering the targets set by each country in the national energy and climate plans (NECP) for the implementation of solar photovoltaic systems is accomplished. The thesis addresses the questions “when will large amounts of panel waste be generated in the EU countries and what will their composition be?” Also, a timescale for starting an economically viable recycling industry for PV panel waste in EU based on the annual PV waste generated in each country is estimated.

Finally, a novel hydrothermal technique for the recycling of c-Si PV waste panels with a focus on Ag recovery is developed. In the past, research on panel waste recycling has focused on Si, glass and Al recovery, but more recent studies also aim at Ag leaching and recovery, due to the high value of Ag. This thesis investigates Ag and Al leaching from waste monocrystalline and polycrystalline silicon photovoltaic (PV) panels, focusing on both cells and ribbons, by hydrothermal process using mild HNO_3 solutions. Under the range of tested parameters, treatment time was the most important factor, followed by HNO_3 normality and S/L ratio, while process temperature (100-140 ^oC) was not statistically significant. Al leaching was satisfactory under the hydrothermal conditions. Under the optimal hydrothermal conditions Ag can be efficiently leached (100%).

ΠΕΡΙΛΗΨΗ

Λόγω της μεγάλης αύξησης του όγκου των ηλεκτρονικών αποβλήτων, η εκτίμηση των μελλοντικών ποσοτήτων αυτών των αποβλήτων καθώς και των βασικών μετάλλων που περιέχονται σε αυτά είναι πρωταρχικής σημασίας για τους υπεύθυνους χάραξης πολιτικής, τους παραγωγούς και τους κατασκευαστές, ώστε να προωθηθούν δράσεις και νομοθετικές ρυθμίσεις που σχετίζονται με τη βιώσιμη διαχείρισή τους. Προκειμένου να αντιμετωπιστεί η αυξανόμενη ζήτηση σε πρώτες ύλες, η ανακύκλωση των απορριπτόμενων συσκευών μπορεί να εξασφαλίσει τον δρόμο προς τη βιωσιμότητα. Για να αξιολογηθεί η σκοπιμότητα της αστικής εξόρυξης, πρέπει να απαντηθούν τρία ερωτήματα: Ποιες ποσότητες βασικών μετάλλων θα είναι δυνητικά διαθέσιμες σε απορριπτόμενες συσκευές στο μέλλον; Ποια θα είναι τα οικονομικά έσοδα από την ανακύκλωση και για ποια μέταλλα είναι οικονομικά βιώσιμη η διαδικασία; Πώς μπορούμε να βελτιώσουμε την αποτελεσματικότητα της ανακύκλωσης των διαφόρων βασικών μετάλλων; Η διδακτορική διατριβή εστιάζει στην εκτίμηση των μελλοντικών ποσοτήτων 4 κατηγοριών ηλεκτρονικών αποβλήτων: **φορητών υπολογιστών** και **κινητών τηλέφωνων** λόγω της ταχέως αυξανόμενης ροής αυτών των αποβλήτων, **μπαταριών ιόντων λιθίου** από ηλεκτρικά οχήματα, καθώς και **φωτοβολταϊκών απόβλητων** τα οποία είναι μελλοντικά ηλεκτρονικά απόβλητα. Τέλος, από αυτές τις τέσσερις κατηγορίες ηλεκτρονικών αποβλήτων επιλέχθηκαν τα φωτοβολταϊκά απόβλητα για την πειραματική ανάκτηση του Ag, που αν και βρίσκεται σε πολύ μικρές ποσότητες, θα προσφέρει σημαντικά οικονομικά έσοδα.

Αρχικά πραγματοποιείται μια λεπτομερή εκτίμηση των μελλοντικών ποσοτήτων αποβλήτων λάπτοπ/τάμπλετ στην Ελλάδα λαμβάνοντας υπόψη διάφορες παραμέτρους και διευκρινίζοντας την επίδρασή τους στις εκτιμώμενες ποσότητες των ηλεκτρονικών αποβλήτων. Εκτιμάται η περιεκτικότητα σε πολύτιμα (PMs) και κρίσιμα μέταλλα (CRMs) σε αυτά τα ηλεκτρονικά απόβλητα, γεγονός που αποκαλύπτει ότι υπάρχουν μεγάλες δυνατότητες ανάκτησης πολύτιμων και κρίσιμων μετάλλων.

Επιπλέον, υπολογίζονται οι ποσότητες των απορριμμάτων κινητών τηλεφώνων και των ενσωματωμένων βασικών μετάλλων τους (PMs και CRMs) στην Ελλάδα μέχρι το 2035 (2010-2035), διαχωρίζοντας τα τηλέφωνα σε παλαιού τύπου (feature

phones) και smartphones. Υπολογίζονται οι ποσότητες κρίσιμων και πολύτιμων πρώτων υλών σε απόβλητα smart και feature phones στην Ελλάδα μέχρι το 2035, και συμπεραίνεται ότι η αποτελεσματική ανακύκλωση των απόβλητων τηλεφώνων (1995-2020) μπορεί να καλύψει τη ζήτηση βασικών μετάλλων για νέα smartphones στην Ελλάδα για περισσότερο από μια δεκαετία.

Λαμβάνοντας υπόψη την ταχεία προώθηση των ηλεκτρικών οχημάτων (EV) στην Ευρωπαϊκή Ένωση (ΕΕ), μια νέα κατηγορία ηλεκτρονικών αποβλήτων αναδύεται, οι μπαταρίες ιόντων λιθίου (LIBs). Υπολογίστηκαν οι μελλοντικές ποσότητες αποβλήτων μπαταριών ηλεκτρικών οχημάτων (EV) στις χώρες της ΕΕ-27. Οι LIB απαιτούν σωστή διαχείριση μέσω επιχειρηματικών μοντέλων κυκλικής οικονομίας. Αυτά περιλαμβάνουν την ανακατασκευή, την επαναχρησιμοποίηση και την ανακύκλωση των LIBs για παράταση της διάρκειας ζωής τους προτού ανακτηθούν πολύτιμα υλικά. Η ανάλυση ροής υλικού με συνάρτηση κατανομής Weibull 3 παραμέτρων χρησιμοποιείται για την ποσοτικοποίηση όλων των ροών απορριμμάτων LIBs και των ενσωματωμένων υλικών τους. Υπολογίζονται οι διαθέσιμες μπαταρίες λιθίου για ανακατασκευή (για χρήση σε ηλεκτρικά αυτοκίνητα) και για επαναχρησιμοποίηση (σε άλλες εφαρμογές). Τα μέταλλα Li, Co, Ni και Cu που μπορούν να ανακτηθούν από τα απόβλητα LIBs υπολογίζονται λαμβάνοντας υπόψη την Οδηγίας της ΕΕ 2020 για τις μπαταρίες και τις αποδόσεις ανακύκλωσης των μετάλλων.

Επιπλέον, υπολογίζονται οι μελλοντικές ποσότητες φωτοβολταϊκών (ΦΒ) αποβλήτων στις χώρες της ΕΕ-27 λαμβάνοντας υπόψη τους στόχους που έχει θέσει κάθε χώρα στο Εθνικό Σχέδιο Ενέργειας Κλίματος για την εγκατάσταση ηλιακών φωτοβολταϊκών συστημάτων. Η διατριβή απαντά στα ερωτήματα «πότε θα δημιουργηθούν μεγάλες ποσότητες απορριμμάτων ΦΒ πάνελ στις χώρες της ΕΕ και ποια θα είναι η σύνθεσή τους;» Επίσης, εκτιμάται ένα χρονοδιάγραμμα για την έναρξη μιας οικονομικά βιώσιμης βιομηχανίας ανακύκλωσης για τα απόβλητα ΦΒ πάνελ στην ΕΕ με βάση τα ετήσια απόβλητα ΦΒ που παράγονται σε κάθε χώρα.

Τέλος, πραγματοποιούνται πειράματα στην ανακύκλωση φωτοβολταϊκών πάνελ με έμφαση στην ανάκτηση Ag. Στο παρελθόν, η έρευνα για την ανακύκλωση απορριμμάτων πάνελ είχε επικεντρωθεί στην ανάκτηση Si, γυαλιού και Al, αλλά πιο πρόσφατες μελέτες στοχεύουν επίσης στην ανάκτηση Ag, λόγω της υψηλής αξίας του. Η παρούσα διατριβή διερευνά την εκχύλιση Ag και Al από απόβλητα μονοκρυσταλλικών και πολυκρυσταλλικών φωτοβολταϊκών (PV) πάνελ πυριτίου,

εστιάζοντας τόσο σε κυψέλες όσο και σε ημιαγωγούς, με υδροθερμική επεξεργασία χρησιμοποιώντας ήπια διαλύματα HNO_3. Τα αποτελέσματα έδειξαν ότι ο χρόνος επεξεργασίας ήταν ο σημαντικότερος παράγοντας, ακολουθούμενος από την κανονικότητα του HNO_3 και την αναλογία S/L, ενώ η θερμοκρασία διεργασίας (100-140 ^oC) δεν ήταν στατιστικά σημαντική. Η εκχύλιση του Al ήταν ικανοποιητική υπό τις υδροθερμικές συνθήκες. Υπό τις βέλτιστες υδροθερμικές συνθήκες, ο Ag μπορεί να εκχυλιστεί αποτελεσματικά (100%).

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NOMENCLATURE

AAGR	Average annual PV growth rate
a-Si	Amorphous silicon
B2L	Battery’s second life
B2R	Battery to recycle
BBD	Box-Behnken design
BEVs	Battery Electric Vehicles
CdTe	Cadmium telluride
CE	Circular economy
CIGS	Copper indium gallium selenide
COVID-19	Coronavirus 2019
CPV	Concentrator photovoltaics
CRMs	Critical Raw Materials
c-Si,	Crystalline silicon
dMFA	dynamic Material flow analysis
EEE	Electrical and electronic equipment
EL	Early Loss
EoL	End-of-Life
EU	European Union
EV	Electric vehicles
EVA	Ethyl vinyl acetate
EVs	Electric Vehicles
FCEVs	Fuel cell electric vehicles

HDDs	Hard Disk Drives
HL	High Lifetime
HREEs	Heavy Rare Earth Elements
ICT	Information and communications technology
IRENA	International renewable energy agency
L/S	Liquid to solid ratio
LFP	Lithium iron phosphate
LIBs	Lithium-ion batteries
LL	Low lifetime
LMO	Lithium manganese oxide
LREEs	Light Rare Earth Elements
MFA	Material Flow Analysis
m-Si	Monocrystalline silicon
MTTF	Mean Time to Failure
NCA	Lithium nickel cobalt aluminum
NECP	National Energy Climate Plan
NMC	Lithium Nickel Manganese Cobalt Oxides
NMC	Nickel manganese cobalt
NWMP	National Waste Management Plan
OF	Objective Function
OPV	Organic photovoltaics
PCBs	printed circuit board
PET	Polyethylene terephthalate
PHEVs	Plug-In hybrid Electric Vehicles
PMs	Precious metals
POM	Put on Market

p-Si	Polycrystalline silicon
PV	Photovoltaic
RL	Regular Loss
RSM	Response Surface Methodology
S	Stock
SDS	Sustainable Development Scenario
SoH	State of Health
SRMs	Secondary raw materials
SSDs	Solid State Disks REEs
STEPS	Stated Policy Scenario
WEEE	Waste Electrical and Electronic Equipment
Γ	Gamma function
ΔG	Gibb’s standard-state free energy
2L	Second Life
3R	Remanufacturing, reuse, recycling

CHAPTER 1

Introduction

Research topics

As the e-waste amounts are increasing, the estimation of future quantities and the key metals contained in them is of primary importance to policy makers, producers and manufacturers, so as to promote actions and legislations related to their sustainable management. In order to cope with the increasing demand for supply resources, recycling of discarded devices can ensure the way to sustainability. To assess the feasibility of urban mining, four questions must be answered: What quantities of key metals will potentially be available in discarded devices in the future? What will the economic revenue of recycling be and for which metals is the process financially viable? How can we improve the recycling efficiency of the various key metals? On this basis, this PhD thesis focuses on estimating future quantities of prime e-waste: laptops and tablets, mobile phones, LIBs from electric vehicles and PV waste. Last, the experimental recovery of precious Ag from crystalline silicon PV waste, which offers significant economic revenue, is investigated.

E-waste represents an ever-increasing waste stream due to the increasing ownership and numerous applications. E-waste, i.e., smartphones, laptops, etc. are among the most valuable waste because of their extremely high content of numerous key metals, specifically in the printed circuit board (PCBs) and magnets. To estimate the Critical Raw Materials (CRMs) and Precious Metals (PMs) embedded in them, initially estimations of obsolete e-waste quantities are essential. Urban mining is an attractive alternative in dealing with the rising demand for CRMs as many key metals in the scrap e-waste have a concentration multiple times more than that of rich primary ores, especially in the PCBs and magnets.

In Europe, the Waste Electrical and Electronic Equipment (WEEE) Directive states that, from the year 2019, 65% of the average weight of EEE placed on the market in three preceding years or 85% of WEEE generated should be the minimum annual country collection rate. The Directive subdivides the e-waste into six categories and poses separate recycling targets for each category on a mass basis. In order to accomplish the targets, the recyclers, are mostly interested in materials present in great amounts in the waste flow, such as ferrous metals or plastics. In this way, materials existent in small amounts are often neglected (Bigum et al., 2012; Sun et al., 2016).

The European Commission started in 2008 the Raw Materials Initiative in order to encounter the expanding concern of assuring valuable raw materials for the EU economy. CRMs are those materials that combine a high economic significance to the EU with a great risk of supply disruptions. Many of these materials are, at present, only derived from a few countries. Among these, China is the leading provider and consumer of various important raw materials e.g., rare earth metals (REEs), antimony, bismuth, magnesium, etc. Thus, this grows the risk of supply scarcity and susceptibility along the value chain. Moreover, in December 2019 the EU adopted the Green Deal Communication that acknowledges access to resources as a vital security issue to accomplish its aspiration towards 2050 climate neutrality (EU, Final Report 2020).

Especially small WEEE, like laptops, tablets or mobile phones represent a perpetually growing waste stream that contains both valuable and hazardous materials. Considering the large amount of these waste categories, the CRMs and PMs present in them, although in tiny amounts, are substantial. Special focus is, therefore, placed on rapidly growing e-waste i.e., laptops and mobile phones and emerging e-waste i.e., lithium-ion batteries (LIBs) from Electric Vehicles EVs and solar photovoltaic (PV) waste.

Due to the rapid promotion of electric vehicles (EV) in the European Union (EU), a new e-waste category is emerging, the lithium-ion batteries (LIBs). LIBs require proper management through circular economy business models. These include Remanufacturing, Reuse and Recycling of LIBs to extend their life before valuable materials are recovered. Lithium-Ion batteries (LIBs) manufacturing requires many different raw materials, some of which are Critical Raw Materials (CRMs) for the EU due to their high economic importance and associated supply risks (Lebedeva et al., 2017). The potential recycling of LIBs will contribute to the sustainable and secure supply of secondary raw materials (SRMs) in the EU. The economic feasibility of the 3R options for EoL LIBs management is largely dependent on the number of discarded LIBs available for each option, as well as the evolution of prices of new cells and raw materials (Rohr et al., 2017). The assessment of EoL EVs batteries is a key factor for their proper management in the EU-27. Assessments of waste streams (remanufacturing, reuse, recycling) will guide process developments and related infrastructure investments. Different battery technologies in waste streams will improve decision-making regarding their management and the recovery of secondary raw materials. The quantities of valuable materials (Co, Li, Ni, and Cu) that can be recovered from the waste LIBs taking into account the new recycling efficiency targets set by the EU battery Directive (2020) are crucial for their management.

Moreover, the decarbonisation of the energy sector is a priority in the EU and this can be accomplished by mainly investing on renewable sources. Increasing offshore wind production, as well as photovoltaic installations is essential. Solar photovoltaic (PV) panels are one of the fastest-growing future waste streams under the category of large electronic waste (WEEE). It is also one of the most important waste streams, as it contains valuable elements like selenium, tellurium, gallium, molybdenum, and indium (Guo and Yan, 2017). The assessment of future PV waste amounts is of primary importance to plan their efficient management.

A PV waste recycling plant aims to maximize profits from the sales of recovered materials from panel waste, while reducing costs related to processes, stocks, transfers and capital investment. It is crucial, therefore, to develop and create an economically viable and environmentally sustainable recycling infrastructure for the emerging photovoltaic industry in conjunction with the accelerating market of these new technologies. The economic viability of such recycling program is closely associated to the geographical quantities of PV waste, the distance from the recycling plant and the amount of valuable materials for sale.

Finally, considering PV waste, it is estimated that, due to the high economic value of Ag, although it is present in a small amount in PV panels, it contributes significantly to the revenue from recycled panel materials. Particular emphasis is placed on how the recycling efficiency of Ag from PV waste can be improved.

Among PV waste, first-generation crystalline silicon (c-Si) modules have had an 80–90% market share over the last 40 years and will thus dominate the PV waste stream. Thus, the recycling of waste c-Si PV panels has been studied with the aim of recovering Ag, a precious metal (PM), from the waste panels. Although Ag is present in tiny amounts in PV panels, it contributes significantly to the revenue from recycled panel materials, due to its high economic value. Silver will be depleted in the next 50 years thus efficient recycling of this metal is crucial (Hunt et al., 2015; Farrell et al., 2020).The mainly used method for Ag recovery from solar cells is etching or leaching in acidic solution based on nitric acid (HNO_3) at concentrations of 2.3-14 N and temperatures 25-80 ^{o}C. Other studies use electrowinning of Ag with methanesulfonic acid solution as the electrolyte. However, no hydrothermal Ag and Al leaching tests from PV panels have been performed to date, so the present study will fill this research gap, focusing on utilizing weak oxidizing agents to avoid the generation of hazardous waste.

Objectives of PhD thesis

The objectives of this PhD thesis are:

To make a detailed estimation of future EoL tablet/laptop amounts in Greece by considering various parameters and clarifying their effect on the estimated amounts. To estimate the content of Precious Metals (PMs) and Critical Raw Materials (CRMs) in these obsolete e-wastes both for average and dynamic changing tablet/laptop weight in order to reveal the great potential of PMs recovery.

To estimate waste mobile phone generation and their embedded key metals (CRMs and PMs) in Greece until 2035 (2010-2035), differentiating between feature phones and smartphones. To calculate the quantities of critical and precious raw materials in obsolete smart and feature phones in Greece until 2035. To decipher whether the effective recycling of obsolete phones (1995-2020) can cover the demand for key metals in new smartphones in Greece.

To estimate the future electric vehicles (EV) battery waste in the EU-27 countries. Considering the rapid promotion of electric vehicles (EV) in the European Union (EU), a new e-waste category is emerging, the lithium-ion batteries (LIBs). LIBs require proper management through circular economy business models. These include Remanufacturing, Reuse and Recycling of LIBs to extend their life before valuable materials are recovered. Material and substance flow analysis with a 3-parameter Weibull distribution function are employed to quantify all battery waste flows and their embedded materials. The available LIBs for remanufacturing and the capacity of Second Life (2L) LIBs are calculated. The recycled metals Li, Co, Ni and Cu in the waste LIB are calculated considering the recycling efficiencies of the 2020

EU Battery Directive. Two case studies are explored, Germany and France which have the highest EVs adoption in the EU-27. The calculated 2L LIBs capacity in these countries is examined whether it can effectively meet the demand for energy storage for photovoltaic systems in these countries,

To estimate the future photovoltaic (PV) waste amounts in EU-27 countries considering the targets set by each country in the NECP for the implementation of solar photovoltaic modules. To answer the questions "when will large amounts of panel waste be generated in the EU countries and what will their composition be?" Also, to estimate a timescale for starting an economically viable recycling industry for PV panel waste in EU based on the annual PV waste generated in each country.

To recycle PV waste panels focusing on Ag recovery. In the past, research on panels waste recycling has focused on Si, glass and aluminum recovery, but more recent studies also aim at Ag leaching and recovery. To investigate silver and aluminum leaching from waste monocrystalline and polycrystalline silicon photovoltaic (PV) panels, focusing on both cells and ribbons, by hydrothermal treatment using mild HNO_3 solutions.

Structure of PhD thesis

The PhD thesis is comprised of seven chapters. A brief description of the contents of each chapter is presented in this section.

Chapter 1 presents the research topics, the objectives and innovation of the PhD thesis.

Chapter 2 presents the theoretical background and literature review on the research subjects.

Chapter 3 presents a detailed estimation of future EoL tablet/laptop amounts in Greece. Various parameters are considered and their effect on the estimated amounts is clarified. Last, the content of precious metals (PMs) and Critical Raw Materials (CRMs) in these obsolete e-wastes is estimated both for average and dynamic changing tablet/laptop weight, revealing a great potential of PMs and CRMs recovery.

Chapter 4 presents a study on waste mobile phone generation and their embedded key metals (CRMs and PMs) in Greece until 2035 (2010-2035) differentiating between feature phones and smartphones. The quantities of critical and precious raw materials in obsolete smart and feature phones in Greece are calculated until 2035. The effective recycling of obsolete phones (1995-2020) can cover the demand for key metals in the new smartphones for more than a decade in Greece.

Chapter 5 presents the estimation of future electric vehicles (EV) battery waste in the EU-27 countries. Considering the rapid promotion of electric vehicles (EV) in the European Union (EU), a new e-waste category is emerging, the lithium-ion batteries (LIBs). LIBs require proper management through circular economy business models. These include Remanufacturing, Reuse and Recycling of LIBs to extend their life before valuable materials are recovered. Material and substance flow analysis with a 3-parameter Weibull distribution function are employed to quantify all battery waste flows and their embedded materials. The available LIBs for remanufacturing and the capacity of Second Life (2L) LIBs are calculated. The recycled metals (Li, Co, Ni and Cu) in the waste LIB are calculated considering the recycling efficiencies of the 2020 EU Battery Directive.

Chapter 6 presents the future photovoltaic (PV) waste amounts in EU-27 countries considering the targets set by each country in the NECP for the implementation of solar photovoltaic modules. The study answers the questions "when will large amounts of panel waste be generated in the EU countries and what will their composition be?" Also, a timescale for starting an economically viable recycling industry for PV panel waste in EU is estimated based on the annual PV waste generated in each country.

Chapter 7 presents the research on PV waste panels recycling focusing on Ag recovery. The aim was to investigate silver and aluminum leaching from waste monocrystalline and polycrystalline silicon photovoltaic (PV) panels, focusing on both cells and ribbons, by hydrothermal treatment using mild HNO_3 solutions.

Chapter 8 summarizes the main conlusions and outines future research recommendations.

Contribution and novelty of PhD thesis

• Reliable estimation of future generation of EoL laptops/tablets in Greece and the precious metals (PMs) and Critical Raw Materials (CRMs) content in these, which is absent in the scientific literature. The PMs and CRMs estimation in the laptop/tablet waste illustrates the potential of recovering valuable resources that exist in EoL laptop/tablets in Greece, which is missing in the literature.

• Consideration of dynamic changes in parameters like penetration rate, population, laptop/tablet weight and product lifespan, which is lacking in the literature. Most reported works apply simplifications of average product weights and/or average lifespan. Clarification of how population, product lifespan and laptop/tablet weight affect the estimated future quantities and to what extent.

• Assessment of the impact of the coronavirus pandemic on Electrical and Electronic Equipment (EEE) demand and their waste.

• The application of a new approach for the precise determination of time-varying lifespan values (1983-2015) by numerically solving the corresponding mathematical equations as suggested in E-Waste Statistics (Forti et al., 2018) given that high quality data are available instead of adopting a survey method. This approach has not been applied in the literature studies. The survey method is impossible to apply over an extended time period (32 years) and may introduce bias if not appropriately conducted. Such a long period can reveal the lifetime dynamics and the economic and social changes reflected in the products lifespan.

• The estimation of waste mobile phone generation and their embedded key metals (CRMs and PMs) in Greece until 2035 (2010-2035), differentiating between feature phones and smartphones which is absent in the scientific literature.

• A new simple technique for distinguishing between feature and smart phones (when only total sales data are available) is presented that can be easily employed in similar cases in other countries.

• For the first time a dynamic 3-parameter Weibull distribution lifespan is adopted for the electric battery life which considers the year of battery production instead of a uniform lifespan for all batteries independently of production year. This has not been applied to EU studies, as they employ a uniform Normal distribution lifespan for all years or fixed or truncated lifetime or a 2-parameter Weibull distribution.

• The assessment of available EoL LIBs for Remanufacturing, Reuse and Recycling in the EU-27, which is rarely found in the literature.

• The determination of future PV waste amounts in the EU-27 countries considering the targets set by each country in the NECP for the implementation of solar photovoltaic modules, rather than making projections based on historical trends that may not capture the future policies promoting renewable sources. This approach is applied for the first time in the literature.

• The indication of the EU candidate countries for the successful establishment of a photovoltaic recycling plant based on the annual PV waste generated in each country and the proposal of a timescale for starting a viable PV recycling industry in the EU-27 is reported for the first time for the EU-27 countries.

• The hydrothermal Ag and Al leaching from waste PV panels (cells and ribbons) utilizing weak oxidizing agents to avoid the generation of hazardous waste, is applied for the first time in the literature.

Publications

a. Scientific publications in journals

1. Eleni Kastanaki, Apostolos Giannis, Dynamic estimation of future obsolete laptop flows and embedded critical raw materials: The case study of Greece, Waste Management, 2021 (132) 74-85.

2. Eleni Kastanaki, Apostolos Giannis, Forecasting quantities of critical raw materials in obsolete feature and smart phones in Greece: a path to circular economy, Journal of Environmental Management, 2022 (307)114566.

3. Eleni Kastanaki, Apostolos Giannis, Energy decarbonisation in the European Union: Assessment of photovoltaic waste recycling potential, Renewable Enegy, 2022 (192) 1-13.

4. Eleni Kastanaki, Apostolos Giannis, Dynamic estimation of end-of-life electric vehicle batteries in the EU-27 considering reuse, remanufacturing and recycling options, Journal of Cleaner Production, 2023 (393) 136349.

5. Eleni Kastanaki, Emmanuel Lagoudakis, Giorgos Kalogerakis and Apostolos Giannis, Hydrothermal silver and aluminum leaching from mono- and poly-crystalline photovoltaic waste panels, Applied Sciences, 2023, 13(6), 3602.

b. Participation in conferences

1. Eleni Kastanaki, Apostolos Giannis, Energy decarbonization in the European Union: Assessment of photovoltaic waste recycling potential, International Conference on Resource Sustainability (icRS 2021), 19 - 23 July 2021.

2. Eleni Kastanaki, Emmanuel Lagoudakis, Giorgos Kalogerakis and Apostolos Giannis, Hydrothermal silver leaching from monocrystalline photovoltaic waste panels, 1st International Conference on Sustainable Chemical and Environmental Engineering (SUSTENG 2022), 1-4 September 2022, Rethymno Crete.

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CHAPTER 2

Theoretical Background

1. Material flow analysis

Effective management of e-waste is crucial to minimize its negative impact on the environment and human health, as e-waste contains toxic substances such as lead, mercury, and cadmium that can cause harm if not handled properly. Many methods accomplish estimations of e-waste generation in the literature. These involve Material Flow Analysis (MFA) that estimates the Input-Output of materials within the system boundaries. MFA can be static when it involves average values or dynamic when it includes time-varying parameters (Wang et al., 2013; Guo and Yan, 2017; Islam and Huda, 2019a). Two data categories are essential to reliably estimate future e-waste generation: future sales and future lifespan distribution data. The uncertainties embedded in the future sales and lifetime data will affect the future waste estimations, therefore it is important to use reliable methods to obtain reliable data. Regarding obsolete mobile phones generation, researchers have used various ways to estimate future sales, but there are several limitations and uncertainties.

Estimation of product adoption is accomplished by the S-shaped growth models such as the logistic model. It has been illustrated that new technology diffusion, market adoption of durable products and users of subscription services have a sigmoidal growth (Meyer et al., 1999; Sokele, 2008, Yang and Williams, 2009). The logistic model is an extensively used growth model with many useful properties for technological and market penetration forecasting. It has been employed to predict the future sales, possession or waste generation of electronic goods (Yang and Williams, 2009; Dwivedy and Mittal, 2010; Guo and Yan, 2017). Contrary to S-shaped cumulative adoption, adoption per period (sales) is a bell-shaped curve and is proportional to the first derivative of cumulative adoption. Thus, the possession is successfully modelled by the logistic curve while sales should be modelled by a bell shaped curve incorporating the phases of growth-saturation-decline (Sokele, 2008).

Moreover, the Norton-Bass model, describes both adoption and substitution, thus the decline phase is also included (Norton and Bass, 1987). The model was used to forecast adoption of electronic goods such as LCD TVs and desktop display screens (Tsai, 2013; Lu et al., 2015). However, this model best fits when modelling direct replacement by consecutive technology generations is the case, which does not always apply on consumer electronics (Tseng et al., 2009).

In order to forecast the WEEE generation, several time-series analysis methods are employed like exponential smoothing, linear regression and moving averages autoregressive models. These models must consider data variations due to trends like seasonality (Rodrigues and Werner, 2019). However, they exhibit low accuracy in long term predictions (Mentzer and Cox, 1984).

1.1 Material flow analysis: case study mobile phones

In the case of waste mobile phone generation estimations, He et al. (2021) used the historical sales data in India till 2019 and extrapolated the average historical growth rate till 2035 considering the global prediction for coronavirus pandemic affecting annual mobile phones sales in 2020, 2021 and 2022. For smart phones only the last 2-year average historical growth rate was used for the projection, while for feature phones the last 10-year average growth rate. They assumed that growth rates will remain steady and concluded that both smart and feature phones sales will increase in the future (after 2024) till 2036. The accumulated obsolete cellphones in 2009-2019 were calculated at 632 million units (79% feature phones and 21%

smartphones). Future obsolete cellphones generation (2020-2030) was calculated at about 3.34 billion units (51% feature phones and 49% smartphones). Although extrapolation of past sales data trend is a simple method for forecasting, it can be unreliable when significant fluctuations in historical data exist or the purchasing power per person is not taken into account (Balde et al., 2017). Also, the prediction method may fail if the past trend doesn't continue in the future, as is the case with mature technologies substituted by emerging ones. Therefore, it is more suitable for short-term forecasting (Mentzer and Cox, 1984; McCarthy et al., 2006). Similarly, He et al. (2018) used the historical sales data in China till 2017 and extrapolated the 6 year-average historical decline rate till 2035 for feature phones and adopted the annual growth rate reported by the Chinese Academy of Environmental Planning for smartphones. However, as previously referred, the extrapolation of past sales data is more suitable for short-term forecasting. They also accounted for smuggled and counterfeit mobile phones adopting their sales growth from previous studies. In the period from 1987 to 2016, the accumulated obsolete mobile phones were 2.34 billion units for feature phones and 987 million units for smart phones (70.3% feature and 29.7% smart phones). In the period from 2017 to 2035, the cumulative quantity of obsolete feature and smart phones is 339 million units and 1.43 billion units, respectively (19% feature and 81% smart phones).

Polak et al. (2012) used sales data 1996-2010 in Czech Republic obtained from Intrastat and extrapolated them by the logistic curve till 2020. However, observing their data, it is obvious that the mobile phone sales had almost reached a plateau in 2010 (saturation phase) and this plateau was just extended to future years. The S-shaped logistic curve, if applied to sales, describes the product's growth till it reaches saturation and does not capture the decline of sales due to substitution by new technological products (Althaf et al., 2019). They estimated that in 2000–2010, 6.5 million pieces were accumulated and in 2010-2020 about 26.3 million obsolete pieces will be generated. Althaf et al. (2019) also used S-shaped logistic curve to simulate sales data but they used the decay rate after the growth rate and saturation, predicting that sales would drop because new emerging technologies are introduced. They estimated that for mature products future e-waste generation will gradually decrease after reaching a peak. They demonstrated that i.e., CRT and LCD TVs sales will drop, whereas LED TVs sales will become significant in USA.

Guo et al. (2017) used the S-shaped logistic curve to describe mobile procession in China and not sales data. Possession data are successfully described by the logistic curve and maximum capacity is estimated relatively easily considering population and mobile subscription data. In their case, the possession logistic curve for China was at the growth phase (data till 2015). However, there was no distinction between feature and smart phones, and this causes inaccuracies as the feature phones adoption may be reduced and substituted by the rising smartphones. They estimated that 781 million units of obsolete cellular phones were generated in 2015 and predicted 878 and 938 million units will be generated in 2020 and 2025, respectively. Rahmani et al. (2014) used the logistic curve to directly simulate the obsolete mobile phones generation in Iran predicting that obsolete devices will reach a plateau in 2035. They estimated 39 million mobile phones will be discarded in 2014 and 90 million units in 2035. However, the logistic curve is suitable to describe technology diffusion rather than waste generation (Meyer et al., 1999).

The logistic model is an extensively used growth model with many useful applications for technological and market penetration forecasting. It has been employed to predict the future sales and possession or waste amounts of electronic goods (Yang and Williams, 2009; Dwivedy and Mittal, 2010; Guo and Yan, 2017). In the beginning, the growth of the logistic model is identical to the exponential one, but later the gradient slows down as growth approaches the market capacity limit. Contrary to S-shaped cumulative adoption, adoption per period (sales) is a bell-shaped curve and is proportional to the first derivative of cumulative adoption. Thus, the possession is successfully modelled by the logistic curve while sales should be modelled by a bell-shaped curve incorporating the phases of growth-saturation-decline (Sokele, 2008). Table 1 summarizes the findings of the literature review on forecasting methods applied for sales. It is evident that the most suitable method is the adoption of the logistic curve incorporating phases of growth-saturation-decline, which is applied in this work.

Furthermore, the lifespan of the electronic goods is fundamental in MFA calculations. Lifespan can be static or dynamic. Static lifespan doesn’t follow any statistical distribution. However, this estimate is inaccurate as it fails to reflect the dynamic patterns of product life (Babbitt et al., 2009). On the other hand, the dynamic lifespan captures the active changes as it follows a statistical distribution such as the exponential, Rayleigh, lognormal, normal distribution or the Weibull. Nevertheless, all the aforementioned distributions are approximated by the Weibull for different values of the shape parameter (Almalki and Nadarajah, 2014). Therefore, the Weibull distribution is appropriate to describe the products lifespan, as it obtains the best fit for most devices (Walk, 2009). Most studies adopt lifespan data for a specific year from literature sources and neglect the time variation (Yu et al., 2010; Tran et al., 2018; Islam and Huda, 2019b). As the product life cycle for electronic goods is decreasing, this approach is not realistic (Trappey and Wu, 2008). Other studies use the survey method to determine the lifetime of household electronic appliances either neglecting time variance or considering a limited one (Abbondanza and Souza, 2019; Kosai et al., 2020). However, the survey method has some disadvantages as it depends on consumer behaviour (Kang and Schoenung, 2006) and is difficult to estimate the lifespan for an extended period of successive years. For a lifespan survey to be reliable it should be conducted on an annual basis at an extensive national level and at waste-disposal points by recording both the date of device manufacture and disposal (Islam and Huda, 2019b). Nevertheless, it is impractical to apply this approach to estimate the product lifespan variation over an extended period of time. In E-Waste Statistics it is proposed that the most precise approach is to numerically solve the corresponding equations to determine lifespan, given that high quality data [Put on Market (POM) and stock (S] are available to produce realistic outcomes (Forti et al., 2018).

He et al. (2018) used lifetime information for feature and smart phones in China from other studies and applied the shape and scale parameters of Weibull distribution of 2.19 and 3.96 for feature and 2.45 and 2.83 for smart phones, respectively. The feature phones and smartphones "service" lifespan in China was 3.51 and 2.51 years and no other time variation was considered for the period 2000-2035. He et al. (2021) also used lifetime information from previous studies and applied a service lifespan described by the Weibull distribution of 6.5 and 8.5 years for the period of 2010-2035 for smart and feature phones in India, respectively. Rahmani et al. (2014) used a questionnaire survey and estimated "service" a lifespan of 2.9 years depicted by Weibull distribution for mobile phones in Iran without distinguishing between feature and smart phones. The service lifespan was considered stable for the period of 2005-2040.

Guo et al. (2017) also adopted a questionnaire survey and evaluated a “service” lifespan of 1.73 years for mobile phones in China to fit the Weibull distribution. No further time variation was considered (during 1995-2025) or any distinction between smart and feature phones. Polák et al. (2012) followed a questionnaire survey and estimated, by fitting the Weilbull distribution, a “total” lifespan of 7.99 years for mobile phones in the Czech Republic, while the service lifespan was assessed at 3.64 years. No further time variation was considered (during 1995-2020) or any differentiation between smart and feature phones.

Table 2.1 summarizes the literature review data on the prediction of the mobile phones lifespan. It is concluded that the most suitable method is the adoption of the Weibull distribution considering a time variance in shape and scale parameters and making a differentiation in the lifespan of feature and smart phones, which is applied in this research work.

Table 2.1 A literature review on the methods used to evaluate future sales and mobile phones lifespan.

Reference/ country of study	***Future Sales calculation method***	***Advantages***	***Disadvantages***
He et al. (2021)/ India	Extrapolated the average historical growth rate till 2035. For smart phones only the last 2-year average historical growth rate was used For feature phones the last 10-year average growth rate was used.	Simple method for forecasting	• Can be unreliable when significant fluctuations in historical data exist • Can be unreliable when the purchasing power per person is not taken into account • Suitable for short-term forecasting
He et al. (2018) /China	Extrapolated the average historical growth rate till 2035. For feature phones the 6 year-average historical rate was used For smartphones the annual growth rate by the Chinese Academy of Environmental Planning was adopted	Simple method for forecasting	• Can be unreliable when significant fluctuations in historical data exist • Can be unreliable when the purchasing power per person is not taken into account • Suitable for short-term forecasting
Polak et al. (2012)/ Czech Republic	Extrapolated sales data by the logistic curve till 2020	The logistic curve is suitable to describe sales, but phases of growth-saturation-decay must be incorporated.	• If sales have reached saturation phase -plateau (2010) the decline phase should be taken into account instead of extending the plateau (for 10 more years) • The decline phase is ignored in this work: the decline of sales due to substitution by new technological products (smartphones substituting feature phones) is not considered • No distinction between feature and smart phones is considered.
Althaf et al. (2019)/ USA	Used the logistic curve to simulate sales incorporating growth-saturation-decay phases	The logistic curve is suitable to describe sales, phases of growth-saturation-decay must be incorporated	
Guo et al. (2017)/ China	Used the logistic curve to describe mobile procession and not sales.	Possession data are successfully described by the logistic curve	• No distinction between feature and smart phones causing inaccuracies as the feature phones possession may be reduced and substituted by the rising smartphones.
Rahmani et al. (2014)/ Iran	Used the logistic curve to directly simulate the waste mobile phones generation		• The logistic curve is suitable to describe technology diffusion (sales/adoption) rather than waste generation
Reference/ country of study	***Lifespan calculation method***	***Advantages***	***Disadvantages***
He et al. (2018)/ China	Weibull distribution with fixed values for shape and scale parameters adopted from literature.	Weibull distribution is appropriate to describe the products lifespan	• Time variance in shape and scale parameters is neglected during 2000-2035. • Values adopted from literature may not be suitable for the period applied in the study.
He et al. (2021)/ India	Weibull distribution with fixed values for shape and scale parameters adopted from literature.	Weibull distribution is appropriate to describe the products lifespan	• Time variance in shape and scale parameters is neglected during 2010-2035. • Values adopted from literature may not be suitable for the period applied in the study.
Rahmani et al. (2014)/ Iran	Weibull distribution with fixed values for shape and scale parameters calculated by questionnaire survey	Weibull distribution is appropriate to describe the products lifespan	• Time variance in shape and scale parameters is neglected during 2005-2040. • The survey method has some disadvantages as it depends on consumer behaviour and is difficult to estimate for an extended period. • Lifespan differentiation between smart and feature phones is neglected
Guo et al. (2017)/ China	Weibull distribution with fixed values for shape and scale parameters calculated by questionnaire survey	Weibull distribution is appropriate to describe the products lifespan	• Time variance in shape and scale parameters is neglected during 1995-2025. • The survey method has some disadvantages as it depends on consumer behaviour and is difficult to estimate for an extended period. • Lifespan differentiation between smart and feature phones is neglected.
Polák et al. (2012)/ Czech Republic	Weibull distribution with fixed values for shape and scale parameters calculated by questionnaire survey	Weibull distribution is appropriate to describe the products lifespan	• Time variance in shape and scale parameters is neglected during 1995-2020. • The survey method has some disadvantages as it depends on consumer behaviour and is difficult to estimate for an extended period. • Lifespan differentiation between smart and feature phones is neglected.

1.2 Material flow analysis: case study laptops/tablets

As can be seem in Table 2.2, a lot of works deal with historic laptop waste estimations and not future projections. These works use average laptop weights that apply for the period under examination and average lifespan. More recent studies make future laptop waste estimations using material flow analysis and Weibull distribution function for laptop lifespan. In the study of Althaf et al. (2019), the logistic function is used to predict future sales incorporating phases of growth, saturation and decline.

Table 2. 2 Literature studies on laptop waste estimation

Reference	Country	Laptop weight	Metals	Examined period	Average lifespan
Van Eygen et al., 2016	Belgium	Average weight 2.84 kg	Ag, Au, Pd, Ni, Fe, Cu, Al	2013	ND
Kahhat and Williams, 2012	USA	2.7 kg		2010	ND
Agamuthu et al., 2015	University of Malaya, Malaysia	ND		2012	5 years
Duygan and Meylan, 2015	Switzerland	3.5 kg	Ag, Au, Pd, Cu	2011	4 years
Wang et al., 2013	the Netherlands	4.6 kg (1995) 3.7 kg (2005)		1995 and 2005	5 years (1995) 4.7 years (2005)
Althaf et al., 2019	USA	3 kg		2000-2025	4 years

1.3 Material flow analysis: case study Lithium Ion Batteries

In the case of LIB waste estimation, Table 2.3 summarizes the main parameters of the literature studies concerning the EU region, the timeframe, the main assumptions, the system boundary inputs and outputs and compares them to the present study. Most of the existing studies examine battery waste flows with a focus on the entire European continent (Abdelbaky et al., 2021; Drabik & Rizos, 2018) or a specific country (Ireland) (Fallah et al., 2021) or region (Catalonia) (Crespo et al., 2022). There are rarely studies that examine the 27 European countries and correlate the available capacities of second life (B2L) LIBs with and the required battery capacities to support stationary storage for PV Energy production for specific countries like Germany and France, the member countries with the higher EV adoption.

Moreover, only two studies consider the remanufacturing option (but this is examined at a fixed and not a dynamic rate) (Abdelbaky et al., 2021; Bobba et al., 2019) and only one examines the dynamic recycling efficiency for the metal recovery in Catalonia region (Crespo et al., 2022). There is no study adopting the 3-parameter Weibull distribution for the batteries lifetime in the EU region. The different cell technologies are either not considered or only 3 or 5 are examined. Only the present study and the study of Crespo et al. (2022) for the region of Catalonia examine eight different technologies.

Table 2.3. Comparison of this work with literature studies for the EU region on LIB waste. (B: Battery; EB: Electric battery).

.	Region	Reference	Forecast	B lifespan scenario Discard Probability Function (1^{st} use)	B lifespan scenario Discard Probability Function (2^{nd} use)	System boundary	Residual capacity for 2U	Vehicle type	Waste results per B cell technology	Metals & materials	Metals recycling efficiency	B Inflows	EoL Volume
1	Europe	Abdelbaky et al., 2021	2040	normal distribution (μ=9-13 y)	normal distribution (μ=3-6 y)	Remanufacturing (fixed rate) for BEV+PHEV, Reuse for BEV+PHEV, Recycling	BEV+ PHEV 80 %	BEVs, PHEVs	8 (NMC 111, NMC 532, NMC622, NMC 811, NCA, LMO, LFP, advanced and beyond Lithium ion)	Li, Ni, Co, Cu, graphit	not considered	One scenario: Europe EV market share at 22 % of global sales until 2030 . Annual EV sales 2.8 million vehicles (2025) and 5 million (2030).	For recycling: 3 million B /125 GWh (2040). For 2U (2040 annual)>70 GWh
2	EU	Bobba et al., 2019	2005-2035	truncated lifetime: 10 % of the batteries 6 y, 40 % 8y, 40 % 10y, 10 % >12 y	fixed lifetime (either 5 or 12 years)	Remanufacturing (0 and 20 %) for BEV+PHEV, Reuse (0:-20 %:2005-2030) and 70 % for BEV+PHEV, Recycling	BEV+ PHEV 60-80 %	BEVs, PHEVs	5 (NMC 111, NMC 532, NMC622, NMC 811, NCA)	Li, Co	not considered	One scenario: Annual EV sales 4 million vehicles (2030).	2U: 0.5-1.75 million B / 9-34 GWh (2035)
3	Catalonia (Spain)	Sanclemente Crespo et al., 2022	2017-2050	2-par Weibull distribution (lifetime: 10 y and 10-16 y)	Weibull distribution (lifetime: 4 y and 4-12 y	Reuse (10-25 %: 2017-2030) and (10-75 %:2017-2030) for BEV, Recycling	BEV 80 %	BEVs, PHEVs	----	Li, Ni, Co, Cu	dynamic	Two scenarios: Low and High	EoL LIBs by 2030: 2.4-8.4 thousand, by 2040:7.4-55 thousand,
4	Ireland	Fallah et al., 2021	2010-2050	2-par Weibull distribution (lifetime: 11 y)	-	Reuse (0-40 %:2010-2050) and (0-80 %: 2010-2050) for BEV, Recycling	EV 80 %	EV	--	--	--	Three scenarios taking into account regulations	Available for 2U: 413-1387 MWh in 2050
5	U.K.	Kamran et al., 2021	2018-2050	fixed 14 y & 3y for shared mobility cars	5 y	Reuse & Recycling (0-80 % (2020)-99 % (2030)). The study accounts for EoL EVB and EB from grid storage (10 y lifetime)	EV 80 %		3 (NMC622, NMC 811+ LMO)	Li, Co, Ni, Mn	Fixed (94-99 % for all metals)	Three scenarios taking into account penetration of shared mobility	EoL LIBs from EV+ grid storage applications 0.99 million in 2046, 0.1 million 2U
6	Europe	Drabik & Rizos, 2018	2010-2040	fixed lifetime of 8 years	fixed lifetime of 10 years		EV 80 %		----	Li, Co, Ni, Al	Two fixed scenarios and 2 collection rates (65-85 %)	One scenario, 1 million (2020), 2.5 million (2025) and 5 million (2030)	EoL LIBs 1.2 million /46 GWh (2030), 2.6 million/104 GWh (in 2035) and 5.4 million/215 GWh (in 2040)
7	EU-27, Germany, France	present study	2010-2040	3-par Weibull distribution (two lifetime scenarios: 4-8-12 y & 4-10-16 y)	Weibull distribution (8 y)	Remanufacturing (SoH>90 %) for BEV+PHEV, Reuse for BEV+PHEV, Recycling	BEV+ PHEV 80 %	BEVs, PHEVs	8 (NCA, LMO, LFP, NMC111, NMC523, NMC622, NMC811, NMC955)	Li, Ni, Co, Cu	dynamic	Two scenarios: Low (STEPS) and High (SDS) 5.4-10.3 million (2030) EU-27.	EoL LIBs 0.82-1.3 million/24-43 GWh (in 2030), 1.14-3.3 million/ 38-137 GWh (in 2035)

1.4 Material flow analysis: case study solar photovoltaic (PV) waste

As far as solar photovoltaic (PV) waste is concerned, PV panels are one of the fastest-growing future waste streams under the category of large electronic waste (WEEE). It is also one of the most important waste streams, as it contains valuable elements like selenium, tellurium, gallium, molybdenum, and indium (Xu et al., 2018). The assessment of future PV waste amounts is of primary importance to plan their efficient management. Previous works have estimated PV waste in Italy, Spain, Austria, Flanders region in Belgium, Mexico, India, Australia, USA and the OECD countries (Paiano, 2015; Santos et al.,2018; Dobra et al., 2020; Peeters et al, 2017; Domínguez, and Geyer, 2017; Gautam et al., 2021; Mahmoudi et al., 2019; Dominguez and Geyer, 2019; Mahmoudi et al, 2021). The differences in the methodology and the main parameters used in this study compared to the literature are reported in Table 2.4.

A key difference from all previous studies is that this is the first work to estimate future panel waste based on future PV deployment capacity commitments as accurately referred in the NECP of each country and not by projections based on historical trends. Thus, the estimations of future PV waste amounts in this study are more precise as the limitations of forecasts based on historical trends, are overcome. Indeed the latter forecasting method may fail if the past trend changes in the future, as is the case with the promotion of renewable energy technologies in the EU. Second**,** all previous studies used either a fixed lifetime or a probability distribution considering an Early and Regular Loss scenarios as proposed by IRENA (Weckend et al., 2016). This is the first study that, in addition to the previous lifetime scenarios, also examines the scenario proposed by EU WEEE Directive and E-waste Statistics (EU, 2017; Forti et al., 2018). Applying a fixed lifetime is not accurate as the failure pattern of a panel is not constant over time, but rather follows a probability distribution. Indeed, when real life data were evaluated, the use of a lifespan following a probability distribution was more suitable to describe PV panels' failure mode (Weckend et al., 2016; Solar America Board, 2013). The Regular Loss (RL) and Early Loss (EL) scenarios are widely used in the literature but there are no reported works for EU WEEE scenario. The EL and RL scenarios employ a characteristic lifetime of 30 years, while the EU-WEEE adopts 25 years resulting in average PVs lifetimes of 26.6, 27.7 and 22.5 years, respectively. The last scenario adopts a shorter lifetime, but this can reliably model the PV panel failures in conditions of transportation, installations and use in the EU.

Third, the waste per PV technology is not considered in the case of the EU countries such as Spain, Austria and Belgium (Santos et al., 2016; Dobra et al., 2020; Peeters et al., 2017)9. In the case of Italy, only five different technologies (c-Si, a-Si, Cd-Te, CISG, CPV) are considered (Paiano, 2015), as opposed to eight in the current study (c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other). The evaluations for India and Mexico consider only three to five different technologies (Domínguez and Geyer, 2017; Gautam et al., 2021) and only the assessments for Australia and OECD countries consider eight technologies (Mahmoudi et al., 2019; Mahmoudi et al., 2021). As these PV panels have different structure and materials, different approaches are required regarding processing, recycling, and treatment to recover the valuable components. A quantification analysis of the respective shares is important for the PV-recyclers. Thus, the current work presents significant information for all EU countries. Fourth, this study considers 22 different metals and materials including critical metals (e.g., gallium, indium and tellurium), precious metals (e.g., silver), and toxic metals (e.g., cadmium, lead, selenium). Previous studies for EU countries either don’t assess any material or evaluate less metals and materials (5-14 materials). The studies for non-EU and OECD countries also assess 21-22 metals and materials (Mahmoudi et al., 2019; Mahmoudi et al., 2021). Fifth, the conversion from installed PV power to mass is either fixed or fixed per technology or panel architecture in most studies, not accounting for the time factor that includes the technological improvement in the panels’ efficiency and the material savings in new panels. In the current study (and in the case of Spain (Santos et al., 2018)), the conversion factor (tonnes per MW) was fitted to an exponential decay function as proposed by IRENA (Weckend et al., 2016). This makes the estimations more accurate and adds value to the presented results. Sixth, the present study and the OECD countries study are the only ones that provide a timescale for launching an economically viable recycling industry for PV panel waste. In the past, the low PV panel waste amount was the main reason behind the unsuccessful attempt of the German recycling company Solar World (Klugmann-Radziemska et al., 2010). For a recycling industry to be successful it needs reliable predictions on panel waste amounts, as well as their composition.

This will eliminate the investment risks associated with operating a recycling industry (Peeters et al., 2017).

Table 2 .4. Comparison of this work with literature studies on solar photovoltaic (PV) waste.

	Country	Reference	PV lifespan scenario	Waste results per PV technology	PV waste	Forecast	Power to mass conversion	Metals and materials
1	**Italy**	3	fixed 25 years	c-Si, a-Si, Cd-Te, CISG, Emerging-CPV	PV modules	2050	fixed per technology	14 (Al, Si, Cu, Sn, Pb, Zn, Ag, In, Ge, Se, Ga, Cd, glass, EVA)
2	**Spain**	4	Regular Loss, Early Loss	no data	PV modules	2050	exponential decrease**	5 (Al, Si, Cu, Ag, glass)
3	**Austria**	5	Regular Loss, Early Loss	no data	PV modules	2050	declining ***	no data
4	**Flanders Belgium**	6	fixed 20-35 years for different scenarios	no data	PV modules	2040	fixed per different PV panel architectures	8 (Cu, Ag, Si, Al, EVA, glass, PET, ABS)
5	**Mexico**	7	fixed 30 years	c-Si, a-Si, CdTe, CIGS, Other	BOS*	2045	fixed	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)
6	**India**	8	fixed 25 & 30 years, Regular Loss, Early Loss	Si-based, CdTe, CiGs	BOS*	2047	fixed per technology	21 (Ag, Au, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb,Ta, Se, Mg, Ga, In, Te, Si)
7	**Australia**	9	fixed 30 years, Regular Loss, Early Loss	c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other	PV modules	2060	fixed per technology (Si-based, CdTe, CiGs)	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)
8	**USA**	10	25-30 years (fixed)	no data	BOS*	2050	fixed per technology	21 (Ag, Au, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb,Ta, Se, Mg, Ga, In, Te, Si)
9	**OECD**	11	Regular Loss, Early Loss	c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other	PV modules	2058	fixed per technology (Si-based, CdTe, CiGs)	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)
10	**EU-27**	this study	Regular Loss, Early Loss and EU-WEEE	c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other	PV modules	2050	exponential decrease**	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)

**BOS: modules, inverters, transformers, **adopted from [12].*

2. Leaching and recovery of valuable metals from PV panels

Regarding the recycling of waste PV, there is a focus on Ag recovery from c-Si waste panels. Table 2.5 summarizes the leaching conditions used in various studies on Ag leaching from PV panels. The most common method for Ag recovery from solar cells is etching/ leaching in acidic solution using nitric acid (HNO_3). Various pre-treatment techniques are applied for EVA removal, then milling and sieving. As seen in Table 1, the optimized HNO_3 concentration varies from 2.3-14 N, temperature from 25-80 °C and time from 2 h to complete dissolution.

Various Al leaching conditions are also found in the literature. Theocharis et al. (2022) reported that the use of 3 N HNO_3 for 2 h at 25 ^oC could quantitatively leach Al from PV powder and flakes. However, Chen et al. (2021) leached 76% of Al using 5 M HNO_3 for 1h at 80 ^oC, as complete dissolution of Al was not possible due to Al-Si alloy formation. Since HNO_3 could also leach Ag, if Ag is to be electrowon, the simultaneous extraction of Al and Ag should be avoided. In this case, selective dissolution of Al can be accomplished by H_2SO_4 (12 M, 3h, 25 ^oC) (Theocharis et al., 2022; Park and Park, 2014).

After successful leaching of Ag, recovery of metallic Ag can be achieved by electrochemical precipitation, in case the concentration in the solution is high enough (Kuczyńska-Łażewska et al., 2018), or by chemical precipitation and subsequent reduction of the silver. For the electrochemical silver precipitation, Dias et al. (2016) processed 50 mL of leached solutions using a platinum plate (7 cm x 2 cm), a steel plate (7 cm x 2 cm) with a density of 60 A/cm^2, and space between the electrodes 30 mm for 1 h. For the chemical precipitation of Ag, 37% HCl was used. Then, AgCl was converted to Ag metal powder, first reacting with an aqueous NaOH solution (2% wt.) while stirring at 200 rpm for 1 h, followed by the addition of H_2O_2 (30% wt.) and further reaction for 0.5 h. The recovered Ag metal powder can be manufactured into an ingot using a thermal torch (Yang et al., 2017). Alternatively, high purity Ag powder can be obtained by ammonia dissolution – hydrazine hydrate reduction of the AgCl precipitate (Luo et al., 2021).

Table 2. 5. Leaching conditions for Ag dissolution from PV panels.

Reference	Leaching agent	Temperature (°C)	Time (h)
Kuczyńska-Łażewskaa et al., 2018	3 M HNO_3	50	3
Yang et al., 2017	90:10 methanesulfonic acid (99 wt.%):H_2O_2 (30 wt.%)	25	4
de Oliveira et al., 2020	2.3 N HNO_3	55	2
Chen et al., 2020	5 N HNO_3	80	1
Dias et al., 2016	13.8 N HNO_3	25	2
Huang et al., 2017	2.6 N HNO_3	60	until complete dissolution

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CHAPTER 3

Dynamic estimation of future obsolete laptop/tablet flows and embedded critical raw materials: the case study of Greece

The coronavirus pandemic has turned school and university learning system from classroom-based to exclusively online all over the world. As this change is accompanied by a spike in demand of laptops, an excessive amount of obsolete devices will be witnessed in the near future. Laptops are the most valuable e-waste category containing a high content of numerous critical raw materials, thus their waste management is crucial. Considering the impact of the coronavirus pandemic on the laptop lifespan, the future quantities and pieces of obsolete laptops in Greece are estimated (2016-2040), as well as the critical raw materials (CRMs) and precious metals (PMs) embedded in them, to illustrate the potential of recovering useful resources, thus contributing to a circular economy. To this end, dynamic material flow analysis is adopted, lifespan distribution is evaluated and future sales are predicted by the logistic model utilizing a bounding analysis. Then, the future End-of-Life (EoL) laptop quantities are estimated taking time-varying parameters into consideration such as penetration rate, population, laptop weight and lifespan. This study is a dynamic estimation that avoids using average values adopted from literature that are not country specific. The provided information is useful for implementing national plans, improving the management of the most valuable category, EoL laptops, enhancing resources efficiency and contributing to a circular economy. The coronavirus pandemic has a similar impact on laptop sales in other countries, affecting their future laptop waste as well.

1. Overview

We are living in an era affected by the coronavirus pandemic which rapidly transformed the educational system from classroom-based into a virtual learning environment. In Greece, the majority of university lectures and school lessons are currently still held online. This change in the education mode has driven all students but also all kinds of workers to go online. Globally, this technology demand is accompanied by a rise in computer sales especially laptops, notebooks and tablets. The estimated impact on notebook computer shipments worldwide due to the coronavirus (COVID-19) outbreak in February and the whole of 2020 is 5.7 and 150.1 million units, respectively, indicating an increase in sales (Statista, 2020a). In Italy, in the first week of May 2020, the sell-out value of laptops increased by 154% compared to the same period of the previous year. Similarly, tablets sales in Italy grew by 61% (Statista, 2020b). In Greece, 8,400 laptops and tablets were distributed in 2020 to schools to cover the emerging needs. As this spike in demand for laptops, notebooks and tablets has left stores with empty selves, an excessive amount of obsolete devices will be witnessed in the near future. This study focuses on Greece, but the impact of the pandemic on laptop sales will affect the generated laptop waste in other countries as well.

In Europe, the Waste Electrical and Electronic Equipment (WEEE) Directive states that, from the year 2019, 65% of the average weight of EEE placed on the market in three preceding years or 85% of WEEE generated should be the minimum annual country collection rate. The Directive subdivides the e-waste into six categories and poses separate recycling targets for each category on a mass basis. In order to accomplish the targets, the recyclers, are mostly interested in materials present in great amounts in the waste flow, such as ferrous metals or plastics. In this way, materials existent in small amounts are often neglected (Bigum et al., 2012; Sun et al., 2016). Especially small WEEE, like laptops, tablets or mobile phones represent a perpetually growing waste stream that contains both treasured and hazardous materials. Considering the large amount of these waste categories, the CRMs and PMs present in them, although in tiny amounts, are substantial.

The European Commission started in 2008 the Raw Materials Initiative in order to encounter the expanding concern of assuring valuable raw materials for the EU economy. CRMs are those materials that combine a high economic significance to the EU with a great risk of supply disruptions. Many of these materials are, at present, only derived from a few countries. Among these, China is the leading provider and consumer of various important raw materials e.g. rare earth metals (REEs), antimony, bismuth, magnesium, etc. Thus, this grows the risk of supply scarcity and susceptibility along the value chain. Moreover, in December 2019 the EU adopted the Green Deal Communication that acknowledges access to resources as a vital security issue to accomplish its aspiration towards 2050 climate neutrality (EU, Final Report 2020).

CRMs are extensively used in electrical and electronic equipment (EEE), particularly in high-tech devices. The renewed 2020 CRMs list includes 30 metals among which are Cobalt, Palladium, Platinum, Neodymium, Praseodymium, Dysprosium Antimony, Yttrium, Indium and Tantalum, all embedded in laptops. Laptops, notebooks and tablets, (together with desktops and servers), are the most valuable WEEE because of their extremely high content of numerous key metals in some of their main sub-systems. Indeed the number of CRMs and PMs embedded in them surpasses other e-waste categories (Cucchiella et al, 2015). Especially the Nd and Ta concentration in notebooks/laptops units is the highest among all EEE. Interest for CRMs will unavoidably rise with the broad penetration of consumer goods. Thus, laptop wastes are a valuable resource for CRMs and PMs and the estimation of their future EoL quantities is vital for their efficient management to ensure that resources will not be lost. Endorsement of the circular economy is a solution to materials criticality challenge, in which products and their CRMs, are kept within the economy over numerous product life cycles via reuse and effective recycling. In this way the economic productivity of CRMs is maximized and global demand is reduced (Krystofik et al., 2018; Charles et al., 2020).

Estimation of e-waste generation can be accomplished by many methods. These have been summarized by Wang et al. (2013), Guo and Yan (2017), Islam and Huda (2019a) and involve Material Flow Analysis (MFA) that estimate the Input-Output of materials within the system boundaries. MFA can be static or dynamic when it involves average values or time-varying parameters, respectively. Briefly the methods are: (1) the Time Step Model that requires sales and stock data, (2) the Market Supply Model requiring sales and average lifespan data, (3) the Market Supply A, involving sales data and a lifespan following a Weibull distribution, (4) the Carnegie Mellon model that applies discrete average lifespan for reuse, household stock, recycling or landfill, (5) the Simple Delay Model using sales and average lifespan data, (6) the Leaching Model for saturated markets and products with short lifespan. All these methods can be applied to estimate e-waste amounts that have been generated in past years.

However, as the amount of e-waste is increasing, the estimation of future quantities and the key metals contained in them is of primary importance to policy makers, producers and manufacturers to promote actions and legislations at local and national level related to their sustainable management. Specifically, this study focuses on laptops as these are consumer goods with rising adoption rate in Greece, especially nowadays due to coronavirus pandemic and they are the most valuable e-waste in terms of CRMs content, and last, their EoL management engages diverse stakeholders. The generation of obsolete amounts of laptops, notebooks and tablets (referred to as "laptops" hereafter) in Greece until 2040 (2016-2040) is presented. Both precious and hazardous compounds are present in waste laptops that require proper management in order to recover most of the valuable materials, while minimizing the risk of exposure to toxic substances.

Thus, the objectives of this chapter are: (i) to precisely determine time-varying lifespan values (1983-2015) by numerically solving the corresponding mathematical equations as suggested in E-Waste Statistics (Forti et al, 2018) given that high quality data are available instead of adopting a survey method. The latter is impossible to apply over an extended time period (32 years) and may introduce bias if not appropriately conducted. To the best of our knowledge, this is the first time this approach is applied. Such a long period can reveal the lifetime dynamics and the economic and social changes reflected in the laptop lifespan Furthermore, in order to check the validity of calculated lifespan profiles, since no specific data were available for Greece, the same analysis was performed for laptops in the Netherlands, where lifespan data existed in the literature and consistency check could be applied. Also the same evaluation was performed for Sweden for comparison reasons. Next, (ii) to reliably estimate by dynamic material flow analysis (dMFA) the future generation of EoL laptops in Greece, which is absent in the scientific literature, (iii) to consider dynamic changes in all parameters that are country specific like penetration rate, population, laptop weight and product lifespan. Furthermore, to assess the impact of the coronavirus pandemic on the lifespan of laptops and the estimated amounts of EoL laptops. Most reported studies apply simplifications of either the average product weight or the average lifespan (Polák and Drápalová, 2012; Zhang et al., 2012; Duygan and Meylan, 2015) and very few consider dynamic changes in more parameters (Wang et al, 2013). Further, (iv) to clarify how population, product lifespan and laptop weight affect the estimated future quantities and to what extent, (v) to estimate the CRMs and PMs content of future obsolete laptops (and tablets) and therefore illustrate the potential of recovering valuable resources that exist in EoL laptops in Greece, which is missing in the literature.

The outcome of this work will be valuable for Greek authorities, policy makers and electronic goods dealers and may contribute to improved management of EoL laptops in Greece, benefiting society and the environment.

2. Methodology

2.1 Calculation of EoL laptop amounts in historical years (1983-2015)

The EoL products output at time t is expressed as a function of product inputs and the change in product stock. This is expressed by the equation (Time Step model):

$$O_t = S_t - (St_t - St_{t-1}) \qquad (1)$$

Where O_t: obsolete product in year t, S_t: Sales of product in year t and St_t: stock of product in year t.

Also, the EoL products output can be given as a function of product inputs and the lifespan probability distribution (Market Supply A):

$$O_t = \sum_{i=0}^{n} (S_{t-i} \times f_i) \qquad (2)$$

Where f_t: lifespan probability distribution function and n: maximum possible lifespan.

The lifespan distribution is depicted by the Weibull distribution function. The Weibull probability distribution function is given by:

$$f(t,a,\beta) = \frac{a}{\beta^a} t^{a-1} \exp(-\left[\frac{t}{\beta}\right]^a) \qquad (3)$$

where α: the shape parameter that describes the gradual ageing of the product and β: the scale parameter describing the characteristic life of the product that is, it defines the years (age) when 63.2% of the products are expected to have failed.

The Mean Time to failure (MTTF), or else average lifespan is estimated by:

$$MTTF = \beta \times \Gamma(1 + 1/\alpha) \qquad (4)$$

where Γ: gamma function.

The product lifespan in this work represents the total time the product stays in the possession of a person, including stock time after the product ceases to be used or donation to another person. After the product lifetime has elapsed the product is discarded in appropriate collecting centers.

2.2 Estimation of future possession of laptops

In order to predict future possession of laptops the three parameter logistic model was employed. For a non mature market like laptops in Greece, the logistic curve includes the phases of growth and saturation. Possible decline phase can be expected in the very distant future, especially if this technology is substituted by a new one, however, this is quite difficult to foresee. The general logistic equation for laptop procession modeled as penetration rate (items per inhabitant), is expressed by (Yang and Williams, 2009):

$$\frac{dN}{dt} = rN(1-\frac{N}{K}) \qquad (5)$$

where N: penetration rate, number of laptops owned per inhabitant, r: growth rate, K: maximum penetration value. Solving this equation gives the penetration rate:

$$N_t = \frac{K}{e^{-(rt+C)}+1} \qquad (6)$$

where

$$C = \ln(\frac{N_o}{K-N_0}) \qquad (7)$$

and N_0: the penetration rate at time 0.

The penetration rate for year t is calculated by:

$$N_t = \frac{St_t}{Q_t} \qquad (8)$$

where Q_t: population at time t.

Using the simplification demonstrated (Meyer et al, 1999), the growth rate, r, was substituted by ln(81)/Δt, where Δt is the time required to grow from 10% to 90% of K, and –C/r was substituted by t_m, the time needed to reach K/2 (Althaf et al., 2019) . Thus, the logistic curve becomes:

$$N_t = \frac{K}{e^{(-\ln(81/\Delta t)*(t-t_m))} + 1} \qquad (9)$$

The most critical parameter for the logistic curve is the maximum penetration rate K. The solution to this equation is given by non linear regression by minimizing the Objective Function (OF):

$$OF = \sum_0^t \left(\frac{K}{e^{(-\ln(81/\Delta t)*(t-t_m))} + 1} - \frac{St_t}{Q_t}\right)^2 \qquad (10)$$

Using the bounding approach the lowest and highest value for K were estimated, and then the problem was solved for Δt and t_m.

Furthermore, the fit of the calculated curve to the historic data curve, at the optimal set of parameters, was determined by:

$$Deviation\% = 100\frac{\sqrt{OF(z-N)}}{\max(N_t)} \qquad (11)$$

where z: number of data points, N: number of parameters engaged in the model, N_t: historic penetration data.

The procedure followed is described hereafter. Initially, obsolete amounts of laptops were calculated by Eq. 1 using sales and stock data for Greece from Statistics Netherlands (Van Straalen et al, 2016) which are retrieved from Eurostat website. The categorization applied follows the European Prodcom (Production Statistics database) and Eurostat Ramon Database. Sales were calculated as imports minus exports, since there is no laptop production in Greece. Once the obsolete amounts were calculated, non linear regression was applied to estimate α, β for various years by minimizing the Objective Function (OF_1):

$$OF_1 = \sum_{t,i}^n \left\{ [S_{t-i} \times (F_{i+1} - F_i)] - [S_t - (St_t - St_{t-1})] \right\}^2 \qquad (12)$$

Where Fi: Weibull cumulative distribution function that gives the cumulative probability a product fails at time t.

The deviation for the curve fitting was calculated by (10) using O_t, as found by (1), instead of N_t in the denominator. Several constraints were applied in order to solve the problem as α, β>0 and α<4.5. The shape parameter α was taken as α<4.5, because α>4.5 is unlikely as this denotes rapid wear-out of products associated with severe problems in manufacturing process (Zhu, 1991).

In addition, another constraint applied was that the total obsolete amount calculated in each year, as a sum of products that were sold several years before multiplied by the probability *f(t,α,β)* that the product is discarded in a particular year, was forced to be $100 \pm 5\%$ of the amount calculated by

$$\sum_{t=t}^{n} [S_t - (St_t - St_{t-1})] \qquad (13)$$

for the same years.

Once the Weibull parameters *(α,β)* were calculated for several years (1983-2015), their trends were extrapolated in the future considering two cases, (i) there is or (ii) there is no impact of the coronavirus pandemic on laptop lifespan. The future possession of laptops was calculated till 2040 by the logistic curve and the future obsolete laptops amounts of (2016-2040) were calculated by iteratively solving Eq. 2 and 1 (Liu et al., 2006).

2.3 Substance flow analysis of EoL laptops

The main components of interest in laptops are printed circuit boards (PCBs), Hard Disk Drives (HDDs) or Solid State Disks (SSDs) -which are continuously gaining ground substituting HDDs- LCD or LED screens and last lithium-ion batteries. PCBs contain PMs, base metals (i.e copper) and toxic metals (antimony, lead etc.) as well as ceramic compounds and plastics (Cucchiella et al., 2015). HHDs contain magnets with high amounts of REEs. The material composition of laptops/notebooks and tablets is presented in Table 3.1 (Cucchiella et al., 2015; Van Eygen et al., 2016; Gaustad et al., 2020). In this Table the percentage of EU dependence on imports and the input recycling rate for EoL devices targeting CRMs are also presented (EU, Final Report 2020, Talens Peiro et al., 2018). Gold and silver are not considered CRMs for the EU economy as they present an insignificant supply risk. As can be observed, significant differences exist in CRMs and PMs concentration between laptops/notebooks and tablets. For this reason the percentage of tablets in the total calculated pieces is clarified and their content is calculated separately for EoL laptops and tablets. As far as notebooks and laptops are concerned an average value of key metals is used, as these were quite similar for these devices. The share of Greek households owning a tablet and laptop in 2017 was 30% and 48% respectively (Statista, 2018), while the number of households in Greece was 4,134,540 (ELLSTAT), thus the total number of possessed tablets can be calculated for 2017. Tablets were introduced in Greece around 2010, thus their number can be estimated assuming a linear increase in possession till 2017. Then the number of possessed laptops was deduced from the total number (laptops + tablets). In this way the relative percentages of the possessed tablets and laptops are clarified. After 2019 it is assumed that the tablets and laptops are equally possessed by Greeks. The CRMs and PMs content in EoL devices is calculated separately for laptops and tablets.

Then the quantity of metal i in year t was calculated by (Guo et al., 2017):

$$m_{i(t)} = C_i \times O'_t \qquad (14)$$

Where C_i: quantity of metal i in year t per device, O'_t: obsolete pieces in year t.

2.4 Data sources and collection

Data on past sales and stock were acquired from Statistics Netherlands (Van Straalen et al., 2016). Data on population were obtained from ELLSTAT, CIA-World Factbook and IndexMundi and data on employment in Greece from Eurostat and ELLSTAT.

The average laptop weight was estimated in each year (1995-2015) based on historic stock and population data. This was extrapolated to predict the future trend. Data showed that there was a decreasing trend, as anticipated. Since weight was continuously decreasing at an almost stable rate, 2 cases were considered: case 1, weight continuously decreasing till 2023 (0.44 kg) and then stable afterwards, or case 2, weight continuously decreasing till 2025 (0.14 kg) and then stable afterwards. The weight data calculated agree reasonably well with the weight data of Wang et al. (2013) and average EU-28 data report (Magalini et al., 2014). Indeed the calculated laptop weight in Greece is estimated to be 4.63 kg in 1995, while Wang et al, 2013 reported 4.6 kg in the Netherlands in 1995 and EU report, 4.5 kg for EU-28 for the same year. Furthermore, the estimated weight is 3.88 kg for 2000, 3.13 kg for 2005, 2.38 kg for 2010, 1.63 kg for 2015 and 1.48 kg for 2015, while the corresponding EU-28 values are 4.14, 3.68, 2.13, 1.26 and 1.26 kg respectively. However, as most studies estimate the generated waste using an average weight value, this approach was also employed for comparison reasons. Thus, the average value calculated for the years 2006-2016, 2.23 kg, was used as the fixed average weight from 2016 onwards. The obsolete laptop weight was calculated by the same approach to ensure realistic calculations as items that become obsolete in one year do not have the same weight as the new ones purchased in the same year, but a little higher as these were bought a few years before.

Furthermore, the growth/decline rate of population in Greece was estimated till 2015 and this rate was extrapolated to give an approximation of future population. It is estimated for Greece that the population decreases from 11,123,000 inhabitants in 2011 to approximately 10,351,937 inhabitants in 2040.

All calculations in this work were performed by Excel software and Solver add-in program.

Table 3.1CRMs and PMs in LCD/LED notebooks, laptops and tablets and EU indices. (Sources: Cucchiella et al, 2015; Van Eygen et a.l, 2016; Gaustad et al., 2020; EU,Final Report 2020; Talens Peiro et al., 2018).

Metal	**LCD/LED notebooks (g/unit)**	**Laptops (g/unit)**	**Average (notebooks/ laptops) (g/unit)**	**Tablets (g/unit)**	**% EU Import reliance**	**% EU EoL recycling input***	**EU Supply Risk (SR)**	**EU Economic Importance (EI)**
Antimony- Sb	0.77	1.95	1.36	0.154	100*	28	2	4.8
Neodymium-Nd	2.10	2.23	2.16	0.427	100	3**	6.1	4.8
Praseodymium- Pr	0.274	0.13	0.20	0.055	100	3**	5.5	4.3
Dysprosium- Dy	0.05	0.05	0.05		100	8***	6.2	7.2
Yttrium -Y	0.002	0.002	0.002	0.0009	100	8***	4.2	3.5
Indium-In	0.04	0.04	0.04	0.008	0*	0	1.8	3.3
Tantalum-Ta	1.70	1.70	1.70	-	100*	1	1.4	4
Cobalt- Co	0.065	0.065	0.065	0.013	32*	22	2.5	5.9
Palladium- Pd	0.04	0.03	0.035	0.008	93	14	1.3	7
Platinum- Pt	0.004	0.004	0.004	-	98	14	1.8	5.9
Gold-Au	0.22	0.22	0.22	0.044		20	0.2	2.1
Silver- Ag	0.25	0.57	0.41	0.05		55	0.7	4.1

*EU data refer to 2020. EU data marked * refer to 2017.*
*** refers to LREEs group, ***refers HREEs to group.*
SR on a scale of 0-6.2. EI on a scale of 0-8.1. The thresholds for the criticality assessment are set at 1 for SR and 2.8 for EI index

3. Analysis and Discussion

3.1 Bounding analysis for maximum penetration rate

Since laptop sales are continuously increasing in Greece, indicating that the saturation phase has not been reached (Fig. 3.1, historical data till 2015), the estimation of K was performed by the bounding approach of Yang and Williams (2009). This approach finds parameter values which serve as the lowest and highest conceivable values and calculates a range of results based on the two bounds. The upper bound is the maximum reasonably expected value for K and the lower bound is the minimum. Thus, the future possession of laptops will be within this range.

The upper bound for K is defined by assuming that due to coronavirus pandemic that changed the traditional education system to online, every pupil (from the age of 10) and university student owns a laptop. Also, all persons aged till 80 years old own a laptop for their personal needs and last, all employed persons own a laptop either at work or at home for remote working. According to ELLSTAT the population of Greece in 2011 aged between 10-80 years old was 9,183,113 people. In addition, as reported by Eurostat Employment Database, the number of employed persons in Greece was 4,083,030 (2016). Additionally, the total population of Greece in 2011 was 10,816,286 while in 2016, it was 10,835,000 people. Thus, the upper bound of K is estimated to be about 1.23. Yang and Williams (2009) estimated the upper bound of 1.3 personal computers per capita in USA according to 2007 data sets.

The lower bound is estimated assuming that only university students, professors, companies, information and telecommunications workers (ITC) and only the financially sound share of the population own a laptop. The number of Greek companies in 2016 (ELLSTAT) was 233,151 while the university students and professors were 461,613. The people working on ITC sector were 83,970 (Eurostat, 2016), while the number of people that could make ends meet, and thus could afford a laptop were estimated about 5,374,902 (Eurostat, 2017). Thus the lower bound for K is about 0.62.

In 2018 the laptop penetration level reported in Denmark was 0.64 laptops/capita (Zhilyaev et al., 2021), while that predicted by the logistic curve for

Greece, is 0.577 to 0.78 pieces/capita for the lower and upper bound scenario, respectively. Moreover, considering the effect of coronavirus pandemic on the laptop sales (2020), it is more likely that the penetration level in Greece will move closer to the upper bound scenario.

3.2 Estimating future penetration rate by logistic model

Figure 3.1 depicts the estimated future laptop penetration rate for the upper bound (K=1.23), the lower bound (K=0.62) as well as the existing data (till 2015). As can be seen the penetration of laptops is at a growth phase as it continuously increases and has not passed through an inflection point yet. This indicates that from the perspective of technological life cycle, diffusion of laptops is still in its early stage.

It is observed that the penetration rate of 0.62 laptops per inhabitant will be reached in 2027 that is in 6 years time, while that of 1.23 laptops per inhabitant, in 2040, in approximately19 years time. Comparing the deviation value as calculated by Eq. 10, the upper bound curve gives a better fitting than the lower bound, (2.62% compared to 3.25%). However, as deviation is calculated by comparing historic penetration data till 2015, both values are within acceptable limits. It is calculated that in 2023 1 and 0.62 laptop will be owned per inhabitant in Greece in the upper and lower bound scenario, respectively. The calculated values of Δt and t_m are 19.46 and 35.57 for the upper bound scenario and 13.19 and 30.22 years for the lower bound scenario.

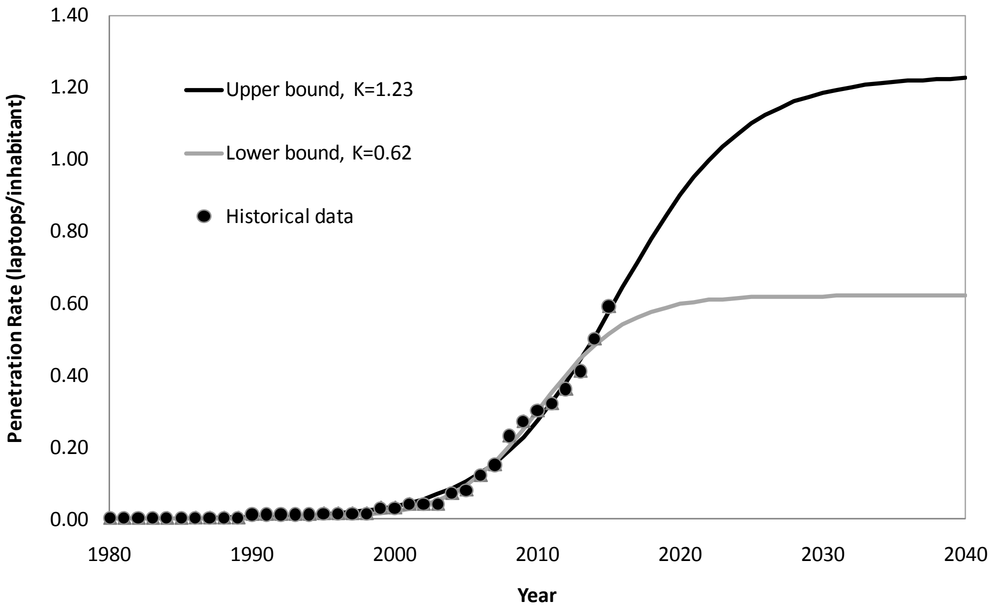

Figure 3. 1. Estimation of laptops penetration rate.

3.3 Estimation of the Weibull parameters and the laptops lifespan

Following the procedure depicted in 2.1, the Weibull parameters are estimated for the years 1983-2015, as explained hereafter. The obsolescence rate fi+1-fi, was calculated by Eq. (1) and (2) using historical data on sales and stock (Wang et al., 2013). The calculation was performed for successive years by considering a 12 year time period that is (1983+ i)-(1995+i), where i: 0-20. Once the Weibull shape and scale parameters are calculated, the lifespan can be estimated (Table 3.2). The values calculated for the Netherlands agree with the shape and scale parameters calculated by Wang et al. (2013) for years 1995 and 2005. Specifically, α, β are reported as 1.6 and 5.6 for 1995 which reasonably agrees with 1.14 and 5.47 estimated for years 1991-2003. Also, α, β are declared as 1.5 and 5.2 for 2005 which complies well with 1.22 and 5.41 (2001-2013). The shape, scale parameter and the MTTF values for EoL laptops in Greece and the Netherlands are given in Table 3.2.

Lifespan is a dynamic parameter that depends on the socio-economic situation in each country, the diffusion of new technology, as well as the habits and awareness of the people (Islam and Huda, 2019a). This is depicted by MTTF values estimated for Greece, which are portrayed in Figure 3.2. As can be seen, the lifespan of laptops in Greece is higher than in the Netherlands and Sweden, which shows that laptops are being replaced at a slower rate in Greece than in the other two countries. This indicates that the purchasing power is lower in Greece, laptops are probably stored for a longer period at home, the take-back system may work less efficiently and public awareness on e-waste recycling is lower. In Fig.3.2 the lifespan of laptops increases around 2008, probably due to the economic crisis in Greece, indicating that people did not buy new products to substitute their old ones. However, as this financial crisis also influenced other Eurozone countries to some extent, a similar increase in lifespan is also observed for the Netherlands in 2009, although less protrusive. On the other hand, the lifespan of laptops is continuously decreasing in Sweden from 2002 to 2015 implying that Sweden was not affected by the European debt crisis, perhaps because it does not employ the Euro currency.

Table 3.2 Weibull lifespan distribution parameters and MTTF for laptops in Greece and the Netherlands.

		GREECE				NETHERLANDS			
		a	**b**	**MTTF**	**dev(%)**	**a**	**b**	**MTTF**	**dev(%)**
1983	1995	1,98	8,90	7,89	6,12	2,44	8,73	7,74	4,9
1984	1996	1,92	8,30	7,37	6,66	1,88	8,26	7,33	6,7
1985	1997	1,75	7,72	6,88	6,59	1,76	7,66	6,82	6,5
1986	1998	1,59	7,26	6,51	7,28	1,62	7,09	6,35	7,2
1987	1999	1,50	7,03	6,35	8,09	1,50	6,60	5,96	8,0
1988	2000	1,42	6,99	6,36	9,15	1,37	6,19	5,66	8,9
1989	2001	1,37	6,97	6,38	10,26	1,26	5,79	5,38	9,9
1990	2002	1,37	6,96	6,37	11,21	1,18	5,51	5,20	11,9
1991	2003	1,42	7,20	6,55	11,77	1,14	5,47	5,22	12,6
1992	2004	1,35	7,78	7,13	12,13	1,14	5,73	5,47	13,3
1993	2005	1,38	8,61	7,87	13,89	1,19	6,15	5,79	13,8
1994	2006	1,47	9,25	8,37	14,26	1,30	6,56	6,06	14,2
1995	2007	1,50	9,72	8,77	13,18	1,41	6,86	6,25	12,9
1996	2008	1,56	10,03	9,02	13,17	1,42	6,97	6,34	14,2
1997	2009	1,69	9,96	8,89	12,58	1,62	7,08	6,34	12,4
1998	2010	1,80	9,60	8,54	11,76	1,43	6,84	6,21	10,9
1999	2011	1,83	9,08	8,07	10,99	1,42	6,54	5,95	12,0
2000	2012	1,77	8,43	7,51	10,42	1,67	6,30	5,63	11,5
2001	2013	1,61	7,70	6,90	10,22	1,22	5,41	5,07	11,5
2002	2014	1,41	6,92	6,30	10,27	1,03	4,58	4,52	11,2
2003	2015	1,20	6,09	5,74	10,70	1,01	4,37	4,36	13,5

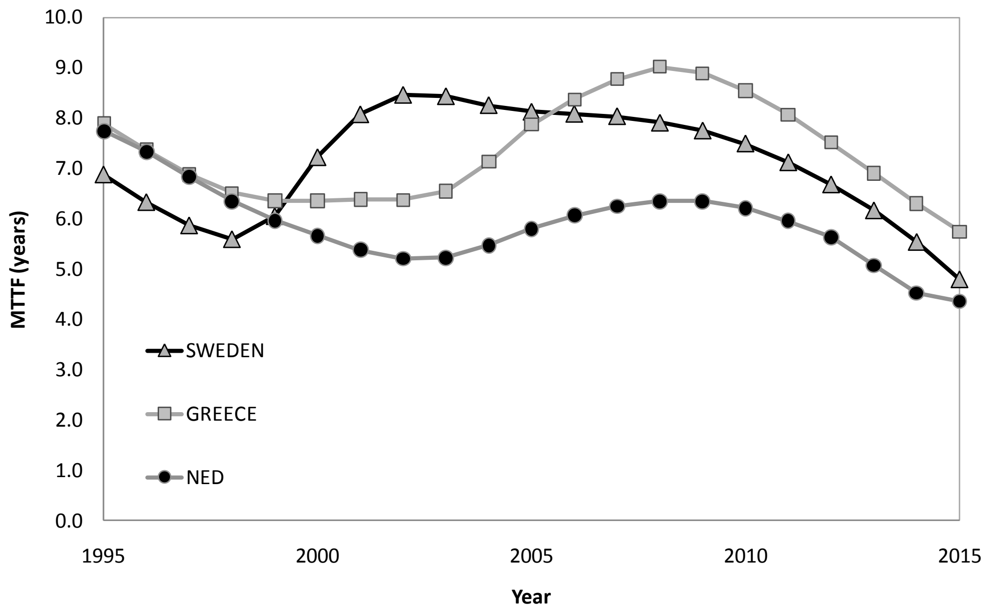

Figure 3.2 Dynamic profile of laptop Average Lifespan (MTTF) in Greece, the Netherlands and Sweden.

The variation in the laptop lifespan in Greece is also illustrated in Figure 3.3. It is obvious that the probability of laptops becoming obsolete has shifted to almost 5.74 years (2015) from 8 years in the past (1995), meaning that more laptop waste is currently generated. Forti et al. (2018) reported the average laptop lifespan for the Netherlands, France and Belgium was 5.9 years in 2016. Further, the estimation of lifespan distribution parameters for 2016-2040 was accomplished by extrapolating the existing trend (1983-2015). Two cases were considered: the lifespan distribution follows a curve-pattern reflecting the effect of coronavirus pandemic or a continuously decreasing pattern not affected by the coronavirus pandemic.

As can be seen in Fig. 3.4, the time profile of the average lifespan presents a peak in year 2008, correlated to the economic crisis in Greece. Nowadays, a financial recession is witnessed due to the coronavirus crisis. Therefore, it is anticipated that a similar pattern will be observed in the lifespan profile in the future. Thus, the lifetime of laptops, after decreasing till 2015, will slowly increase to peak around 2020-2021. Thus, two cases are taken into account: the generation of obsolete laptops when, (i) the lifespan distribution follows a curve-pattern and (ii) a continuously decreasing pattern. The lifespan profiles (scale parameter) employed are presented in Fig. 3.4. As regards the shape parameter, α, this followed a slight decrease over the years 1995-2015 and this was extrapolated to the future.

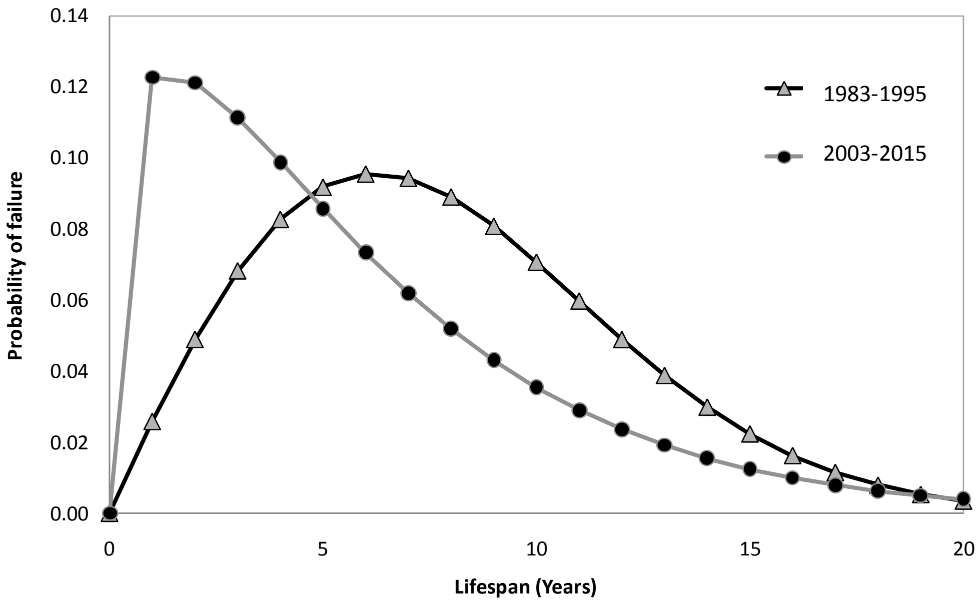

Figure 3.3. Probability of obsolete laptop/tablets generation in Greece in 1995 and 2005.

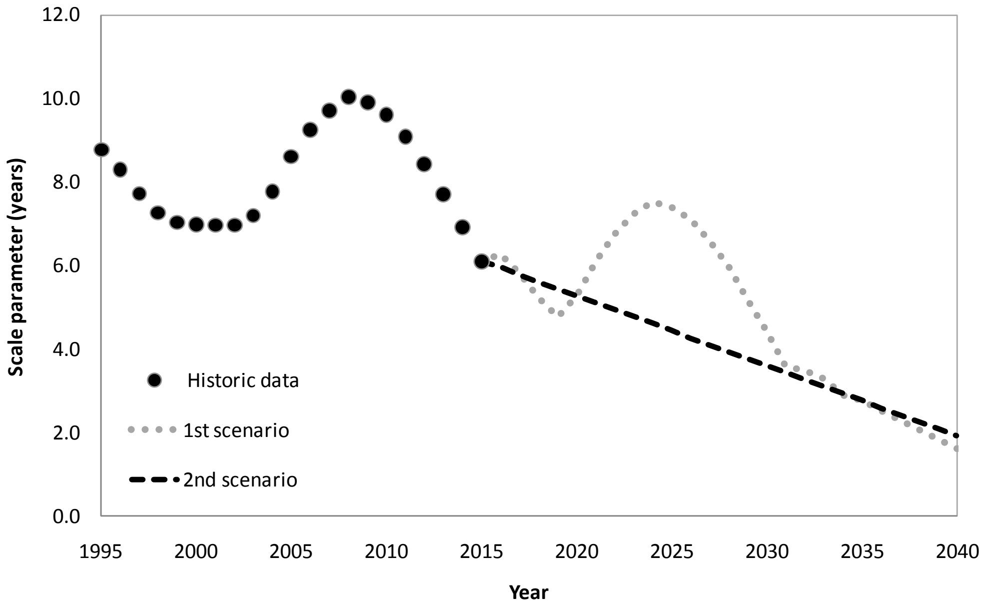

Figure 3.4. Weibull scale parameter: calculated values and values extrapolated in the future. In the 1st scenario lifespan is affected by the coronavirus pandemic, in the 2nd scenario it is not affected.

3.4 Sensitivity and uncertainty analysis

The future generation of laptop waste was calculated according to the scenarios presented in Table 3.3. The examined parameters were: laptop penetration rate (0.6 or 1.23 pieces/inhabitant), laptop weight (either average value of 2.23 kg/piece or decreasing till 0.44 or 0.14 kg/piece), Weibull parameters (either changing according to curve or decreasing pattern, as referred in section 3.3) and population change (either decreasing as described in 2.4 or stable from 2020 and on). In this way the effect of the examined parameters on the estimated obsolete laptop amounts is clarified and light is shed on the most influential parameters of the analysis.

First, regarding the lower bound penetration capacity (K=0.62), the results depicted in Fig. 3.5 (a), show that scenario A1 (0.44 kg) and A2 (0.14 Kg) yield almost identical results (deviation <1%), which means that for a reducing future laptop weight, there is insignificant effect after 2025. However, the A3 scenario, (average weight 2.23 kg) yields different results as there is a significant deviation from 2027 (0.5%) that gradually increases to 27% by 2040. Thus, the simplification of considering an average weight leads to overestimation of obsolete amounts of laptops. Comparing A1to A2 and A3 scenario it is evident that the accumulated amounts of EoL laptops in years 2016-2040, are almost 29,696, 29,719 and 33,476 tonnes for scenario A1, A2 and A3 respectively and the maximum EoL laptops amount will be generated by 2020 and then will gradually decrease. According to A1 scenario the generated EoL laptops in 2000 are about 96 tonnes while by 2010 they are 8 times more (762 tonnes). Moreover the increase in the next 10 years (2010-2020) is estimated at 105%. The annual growth rate from 2013-2020 is below 10%, while it depicts a reducing trend in 2020-2040. The same trends are observed in A3 scenario except that the reducing trend is minor.

Also, there is a considerable difference regarding the calculated units in A3 scenario compared to A1, as can be seen in Fig.3.6 (a) and (b). In the case of average weight (A3), the unit curve follows the pattern of the weight curve, while on the contrary, when the reducing weight is considered, the shape of the two curves is different. For the reducing weight, a peak in the amount of pieces will occur in 2025, corresponding to 2,866,241 pieces, while for the average weight case, the peak occurs in 2020, corresponding to 740,883 pieces. Also, the accumulated pieces in 2016-2040 will be 47,844,127 and 15,011,523 in scenario A1 and A3 accordingly. The accumulated pieces by 2015 are only 7.6% and 25% of the accumulated pieces in 2016-2040 in A1 and A3 scenario accordingly, revealing that enormous amounts of laptop waste are anticipated in the future. In A2 scenario, the maximum number of pieces is 9,641,969 items in 2025 and the accumulated pieces in 2016-2040 are 138,006,988 (data not shown). Thus the scenario A1 seems more plausible than A2 or A3, or the reducing weight till 0.44 kg, is more realistic.

The comparison of A4 to A1 scenario reveals a deviation of 7-15% (effect of the Weibull parameters, Fig. 3.7) while the comparison of the other scenarios A3 to A6, A3 to A31 and A1 to A11 reveal insignificant fluctuations and are all reported hereafter. Comparison of scenario A4 to A1, clarifies how the Weibull parameters affect the estimated amounts of EoL laptops. When the Weibull parameters are gradually decreasing (A4), the accumulated obsolete laptops in 2016-2040 are 31,869 tonnes as compared to 29,696 tonnes when the Weibull parameters follow the curve pattern (A1). In Fig. 3.7 (a) there is a deviation of 7% in 2023 which gradually increases to about 15% in 2040.The estimated amounts of obsolete laptops are 34695 tonnes for A6 scenario (average weight 2.23 kg, decreasing Weibull parameters) as opposed to 33476 tonnes for A3. The deviation is less than 10% when the average mass (2.23 kg) scenario is examined. The scenario that employs the reducing Weibull parameters presents the higher amount of EoL laptops.

Furthermore, inspection of scenarios A1 and A11 can explain if there is a significant deviation when the population is considered stable instead of slightly decreasing after 2020. The results reveal a negligible deviation of maximum 2% in estimated amounts of EoL laptops. The total amount estimated in 2016-2040 is 29803 tonnes in the case of A11 as opposed to 29696 tonnes (A1). Also examination of scenarios A3 and A31 reveal a negligible deviation, except for the last 3 years (2038, 2039, 2040, deviation 9%, 12% and 16% respectively).

Table 3.3 Different scenarios examined in the generation of obsolete laptops in Greece.

Parameter	Scenario A1	Scenario B1	Scenario A2	Scenario B2	Scenario A3	Scenario B3	Scenario A4	Scenario B4	Scenario A5	Scenario B5	Scenario A6	Scenario B6	Scenario A11	Scenario B11	Scenario A31	Scenario B31
max penetration rate (pieces/inhabitant)	0.62	1.23	0.62	1.23	0.62	1.23	0.62	1.23	0.62	1.23	0.62	1.23	0.62	1.23	0.62	1.23
laptop/tablets weight	decreasing till 0.44 kg (2023) and then stable		decreasing till 0.14 kg (2025) and then stable		average 2.23 kg (from 2016 and on)		decreasing till 0.44 kg (2023) and then stable		decreasing till 0.14 kg (2025) and then stable		average 2.23 kg (from 2016 and on)		decreasing till 0.44 kg (2023) and then stable		average 2.23 kg (from 2016 and on)	
Weibull parameters	according to scenario 1 (curve pattern)*		according to scenario 1 (curve pattern)*		according to scenario 1 (curve pattern)*		according to scenario 2 (decreasing pattern)		according to scenario 2 (decreasing pattern)**		according to scenario 2 (decreasing pattern)**		according to scenario 1 (curve pattern)*		according to scenario 1 (curve pattern)*	
Population	Decreasing from 2010 and on		Decreasing from 2010 and on		Decreasing from 2010 and on		Decreasing from 2010 and on		Decreasing from 2010 and on		Decreasing from 2010 and on		Stable from 2020 and on		Stable from 2020 and on	

*Curve pattern reflects the impact of the coronavirus pandemic, ** Decreasing pattern is not affected by the coronavirus pandemic

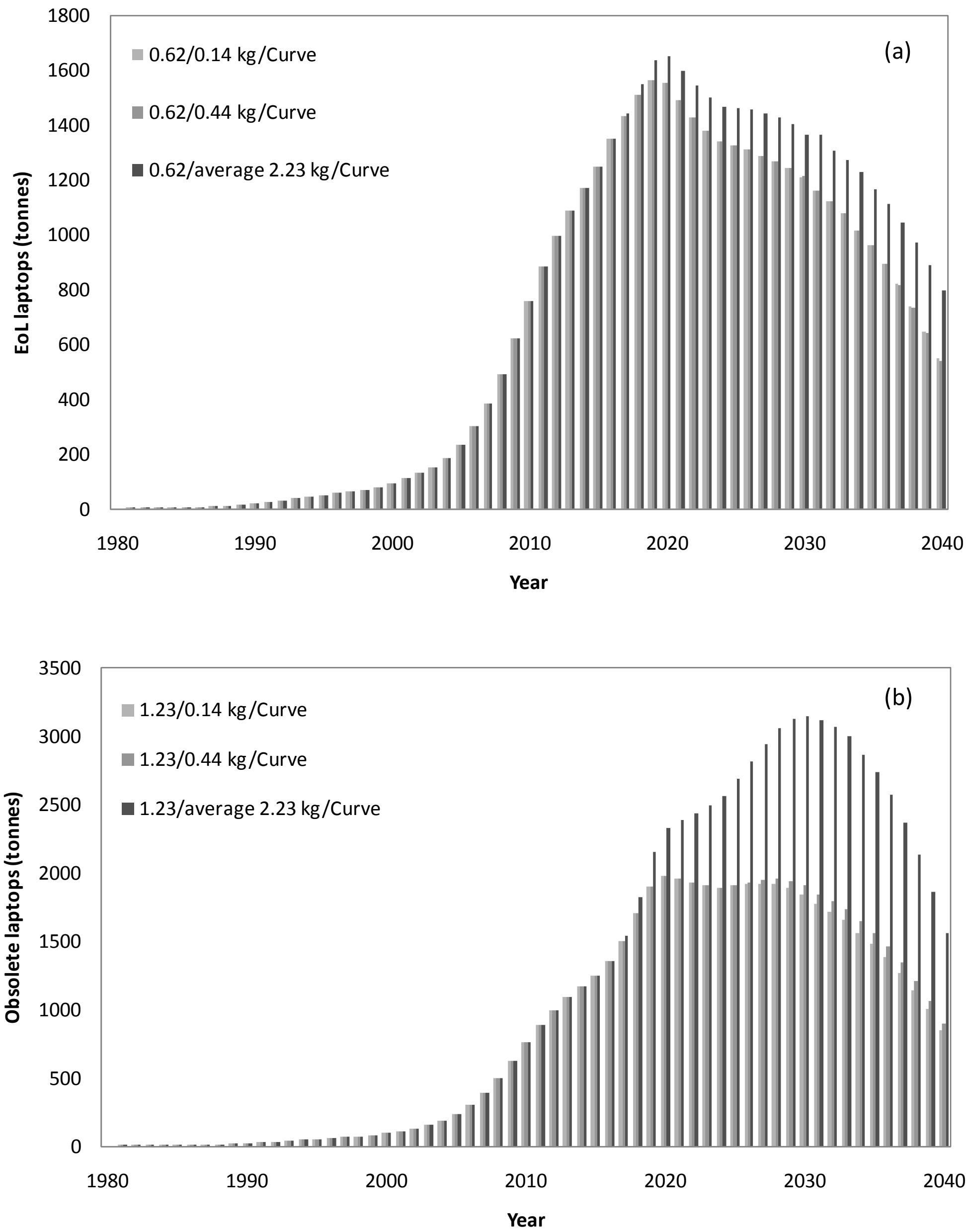

Figure 3.5. Estimation of obsolete laptops according to the scenarios: (a) A1, A2 and A3 (penetration rate 0.62/final weight/Weibull parameters pattern) and

(b) B1, B2 and B3 (penetration rate 1.23/final weight/Weibull parameters pattern)

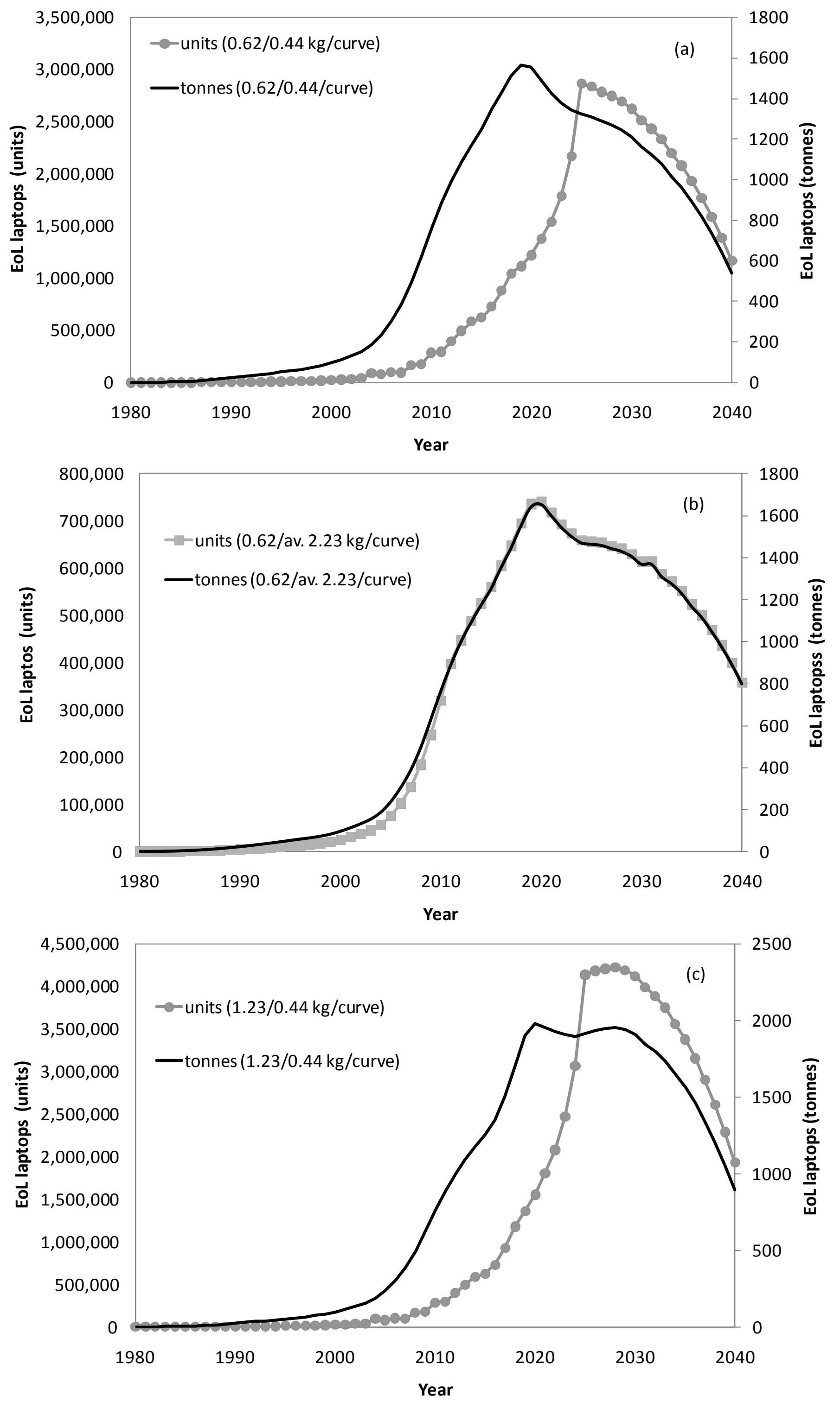

Figure 3.6. Estimation of obsolete laptops by units and weight (tonnes) according to scenario (a) A1 (0.62/0.44 kg/Curve), (b) A3 (0.62/2.23 kg/Curve) and (c) B1 (1.23/0.44 kg/Curve).

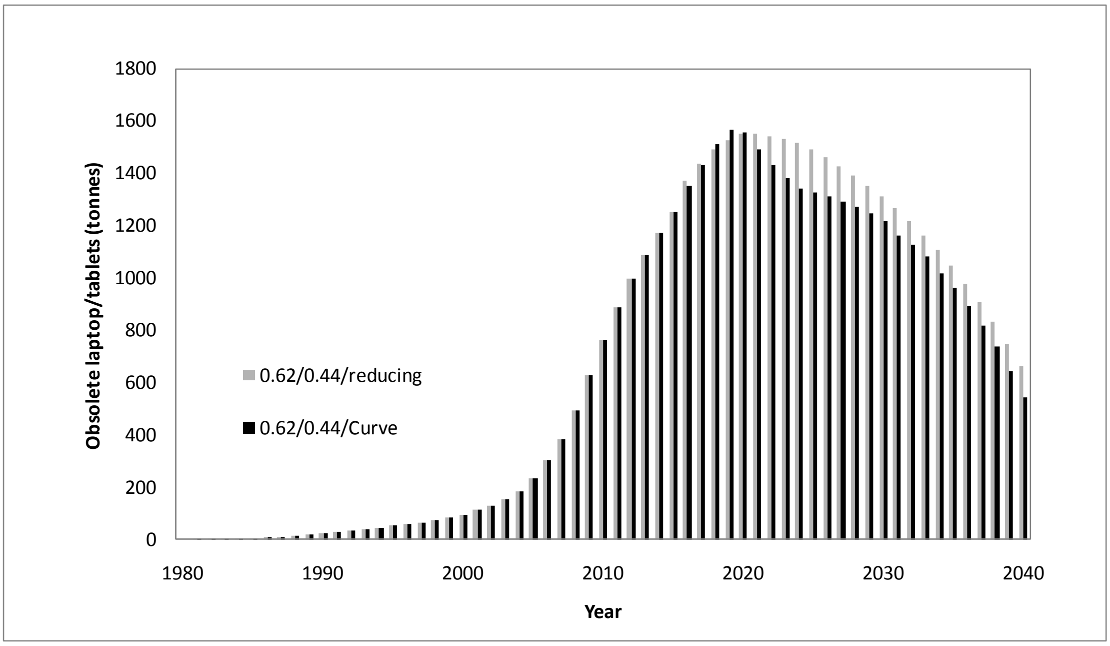

Figure 3.7. Effect of the Weibull parameters: Estimation of obsolete laptops according to scenario (a) A1 and A4 (penetration rate 0.62/final weight 0.44 kg/Weibull parameters pattern).

As far as the upper bound penetration capacity is concerned (K=1.23), Fig.3.5 (b) presents the results from scenarios B1, B2 and B3. In the upper case capacity, significantly more amounts of obsolete laptops are accumulated in 2016-2040, which are 42,228, 41,350 and 62,122 tonnes for B1, B2 and B3 scenario respectively. Also, considerable and increasing amounts of obsolete laptops will be generated till 2030 and then the amounts will gradually decrease. Analyzing the data in Fig. 3 (b), it is concluded again (as in lower bound scenario) that, B1 and B2 yield almost similar results (dev. 1.23-3.89% in 2028-2040), while the deviation in calculated results in B3 (compared to B1) is 1.4-39% in 2017-2040. According to B1 scenario the generated EoL laptops in 2010 are 762 tonnes, while the increase in the next 10 years (2010-2020) is estimated at 160%. The 10-year change rate is -6.6% (2020-2030) while the annual rate in the same period ranges between -3.9 to 1%. The annual rate is constantly declining after 2030. In B3 scenario the 10-year growth rate is 206% (2010-2020) and then 35% (2020-2030).

In B1 scenario (0.44 kg), the generated amount of obsolete laptops is described both in pieces and weight [Fig. 3.6 (c)]. It is observed that the maximum number of obsolete pieces will be 4,223,002 in 2028. Also, the accumulated pieces in 2016-2040 will be 71,617,824, while the accumulated pieces by 2015 are only 5.1% of the accumulated pieces in 2016-2040. This shows that huge amounts of laptop waste are expected in the future. In B2 and B3 scenario the peak in estimated pieces will be in 2025 (13,890,446 pieces) and 2030 (1,412,551 pieces), respectively and the total accumulated pieces in 2016-2040 will be 204,967,672 and 31,649,463 accordingly, (Fig. 3.8 and Fig. 3.9). While scenario B3 underestimates the number of EoL pieces, B2 seems to overestimate, thus the more realistic scenario is B1.

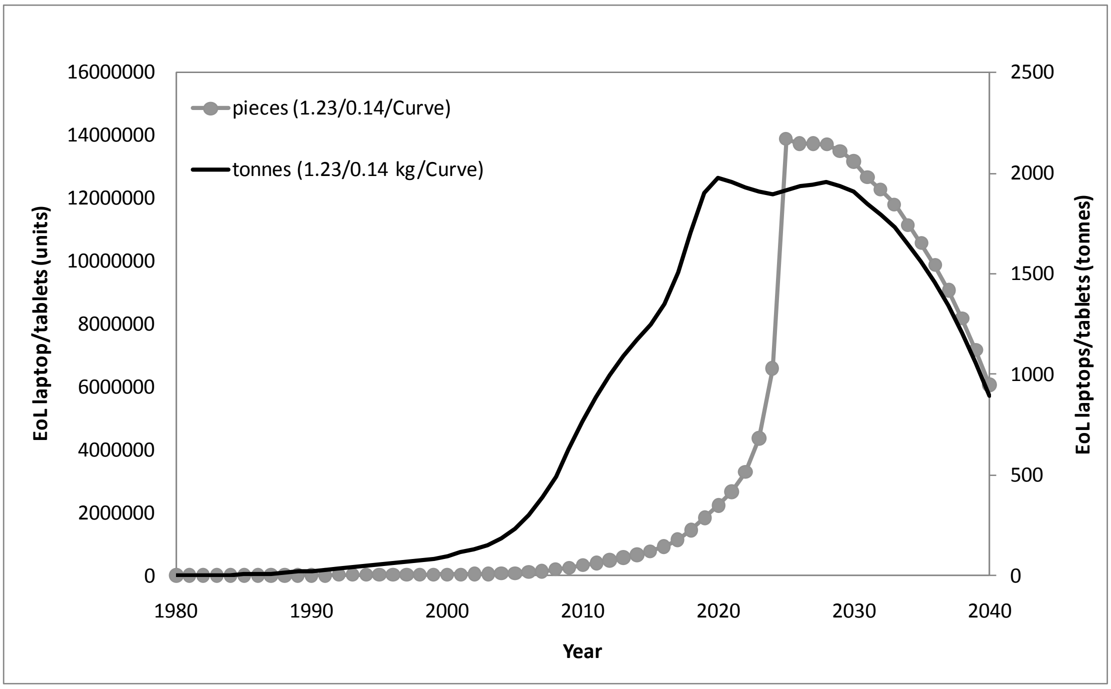

Figure 3.8. Estimation of obsolete laptops/tablets by units and weight (tonnes) according to scenario B2.

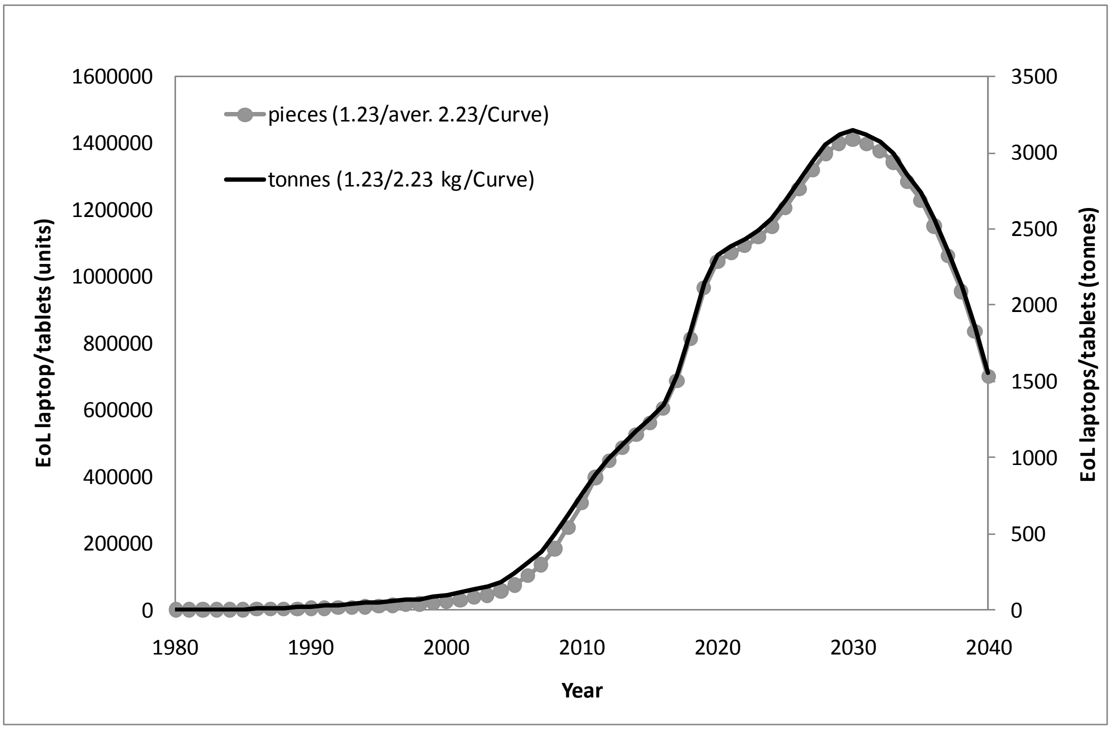

Figure 3.9. Estimation of obsolete laptops/tablets by units and weight (tonnes) according to scenario B3.

Furthermore, regarding the effect of the Weibull parameters pattern, Fig. 3.10 compares scenario B1 to B4. In B4 the accumulated amounts of obsolete laptops are 47,102 tonnes (2016-2040), substantially higher than 42,228 tonnes in B1. Thus, the effect of the Weibull parameters on the estimation of laptop waste is significant. Scenario B1 takes into account the effect of coronavirus pandemic on the laptop lifespan, while B4 does not. The deviation in years 2016-2040 in curves B1 and B4 is between 0.24-17%.

When the Weibull parameters follow a reducing pattern the generated amounts of obsolete laptops are depicted in Fig. 3.11 regarding B4, B5 and B6 scenarios. As can be seen, the generated amounts are higher in this case and there is a deviation between 10-19.5% in years (2022-2040) between B4 and B6. In B4 scenario the generated EoL laptops in 2020 are 1961 tonnes, while the increase in the next 10 years (2020-2030) is estimated at 9.3%. In B6 scenario the generated EoL laptops in 2020 are 2311 tonnes, and the 10-year growth rate is 61% (2020-2030).

In addition, comparison of B11 to B1 scenario reflects the population fluctuations (stable after 2020 or decreasing) in the estimated amounts of obsolete laptops. This is insignificant as deviation is less than 1% till 2036, and then less than 2.5% till 2040. Thus, the population change regarded does not affect the waste amounts considerably.

In conclusion, the most important parameter affecting the calculated obsolete laptop quantities is the weight of the devices, which should be considered a dynamic changing parameter instead of an average one. Following, the Weibull parameters are significant as their pattern (affected by the coronavirus pandemic or not) substantially influences the estimated waste amounts. As for the population variation and the final considered weight (0.14 or 0.44 kg), these had a minimum effect.

A comparison of quantities of laptop waste with other EU countries reveals that the generated quantities in recent years in Greece are lower than in Belgium and the Netherlands. Van Eygen et al. (2016) calculated 2,449 tonnes laptop wastes were generated in Belgium in 2013 contrary to 1,090 tonnes in Greece. Wang et al., 2013 estimated 5,137 tonnes EoL laptops in the Netherlands in 2010 contrary to 762 tonnes in Greece. Also Cuchiella et al., 2015 estimated that there will be an increase in EoL notebooks and tablets in the EU of 42% in 2014-2020, while the calculated increase in Greece will be 33% in A1 and 41% in A3 scenario, 68% in B1 and 92% in B3 scenario. This information is important for waste management systems to be able to tackle forecoming obsolete amounts and prepare suitable collection and treatment schemes.

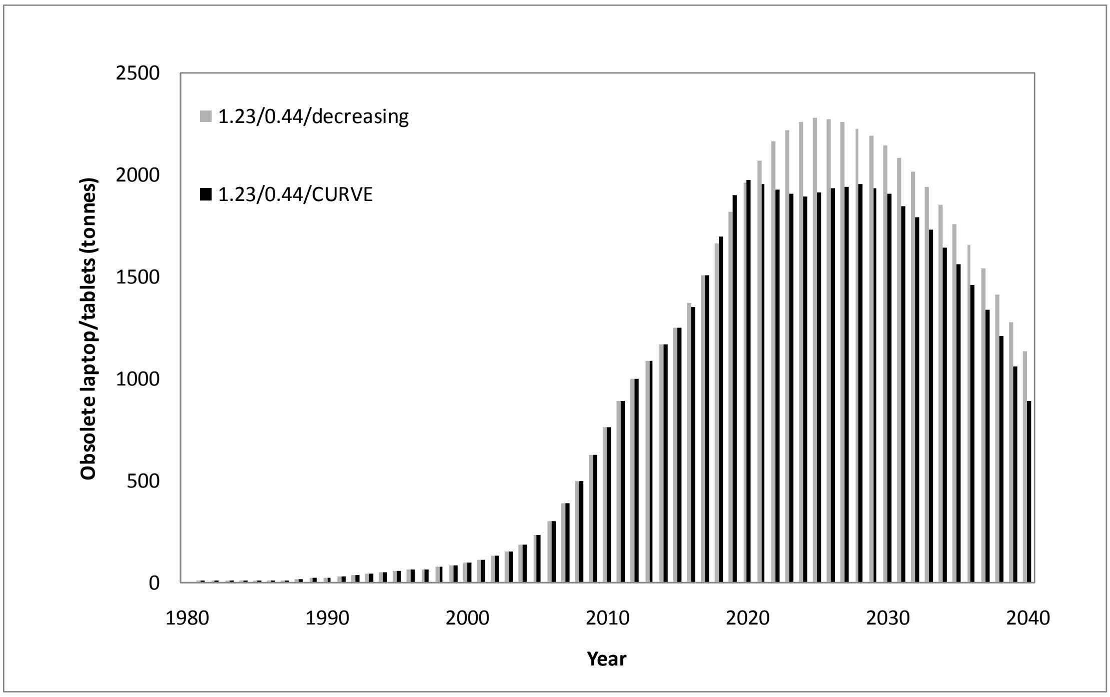

Figure 3.10. Effect of the Weibull parameters: Estimation of obsolete laptops according to B1 and B4 scenario (penetration rate 1.23/final weight 0.44 kg/Weibull parameters pattern).

3.5 Estimation of CRMs and PMs in the EoL laptops

An estimation of CRMs and PMs content in the generated EoL laptops is presented, to illustrate the potential of key metals recovery in EoL laptops. Table 3.1 presents CRMs and PMs content in LCD/LED notebooks, laptops and tablets. As observed, laptops and notebooks include many valuable metals in high concentrations, while the concentrations in tablets are lower. Although future changes in the products design may slightly affect the CRMs and PMs content, the results are indicative of the valuable materials embedded in these electronic devices. These changes may not be dramatic, i.e. Charles et al. 2017, estimated that the trend in random-access memory (RAM) modules in PCBs is that gold and silver content remains roughly stable while palladium content decreases (1988-2010). However, technological innovations in the field of displays like flexible Organic Light Emitting Diodes (OLED) screens or rechargeable batteries like lanthanum-nickel-hydride (La-Ni-H) may change the average composition (Zhang et al., 2017), but predicting the future changes is challenging. Also, composition modifications may occur due to the substitution of scarce or expensive elements by more abundant or cheaper ones, like replacement of Pr and Nd by La and Ce (Zhang et al., 2017, Lixandru et al., 2017).

A possible future technology change in laptops will lead to new waste products of slightly different composition. The recycling industry must invest in research and development to adapt to new waste devices to ensure better resource management. An example of a technological shift in the past was the evolution of CRTs to LCDs TV screens. Ecological design, creation of devices that can be easily processed to recover embedded materials and cooperation between producers and recyclers are prerequisites for the success of future recycling (Kalmykova et al., 2015; Shittu et al., 2021).

Table 3.1 also includes various indices that illustrate the significance of these metals in the EU economy, as most of them are 100% imported, while they are recycled at a low level (<10%). The Supply Risk (SR) index is especially high for RREs while the Economic Importance (EI) index is very high for RREs and platinum group metals. This information points to the fact that EoL laptops are 'treasured' materials that must be collected, treated, effectively recycled to ensure that their valuable metals stay within the EU, contributing to a circular economy. Emphasis must be placed on improving recycling, as increasing the recycling rate, reduces the supply risk of materials. For instance, the EoL recycling rate of antimony is 28% in 2020 (while <5% in 2011) thus the SR decreased from 2.6 in 2011 to 2 in 2020, as recycled Sb can enter the supply chain, relieving the import reliance stress (100% imported in EU in 2017). At the same time the EI has fallen from 5.8 in 2011 to 4.8 in 2020, as substitute materials are partly used, mitigating the risk of supply disruptions. Substitute materials can help reduce both EI and SR indices. For instance, compounds of Sn, Ti, Cr, Zn and Zr can replace Sb in the manufacture of pigments and glass. However, in its principal application as flame retardant, substitution of antimony is much harder. (EU, Final Report 2020, EU factsheets on CRMs, 2020). With the increasing amounts of e-waste, (laptop waste in particular), developing non-polluting, efficient and economical recycling technologies is crucial to recovering their valuable metals (Kiddee et al, 2013; Kaya, 2016). In order to facilitate recycling, it is necessary to design devices for recycling, promoting disassembly and recovery (Shittu et al., 2021).

Regarding EoL laptops, the estimations of key metals (from 2000 to 2040) are illustrated in Figure 3.12 for B1 scenario and in Figure 3.13 (for B3 scenario). In B1 scenario the number of generated pieces is far greater because the weight of the devices is considered reducing instead of stable as in B3. In B1 scenario the 10 year growth rate (2010-2020) of the key metals in EoL laptops is about 170%, while it is predicted that the same growth rate will be achieved in only 5 years time in the future (2020-2025). From 2025-2040 the annual growth rate declines to almost below 1%. The generated amounts in 2020 are 1055 kg Sb, 1675 kg Nd, 155.2 kg Pr, 38.8 kg Dy, 1.6 kg Y, 31 kg In, 1319 kg Ta, 50.4 kg Co and the precious metals 27.2 kg Pd, 3.1 kg Pt, 171 kg Au and 318 kg Ag, while in 2025 they are predicted 2.7 times more. The total accumulated amount in waste laptops in 2020-2040 will be 46 t Sb, 73 t Nd, 6.7 t Pr, 1.7 t Dy, 0.07 t Y, 1.35 t In, 57.3 t Ta, 2.2 t Co and 1.18 t Pd, 0.13 t Pt, 7.4 t Au and 13.8 t Ag. The accumulated amounts till 2019 are only 16% of the accumulated amounts in 2020-2040, showing the great potential for urban mining in the near future, which should not be lost.

Concerning the key metals in EoL tablets the estimations for B1 scenario are depicted in Figure 3.14 and for B3 scenario in Figure 3.15. The annual growth rate in key metals in 2015-2020 is estimated between 14-59%, while it will range between 14-35% in 2020-2025. The estimated amounts in 2020 are 119.5 kg Sb, 331 kg Nd, 0.7 kg Y, 6.2 kg In, 10 kg Co, 6.2 kg Pd, 34 kg Au and 39 kg Ag. In 2025 the predicted amounts are almost three times more. The total accumulated amounts in waste tablets in 2020-2040 will be 5.19 t Sb, 14.4 t Nd, 0.03 t Y, 0.27 t In, 0.44 t Co, 0.27 t Pd, 1.48 t Au and 1.69 t Ag. The accumulated amounts of key metals in the past (till 2019) are only 7% of the future accumulated amounts (2020-2040), revealing that we must invest on collecting and recycling to maintain the secondary resources within the economy.

Regarding EoL laptops in B3 scenario (2.23 kg) the annual growth rate of key metals ranges from 21-35% (2000-2011), as depicted in Fig. 3.14. Then the annual growth rate decreases and is about 14% in 2019 and then lowers down to below 10%. The generated amounts in 2011 of Sb, Nd, Pr, Dy, Y, In, Ta, Co and the precious metals Pd, Pt, Au and Ag are 525.2, 834.2, 77.2, 19.3, 0.8, 15.4, 656.5, 25 and 13.5, 1.5, 85 and 158.3 kg respectively, while the values in 2030 are almost double. EoL laptops contain significant amounts of Nd, Ta and Sb as well as Ag and Au. The total accumulated amount in waste laptops in 2020-2040 will be 16.9 tonnes Sb, 27.8 t Nd, 2.5 t Pr, 0.62 t Dy, 0.02 t Y, 0.50 t In, 21.1 t Ta, 0.81 t Co and 0.43 t Pd, 0.05 t Pt, 2.73 t Au and 5.1 t Ag.

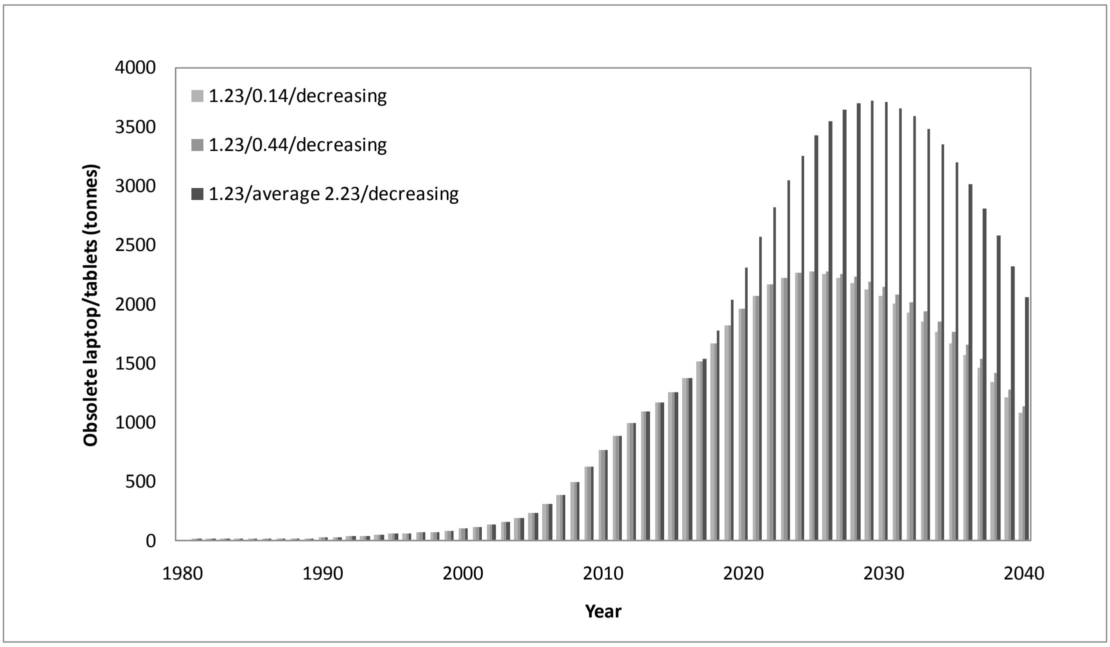

Figure 3.11 Estimation of obsolete laptops/tablets according to scenario B4, B5 and B6 (penetration rate 1.23/final weight / decreasing Weibull parameters pattern).

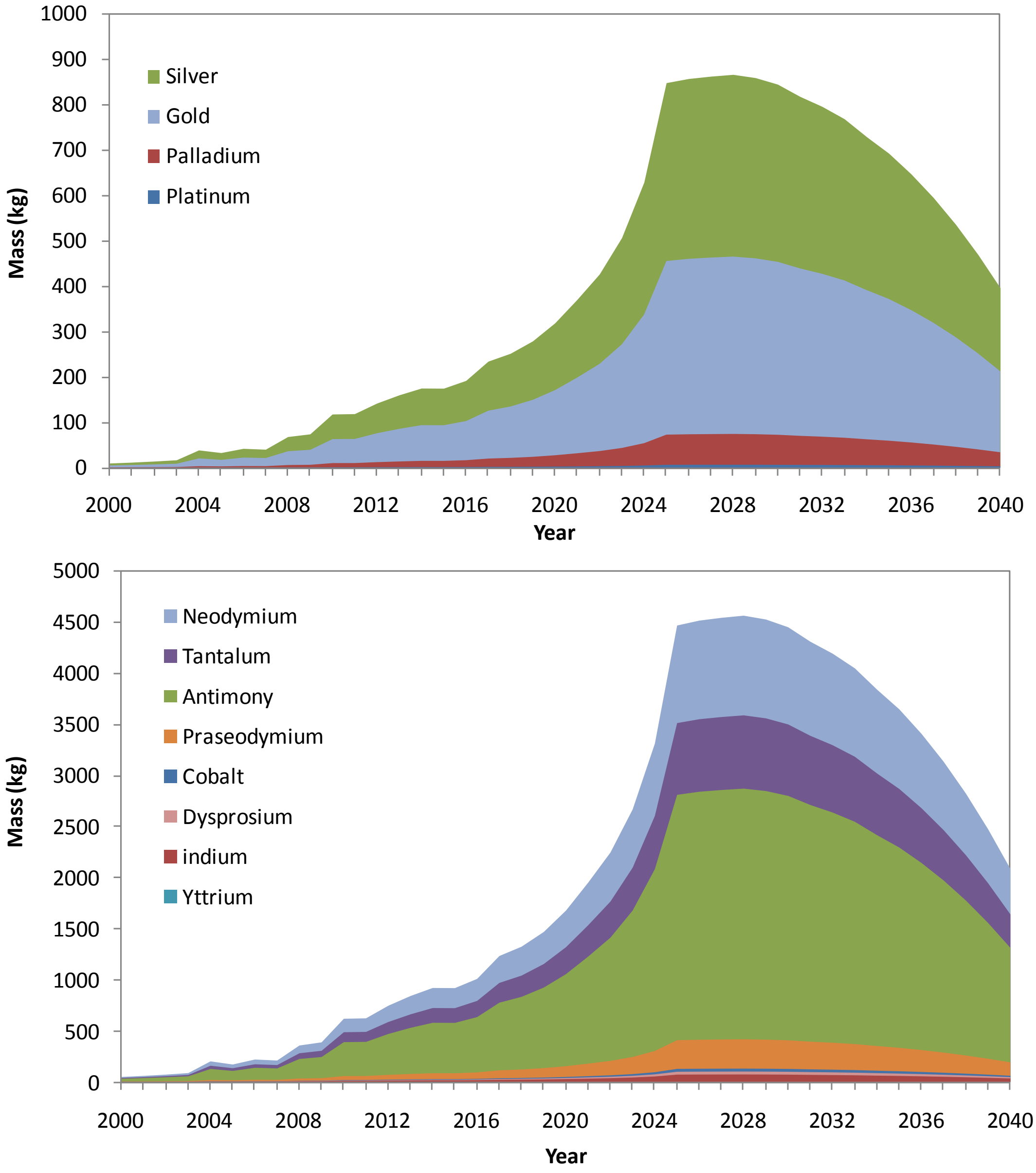

Figure 3.12. Estimation of key metals in obsolete laptops according to B1scenario.

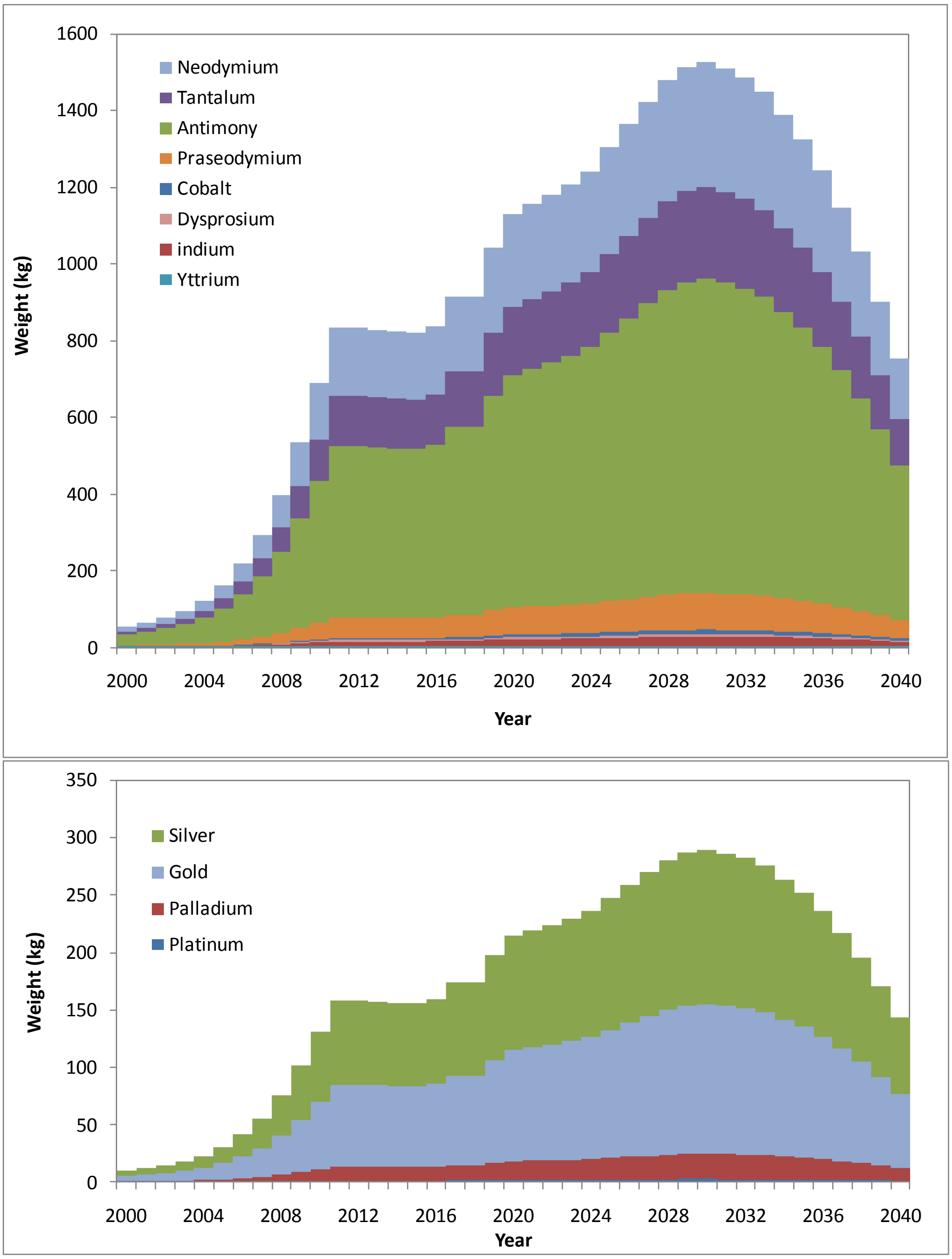

Figure 3.13. Estimation of key metals in obsolete laptops according to scenario B3.

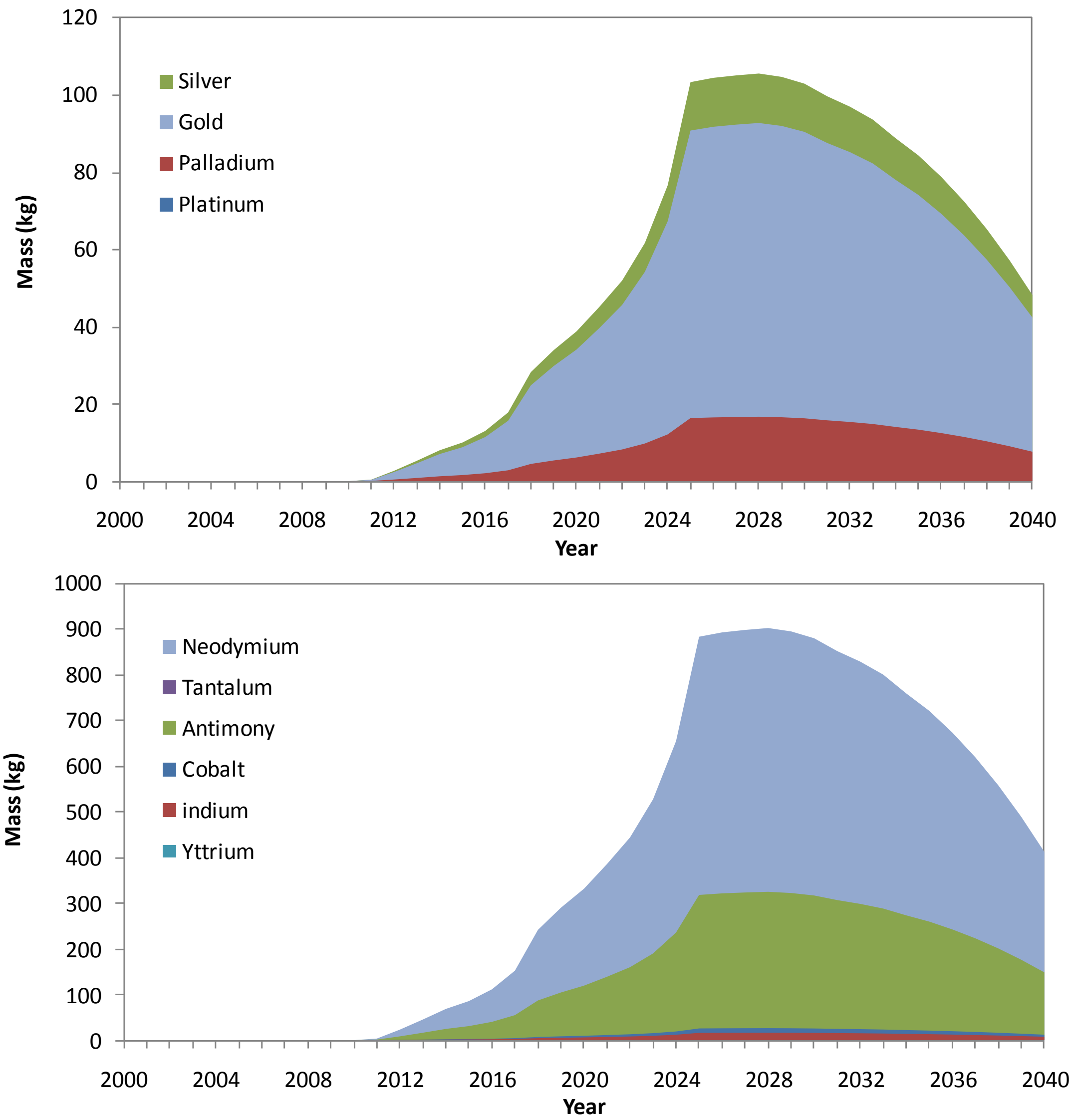

Figure 3.14. Estimation of key metals in obsolete tablets for B1 scenario.

The estimation of CRMs and PMs in generated EoL tablets according to B3 scenario is depicted in Fig 3.15. Concerning EoL tablets the annual growth rate in 2013-2019 varies between 20-73% in B3 scenario. After 2019 the annual growth rate reduces to below 10%. The generated amounts in 2018 were 113 kg Sb, 40.8 kg Nd, 0.2 kg Y, 2.1 kg In, 3.4 kg Co, 2.1 kg Pd, 11.6 kg Au and 13.2 kg Ag. These amounts are predicted to almost triple in 2030. The total accumulated amount in waste laptops in 2020-2040 will be 1.91 t Sb, 5.29 t Nd, 0.01 t Y, 0.10 t In, 0.16 t Co, 0.10 Pd, 0.55 t Au and 5.08 t Ag.

The calculated metal contents depict the potential that exists in the obsolete waste, while there is still a long way till efficient recycling can be accomplished for most of these metals. Indeed, the EoL recycling input for Pd and Pt is currently at 14%, while it is 0-8%for

CRMs. The only exceptions are Sb with a recycling input ratio of 28% and the precious metals Ag and Au with 55% and 20% respectively. It is evident that urban mining is an attractive option in order to meet future resource needs within the circular economy concept (Zeng et al., 2020), but new efficient recycling methods must be applied.

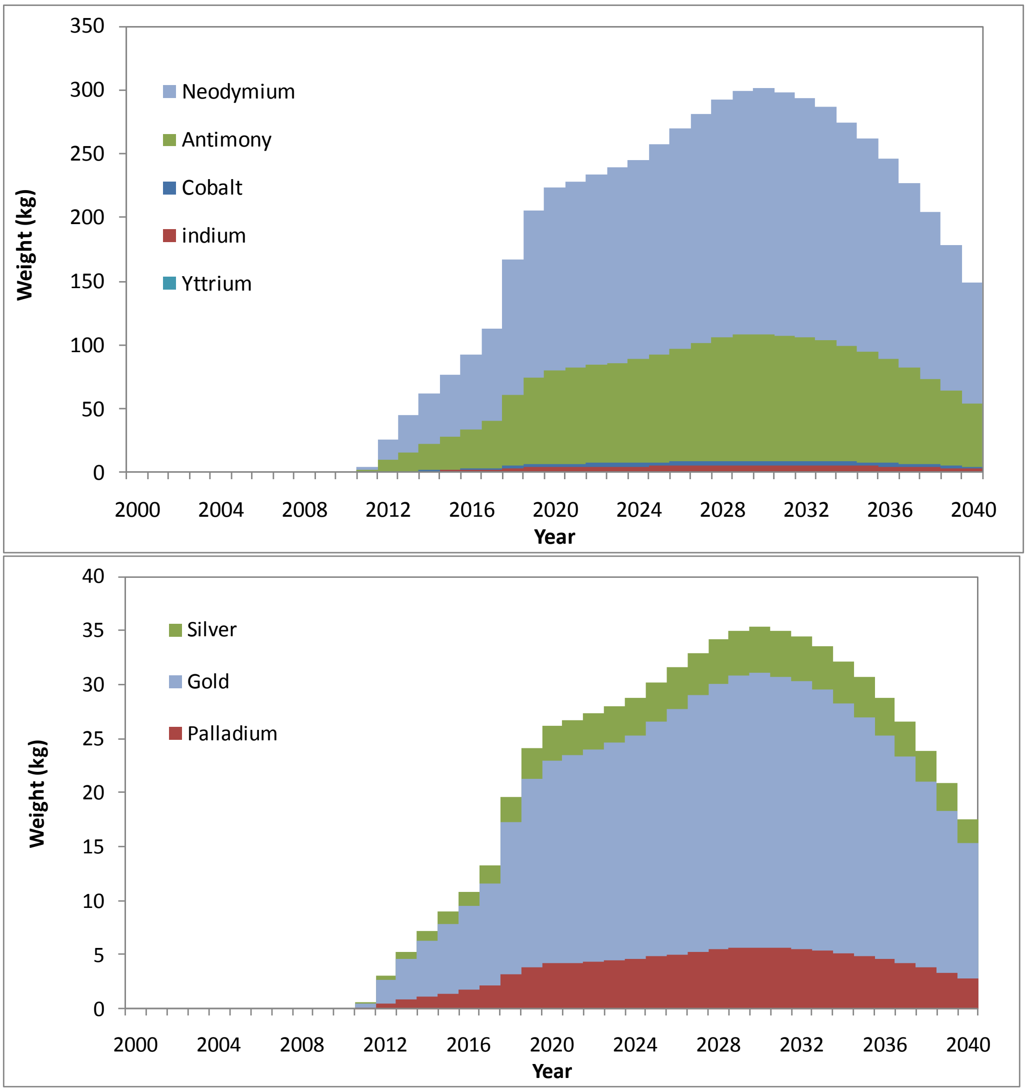

Figure 3.15 Estimation of key metals in obsolete tablets for B3 scenario.

4. Policy implications

This study has significant implications for developing appropriate e-waste policies in Greece, especially for category 3 (ICT devices). The estimation of future EoL laptops (particularly in the long run) is a prerequisite to better organize collection and treatment processes and ensure the appropriate infrastructure. Our study shows that significant waste laptop quantities are anticipated in the near future, far greater than the laptop waste generated in the past. This opportunity of urban mining should not be lost, as great amounts of key metals are contained in this waste.

Moreover, the 2015 National Waste Management Plan (NWMP) states that in 2011 about 73 thousand tonnes of WEEE were generated and 15 thousand were promoted to other countries for further treatment. Also the prediction of e-waste increase in 2011-2020 is 9.1%, while according to our calculations, only EoL laptops will present an increase of about 123-163% according to B1 and B3 scenario and 71-86% in A1 and A3 scenario for the same years. The calculated laptop waste shows that great amounts will be generated in the future. The 2020 NWMP states that there is no reliable method to estimate WEEE and that the EEE POM quantities are used to estimate future quantities, however our study shows that future quantities of e-waste can be reasonably calculated. .

The next important step in e-waste management is waste collection. As far as the collection, recovery and recycling targets in Greece are concerned, these were not met for category 3 in 2018 (collection 44.6%, recovery 83.8% and recycling 73.8%) and it is very unlikely that they will be fulfilled in 2020 since collection target rises from 45 to 65% (based on POM quantities). As illustrated in our work, EoL laptops are among the most important e-wastes from a CRMs and PMs recovering perspective, thus their inefficient collection plan is not promoting circular economy in Greece, as valuable resources are lost. In order to enhance the collection several actions can be adopted to motivate people to participate in WEEE collection instead of storing non functional electronic devices at home. Financial incentives may include vouchers for individuals when they hand in their old devices, discounts for a new buy or reduction in waste fees for residents or communities willing to separate more e-waste at source. For the latter legal regulation at local level is a key factor for the implementation. Also, all municipalities in a prefecture can organize e-waste collection campaigns with prizes for the citizens (Dri et al., 2018). All these actions are important as Greece includes many islands that hinder efficient collection, thus establishing multiple collection points and increasing public awareness is vital.

5. Conclusions

Future EoL laptops amounts in Greece are estimated considering time-varying parameters of penetration rate, population, laptop weight and lifespan. The impact of the coronavirus pandemic on the lifespan of laptops is also assessed. It is concluded, the most important parameter affecting the calculated obsolete laptops amounts is the weight of the devices, followed by the Weibull parameters (lifespan). The estimated CRMs and PMs amounts in these wastes reveal a great potential of key metals recovery. The results point out that significant amounts of EoL laptops will be generated in the future in Greece and existing compilation schemes should be improved to enhance collection as much as possible. In this way valuable resources, like key metals will not be lost and the EU economy can become more material sufficient in accordance with the circular economy concept.

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CHAPTER 4

Forecasting quantities of critical raw materials in obsolete feature and smart phones in Greece: a path to circular economy

Mobile phones represent an ever-increasing waste stream due to the increasing ownership and short lifetime. In particular, smartphones are among the most valuable e-waste because of their extremely high content of numerous key metals, specifically in the printed circuit board and magnets. As feature and smart phones contain different key metals at different concentrations, it is important to distinguish between the two phone types to make reliable estimations. This study presents estimations of obsolete mobile phones quantities, generated in Greece in 1995-2035 and the Critical Raw Materials (CRMs) and Precious Metals (PMs) embedded in them, making a differentiation between feature and smart phones. The dynamic material flow analysis is adopted, the lifespan is evaluated by the Weibull distribution and future sales are predicted by the logistic model incorporating phases of growth, saturation and decline. Then, the future wastes are predicted by the Market Supply A model. According to the results, the generation of obsolete smartphones is constantly increasing, while the waste flows of feature phones are declining. Efficient recycling of obsolete phones (1995-2020) can cover the demand for key metals (Au, Pd, Co) in the new

smartphones for more than a decade in Greece, while the demand for Ag, Sb, Si, Zn, Be, Ti will be covered for more than 15 years. In 2020-2035 the accumulated amounts of CRMs and PMs, only from the smartphone waste, will be 1292.02 and 14.11 tonnes, respectively. The findings can contribute to the management of a valuable e-waste category closing the loop between resources-products-wastes.

1. Overview

As the e-waste amounts are increasing, the estimation of future quantities and the key metals contained in them is of primary importance to policy makers, producers and manufacturers so as to promote actions and legislations related to their sustainable management. In order to cope with the increasing demand for supply resources, recycling of discarded devices can ensure our way to sustainability. To assess the feasibility of urban mining, two questions must be answered: What quantities of key metals will potentially be available in discarded devices in the future? How can we improve the recycling efficiency of the various key metals? The present study aims to answer the first question focusing on mobile phones as they represent a perpetually growing waste stream because of the increasing ownership and short product innovation cycle (Yu et al., 2010). Especially smartphones (and earbuds), followed by laptops and tablets are among the most valuable WEEE because of their extremely high content (in ppm) of numerous key metals (Gaustad et al., 2020). Urban mining is an attractive alternative in dealing with the rising demand for Critical Raw Materials (CRMs) as many key metals in the scrap mobiles have a concentration multiple times more than that of rich primary ores, especially in the printed circuit boards (PCBs) and magnets (Rosa and Terzi, 2016; Bookhagen et al., 2020).

The European Commission initiated in 2008 the Raw Materials Initiative aiming to confront the expanding concern of valuable raw materials for the EU economy. CRMs are the materials that combine a high economic significance to the EU with a great risk of supply disruptions. Currently, many of these materials are only extracted in a few countries, among which China is the leading provider and consumer. This grows the risk of supply scarcity of various important raw materials e.g., rare earth metals (REEs) etc. Furthermore, the EU endorsed the Green Deal Communication (in 2019) accepting access to resources as a vital security issue for realizing its aspiration towards 2050 climate neutrality (EU, Final Report 2020).

CRMs are widely used in electrical and electronic equipment (EEE), specifically in high-tech devices. The renewed 2020 CRMs list includes 30 metals/ materials among which are Cobalt, Palladium, Platinum, Neodymium, Praseodymium, Cerium, Europium, Antimony, Indium, Lithium, and Tantalum which are all embedded in smartphones (Cucchiella et al., 2015; Holgersson et al., 2018). Specifically, smartphones contain at least 22 out of 30 CRMs. Thus, the obsolete phones are considered valuable resources for CRMs and PMs, and the estimation of their future End-of-Life (EoL) quantities is vital for their efficient management to ensure that these resources will not be lost (Charles et al., 2020).

In order to make reliable estimations of the quantities of key metals in mobile phones, differentiating between feature and smart phones is crucial, as these mobile phones have different functionalities and therefore different composition. However, most published cell phone waste projects do not distinguish between feature and smart phones (Polak et al., 2012; Rahmani et al., 2014; Guo et al., 2017) except for two studies (He et al., 2018; He et al., 2021). Nevertheless, it is important to make this distinction because feature and smart phones future flows have different trends. Indeed, in developed countries the feature phone market is declining while the smartphone adoption is constantly rising, indicating that their wastes also follow different trends. Thus, the estimation of waste mobile phones as a single category without considering the different trends and composition of the sub-components may lead to inaccurate results. Also, most published works only deal with waste mobile phones generation or estimation of common metals contained in them, while the research on the present and future flows of CRMs and PMs embedded in waste mobile phones, is extremely limited and concerning only non-European countries (He et al., 2018; He et al., 2021). This study addresses the research gap of reliably estimating future trends of CRMs and PMs in waste cellphones in an EU country, differentiating between smart and feature phones, by assessing the future mobile phone market trends.

To the best of our knowledge, this is the first time a study on waste mobile phone generation and their embedded key metals (CRMs and PMs) in Greece until 2035 (2010-2035) is reported, differentiating between feature phones and smartphones. This study also presents a simple technique for distinguishing between feature and smart phones, easily employed in similar cases in other countries.

Additional objectives are: (i) a detailed estimation of time-varying lifespan values for feature and smart phones in Greece which is absent in the literature. The estimation is achieved by the numerical solution of the corresponding mathematical equations, as suggested in E-Waste Statistics (Forti et al., 2018), since high quality data are available. This is preferable than adopting a survey method that is impossible to apply for an extended period of time (25 years) and can be biased if not properly conducted. To the best of our knowledge, this is the first time this approach is used to determine the lifespan of mobile phones. Also (ii) to estimate CRM and PMs amounts in future waste phones (feature and smart phones) and (iii) assess whether efficient recycling of obsolete phones (1995-2020) can meet the demand for key metals in the new smartphones sales (2021-2035). This information is vital to securing a sustainable and reliable source of raw materials in the EU, which will help to tackle the huge increase in demand for minerals and metals. Moreover, the outcomes will assist the Greek authorities, policy makers and electronic good dealers to improve management of EoL feature and smart phones in Greece, benefiting society and environment and ensuring the pathway to sustainability.

The chapter is organized as follows: Section 1 sets the introduction and background of waste mobile phones estimation. Section 2 presents the methodology adopted in this study and Section 3 provides the analysis and discussion of the results. Section 4 presents the policy issues and relative recommendations while Section 5 displays the conclusions reached from this study.

2. Methodology

2.1 Calculation of obsolete mobile phone amounts in the past years (1985-2010)

The obsolete mobile phone output is expressed as a function of mobile phone inputs and the change in product stock (Time Step model):

$$O_t = S_t - (St_t - St_{t-1}) \qquad (1)$$

Where O_t: obsolete product in year t, S_t: Sales of product in year t, and St_t: stock of product in year t.

Also, the obsolete mobile phone output is given as a function of product inputs and the lifespan probability distribution (Market Supply A):

$$O_t = \sum_{i=0}^{n} (S_{t-i} \times f_i) \qquad (2)$$

Where f_t: lifespan probability distribution function, and n: maximum possible lifespan (Wang et al., 2013).

The lifespan distribution is depicted by the Weibull distribution (Kalmykova et al., 2017) and is given by:

$$f(t, a, \beta) = \frac{a}{\beta^a} t^{a-1} \exp(-\left[\frac{t}{\beta}\right]^a) \qquad (3)$$

where α: the shape parameter that describes the gradual ageing of the product, and β: the scale parameter describing the characteristic life of the product that is, it defines the age when 63.2% of the products are expected to have failed.

The average lifespan or else Mean Time to Failure (MTTF), is estimated by:

$$MTTF = \beta \times \Gamma(1 + 1/\alpha) \qquad (4)$$

where Γ: gamma function.

The product lifespan in this work represents the total time the mobile phone stays in the possession of a person, including stock time after the product ceases to be used or donation to another person. Some studies approach mobile phone lifespan as “service lifespan” not taking into account storage of non-functional phones (Rahmani et al., 2014; Guo et al., 2017). The total lifespan is more essential information than “service lifespan” in designing the waste management and recycling system (Murakami et al., 2017). After the mobile phone total lifetime has elapsed the product is discarded in appropriate collecting centers.

2.2 Estimating future sales of mobile phones

In order to assess mobiles phone sales, the three parameter logistic model was employed. For a mature market like feature phones in Greece, the logistic curve includes the phases of growth, saturation (eq. 5), and decline (eq. 6). The smartphones market is at a growing phase and is described by eq. (5). The product sales are expressed by (Althaf et al., 2019):

$$S_t = \frac{K}{e^{-(r_1 t + C_1)} + 1} \qquad \text{when} \qquad t \le t_{peak} \qquad (5)$$

$$S_t = \frac{K}{e^{-(-r_2 t + C_2)} + 1} \qquad \text{when} \qquad t \ge t_{peak} \qquad (6)$$

where

$$C_i = \ln\left(\frac{S_{0i}}{K - S_{0i}}\right) \qquad (7)$$

and K: maximum sales units, S_{0i}: sales at time 0, i:1 or 2.

As demonstrated by Meyer et al. (1999) the growth rate, r, is substituted by ln(81)/Δt, where Δt is the time required to grow from 10% to 90% of K, and –C/r is substituted by t_m, the time needed to reach K/2 (Althaf et al., 2019).

The solution to this equation is given by non-linear regression by minimizing the Objective Function (OF):

$$OF = \sum_{0}^{t} (S_t - S_t^{'})^2 \qquad (8)$$

Where $S_t^{'}$: historical sales data. Furthermore, the fit of the calculated curve to the historical data curve, at the optimal set of parameters, was determined by:

$$Deviation\_\% = 100 \frac{\sqrt{OF(z-N)}}{\max(S_t^{'})} \qquad (9)$$

where z: number of data points, N: number of parameters engaged in the model.

The penetration rate for year t is calculated by:

$$N_t = \frac{St_t}{Q_t} \qquad (10)$$

where Q_t: population at time t.

Initially, the historical amount of mobile phone waste was calculated by Eq. (1) using sales and stock data. Then, non-linear regression was applied to estimate α, β for various years by minimizing the Objective Function (OF_1):

$$OF_1 = \sum_{t,i}^{n} \{ [S_{t-i} \times (F_{i+1} - F_i)] - [S_t - (St_t - St_{t-1})] \}^2 \qquad (11)$$

Where Fi: Weibull cumulative distribution function that gives the cumulative probability a product fails at time.

The deviation for the curve fitting was calculated by (11) using O_t, as found by (1), in the denominator. Several constraints were applied in order to solve the problem as α, β>0 and 1<α<4.5. The shape parameter α was taken as α<4.5, because α>4.5 is unlikely as this denotes rapid wear-out of products associated with severe problems in manufacturing process. Also, α <1 indicates infant mortality due to quality problems in components or manufacturing, therefore α was considered α >1 (Zhu, 1991).

Additionally the obsolete amount calculated in each year by Eq. (2), should be equal to the amount calculated by

$$\sum_{t}^{n}[S_t-(St_t-St_{t-1})] \qquad (12)$$

for the same years.

Once the Weibull parameters *(α,β)* were calculated for several years (1985-2010), their trends were extrapolated in the future. The future obsolete mobile phones amounts (2011-2035) were calculated by iteratively solving Eq. (2) and (1) (Liu et al., 2006).

2.3 Differentiation between feature and smart phones

Although total sales data are available, it is important to differentiate between feature and smart phone sales. The calculation of feature and smart phone sales data when only total sales data are available is described hereafter. First the total historical possession data are calculated by eq. (10). Mobile phones manufactured and sold before 2004 are treated as feature phones because smartphones were first introduced in Greece in 2004. Therefore, sales data up to 2004 only applies to feature phones. The adoption growth of smartphones is described by the logistic model, which is, in the beginning, identical to the exponential growth. The data on smart phone penetration in Greece for the years 2011 and 2017 were collected from EKKT (2011) and Newzoo (2017). These were well-fitted by exponential growth to fill in the missing data from 2004-2017. Then, the feature phones possession was calculated by deducting from total possession the smartphone possession. Also, the annual percentage of smart phone possession was calculated. Sales data before 2004 regard only feature phones. The trend in historical sales data agrees to the historical possession trend as seen in Fig. 4.1, and the percentage of the annual sales agrees to the possession one.

The percentage of sales concerning smartphones from 2004-2015 was calculated by multiplying the smart phone possession percentage by total sales. Furthermore, the feature phone sales (after 2004) were calculated by deduction of total sales minus smart phone sales. It can be clearly observed that the feature phone sales data have reached a peak and are currently on the decay phase, while the smartphone sales are in the growth phase (see section 4.2 and Fig. 4.2). Feature phone sales were simulated by the logistic curve including phases of growth, saturation, and decay. Smartphone sales till 2035 were simulated by the logistic curve (only growth phase till saturation). Also, a constraint applied was that sum of sales equals historical sales (data till 2015). The European prediction for coronavirus pandemic affecting annual smart phones sales in 2020, 2021 and 2022 was taken into account (-24, -12, -4%, respectively) (Statista, 2020; Counterpointresearch, 2020; Android authority, 2020).

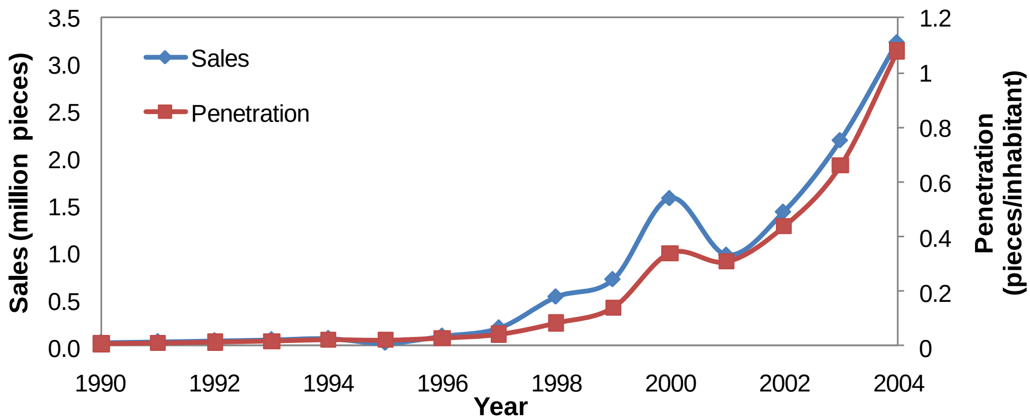

Figure 4.1. Histrorical penetration and sales data of feature phones.

2.4 Substance flow analysis of obsolete mobile phones

The components of interest in mobile phones are PCBs, LCD or LED screens, microphone and speakers and lithium-ion batteries. Material composition of feature and smart phones is presented in Table 4.1. As can be observed, there are significant differences in CRMs and PMs concentration between feature and smart phones. Therefore, the key metals content is calculated separately for obsolete feature and smart phones. The quantity of metal i in year t is calculated by (Guo et al., 2017):

$$m_{i(t)} = C_i \times O'_t \qquad (13)$$

Where C_i: quantity of metal i in year t per device, O'_t: obsolete pieces in year t.

2.5 Data sources and collection

Past sales and stock data were acquired from Statistics Netherlands which are retrieved from Eurostat website. The applied categorization follows the European Prodcom (Production Statistics database) and Eurostat Ramon Database (Van Straalen et al., 2016).

Population data were obtained from ELLSTAT and CIA-World Factbook.

The average mass of mobile phones (1995-2010) was adopted from the EU-28 data report (Magalini et al., 2014). The average mass of a mobile phone is reducing through the years (0.6 kg in 1995, while 0.1 kg in 2005). The average mass was taken as 0.1 kg from 2005 to 2035 for feature phones and 0.15 kg from 2004 to 2035 for smartphones, after extensive on-line assessment of technical characteristics. Guo et al. (2017) considered the average mass of mobile phones in China as 0.15 Kg from 2015-2025.

All calculations were performed by Excel software and Solver add-in program.

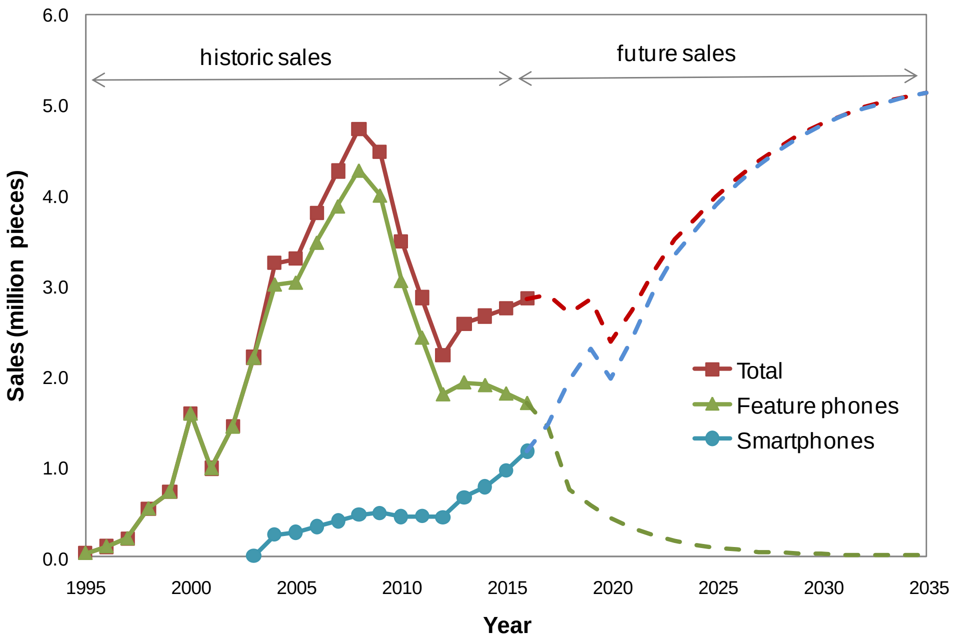

Figure 4.2 Feature phones and smartphone sales: historical data and projections (dashed lines).

Table 4.1 Composition of feature and smart phones.

Element (g/unit)	1	2	3	4	Average feature phones	5	6	7	8	9	Average smart phones
	Feature phones					**Smartphones**					
Fe	4.66	9.74	11.0		8.47	15.33	0.88		8.0		8.07
Si						9.32					9.32
Mg						7.84	5.54				6.69
Cu	14.99	21.00	26.0		20.66	6.37	15.12	10.0	14.0		11.37
Al	4.32	4.92	12.0		7.08	6.33	22.18	3.0	2.9		8.60
Ni	1.72	1.41	1.0		1.38	2.47			1.50		1.98
Cr	0.46	0.43			0.45	1.96					1.96
Sn	0.61	0.86			0.74	0.62	1.21	1.0			0.94
Zn	0.60	0.80	4.0		1.80	0.57			1.0		0.79
Sr						0.45					0.45
Ba						0.35					0.35
W						0.26	0.44				0.35
Mn						0.18					0.18
Ti			1.0		1.00	0.11			1.0		0.56
Co			3.9		3.90	0.05	5.38	5.0	6.30	6.3	4.61
Ta		0.005			0.005	0.04	0.02				0.03
Zr						0.04					0.04
Mo						0.04					0.04
Au	0.03	0.05	0.02	0.02	0.03	0.02	0.03		0.04	0.03	0.03
V						0.01					0.01
Ag	0.51	0.43	1.0	0.25	0.55	0.01	0.31		0.24	0.31	0.22
Pb	0.54	0.38	1.0		0.64	0.01			0.60		0.30
Ga						0.01	0.0004				0.004
Nb						0.004					0.004
As		0.001			0.001	0.003					0.003
In						0.003	0.01				0.01
Pd	0.045	0.02	0.01	0.01	0.02	0.002	0.01		0.02		0.01
Li						0.001		4.0			2.00
Hf						0.001					0.001
Bi		0.038			0.038	0.001					0.001
Pt						0.001			0.004		0.002
Sb		0.11			0.11	0.0004			0.08		0.04
Ge						0.0003					0.0003
REE (sum)						0.30					0.30
Nd							0.05		0.05	0.05	0.05
Pr							0.01		0.01	0.01	0.01
Gd							0.0002				0.0002
Hg			1.0		1.00						
Eu							0.0001				0.0001
Ce							0.00003				0.00003
Be		0.004			0.004				0.003		0.003
Cd		0.00003			0.00003						

1. Geyer & Blass, 2010
2. Huisman, 2004
3. Cucchiella et al., 2015
4. Buchert et al.,2012
5. Bookhagen et al., 2020
6. Manhart et al., 2016;
7. Basel Convention, 2012
8. Cucchiella et al., 2015
9. Buchert et al.,2012

3. Analysis and Discussion

3.1 Historical possession of mobile phones in Greece

The mobile phone subscriptions in Greece, the EU-28 and developed countries was 118.8, 118.4 and 128.9 subscriptions per 100 inhabitants in 2019, respectively (ITU, 2019a; ITU, 2019b). However, once the number of subscriptions in Greece in the period of 1993-2019 is considered, a peak is observed in 2008, corresponding to almost 138 subscriptions per 100 inhabitants (ITU, 2019b). Every subscription coincides to a mobile phone, although nowadays there exist also dual-sim phones.

Fig. 4.3 shows the total procession of mobile phones in Greece from 1995 to 2015 and the number of subscriptions. It is observed that the annual growth rate of mobile phone ownership in Greece was significant in the past years, i.e. almost 68.4% and 142.5% increase in 1999 and 2000, and then slowed down below 10% in the following years. Indeed, the average possession in 1999 was about 1,513,000 units, while in 2000 and 2004 it was 3,668,400 and 11,822,400 units, respectively.

A comparison of the subscription with the total possession data reveals information about possible storage of non-functional mobile phones at homes. It is observed that in 2004-2012 the subscriptions per inhabitant (SpI) are significantly lower than possession per inhabitant (PpI), implying that mobile phones are not delivered to the e-waste management system. However, after 2012 SpI almost coincide with PpI and in 2015 SpI (1.26) are slightly higher than PpI (1.20), meaning that some inhabitants own one mobile phone but more than one subscription, and storage of non functional mobiles is declining.

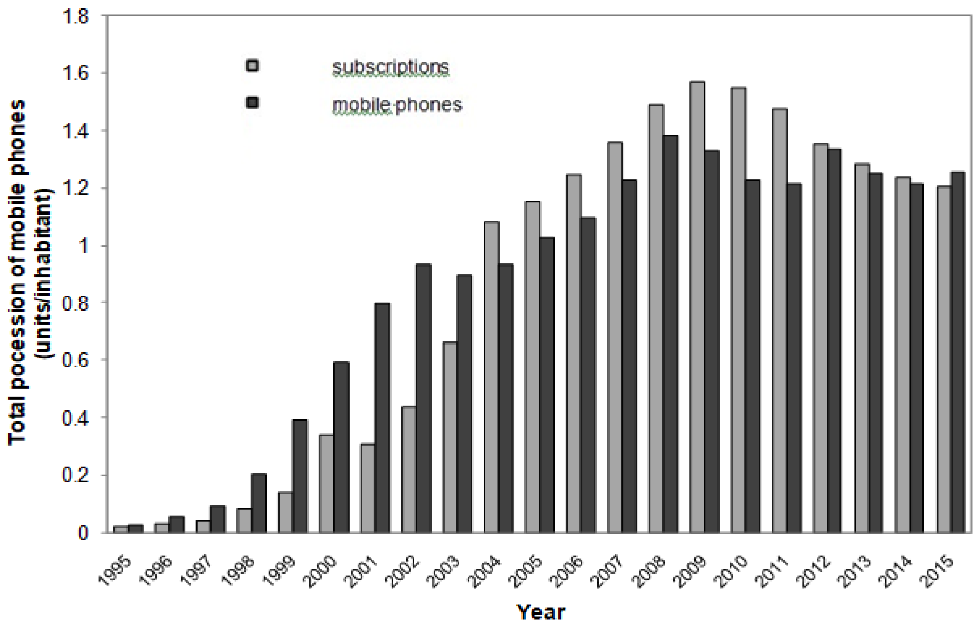

Figure 4. 3 Total possession of mobile phones and number of subscriptions per inhabitant.

3.2 Estimation of historical and future mobile phone sales by logistic model

In Greece the feature phone market has reached the saturation phase and is currently declining, while the smartphone market is growing. Mobile phone sales were simulated by the logistic curve, as described in Section 2.2, including phases of growth, saturation and decay for feature phones, and growth followed by saturation phase for smartphones (till 2035). The results are depicted in Fig. 4.2. The maximum sales for feature phones were 4,353,000 items in 2008, while Δt and t_m were 9.09 and 11.74 years, respectively. For the decay phase the logistic curve coincided to the historical data for Δt and t_m, -12.95 and 4.87 years, respectively, showing a sharper decrease rate compared to the growth phase. This is reasonable because during the feature phone growth phase there was no competing mobile phone technology, while after 2008 smart phones had already been introduced and started substituting the old mobile phones, causing a rapid decay rate in feature phone market. Feature phones sales will decline rapidly (below 100,000 units) in 2024 and even more (below 10,000 units) in 2031, while in 2035 the feature phone sales will be possibly out of the market.

The total peak sales were 4,732,000 in 2008 (historical data) representing sales of both feature and smart phones. Thus, the peak sales of smartphones in the future are considered slightly higher than that since only smart phones will be sold. The influence of the pandemic on sales was considered in 2020-2022 (section 2.5).

The maximum sales value for smartphones is 5,300,000 units, while Δt and t_m are calculated at 26.37 and 20.34 years, respectively, indicating that the saturation phase of smartphones is in the rear future, i.e., after 2050. The annual growth rate of smartphone sales was between 15-29% till 2008, 50% in 2009, 48% in 2013 and from 2024 onwards it will slow down below 10%.

3.3 Estimation of the lifespan of mobile phones

The Weibull parameters are estimated for the years 1985-2010 using the methods discussed in Section 3.1. Once the Weibull shape and scale parameters are calculated, the average lifespan (MTTF) can be estimated. Lifespan is a dynamic parameter that depends on the socio-economic situation in each country, the diffusion of new technology, as well as the habits and awareness of the people (Islam and Huda, 2019a). This is depicted by the estimated MTTF values for Greece, which are illustrated in Fig. 4.4.

As observed, the total lifespan of mobile phones in Greece is constantly decreasing, from about 8.1 years in 2000 to 5.2 years in 2010, meaning that more waste is generated (Table 4.3). The decrease in these years is almost linear. The detailed lifespan values for mobile phones, reported in relevant literature studies, are presented in Table 4.4.The differences observed in average lifespan are attributed first to the definition of lifespan in each study ("service" lifespan or "total" which includes storage time) as well as differences in consumer behavior, economic conditions and take-back systems in each country. Our results agree well with Forti et al. (2018) who reported mobile lifespan of 5.7 years for Netherlands, France and Belgium and 5.1 for Italy in 2016.

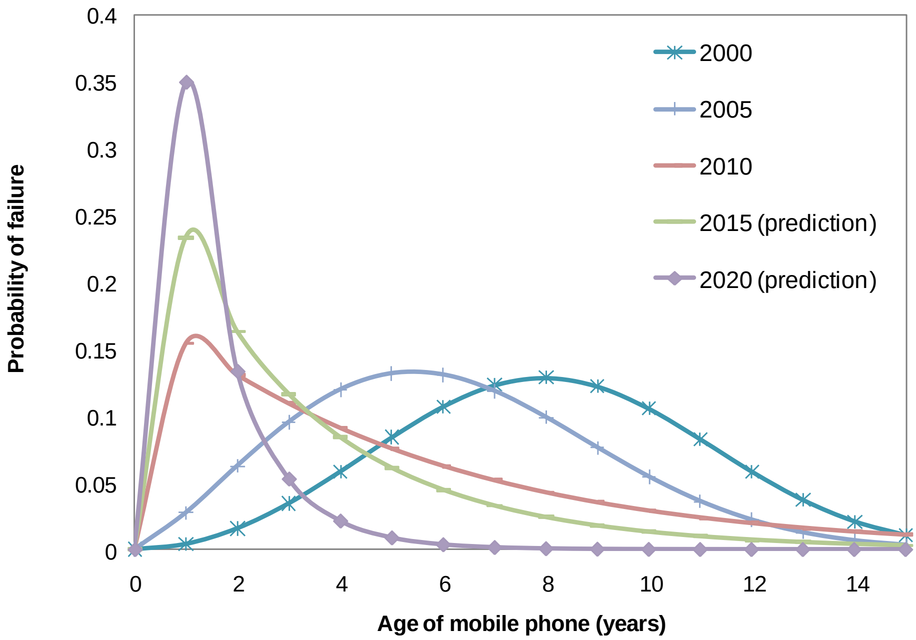

Figure 4. 4 Lifespan of mobile phones in historical years and future predictions.

Table 4.2 Calculated mobile lifespan values

Year	α (shape parameter)	β (scale parameter)	Lifespan (MTTF)	dev(%)
1995	2.34	6.96	6.17	2.29
1996	2.25	6.58	5.83	7.18
1997	2.46	7.67	6.80	10.67
1998	2.52	8.69	7.72	12.51
1999	2.98	9.13	8.15	12.76
2000	2.99	9.11	8.14	12.01
2001	2.93	8.82	7.86	10.87
2002	2.78	8.44	7.52	9.61
2003	2.62	8.03	7.13	8.39
2004	2.44	7.55	6.70	7.52
2005	2.26	7.02	6.22	7.05
2006	2.08	6.52	5.77	6.88
2007	1.70	6.13	5.47	7.27
2008	1.47	5.85	5.30	8.47
2009	1.37	5.59	5.11	10.69
2010	1.03	5.31	5.25	15.14

Table 4.3. The lifespan values of mobile phones as reported in relevant literature studies.

Reference	Country of study	Method / period	Lifespan value (years)	Service or total lifespan
Rahmani et al., 2014	Iran	Telephone interviews/-	2-5	service
Guo et al., 2017	China	Questionnaire survey/ Dec. 2014-April 2015	1.76	service
Deng et al., 2017	Hong Kong	Questionnaire survey/ October 2014-March 2015	1.92	service
Polak and Drapalova, 2012	Czech Repubic	Survey (2008) and sampling of 3362 mobile phones in 2010.	7.99	total (including storage)
Wang et al., 2013	Netherlands	Survey (2006-2007)	8.95 (1995) 9.62 (2005)	total
Forti et al. (2018)	Netherlands, France & Belgium	Survey/-	5.7 (2016)	total
Forti et al. (2018)	Italy	Survey/-	5.1 (2016)	total
This study	Greece	Numerical solution of Eq. (1) and (2)	8.1 (2000) 5.2 (2010)	total

Further, in this work, the estimation of lifespan distribution parameters for 2010-2035 was accomplished by extrapolating the existing trend (1985-2010) instead of adopting a fixed value. After 2004, specific values for feature and smart phones were calculated by incorporating the percentage of smartphones in total mobile stream.

The shape values for feature phones were optimized for historical years according to eq. (2). Thus, different lifetime was estimated for feature phones and smartphones. The shape and scale parameters decrease linearly over time from 2004 to 2010 (R^2>0.9). The scale parameter was extrapolated to the future until 2020 and then kept constant at 2.6 years for feature phones and 1.6 years for smartphones. Then, considering that lifespan is taken as 'total lifespan' including storage, the scale parameter was not allowed to reduce anymore and was further kept constant. The shape parameter was extrapolated to the future and then kept constant further on for smartphones at 1, while for feature phones it was optimized at 1.46. Values of shape parameter less than 1 are associated with infant mortality of the devices that is, failure in the first year of their life, thus the shape parameter was not allowed to decline and was kept constant further on.

3.4 Estimation of obsolete mobile phones generation

The generation of obsolete feature phones (1995-2035) and smartphones (2004-2035), calculated by iteratively solving eq. (2) and (1) of Section 3.1, is depicted in Fig. 4.5 (both in mass and units). The information is important since the number of the devices is huge; their total obsolete amount by mass is not that great due to their small size. Since the feature phone market has surpassed the saturation phase in Greece, it is observed that feature phone waste flows are declining in future years. Similar results of declining waste flows for markets with competing technologies are reported by Althaf et al. (2019).

As presented in Fig. 4.5, there was a 42% increase between 2005-2011 in the flow of obsolete feature phones, reaching a peak in 2011 and then decreasing by 27.3% in 2015, followed by a further reduction of 63.6% in 2020. About 3.2, 2.4 and 0.85 million units of obsolete feature phones were discarded in 2011, 2015 and 2020, corresponding to 321, 233.4 and 85 tonnes, respectively. In 2025 and 2030, the amount of generated obsolete pieces will be 0.19 and 0.04 million units or 18.5 and 3.9 tonnes, respectively. The accumulated amounts in 1995-2009 and 2010-2020 were 19.5 and 24.6 million feature phones, respectively, while the amount will be only 2.3 million units in 2021-2035.

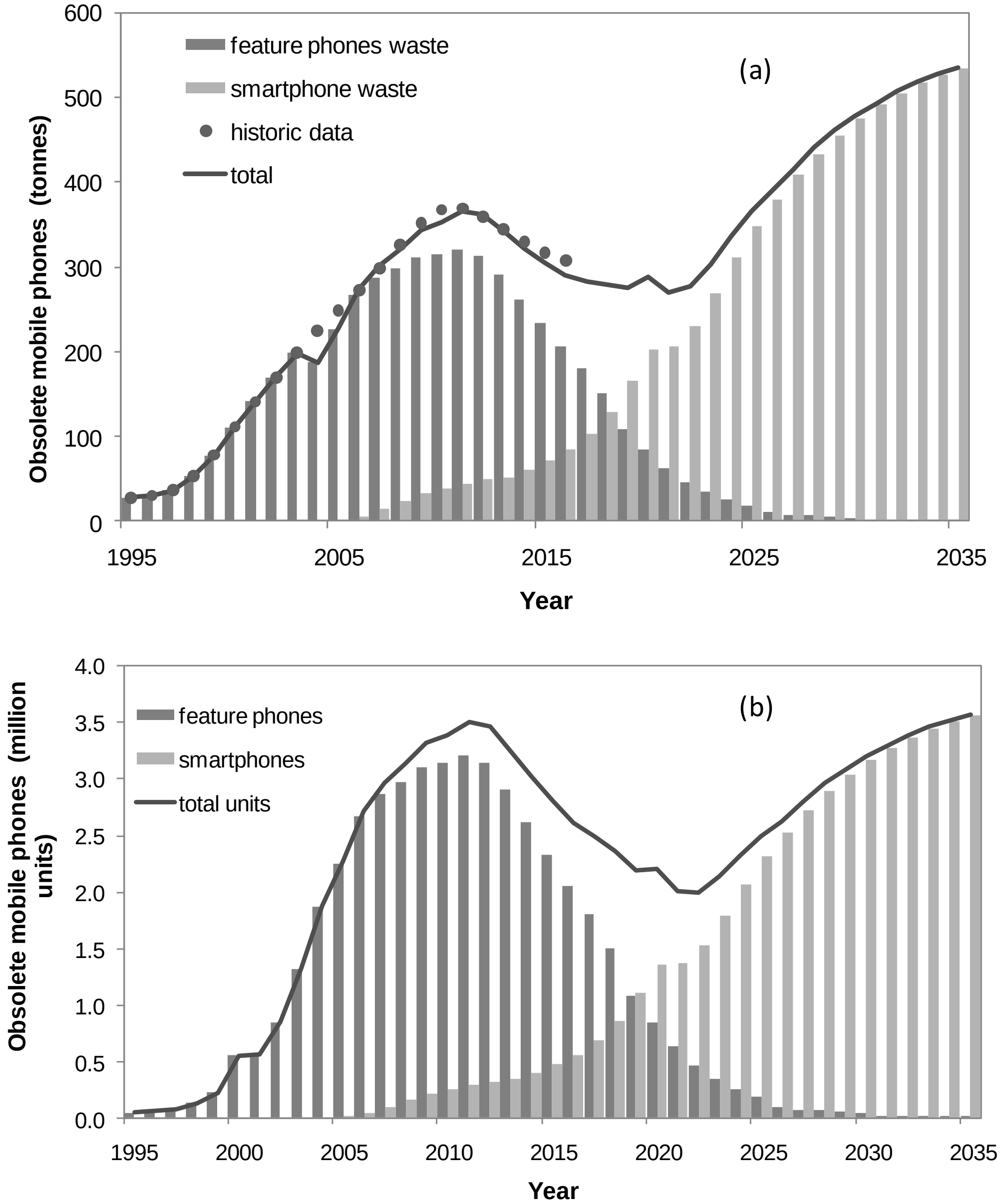

Figure 4. 5 Calculated amounts of obsolete feature phones and smartphones by (a) mass and (b) units.

On the contrary, the generation of obsolete smartphones is continuously increasing as seen in Fig. 4.5. Starting from 2010 there is, every five years, a continuous increase of 86.7% (2015), 186.9% (2020), 70.8% (2025), 36.3% (2030) and 12.5% (2035) of obsolete smartphones (tonnes). About 0.47 and 1.35 million units of obsolete smartphones were discarded in 2015 and 2020 corresponding to 71 and 203 tonnes, respectively. In 2025 and 2030, 2.3 and 3.2 million units of obsolete smartphones will be discarded corresponding to 347 and 475 tonnes, respectively. The accumulated amount in the years 2010-2020 was 6.6 million units (corresponding to 997.3 tonnes), while the amount will be 40.7 million units in 2021-2035 corresponding to 6089.3 tonnes, respectively. The generated obsolete smartphone pieces in 2010-2020 and 2021-2035 correspond to 21.1% and 94.65% of the total mobile waste. Similar percentage of smartphone waste, 21%, was calculated in India during 2009-2019 (He et al., 2021) while the corresponding percentage in 2020-2030 in India was estimated at 49%. He et al. (2018) estimated in China, 81% smartphone waste in the period from 2017 to 2035.

The total generated waste mobile phone amount in 2010 in Greece was 0.03 kg per inhabitant, while Wang et al., (2013) reported 0.02 kg per inhabitant in the Netherlands for the same year. The total amount (kg per inhabitant) will be about 0.034 till 2025, but as smartphones will continuously substitute feature phones the amount will increase to 0.045 in 2030 and 0.051 kg per inhabitant in 2035. Polak and Drapalova (2012) estimated 6.5 and 26.3 million mobile phones in years 2000-2010 and 2010-2020, respectively, in the Czech Republic compared to 23 and 27.9 million mobile phones estimated in Greece. Ongondo and Williams (2011) estimated 18 million units in 2008 in the United Kingdom, corresponding to 0.29 obsolete pieces per inhabitant. He et al. (2018) calculated for China 109.5 and 719.8 million obsolete phone units in 2008 and 2025 or 0.08 and 0.49 PpI, respectively. He et al. (2021) calculated for India 181 and 224 million obsolete phone units in 2020 and 2035 or 0.13 and 0.14 PpI, respectively. In Greece, it is calculated 2.2 and 3.6 million obsolete phone units in 2020 and 2035 or 0.21 and 0.34 PpI, respectively.

3.5 Estimation of CRMs and PMs in obsolete mobile phones

An estimation of CRMs and PMs content in the obsolete feature and smart phone amounts is presented to illustrate the potential recovery of key metals. Table 4.1 presents CRMs and PMs content in feature and smart phones from various sources. Smartphones include many valuable metals at high concentrations, while feature phones contain less CRMs. Although future changes in the product design may slightly affect the CRMs and PMs content, the results are indicative of the valuable materials embedded in these electronic devices.

The estimation of key metals for obsolete smartphones from 2005 to 2035, calculated using the methods in section 3.4, is illustrated in Fig. 4.6. Regarding smartphones, the annual growth rate of key metals ranges from 37-310% (2004-2009), 10-30% (2010-2020), and 1.5-17% (2021-2025). Afterwards, the annual growth rate falls below 10%. The estimated amounts in 2020 of Si, Al, Mg, Co, Ti, Sr, W, Nd, Sb, Ta, V, Pr, In, Ga, Nb, Be and the precious metals Pd, Pt, Au and Ag are 12623, 11675, 9061, 6244, 758, 609, 474, 68, 54, 41, 16, 14, 8, 5.7, 5.6, 4 kg and 27, 2.7, 41, 298 kg, respectively, while the values in 2035 are 2.6 times more. The total accumulated amount of Si, Al, Mg, Co, Ti, W, Nd, Sb, Ta, V, Pr, In, Ga, Nb, Be and the precious metals Pd, Pt, Au and Ag in obsolete smartphones in 2020-2035 will be 391, 362, 280.6, 193.4, 23.5, 18.9, 10.9, 2.1, 1.7, 1.7, 0,5, 0.42, 0.3, 0.2, 0.17, 0.13 tonnes and 0.8, 0.08, 2.7, 1.3, 9.23 tonnes, respectively, which are 7.2 times more than the accumulated amounts in the last 15 years (2004-2019). Smartphones are considered urban resources of Rare Earth Elements (Nd, Pr, Eu, Ce, Gd) and precious metals (Pt) which are not present in feature phones, therefore high effort should be given on their recycling.

As for obsolete feature phones, the assessment of key metals, calculated according to the method in section 3.4, is presented in Fig. 4.7. Feature phones are significantly richer in Ag and Sb, than smartphones, thus significant amounts can be collected and recycled. Antimony as a semi-conductor it is used in the industry as a dopant for Si wafers. It is a hazardous substance among others (mercury (Hg), beryllium (Be), lead (Pb), cadmium (Cd), arsenic (As) etc. (Hagelüken & Corti, 2010). Most of the hazardous substances appear in The RoHS Directive (2011/65/EU) which regulates that toxic substances are increasingly phased out in newer devices

(RoHS Directive, 2011). For example, the Pb content per device is double in feature phones compared to smartphones. Almost all hazardous substances appear in larger concentration in feature phones compared to smartphones, except for As. In feature phones, the annual growth rate of key metals ranges from 32-150% (1995-2000) and 1-56% (2001-2011). Then, the annual rate declines and is always negative (2012-2035). The generated amounts in 2011 of Al, Co, Ti, Sb, Ta, Be and the precious metals Pd, Au and Ag are 22720, 12515, 3209, 353, 16, 12.8 kg and 64.2, 96.3, 1765 kg, respectively, while the values in 2020 are almost 4 times less. The total accumulated amounts of Al, Co, Ti, Sb, Ta, Be and the precious metals Pd, Au and Ag in obsolete feature phones in 1995-2011 were 184, 101, 26, 3, 0.13, 0.1 tonnes and 0.5, 0.8 and 14.3 tonnes, respectively, which are 0.79 times more than the accumulated amounts in the next 23 years (2012-2035).

Fig. 4.7 presents the total amount of key metals in obsolete feature and smart phones and their future demand in new smartphones for the period 2021-2035. By efficient recycling (100%) of obsolete phones (1995-2020), the demand for key metals Au, Pd, Co and Ag, Sb, Si, Zn, Be in the new smartphones can be covered for more than a decade and more than fifteen years in Greece, respectively. According to the estimated smartphone sales, the demand (from 2021) for Pd could be covered till 2033, for Au till 2033, for Co till 2032 and for Al almost till 2032. Also, the accumulated Ag is almost two times more than the quantity required for new smartphones according to sales in 2021-2035, while Sb, Si, Zn, Be and Ti are 2.13, 1.13, 1.43, 1.11 and 1.42 times more, respectively. In the case of Ta only the 24% of the demand is covered.

The calculated metal contents depict the great potential for their recovery, but there is still a long way till efficient recycling can be accomplished for most of these metals. The EU EoL recycling rate for Pd and Pt is currently 14%, while it is 0-8% for CRMs. The only exceptions are Co and Sb with a recycling rate of 22 and 28% and the precious metals Ag and Au with 55% and 20%, respectively (EU, Final Report 2020). It is evident that urban mining is an attractive option to meet future resource needs within the circular economy concept, but new efficient recycling methods must be applied (Zeng et al., 2020).

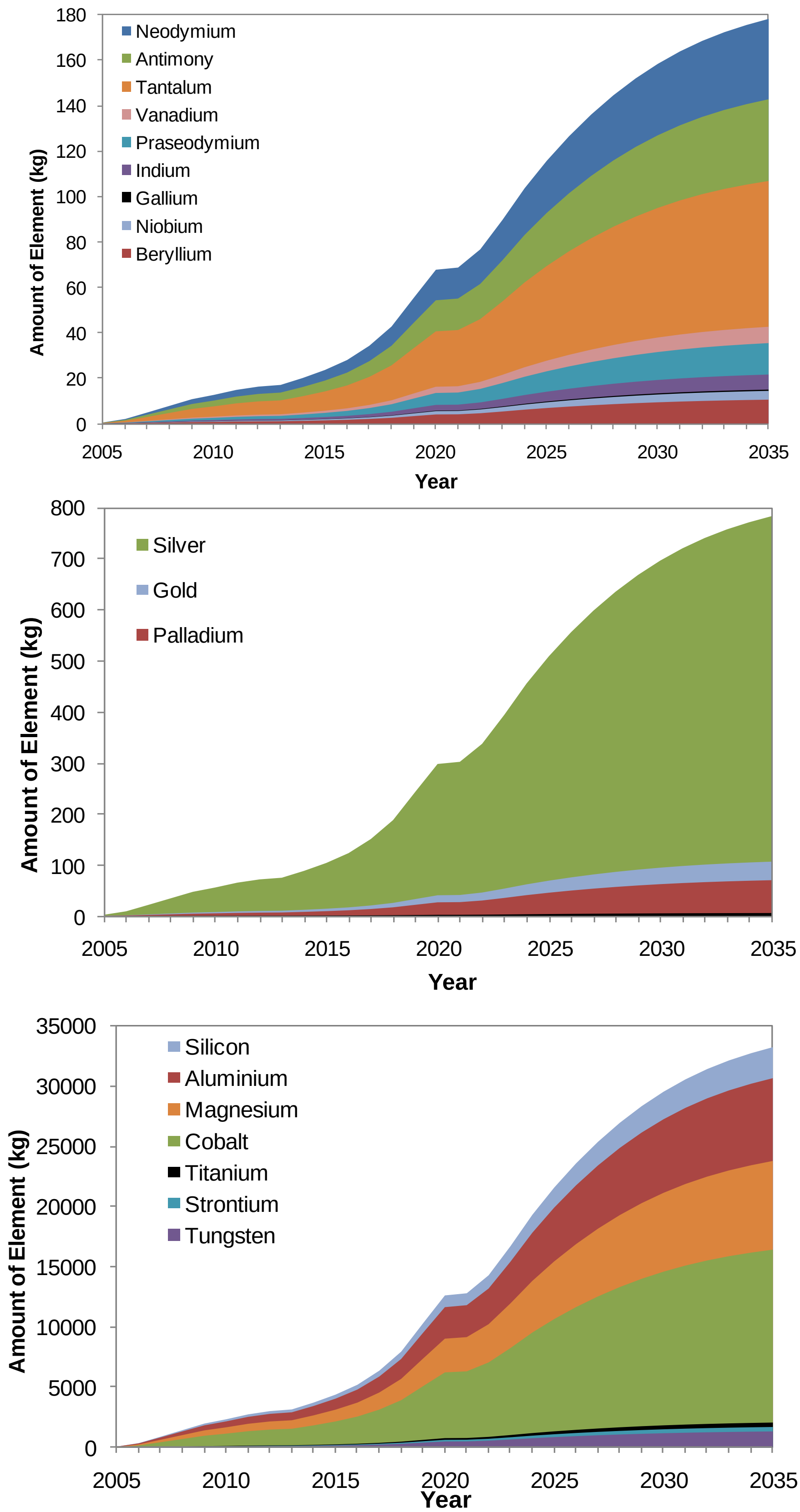

Figure 4. 6 Amounts of key metals in smartphones.

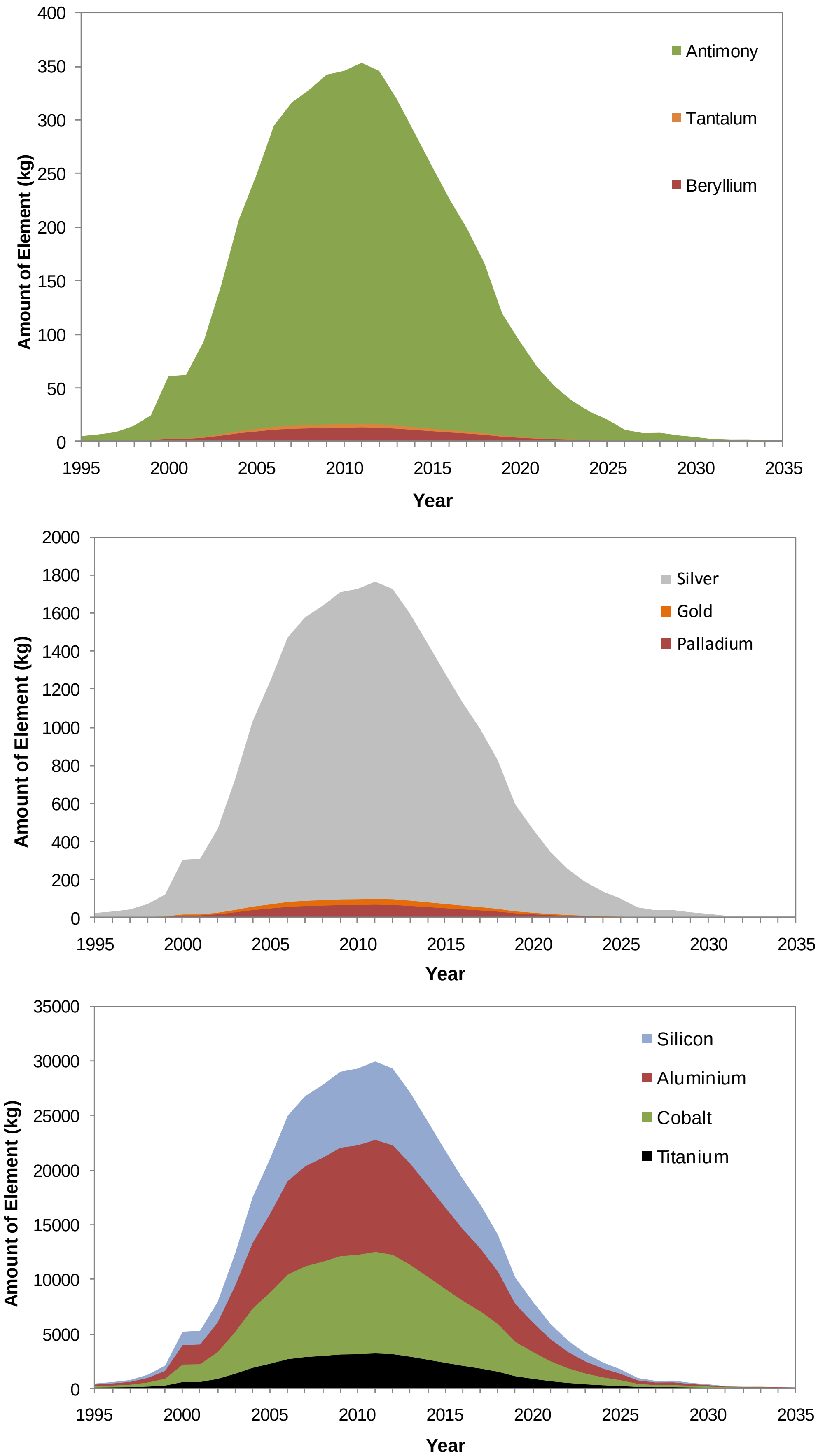

Figure 4. 7 Amounts of key metals in feature phones.

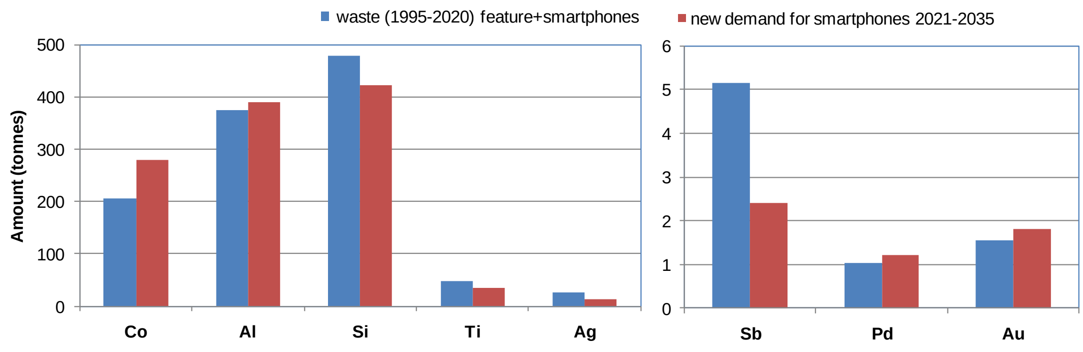

Figure 4. 8 Metal quantities in waste feature and smart phones (1995-2020) and metal demand in new smartphones (according to sales in 2021-2035).

3.6 Sensitivity and uncertainty analysis

The future obsolete mobile phones generation is further calculated according to the scenarios presented in Table 4.5. The examined parameters are: Weibull parameters (scale parameter after 2020: 2.6+/-0.6 for feature phones and 1.6 +/-0.6 for smartphones) and changing composition of elements by +/-10% in feature and smart phones in the future to account for elements content fluctuations in future designs. In this way, the effect of the examined parameters on the estimated obsolete waste amounts are presented and compared to the basic scenario (presented in 3.4 and 3.5).

In Fig 4.9 it is observed that for feature phones scenario 2 causes a deviation of 18-20% in EoL feature phones generation (tonnes) after 2028. On the other hand, for smartphones scenario 2 causes a deviation of 11-13.7% in EoL smartphones generation (tonnes) after 2021. The deviation between total EoL mobile phones is between 8.3-13.7% after 2021.

In Fig 4.10 it is observed that for feature phones scenario 2a causes a deviation of 13-38% in EoL feature phones generation (tonnes) in 2021-2028, 17-39% in 2029-2032, 15-17% in 2033-2035. For smartphones scenario 2a causes a deviation of 2.7-

5.8% in EoL smartphones generation (tonnes) in 2021-2035. The deviation between total EoL mobile phones is between 2.5-5.8% after 2021

Concerning the CRMs content the results are depicted in Fig 4.11 and Fig 4.12. In scenario 3 the deviation in CRMs content (kg) is +6.7% to the basic scenario, while in scenario 4 the deviation in CRMs content (kg) is -6.7% to the basic scenario. In scenario 5 the deviation in CRMs content (kg) is between 3.2-6.8% to the basic scenario (increase) for 2004-2035. Finally in scenario 5 the deviation in CRMs content (kg) is between 3.2-6.8% to the basic scenario (decrease) for 2004-2035.

Table 4.4 Different scenarios examined in the generation of obsolete feature and smart phones in Greece.

Parameter	**Scenario 1 (basic scenario)**	**Scenario 2**	**Scenario 2a**	**Scenario 3**	**Scenario 4**	**Scenario 5**	**Scenario 6**
Weibull parameters	scale parameter after 2020: 2.6 years for feature phones and 1.6 for smartphones	scale parameter after 2020: 2 years instead of 2.6 for feature phones and 1.0 instead of 1.6 for smartphones	scale parameter after 2020: 3.2 years instead of 2.6 for feature phones and 2.2 instead of 1.6 for smartphones	According to basic scenario	According to basic scenario	scale parameter after 2020: 2 years instead of 2.6 for feature phones and 1.0 instead of 1.6 for smartphones	scale parameter after 2020: 2 years instead of 2.6 for feature phones and 1.0 instead of 1.6 for smartphones
Elements composition		According to basic scenario	According to basic scenario	+10%	-10%	+10%	-10%

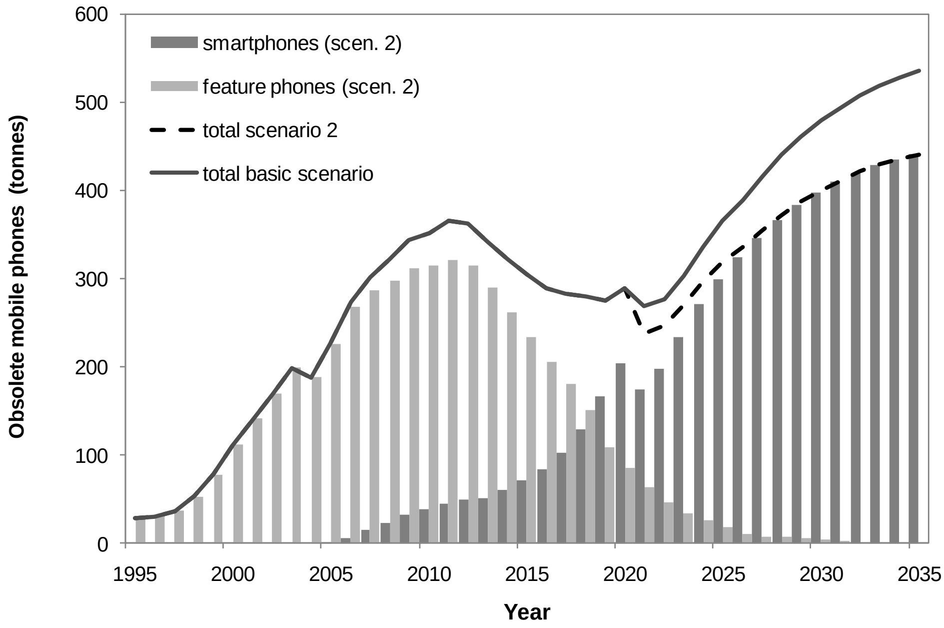

Figure 4. 9 Mobile phones waste generation according to Scenario 2.

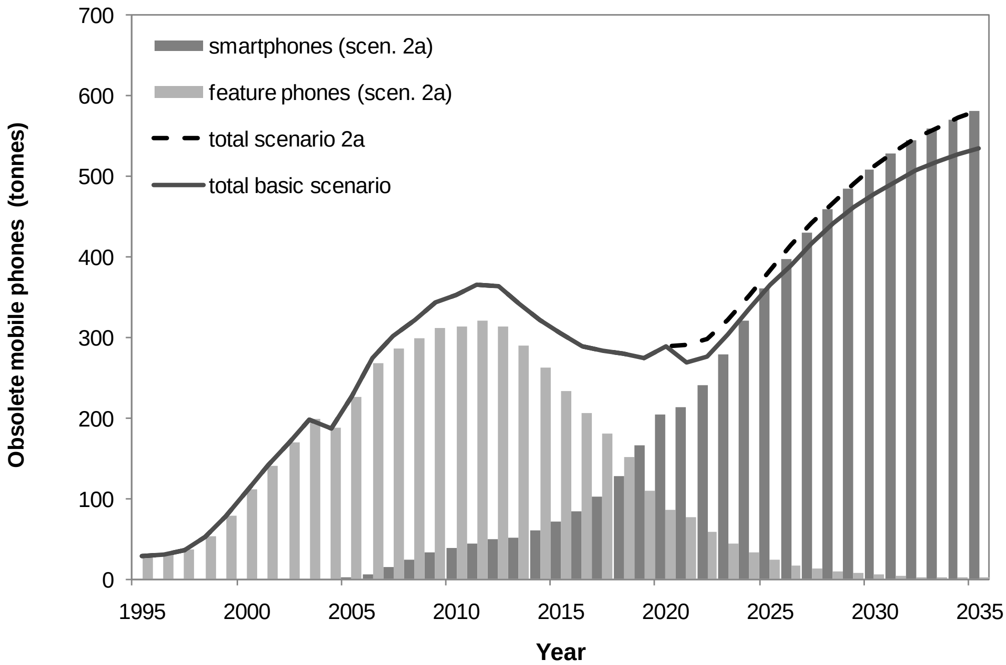

Figure 4. 10 Mobile phones waste generation according to Scenario 2a.

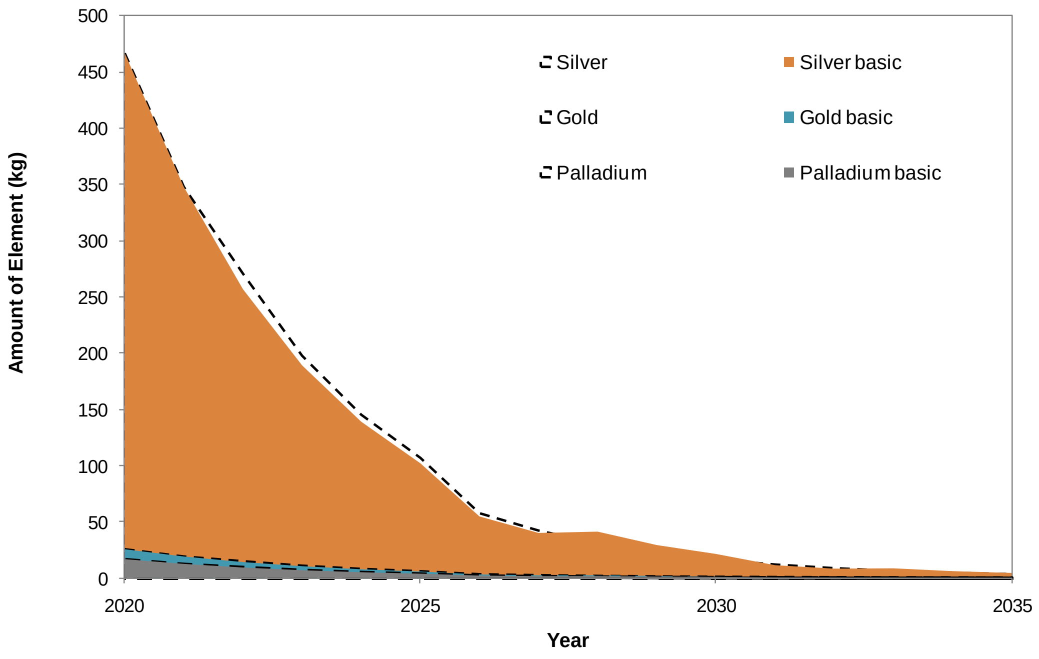

Figure 4. 11 Feature phones waste composition according to scenario 2 compared to basic scenario.

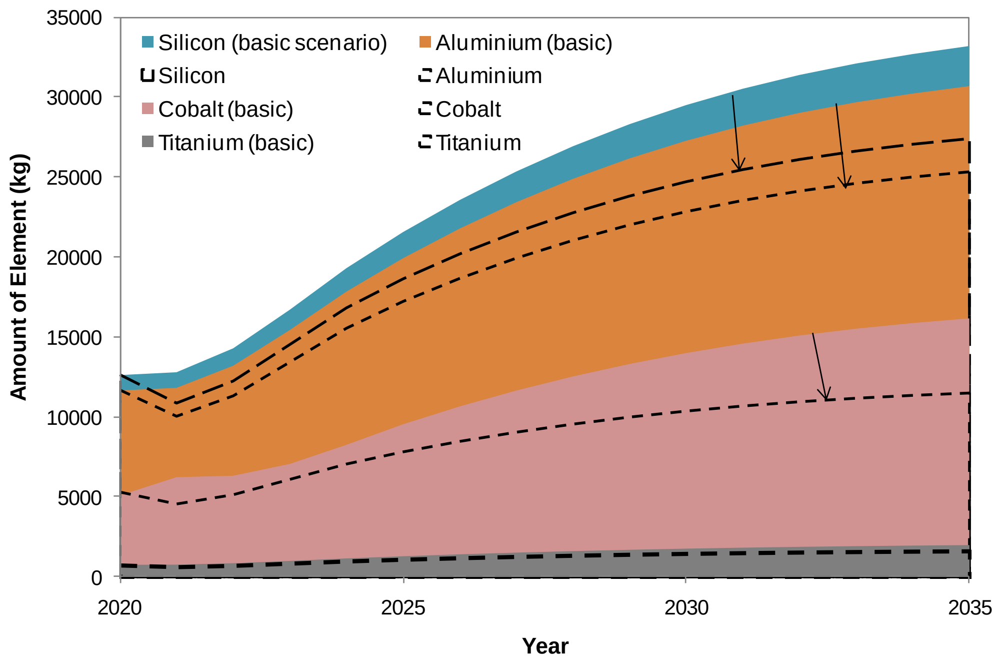

Figure 4. 12 The Smartphones waste composition according to scenario 2 compared to basic scenario. The deviation is between 11-13 % between the two scenarios.

4. Policy issues and recommendations

The composition of the future e-waste stream in Greece is expected to change rapidly. This should be considered by the authorities when designing the recycling systems. Material composition analysis of the mobile phones and their components is important as the composition of the same kind of product can vary due to different performance requirements, as in the case of smartphones and feature phones.

This study has a significant impact on the development of appropriate e-waste management policies in Greece, especially for category 3 (ICT devices). The estimation of future e-waste quantities (particularly in the long run) is a prerequisite for better organization of collection and treatment processes and the provision of the appropriate infrastructure. The National Waste Management Plan-NWMP (2020) stated that there is no reliable method to estimate WEEE and that the EEE POM quantities were used to estimate future quantities to achieve collection, recycling, and recovery targets.

Considering these targets in Greece, they were not achieved for category 3 in 2018 (collection 44.6%, recovery 83.8% and recycling 73.8%), and it is very unlikely that they will be met in 2020, as the collection target rises from 45 to 65% (based on POM quantities). Particular attention should be placed on mobile phones as they generally represent the fraction of high-tech devices with the lowest collection rate (Chancerel, 2010). Efficient collection schemes should pose separate targets for the collection and recycling of mobile phones focusing on the CRMs (on an element wt. basis).

Obsolete mobile phones and especially smart phones are among the most important e-wastes in terms of CRMs and PMs recovery, thus promoting circular economy involves their efficient collection. In order to improve the collection, several actions should be adopted to motivate people participate in the WEEE collection instead of storing non-functional electronic devices at home. Financial incentives may include vouchers for individuals for handing over their old devices, discounts on a new buy or a reduction in waste fees for residents or communities willing to separate more e-waste at source. Legal regulation at the local level could be the key factor for implementation (Dri et al., 2018). All these actions are important as Greece includes many islands that hinder efficient collection, thus establishing multiple collection points and increasing public awareness are vital.

Information about the quantities of critical and precious raw materials in obsolete mobile phones indicates the recycling potential. Especially Au and Pd contents and Au and Pd market prices are, among other parameters, critical for a recycling plant to be economically viable (D'Adamo et al., 2016). As the EU is 100% reliant on imports for REEs, Antimony, Tantalum, and 98% for Platinum and 93% Pd supply, e-waste collection and recycling must be intensified (EU, Final Report 2020). Currently, the recycling potential for REEs and Ta is low, thus new recycling methods must be introduced and the efficiency for Pd, Pt and other metals must be improved. For example, the EoL recycling rate for Co in the EU is only 22%, while in non-EU countries is 68% (World Bank, 2020). To accomplish improved recycling rates, first pressure through legislations must be put on producers to design devices for easy disassembly and recycling, facilitating recyclers to reach the metals included in the devices (Hagelüken and Corti, 2010). Furthermore, policy makers must take into account that the metal grade in primary ores is continuously decreasing, thus recycling or possible metal substitution is crucial to cope with future demand. On the other hand, metal recycling has many environmental benefits as the carbon footprint of secondary metal production is only a small part of the primary ore production (World Bank, 2020).

5. Conclusions

The quantities of critical and precious raw materials in obsolete smart and feature phones in Greece are presented until 2035. The composition of the future mobile phone waste stream is predicted to change rapidly and a great number of obsolete smartphones will be generated in the future. The accumulated waste mobile phones were 31.2 million units in 2010-2020, including 24.6 million feature phones and 6.6 million smartphones. The amount will grow to 43 million units in 2021-2035 (2.3 million feature phones and 40.7 million smartphones). In 2020-2035 the accumulated amounts of CRMs and PMs will be 1292.02 and 14.11 t., respectively, from waste smartphone only. The effective recycling of obsolete phones (1995-2020), can cover the demand for key metals in the new smartphones for more than a decade in Greece.

Existing compilation schemes should be improved to enhance collection as much as possible. In this way valuable resources, like key metals will not be lost and the EU can become more material sufficient in accordance with the circular economy concept. Our path to a sustainable future comes through zero waste policies. Currently, the recycling potential for REEs and Ta is low so closing the loop between resources, materials and their waste demands new efficient innovative recycling processes. The information presented can contribute in creating a data set about metal demand and secondary supply which is needed to move to a circular economy.

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CHAPTER 5

Dynamic estimation of end-of-life electric vehicle batteries in the EU-27 considering reuse, remanufacturing and recycling options

With the rapid promotion of electric vehicles (EV) in the European Union (EU), a new e-waste category is emerging, the lithium-ion batteries (LIBs). LIBs require proper management through circular economy business models. These include Remanufacturing, Reuse and Recycling of LIBs to extend their life before valuable materials are recovered. Material and substance flow analysis with a 3-parameter Weibull distribution function are employed to quantify all battery waste flows and their embedded materials. The available LIBs for remanufacturing will be 113-301 thousand (6-31 GWh) by 2024-2034, while the capacity of Second Life (2L) LIBs will be 212-617 GWh by 2040. The available 2L capacity in Germany and France could effectively cover the demand for energy storage for photovoltaic systems in these countries, which have the highest EV adoption in the EU-27. The recycled metals Li, Co, Ni and Cu could meet 5.2-11.3 % of the demand for new materials considering the recycling efficiencies of the 2020 EU Battery Directive.

1. Overview

The New Green Deal in the European Union (EU) aims to decarbonize all sectors of economy, including energy and transport, in order to create a climate neutral economy. In this concept, many EU countries have set mandatory targets to eventually end all sales of internal combustion engine vehicles by 2030 or 2040, promoting electric vehicles (EV). As EV sales are expected to increase, so will the demand for EV batteries and subsequently, the materials required for their production (Drabik and Rizos, 2018).

The efficient use of material sources and the efficient waste treatment dictate circular economy (CE) models that establish practicable and profitable management. When handling End-of-Life (EoL) EV batteries, the best circular business model is Remanufacturing (to use in EVs), Reuse (in other applications), Recycling and last waste management (Wrålsen et al., 2021). The present study considers all the CE options of 3R (Remanufacturing, Reuse and Recycling).

Lithium-Ion batteries (LIBs) manufacturing requires many different raw materials, some of which are Critical Raw Materials (CRMs) for the EU due to their high economic importance and associated supply risks (Lebedeva et al., 2017). Among the materials used in LIBs, only Co and less likely Li may be constrained by future resource limitations (Olivetti et al., 2017; Xu et al., 2020). These materials are included in the EU CRMs List (EU, 2020a), being of strategic importance for the EU economy. The potential recycling of LIBs will contribute to the sustainable and secure supply of secondary raw materials (SRMs) in the EU.

As the manufacturing of a new EV battery causes high environmental impacts, the extension of the battery life after reaching its EoL in the vehicle is a prerequisite according to the CE models (Peters et al., 2017). The LIB second life could ‘slow the material loop’ by life extension, before finally ‘closing the loop’ by recycling to recover valuable materials (Emilsson et al., 2019). Extending the battery life can occur either by remanufacturing, if the battery has a State of Health (SoH)>85 (Castro et al., 2021) or by using it in a second application, extending its life before recycling to acquire secondary raw materials.

As the EU has insignificant LIBs production (only 3 % of global production), it is depended on batteries mainly produced in Asia. However, there are plans to increase domestic LIBs production to 7 % and 25 % by 2022 and 2025, respectively (Tsiropoulos et al., 2018). Since there is currently, no official LIBs remanufacturing in the EU, remanufacturing could be an appealing option considering that most EV cars are sold in EU (39.8 %), followed by China (39.6 %) and North America (10.7 %) (Kotak et al., 2021). To achieve the CE remanufacturing and repurposing options, there is a necessity to design the batteries for disassembly and remanufacturing, including ease of diagnosing the SoH (Albertsen et al., 2021). This is likely to materialize in the forthcoming years. Remanufactured batteries could have a potential cost reduction of 40 %, for a 10 % cell replacement (Standridge and Corneal, 2014).

LIB reuse, after the EV battery ends its (first) life, is possible as it still retains about 80 % of its initial capacity and could be utilized for other, less demanding, applications (Ziemann et al., 2018; Rallo et al., 2020). Therefore, the battery lifetime can be prolonged by 5–10 years mitigating the demand for raw materials needed to manufacture new batteries (Bobba et al., 2019; Olivetti et al., 2017). Possible second life (2L) applications include Energy Storage Systems (ESS) to support the production of electricity from renewable energy sources, storage for domestic purposes or other uses for micro or urban electro-mobility (street lighting, refrigerated vehicles, forklifts, etc.). Extending battery life is worthwhile as it delays recycling until the associated processes evolve to become more mature (Kotak et al., 2021; White et al., 2020; Albertsen et al., 2021).

The economic feasibility of the 3R options for EoL LIBs management is largely dependent on the number of discarded LIBs available for each option, as well as the evolution of prices of new cells and raw materials (Rohr et al., 2017). The proposal of the new EU Battery Regulation aims to facilitate reuse, remanufacturing and repurposing of EV batteries, claiming that batteries include a management system that stores information and data on the SoH and expected lifetime. This data could be accessed by a person or corporation that has legally purchased the battery (EU, 2020b).

According to the EU Battery Directive (2020) the recycling efficiency of LIB is expected to increase to 65 % by 2025 from 50 % in 2006 (EU, 2006; EU 2020b). The option of recycling will close the materials loop as valuable metals will be recovered as SRMs. In particular, the recovery of Ni and Co is the economic driving force for LIBs recycling (Kwade and Diekmann, 2018). As Co is a rather rare metal, its growing demand can cause the price of the metal to rise and the battery industry aims to minimize or replace it with Ni, Mn or iron phosphate. Many cell chemistries have been established in recent decades, aiming to improve efficiency and reduce costs. Lithium Nickel Manganese Cobalt Oxides (NMC) is the cathode chemistry with the most considerable market share in small commercial and passenger EVs in Europe (Tsiropoulos et al., 2018; Bobba et al., 2019). NMC batteries use 1:1:1, 4:2:4, 5:3:2, or 6:2:2 ratios for nickel, manganese, and cobalt, respectively. As technology progresses there is a shift from NMC 111 to NMC 622 or NMC 811 (8:1:1), and in the future NMC 955 (9:0.5:0.5), minimizing the content of Co in the batteries, while retaining high energy density (Olivetti et al, 2017; Abdelbaky et al, 2021).

The assessment of EoL EVs batteries is a key factor for their proper management in the EU-27. Assessments of waste streams (remanufacturing, reuse, recycling) will guide process developments and related infrastructure investments. Different battery technologies in waste streams will improve decision-making regarding their management and the recovery of secondary raw materials. The optimized management of retired LIBs requires regional planning at the country level. To investigate the influence of 3R options regarding the management of EoL LIBs, this study addresses the following questions: (I) what quantities and capacities of LIBs will be generated in the EU-27 and the 2 member countries with the higher EV adoption (Germany and France)? This is the first time a study focuses on the EU-27 using a 3-parameter Weibull distribution lifetime with a dynamic material flow analysis, examining the 3R options in LIBs waste management. The 3-parameter Weibull distribution allows a more accurate description of LIBs lifespan compared to the 2-parameter model. How many LIBs and of what technology and capacity will be available (II) for remanufacturing, (III) for a second life and (IV) for recycling? The option of remanufacturing has been scarcely considered in the literature and only at a fixed rate (Abdelbaky et al., 2021) and the present study will cover a significant research gap. In addition, the available capacities of second life (B2L) LIBs are estimated for Germany and France, and the share of B2L for stationary storage to support Photovoltaic Energy (PV) production according to the announced PV installations by 2030 in the two countries is identified. (V) What quantities of valuable materials (Co, Li, Ni, and Cu) can be recovered from the waste LIBs taking into account the new recycling efficiency targets set by the EU battery Directive (2020)? Only one study in the literature refers to dynamic metal recycling rates but it concerns exclusively the region of Catalonia in Spain (Crespo et al., 2022). So far, there is no study quantifying Co, Li, Ni, and Cu recovery from LIBs in the EU considering dynamic recycling efficiencies, so an important research gap is filled. The answers to the above questions are highly important for the EU-27 to direct the battery value chain action plan for the efficient management of EoL LIBs.

To answer the above questions, a dynamic 3-parameter Weibull distribution lifespan is adopted for the battery life which considers the year of battery production instead of a uniform lifespan for all batteries independently of production year. This has not been applied to EU studies, as they employ a uniform Normal distribution lifespan for all years or fixed or truncated lifetime or a 2-parameter Weibull distribution. Future EV sales are projected according to two scenarios employed by the International Energy Agency (IEA, 2020). The scenarios are applied for the first time in the EU, while the impact of covid pandemic on sales is also taken into account. Future EV sales (and EoL batteries) are calculated separately for Battery EVs (BEVs) and Plug-In hybrid EVs (PHEVs) considering the 2025 and 2030 projection targets of the two scenarios for BEVs and PHEVs. This yields more precise results than calculating the total EV sales and making assumptions for the 2030 share of BEVs and PHEVs (Abdelbaky et al., 2021). Then, the EoL LIBs are estimated by material flow analysis using two dynamic Weibull lifetime distribution scenarios, a Low lifetime (LL) and a High Lifetime (HL). The EoL LIBs are presented for eight battery cell chemistries (lithium nickel cobalt aluminum (NCA), lithium manganese oxide (LMO), lithium iron phosphate (LFP), nickel manganese cobalt NMC 111, NMC 532, NMC 622, NMC 811, and NMC 955). The specific EU Battery Directive (2020) recycling targets are used, considering the future improvement in the recycling processes. The recovered amounts of Li, Ni, Co, and Cu are then calculated, and the recycled metal quantities are compared to the annual material demand according to new EV sales.

2. Methodology

The research methodology followed in this work is illustrated in Fig. 5.1. As BEV and PHEV batteries have different sales trends and different shares of battery technologies and capacities, all calculations are performed for BEV and PHEV batteries separately. This is more accurate than considering an average for BEV and PHEV batteries together. First, the historical BEV and PHEV sales data are collected (2010-2020). Future projections (2020-2030) are then estimated under two scenarios, the Stated Policy Scenario (STEPS) and the Sustainable Development Scenario (SDS). The STEP Scenario incorporates existing government policies while the SD Scenario takes into account the targets set by each country for EV car deployment by 2030 in compliance with the climate goals of the Paris agreement (IEA, 2020). Second, the relative shares of eight battery technologies (NCA, LMO, LFP, NMC 111, NMC 532, NMC 622, NMC 811, and NMC 955) and the battery capacities (kWh/kg) in BEV and PHEV are regarded in 2010-2030. Then, the EoL LIBs are estimated by material flow analysis using two dynamic Weibull battery lifetime distribution scenarios: a Low (LL) and a High (HL) (see Section 3.5). Then, a part of the EoL LIBs will be remanufactured if it is discarded early (<5 y) so as to retain SoH> 90 %. Another part will be reused in other applications according to a dynamic reuse pattern and will undergo a second life (B2L) and the rest will be recycled (B2R) (section 3.6 and 3.7). After using the B2L according to the second life Weibull distribution model, the generated obsolete batteries will also be recycled. Finally, taking into account the recycling efficiencies, the materials embedded in the EoL batteries (BEV and PHEV) are estimated to reveal the potential of recovering valuable materials.

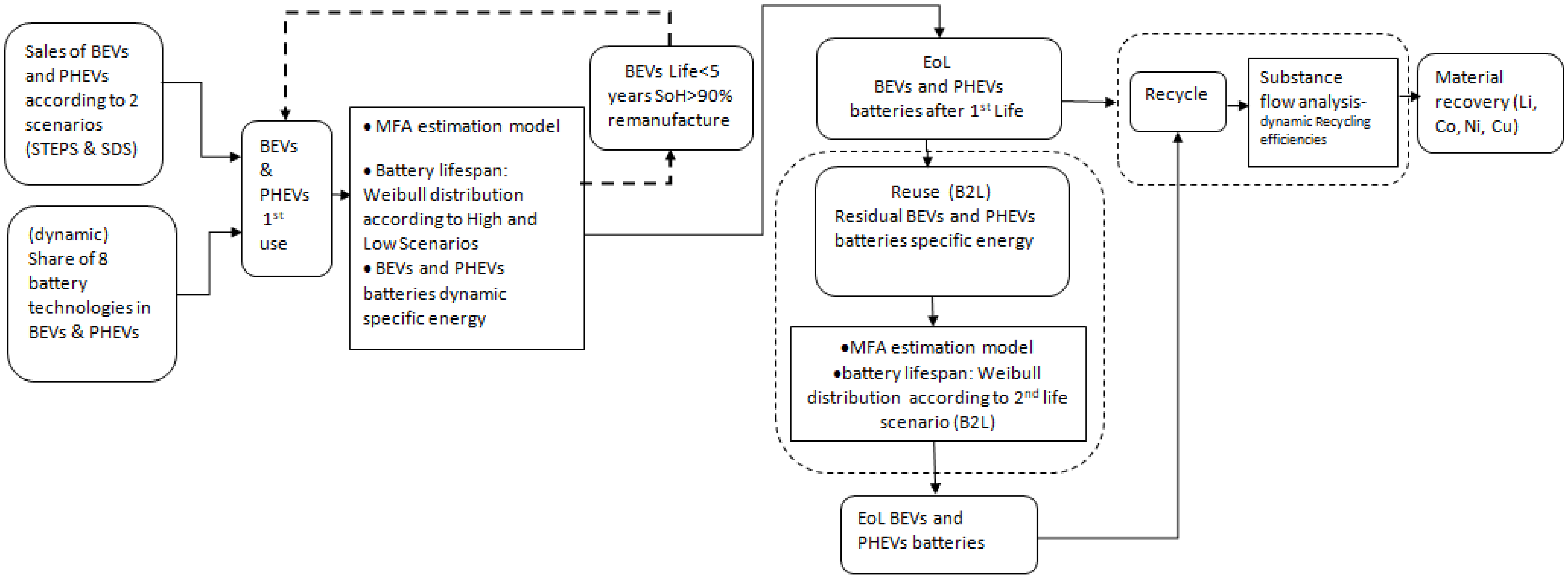


Figure 5.1. Methodology flowchart considering the 3R options (Remanufacturing, Reuse, and Recycling).(BEVs: Battery Electric Vehicles; PHEVs Plug-In Hybrid Electric Vehicles; STEPS: Stated Policy Scenario; SDS: Sustainable Development Scenario; B2L: Battery Second Life; B2R: Batteries to Recycle; MFA: Material Flow Analysis).

The analysis is performed for the EU-27 as well as the EU continent (including Norway, Iceland, Switzerland and the UK) for comparison. In addition, the analysis is performed on a country basis for Germany and France, as they possess the largest share of EVs in the EU-27 (about 34 and 18 % in 2020, respectively) to demonstrate the possibility of integrating 2L LIBs into ESS for PV systems, according to the announced PV installations in these countries by 2030. The details of the methodology are further explained in the following sections.

2.1 EV batteries sales from 2010 to 2030

Historical EV batteries sales in Europe are derived from IEA vehicle sales data (IEA, 2021). The sales apply to the European continent, including the EU-27 countries and Norway, Iceland, Switzerland and the UK. The analysis in this study is performed for the EU-27 (and the European continent as a comparison) and Germany and France (country level). The increase in annual EV sales in the European continent, between 2010 and 2020, follows an exponential curve. Forecasts of future sales are based on 2 scenarios: the Stated Policy Scenario (STEPS) and the Sustainable Development Scenario (SDS). The derived data include 5-year projections based on STEP and SD Scenario (European continent). The increase between 2020-2030 follows an exponential curve for BEVs and a linear trend for PHEVs (in both models). For example, Denmark has set the target of 1 million EV sales by 2030, while France 4.8 million and Italy 6 million EV sales accordingly. The influence of Covid-19 pandemic on EV sales in 2020-22 is also considered. It appears that electric car sales in 2020 have been more resilient than the overall car market. One reason for this was the financial incentives from EU governments in the form of subsidies or tax reductions on EV sales (IEA, 2020). For example, Germany, France and Italy announced additional subsidies to boost EV sales after the pandemic in 2020. Thus, as these subsidies are announced to continue till 2022, the pandemic crisis slightly affects the EV sales (IEA, 2021).

According to the 2 scenarios (STEPS and SDS), the BEV and PHEV sales across the European continent (including the 4 non-EU countries) will be 3.2-5.1 million vehicles in 2025 and 5.9-11.3 million in 2030. In STEPS, there is a forecast of

138 % increase in 2025 EV sales and 329 % in 2030 compared to 2020 sales. In SDS, there is a forecast of 272 % increase in 2025 EV sales and 722 % in 2030 sales compared to 2020. Abdelbaky et al. (2021) used a forecast of EV sales in Europe as share of global EV sales until 2030. They estimated the annual EV sales in Europe at 2.8 million vehicles in 2025 and 5 million in 2030.

Corresponding sales for the EU-27 are calculated at 3-4.7 million vehicles in 2025 and 5.4-10.3 million in 2030 in STEPS and SDS scenarios. The share of EV sales in UK, Switzerland, Iceland and Norway decreased from about 50 % in 2010 to around 23 % in 2030. Historical EV sales (BEV and PHEV) for the European continent, EU-27 and the four countries UK, Switzerland, Iceland and Norway by 2020 are depicted in Fig. 5.2

Fuel cell electric vehicles (FCEVs) are not considered in the analysis as their sales represent only 0.77 % of total sales in Europe (BEV+PHEV+FCEV) between 2010-2020

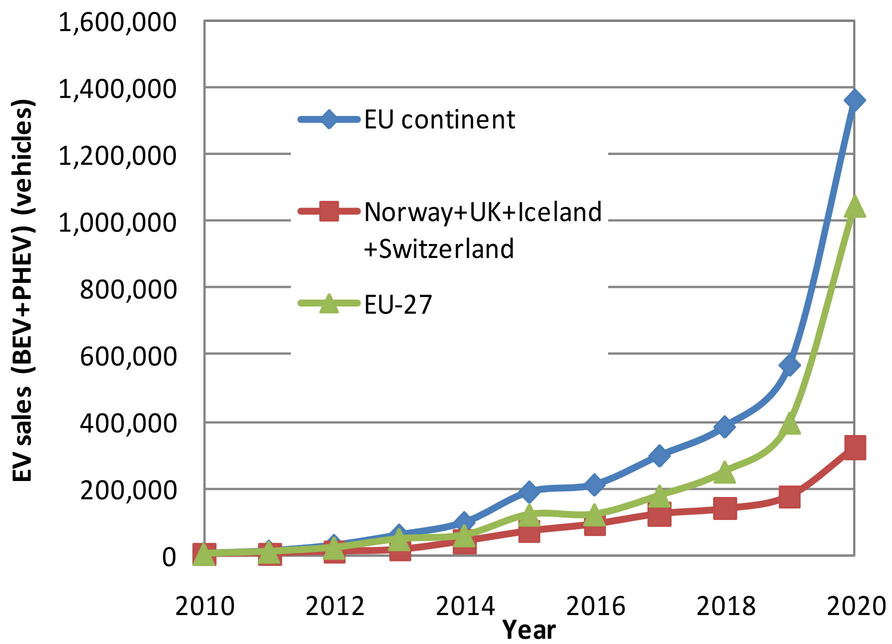


Figure 5.2. Historical EV sales (BEV and PHEV) for the European continent, EU-27 and the four countries UK, Switzerland, Iceland and Norway (IEA, 2021).

2.2 EV batteries sales in Germany and France

Germany is presented as a case study to examine the possibility of covering ESS for PV systems by second-life EV batteries. Germany possesses the largest share of EVs in the EU-27 (about 38 % in 2020) and the largest share of installed PV capacity in EU-27 (51 % by 2020). The targeted PV deployment capacity in Germany, as retrieved from the German National Energy Climate Plan (NECP) by 2030 will be 98 GW (Kastanaki et al., 2022).

Considering the increased rate of EV sales in Germany between 2017-2020 and the targeted EV sales by 2030 (IEA, 2020), the BEVs and PHEVs sales will be about 0.97 and 0.95 million in 2025 and 3.4 and 2.9 million in 2030, respectively. These targets are consistent with the SD Scenario. The increase in BEV and PHEV sales follows an exponential and a linear trend in 2020-2030, respectively (R^2>0.99).

France is the second country in the EU-27 in EV adoption (about 18 % in 2020). The targeted sales in 2025 are 660 and 500 thousand BEVs and PHEVs, while in 2030, 3 and 1.8 million BEVs and PHEVs, accordingly. The increase in both BEVs and PHEVs is exponential between 2020-2030 (R^2>0.99). The planned PV deployment capacity in France, as retrieved from the French National Energy Climate Plan (NECP) by 2030 will be 42 GW (Kastanaki and Giannis, 2022).

2.3 Market share of batteries technologies

The market share of the various battery technologies from 2010-2030 is presented in Table 5.1 . Data are gathered from various sources (Bobba et al., 2019; Statista, 2019; Xu et al., 2020; Abdelbacky et al., 2021). In order to combine the data from various sources, the procedure hereafter was followed. First, data on the market share of various batteries technologies regarding Europe are retrieved from Statista (2019). The data include NCA, LMO, LFP, NMC 111, NMC 622, NMC 811, and NMC 955 and are given on a 5-year basis from 2019-2030. However in these data the market share of NMC 532 is not reported. Considering the data of Bobba et al.,

(2019) and Abdelbacky et al., (2021), it is evident that in Statista data for NMC 532 and NMC 622 are combined. Thus, the data of Bobba et al and Abdelbacky et al., are utilized to clarify the separate NMC 532 and NMC 622 market shares. Finally, as these data are given on a 5-year basis, Xu et al., the (trend) data of (on a global scale) which are reported on an annual basis are utilized to fill in the gaps between the 5-year periods. In this way, annual data for all chemistries are obtained.

Last, Abdelbacky et al., distinguished between the Lithium Manganese Oxide (LMO) and LMO blend market shares, however in this study these two are combined (since LMO blend share is greater than LMO, as reported in Abdelbacky et al. (2021)). For example in 2018 market share LMO blend is 20%, and LMO 9%. (Abdelbacky et al., 2021), LMO battery cell chemistry in EVs is used due to its efficient performance in charging/recharging and also Mn is cheaper than Co. The performance of LMO cells is improved by blending with Lithium Nickel Manganese Cobalt oxides (LMO blend).

As can be observed the dominant battery technology in vehicles sold in 2010 is the NMC 111. In 2019, NCA and NMC 523 (Ni:Mn:Co 5:2:3) prevail, while in 2025, NMC 811 batteries. As the development of low content cobalt batteries progresses, NMC 955 batteries will dominate over older and more expensive technologies by 2030.

Global research aims to develop several future chemical cells with improved capacity and reduced cost. Chemistries that can advance the battery technology beyond Li-ion include the Solid State, Lithium-air batteries, etc. (Lebedeva et al., 2017), but these will be introduced after 2030 and are not considered.

The average battery capacity in BEVs increases from 34 kWh in 2014 to 72 kWh in 2030 and in PHEVs from 9.8 kWh to 15 kWh, respectively (Ai et al., 2019; Abdelbaky et al., 2021; IEA, 2020). These data are presented in Table 5.2 In order to combine the data from various sources, the procedure hereafter was followed. Abdelbaky et al., (2021); reported the 2018-2030 battery capacity for BEVs is 44 - 60kWh and for PHEV 10-15 kWh. Ai et al. (2019), reported that the capacity ranges from 2005-2030 at 5-year intervals for BEVs (22-56.5 kWh) and PHEVs (7.8-16.2 KWh).

The IEA (2020) report stated that for PHEVs the average battery capacity was almost constant at 11 kWh (2016-2020). Also, there is a trend to increase the capacity to 70-80 kWh by 2030 for BEV batteries and to 15 kWh for PHEV batteries. The above data are combined and the gaps between the intervals, where there is no data, are filed by linear extrapolation, taking into account an increasing trend in the capacity of the batteries.

Table 5.1. The market share (%) of the various battery technologies from 2010-2030 (Bobba et al., 2019; Statista, 2019; Xu et al., 2020; Abdelbacky et al., 2021).

			Battery chemistry share					
Year	NCA	LMO	LFP	NMC111	NMC532	NMC622	NMC811	NMC955
2010	19		24	57				
2011	19		24	57				
2012	11		14	33	41			
2013	16		18	3	35			
2014	22		24	26	28			
2015	23		29	23	24			
2016	36	10	1	28	21	0.2	5	
2017	31	2	8	21	21	9	8	
2018	30	2	4	11	21	12	20	
2019	28	2	2	6	25	13	25	
2020	25	1	2	2	30	10	32	
2021	25	1	2	2	19	32	19	
2022	25	1	1	2	11	30	30	
2023	25	1	2	2	11	27	32	
2024	25	1	2	2	10	27	33	
2025	24	1	2	1	9	28	35	
2026	22	1	1	3	8	23	33	9
2027	21	1	1	3	6	26	31	12
2028	20	1	1	3	5	26	30	14
2029	18	1	1	2	5	27	28	19
2030	14	1	1	1	5	18	25	37

Table 5.2. The average battery capacity in PHEVs from 2010-2030 (Ai et al., 2019; Abdelbaky et al., 2021; IEA, 2020).

Year	BEV kwh	PHEV kwh
2010	24.3	8.6
2011	26.7	8.9
2012	29.1	9.2
2013	31.5	9.5
2014	34.3	9.8
2015	36.5	10.0
2016	37.1	11.0
2017	41.0	11.0
2018	44.0	11.0
2019	45.8	11.0
2020	48.1	11.7
2021	48.1	12.0
2022	52.9	12.4
2023	55.3	12.7
2024	57.7	13.0
2025	60.1	13.3
2026	62.4	13.6
2027	64.8	13.9
2028	67.2	14.2
2029	69.6	14.6
2030	72.0	15.0

2.4 Material and substance flow analysis

The obsolete batteries output (O) is given as a function of batteries input (S) and the lifespan probability distribution (Market Supply A):

$$O_t = \sum_{i=0}^{n} (S_{t-i} \times f_i) \qquad (1)$$

Where f_t: battery lifespan probability distribution function, and n: maximum possible lifespan (Wang et al., 2013).

The battery lifespan distribution is depicted by the Weibull distribution (Kalmykova et al., 2017; Ai et al., 2019). The Weibull distribution has proven to be suitable for describing the electronic products lifespan as it provides the best fit for most devices. It also captures the lifespan active changes due to its dynamic nature (Kastanaki and Giannis, 2022). The 2-parameter Weibull is widely used for electric and electronic equipment such as televisions, laptops, batteries etc. (Islam and Huda. 2019; Kastanaki and Giannis, 2021; Kalmykova et al., 2017) and is simpler than the 3-parameter Weibull. E-waste statistics also suggests using the 2-parameter Weibull when modeling e-waste flows (Forti et al., 2018). The 2-parameter Weibull only employs two parameters (α: the shape parameter and β: the scale parameter), while the 3-parameter additionally uses the minimum lifespan parameter, γ. The more complex 3-parameter Weibull better reflects the real life data (Ai et al., 2019). The 3-parameter Weibull distribution is a probability model for simulating variation in battery life and is given by:

$$f\left(t,a,\beta,\gamma\right)=\frac{a}{\beta^{a}}(t-\gamma)^{a-1}\exp(-\left[\frac{(t-\gamma)}{\beta}\right]^{a}) \qquad \text{if } t>\gamma \tag{2}$$

And f=0 if $t<\gamma$

where α: the shape parameter that describes the gradual ageing of the product, γ: the minimum lifespan or threshold value (years), and β: the scale parameter describing the characteristic life of the product that is, it defines the age when 63.2 % of the products are expected to have failed. Xu et al. (2020) assessed the minimum, lifespan for LIBs (of all chemistries) from 2005-2050 is 1 year on a global scale. Also Hoarau and Lorang (2022) assessed a minimum lifespan of 1 year for LIBs in Europe. The present study considers γ=1 year.

When the average lifetime is known (or expected according to the warranty period) the scale parameter is calculated by:

$$\beta=(Average\_Lifetime-\gamma)/\Gamma(1+1/\alpha) \tag{3}$$

where Γ: gamma function.

The Average Lifetime (AL) does not coincide with the scale parameter (β) when the minimum lifespan (γ) is taken as 0. For instance, when γ=0, AL=12 years and α=3.5, then β=13.34 years. However, when γ=1, AL=12 years and α=3.5, then β=12.23 years. In the latter case, the AL almost coincides to β. Various studies (Ai et al., 2019; Shafique et al., 2022; Crespo et al., 2022) have considered AL= β, however this is approximately the case for the 3-parameter Weibull function, as explained.

For the substance flow analysis, the composition of recycling waste stream is calculated from the BEVs and PHEVs recycled after the batteries' first and second life.

For example, for BEVs batteries after first life:

$$RC^{BEV}{}_{t,x} = \sum_{i=0}^{n} \left\{ [(S^{BEV}{}_{t-i} \times Sh_{t-i}^{BT}) \times f_i] \times sh_{t-i} \times Cap^{BEV}{}_{t-i} \times C_x \right\} \quad (4)$$

where S^{BEV}: sales of BEVs, Sh^{BT}: share of battery technology (NCA, LMO, LFP, NMC111, NMC523, NMC622, NMC811, NMC955), sh: share of recycling, Cap^{BEV}: capacity (kWh), C_x: mass fraction of material x (kg/kWh).

In a similar way, the composition of BEV batteries after second life is estimated. In that case, all batteries after second life are recycled. The same apply for PHEVs batteries.

2.5 Battery lifespan

EV battery life is a critical factor in estimating the amount of obsolete batteries. Many factors affect the battery lifetime, i.e., battery capacity and degradation rate, technology advancement and materials used and the pattern of usage (driving behavior, charging frequency, road condition and maintenance) (Ai et al., 2019; Qiao et al., 2020; Shafique et al., 2022).

EV batteries of different technology have their own lifespan, which may not coincide to the average EV battery lifespan. The Weibull distribution function regards various product life patterns to shape realistic lifetime distributions rather than considering a fixed lifetime (Ai et al., 2019). Two dynamic scenarios are used in this study to account for changes in battery life pattern as technology improves, a High and a Low Lifespan scenario (HL, LL). In the HL scenario, the average lifespan of EV batteries is considered according to the production year of the battery as follows: the average lifetime in 2009–2016, 2017–2022, 2023–2030 are 4, 10 and 16 years (Shafique et al., 2022), while the shape parameter is equal to 3.5., resulting in the scale parameters of 3.33, 10.00 and 16.67, respectively. Customer surveys revealed that battery lifespan can be as short as 4–6 years in early EV models (Ai et al., 2019), while the current battery lifespan is around 10 years and is expected to increase in the future. Other studies have also used lifespans of 5-15 years (Ahmadi et al., 2014; Neubauer et al., 2015; Sathre et al., 2015) and the Association of European Automotive and Industrial Battery Manufacturers expect the LIB lifetime to reach 15 years in 2030 (EUROBAT, 2015). Note that when α =3.44 the Weibull distribution approximates the Normal distribution function. The value α =3.5 has been estimated by reliability assessment of experimental data failure pattern of LIBs (Eom et al., 2007) and was used in various studies (Ai et al., 2019; Shafique et al., 2022; Crespo et al., 2022). This value accounts for various losses like premature battery failure or road accidents (Abdelbaky et al., 2021). Xu et al. (2020) used, after 2020, a shape parameter of 6.3 and a lifetime of 15 years on a global scale.

After carefully examining the warranty periods given for EV batteries from various companies (MyEV.com), a Low Lifetime scenario was also used corresponding to an average lifetime of 4, 8, 12 years in 2009–2016, 2017–2022, 2023–2030 (Shafique et al., 2022). The LL scenario data are similar to the lifetime considered by the US National Academy of Sciences (Shafique et al., 2022).

2.6 EV batteries available for remanufacturing and repurposing

Regarding remanufacturing and reuse, Kotak et al. (2021) have excluded PHEV batteries for economic and SoH reasons. In the present study, remanufacturing is only considered for BEV batteries with a SoH>90 %, as this can be economically viable. Battery remanufacturing (for reuse in EVs) is considered after 2023 in the EU-27. As discussed in Albertsen et al. (2021) currently there is insignificant repair-remanufacturing of EV batteries, but this is expected to increase as the original equipment manufacturers (OEMs) pursue closed loop management strategies. In order to make sure that SoH> 90 %, only the portion of batteries that are discarded within five years after first use are considered. Since EV batteries lose about 20 % of their capacity in about 10 years, resulting in a SoH of 80 %, batteries discarded within 5 years could have SoH>90 %. Finally, only a 50 % share of these batteries is ultimately considered suitable for remanufacturing for various reasons.

For repurposing options (B2L), a dynamic second use rate is considered as follows: 10 % until 2019, then linearly increasing from 10 % to 50 % in 2030 and remaining constant after 2030. This scenario agrees with an average scenario used in other studies (Abdelbaky et al., 2021; Crespo et al., 2022). The average battery lifespan is considered 8 years and the shape parameter α=3.5 (Castro et al., 2021) and γ=0, therefore, the scale parameter is β=8.89. A higher (than 50 %) repurposing rate has not been considered, as with the expected decrease in the price of LIBs, the repurposed batteries may not be profitable (Melin, 2021). Both BEV and PHEV batteries can be used as B2L to support grid balancing (EASE, 2017).

2.7 EV battery specific content of relevant materials – Recycling potential

The mass fractions (kg/kWh) of the relevant materials in different battery technologies in BEVs and PHEVs are listed in Table 5.3 (Olivetti et al., 2017; IEA, 2018; Abdelbaky et al., 2021). Since battery capacity (kWh) depends on the battery composition, the values are combined with dynamic battery capacity to yield the time-dependent material composition. The values are critical for evaluating the waste stream recycling potential of LIBs.

In order to combine the data from various sources, the procedure hereafter was followed. LMO refers to (LMO blend +LMO) in this study; the two are combined for simplicity (since LMO blend share is greater than LMO, as reported in Abdelbacky et al). Olivetti et al. (2017) also report the LMO blend mass fractions of the relevant materials in agreement to this study.

Abdelbacky et al. (2021) and IEA (2018) reported the mass fractions (kg/kWh) of the relevant materials in NCA, LMO, LFP, NMC 811, NMC 622, NMC 111, NMC

532 but not NMC 955. The materials mass fractions (kg/kWh) for NMC 955 are retrieved from Olivetti et al. (2017).

In order to estimate the recycling potential of Li, Ni, Co, and Cu, the recycling efficiency of the metals is taken into account. Although the recycling efficiency of Co and Ni is about 85 % through pyrometallurgical and hydrometallurgical processes, the recycling of Li is extremely low. In 2015 no lithium recycling occurred in Europe, but this will change as directed by the targets set in the European Batteries Regulation proposal (EU, 2020b). For instance this proposal sets a target of 35 % recycling efficiency for Li in 2025 and 70 % in 2030 for the EU region, while for both Ni and Co 90 % and 95 %, respectively. Specific recycling targets applicable to the EU have been adopted which consider the future improvement of the recycling processes. The dynamic parameters are presented in Table 5.3 (EU, 2020a; Crespo et al.2022; Xu et al., 2020). Currently, graphite is not recycled as the process is not economically viable, so this material is not examined (Castro et al., 2021). However, possible future imbalance in the supply and demand of these metals may affect the efficiency of recycling, especially for Li. This may cause a further increase in recycling efficiency, which will affect the amounts of recovered metal. However, as this study focuses on the EU-27 region, the recycling targets set by the EU are adopted in this study.

Table 5.3. Mass fractions (kg/kWh) of the relevant materials in different battery technologies in BEVs and PHEVs (Olivetti et al., 2017; IEA, 2018; Abdelbaky et al., 2021).

	Li [kg/kWh]	Ni [kg/kWh]	Co [kg/kWh]	Cu [kg/kWh]
NCA	0.10	0.67	0.13	0.76
LMO	0.11	0.07	0.07	0.96
LFP	0.10	-	-	0.90
NMC 811	0.11	0.75	0.09	0.77
NMC 622	0.13	0.61	0.19	0.76
NMC 111	0.15	0.40	0.4	0.82
NMC 955	0.10	0.70	0.04	0.76
NMC 532	0.14	0.59	0.23	0.80

Table 5.4 1Recycling efficiencies (%) (Lebedeva et al., 2017; Sanclemente Crespo et al.2022; Xu et al., 2020).

	Li	Ni	Co	Cu
2013	0.0	75.5	75.5	75.5
2014	0.0	76.7	76.7	76.7
2015	0.0	77.8	77.8	77.8
2016	0.0	79.0	79.0	79.0
2017	0.0	80.0	80.0	80.0
2018	4.4	81.3	81.3	81.3
2019	8.7	82.5	82.5	82.5
2020	13.1	83.7	83.7	83.7
2021	17.5	84.8	84.8	84.8
2022	21.9	86.0	86.0	86.0
2023	26.2	87.1	87.1	87.1
2024	30.6	88.3	88.3	88.3
2025	35.0	90.0	90.0	90.0
2026	42.0	90.6	90.6	90.6
2027	49.0	91.8	91.8	91.8
2028	56.0	93.0	93.0	93.0
2029	63.0	94.1	94.1	94.1
2030	70.0	95.0	95.0	95.0
2031	77.0	96.4	96.4	96.4
2032	84.0	97.6	97.6	97.6
2033	90.0	98.0	98.0	98.0
2034	90.0	98.0	98.0	98.0
2035	90.0	98.0	98.0	98.0
2036	90.0	98.0	98.0	98.0
2037	90.0	98.0	98.0	98.0
2038	90.0	98.0	98.0	98.0
2039	90.0	98.0	98.0	98.0
2040	90.0	98.0	98.0	98.0

3. Results and discussion

Utilizing the material and substance flow analysis and the data parameters as described in Section 3, the EoL LIBs are estimated on an EU-27 level and a country level for Germany and France. Sensitivity analysis involves investigating how future EVs sales and different lifespans affect the estimations of batteries waste. The impact of two sales models and two lifespan scenarios is investigated.

3.1 EV batteries reaching the end of their life

The EV batteries reaching their EoL are calculated by Eqs. (1) and (2) using the annual sales data for EV batteries in the EU-27 (STEPS and SDS) and the two lifespan scenarios (LL and HL). The results for the four scenarios are presented in Fig. 5.3. As can be seen, the LIBs predicted by the STEPS model are lower than those of the SDS model and those predicted by the HL scenario are lower than the corresponding of the LL scenario. It is estimated that between 2011-2035 about 9-14 million EoL LIBs will be accumulated in the EU-27 according to the STEPS model for LL and HL scenarios (17-27 million in 2011-2040). For the European continent (including UK, Iceland, Norway and Switzerland), the accumulated EoL LIBs will be about 20-30 million by 2040 for the same scenarios (STEPS model for HL, LL). Until 2017, the EVs sales share in the four non-EU countries was between 23-49 % and has then started to decrease, explaining the higher predicted amounts of EoL batteries.

For the SD Scenario, the accrued batteries in the EU-27 by 2035 will be 10.5-18 million EoL LIBs (23.5-39.4 million by 2040) for HL and LL scenarios. As for the entire European continent the accumulated batteries will be 26-43 million EoL batteries by 2040.

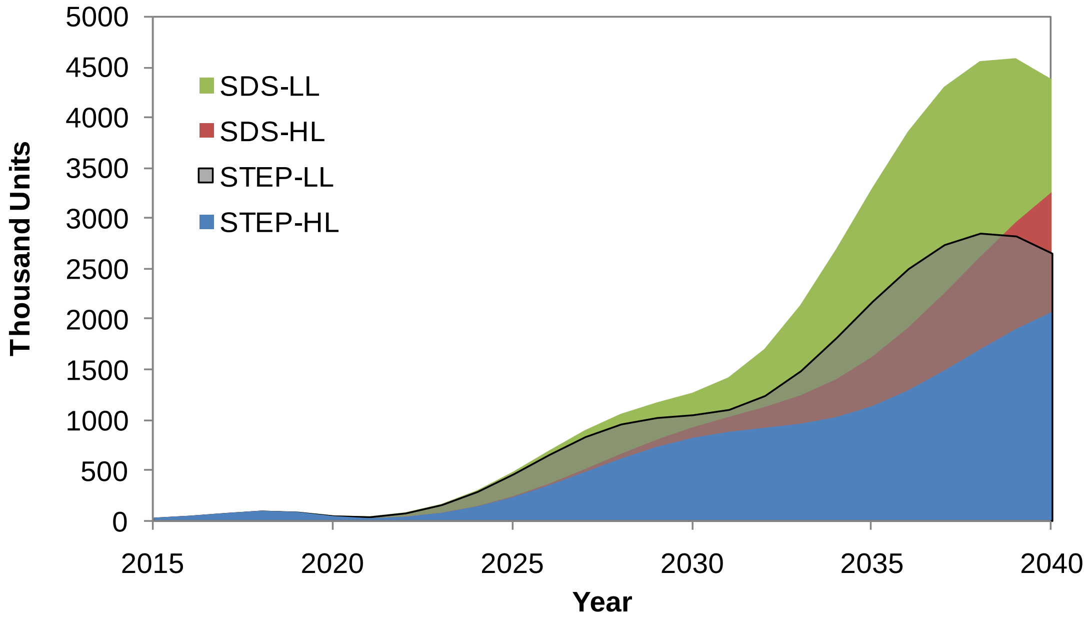


Figure 5.3. Amounts of EV batteries reaching their EoL according to the examined scenarios.

In the EU-27, 0.82-1.3 and 1.14-3.3 million EoL batteries will be generated in 2030 and 2035, respectively, while 2.1-4.4 million EoL batteries in 2040 (2.2-4.8 million in the European continent). The results of the present study reasonably agree with Drabik and Rizos (2018) who estimated 1.2 million EoL batteries in 2030 and 2.6 million in 2035. Drabik and Rizos (2018) have adopted a fixed lifespan of 8 years until 2040 (instead of a distribution) and one sales scenario.

As the waste calculations are based on EV sales data till 2030, differences may occur according to new sales (after 2030) and the scenario considered. The new sales will start to significantly impact the results after 2035 for STEPS-LL and SDS-LL, and after 2036 for STEPS-HL and SDS-HL scenarios. For example, if a linear increase in EV sales is assumed from 2030 to 2040 (based on the 2017-2030 trend) in STEPS-HL scenario, the LIB waste will increase by 2 % in 2034, 5 % in 2035 and 9 % in 2036. For SDS-LL scenario, assuming a linear increase in sales in 2030-2040 (based on the 2020-2040 trend) the LIB waste will increase by 1.7 % in 2033, 4.7 % in 2034 and 9.6 % in 2035. However, due to the global economic and political situation long term forecasts on sales data (esp. after 2035) may not be accurate; this study utilizes as reliable sales and capacity data as possible, focusing on obsolete LIBs that will be generated in the period 2020-2035. In addition, long term projections on the share of new cell technologies and capacities after 2030 would be required; however, these may involve high uncertainty.

3.2 EV batteries available for remanufacturing and repurposing

LIBs remanufacturing could be practically addressed by the manufacturing companies, as the battery cells are not usually labeled. Dismantling into cells is initially performed to check the SoH and later the suitable cells are reassembled and usually new battery management and control systems need to be implemented. The cost rises due to manipulation, testing and implementation of new control systems that may make remanufacturing unprofitable (Kotak et al., 2021).

Considering only the EoL BEV batteries with a SoH>90 %, as explained in Section 3.4 and using the eq. (4) as adjusted, the batteries available for remanufacturing are calculated. By 2024-2034, 113-301 thousand BEVs batteries will be remanufactured (136-361 thousand for the European continent) according to STEPS model and about 202-553 thousand BEV batteries according to the SDS model, respectively. The total accumulated capacity of remanufactured batteries will be 6-31 GWh. In 2030, 8-47 thousand batteries will be remanufactured, accounting for 0.14-0.45 % of the new EV sales in that year. In 2033, 10-76 thousand batteries will be directed to remanufacturing (annually). In SDS-LL, 23-76.3 thousand LIBs will be remanufactured annually from 2024-2034, while in the STEPS-LL scenario 14.5-42.4 thousand LIBs. Rohr et al. (2017) estimated, considering a factory capacity for remanufacturing of 5,000 batteries per year in Germany (and all associated labour and logistics costs), that remanufacturing would be profitable after 2017 in Germany (examining the period 2015-2030). The remanufactured LIBs in Germany will be 7-29 thousand annually in 2024-2034 (see section 4.4), as this works considers remanufacturing will take place by 2023 in the EU. The calculated LIBs exceed 5,000 unites, thus remanufacturing seems profitable. The BEVs sold in the EU-27 in 2018 and 2020 were 133 and 543 thousand, respectively, but sales are increasing exponentially in the forthcoming years. Although the number of remanufactured LIBs is relatively small, remanufacturing is a promising option to strengthen the EU as battery seller.

Regarding the LIBs reuse, not all retired batteries are suitable for repurposing options (2L). To address LIBs for a second life, battery diagnostics on SoH, and battery control systems must be performed. Since batteries for a 2L are retired batteries not all of them will pass a safety check and not all batteries will be accepted for reuse. Dismantling into cells will enable evaluation of electrical capabilities and selection of the appropriate use cases and exclusion of unsuitable cells (Kotak et al., 2021; Wöhrl et al., 2021). Employing the reuse rates as described in Section 3.4 and eq. (4), the accumulated BEV Batteries available for a Second Life (B2L) will be 1.7-2.5 million by 2035 and 3.4-5.4 million by 2040 according to STEPS model. The accumulated BEV B2L will be 2.4-4.2 million by 2035 and 6-13.7 million by 2040 for the SDS model. On the other hand, the PHEV batteries B2L will be 2.2-3.5 million by 2035 and 4.7-7.4 million by 2040 in STEPS, and 2.3-3.9 and 5.1-8.4 million by 2035 and 2040, respectively, for SDS model. Fig. 5.4 (a) presents the total batteries (BEV and PHEV) available for B2L according to all models. In 2035, 0.5-1.7 million LIBs will be available for B2L which reasonably agrees with the results of Bobba et al. (2019).

Fig. 5.4 (b) presents the energy storage capacity available for a second life (B2L). This ranges from 94-152 GWh accumulated by 2035 (2011-2035) in STEPS, and 131-233 GWh in SDS, respectively. By 2040, around 212-347 GWh B2L will be accumulated (2011-2040) in STEPS and 345-617 GWh in SDS, respectively. In 2035, B2L capacity ranges from 15-55 GWh compared to 9-34 GWh reported by Bobba et al. (2019). They have used lower capacity for BEV and PHEV batteries, i.e., they adopted 20-57 kWh from 2005 to 2030 for BEV batteries capacity as opposed to 24-72 kWh from 2010 to 2030 in the current study. In 2040, Abdelbaky et al. (2021) have calculated 20-70 GWh available for B2L as compared to 29-80 GWh in this study, although they used sales data till 2040 (instead of 2030 in this study). The share of battery technologies for B2L is depicted in Fig. 5.5 for STEPS-LL and SDS-LL scenarios. As observed, until 2019 the dominant technology among batteries available for B2L was NMC 111 (21.1-56.8 %), then LFP (18.3-27.3 %), while from 2020-2025 the NCA (21.8-26.2 %), LFP (21.7-26 %) and from 2026-2040 the NMC 811 (20.1-28.1 %), followed by NMC 622 (16.4-23.2 %). Abdelbaky et al. (2021), Bobba et al. (2019) and Drabik and Rizos (2018) have not estimated the technology share for B2L, so there is no direct comparison. Nowadays, there are several real-world successful projects of B2L LIBs that provide energy storage and grid services, like the 2.8 MWh ESS installed in 2018 at “Johan Cruyff Arena” in Amsterdam (Robb, 2018; Pagliaro and Meneguzzo, 2019). Another successful example is the SUNBATT project in Spain led by SEAT with 2L LIBs as ESS. The SUNBATT container supports an 8 kW solar carport, 3 EV chargers, 1 Fast EV charger and the grid, offering 90 kW peak power (Casals et al., 2018). The real life cases show that utilizing B2L as stationary applications offers environmental and economic benefits.

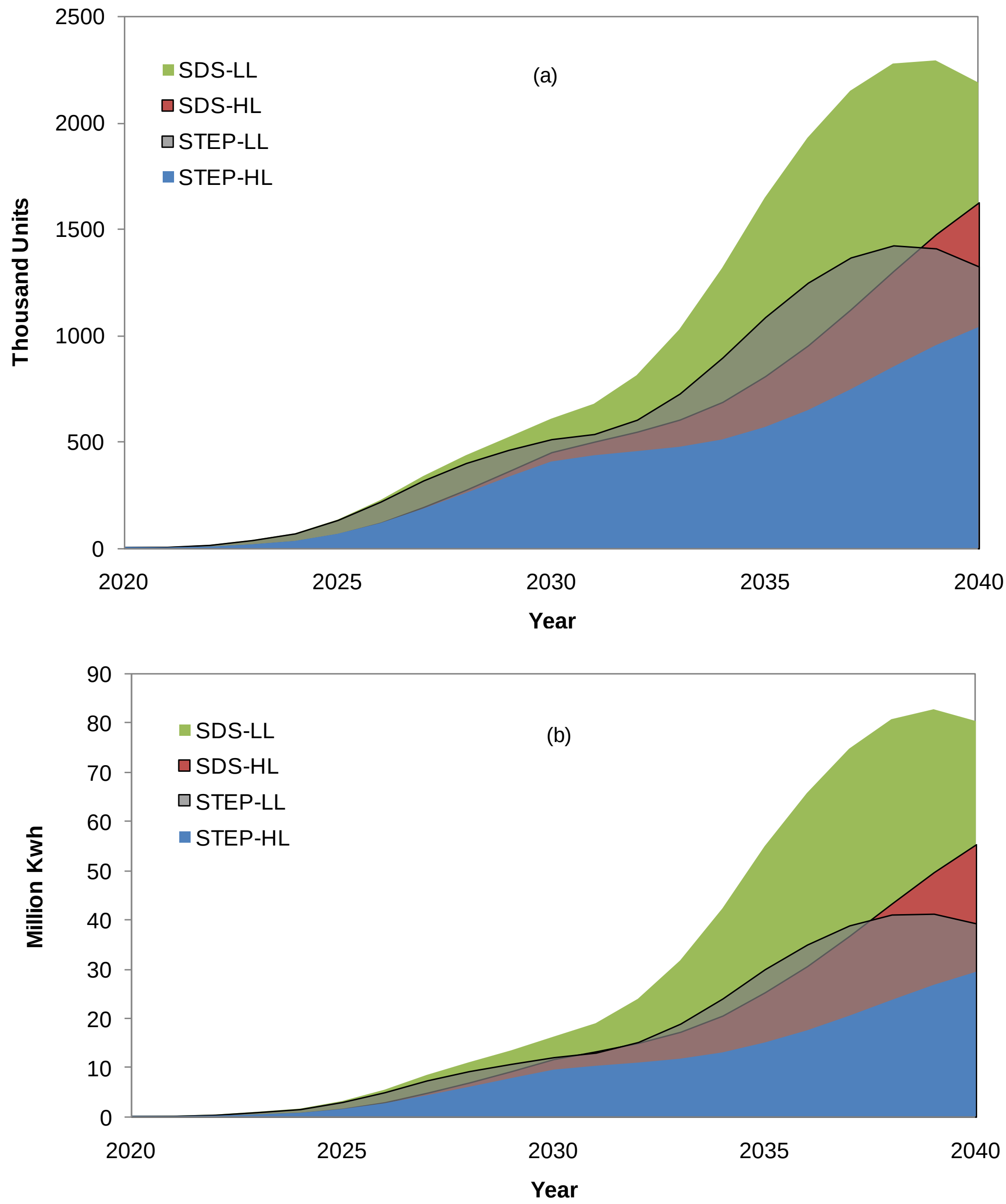


Figure 5. 4 (a) Total batteries (BEV and PHEV) available for a second life (B2L) according to all scenarios and (b) Energy storage capacity available for second life.

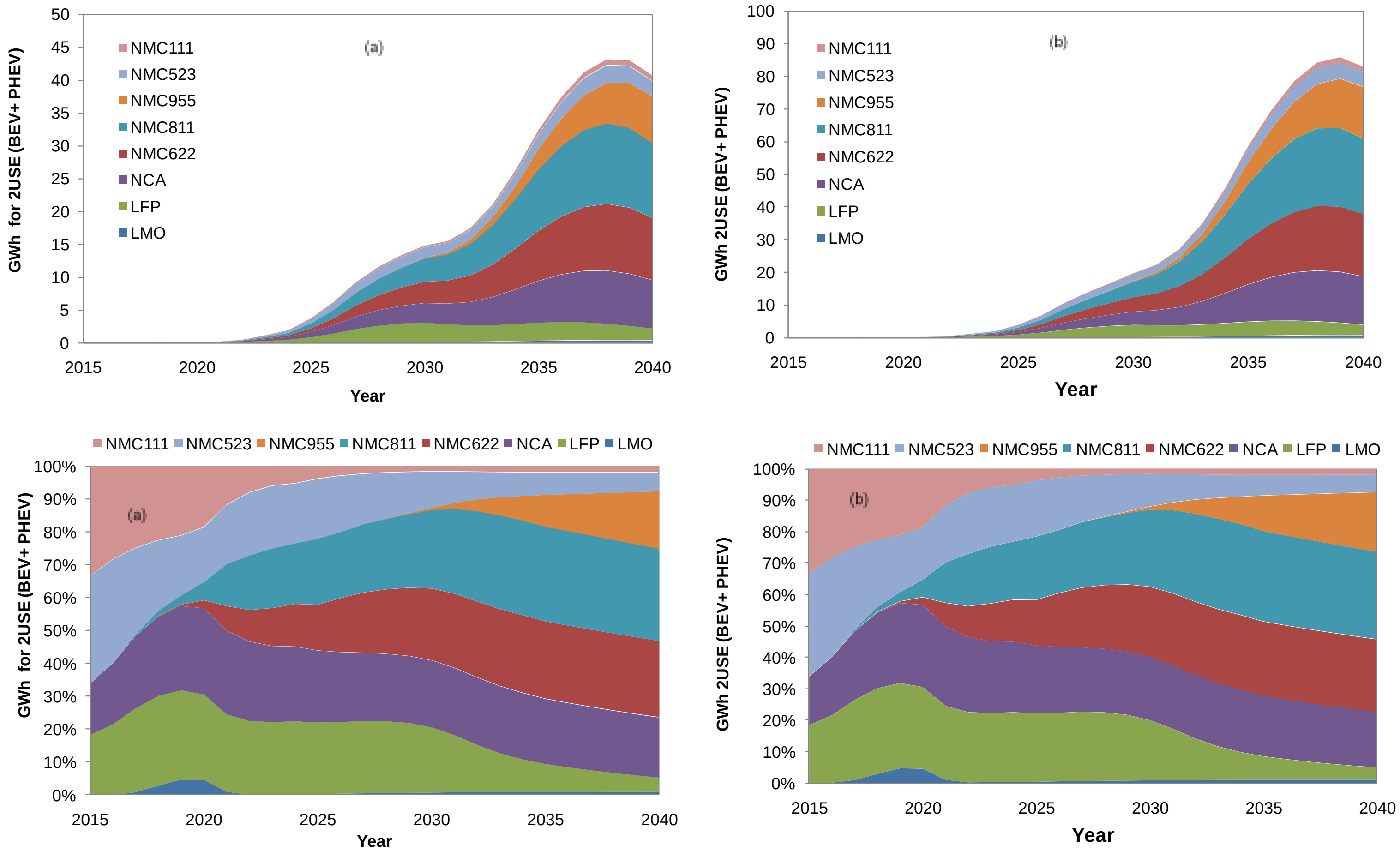


Figure 5. 5 The share of battery technologies available for B2L according to (a) STEPS-LL (b) and SDS-LL models. (Lithium Nickel Manganese Cobalt Oxide (NMC): NMC 111 (1:1:1), NMC 523 (5:2:3), NMC 622(6:2:2), NMC 811 (8:1:1), NMC 955 (9:0.5:0.5); Lithium Nickel Cobalt Aluminum (NCA); Lithium Manganese Oxide (LMO); Lithium iron Phosphate (LFP).

3.3 EV batteries available for recycling and secondary raw materials

The batteries available for Recycling (B2R) are calculated as explained in Section 3.4, eq. (4) and Fig. 5.1. The batteries ending up for recycling are the share of batteries after their 1st use and waste batteries after B2L (eq. 4). Fig. 5.6 shows the sum of B2R after 1st and 2nd Life for all scenarios. In addition, Fig. 5.7 illustrates the capacity and relative shares of B2R, B2L and B Remanufactured (for the scenarios STEPS-HL and SDS-LL). The available capacity of B2R is greater, as the batteries after 2L will also be recycled. Batteries of 182-442 GWh will be directed to recycling between 2011-2035 (391-1100 GWh between 2011-2040). By 2030, the recycling infrastructure with an annual recycling capacity of 15-28 GWh will be necessary under the considered scenarios. This capacity is greater than the 10 GWh in 2030 predicted by Abdelbaky et al. (2021) for Europe (for the same recycling rate). By 2035 and 2040, the annual recycling capacity will reach 26-90 GWh and 53-146 GWh, respectively, compared to 48 GWh in 2035 and 125 GWh in 2040 predicted by Abdelbaky et al. (2021) for Europe (for the same recycling rate). The B2L, B2R quantities in the SDS scenario are larger than in STEPS as the EV sales under the SDS scenario are larger, resulting in more battery waste, since the waste calculations are based on sales data until, 2030 (section 4.1,), there will be slight differences (increase) in resulting waste when sales data after 203 0 are regarded. In this case, there will be a continuous increase in B2L and B2R after 2035 (rather than a slight decrease in 2039 as depicted in Fig. 5. 7(b)). The % differences for a linear increase in sales data after 2030 are commented in section 4.1. However as long term forecasting of EVs sales involves high uncertainty, this study chose to use as reliable data as possible. Fig. 5.8 shows the share of battery technologies for B2R for STEPS-HL and SDS-LL models. Until 2020, the dominant technology in the recycling stream is NMC 111 (19-57 %), while from 2021-25 is NCA (21-25%), followed by LFP (22-24 %). During 2026-2030, LFP (18-22 %), NCA (20-21.5 %), and NMC 811 (20-24 %) have similar shares, while from 2031-2040 NMC 811 dominates (23-28 %), followed by NMC 622 (20-23 %). Overall, NMC cathodes represent the largest share of the waste stream (42-78 %), highlighting the need to develop recycling technologies for this type of cathodes.

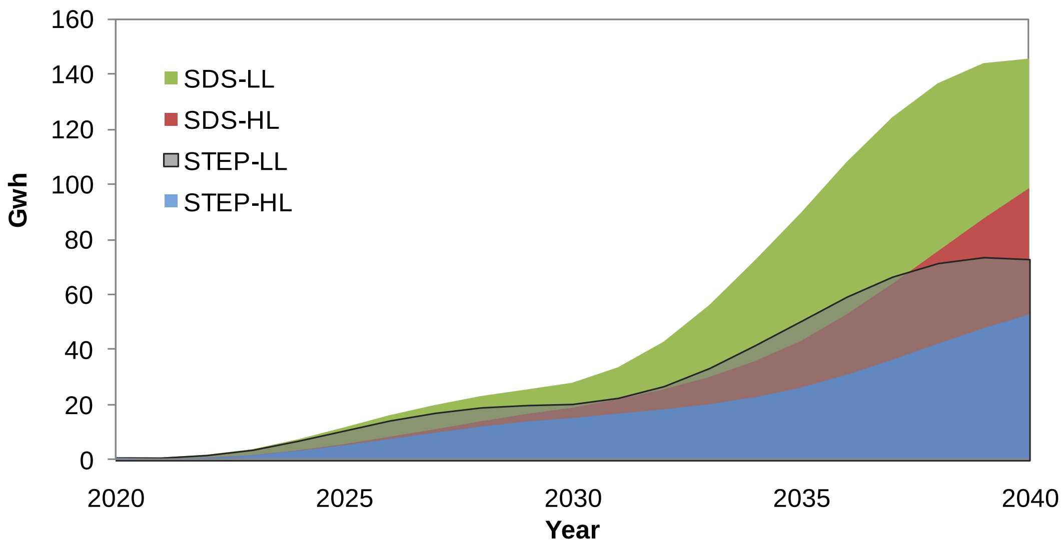


Figure 5. 6.Total (BEV+PHEV) batteries to Recycle (B2R) after 1st and 2nd use.

Abdelbaky et al. (2021) also concluded that NMC cathode batteries will prevail from 2025 onwards. However, the results are slightly different due to different market shares of LIB cells considered in each study.

Taking into account the recycling efficiency of the metals in the LIBs, as explained in Section 3.7, Li, Co, Ni and Cu are calculated in the B2R stream. The results are presented in Fig. 5.9 for all scenarios. The quantities that can be recycled according to SDS model are more significant than STEPS. In 2030, the annual recycled quantities of Li, Co, Ni and Cu will range between 1.29-2.35, 2.4-4.2, 9.35-17.6, 11.4-20.8 kt, respectively, according to the examined scenarios. The reported Li quantities roughly agree with Abdelbaky et al. (2021) in 2030 who reported 1.3 kt available in 2030 for the average recycling scenario. They also calculated 2 kt Co, 5 kt Ni and 8.3 kt Cu in 2030, which are less than the calculated quantities in this study. In 2030, Bobba et al. (2019) estimated that 3-6.5 kt Co will enter the recycling stream. However, in contrast to the previous mentioned studies that calculate the maximum recycling potential, the current study takes into account the recycling efficiencies set by the EU battery directive (70 % for Li in 2030 and 95 % for Co, Ni, and Cu). The recycled quantities of Li, Co, Ni and Cu in 2030 correspond to 5.2-6.2, 9-11.3, 6-6.9

and 6.1-7.2 % of the annual demand according to the sales of new EVs in the same year. Abdelbaky et al. (2021) calculated for Li, Co, Ni and Cu that the annual recycled quantities correspond to 5 % of the sales demand in 2030. The annual recycled quantities of Li, Co, Ni and Cu in 2035 will be 2.9-9.6, 4-12.8, 17.4-59.5, 20.7-69.5 kt, respectively. Also, the annual recycled quantities of Li, Co, Ni and Cu in 2040 will be 5.8-15.7, 7.7-19.9, 35.8-98.8, 42.2-114.7 kt, respectively. In 2040, Abdelbaky et al. (2021) estimated 15 kt Li, which agrees to the calculations of the present study. However, Abdelbaky et al. used EV sales data till 2040 instead of 2030, which yields more LIBs waste after 2035 compared to the present study that considers sales till 2030 and also they also calculated the maximum recycling potential and did not consider the metals' recycling efficiencies. The recovered quantity of Li has significantly increased because of the increase in the recycling efficiency as dictated by the EU battery directive from 13 % in 2020 to 70 % in 2030 and 100 % in 2035. Since global Li production in 2019 was 77 kt, secondary raw materials from B2R option will emerge as a significant supply (Abdelbaky et al., 2021).

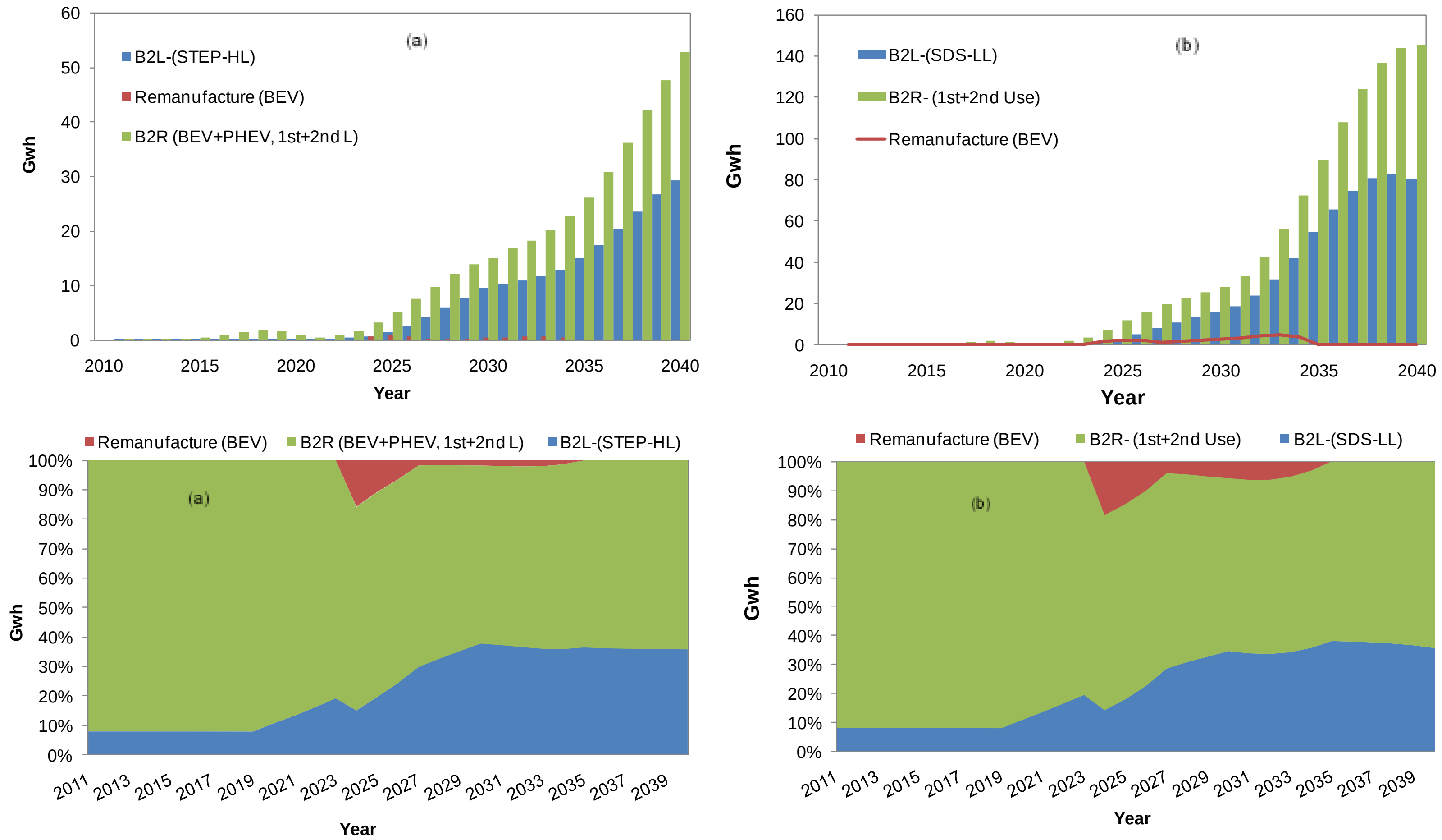


Figure 5. 7 B2L, B Remanufacturing and B2R after 1st and 2nd use (BEV+PHEV) according to (a) STEPS-HL and (b) SDS-LL scenarios.

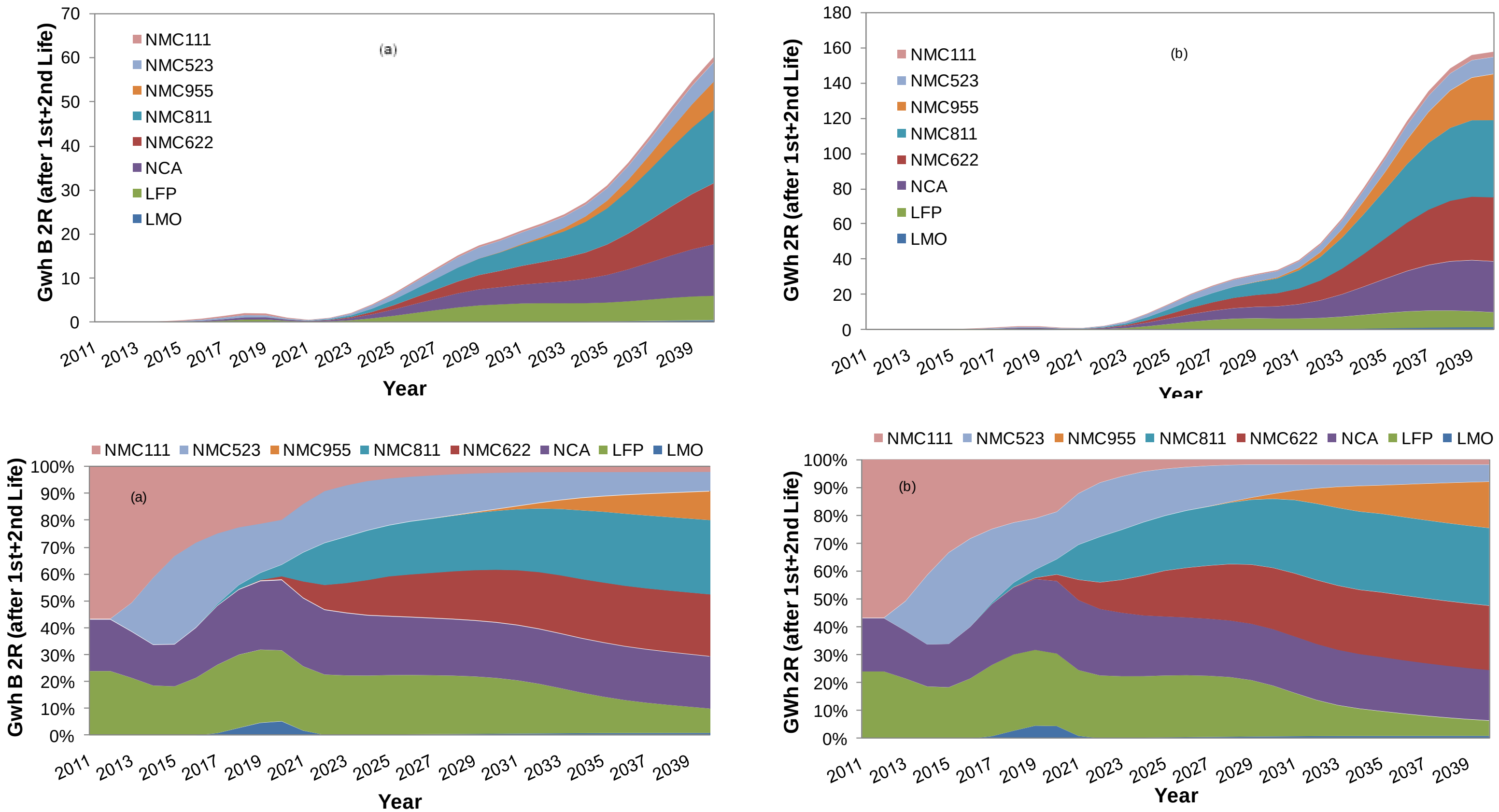


Figure 5. 8 Share of battery technologies available for recycling after 1st and 2nd life according to (a) STEPS-HL and (b) SDS-LL models. (Lithium Nickel Manganese Cobalt Oxide (NMC): NMC 111 (1:1:1), NMC 523 (5:2:3), NMC 622(6:2:2), NMC 811 (8:1:1), NMC 955 (9:0.5:0.5);Lithium Nickel Cobalt Aluminum (NCA); Lithium Manganese Oxide (LMO); Lithium iron Phosphate (LFP).

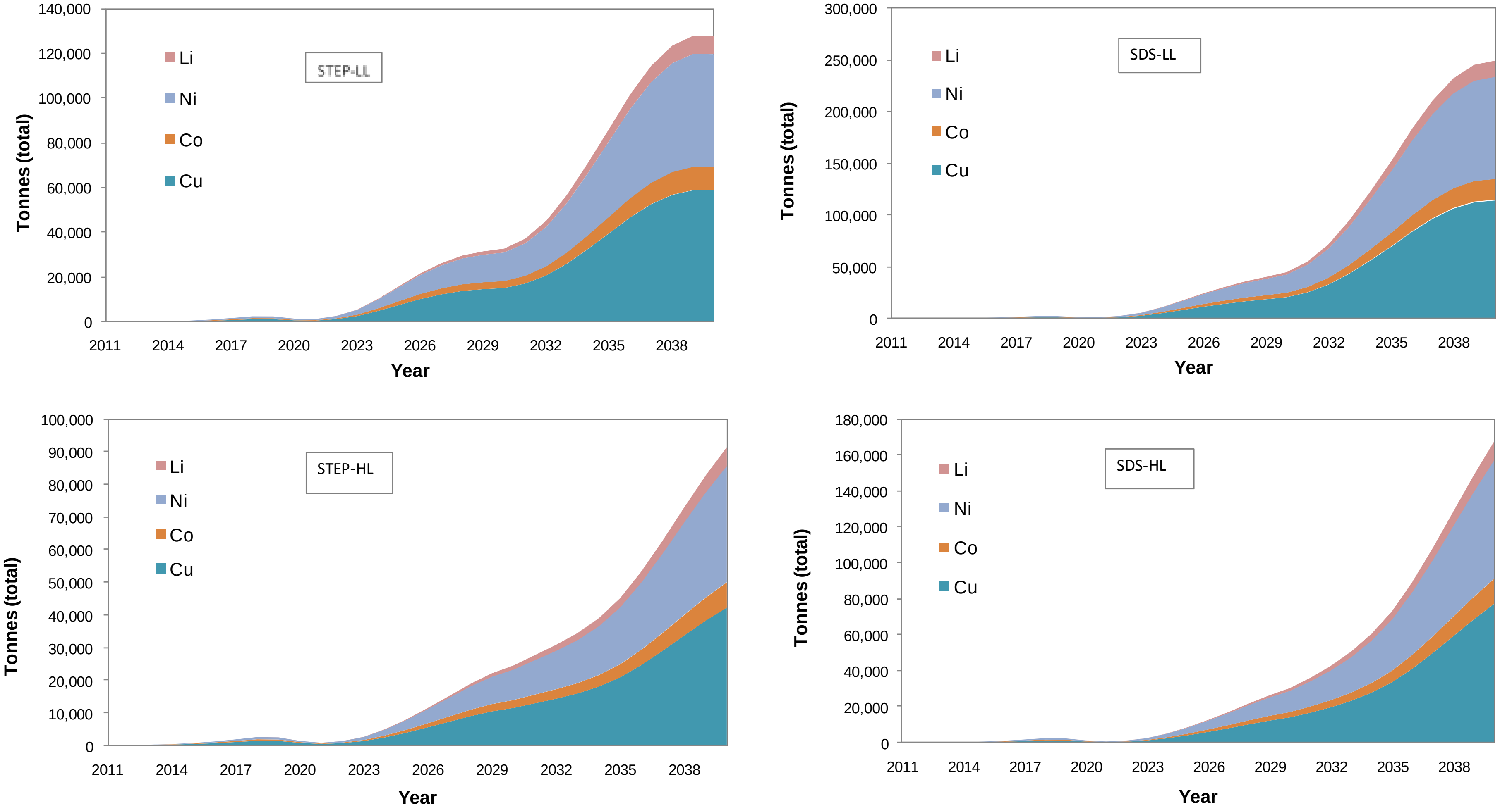


Figure 5. 9 Metals recovery from LIB 2R according to STEPS-LL, STEPS-HL, SDS-LL and SDS-HL scenarios.

3.4 Batteries available for second life in Germany and France

The concept of coupling retired LIBs with PV energy production is further assessed. A key obstacle to achieving high power efficiency for renewable energy systems, such as solar PV, is the potential intermittency that causes grid fluctuations. Stationary batteries can be used as an ESS to overcome this drawback (Volan et al, 2021). A primary driver for the cost of repurposing LIBs for a second life is the logistics involved in the collection, apart from SoH testing, physical breakup and repacking of cells (Zhu et al., 2021). Local collection is more cost effective and reuse within a country is preferable, if there is a market to absorb the offered B2L.

The total available battery capacity for a second life in Germany for LL and HL scenarios is shown in Fig. 5.10. In Fig. 5.11 and Fig. 5.12, the capacity of EoL LIBs, B2R and B remanufacture are depicted for Germany. The cumulative B2L capacity is 13.7-29.1 GWh until 2030 and 38.4-89 GWh from 2031-2035 (140-293- GWh from 2031-2040). Assuming that the cumulative battery capacity could support the integration of PV power sources until 2030 (98 GW PV installations in Germany in 2030), there would be a need of 49 GWh battery capacity, according to the IRENA report (Kempener and de Vivero, 2015), as for 100 kW PV capacity, 50 kWh batteries with 25 % capacity loss and 80 % depth of discharge are required. In this way, until 2030, the accumulated B2L could cover between 28-60 % of the required battery capacity to support PV installations. In the forthcoming years (2031-2040), the available batteries for 2nd Life increase significantly, making integration with renewable energy installations very attractive. However, by 2032 the accumulated B2L will be 24.7-50.8 GWh covering 50-104 % of stationary battery storage needs. The cumulative capacity of B2L in 2031-2036 increases to 52.4-123 GWh, enough to adequately support PV renewable energy production (107-253 %). Between 2032-2036, 12-35 GWh B2L will be available annually, while from 2037-2040, 37-43 GWh (LL scenario). Thus, a significant opportunity is presented to cover the stationary energy needs of PV systems with the available capacity from B2L.

The total available B2L capacity in France for LL and HL scenario is shown in Fig. 5.13. Also, in Fig. 5.14 the capacity of EoL LIBs, B2R and B remanufacture are depicted for France. The cumulative B2L capacity is 5.4-18.22 GWh until 2030 and 11-42 GWh from 2031-2035 (44.7-139.2 GWh from 2031-2040). To support the 42 GW PV installations in France by 2030, 21 GWh battery capacity would be needed, as previously explained. Therefore by 2030, the accumulated B2L could cover between 26-87 % of the required battery capacity to support PV installations. However, by 2031 the accumulated B2L will be 20.74-23.3 GWh covering 99-111 % of stationary battery storage needs. France is the third country in the EU-27 in planned PV deployment capacity, and the second in EVs adoption. Thus, the capacity of B2L is significant and can effectively support stationary battery storage needs.

LIBs have been widely incorporated into renewable energy systems due to their advantages such as high energy and power density, high reliability and long operating time (Hua et al., 2021). Retired NMC and LFP batteries have been proven reliable for energy storage with a proper battery management system (Hart et al., 2014). For example, in Europe a real B2L project for stationary storage coupled to PV power station has been implemented in 2015 by Bosch, BMW and Vattenfall (Gohla-Neudecker et al., 2015). More than 100 retired LIBs have been used as the storage unit with an output of 2 MW and an installed capacity of 2.8 MWh in Hamburg. Numerous studies have concluded, by assessing the battery degradation patterns, that coupling retired LIBs with PV systems is technically and economically feasible (Tong et al., 2015; Assunção et al., 2016; Lee et al., 2022).

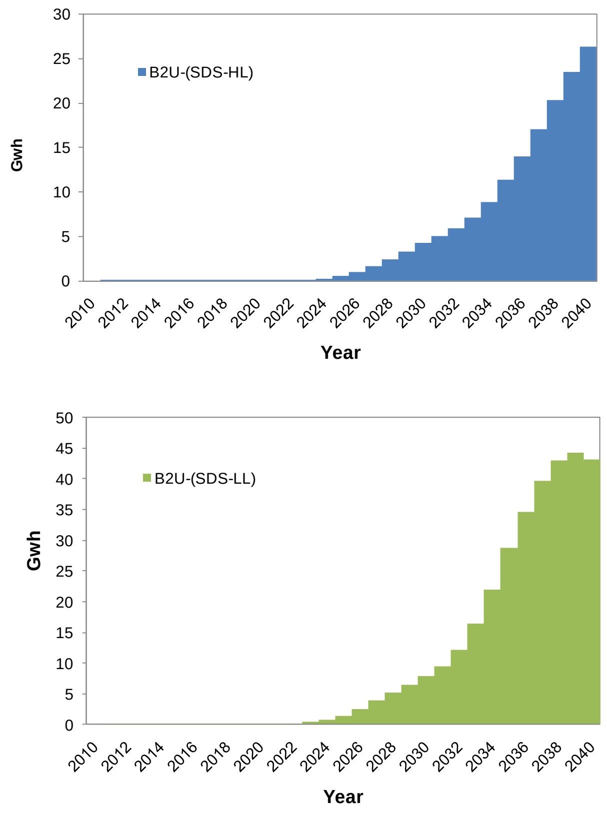


Figure 5. 10 The total available battery capacity for a second life in Germany for LL and HL

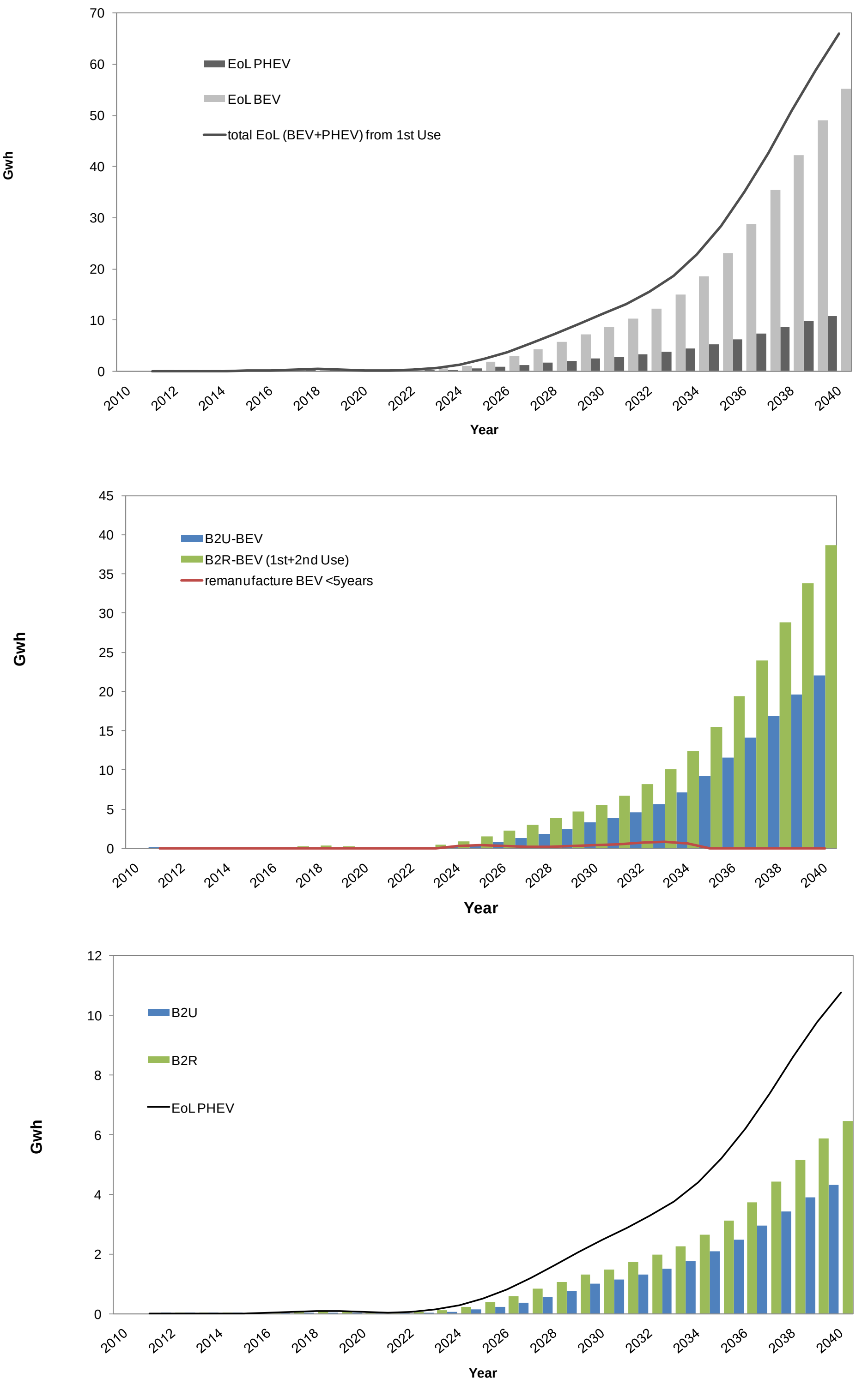


Figure 5. 11. EoL LIBs capacity and B2R and remanufacture for Germany (HL scenario).

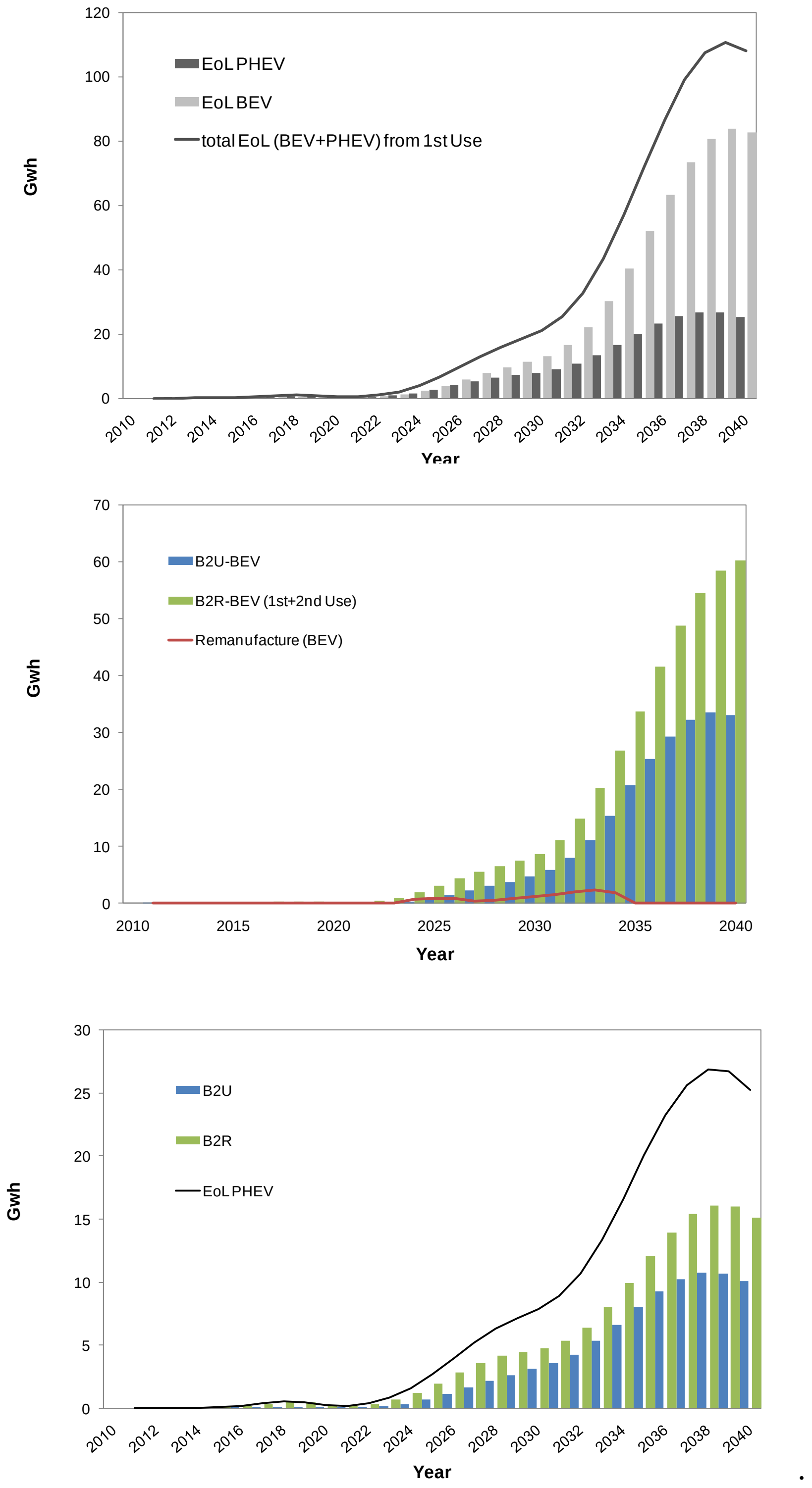
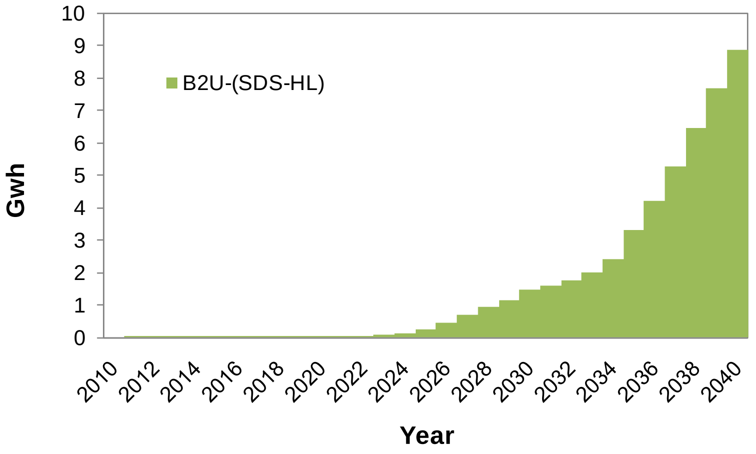


Figure 5. 12 EoL LIBs capacity and B2R and remanufacture for Germany (LL scenario).



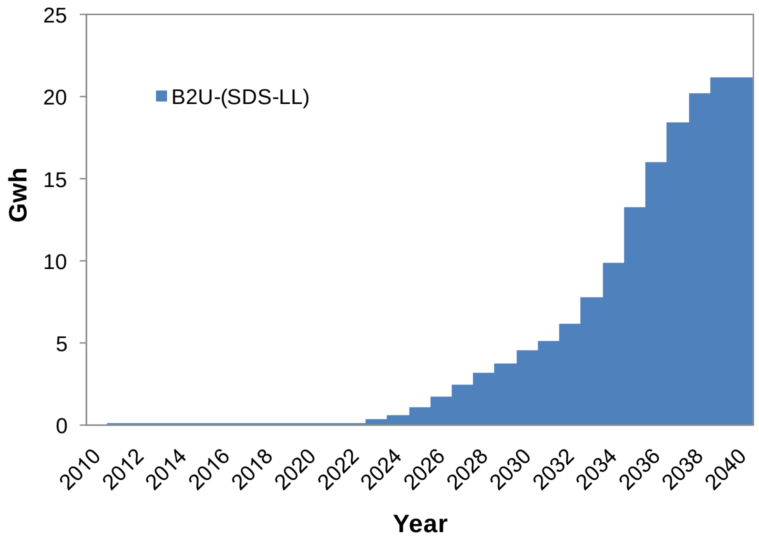


Figure 5. 13. Total available battery capacity for a second life in France for LL and HL scenario.

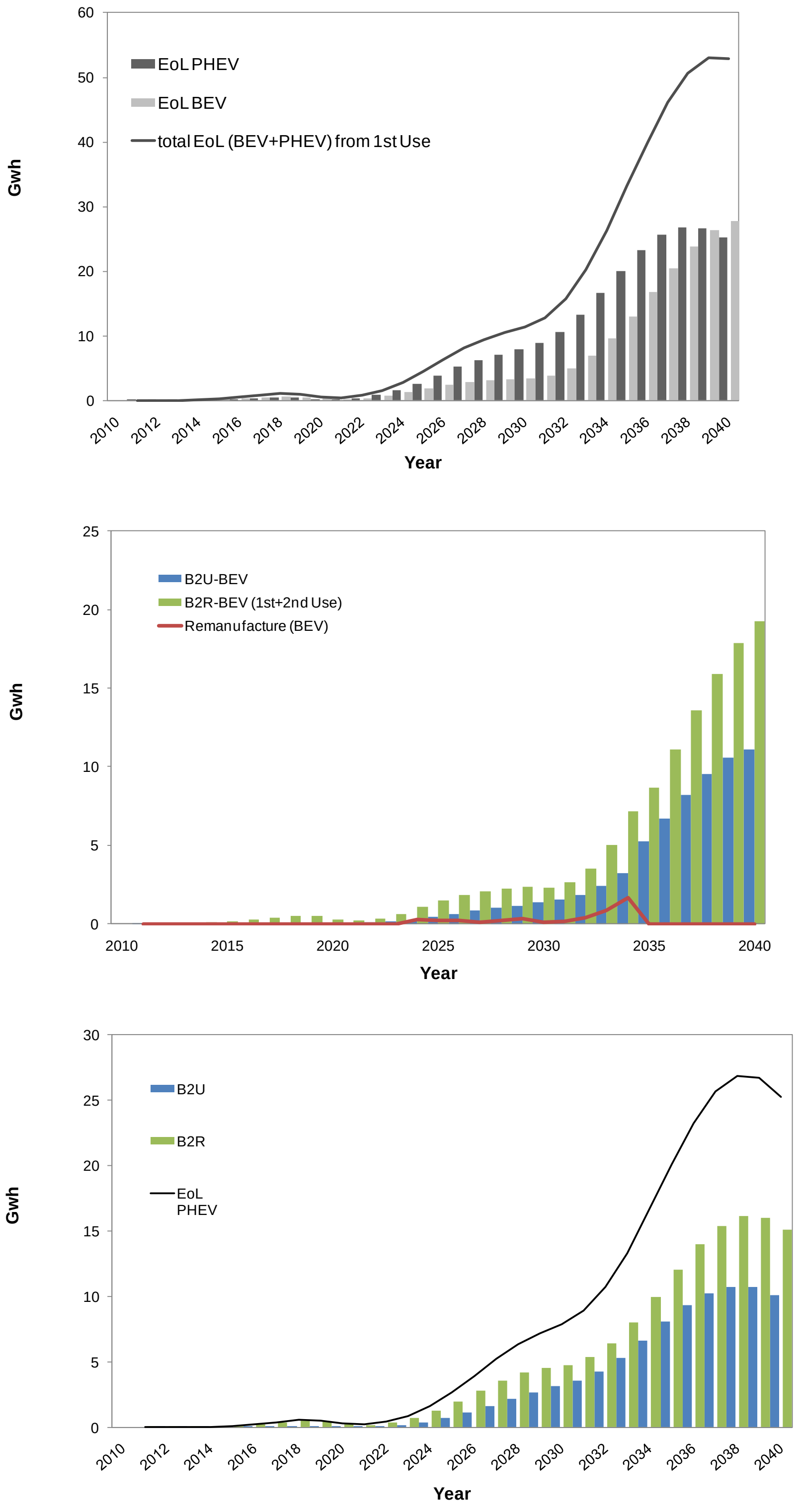


Figure 5. 14 EoL LIBs capacity and B2R and remanufacture for France (LL scenario).

4. Conclusions

The adoption of circular economy business models is necessary to deal with the increasing volumes of EoL LIBs from electric vehicles. These models involve Remanufacturing, Reuse and Recycling (3R) of the waste batteries to extend their life before recovering valuable materials through recycling. This study forecasts the amounts and capacity of batteries directed to Remanufacturing, Reuse and Recycling in the EU-27, as well as in Germany and France at country level, which are the countries with the highest EVs adoption. Between 2011-2035, about 9-17 17-39.4 million EoL LIBs will be accumulated in EU-27 (17-39.4 million in 2011-2040), while 113-553 thousand BEVs will be remanufactured by 2024-2034 (6-31 GWh). The accumulated LIBs available for a second life (2L) will be 3.9-7.9 million by 2035 (8-18 million by 2040) with an energy storage capacity of 212-617 GWh. The dominant technology among batteries available for 2L until 2019 is NMC 111, while from 2020-2025 the NCA, followed by LFP and from 2026-2040 the NMC 811, followed by NMC 622. About 182-442 GWh LIBs will be directed to recycling in 2011-2035 (391-1100 GWh between 2011-2040). By 2030, a recycling infrastructure with an annual recycling capacity of 15-28 GWh will be necessary. The recycled quantities of Li, Co, Ni and Cu in 2030 correspond to 5.2-6.2, 9-11.3, 6-6.9 and 6.1-7.2 % of the annual demand according to the sales of new EVs in the same year.

Future investments in battery remanufacturing, reuse and recycling infrastructures are necessary to deal with the increasing amounts of LIBs waste. The batteries available for a second life offer a great opportunity to cover the stationary energy needs of PV systems. In Germany, the accumulated B2L capacity until 2030 could cover between 28-60 % of the required battery capacity to support 98 GW PV installations. In France, the batteries available for a second life could cover 26-87 % of stationary storage needs of 42 GW PV installations by 2030.

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CHAPTER 6

Energy decarbonisation in the European Union: Assessment of photovoltaic waste recycling potential

The Renewable Energy Directive delineates policies for energy production from renewable sources by at least 32% in European Union (EU) by 2030. All member states have established National Energy and Climate Plans (NECPs) for 2021-2030 to decipher how they will cover their energy needs from renewable sources. The present work considers the targets set by each of the EU-27 countries to implement, in particular, solar photovoltaic (PV) modules to cover their energy needs. Then, the future PV waste amounts are assessed considering the widely used Early Loss and Regular Loss scenarios, as well as the noteworthy scenario proposed by the EU WEEE Directive. The study examines the questions “when will large amounts of panel waste be generated in the EU countries and what will their composition be?” In addition, a timescale for starting an economically viable recycling industry for PV panel waste in the EU is estimated based on the annual PV waste generated in each country. By 2050, 14.3-18.5 Mt PV waste will be generated in EU-27 while the profit of PV recovered materials will be 21.98-27.36 billion USD. The findings contribute to the efficient management of the forthcoming e-waste category, according to circular economy principles, ensuring the pathway to sustainability.

1. Overview

The European Union (EU) aims to develop its strategy and infrastructure for further decarbonisation of the energy system towards 2050. The European Green Deal is a new growth strategy intending to transform the EU into a sustainable, equitable and prosperous society, efficient in resources and without net emissions of greenhouse gases by 2050. It outlines ways to accelerate the development and adoption of low-carbon technologies with respect to the targets set for 2030 and 2050, while the Renewable Energy Directive delineates policies for energy production from renewable sources by covering at least 32% of EU's total energy needs with renewable energy by 2030 [1].

The decarbonisation of the energy sector is essential as the production and use of energy are responsible for more than 75% of the greenhouse gas emissions in the EU. Energy adequacy is a priority in the EU and this can be accomplished by mainly investing on renewable sources. Increasing offshore wind production, as well as photovoltaic installations is essential. In the EU, all member states have established National Energy and Climate Plans (NECPs) for 2021-2030 to decipher how they will cover their energy needs by renewable sources.

Solar photovoltaic (PV) panels are one of the fastest-growing future waste streams under the category of large electronic waste (WEEE). It is also one of the most important waste streams, as it contains valuable elements like selenium, tellurium, gallium, molybdenum, and indium [2]. The assessment of future PV waste amounts is of primary importance to plan their efficient management. Previous works have estimated PV waste in Italy, Spain, Austria, Flanders region in Belgium, Mexico, India, Australia, USA and the OECD countries [3-11]. However, there is no evaluation study for future quantities of PV waste in all EU-27 countries. This work comes to fill the research gap of PV waste assessment in all EU-27 countries. The differences in the methodology and the main parameters used in this study compared to the literature are reported in Table 1.

A key difference from all previous studies is that this is the first work to estimate future panel waste based on future PV deployment capacity commitments as accurately referred in the NECP of each country and not by projections based on historical trends. Thus, the estimations of future PV waste amounts in this study are more precise as the limitations of forecasts based on historical trends, are overcome. Indeed the latter forecasting method may fail if the past trend changes in the future, as is the case with the promotion of renewable energy technologies in the EU.. Second**,** all previous studies used either a fixed lifetime or a probability distribution considering an Early and Regular Loss scenarios as proposed by IRENA [12]. This is the first study that, in addition to the previous lifetime scenarios, also examines the scenario proposed by EU WEEE Directive and E-waste Statistics [13, 14]. Applying a fixed lifetime is not accurate as the failure pattern of a panel is not constant over time, but rather follows a probability distribution. Indeed, when real life data were evaluated, the use of a lifespan following a probability distribution was more suitable to describe PV panels' failure mode [12, 15]. The Regular Loss (RL) and Early Loss (EL) scenarios are widely used in the literature but there are no reported works for EU WEEE scenario. The EL and RL scenarios employ a characteristic lifetime of 30 years, while the EU-WEEE adopts 25 years resulting in average PVs lifetimes of 26.6, 27.7 and 22.5 years, respectively. The last scenario adopts a shorter lifetime, but this can reliably model the PV panel failures in conditions of transportation, installations and use in the EU.

Third, the waste per PV technology is not considered in the case of the EU countries such as Spain, Austria and Belgium [4-6]. In the case of Italy, only five different technologies (c-Si, a-Si, Cd-Te, CISG, CPV) are considered [3], as opposed to eight in the current study (c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other). The evaluations for India and Mexico consider only three to five different technologies [7, 8] and only the assessments for Australia and OECD countries consider eight technologies [9, 11]. As these PV panels have different structure and materials, different approaches are required regarding processing, recycling, and treatment to recover the valuable components. A quantification analysis of the respective shares is important for the PV-recyclers. Thus, the current work presents significant information for all EU countries. Fourth, this study considers 22 different metals and materials including critical metals (e.g., gallium, indium and tellurium), precious metals (e.g., silver), and toxic metals (e.g., cadmium, lead, selenium). Previous studies for EU countries either don't assess any material or evaluate less metals and materials (5-14 materials). The studies for non-EU and OECD countries also assess 21-22 metals and materials [9, 11]. Fifth, the conversion from installed PV power to mass is either fixed or fixed per technology or panel architecture in most studies, not accounting for the time factor that includes the technological improvement in the panels' efficiency and the material savings in new panels. In the current study (and in the case of Spain [4]), the conversion factor (tonnes per MW) was fitted to an exponential decay function as proposed by IRENA [12]. This makes the estimations more accurate and adds value to the presented results. Sixth, the present study and the OECD countries study are the only ones that provide a timescale for launching an economically viable recycling industry for PV panel waste. In the past, the low PV panel waste amount was the main reason behind the unsuccessful attempt of the German recycling company Solar World [16]. For a recycling industry to be successful it needs reliable predictions on panel waste amounts, as well as their composition. This will eliminate the investment risks associated with operating a recycling industry [6]. The present study answers the questions "when will large amounts of panel waste be generated in the EU and what will their composition be?" This is the first work that gives answers for all the EU countries in detail.

A PV waste recycling plant aims to maximize profits from the sales of recovered materials from panel waste, while reducing costs related to processes, stocks, transfers and capital investment. It is crucial, therefore, to develop and create an economically viable and environmentally sustainable recycling infrastructure for the emerging photovoltaic industry in conjunction with the accelerating market of these new technologies. The economic viability of such recycling program is closely associated to the geographical quantities of PV waste, the distance from the recycling plant and the amount of valuable materials for sale. Choi and Fthenakis estimated that a PV recycling plant is economically viable when the incoming PV waste reach a minimum annual amount of 20 kt [17].

Thus, the objectives of this chapter are: (i) to determine the future PV waste amounts in EU-27 countries considering the targets set by each country in the NECP for the implementation of solar photovoltaic modules, rather than making projections based on historical trends that may not capture the future policies promoting renewable sources, (ii) to use a lifespan distribution function proposed by the EU WEEE Directive in addition to the EL and RL scenarios proposed by IRENA, (iii) to assess the waste per PV technology (c-Si, a-Si, CdTe, CIGS, CPV, OPV, advanced c-Si and other) in EU-27, (iv) to make estimations of the accumulated amounts of 22 metals and materials including critical metals that could be recovered from panel waste and also assess the relative financial revenue, (v) to perform a sensitivity analysis regarding the material composition of PV panel waste and the material recycling yields, and (vi) to indicate EU candidate countries for the successful establishment of a photovoltaic recycling plant based on the annual PV waste generated in each country and to propose a timescale for starting a viable PV recycling industry in the EU-27.

The findings can contribute to the efficient management of PV waste in the EU, closing the loop between resources, products and waste. This can ensure a sustainable and reliable source of raw materials, which will support the increasing demand for minerals and metals associated with low-carbon technologies.

1.1 Historical and future PV installations in Europe

Historical installation data until 2019 are retrieved from IRENA [18]. Then, in order to overcome the uncertainties associated with future projections about PV deployment in EU 27 countries, data on targeted PV capacity by 2030 are retrieved from the national NECP of member countries. Some countries have included data on planned PV installations by 2040 (Austria, Luxembourg, Cyprus, Poland, and Ireland), however only installations till 2030 are considered for consistency reasons. Some NECPs enclosed detailed annual or 5-year-period targets. When no annual installation data are available, they are completed to match the 5-year-period or final (2030) installation capacity targets given in the NECPs.

Germany has the largest share of installed PV capacity in EU-27, ranging between 60-91% of the installed MW in the EU in the period 2000-2010 with an average annual PV growth rate (AAGR) of about 70%. The share then fell to 40-50% in 2011-2020 and will fall further between 20-30% in 2021-2030, as more EU countries will install PVs to reach the 2030 decarbonisation target set by the European Commission. The 10-year AAGR in Germany was 11.8% in 2011-2020, but it is expected to decrease to 6.7% in 2021-2030. The installed PV capacity will be about 98 GW in 2030, almost double compared to 2020.

Italy is the second country in PV deployment capacity in the EU-27 with a share of about 5, 21, and 14% of the installed MW in the EU in the decades 2000, 2010, 2020, respectively. AAGR peaked at 219% in 2007-2011, as there have been significant subsidies or feed-in-tariff support programs during these years. According to the NECP, the installed PV MW by 2030 (52 GW) will be more than double compared to 2020 (22 GW).

A similar trend was observed in France as there was an unprecedented rise in AAGR of 230% in 2008-2011 due to solar subsidies. France is the third country regarding PV installations in the EU-27 with a share of 1.7, 8 and 12% in the decades 2000, 2010 and 2020, respectively. The NECP sets a target of 42.3 GW in 2030 which is more than triple the installed capacity of 2020 (13.5 GW). Significant AAGRs were observed in most member states during 2008-2012.

The cumulative deployment power (GW) in the EU-27 during 2000-2020 and the forecasts according to the NECP of each country till 2030 are presented in Fig.6.1. As shown, the countries with largest share in 2030 are Germany (29%), Italy (15.4%), France (12.5%), Spain (11.6%), Netherlands (8%), Belgium (2.9%), Austria (2.9%), Portugal (2.7%), Denmark (2.3%) and Greece (2.3%). The total EU installed PV capacity grew from 0.17 GW in 2000 to 117.4 GW in 2019. Due to the EU commitment for energy decarbonisation in member countries and promote the New Green Deal, the total solar PV installations will almost triple in 2030 (338.3 GW).

As future PV panel installation data are retrieved from NECPs, they are country specific and all projections by 2030 are accurate. Thus, the PV panel waste is predicted accurately. However, uncontrollable factors may trigger a reconsideration of national binding targets. Nonetheless, even if the annual targets change, the long term 5 or 10 year targets may not change significantly, as the countries and the EU must guarantee a safe and predictable environment for investing in renewable energy since it involves high capital costs. Currently, world circumstances favour the promotion of renewable energy sources and the elimination of imported oil/gas dependence in EU.

Table 6 1. Comparison of this work with literature studies.

	Country	Reference	PV lifespan scenario	Waste results per PV technology	PV waste	Forecast	Power to mass conversion	Metals and materials
1	**Italy**	3	fixed 25 years	c-Si, a-Si, Cd-Te, CISG, Emerging-CPV	PV modules	2050	fixed per technology	14 (Al, Si, Cu, Sn, Pb, Zn, Ag, In, Ge, Se, Ga, Cd, glass, EVA)
2	**Spain**	4	Regular Loss, Early Loss	no data	PV modules	2050	exponential decrease**	5 (Al, Si, Cu, Ag, glass)
3	**Austria**	5	Regular Loss, Early Loss	no data	PV modules	2050	declining ***	no data
4	**Flanders Belgium**	6	fixed 20-35 years for different scenarios	no data	PV modules	2040	fixed per different PV panel architectures	8 (Cu, Ag, Si, Al, EVA, glass, PET, ABS)
5	**Mexico**	7	fixed 30 years	c-Si, a-Si, CdTe, CIGS, Other	BOS*	2045	fixed	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)
6	**India**	8	fixed 25 & 30 years, Regular Loss, Early Loss	Si-based, CdTe, CiGs	BOS*	2047	fixed per technology	21 (Ag, Au, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb,Ta, Se, Mg, Ga, In, Te, Si)
7	**Australia**	9	fixed 30 years, Regular Loss, Early Loss	c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other	PV modules	2060	fixed per technology (Si-based, CdTe, CiGs)	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)
8	**USA**	10	25-30 years (fixed)	no data	BOS*	2050	fixed per technology	21 (Ag, Au, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb,Ta, Se, Mg, Ga, In, Te, Si)
9	**OECD**	11	Regular Loss, Early Loss	c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other	PV modules	2058	fixed per technology (Si-based, CdTe, CiGs)	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)
10	**EU-27**	this study	Regular Loss, Early Loss and EU-WEEE	c-Si, a-Si, CdTe, CIGS, CPV, OPV, Advanced c-Si, Other	PV modules	2050	exponential decrease**	22 (Ag, Al, Cu, Ni, Fe, Ti, Sn, Zn, Cr, Mn, Mo, Cd, Pb, Se, Mg, Ga, In, Te, Si, Steel, EVA, Glass)

**BOS: modules, inverters, transformers, **adopted from [12].*

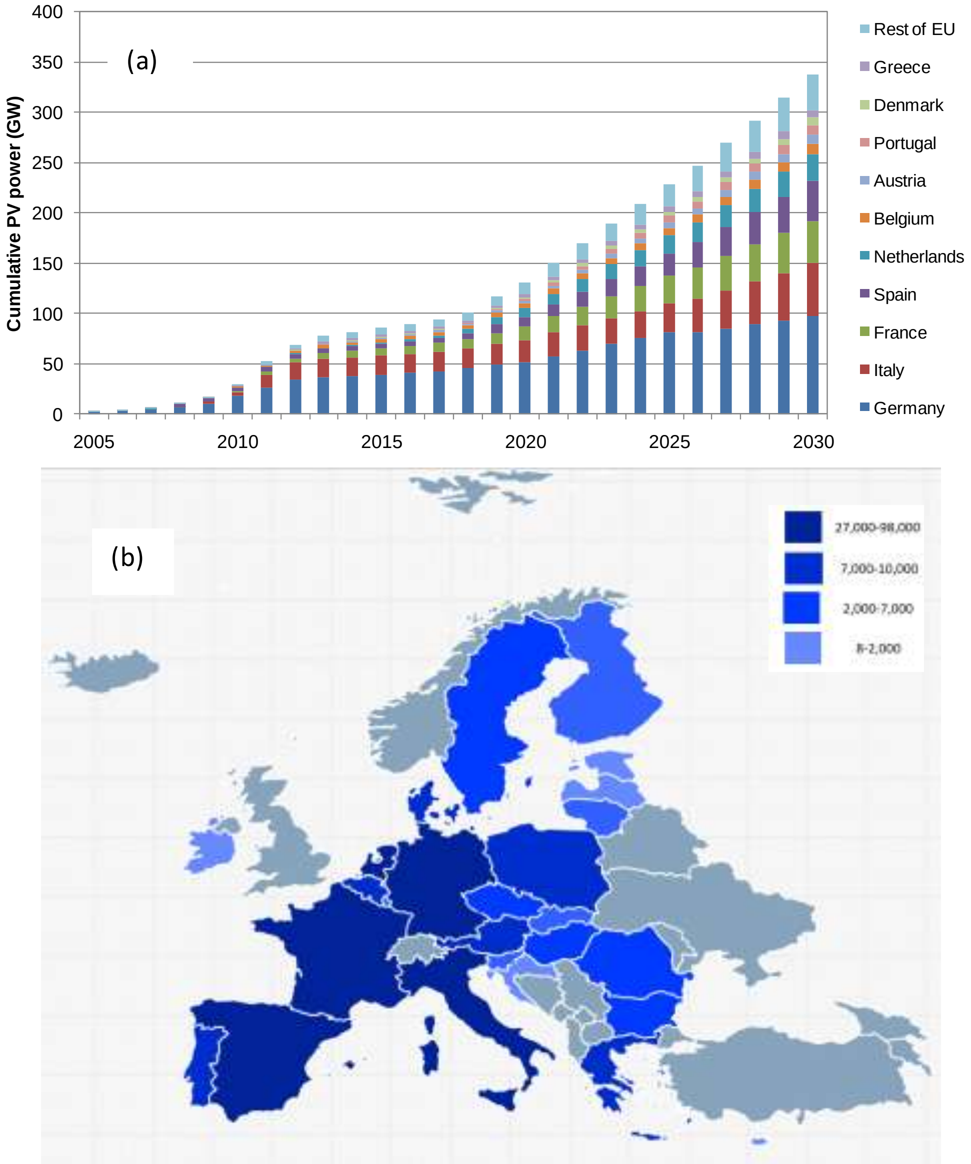


Figure 6. 1 Cumulative PV capacity (GW) in Europe in historical years and forecasts till 2030 according to the NECPs of member states and b) installed PV capacity (MW) in Europe by 2030.

1.2 PV waste regulations in EU

The EU specifically enforces photovoltaic waste regulations, including collection, recovery, and recycling targets, unlike most other countries that treat photovoltaic waste as general or industrial. According to the extended producer responsibility principle (EPR), producers are responsible for treating their End-of-Life products (EoL). EPR organizations require compliance with EU standards (CENELEC or WEEELABEX) by contract operators. EU prohibits mixed collection of photovoltaic panels with construction and demolition waste and requires separate treatment of silicon and non-silicon PV panels. Furthermore, it imposes certain de-pollution requirements concerning metals like cadmium, selenium, and lead. For example, glass fractions of silicon photovoltaic panels for recycling or recovery should not contain more than 1 mg/kg Cd, 1 mg/kg Se or 100 mg/kg Pb. The regulations also require that during thermal, chemical and mechanical treatment of PV panels, special care should be taken for the extraction of exhaust air and the removal of dust and related dangerous substances for the protection of human health and the environment [15, 19].

2. Methodology

2.1 Conversion of PV power into mass

The amounts of EoL PV panel are predicted based on historical installation data until 2019 [18], while PV capacity projections by 2030 are retrieved from the National Energy and Climate Plans (NECPs) of each of the 27 European countries.

Initially, the PV capacity (MW) is transformed into mass (tonnes). The annual conversion factor of PV mass per panel capacity (t/MW) was adopted, which was calculated from main PV producers' data for a period of 33 years [4, 12]. The factor considers the technological advancement that makes the panels more efficient and lighter over time, resulting in an exponential relationship of mass per panel capacity (t/MW) over time. This approach is more precise than using a constant conversion factor that is independent of time. The factor mass per panel capacity in year t is given by:

$$C(t) = mass\ per\ panel\ capacity\ (t) = A * e^{-t/B} \quad (1)$$

where A: 1.11*1,020 (tonnes/MW) and B: 48.24 years.

Then, the annual installed PV mass is:

$$mass\ PV_{\ annual}\ (t) = power\ PV_{annual}\ (t) * C(t) \qquad (2)$$

2.2 PV panel waste model

The failure mode of a PV panel is not constant over time, but rather follows a probability distribution. The Weibull distribution function has proven suitable in describing PV panels' failure when real life data were evaluated, successfully simulating early, mid and later-life failures [12, 15].

The lifespan distribution is depicted by the Weibull distribution:

$$f(t,a,\beta) = \frac{a}{\beta^a} t^{a-1} \exp(-\left[\frac{t}{\beta}\right]^a) \quad (3)$$

where α: the shape parameter that describes the gradual ageing of the product, and β: the scale parameter describing the characteristic life of the product that is, it defines the age when 63.2% of the products are expected to have failed.

The average lifetime (AL) is estimated by:

$$AL = \beta \times \Gamma(1 + 1/\alpha) \qquad (4)$$

where Γ: gamma function.

The annual PV panel waste outputs O_t (tonnes) is given as a function of PV inputs and the lifespan probability distribution:

$$O_t = \sum_{i=0}^{n} (S_{t-i} \times f_i) \quad \text{or} \quad O_t = \sum_{i=0}^{n} S_{t-i} \times (F_{i+1} - F_i) \qquad (5)$$

Where S_t: installed annual PV capacity in tonnes, f_i: Weibull lifespan probability distribution function, n: maximum possible lifespan and F_i: Weibull cumulative distribution function that gives the cumulative probability an installed PV panel fails at time t:

$$F(t) = 1 - e^{-(\frac{t}{\beta})^{\alpha}} \qquad (6)$$

The cumulative PV waste in year t is then calculated by:

$$O^{cum}{}_t = \sum_t^n (O_t) \quad (7)$$

Furthermore, the PV panels were classified into eight main categories (crystalline Silicon (c-Si), amorphous Silicon (a-Si), thin film Cadmium Telluride (CdTe), thin film Copper Indium Gallium Selenide (CIGS), Organic PV (OPV), concentrating solar (CPV), advanced c-Si and other), while the panel waste belonging into each category for EU-27 were determined considering the market share of these technologies [11, 12]. The eq. (8) was used to calculate the annual PV mass of each category:

$$mass\ PV_{\ annual\ ,j}\,(t) = power\ PV_{annual}\ (t) * C(t) * m_j \qquad (8)$$

where m_j: market share (%) of technology j (j: c-Si, a-Si, CdTe, CIGS, OPV, CPV, advanced c-Si and other). Then, the eq. (5) was used to calculate the corresponding amount of PV waste.

Last, the panel composition of these technologies [7, 9] was used to estimate the amounts of materials contained in the corresponding waste per PV technology.

2.3 Model assumptions

The Early and Regular Loss scenarios assume a characteristic PV lifetime of 30 years, while EU WEEE assumes 25 years. The shape factor (α) is smaller in EL scenario (2.4928), making the probability of failure steeper, while EU WEEE and regular have a shape factor of 3.50 and 5.3759, respectively, causing the corresponding curves to ascend smoothly (Fig. 6.2). The characteristic and the resulting average lifetime in EU WEEE, EL and RL scenario are 25, 30, 30 and 22.5, 26.6, 27.7, respectively.

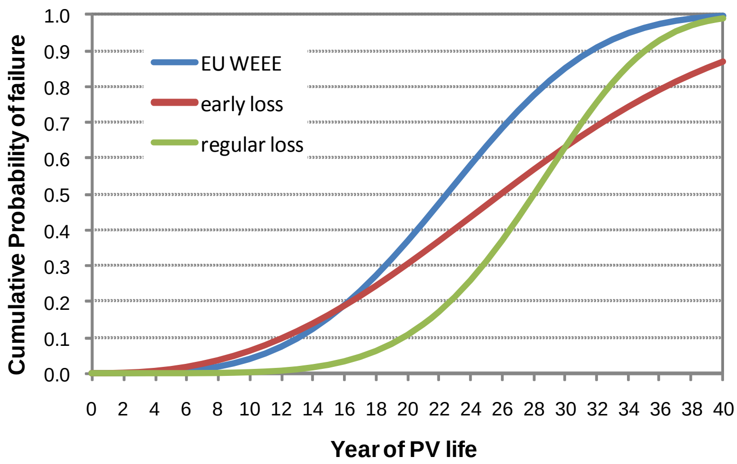


Figure 6. 2. Weibull cumulative probability of PV failure according to EU WEEE, Regular and Early loss scenarios.

The EU WEEE and EL curves intersect at about 16 years. EL scenario predicts higher probability of failure before 16 years, while the EU-WEEE predicts greater failure probability after 16 years of PV life. Comparing EL and RL curves, these intersect at 30 years, which is the characteristic time of these two scenarios, corresponding at 63.2% PV panels' failure at that time. After 30 years of PV life, the regular loss predicts quicker failure will occur. However, when failure occurs, it is not certain that the panels will reach the waste flow, as loss of power may be tolerated due to economic or other reasons. Thus, the EU WEEE scenario may predict actual losses, while the RL scenario predicts the amounts of waste that eventually enter the waste stream.

When PV panels are 33 years of age, regular loss predicts 81% of panels will have failed compared to 72% in the early loss scenario. Overall, comparing the three scenarios, the EU WEEE predicts quicker panels failure especially after 16 years of life. When panels are 40 years old, regular loss and EU WEEE scenario predict that practically all panels will have failed, while EL predicts 87% failure. For the EL scenario, practically all panels will fail at 58 years. The RL scenario predicts that 90% of installed panels will operate for about 20 years after installation, while EL and EU-WEEE scenario for 12 and 13 years, respectively.

3. Results and discussion

3.1 PV waste assessment in EU-27 countries

The assessment of PV waste in the EU is determined on the basis of the PV installations in the member countries until 2030 for the three scenarios (RL, EL and EU-WEEE). The annual and cumulative PV waste in the EU-27 according to these scenarios are shown in Fig. 6.3-6.5. These waste are calculated as explained in section 2.2 by the Eq. 5 (annual waste) and Eq. 7 (cumulative waste), incorporating the model assumptions detailed in Section 2.3. As can be seen, higher amounts of PV waste are predicted by 2040 according to EU WEEE scenario, followed by EL and RL scenarios. The most conservative predictions under RL scenario may reflect the reality from the waste collection point, while EU-WEEE scenario reflects losses occurring at all stages of transportation, installation and use, which may not all end in EoL management plants.

Germany has the largest accumulated quantities of PV waste in EU by 2040, ranging from 2.50 to 3.96 Mt in RL and EU-WEEE scenarios. It is followed by Italy with 1.05-1.70 Mt PV waste according to RL and EU-WEEE scenarios, followed by France (0.47-1.02 Mt), Spain (0.43-0.84 Mt), the Netherlands (0.20-0.57 Mt), Belgium (0.20-0.35 Mt), Greece (0.13-0.24 Mt), the Czech Republic (0.125-0.175 Mt), Austria (0.07-0.184 Mt), Denmark (0.06-0.164 Mt), Romania (0.060-0.13 Mt), Portugal (0.06-0.17 Mt) and then Bulgaria, Poland, Hungary, Slovakia, Sweden, Slovenia, Luxembourg, Finland, Cyprus, Malta, Lithuania, Croatia, Estonia, Ireland and Latvia.

The results are consistent with IRENA report [12] that roughly calculates 2.2 Mt for Germany by 2040 for RL scenario as compared to 2.5 Mt in the present study, 1 Mt for Italy (1.05 Mt in the present study), 0.4 Mt for France (0.465 Mt in the present study) and 0.04 Mt for Denmark (0.062 Mt in the present study). On the other hand, Paiano et al. [3], using the assumption of a fixed PV lifetime and the extrapolation of historical PV installation data, overestimated the generated waste in the case of Italy compared to the current study. They calculated 2.74 Mt PV panel waste by 2040 in Italy, which is 1.6-2.7 times higher the calculated amounts in our study (for RL – EL scenario). Santos et al. [4] using the RL and EL scenario estimated 0.35-0.42 Mt by 2040 for Spain as compared to 0.43-0.72 Mt in our study. This study adopted the official forecast of the Spanish government (2020) for PV deployment capacity and then used the assumption of 27.4 GW cumulative PV capacity by 2050. However, according to the Spanish NECP (used in our study) the cumulative installed power will be 39.2 GW just by 2030, explaining the underestimation by Santos et al. [4]. Dobra et al. [5] estimated 18,000-22,000 t in year 2050 (annual panel waste generation) in comparison to 20,101-31,081 t in our study. This study employed historical installation data till 2018 and assumed 400 MW of annually installed PV capacity till 2050. However, the targets set by the Austrian NECP anticipate an annual installation capacity of 600-900 MW from 2020-2050, revealing that the annual quantities are underestimated in [5].

Countries that installed high PV capacity in 2010-2013 because of subsidies (i.e., Belgium) will witness a huge amount of PV waste in 2035, while others that installed a great capacity after 2020 (i.e., the Netherlands) will witness great amounts of waste after 2040-45. For example, until 2017 Belgium had a higher installed capacity compared to the Netherlands, but after 2018 and especially after 2020 the Netherlands plans to install even larger PVs. So, the predicted waste (RL scenario) by 2040 for these two countries are comparable, while the installed capacity by 2030 in the Netherlands will be 27 GW compared to 9.8 GW for Belgium.

By 2030, Germany will generate more than half the amount of annual PV waste in EU according to the RL scenario. The German annual PV waste share is 61% in 2020, 50% in 2030 and then the share reduces to 39% in 2040 and 26% in 2050. Italy contributes to 14% in 2020, 19% in 2030, 18% in 2040 and 12% in 2050. France generates 3.8% in 2020, 6.7% in 2030, 10% in 2040 and 14% in 2050 of the annual PV waste in EU. The annual share of PV waste in Spain accounts for 10.7% in 2020, 8.3% in 2030, 7.3% in 2040 and 12% in 2050. The Netherlands contributes to less than 10% till 2049 (while in 2050, exactly 10%). The annual PV waste share in Belgium and Austria represents less than 3.7 and 3.2% from 2000 till 2050, respectively. Greece and the Czech Republic annually contribute to an average of 2.2 and 2% from 2020 till 2050. The annual PV waste share for the rest of the EU countries corresponds to 2.5-9.9% from 2020 to 2040 and then gradually increases to 17% in 2050. The total accumulated PV waste in the EU-27 will be 5.6 and 14.3 Mt in 2040 and 2050, respectively, according to RL scenario. As for EL and EU-WEEE scenarios, the total accumulated PV waste will amount to 8.1 (1.4 times the amount of RL scenario) and 10.0 (1.78 times the amount of RL) Mt in 2040 and 14.9 and 18.5 in 2050, respectively. Mahmoudi et al. [11] calculated 24.05-27.32 Mt PV panel waste for OECD countries according to RL and EL scenario by 2058.

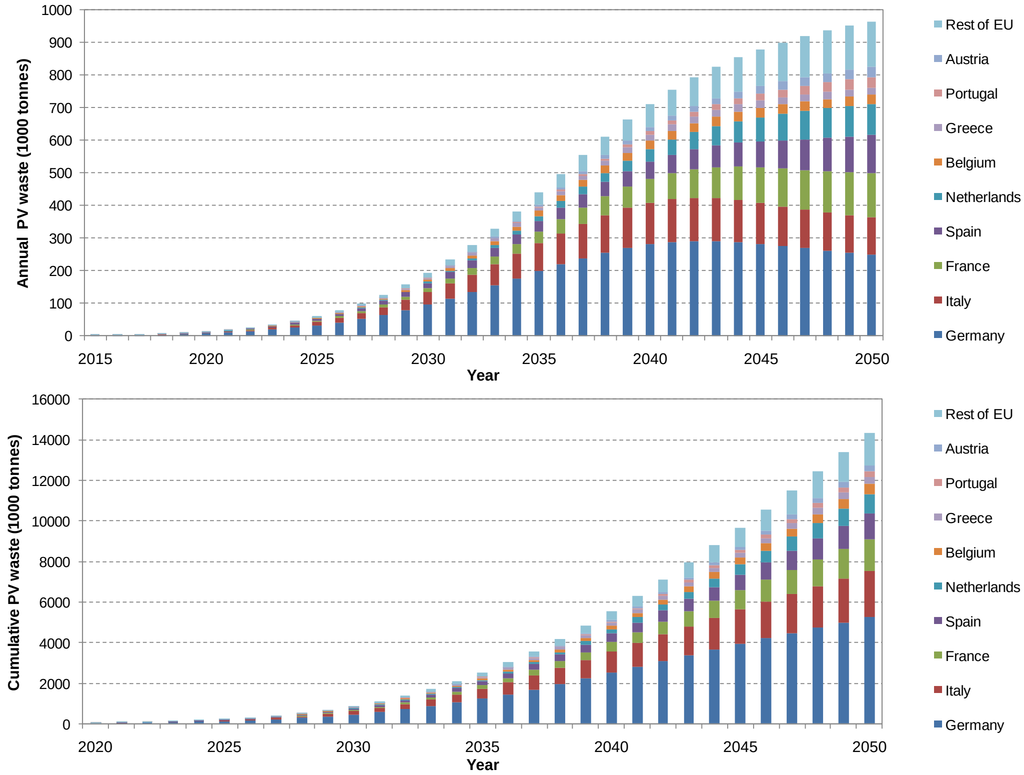


Figure 6. 3 Annual and cumulative PV waste in EU-27 countries according to Regular Loss scenario.

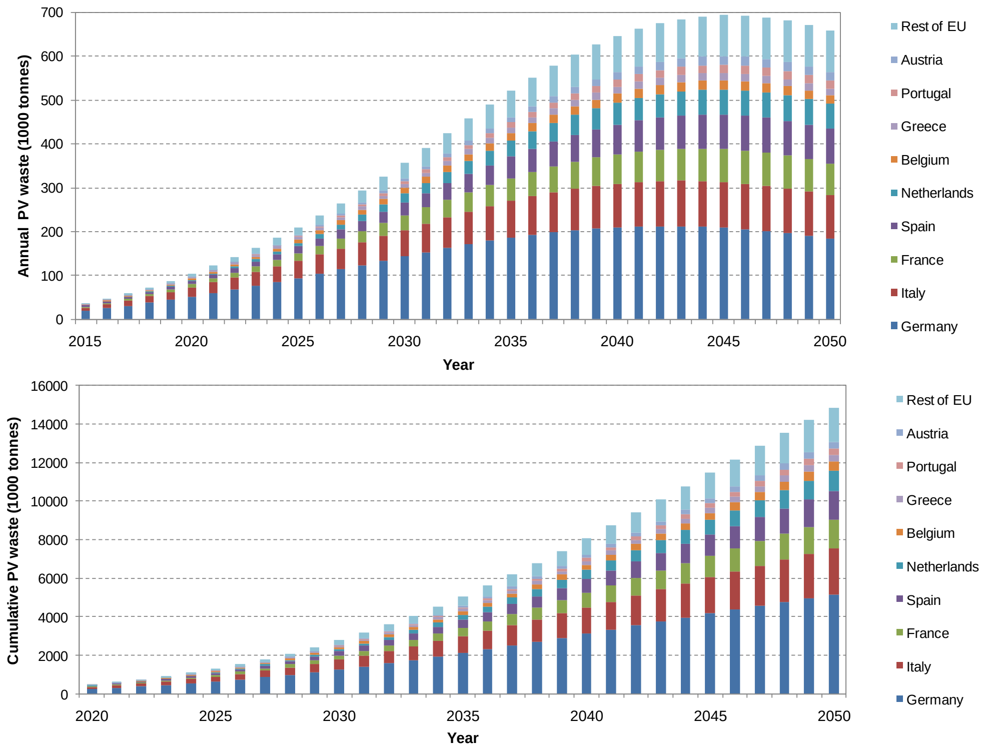


Figure 6. 4 Annual and cumulative PV waste in EU-27 countries according to Early Loss scenario.

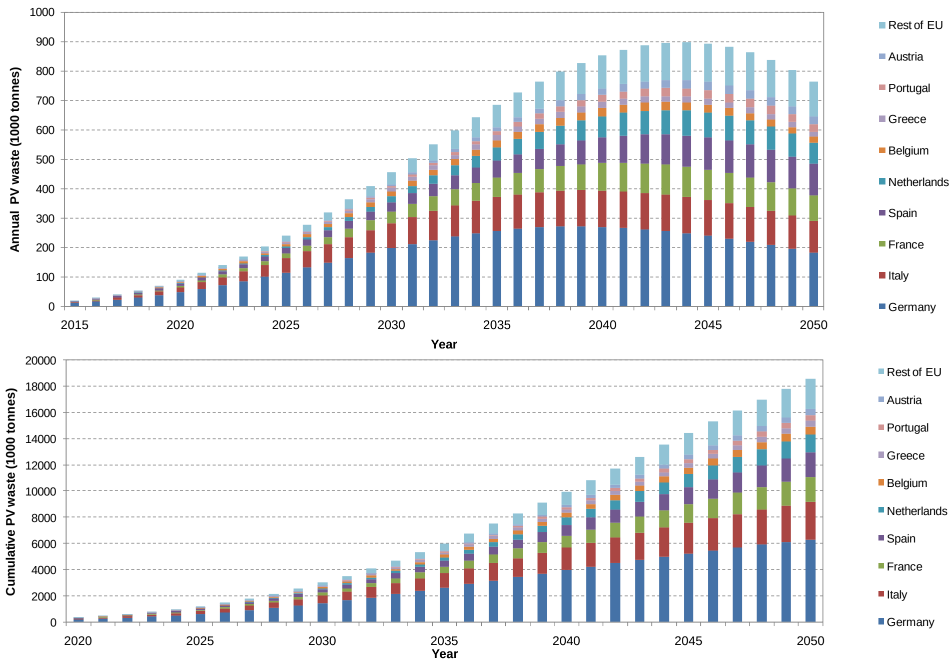


Figure 6. 5 Annual and cumulative PV waste in EU-27 countries according to EU-WEEE scenario.

As Germany is the leading country in PV installations, the evaluation is analyzed in more detail. The annual and cumulative PV panel waste generated in Germany is shown in Fig. 6.6. Annual and cumulative PV waste are calculated as explained in section 2.2 by the Eq. 5 and 7, respectively, incorporating the model assumptions detailed in Section 2.3. As can be seen, the annual waste curves decline after 2038-2042, as there were significant annual PV installations between 2010-2012 (average 7.84 GW) and then the annual installed capacity decreased to an average of 2.19 GW till 2020 and then 4.6 GW till 2030. According to the RL scenario, the accumulated waste in Germany will be 0.46, 2.50 and 5.24 Mt in 2030, 2040 and 2050, respectively. The EL scenario and EU-WEEE predict 1.25, 3.12, 5.15 and 1.43, 3.96, 6.27 Mt in 2030, 2040 and 2050, respectively. However, as the waste calculations are based on installed PV capacity till 2030, slight differences may occur according to the new installations (after 2030) and the scenario considered (affecting the results after 2043). In case new PV panels will be installed after 2030, these would gradually generate waste according to the Weibull distribution function that would add new waste from 2043 and onwards. The RL scenario predicts that small percentages of waste (about 1.5 %) are generated after 13 years have elapsed from installation time, while EL after 5 years and EU-WEEE after 7 years. The generated PV panel wastes then gradually increase according to each scenario. However, if 4.4 GW annual installations are assumed after 2030, there is a difference of about 1% in the calculated waste in 2044 (gradually increasing to 9% in 2050) for RL scenario. In EU-WEEE scenario, the impact will be 3% increase in 2040, 10% in 2044 and 44% in 2050, while in EL, 6.4%, 14.4% and 36%, respectively. In conclusion, the estimations presented for PV waste are accurate till 2045 for RL scenario, while for EL and EU-WEEE scenarios they are accurate till 2040 as deviations according to the new installed capacities may occur (after 2030).

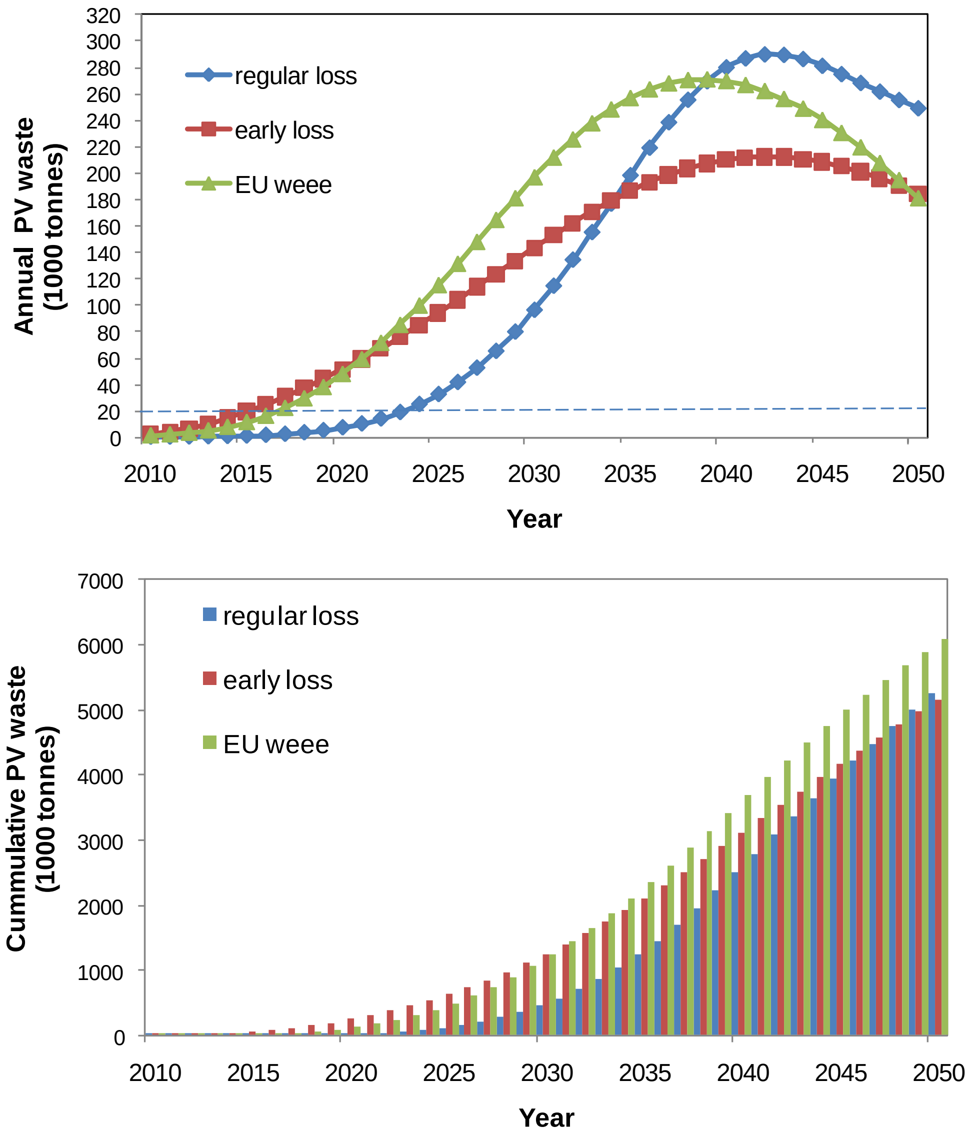


Figure 6.6 Annual and cumulative PV waste generation in Germany according to Regular Loss, Early Loss and EU-WEEE scenarios.

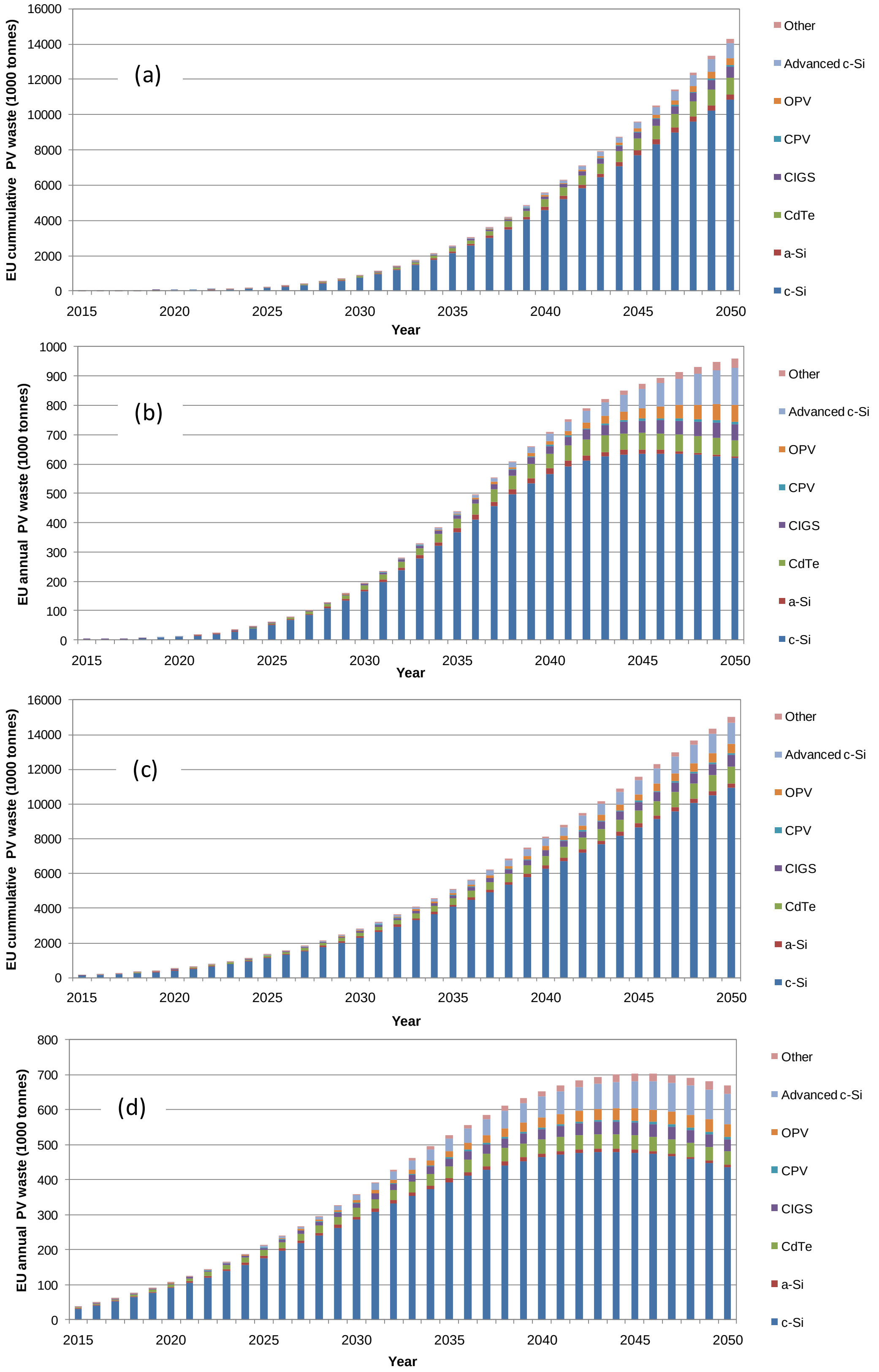
3.2 Waste assessment of different PV technologies in EU-27

Fig. 6.7 presents the annual and cumulative PV waste in EU for the various PV technologies (silicon-based: c-Si, a-Si, thin-film: CdTe, CIGS, advanced c-Si, OPV, CPV and other) for the RL, EL and EU-WEEE scenarios, respectively. The PV panel classification is important as different treatment, recycling scheme and regulations are applied for different PV panels. The oldest and most widely used PV technology is c-Si, which also dominates the panel waste.

In RL scenario, the c-Si panel waste has an average share of 90.9% of the total accumulated panel waste until 2019. Then, as new technologies enter the solar market, the total share of the c-Si waste reduces to 87.4% (2020-2030), 84.8% (2031-2040) and 79.5% (2041-2050). The accumulated c-Si panel waste by 2040 and 2050 will be 4.61 and 10.85 Mt, respectively. The second most important technology is CdTe PV panels as their waste amounts to only 1.5% before 2019, but then gradually increases to 6.6% (2020-2030), 7% (2031-2040) and 7.3% (2041-2050). The accrued CdTe panel waste by 2040 and 2050 will be 0.41 and 0.96 Mt, respectively. The average share of accumulated a-Si panel waste is 3.8% and 2.6% for 2020-2040 and 2041-2050, respectively, while the CIGS panel waste is 2.1% and 3.7%, respectively. The accumulated a-Si panel waste by 2040 and 2050 will be 0.18 and 0.30 Mt, while for CIGS panel waste 0.17 and 0.60 Mt, respectively. The accumulated Advanced c-Si panel waste contributes on average to 0.9% (2020-2040) and then to 3.9% (2041-2050). The Advanced c-Si panel waste by 2040 and 2050 will be 0.10 and 0.86 Mt, respectively. The OPV, CPV and other PV technology wastes contribute to a lesser extent. After 2045, the annual Advanced c-Si panel waste share becomes significant, with an average share of 11% (2045-2050).

In EL and EU-WEEE scenarios, the average share of accumulated a-Si panel waste is significantly higher (2000-2050) followed by CdTe, Advanced c-Si, CIGS, a-Si, OPV, other and CPV. For EL and EU-WEEE scenarios, the average share of accumulated a-Si waste panels is significantly higher (2000-2050) followed by CdTe, Advanced c-Si, CIGS, a-Si, OPV, other and CPV. In EL scenario, the accumulated c-Si panel waste by 2040 and 2050 will be 57.8 and 144.3 Mt, respectively, while CdTe waste will be 5.1 and 12.9 Mt, respectively. Also, the total Advanced c-Si panel waste will be 2.4 and 11 Mt by 2040 and 2050, accordingly, and the CIGS waste 2.4 and 7.5 Mt, respectively. The annual Advanced c-Si panel waste share in 2040-2050 is important, 11.3 and 13.3% in EL and EU WEEE scenario, respectively. In EU WEEE scenario, the accumulated c-Si panel waste by 2040 and 2050 will be 66.7 and 177.9 Mt, respectively, while CdTe waste will be 5.8 and 15.7 Mt, accordingly. Furthermore, in EL scenario the total Advanced c-Si panel waste will be 2.1 and 12.6

Mt by 2040 and 2050, respectively.Similar results on the share of PV technologies in waste for EL and RL scenario are reported by Mahmoudi et al. [11]. As these PV panels have different structure and materials, the quantitative analysis of the respective shares is important for the PV-recyclers, as different approaches are required to recover the valuable components.



(a)
EU cummulative PV waste (1000 tonnes)
0
2000
4000
6000
8000
10000
12000
14000
16000
2015
2020
2025
2030
2035
2040
2045
2050
Year
Other
Advanced c-Si
OPV
CPV
CIGS
CdTe
a-Si
c-Si
(b)
EU annual PV waste (1000 tonnes)
0
100
200
300
400
500
600
700
800
900
1000
(c)
(d)

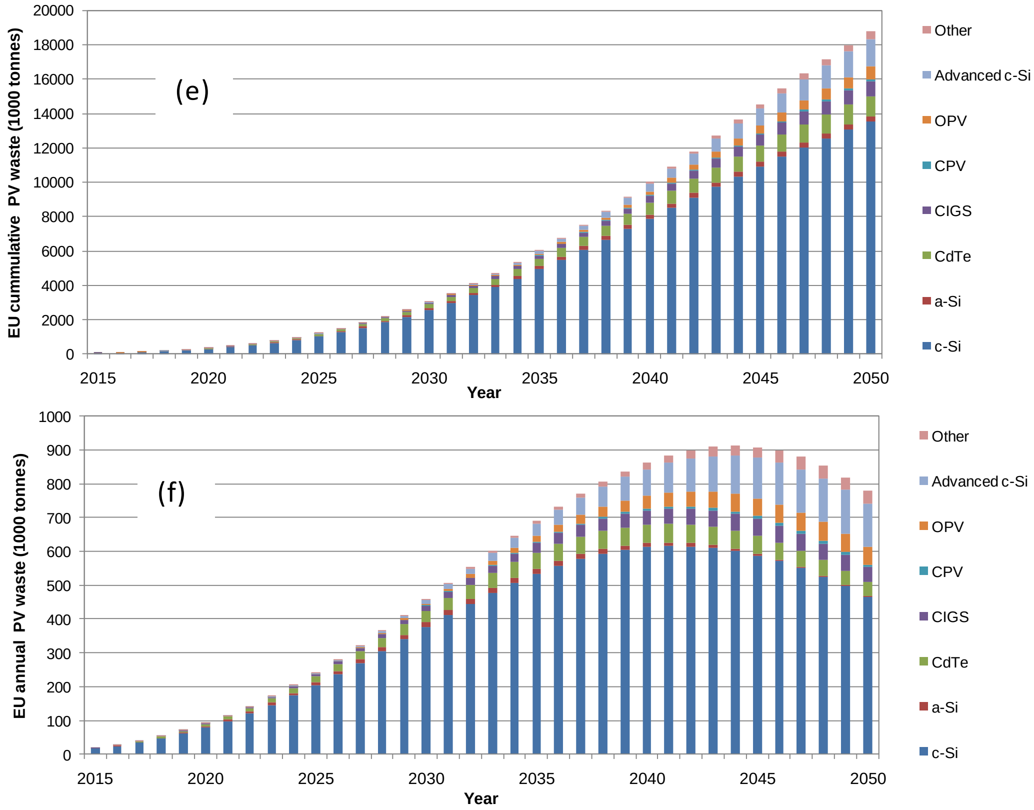


Figure 6.7 Cumulative and annual PV waste in EU-27 countries for the various PV technologies (silicon-based: c-Si, a-Si, thin-film: CdTe, CIGS, advanced c-Si, OPV, CPV and other) according to (a, b) Regular Loss scenario, (c, d) Early loss scenario, and (e, f) EU-WEEE scenario.

3.3 Material composition of different PV technologies waste in EU-27 and financial revenue

Based on the material composition of the four main PV technologies, c-Si, a-Si, CdTe, and CIGS [9, 10], the estimated materials in the PV panel waste in the EU are calculated. The accumulated amounts by 2040 and 2050 are presented in Table 2 according to RL scenario. As can be seen, the accumulated amount of glass will be 3.5 Mt by 2040, while the amount will be more than double by 2050. Aluminum and steel will approximate 0.85 and 0.51 Mt by 2040, respectively, while the quantities will be 2.3 times more by 2050. The Ethylene-vinyl acetate (EVA) and Cu will approach 0.35 and 0.05 Mt by 2040, accordingly, and the quantities will be more than double by 2050. Moreover, significant quantities 0.03 and 0.06 Mt of Mg will be accumulated by 2040 and 2050, respectively. Considerable amounts of Ag, Si, Ga, In, Mn, and Ti, which are included in the EU Critical Raw Materials 2020 list (CRM) [20] are accrued. Furthermore, mainly due to the thin-film panels, toxic Cd will be 0.78 and 2.19 thousand t by 2040 and 2050, respectively. The treatment of PV panel waste with hazardous substances requires special procedures for hazardous materials to ensure that human health and the environment are protected.

In the EU-WEEE scenario, more waste (and materials) is generated compared to RL and EL scenarios. For instance, based on EU-WEEE scenario, the accumulated amounts by 2040 of Ag, Al, Cu, Cr, Te, Pb, Mg, Si, Cd will be on average 70% higher, In 101%, and Sn, Zn, Mo, Se, Ga about 127% compared to RL scenario.

As reported in Table 2 (RL scenario), the accumulated materials by 2040 with the largest mass share are glass (65.85%), Al (15.8%), steel (9.6%), EVA (6.6%), Cu (0.9%) and Mg (0.5%). The calculated mass shares fairly agree with the corresponding shares calculated for OECD countries [11]. However, when the financial revenue is calculated, taking into consideration the recycling yield of each material and the commodity price [8], the precious metals with a small mass share significantly contribute to the total revenue. Indeed, Ag with a mass share of 0.05% adds 18% to the revenue share, while Al with a mass share 15.9% also adds 18% to the revenue share. This indicates that recycling treatment (and regulations), based on a mass basis, may lose precious and economically important metals present in small quantities. Recycling requirements could be revised to include metals with the highest economic share. Also, the economic shares of Te, Si, In, Ga become important although their mass share is insignificant. Metals like Te, In, and Ga are considered CRMs due to their high economic significance and great supply risk [21] especially for EU economy [22]. Although they have a very small mass share in PV panels, they are important in PV solar industry because of their properties. The total gross value of PV panel waste will be 9.3 billion USD by 2040, while it will reach 22 billion USD by 2050. Mahmoudi et al. [11] estimated 17 and 36.66 billion USD by 2040 and 2058, respectively, in OECD countries for RL scenario. According to the EU-WEEE scenario, the total gross value of PV panel waste will be 15.96-27.36 billion USD by 2040-2050.

Table 6 2 Cumulative material amounts (tonnes) in 2040 and 2050 according to Regular Loss scenario in EU-27 for c-Si, a-Si, CdTe and CIGS panel waste .

Metal/ material	c-Si		a-Si		CdTe		CIGS		Total (tonnes)		Mass share % (by 2040)	Revenue USD (by 2040)	Share economic value % (by 2040)	Recycling yield
	2040	2050	2040	2050	2040	2050	2040	2050	2040	2050				
Glass	3014100	7097188	810	1375	371811	879640	142536	508497	3529257	8486701	65,85	2648707559	28,36	95
Al	760438	1790575	73445	124586	367	869	14337	51148	848588	1967178	15,83	1692084142	18,11	99,7
Ag	2659	6262							2659	6262	0,05	1644600384	17,61	95
Steel	438289	1032022	70267	119195	4876	11536			513432	1162754	9,58	1483768262	15,88	95
Si	36455	85839	4,54	7,70	1223	2893			37683	88740	0,70	1129284219	12,09	99,9
EVA	299567	705378	28072	47618	14669	34705	8555	30522	350863	818223	6,55	312267974	3,34	100
Cu	33689,7	79328	1587,2	2692	12231	28937	475	1693	47983	112650	0,90	309967725	3,32	100
Te			11,3	19,2	488	1154			499	1173	0,01	42186655	0,45	95
In			20,5	34,7			47,5	169	68	204	0,001	28125548	0,30	90
Ga							94,9	339	95	339	0,002	25199243	0,27	90
Mg	23965	56430	2313	3923			446	1592	26724	61945	0,50	17726226	0,19	33
Se							94,9	339	95	339	0,002	4248944	0,05	89
Cd			9,1	15,40	488	1154	286	1019	782	2188	0,01	817626	0,009	95
Sn	2,7	6,4			0,01	0,01	94,9	339	98	345	0,002	604143	0,006	32
Pb	215	507			17,1	40,6			232	547	0,004	517546	0,006	96
Ni	48,9	115			17,1	40,6			84	143	0,002	497248	0,005	41
Mo							94,9	339	95	339	0,002	304099	0,003	18
Cr			1	1,70	73,5	174			75	176	0,001	147603	0,002	20
Zn	0,40	0,80	0,7	1,10	0,001	0,002	94,9	339	96	341	0,002	67602	0,001	27
Fe			1,3	2,20					1,3	2,2	0,00002	710	0,00001	90
Ti	0,24	0,57			0,001	0,001			0,2	0,6	0,00001	136	0,000001	52
Mn			1,7	2,80					1,7	2,8	0,00003	3	0,00000003	37

3.4 Sensitivity analysis

The estimated amount of materials and metals embedded in panel waste is influenced by factors such as the recycling yields and the future composition of PV panels. The progress in recycling technology can improve the recycling yields, in turn affecting the amounts of materials recovered. The amount of recovered materials considering low, average, and maximum recycling yields, as reported in [7], are depicted in Fig. 6.8. For example, the cumulative recovered amount of Ga by 2040 ranges between 31 and 94 tonnes according to low (33%) and maximum (99%) recycling yield. The variations in recovered materials were estimated taking into account the low, average and maximum levels for the ‘recycling yield’ parameter for various metals as follows: 22, 54.8, 100% for Ag, 14, 70, 100% for Al, 30, 77.3, 100% for Cu, 14, 75.2, 100% for Cd, 63, 79.5, 96% for Pb, 38, 69.5, 90% for Se, 33, 63, 99% for Ga, 15, 64.7, 99% for In, 80, 89.6, 100% for Te and 74, 86.7, 100% for Si.

Furthermore, considering the evolution of materials per PV technology as reported by IRENA [12], it is evident that the c-Si technology panels will present the maximum variation of materials between 2014 and 2030. However, the PV panels deployed in 2030 will start contributing to waste after 2040, thus the variation in the materials will not be evident before 2040 and the calculated waste till 2040 will not be affected. The most important changes are a 40% reduction in Si content, 1% decrease in Al, 0.01% decrease in all other metals, and 0.01% or slightly higher decrease in Ag. On the other hand, there will be a 4% increase in the glass content. The amounts of affected materials (after 2040 to 2050) are depicted in Fig. 6.9.

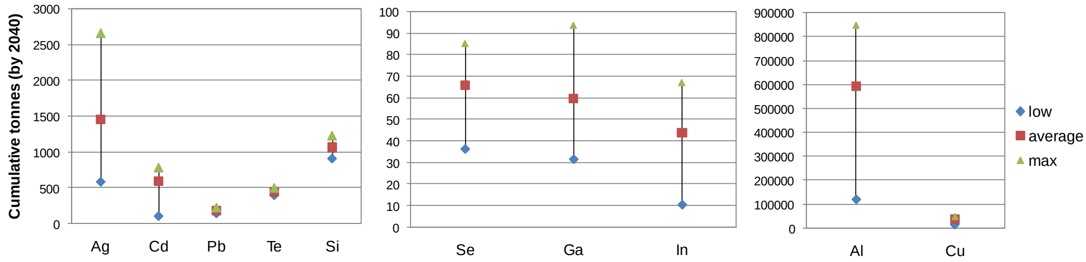


Figure 6. 8. Variation in metal recovery according to recycling yields (low, average, maximum).

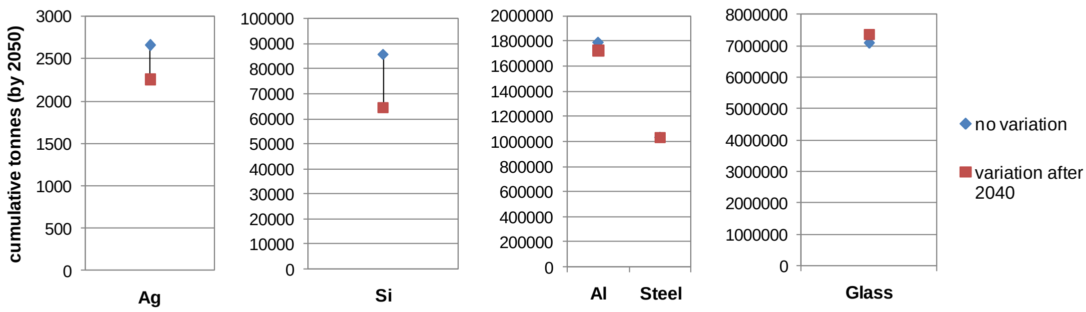


Figure 6. 9. Comparison in accumulated metal amounts by 2050 due to changes in the panel composition after 2030 as compared to fixed composition.

3.5 Timescale for operation of PV recycling industry in EU-27

Currently, the solar PV industry in the EU is expanding aiming to capture the additional solar energy demand. A recycling plant has started operation in France (Veolia) in 2018 as significant volumes of end-of-life PV panels are anticipated in the next years [5], whereas unofficial data show that PV recycling takes place in 6 EU countries. Candidate countries for the establishment of a photovoltaic recycling plant are indicated based on the annual PV waste generated in each country until 2030, then corporations can be set up to serve neighbouring countries. Germany is the first country to accumulate a large amount of PV waste. For a PV recycling plant to be economically viable, the minimum annual amount of waste is estimated at 20 kt [17]. This will be satisfied in 2024 according to Regular Loss scenario, while the Early loss and EU WEEE scenarios predict that the required PV waste amount has already been generated in 2016 and 2018, respectively (Fig. 6.6). By the implementation of efficient collection and recycling schemes, the recovered materials can be used in the manufacture of new PVs within EU closing the material loop and therefore strengthening economy.

Some countries accumulate a great amount of PV waste but they are below the limit of 20 kt annually (i.e., the Czech Republic), while others reach the annual limit although their accumulated waste by 2040 is less than the previous countries (i.e., Austria). The year (and amount) in which the waste is generated is related to the year (and size) of the PVs installation. Thus, corporations between countries are needed to sustain viable recycling.

Table 3 summarizes the cumulative waste data and the year that satisfies the annual limit of 20 kt for all countries. Based on the RL scenario, which provides conservative predictions on the waste generation compared to the other two scenarios, the year to satisfy the annual limit of 20 kt PV waste is 2024 for Germany. It is followed by Italy (2028), France (2032), Spain (2032), the Netherlands (2037), Belgium (2037), Greece (2042), Austria (2044), Denmark (2045), Portugal (2045), Poland (2046), Hungary (2049) and then (after 2050) the Czech Republic, Romania, Bulgaria, Slovakia, Sweden, Slovenia, Luxembourg, Finland, Cyprus, Malta, Lithuania, Croatia, Estonia, Ireland, and Latvia. The EU-WEEE scenario also predicts almost the same order but a little earlier. The countries that reach the limit before 2050 (in RL scenario) are predicted 5-7 years earlier in the EU-WEEE scenario, except for Hungary (4 years earlier) and Greece (3 years earlier). Overall, the mature year to operate a PV recycling plant according to EL and EU WEEE scenarios is 0-8 years earlier than the RL scenario. The results of this study partially agree with those reported for the OECD countries [11]. Specifically, the operation year is the same for France and Spain (2032), very close for Italy and Belgium and the Netherlands (2027, 2036 and 2040, respectively in [11]), but very different for Germany (2035 in [11] as opposed to 2024 in this study). Significant differences also exist in [11] for Austria

(2060 while 2044 in this study), Denmark (2060 while 2045 in this study), Greece (2060 while 2042 in this study), Hungary (2060 while 2049 in this study), Portugal (2060 while 2045 in this study) and Poland (2060 while 2046 in this study). These differences are due to the different future deployment capacity considered, which in turn yields different amount of waste (for the same scenario). The current study accurately considers the future PV deployment capacity from the NECPs as opposed to projections on historical data used in other studies.

Fig. 6.10 presents the map with the leading countries that can start a viable recycling industry by 2024-2032 (Germany, Italy, France and Spain). Eight more countries will be able to successfully install PV recycling plants by 2037-2049 (the Netherlands, Belgium, Greece, Austria, Denmark, Portugal, Poland, and Hungary). The remaining fifteen countries will not be able to operate a photovoltaic recycling plant until 2050; therefore, partnerships with neighbouring countries are necessary. The logistics about transportation and storage of PV waste must be incorporated into the recycling methodology to determine the exact location of the recycling plant in each country.

Table 6 3 Installed MW, accumulated PV waste and year of 20kt annual PV waste in EU countries according to RL, EL and EU-WEEE scenarios.

		installed MW		Regular Loss Scenario Sum waste (tonnes)				Early Loss Scenario Sum waste (tonnes)				EU Weee Scenario Sum waste (tonnes)			
		2020	**2030**	**2030**	**2035**	**2040**	**waste 20kt**	**2030**	**2035**	**2040**	**waste 20kt**	**2030**	**2035**	**2040**	**waste 20kt**
1	**Germany**	51,568	97,924	456,837	1,236,249	2,499,618	2024	1,253,081	2,105,070	3,117,935	2016	1,437,156	2,617,928	3,961,588	2017
2	**Italy**	22,065	52,000	156,949	480,493	1,047,889	2028	511,587	882,983	1,350,576	2020	580,185	1,098,897	1,704,056	2021
3	**France**	13,580	42,280	50,784	178,464	465,240	2032	236,578	488,527	847,178	2027	230,692	539,537	1,021,261	2026
4	**Spain**	9,071	39,181	77,310	206,488	429,020	2032	217,288	422,659	724,811	2028	236,067	470,771	843,399	2026
5	**Netherlands**	9,000	27,000	13,781	59,594	198,006	2037	130,722	258,105	483,972	2030	86,050	258,041	569,812	2030
6	**Belgium**	5,096	9,823	27,628	87,005	197,935	2037	98,601	176,664	274,153	2039	106,904	214,135	347,572	2032
7	**Greece**	3,000	7,700	16,502	55,347	132,353	2042	65,531	120,889	192,663	2050	69,741	144,688	241,937	2039
8	**Czech Republic**	2,082	3,975	22,681	63,618	125,156	2050	60,590	96,142	136,276	2050	73,201	125,474	175,285	2050
9	**Austria**	2,294	9,695	7,078	25,168	71,569	2044	37,191	85,346	159,755	2046	33,395	87,560	184,777	2039
10	**Denmark**	1,580	7,842	4,853	20,193	61,939	2045	32,381	74,869	140,610	2050	28,037	76,953	163,906	2040
11	**Romania**	1,698	5,054	5,516	21,632	59,656	2050	30,184	61,675	105,607	2050	29,148	69,338	129,512	2050
12	**Portugal**	2,000	9,000	4,482	17,672	56,386	2045	30,247	76,716	149,132	2050	24,051	73,486	170,044	2039
13	**Bulgaria**	1,042	3,216	6,693	22,190	51,984	2050	25,400	46,298	74,023	2050	27,690	55,926	92,123	2050
14	**Poland**	2,285	7,270	1,445	9,754	40,251	2046	22,315	59,949	118,743	2050	15,363	55,949	136,366	2041
15	**Hungary**	1,170	6,454	1,339	7,885	31,053	2049	16,766	46,235	94,372	2050	11,947	42,491	105,560	2045
16	**Slovakia**	630	1,200	4,375	13,742	30,446	2050	14,926	25,604	38,397	2050	16,750	32,077	49,370	2050
17	**Sweden**	798	2,231	1,131	4,762	15,738	2050	8,500	20,285	38,202	2050	6,874	20,359	44,819	2050
18	**Slovenia**	400	1,650	1,469	5,343	14,374	2050	7,257	15,715	28,489	2050	6,957	16,847	33,399	2050
19	**Luxembourg**	230	950	1,672	4,174	9,101	2050	4,810	9,824	17,290	2050	4,892	10,515	20,071	2050
20	**Finland**	310	1,200	392	1,809	6,586	2050	3,611	9,507	18,851	2050	2,656	8,933	21,447	2050
21	**Cyprus**	360	804	505	2,120	6,571	2050	3,480	7,973	14,636	2050	2,968	8,241	17,379	2050
22	**Malta**	164	266	372	1,516	4,270	2050	2,205	4,340	7,081	2050	2,094	5,011	9,054	2050
23	**Lithuania**	72	864	333	1,338	4,145	2050	2,061	5,551	11,528	2050	1,769	5,183	12,436	2050
24	**Croatia**	96	768	216	993	3,331	2050	1,711	4,496	9,368	2050	1,401	4,266	10,207	2050
25	**Estonia**	100	415	55	401	1,793	2050	995	2,930	6,115	2050	632	2,574	6,772	2050
26	**Ireland**	40	431	51	312	1,333	2050	705	2,208	4,942	2050	471	1,876	5,189	2050
27	**Latvia**	4	8	3	17	63	2050	34	83	153	2050	26	83	184	2050

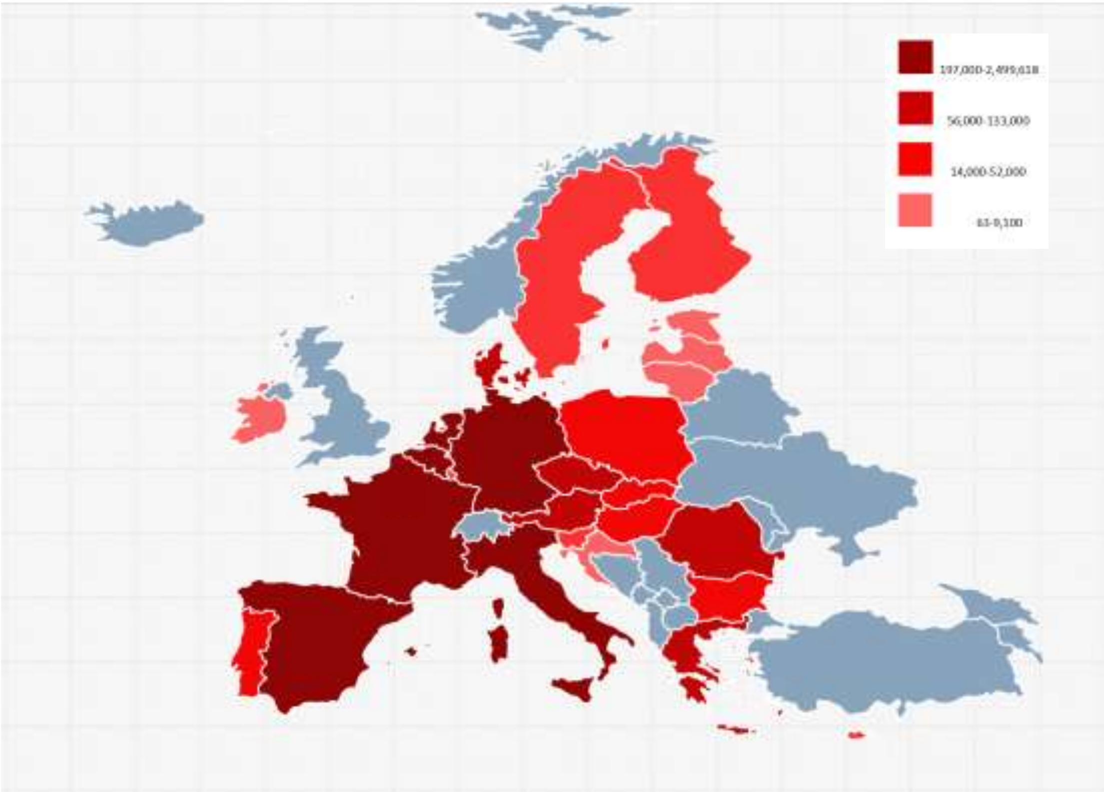


Figure 6. 10. Accumulated PV waste (tonnes) in Europe by 2040 according to the Regular Loss Scenario.

4. Conclusions

PV waste assessments in all 27 EU countries are presented according to three scenarios, the Early and Regular Loss scenarios proposed by IRENA and the scenario proposed by the EU WEEE Directive. By 2050, 14.3-18.5 Mt PV waste will be generated in EU-27 while the profit of PV recovered materials will be 22-27.4 billion USD. C-Si technology will dominate the PV waste (10.85 Mt), followed by CdTe (0.96 Mt). The EU must be prepared to deal with future amounts of PV waste. On this basis, a scheme for the EU-27 countries to launch a viable PV recycling business is presented according to the timetable estimated in this study. A core of 4 countries can start a viable recycling industry by 2024-2032 (Germany, Italy, France and Spain). Eight more countries will be able to successfully operate individual PV recycling plants by 2037-2049 (the Netherlands, Belgium, Greece, Austria, Denmark, Portugal, Poland and Hungary). The remaining fifteen countries will not be able to operate a PV recycling plant until 2050, so PV waste recycling partnerships will need to be established in these countries, taking into account the proximity of existing recycling facilities in neighbouring countries. The recycled materials can be used in the construction of new PVs within EU, closing the material loop and therefore strengthening the economy. This will ensure a sustainable and reliable source of raw materials in the EU, which will contribute to covering the apparent demand for minerals and metals associated with low-carbon technologies.

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CHAPTER 7

Hydrothermal silver and aluminum leaching from mono- and poly-crystalline photovoltaic waste panels

The aim of this study was the hydrothermal leaching of silver and aluminum from waste monocrystalline and polycrystalline silicon photovoltaic (PV) panels using mild HNO3 solutions, from both cells and ribbons. Prior to leaching, pretreatment was applied to remove the fluoropolymer backsheet and then thermally degrade the ethyl acetate (EVA) polymer. Several hydrothermal parameters were investigated like liquid to solid ratio (L/S), HNO3 concentration (N), time (t) and temperature (T). Based on preliminary tests, the HNO3 concentration was set in the range of 1-2 N to reduce hazardous waste effluent. Response Surface Methodology (RSM) was applied to optimize the hydrothermal leaching parameters. It was found that processing time was the most important factor for Ag leaching, followed by HNO3 concentration and S/L ratio, while the processing temperature (100-140 oC) was not a statistically significant factor. Al leaching was efficient under most hydrothermal conditions. For comparison, leaching was also applied at lower temperatures 25-45 oC for prolonged times, however lower efficiencies were observed. Under the optimal hydrothermal conditions, Ag can be completely leached, while Al dissolution was favoured at hydrothermal conditions compared to lower temperature leaching. Silver leaching efficiency was 100% under hydrothermal conditions; however, under conventional lower temperature conditions, it was 80.7–85.3% for m-Si and p-Si waste panels. Under conventional lower temperature conditions, Al leaching efficiency was 56.6–61.3% for p-Si and m-Si waste panels.

1. Overview

Due to the unprecedented worldwide increase in photovoltaic (PV) installations and considering the typical lifetime of PV panels of 25 years, there is a growing concern about emerging PV waste in the near future. It is estimated that 60-78 million tonnes (Mt) of PV waste will be generated worldwide by 2050, while in the EU-27 alone, PV waste will be 14.3-18.5 Mt. First-generation crystalline silicon (c-Si) modules have had an 80-90% market share over the last 40 years and will thus dominate the PV waste stream. Of these c-Si panels, 51% of the PV market share is covered by polycrystalline Si (p-Si) and 41% by monocrystalline Si (m-Si). In the EU-27, the c-Si panel waste was 90% till 2019 and will be 80-87% in 2020-2050 (Weckend et al., 2016; Kastanaki and Giannis, 2022). Thus, research on PV panel recycling should primarily focus on c-Si panels, due to their large volume compared to other panel technologies.

Crystalline silicon PV modules consist of various types of materials such as glass, polymer layers of ethyl vinyl acetate (EVA), crystalline silicon cells, copper and silver metal contacts, a polymer backsheet and an aluminum frame. This layered design hinders the reuse of modules and restricts the recycling options. The mass of a typical PV module consists of more than 90% glass, polymers and aluminium frames (Dias et al., 2017; de Oliveira et al., 2020). However, there are also components in smaller proportions that require more attention such as silver, tin, lead and other metals. Of these finite materials, silver and tin will be depleted in the next 50 years, while other metals will last for another 50-500 years (Hunt et al., 2015; Farrell et al., 2020).

Although Ag is present in tiny amounts in PV panels, it contributes significantly to the revenue from recycled panel materials, due to its high economic value (Kastanaki and Giannis, 2022). According to Tao et al. (2020), Ag represented 35.2% of the economic value obtained from recovering materials from a Si module in 2019. In the EU-27, the economic value of Ag will represent 17.6% of the total revenue from recycled materials by 2040, although its mass share in the accumulated PV waste (considering all PV technologies waste) will be only 0.05% (Kastanaki and Giannis, 2022). The average quantity of silver in a PV panel is estimated around 10 g per square meter. According to US resources, the silver content should surpass 700 g/t when mined to be commercially extracted as a primary product. As a result, Ag recovery from PV panel waste is becoming increasingly important (Huang et al., 2017; Kuczyńska-Łażewskaa et al., 2018; Savvilotidou et al., 2020).

In order to maximize Ag leaching efficiency, the operating parameters of temperature, time, L/S ratio and HNO3 concentration should be optimized. This can be accomplished with appropriate statistical techniques, such as the response surface methodology (RSM) with a Box–Behnken design (BBD) (Tanong et al., 2017). This design investigates the effects of the parameters on silver leaching by simultaneously differentiating all parameters under consideration, instead of differentiating one parameter at a time. Thus, possible interactions between factors can be identified. The Box–Behnken design effectively avoids the realization of the complete experimental design, which would otherwise make the process more demanding in terms of time and resources, without losing any information on the effects of the parameters on the process. In this design, three levels of each factor are defined: low, middle and high (Tanong et al., 2017; Behera et al., 2018. Among the response surface designs, BBD is slightly more efficient compared to the central composite design, and is much more efficient than the three-level full factorial design (Ferreira et al., 2017). This approach (RDM-BBD) is currently missing from the literature to optimize Ag leaching from waste PV panels.

The traditional acid-leaching process involves leaching metals from various matrices in non-pressurized hot water using mainly inorganic acids. In this process, the reaction temperature should be low in order to avoid water evaporation (<80–90 °C). The low leaching temperature requires a long reaction time, as well as a high acid concentration, to ensure complete leaching; however, this increases costs and environmental load (Zheng et al., 2018; Xing et al., 2019; Zhang et al., 2021). Nonetheless, conventional acid leaching has been extensively applied to leach metals from waste PV panels (Dias et al., 2016; Huang et al., 2016).

Initially, research on waste PV panel recycling has focused on Si, glass and aluminum recovery (Klugmann-Radziemska et al, 2010; Shin et al., 2017), but more recent studies also aim at Ag leaching and recovery (Dias et al., 2016; Kuczyńska-Łażewska et al., 2018; de Oliveira et al., 2020). Some of these studies also consider leaching of Al along with Ag from PV cells. Because of the high value of Ag, the main focus is mainly on this metal. Also the largest amount of Al in PV panels is found in the Al frames which are easily recovered. Literature studies on the recycling of PV panels focus on Ag recovery from the cells, ignoring the ribbons. However, as will be highlighted in the present study significant amounts of Ag adhere to the metal ribbons and should be recovered and not lost. Currently, there is little research on Ag recovery from metal ribbons (Chen et al., 2021), thus this research gap will be filled. The most used method for Ag recovery from solar cells is etching or leaching in acidic solution based on nitric acid (HNO3) at concentrations of 2.3-14 N and temperatures 25-80 oC. These experiments are performed in open vessels, with or without agitation. Other studies use electrowinning of Ag with methanesulfonic acid solution as the electrolyte (Lee et al., 2018).

In contrast with traditional leaching methods, the hydrothermal leaching process is not limited by the boiling point of water at 100 °C. Using pressurized hot water as the solvent and with elevated temperatures, the hydrothermal method can efficiently leach metal ions from various matrixes. The high temperature accelerates the process, shortens the reaction time, and decreases the required acid concentration (Zheng et al., 2020). In this way, hydrothermal leaching is more efficient and environmentally friendly than traditional leaching methods (Yuxin et al., 2023). Although hydrothermal leaching is a potential leaching method, so far, the process has not been utilized for PV waste, leaving a significant research gap to be filled.

Hydrothermal metal leaching has been efficiently utilized for other types of waste, like lithium-ion battery cathode materials (Zheng et al., 2020), electroplating sludge (Yuxin et al., 2023) or copper anode slime (Rao et al., 2022). So, the present study will fill this research gap, utilizing weak leaching agents to avoid the generation of hazardous waste effluents. The efficient recovery of precious metals from waste PV panels under mild conditions is necessary to reduce the environmental impact and achieve financial benefits.

For this reason, several hydrothermal experiments were performed investigating the influence of four parameters, liquid to solid ratio (L/S), HNO3 normality (N), time (t) and temperature (T) on the leaching efficiency. To efficiently plan the experiments Response Surface Methodology (RSM) with a Box-Behnken design was used. Thus, the objectives of this study were: (i) to leach Ag and Al from both PV cells and ribbons, (ii) to investigate the influence of four parameters (L/S, HNO3, t, T) on the hydrothermal leaching of Ag and Al, (iii) to compare the hydrothermal to conventional leaching and (iv) to investigate the influence of milling and agitation on leaching efficiency.

2. Materials and Methods

The raw materials in this research were waste mono-crystalline (m-Si) and poly-crystalline silicon (p-Si) PV panels, collected from local PV trading/installation companies located in Chania and Heraklion, Greece, as described in Savvilotidou and Gidarakos (2020). Specifically, the p-Si panel (series SYP230S) was manufactured by Risen Energy Co. Ltd and the m-Si panel (series ESP 60) by ExelGroup Ltd. The operational lifetime of the panels was completed due to external damage caused by extreme weather conditions. However, their multi-layered structure was not destroyed. The panels were stored in the laboratory until used in the experiments.

2.1 Composition of c-Si photovoltaic modules

2.1.1 Pretreatment

The panels were initially dismantled, and the aluminum frames, junction-boxes and cables were removed. Then, the panels were shredded in small pieces of approximately 4×4 cm^2. Before further treatment, the backsheet which consists of layers of superposed fluorinated and terepthalate polymers (polyvinyl fluoride or Tedlar film, Polyethylene terephthalate or PET, in the sequence Tedlar, PET, Tedlar) was manually removed.

The manual method was selected after preliminary thermal treatment testing. Some pieces with and without the backsheet were thermally treated (600 ^oC, 30 min). It was observed that significant amount of ash was generated in the pieces with the fluoropolymer backsheet but almost no ash in the pieces without the backsheet. Thus, to avoid hazardous gas emissions of hydrofluoric acid and fluorinated compounds and to ensure ease of handling the panels, it was decided to manually remove the backsheet before further treatment. Fiandra et al. (2019) also reported that the fluoropolymer backsheet removal prior to thermal treatment allows the avoidance of hazardous fluorinated compound emissions generated from conventional heat treatment. During processing, it was observed that for some panel pieces the back film was easily removed by hand, while for other pieces, a vise and saw were necessary. This could be due to non-uniform weathering degradation of the EVA film, as the backsheet was attached to an EVA film used as an encapsulant (Fiandra et al., 2019; Oliveira et al., 2018). Thus, the possible degradation of EVA by weathering may facilitate the delamination of the backsheet, explaining why detachment was easy for some panel pieces. Badiee at al. (2016) mentioned that the ash produced from the thermal decomposition of EVA is almost zero and this was confirmed in the present study.

2.1.2 Thermal Treatment of EoL Si PV Panels

After removal of the white backsheet (section 3.2), the panels were thermally treated to eliminate the EVA layer and thus separate the PV in glass, silicon cells and metal ribbons. The decomposition of the adhesive resin was carried out in a Nabertherm furnace in an oxidizing atmosphere. The samples were heated at 600 °C for 20 min.

After the thermal pre-treatment, the panels were separated by screening with sieves of different sizes into glass, cells and metal ribbons. Glass can be easily recovered and recycled. The separated cells as well as the metal ribbons were subjected to hydrothermal treatment.

2.1.3 Characterization of PV Panel components

The separated cells and metal electrodes were separately milled with a pulverisette 19 FRITCH mill equipped with a sieve of 0.5 mm and then subjected to X-ray fluorescence analysis (XRF, Rigaku Primus IV) to determine their chemical composition. The results are presented in Table 7.1.

The gravimetric determination of the panel components was also carried out taking into account the volatilization of EVA (Table 7.2). To ensure homogeneity and representativeness in each sample, a PV laminate of 15.8 x 15.8 cm^2 was quartered and the mass of glass, solar cells, metallic ribbons and EVA was determined. Since significant amounts of silver were also found in the metal ribbons (Table 7.3), both cells and ribbons were used in the subsequent treatment. The proportion of cells to ribbons (Table 7.2) remained the same in all samples.

The losses due to milling were also calculated. For this reason, cells and ribbons were mixed in the above proportion (Table 7.2) and then milled. The resulting ground sample (mixture of both cells and ribbons) was subjected to XRF analysis.

Table 7. 1 Chemical composition of cells and ribbons.

Element (% w/w)	p-Si Cell	m-Si Cell	p-Si Ribbon	m-Si Ribbon
Si	64.46 ± 0.22	65.18 ± 0.31	2.78 ± 0.09	4.55 ± 0.13
Al	6.16 ± 0.05	9.87 ± 0.07	5.50 ± 0.05	5.59 ± 0.03
Cu	0.25 ± 0.02	0.93 ± 0.01	38.83 ± 0.11	48.51 ± 0.34
Ag	1.13 ± 0.02	1.37 ± 0.01	4.62 ± 0.06	6.22 ± 0.04
Sn	0.10 ± 0.03	0.28 ± 0.01	8.57 ± 0.06	10.68 ± 0.09
Pb	0.13 ± 0.03	0.35 ± 0.01	9.98 ± 0.05	10.13 ± 0.07

Table 7. 2. Gravimetric determination of panel components.

Material (% w/w)	p-Si Panel	m-Si Panel
glass	88.35 ± 0.18	89.37 ± 0.23
cell	5.25 ± 0.08	5.1 ± 0.20
ribbons	1.16 ± 0.01	0.98 ± 0.01
EVA	5.23 ± 0.11	4.56 ± 0.03

2.2 Experimental design: Response surface methodology

Four different parameters (independent variables) of the hydrothermal leaching process were initially investigated: (i) temperature, (ii) time, (iii) S/L ratio and (iv) HNO_3 concentration. The effect of the independent variables on the leaching yield (dependent variable) was evaluated.

Response surface methodology with a Box-Behnken design (BBD) was used to plan the experiments. The design investigated the effects of the parameters on silver leaching by simultaneously differentiating all parameters under consideration, instead of differentiating one parameter at the time. Thus, possible interactions between the factors were identified. The Box-Behnken design effectively avoided the realization of the complete experimental design that could make the process quite demanding in terms of time and resources, without losing any information on the effect of the parameters on the process. In this design, three levels of each factor were defined, low, middle and high level (Tanong et al., 2017; Behera et al., 2018). Among the response surface designs, BBD was slightly more efficient compared to the central composite design and was much more efficient than the three-level full factorial design (Ferreira et al., 2007). The experimental design was performed by Minitab software and the input settings are presented in Table 7.3. All experiments were duplicated.

Table 7. 3. Response surface methodology with a Box-Behnken experimental design.

Run Order	**HNO_3 (N)**	**Time (min)**	**temp (^oC)**	**L/S**
1	1.5	120	120	10
2	1.5	75	100	20
3	1.5	75	120	15
4	2	120	120	15
5	1.5	75	100	10
6	1	30	120	15
7	1.5	30	140	15
8	1.5	30	100	15
9	1	75	120	20
10	1.5	120	140	15
11	1	75	100	15
12	1.5	30	120	20
13	1.5	75	120	15
14	2	75	120	20
15	1.5	120	120	20
16	2	30	120	15
17	1.5	75	140	10
18	2	75	140	15
19	2	75	100	15
20	1.5	120	100	15
21	1.5	75	120	15
22	1	120	120	15
23	1.5	75	140	20
24	2	75	120	10
25	1	75	120	10
26	1	75	140	15
27	1.5	30	120	10

2.3 Metal leaching

As previously mentioned, prior to metal leaching the samples were milled. This facilitated cleavage of the chemical and mechanical bonds between Ag and Al electrodes and Si layer and made the surface area for reaction larger. To test the effect of milling, two samples were created: a powder sample (size <0.5 mm) and a flake sample (random flakes of about 0.5-1 cm^2). Preliminary studies on silver leaching were carried out in samples for screening the optimal conditions by determining the leaching agent (HNO_3) and its concentration, the temperature and contact time. Ag can be easily dissolved in HNO_3, as the possible chemical reactions of Ag with HNO_3 have negative values of standard-state free energy (ΔG) indicating the spontaneous character of the reactions (Kamberovic et al., 2018). Since Ag is more valuable metal compared to Al, choosing only one type of acid is considered as the most profitable solution.

a. Hydrothermal leaching

The hydrothermal leaching tests were performed in cylindrical steel reactors (Techinstro) internally coated with PTFE with a volume of 100 ml. The sample was mixed with 30 mL HNO_3 according to the desired solid-liquid ratio (S/L) and–was hydrothermally treated at 100-140 ^oC for 0.5-2 h without stirring. The resulting product was allowed to cool down and then was filtered with a 0.45 μm syringe filter for subsequent metal analysis in the Inductively Coupled Plasma-Mass Spectrometry (7500 cx ICP-MS, Agilent).

b. Low temperature metal extraction

The optimized conditions of hydrothermal leaching were used in the next experimental series. Keeping the HNO_3 concentration at 2 N and S/L ratio at 1/10, experiments were conducted at lower temperatures (25-45^oC), increasing the processing time (2-24 h), while flakes were used as processing material. The effect of stirring was also tested at the lower temperature experiments (as hydrothermal reactors could not utilize stirring).

3. Results and Discussion

3.1 Composition of end-of-life PV panels

As seen in Table 7.1, the silicon content in the cells was 64.5-65.2% (w/w) while silver was 1.1-1.4% (w/w). The cells composition was in good agreement with the reported by Theocharis et al. (2022). Aluminum content in the cells was about 5.5-7.2 times the silver content. The silver content in the metallic ribbons was much higher, between 4.6-6.2% (w/w). This was attributed to the silver band attached to the PV ribbon for electrical conduction (Patcharawit et al, 2022).

The mass composition (wt. %) of the panel components (without the backsheet mass which was about 11% of the panel mass) is presented in Table 7.2, whereas the losses due to the milling are calculated hereafter. Although the metallic ribbons and cells represented 1-1.16% and 5.1-5.2% of the panel mass, respectively, the content of silver in the metallic ribbons was comparable to that of the cells because of the higher silver content. The proportion of cells to ribbons was kept constant in all experiments.

The total mass loss due to the milling was 8.19% (w/w). The milled sample (mixture of cells and ribbons) was subjected to XRF analysis and the Ag mass loss in the sample was calculated at 30%. In the powder sample, there was almost insignificant mass loss for Si and Al, but there were significant losses for Cu, Sn and Pb. This could be attributed to the very fine particles which were lost in the dust (Cu (75%), Sn (66%), Pb (71%) and Ag (30%)). Thus, the mass loss due to milling was not proportional for all elements, and Ag loss was quite significant. This implies that leaching using panel flakes instead of powder might be preferred.

3.2 Hydrothermal leaching of c- Si panels

Hydrothermal leaching of Si panels revealed that, within the range of tested parameters, reaction time was the most important factor for Ag leaching, followed by HNO_3 concentration. Pareto charts of the significant factor terms and their interactions in response to leaching efficiency are shown in Figure 7.1. Significant terms are above the dashed red line, while non-significant terms are below the red line. The S/L ratio was the least important factor in the case of m-Si and p-Si panels. In addition, the temperature was not a statistically significant factor in both samples (m-Si and p-Si) and there were no significant interactions between the parameters. The analysis of variance of the model is presented in Table 7.4.

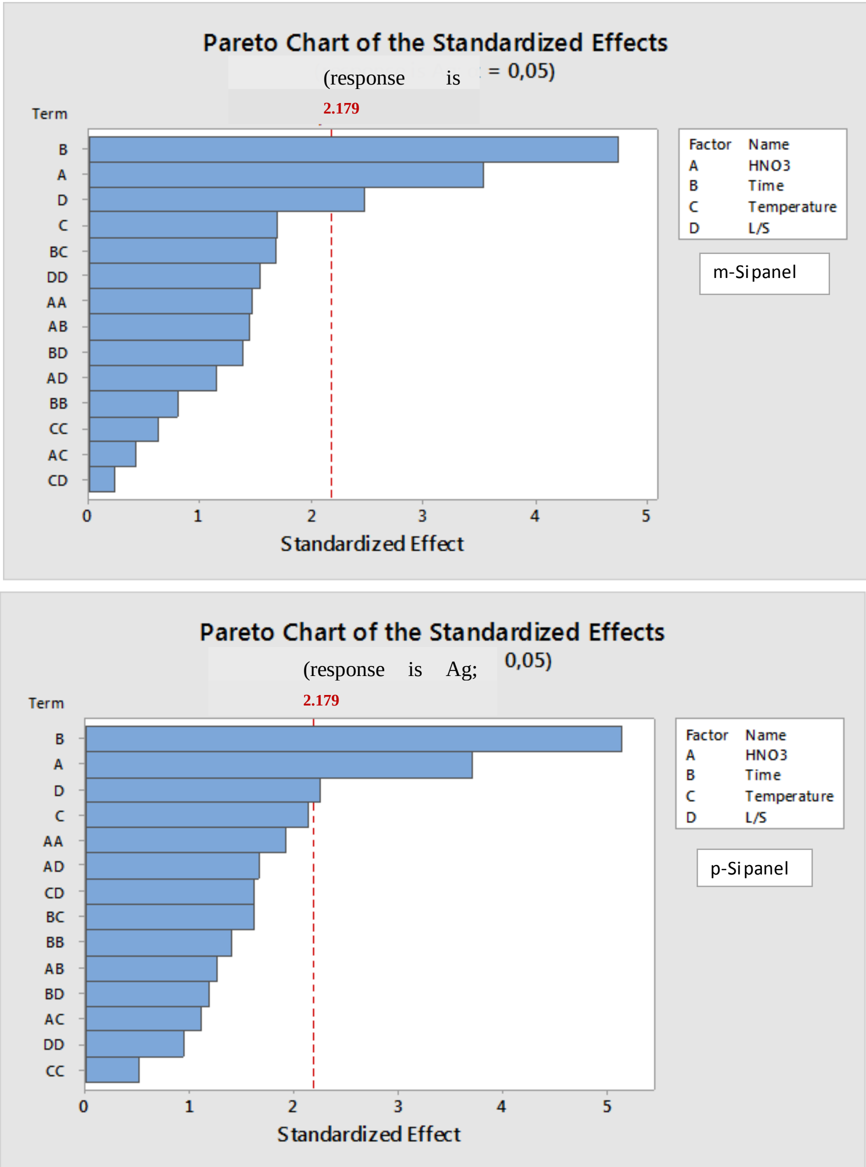


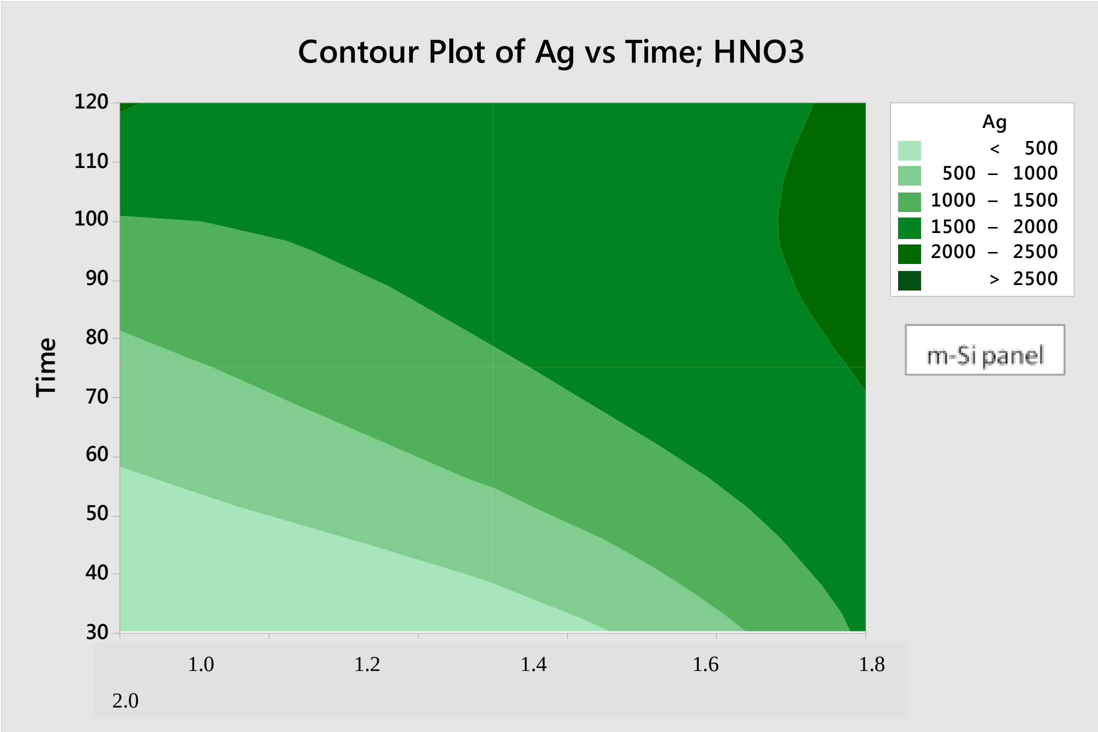
Figure 7. 1. Pareto chart of standardized effects of the factors and interactions for Ag leaching from m-Si and p-Si panels.

Figure 7.2 presents the interaction between time and HNO3 concentration regarding Ag leaching. The Ag leaching efficiency increased significantly with increasing leaching time using 2 N HNO3 for p-Si and m-Si. Figure 7.3 shows other factor interactions. Increasing HNO3 normality and temperature also increase the leaching efficiency, however their effect is less significant than increasing hydrothermal processing time. Reducing L/S ratio increases leaching yield, i.e. the concentration of Ag is higher. The combination of temperatures, times, HNO3 concentrations and S/L ratio in Ag leaching experiments resulted in leaching yields ranging from 0.12-74% for p-Si, while for m-Si the efficiency of Ag leaching ranged between 5.7-66.2%. The highest Ag leaching yield was achieved at 2 N HNO3, S/L = 10, 120 oC and 75 min. The lowest yield value of was achieved at 1.5 N HNO_3, S/L = 15, 100 ^oC and 30 min. Based on optimization obtained by mathematical equation defining the BBD/RSM, the optimized conditions to maximize Ag leaching (100% yield) through hydrothermal treatment were 140 ^oC, 2 h leaching, S/L = 10 and 2 N HNO_3, which was experimentally validated.

Al leaching was also investigated under the same conditions. The main effects plot for the parameters (HNO_3, time, temperature and S/L ratio) revealed that Al leaching favourably increased with processing time and S/L ratio within the range of tested parameters (Fig. 7.4), while the other two factors are less significant. Efficient Al leaching was observed using 1 N HNO_3 under the hydrothermal conditions. For m-Si and p-Si samples, the efficiency of Al leaching ranged from 23-88% and 29-85%, accordingly. The optimized conditions for the maximum Al leaching were 140 ^oC, 2 h leaching, S/L = 10 and 2 N HNO_3. Ag is the most valuable metal in PV panel waste, however favourable Al leaching can be accomplished under the same conditions.

Table 7. 4 Analysis of Variance of the model (Response surface methodology with a Box- Behnken design).

(a)Analysis of Variance (m-Si)	*DF*	*Adj SS*	*Adj MS*	*F-Value*	*P-Value*
Model	14	15647933	1117709	4,2	0,009
Linear	4	11730171	2932543	11,01	0,001
HNO3	1	3326842	3326842	12,49	0,004
Time	1	6014084	6014084	22,58	0
Temperature	1	765697	765697	2,87	0,116
L/S	1	1623548	1623548	6,09	0,03
Square	4	1678834	419709	1,58	0,244
HNO3*HNO3	1	567900	567900	2,13	0,17
Time*Time	1	170053	170053	0,64	0,44
Temperature*Temperature	1	105658	105658	0,4	0,541
L/S*L/S	1	631262	631262	2,37	0,15
2-Way Interaction	6	2238927	373155	1,4	0,291
HNO3*Time	1	562021	562021	2,11	0,172
HNO3*Temperature	1	48457	48457	0,18	0,677
HNO3*L/S	1	353885	353885	1,33	0,272
Time*Temperature	1	748129	748129	2,81	0,12
Time*L/S	1	511250	511250	1,92	0,191
Temperature*L/S	1	15184	15184	0,06	0,815
Error	12	3196596	266383		
Lack-of-Fit	10	1565594	156559	0,19	0,972
Pure Error	2	1631002	815501		
Total	26	18844529			
(b)Analysis of Variance (p-Si)	***DF***	***Adj SS***	***Adj MS***	***F-Value***	***P-Value***
Model	14	8625209	616086	5,1	0,004
Linear	4	6024329	1506082	12,48	0
HNO3	1	1668330	1668330	13,82	0,003
Time	1	3189906	3189906	26,42	0
Temperature	1	552595	552595	4,58	0,054
L/S	1	613498	613498	5,08	0,044
Square	4	1114108	278527	2,31	0,118
HNO3*HNO3	1	444487	444487	3,68	0,079
Time*Time	1	236774	236774	1,96	0,187
Temperature*Temperature	1	31579	31579	0,26	0,618
L/S*L/S	1	108665	108665	0,9	0,361
2-Way Interaction	6	1486771	247795	2,05	0,136
HNO3*Time	1	193108	193108	1,6	0,23
HNO3*Temperature	1	152028	152028	1,26	0,284
HNO3*L/S	1	335452	335452	2,78	0,121
Time*Temperature	1	315618	315618	2,61	0,132
Time*L/S	1	171747	171747	1,42	0,256
Temperature*L/S	1	318818	318818	2,64	0,13
Error	12	1448598	120716		
Lack-of-Fit	10	739026	73903	0,21	0,965
Pure Error	2	709571	354786		
Total	26	10073806			



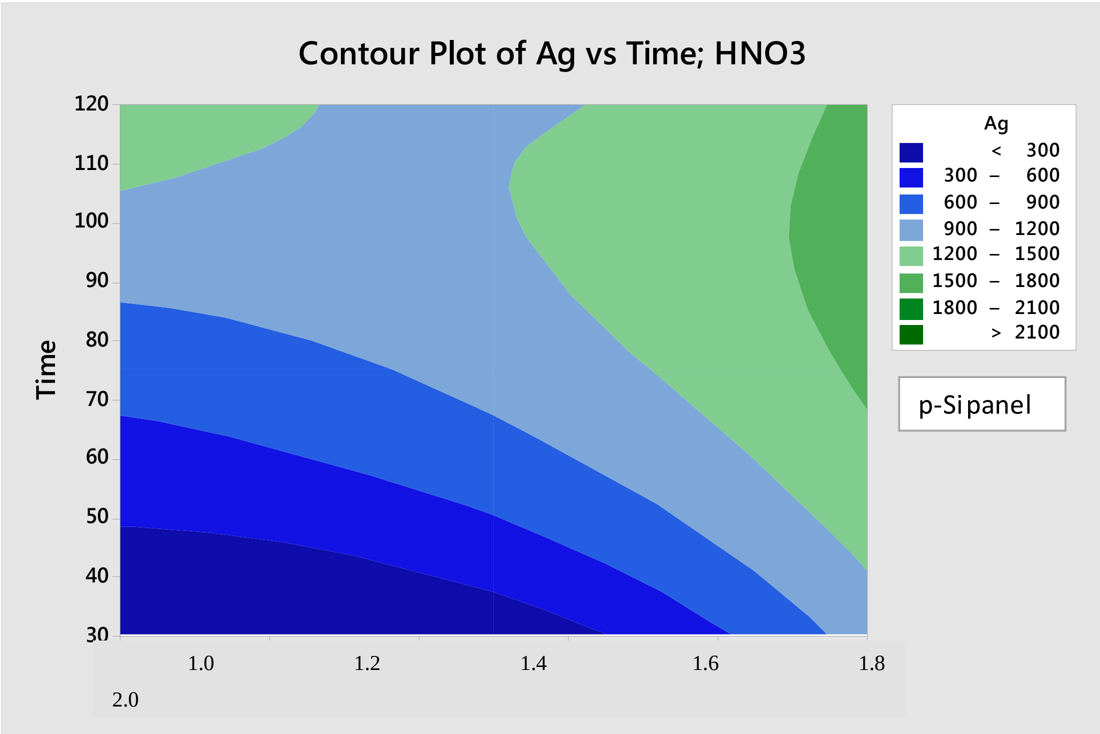


Figure 7. 2. Fitted contour plots of (a) Ag conc. versus time-HNO3 concentration for m-Si and p-Si panels.

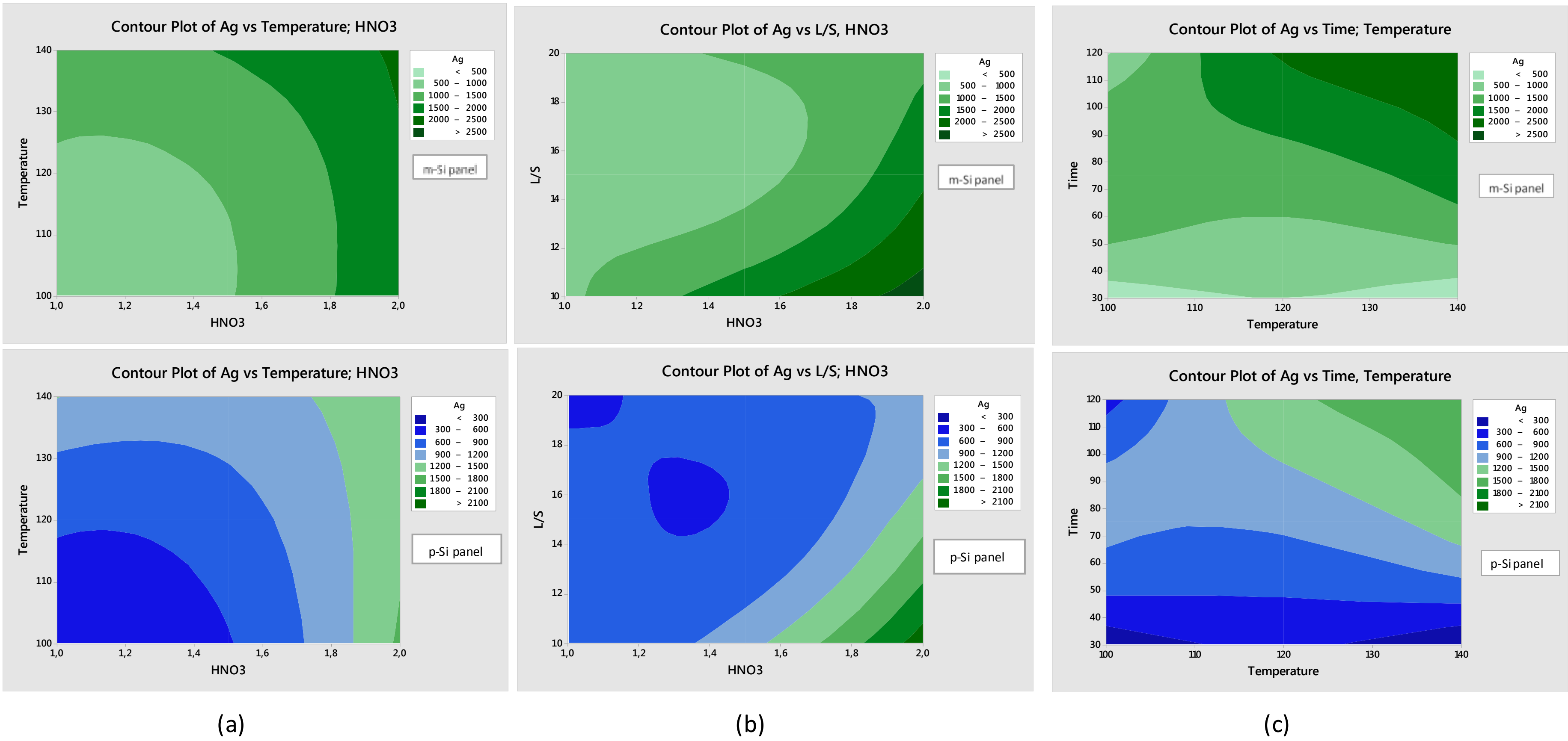


Figure 7. 3. Fitted contour plots of (a) Ag conc. versus temperature-HNO3, (b) Ag conc. versus L/S-HNO3 (b) Ag conc. versus time-temperature for m-Si and p-Si panels.

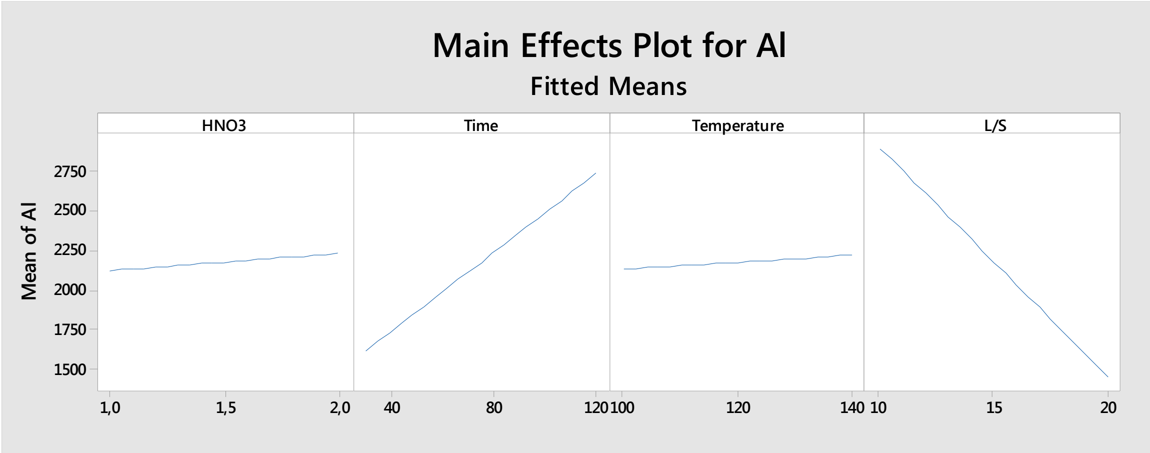


Figure 7.4. The main effects plot for the parameters HNO_3, Time, Temperature and S/L ratio of Al. leaching.

3.3 Conventional leaching of Ag at lower temperatures

Since the elevated temperature of the hydrothermal process was not a statistically significant parameter in the Ag leaching, experiments were conducted at lower temperatures to reduce the energy cost. As time has proved to be an important factor, the effect of extending the leaching time was further investigated. Experiments were conducted at 25 and 45 ^{o}C, keeping the HNO_3 concentration at 2 N and S/L = 10. The effect of stirring was further investigated.

The effect of stirring on Ag leaching was significant as the leaching efficiency increased by 40% when stirring at 200 rpm was applied (2 N HNO_3, S/L = 10, 2 h, 45 ^{o}C), thus stirring was applied in all subsequent experiments at 25 and 45 ^{o}C (Fig. 7.5) (Fig. S2 Appendix). The positive effect of stirring is explained by facilitating the access of metals to the leaching agent (Kamberović et al., 2018). The effect of temperature is illustrated in Fig.7.6a (for m-Si). It was observed that leaching with 2 N HNO_3 for 6 h at 45 ^{o}C had 80% efficiency compared to 63.3% at 25 ^{o}C. In the subsequent step, leaching was performed at 45 ^{o}C with both powdered and flakes m-Si (and p-Si) panels. For m-Si panels, the maximum Ag leaching for flakes was 76.5% at 6 h, while for p-Si panel it was 78% at 6 h (Fig. 7.6b). However, Theocharis et al. (2022) observed almost total leaching (100%) from PV flakes for

5 h leaching, 25 ^{o}C but with a 3 N HNO_3 concentration, while for PV powder 100% leaching was achieved at 25 ^{o}C, 3 N HNO_3, for 2 h. Chen et al. 2020, reported 99.4% Ag leaching at 80 ^{o}C with a 5 N HNO_3 for 1h.

For Al leaching, the efficiency was 61.7% at 45 ^{o}C for 6h compared to 44.7% at 25 oC for 6h. However, for 24 h leaching the leaching efficiency was 61% both at 25 ^{o}C and 45 ^{o}C (Fig 7.7). For the unpulverized sample, the leaching efficiency was 49% at 45 ^{o}C for 8 h (Fig. 7.8). Complete Al leaching was not achieved under low temperature conditions; however, this was favoured under hydrothermal conditions. In the first case, the dissolution of Al may be partially inhibited by the formation of an aluminum oxide (Al_2O_3) layer on the surface and also due to possible Al–Si compounds formation during acid treatment (Šleiniūtė et al., 2020; Yi et al., 2014). After acid leaching, Al can be fully dissolved with NaOH (Yi et al., 2014). Chen et al. (2020) reported 76% Al leaching at 80 ^{o}C with a 5N HNO_3 for 1h, but Theocharis et al. (2022) observed efficient Al leaching 100% for 2 h leaching, at 25 ^{o}C with a 3N HNO_3 concentration.

Limitations of the study

The main problem with e-waste recycling in general, is the complexity of the samples, the tiny amounts of the multiple elements that co-exist on various substrates, as well as utilization of sophisticated alloys make the materials recycling very challenging. In the case of PV cells and ribbons recycling, Ag leaching with HNO_3 causes simultaneous leaching of Al, Cu, Pb and Sn, Selective precipitation of Ag can be accomplished by various steps. Initially, precipitation of Ag and Pb is performed by HCl. Afterwards, H_2S (gas) channelling into the solution until saturation will .create Cu and Sn precipitates. Last, addition of NaOH will precipitate Al. Additional steps are necessary to separate Ag and Pb as well as Cu and Sn precipitates. To separate Ag and Pb precipitates, the precipitate is re-suspended in small amount of water and heated in a boiling water bath to selectively dissolve $PbCl_2$, leaving AgCl almost undissolved. In this way AgCl is obtained (Oxtoby et al., 2011).

Also electrochemical separation of the metal ions is possible through careful selection of cell electrodes and potential (Petrucci, et al., 2007).

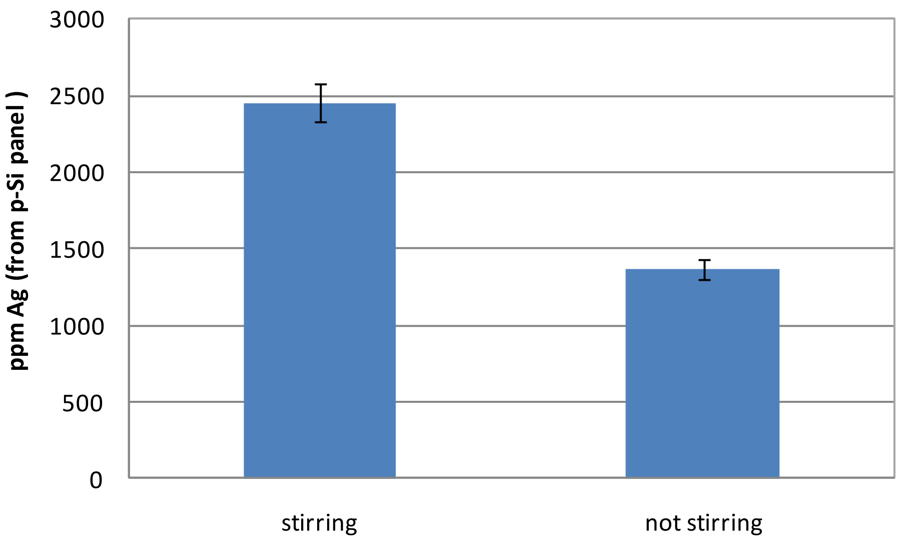


Figure 7. 5 The effect of stirring on Ag leaching (HNO_3 2N, S/L=1/10, 2 h, 45 ^oC).

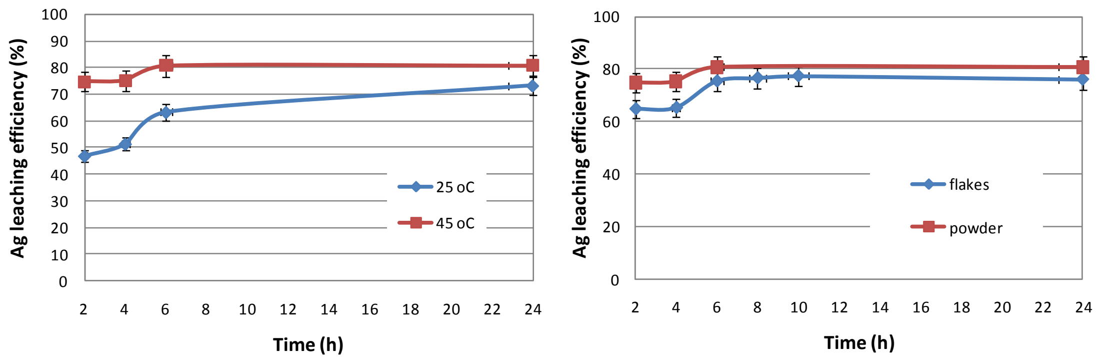


Figure 7. 6 The effect of (a) temperature and (b) milling on Ag leaching (HNO_3 2 N, S/L = 10, m-Si sample).

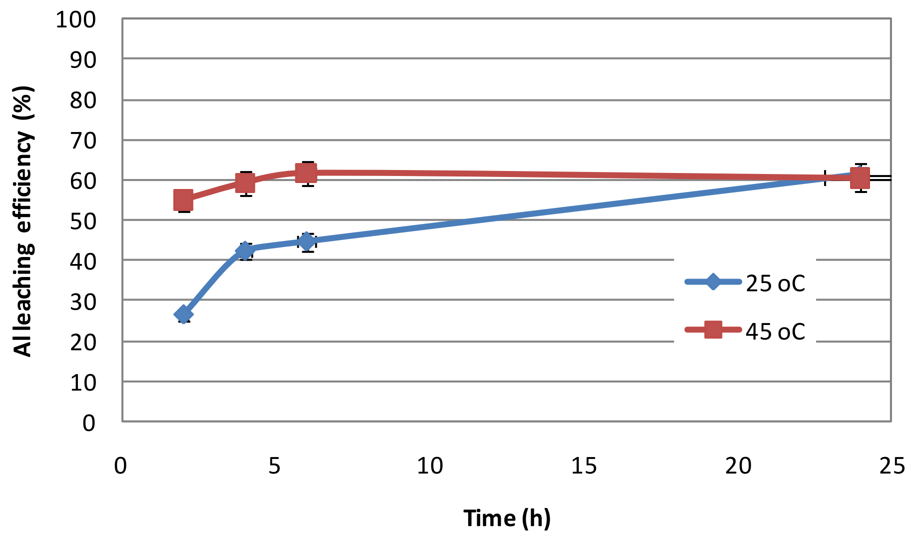


Figure 7. 7 The effect of temperature on Al leaching (HNO_3 2N, S/L=1/10).

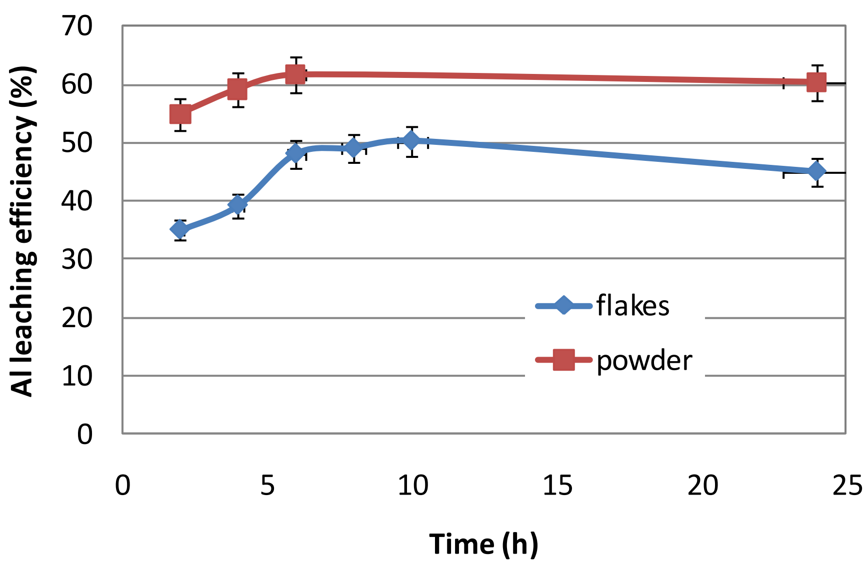


Figure 7. 8. The effect of milling on Al leaching (HNO_3 2N, S/L=1/10, 45 ^oC).

4. Conclusions

PV waste recycling is challenging due to the complexity of the PV structure and the tiny amounts of multiple elements that co-exist in its various components. In this study, hydrothermal leaching experiments were planned using response surface methodology with a Box–Behnken design. The Box–Behnken design effectively avoided the realization of the complete experimental design, which would otherwise make the process more demanding in terms of time and resources, yet without losing any information on the effects of the parameters on the process. The hydrothermal leaching of Ag and Al was studied with the aim of developing an environmentally friendly process that could use weak acids, short leaching times and relatively low reaction temperatures. Before leaching, the fluoropolymer backsheet was manually removed and thermal degradation of the ethyl vinyl acetate (EVA) polymer was applied. The optimized hydrothermal process parameters were L/S = 10, 2 N HNO3, 120 min and 140 °C. Under these parameters, Ag and Al were efficiently extracted. Since temperature was not a statistically significant parameter, further experiments were performed at lower temperatures to compare hydrothermal leaching to conventional low temperature processes. Under the low temperature leaching conditions, Ag could be leached using stirring and longer reaction times to generate a yield of 80.7–85%, while Al leaching was insufficient (at 56.6–61% efficiency) due to the formation of Al–Si compounds. Hydrothermal treatment favors the leaching of valuable metals; furthermore, it is more efficient and environmentally friendly than traditional leaching methods and may create financial benefits through the urban mining of PV waste.

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CHAPTER 8

8.1 Conclusions

A large amount of EoL laptops in Greece will be generated in the future. These are estimated considering time-varying parameters of penetration rate, population, laptop weight and lifespan. The coronavirus pandemic significantly boosted the laptop sales. The most important parameter affecting the calculated amounts of obsolete laptops/tablets is the weight of the devices, followed by the Weibull parameters (lifetime). The estimated CRMs and PMs amounts in these wastes reveal a great potential of key metals recovery.

A great number of obsolete smartphones will be generated in the future, while the waste feature phones will be far less (2.3 million feature phones and 40.7 million smartphones in 2021-2035). The composition of the future mobile phone waste stream is therefore predicted to change rapidly. The accumulated waste mobile phones were 31.2 million units in 2010-2020 (24.6 million feature phones and 6.6 million smartphones). The amount will grow to 43 million units in 2021-2035. The effective recycling of obsolete phones can cover the demand for key metals in the new smartphones for more than a decade in Greece. Currently, the recycling potential for REEs is low so closing the loop between resources, materials and their waste demands new efficient innovative recycling processes. The information can contribute to creating a data set about metal demand and secondary supply which is needed to move to a circular economy.

Existing compilation schemes for e-waste such as laptops and mobile phones especially should be improved to enhance collection as much as possible. In this way valuable resources, like key metals will not be lost and the EU can become more material sufficient in accordance with the circular economy concept. Our path to a sustainable future comes through zero waste policies.

In the future great amounts of discarder LIBs from EVs will be observed. The adoption of circular economy business models is necessary to deal with the increasing volumes of EoL LIBs from electric vehicles. These models involve Remanufacturing, Reuse and Recycling (3R) of the waste batteries to extend their life before recovering valuable materials through recycling. The estimations of the amounts and capacity of batteries directed to Remanufacturing, Reuse and Recycling in the EU-27 are essential to plan their efficient management. By 2030, a recycling infrastructure with an annual recycling capacity of 15-28 GWh will be necessary in the EU-27. The recycled quantities of Li, Co, Ni and Cu in waste LIBs in 2030 may be directed to manufacturing of new |LIBs in the EU covering 5.2-6.2, 9-11.3, 6-6.9 and 6.1-7.2 % of annual demand according to sales of new EVs in the same year.

Future investments in LIB remanufacturing, reuse and recycling infrastructures are necessary to deal with the increasing amounts of waste. The batteries available for a second life offer a great opportunity to cover the stationary energy needs of PV systems. In Germany, the accumulated B2L capacity until 2030 could cover between 28-60% of the required battery capacity to support 98 GW PV installations. In France, the batteries available for a second life could cover 26-87% of stationary storage needs of 42 GW PV installations by 2030.

PV waste assessments in all 27 EU countries are accomplished according to three scenarios, the Early and Regular Loss scenarios proposed by IRENA and the scenario proposed by the EU WEEE Directive. By 2050, 14.3-18.5 Mt PV waste will be generated in EU-27 while the profit of PV recovered materials will be 22-27.4 billion USD. C-Si technology will dominate the PV waste, followed by CdTe. The EU must be prepared to deal with future amounts of PV waste. On this basis, a scheme for the EU-27 countries to launch a viable PV recycling business is presented according to the timetable estimated in this study. A core of 4 countries can start a viable recycling industry by 2024-2032 (Germany, Italy, France and Spain). Eight more countries will be able to successfully operate individual PV recycling plants by 2037-2049 (the Netherlands, Belgium, Greece, Austria, Denmark, Portugal, Poland and Hungary). The remaining fifteen countries will not be able to operate a PV recycling plant until 2050, so PV waste recycling partnerships will need to be established in these countries, taking into account the proximity of existing recycling facilities in neighbouring countries. The recycled materials can be used in the construction of new PVs within EU, closing the material loop and therefore strengthening the economy. This will ensure a sustainable and reliable source of raw materials in the EU, which will contribute to covering the apparent demand for minerals and metals associated with low-carbon technologies.

PV waste recycling is challenging due to the complexity of the PV structure and the tiny amounts of multiple elements that co-exist in its various components. The hydrothermal leaching of Ag and Al was studied with the aim of developing an environmentally friendly process that could use weak acids, short leaching times and relatively low reaction temperatures. Hydrothermal treatment favors the leaching of valuable metals; furthermore, it is more efficient and environmentally friendly than traditional leaching methods and may create financial benefits through the urban mining of PV waste.

8.2 Future work

The research area of e-waste estimations is vast. However, in Greece e-waste estimations are completely lacking, therefore it is suggested that future work should include e-waste estimations for Greece for more waste types, other than laptops, tablets, mobile phones, PV panels and LIBs from EVs. This will enable the observance of collection rates and relative targets set by the EU. Furthermore, future research steps should include the formation of a national platform where future e-waste amounts of all categories can be compiled; this will help the country comply to the EU mandates.

Moreover, the estimations of precious and critical metals in other e-waste categories, as well as the estimation of the associated economic revenue will indicate from which e-waste categories, critical metals other than base metals should be recycled. This information together with the spatial generated quantities will reveal the regional economic potential for recycling, which is important as transportation costs are important for the logistics of the e-waste recycling process.

Future research should focus on leaching and recovery of other metals from e-waste categories other than PV panels, i.e, LIBs or smartphones. The area of e-waste recycling is huge and demanding, but is an opportunity, rather than a problem, and should play significant role in the sustainable use of resources. For instance, the recovery of lithium or cobalt from waste LIBs is an emerging research area.

Ag leaching and recovery from waste PV panels can be further investigated by examining a wider range of parameters and conditions or other leaching agents (or combinations) with hydrothermal treatment. An economic and environmental evaluation of the process could also be the focus of future research.