Critical metals in electronic equipment

A methodology to model end-user stocks and flows

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Abstract

Electronic equipment (EE) contains important material resources, not only bulk materials but also precious metals and critical metals. While the recycling of bulk materials and precious metals is often well established, efforts to specifically recover critical metals from EE are only beginning. They are hampered by low contents per device, limitations of recovery technolo-gies, lack of economic incentives as well as limited knowledge of stocks, flows and disposal pathways of critical metals incorporated in EE. For an efficient management of these resources, it is thus important to know where they are located, how long they are used and when and how they are disposed of. This can be achieved by dynamic material flow analyses (MFAs), which are often used to investigate the development of material cycles over time.

In this thesis, we explore the fate of critical metals in EE used by private end-users in Switzer-land with a focus on the examples of indium and neodymium. Additionally, we include the precious metal gold as a reference metal with already well-established recycling processes. The main objectives of this research are to better understand the metabolism of the anthroposphere regarding critical metals connected to the use of EE, provide a basis to develop appropriate tools and alternatives to manage efficient recycling systems and encourage the recycling and reintegration of critical metals into anthropogenic material cycles. We explore the fate of critical metals and their suitability for urban mining with a special focus on the service lifetime, storage time and disposal pathways of different device types. Within the use phase, we investigate the past and current quantities of electronic devices containing indium, neodymium, and gold in the in-use stock and quantify the flows between the use, storage and disposal phase. We further analyze the reasons for discre-pancies between low collection flows and high sales flows or long phase-out periods of technologies that are no longer sold. Within the collection, recycling and disposal phase, we assess sinks resulting from the dissipation of critical metals due to inappropriate recycling processes. As input data of dynamic MFAs are often acquired from many different sources with varying data reliability, we systematically consider the associated data uncertainties.

Data for the service lifetime, storage time and disposal pathways are collected via a survey and additional interviews. Devices included are mobile phones, smartphones, desktop and laptop computers, monitors, cathode ray tube and flat panel display televisions, DVD players and headphones. Based on the empirical results, the system for the dynamic MFA is deve-loped as a cascade model, with each step consisting of an in-use-stock and a storage stock for new and second-hand devices, respectively. In order to track the three metals from their entry into Switzerland as components of new devices until their recovery, disposal in landfill or dissipation to the environment, the cascade model is extended with the collection, recy-cling, and disposal phase. With statistical entropy analysis (SEA), we further analyze the dilution or concentration of the metals during their route through the current system. Using a customized software tool, we apply Monte Carlo simulation to systematically consider data uncertainty in the calculation of the dynamic MFA.

The cascade model provides new and important insights regarding product lifetimes and the transfer of devices from active use to storage and disposal. We show that not only the service lifetime but also the storage time and the flows between the in-use stock and the storage stock are important to consider in dynamic MFAs. The median service lifetime of new devices varies from 3 years for mobile

phones to 9 years for CRT TVs, the median storage time from 0 years for most device types to 2 years for headphones. Due to reuse and storage, the total time a device stays in the use phase is significantly prolonged. Compared to the median service lifetime of new devices, the median total lifetime increases, for example, from 3 to 7 years for mobile phones and from 5 to 8 and 9 years for desktops and laptops, respectively. The results highlight the importance of the storage stock, which accounts for 25% (in terms of mass) or 40% (in terms of pieces) of the total stock of EE in 2014. Our study thus provides insights into the 'black box' that has been the usual way of modeling the use phase of end-user products so far. Furthermore, by differentiating among device types, the changing composition of outflows due to technology changes is accounted for.

With the extended model, we are able to show the final destinations of indium and neodymium within the collection, recycling and disposal phase and quantify the related stocks and flows. The largest quantities of all three metals are still found in the EE currently in use and amount to 1.7 tonnes of indium, 39 tonnes of neodymium and 4.8 tonnes of gold. The second largest stocks are disposed slags in landfills for indium, slags used for construction for neodymium, and the output of metal recovery processes for gold. The average metal quantities reaching recycling in 2014 were 90 kg for indium, 2800 kg for neodymium and 330 kg for gold. With SEA, we illustrate how indium and neodymium are successfully concentrated during preprocessing, but subsequently lost in smelting and incineration processes. The variable data quality in MFAs is accounted for in a comprehensive and flexible way, by the inclusion of input data uncertainty in the form of probability distributions, by Monte-Carlo simulation and analysis of the resulting probabilistic stocks and flows. The presented approach is a step towards a deeper understanding of the stocks and flows of EE and its incorporated critical metals. The generic model can be customized and applied to any end-user product that is potentially reused and stored after its first service life.

Zusammenfassung

Elektronische Geräte (EG) enthalten wichtige Ressourcen, darunter Basismetalle und Kunststoffe, aber auch Edelmetalle und kritische Metalle. Während das Recycling von Basis- und Edelmetallen schon lange etabliert ist, stehen die Bemühungen, kritische Metalle aus EG zurückzugewinnen, noch am Anfang. Gründe dafür sind geringe Metallgehalte pro Gerät, thermodynamische Grenzen der Rückgewinnungsprozesse, fehlende ökonomische Anreize sowie geringe Kenntnisse über die Lager, Flüsse und Entsorgungswege von kritischen Metallen in EG. Für ein effizientes Ressourcenmanagement ist es also wichtig zu wissen, wo sich die Geräte befinden, wie lange sie verwendet werden und wann sie wie entsorgt werden. Um solche Entwicklungen von Materialzyklen im Zeitablauf zu analysieren, werden häufig dynamische Materialflussanalysen (MFAs) verwendet.

In der vorliegenden Dissertation wird am Beispiel von Indium und Neodym der Verbleib kritischer Metalle in EG, die von privaten Haushalten in der Schweiz genutzt werden, untersucht. Zusätzlich wird das Edelmetall Gold, als Referenzmetall mit bereits existierender Rückgewinnung, in die Untersuchung miteinbezogen. Die Hauptziele dieser Arbeit sind es, den Metabolismus der Anthroposphäre in Bezug auf kritische Metalle als Bestandteile von elektronischen Geräten besser zu verstehen, eine Grundlage für die Entwicklung geeigneter Instrumente für das Management von effizienten Recyclingsystemen zu schaffen, sowie das Recycling und den Rücklauf von kritischen Metallen in anthropogene Materialzyklen zu fördern. Der Verbleib der kritischen Metalle im System sowie ihre Eignung für sogenanntes 'urban mining' wird mit besonderem Fokus auf die Nutzungszeit, die Aufbewahrungszeit und die Entsorgungswege der verschiedenen Gerätetypen bewertet. Innerhalb der Nutzungs-phase untersuchen wir die vergangenen und gegenwärtigen Lager an EG, die Indium, Neodym und Gold enthalten, und quantifizieren die Flüsse zwischen der Nutzungs-, Aufbewahrungs- und Entsorgungsphase. Darauf aufbauend analysieren wir die Gründe für Diskrepanzen zwischen niedrigen Sammelmengen und hohen Absatzströmen oder lange Auslaufzeiten von Technologien, die nicht mehr verkauft werden. Innerhalb der Sammel-, Recycling- und Entsorgungsphase beurteilen wir die Senken, die sich aus der Dissipation von kritischen Metallen aufgrund von ungeeigneten Recyclingprozessen bilden. Da oft nur wenige und unvollständige Daten vorhanden sind, berücksichtigen wir systematisch die damit verbundenen Datenunsicherheiten.

Daten für die Nutzungszeit, die Aufbewahrungszeit und die Entsorgungswege werden über eine Umfrage und zusätzliche Interviews gesammelt. Berücksichtigte Geräte sind Mobil-telefone, Smartphones, Desktop- und Laptop-Computer, Monitore, Röhren- und Flachbild-fernseher, DVD-Player und Kopfhörer. Basierend auf den empirischen Ergebnissen wird das System für die dynamische MFA als Kaskadenmodell entwickelt, wobei in jeder Stufe das Lager in Nutzung und Aufbewahrung für neue bzw. gebrauchte Geräte aufgeteilt wird. Um den Weg der drei Metalle vom Verkauf in die Schweiz als Bestandteil neuer Geräte bis zur Verwertung, Entsorgung oder Dissipation in die Umwelt zu verfolgen, wird das Kaskaden-modell um die Sammel-, Recycling- und Entsorgungsphase erweitert. Zusätzlich wird die Verdünnung der Metalle auf ihrem Weg durch das System mit der Statistischen Entropie Analyse (SEA) untersucht. Mit Hilfe eines maßgeschneiderten Software-Tools wird die Daten-unsicherheit der dynamischen MFA mit Monte-Carlo-Simulation berücksichtigt.

Das Kaskadenmodell liefert neue und wichtige Erkenntnisse über die Produktlebensdauer und den Übergang von EG vom aktiven Gebrauch in die Lagerung und Entsorgung. Wir zeigen, dass nicht nur die Nutzungszeit, sondern auch die Aufbewahrungszeit und die Flüsse zwischen dem aktiv genutzten Bestand und der Aufbewahrung für dynamische MFAs wichtig sind. Die mittlere Nutzungszeit von Neugeräten variiert zwischen 3 Jahren für Mobiltelefone und 9 Jahren für Röhrenfernseher, die mittlere Aufbewahrungszeit zwischen 0 Jahre für die meisten Gerätetypen und 2 Jahren für Kopfhörer. Durch Wiederverwendung und Aufbewahrung wird die totale Aufenthaltszeit in der Nutzungsphase erheblich verlängert. Im Vergleich zur Nutzungszeit von Neugeräten erhöht sich die mittlere totale Aufenthaltszeit zum Beispiel für Mobiltelefone von 3 auf 7 Jahre und für Laptops und Desktops von 5 auf 8 bzw. 9 Jahre. Die Resultate heben zudem die Wichtigkeit der Aufbewahrung hervor, deren Lager 25% (bezogen auf Masse) oder 40% (bezogen auf Stückzahl) des Gesamtlagers an EG in 2014 ausmacht. Unsere Studie bietet also Einblicke in die 'Black Box', die bisher die übliche Art war, die Nutzungsphase von Endverbraucherprodukten zu modellieren. Darüber hinaus wird durch die Einbeziehung verschiedener Gerätetypen die sich ändernde Zusammen-setzung der Abflüsse aufgrund von Technologieänderungen berücksichtigt.

Das erweiterte Modell ermöglicht es, die endgültigen Senken von Indium und Neodym in der Sammel-, Recycling- und Entsorgungsphase aufzuzeigen und die damit verbundenen Lager und Flüsse quantifizieren. Die grössten Lager aller drei Metalle sind nach wie vor in EG in der Nutzungsphase zu finden und machen rund 1.7 Tonnen Indium, 39 Tonnen Neodym und 4.8 Tonnen Gold aus. Die zweitgrössten Lager sind für Indium deponierte Schlacken aus Müllverbrennungsanlagen, für Neodym Schlacken aus Metallhütten welche meist als Baumaterial verwendet werden, und das wiedergewonnene Gold aus Metallrück-gewinnungsprozessen. Rund 90 kg Indium, 2800 kg Neodym und 330 kg Gold sind 2014 in der Schweiz in den Recyclingprozess zurückgeführt worden. Mit SEA können wir veranschaulichen, wie sich die Metalle bei der Vorverarbeitung erfolgreich aufkonzentrieren lassen, aber später in Schmelz- und Verbrennungsprozessen wieder verloren gehen. Die variable Datenqualität in MFAs wird durch die Darstellung der Unsicherheiten von Inputdaten mit Wahrscheinlichkeits-verteilungen, Monte-Carlo-Simulation und die Analyse der daraus resultierenden probabilistischen Lager und Flüsse umfassend und flexibel berücksichtigt. Der verwendete Ansatz ist ein Schritt hin zu einem tieferen Verständnis der Lager und Flüsse von elektronischen Geräten und den darin enthaltenen kritischen Metallen. Das generische Modell kann auch für andere Endverbraucherprodukte angepasst und verwendet werden, welche nach ihrer ersten Lebens-dauer potenziell wiederverwendet und aufbewahrt werden.

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Abbreviations

AG	Aktiengesellschaft
CCFL	Cold cathode fluorescent lamp
CED	Cumulated energy demand
CH	Switzerland (Confoederatio Helvetica)
CHF	Swiss fFanc
CIS	Commonwealth of Independent States
CRT	Cathode ray tube
CSV	Comma-separated values
DOI	Digital object identifier
DVD	Digital video disk
EC	European Commission
EE	Electronic equipment
EEE	Electrical and electronic equipment
EG	Elektronische Geräte
EOL	End-of-life
EPA	Environmental protection agency
EU	European Union
e-waste	Waste electrical and electronic equipment
FPD	Flat panel display
FSO	Swiss Federal Statistical Office
GDP	Gross domestic product
GHG	Greenhouse gas
GIS	Geographic information system
HDD	Hard disk drive
ICT	Information and communication technology
IPCC	Intergovernmental Panel on Climate Change
IT	Information technology
ITO	Indium tin oxide
iu	in use
LCA	Life cycle assessment
LCD	Liquid crystal display
LED	Light-emitting diode
MFA	Material flow analysis
MJ	Megajoule
MWI	Municipal waste incineration
NiMH	Nickel metal hydride
niu	no longer in use
ODD	Overview, design concepts, details
OFCOM	Swiss Federal Office of Communications
ORDEE	Ordinance of the Return, the Take-Back, and Disposal of Electrical and Electronic Equipment
PGM	Platinum group metal
PMS	Precious metal smelter
ppm	Part per million
PRO	Producer responsibility organization

PWB	Printed wiring board
REE	Rare earth element
RQ	Research question
RSE	Relative statistical entropy
SCEA	Swiss consumer electronics association
SD	Standard deviation
SEA	Statistical entropy analysis
SI	Supporting information
SL	Service lifetime
SL2	Second service lifetime
SSD	Solid state drive
ST	Storage time
ST2	Second storage time
TL	Total lifetime
TV	Television
U.S./US(A)	United States (of America)
UK	United Kingdom
UNEP	United Nations Environment Programme
UNU	United Nations University
US$	US Dollar
USGS	United States Geological Survey
WEEE	Waste electrical and electronic equipment

Chapter 1

Introduction

1.1 Critical Metals in Electronic Equipment

The fast pace of innovation cycles for electrical and electronic equipment (EEE) and the falling prices for new devices lead to short product lifetimes and increasing sales.[1] The resulting waste electrical and electronic equipment (WEEE) or e-waste is a fast-growing waste stream that challenges waste management both in developed and developing countries.[2] EEE and the resulting e-waste contain important material resources, including bulk materials (e.g. iron, aluminum, copper or plastics), precious metals (e.g. gold, silver or platinum) and critical raw materials (e.g. indium, tantalum or rare earth elements, REE).[3] Pursuant to a definition of the ad hoc working group on defining critical raw materials of the European Commission (EC)[4,5], a material is termed 'critical' when the risks of a supply shortage and its impacts on the economy are higher compared to most of the other raw materials. Critical raw materials found in EE can be all classified as metals[3,6] and are thus further referred to as 'critical metals'.

The material mix used to manufacture electronic equipment (EE), for example, computer, mobile phones, or televisions (TVs), has become increasingly complex in the last 30 years, as illustrated in Figure 1.1 by the example of the composition of printed wiring boards (PWBs). In the 1980s, a PWB contained about 11 elements, while today, more than half of all stable elements are used.[7]

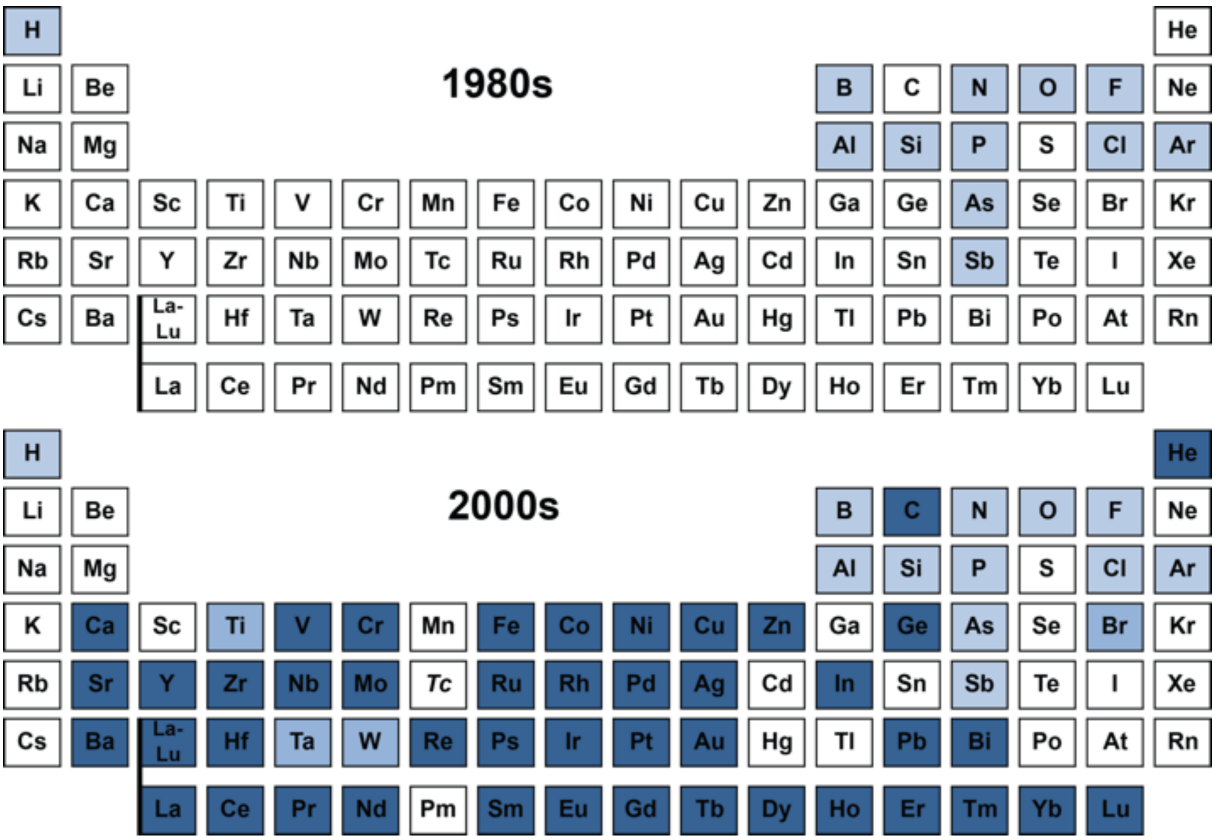

Figure 1.1: Composition of a PWB in the 1980s and 2000s. Adapted from Stamp[7]

The growing demand for EE combined with the increased material complexity has led to an increased demand for critical metals in the past 30 years. Their demand is expected to further increase in the future, not only for the production of EE but also due to their important role for other emerging technologies, such as electric vehicles or energy technologies (solar cells, wind turbines etc.), among others.[7–9] As critical metals are applied in emerging technologies due to specific physical or chemical properties, their substitution is often not possible, which leads to vulnerability to supply shortages.[7,10]

The supply of critical metals may be restricted due to many factors. An important *geological factor* is the trend towards declining ore grades in the past years.[11] A *geopolitical factor* is the uneven distribution of raw material deposits and their production concentrated within a few countries. For example, China has a global market share of over 80% in the REE production and has implemented export quotas in 2010, which widely affected industries worldwide.[12] *Technical factors* include the interlinkage of metal supply chains. Critical metals often naturally occur in ores in combinations. The supply of a critical metal is thus linked to the demand for carrier metals and cannot be independently increased.[13,14] In the waste stream, metal combinations are often more complex and different from naturally occurring combinations in ores. Therefore, the recovery of critical metals from end-of-life (EOL) products and their reintegration into the anthropogenic material cycles is challenging. As the extraction and refining of many critical metals, because of low and decreasing ore grades, is often linked to high environmental impacts, increase in demand is always connected to *environmental factors*, such as regulations, restrictions or certification requirements.[14,15]

In the last ten years, many publications have addressed the issue of supply risks and 'criticalities' of materials.[4–6,16–20] In most of the studies, the criticality is assessed as a two-dimensional matrix. One dimension refers to supply risk, the second to some form of vulnerability to supply restriction.[21] Graedel et al. propose to include environmental implications as a third dimension.[22] As the supply risk, the vulnerability and the environ-mental implications cannot be directly measured, various indicators are used as proxies to quantify these three dimensions.[14] The Committee on Critical Mineral Impacts on the U.S. Economy of the National Research Council[16] and the ad hoc working group of the European Community[5] refer in their studies to the specific economic context of the U.S. and the European Union. In contrast, Behrendt et al., Buchert et al., and the U.S. Department of Energy [3,6,18,20] address specific technologies. Besides clean energy technologies including electric vehicles, energy-efficient lighting, photovoltaics and wind turbines, EE is among the most important applications of critical metals.

Considering all the above-mentioned studies, the most critical raw materials appear to be antimony, beryllium, cobalt, gallium, germanium, indium, lithium, niobium, platinum group metals (PGM), REE, tantalum, tin and tungsten. However, this result has to be interpreted with care, since the scopes and methodologies of the studies addressed differ from each other and the criticality of a metal is subject to high temporal dynamics as it may rapidly change due to altering conditions.[23] Recent studies also criticize current methods regarding the selection of indicators or arbitrary thresholds to separate critical from 'non-critical' metals.[14,21,24] Glöser et al. shows the close relation between criticality matrices and classical risk analysis and its implication to assign criticality levels to specific metals.[14]

While focusing on different aspects, many studies emphasize that consistent and reliable information on critical raw materials still appears to be missing both for primary and secondary supply. Especially the need to improve the availability of credible, consistent stock and flow data has been highlighted in several studies as a prerequisite for sound policy- and decision-making (e.g. refs 5, 18 and 25).

Furthermore, critical issues such as the communication of uncertainties and the validation of results should be addressed.[25,26] The ad hoc working group of the EC specifically recommends to build a European Raw Materials Yearbook together with national geological surveys and mining/processing industries.[5] Regarding secondary production, a review considering data availability of stocks in society has shown that information on the in-use stocks is sufficiently detailed only for five base metals (aluminum, copper, iron, lead and zinc), and sparse or almost inexistent for all other metals. In particular, there seems to be essentially no information on stocks in 'hibernation', in tailings repositories, in industrial stockpiles, or in landfills. Furthermore, only sparse data is available on the product lifetimes for almost the entire periodic table of the elements, which makes it difficult to estimate outflows from the in-use stocks to determine future reuse and recycling rates.[26] A subsequent review regarding research of anthropogenic elemental cycles shows a similarly weak data basis, especially when it comes to dynamic stock and flow information.[27] However, secondary production is expected to become increasingly important because for many materials their extraction from geological resources has significantly increased in recent decades and induced a continuous shift to stocks in the anthroposphere.[28]

1.2 e-Waste Collection and Recycling

The fast growing e-waste streams with their complex material mix pose various challenges when it comes to collection and recycling. Baldé et al.[2] describe four different ways of WEEE collection: the collection in official take-back systems, the disposal of e-waste in mixed residual waste, the collection outside official take-back systems in developed countries and the informal collection in developing countries.[2] They estimate that worldwide around 15% and in Europe around 40% of the e-waste reaches official take-back systems. The WEEE Directive (2002/96/EC)[29], which entered into force in 2003, used to demand a collection target of 4 kg/inhabitant. The recast of the WEEE Directive (2012/19/EU) introduces until 2019 a stepped increase in the collection targets to 65% of the average weight of EEE put on the market in the three preceding years, or, alternatively, 85% of the WEEE generated.[30] The WEEE Directive sets the framework for setting up collection and recycling systems in the European Union (EU), however, the implementation of the WEEE directive differs greatly among member countries. In Switzerland, where the case study investigated in this thesis is located, the Ordinance of the Return, Take-Back and Disposal of Electrical and Electronic Equipment (ORDEE) came into force in 1998.[31,32] In contrast to the WEEE Directive, it does not set specific collection targets. However, Swico Recycling, the national not-for-profit producer responsibility organization (PRO) for taking back EE, claims to collect over 90% of the e-waste generated.[33] Technical requirements in the recycling process such as depollution or recovery targets for different WEEE categories[29,30] are regulated in the CENELEC standard or system-specific technical regulations (e.g., refs 34 and 35). Recovery targets are related to the total mass of EEE of a certain WEEE category and are often met through the recycling of base metals (iron, aluminum and copper) and plastics. Furthermore, the recovery of precious metals is economically incentivized and therefore well established. Specific recovery targets for individual materials to prevent their losses in the recycling process have not yet been established. The low collection rates in many countries, the great differences in how collection and recycling systems are set up, and only mass-related recovery targets are difficult preconditions for the recycling of the increasingly complex WEEE. Hence, the need for the implementation of more efficient collection and recovery systems is prominently addressed in several studies.[5,11,13,16,36,37]

Recycling efforts to specifically recover critical metals are in most cases only beginning and they are thus often lost in the recycling process. Challenges are, first of all, the use of critical metals at very low contents in individual products that leads to highly diluted stocks and flows which are difficult to concentrate and recover in current recycling systems. As shown by UNEP[25], for many critical metals, for example gallium, germanium, indium, osmium REE, tantalum and tellurium, recovery rates were estimated to be below 1%. Various recycling processes are currently under development, but they often imply high costs compared to relatively cheap primary resources.[25] A further challenge arises from the thermodynamic properties of metal combinations in EE that often differ from naturally occurring metal combinations in ores and lead to trade-offs between the recovery of different metals.[7,38,39] Therefore, critical metals are either dissipated into recovered base metals or slags that are disposed of in landfills or used for construction, either within landfills or, for example, for the reinforcement of dams. To allow for a gradual transition from open to closed material cycles, a more product-centric approach in contrast to the usual material-centric approach is demanded. A product-centric approach enables to target specific components of a product, devising ways to separate them and recover metals from the complex interlinkages within a product.[40] In addition, design for recycling, including design for disassembly or design for resource efficiency is required to create highly concentrated feed material for the various recovery processes and take into account fundamental physical and thermodynamic limits of the recycling and recovery processes.[39–41]

1.3 Methods for Analyzing Material Cycles

Various studies indicate that efficient collection and recycling also rely on knowledge of anthropogenic material cycles regarding location, lifetime, disposal pathways, quantities, and qualities of EE.[25,26,39–41] This information enables recycling systems to forecast future mass flows, provide for sufficient recycling capacities, and invest in appropriate recycling technologies.[1] The temporal analysis of material cycles is often based on a dynamic material flow analysis (MFA) approach. The development of stocks and flows are calculated based on inflow or stock data and the product lifetime or lifespan, these terms are used as synonyms in existing literature. An in-depth literature review on dynamic MFA methods of metals in general, including critical metals, is presented in Chapter 2 of this thesis. The main findings relevant for developing a basic model and handling metal dissipation as well as data uncertainty are presented in the following.

It is generally assumed that all processes except the use process transform material. Only within the use process, material is also stocked, which leads to the actual dynamics of the system. Most existing studies apply a 'top-down' approach to calculating stocks from inflow data and lifetime distributions. Due to more extensive data collection, only a few studies use a 'bottom-up' approach, where stocks are assembled from different product groups containing the material of interest. However, by investigating in detail the in-use stock, 'bottom-up' approaches can provide important insights on consumer behavior. This includes, among others, product lifetime, storage time and disposal pathways of products. Existing data regarding these variables are scarce and based on diverging definitions of lifetime as well as different temporal and regional scopes.

Dissipation, losses or emissions of metals to the environment have only in recent years been addressed from a resource point of view and data are still scarce. In the past, they were included in dynamic MFAs mainly focusing on heavy metal pollution, with dissipative flows calculated mainly from inflows and transfer coefficients or stocks and emission factors.

Input data of a dynamic MFA are often acquired from many different sources with varying data reliability. Uncertainty analyses or sensitivity analyses are therefore important to understand the effect of uncertain model input, but are not included in many studies. Probabilistic MFA, a very comprehensive approach to dealing with uncertainty, has not yet been applied to metals.

In many cases, before we are able to calculate stocks and flows of metals, the stock and flow dynamics of the product in which they are incorporated have to be understood. Various studies have shown discrepancies between high sales flows and low collection flows for EE or long phase-out periods of EE that is no longer sold.[31,43–48] These findings suggest that the lifetime of EE might be longer in reality than assumed in existing models. This could be due to reuse or storage that have not been taken into account. Another reason might be that there are other disposal pathways than collection, for example, export for reuse or recycling, waste incineration (via the municipal solid waste collection pathway), or illegal dumping.

Due to the lack of 'bottom-up' studies, the use phase can be viewed as a 'black box'. Only the sales flows and flows to formal collection schemes are usually measured. Information regarding the whereabouts of EE inside the 'black box', that is, lifetime, reuse, storage and disposal pathways diverting from formal collection, is virtually inexistent.

Stocks and flows in recycling processes are well known on the level of device input as well as base metals, plastics and precious metals in the resulting output fractions. Only a few studies have measured the content of critical metals in fractions after preprocessing.[49–51] Recyclers therefore often do not know in which fractions they transfer critical metals within their recycling processes. Due to various reasons, as stated in Chapter 1.2, critical metals are mostly dissipated to slags after incineration or smelting processes. In order to quantify the resulting sinks, for example in landfills, the stocks and flows of critical metals should be quantified from the time they have been first put on the market – incorporated in EE – until today.

The dilution or concentration of a substance in an MFA system due to recycling or dissipation can be measured with statistical entropy analysis (SEA). The SEA is a tailor-made evaluation method for MFA that can be directly applied to an MFA database.[52] It helps to visualize the metabolism of anthropogenic systems and thus better understand, for example, which processes are responsible for substance losses.[53] SEA is based on the information theory founded by Shannon, where the concept of entropy it is understood as a measure of the loss or gain of information about a system.[54] This so-called Shannon–entropy is used in statistics to measure the variance of a probability distribution. Formally, the thermodynamic entropy and the statistical entropy are identical, but there is no physical relationship between the two.[53] Rechberger and Graedel analyzed the contemporary copper cycle and various alternative scenarios with SEA.[53] Bai et al. use MFA and SEA to investigate substance flows in a lead smelting process. Both are static analyses. A SEA of a dynamic MFA of critical metals in the anthroposphere has not yet been conducted, but can serve as an indicator to assess the level of dilution and dissipation of critical metals during their anthropogenic life cycle and its change over time.

1.4 Research Questions and Objectives

If secondary raw material production should contribute to the mitigation of critical materials' supply risks, adequate databases regarding location, lifetime, disposal pathways, quantities, and qualities of emerging technologies are required. The existing data on critical metals are incomplete and often do not offer information on the temporal distribution as well as the uncertainty of their stocks and flows. Therefore, the **guiding research question** is the following:

What are the stocks, pathways, contents, and properties of critical metals in emerging end-user technologies (in particular, electronic equipment) within the anthroposphere and how can they be influenced in view of effective and efficient recycling systems?

Concerning stocks and their recycling potential, the specific research questions (RQ)s are:

- *RQ1: Where are the largest stocks of critical metals in the anthroposphere?*
- *RQ2: How suitable are these stocks for urban mining?*

As most critical metals are not recovered in the current recycling processes, the following specific research questions arise:

- *RQ3: Where are the current sinks of critical metals that are not recovered?*

- *RQ4: How are critical metals diluted in the anthroposphere and how does this change during a critical metals life cycle?*
- *RQ5: Where are possible starting points to improve the current recycling system?*

Due to scarce data availability and various methodological challenges, there is no straightforward approach to providing answers to these empirical questions. The research in this thesis is thus based on a twofold approach as illustrated in Figure 1.2: the empirical thread and the methodological thread.

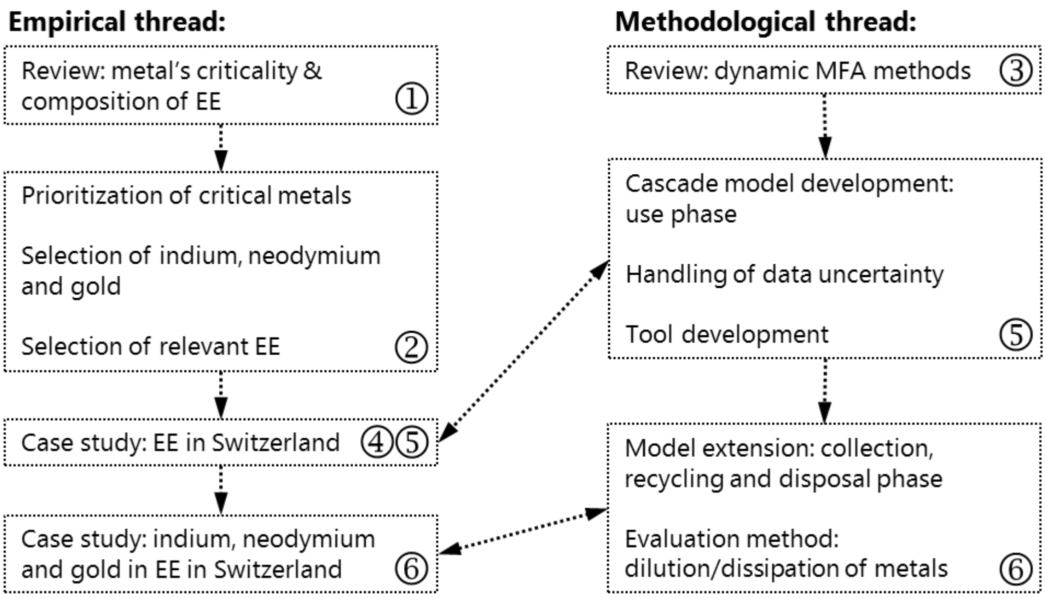

Figure 1.2: The twofold research approach of this thesis. The numbers indicate the chronological order as well as the related chapters in this thesis.

Within the empirical thread, we started with a literature review of metals' criticality and composition of EE, which resulted in the prioritization of critical metals and the selection of two relevant metals: indium and neodymium (Chapter 2). As a reference metal with already well-established recycling processes, we additionally included the precious metal gold. Due to its good recoverability and high economic value, gold is one of the most important economic drivers of the EE recycling system. Gold stocks and flows, therefore, serve as a reference and facilitate the interpretation of indium and neodymium stocks and flows in the context of today's recycling system. Subsequently, we selected the EE types where these metals are incorporated in significant quantities.

Within the methodological thread, a literature review of existing dynamic MFA methods builds the basis for developing a methodology that allows to dynamically model anthropogenic stocks and flows of critical metals and assessing the dilution and dissipation of critical metals during their anthropogenic life cycle (Chapter 3). A special focus is laid on the expression and discussion of associated uncertainties.

Although the methodology should be applicable for metals and emerging end-user technologies in general, it is developed based on the case of end-user EE in private Swiss households and applied to indium, neodymium and gold. Business use of EE in Switzerland differs considerably from privately used EE and was analyzed in a separate study based on the results of this thesis.[55] The case study of EE in private Swiss households comprises an extensive data collection based on the above-mentioned information gaps (Chapter 4) and a detailed dynamic MFA of the current use phase (Chapter 5) as well as collection, recycling, and disposal phase (Chapter 6). In addition, the current system is evaluated with SEA. The goal of the case study is to better understand the metabolism of the anthroposphere regarding critical metals connected to the use of EE, provide a basis to develop appropriate tools and alternatives how to manage recycling systems and encourage the recycling and reintegration of critical metals into anthropogenic material cycles.

As there is no adequate open source software tool for the modeling and simulation of dynamic MFA and SEA, another objective is the development of such a tool to facilitate future applications of the proposed methodology and similar dynamic MFAs.

1.5 Structure of this Thesis

This thesis is structured in seven chapters. Chapter 2 describes the prioritization and selection process to select relevant critical metals whose recovery from e-waste should be examined in detail, with the goal to further sharpen the focus of this thesis. The selection results in the two critical metals indium and neodymium. In addition, we selected the precious metal gold, as the recycling of gold is already well established and an important economic driver in the recycling system.

Chapters 3 to 5 are already published in scientific journals. Chapter 6 is submitted to a scientific journal. An overview of the research articles presented in this thesis is provided in Figure 1.4

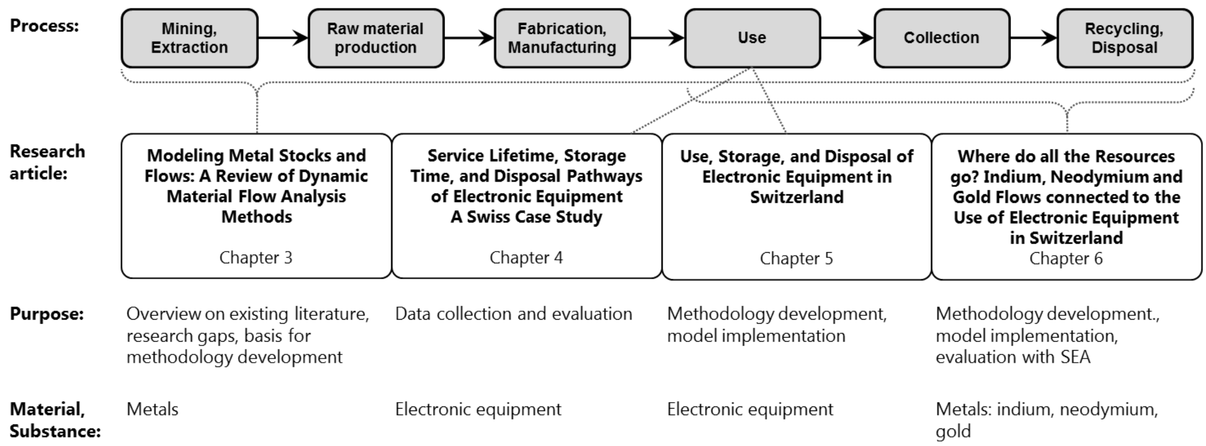

Figure 1.3: Overview of the research articles presented in this thesis

Chapter 3 provides a review of existing dynamic MFA methods. As for critical metals, dynamic MFAs are still scarce, the focus was broadened to metals in general to include more methodological approaches. Reviewed literature covers all processes in the life cycle of metals, from mining and extraction to recycling and disposal. The review evaluates general modeling approaches such as 'top-down' and 'bottom-up' methods, retrospective and prospective approaches and the related extrapolation methods. Further, literature on dissipation, lifetime modeling, spatial MFA and uncertainty is included. The literature review reveals the main research gaps and provides a basis for the methodology development.

Chapter 4 reveals the scarce data availability regarding service lifetime, storage time and disposal pathways of electronic equipment in the use phase. It then presents the results of a survey on the service lifetime, storage time, and disposal pathways of EE that was conducted between 2014 and 2016 with the goal to obtain detailed 'bottom-up' information as a data basis for the model development.

Chapter 5 presents the development of a dynamic MFA approach that identifies the past and current in-use stocks and storage stocks of EE and quantifies in detail the flows between and from the use, storage, and disposal phases. Based on the survey data, this model is able to differentiate between new and (re)used devices. The model is implemented in an open source software tool we developed in the programming language Python 3. The tool applies a probabilistic approach that allows to systematically deal with data uncertainty.

In Chapter 6, the flows of the critical metals indium and neodymium and the precious metal gold, incorporated in EE, are derived from the model presented in chapter 5. The detailed model of the use phase is extended with the collection, recycling, and disposal phase. This allows to track critical metals from their entry into Switzerland (as components of new devices) until their disposal in landfills or dissipation to the environment, identifying the most important sinks and comparing them to current in-use and storage stocks in order to estimate the recycling potential for the different metals. With SEA, we further illustrate the dilution or concentration of each metal during its route through the current system.

Finally, Chapter 7 first provides a discussion of this thesis as a whole, mainly in relation to the methodological thread. In the second part, the contributions and conclusions regarding the methodology developed are presented and the key findings concerning the empirical research questions are recapitulated. The third and last part provides an outlook on future research.

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Chapter 2

Prioritization of Critical Metals in Electronic Equipment

2.1 Introduction

Electronic equipment (EE) contains many different critical metals in various contents. Some metals are concentrated in a few single components while others are spread across many components within a product. With the goal to further sharpen the focus of this thesis, we limited the case study of EE in Switzerland to two critical metals which are highly relevant with respect to EE and have a high recovery potential in the future.

The selection of the relevant critical metals based on a screening and prioritization process that we developed within the framework of the e-Recmet project,[1] commissioned by the Swiss Federal Office for the Environment and Swico. The screening and prioritization process includes various criteria and various stages or 'filter' according to Figure 2.1.

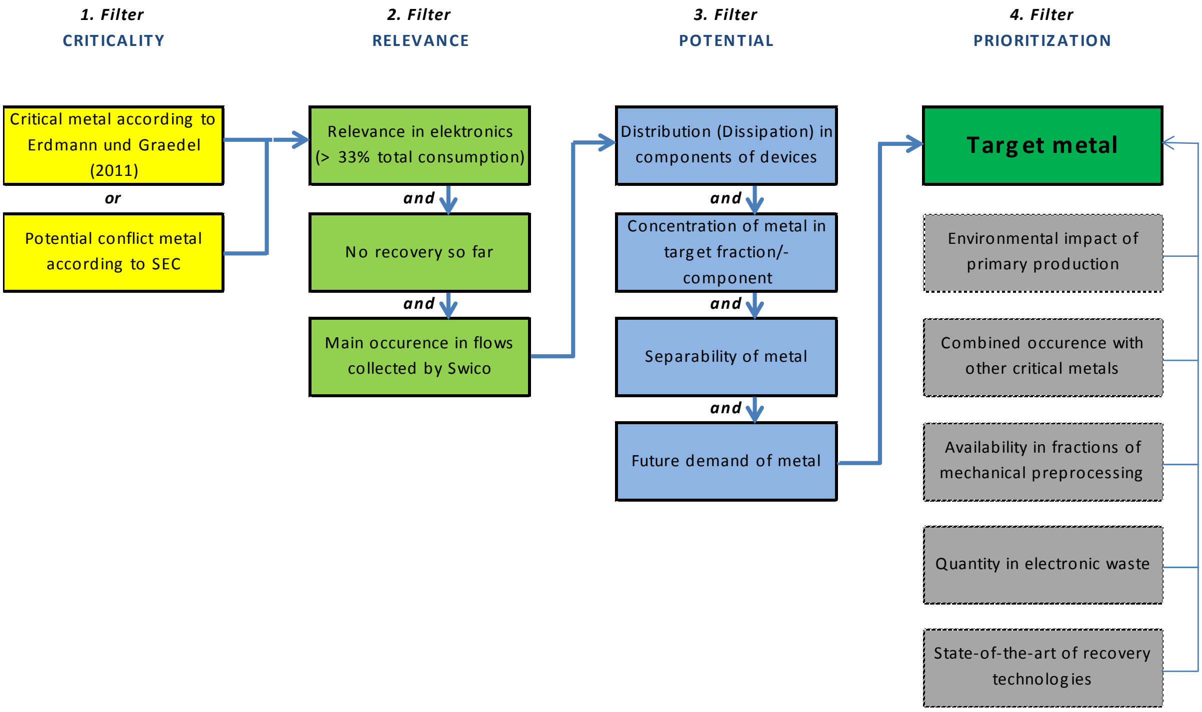

Figure 2.1: Scheme for the selection of relevant critical metals in electronic waste

In the following, we present the different stages with its intermediate results, the final results of the selection process as well as more details about the two selected critical metals.

2.2 Screening and Prioritization Process

2.2.1 Criticality

In a first stage, the spectrum of metals is limited to those that are viewed as critical regarding supply risks and vulnerable to a potential supply disruption. Metals that are mentioned in at least three of the seven studies reviewed by Erdmann and Graedel[2] and that are evaluated as critical with a frequency of at least 30% are regarded as critical metals. In addition, we took into account those metals that could possibly origin from so-called 'conflict minerals' that are mined in conflict regions such as the Democratic Republic of Congo.[3]

The resulting selection includes 37 critical metals, whereof 14 belong to the REE. Metals possibly originating from conflict minerals are tantalum, tungsten, zinc and gold (see Table 2.1).[3]

Table 2.1: Critical metals according to Erdmann and Graedel[2] and conflict metals (*)[3]

Antimony	**Sb**	Holmium	**Ho**	Osmium	**Os**	Thulium	**Tm**
Beryllium	**Be**	Indium	**In**	Palladium	**Pd**	Tungsten*	**W**
Cerium	**Ce**	Iridium	**Ir**	Platinum	**Pt**	Ytterbium	**Yb**
Dysprosium	**Dy**	Cobalt	**Co**	Praseodymium	**Pr**	Yttrium	**Y**
Erbium	**Er**	Lanthanum	**La**	Rhodium	**Rh**	Tin	**Sn**
Europium	**Eu**	Lithium	**Li**	Ruthenium	**Ru**	Zinc*	**Zn**
Gadolinium	**Gd**	Lutetium	**Lu**	Samarium	**Sm**	Zirconium	**Zr**
Gallium	**Ga**	Magnesium	**Mg**	Scandium	**Sc**		
Germanium	**Ge**	Neodymium	**Nd**	Tantalum*	**Ta**		
Gold*	**Au**	Niobium	**Nb**	Terbium	**Tb**		

(grey = REE)

2.2.2 Relevance

In a second stage, critical metals resulting after the first filter are further limited regarding their relevance with respect to EE. Relevant are metals that are predominantly used in electronic devices. The limit is set at a minimum of one-third of the world's annual production of the metal is used in electronic applications. The metals should not have been recovered from WEEE so far. In addition, they were restricted to those occurring in applications collected by Swico (WEEE category 3 and 4).[4]

The 37 critical metals are limited to 13 critical metals considered as relevant with respect to EE, as listed in Table 2.2. Reasons for the exclusion after the 'relevance' filter are listed in Table 2.3.

Table 2.2: Relevant critical metals

Antimony	**Sb**	Gallium	**Ga**	Neodymium	**Nd**	Yttrium	**Y**
Beryllium	**Be**	Holmium	**Ho**	Praseodymium	**Pr**		
Dysprosium	**Dy**	Indium	**In**	Ruthenium	**Ru**		
Gadolinium	**Gd**	Lanthanum	**La**	Tantalum	**Ta**		

(grey = REE)

Table 2.3: Reasons for exclusion after 'relevance' filter

Cerium	**Ce**	< 33% of total consumption in electronics
Erbium	**Er**	Main occurrence in applications not collected by Swico
Europium	**Eu**	Main occurrence in applications not collected by Swico
Germanium	**Ge**	Main occurrence in applications not collected by Swico
Gold	**Au**	< 33% of total consumption in electronics
Iridium	**Ir**	< 33% of total consumption in electronics
Lithium	**Li**	< 33% of total consumption in electronics
Cobalt	**Co**	Recovery possible (Umicore)
Lutetium	**Lu**	Main occurrence in applications not collected by Swico
Magnesium	**Mg**	< 33% of total consumption in electronics
Niobium	**Nb**	< 33% of total consumption in electronics
Osmium	**Os**	< 33% of total consumption in electronics
Palladium	**Pd**	< 33% of total consumption in electronics
Platinum	**Pt**	< 33% of total consumption in electronics
Rhodium	**Rh**	Main occurrence in applications not collected by Swico
Samarium	**Sm**	Main occurrence in applications not collected by Swico
Scandium	**Sc**	< 33% of total consumption in electronics
Terbium	**Tb**	Main occurrence in applications not collected by Swico
Thulium	**Tm**	Main occurrence in applications not collected by Swico
Tungsten	**W**	< 33% of total consumption in electronics
Ytterbium	**Yb**	Main occurrence in applications not collected by Swico
Tin	**Sn**	< 33% of total consumption in electronics
Zinc	**Zn**	< 33% of total consumption in electronics
Zirconium	**Zr**	< 33% of total consumption in electronics

(grey = REE)

2.2.3 Potential

The critical and relevant metals for a future recovery are integrated into components of EE in different ways. The metal should be concentrated in a few single components instead of dissipatedly distributed across different components. They should also occur in contents as high as possible, with the content in a component corresponding to at least the (present / foreseeable future) content/cut-off grade in the ore. The metal containing components should be accessible in the context of a manual or mechanical decomposition, that is, they are not incorporated into a complex matrix, for example as an additive. Finally, their significance should remain high in technical applications in the future.

This third stage results in 8 of the 13 relevant critical metals that are considered as target metals for the prioritization (Table 2.4). Reasons for the exclusion after the 'potential' filter are listed in Table 2.5.

Table 2.4: Target critical metals for prioritization

Dysprosium	**Dy**	Neodymium	**Nd**
Gadolinium	**Gd**	Praseodymium	**Pr**
Holmium	**Ho**	Ruthenium	**Ru**
Indium	**In**	Yttrium	**Y**

(grey = REE)

Table 2.5: Reasons for exclusion after 'potential' filter

		Occurrence	Separability/future significance
Antimony	Sb	Plastics	Distributed to many components, decreasing significance
Beryllium	Be	Copper alloys	Distributed to many components, decreasing significance
Gallium	Ga	Printed wiring boards	Poor separability
Lanthanum	La	Printed wiring boards	Poor separability
Tantalum	Ta	Capacitor	Poor separability

(grey = REE)

2.2.4 Prioritization

The remaining metals are prioritized in the last stage. In contrast to the preceding steps, it is not necessary to consider all criteria. The following criteria were taken into account:

- *Environmental relevance for primary production:* in the case of a high environmental impact assessed by the cumulative energy demand during primary production, the corresponding critical metal is highly prioritized.
- *Combined occurrence with other critical metals:* association with other critical metals in their applications results in increased priority.
- *Presence in fractions of mechanical processing:* critical metals detected in the output fractions indicate that these metals are actually present in measurable contents and are to be followed as a matter of priority.
- *The amount in electronic waste:* If critical metals are found in flows collected by Swico in high quantities, the priority is increased.
- *State of the art for recovery:* If recovery processes are at the very least available as a pilot plant for the corresponding critical metal, the priority increased.

The prioritization of the 8 target critical metals (see Table 2.6) results in **indium** and **neodymium** as the most suitable for further investigations:

➔ Both metals were detected in the fractions resulting from mechanical processing.[5]

➔ The amounts of neodymium and indium, which could be recovered in the Swico channel, are significantly larger in both cases than the amounts of the other target metals.

➔ Both metals are associated with other critical metals, which in the case of manual decomposition or mechanical processing results in a possible additional benefit.

➔ The criterion of environmental impact is not selective enough at this stage. However, it is important to note that detailed databases are missing and approximation considerations are necessary.[6]

➔ In both cases, the most suitable method of final treatment is still in development. However, various research projects for the recovery of indium and neodymium exist.

Table 2.6: Prioritization of 8 target metals

Metal		**Environmental impact CED (MJ/t)** [6]	**Occurrence with other critical metals**	**Results mechanical preprocess-sing** [5]	**Quantities Swico** (first estimation)	**State-of-the-art recovery technology**	**Prio-rity**
Dys-prosium	**Dy**	1'668'000	DyNd, DyFe, DyBr	*Not detected*	16.5 kg (Laptops)	Pyrometal., laboratory (Japan)	II
Gado-linium	**Gd**	1'668'000	GdGe, GdBr, EuGdO, GdLuF	Finest fraction	0.2 kg (only Laptops)	-	III
Holmium	**Ho**	1'668'000	AlDyHo	*Not detected*	-	-	III
Indium	**In**	1'981'000	GaInSn	Various fractions	Ca. 65 kg	Hydrometal., pyrometal. production scrap (Umicore)	I
Neo-dymium	**Nd**	1'668'000	NdFeB, NdYDy, NdPrDy	Finest fraction	Ca. 3'100 kg	Pyrometal., pilot plant (Japan)	I
Praseo-dymium	**Pr**	1'668'000	PrNd, PrMg	Finest fraction	Ca. 75 kg (Laptops)	Batteries (Umicore/ Rhodia)	II
Ruthe-nium	**Ru**	-	RuPt, RuP, RuIr, LnRu	Metal fraction (very low conc.)	-	Industrial scale	III
Yttrium	**Y**	-	GdY, YMg	Finest fraction	Ca. 0.5 kg (Laptops)	Industrial scale	III

(grey = REE)

2.3 Indium

Indium is a silvery white and very soft metal. It possesses a high ductility and easy mallea-bility. As alloying component, it increases already in small concentrations hardness and cor-rosion resistance.[7]

The content of indium in the earth crust lies between 0.05 and 0.25 ppm.[7–11] Indium is most commonly associated with lead, zinc, copper, and tin. The indium content in such ores ranges from 1 ppm to 100 ppm, with indium being extracted as a by-product.[12] Almost the total indium production worldwide origins from treatment and smelting residues of zinc extraction as well as the recycling of dust and gases produced by the melting of zinc.[7]

The most important field of application of indium is indium tin oxide (ITO) in flat screen displays and photovoltaics. The compound is formed from 90% In_2O_3 and 10% SnO_2, which results in a mass fraction of 78% indium in the ITO.[17] ITO is conductive as a metal and at the same time transparent since visible light with a wavelength of 0.4 to 0.8 µm is not reflected. Another advantage is the heat-resisting property of ITO. All these properties make ITO the ideal material for the production of thin-film electrodes for flat-panel displays (FPDs). According to recent investigations, the indium content in LCD modules varies highly, from 56 mg In/m^2 in smartphones to 931 mg In/m^2 in FPD TVs.[1,6,13–16] Figure 2.2 shows manually disassembled LCD panels.

Figure 2.2: LCD panels resulting from manual dismantling[1]

The accessibility of indium in EE is best ensured by manual disassembly, which on the whole leads to a better exploitation of indium, and to considerably lower total costs than mechanical preliminary treatment.[1]

The amounts of indium used in the field of photovoltaics are still low compared to the application in FPDs. Indium is applied as a thin film coating on cadmium telluride and copper indium gallium selenid solar cells.[17] Indium is also a component of light-emitting diodes (LED), for example, as aluminum-indium-gallium-phosphide and as indium-gallium-nitrides with different color properties. Other fields of application include dental medicine, high-temperature thermometers, control rods in nuclear reactors, seals and coatings for improving the resistance of the coated materials, special batteries or infrared reflectors. According to Böni et al.,[1] indium in control rods in nuclear reactors forms the largest indium in-use stock in Switzerland. Due to radioactive radiation, however, it is not suitable for recycling. Therefore EE currently forms the most relevant indium stock in Switzerland.

According to USGS,[12] 755 tonnes of indium were refined in 2015, of which 370 tonnes origin from China and 150 tonnes from Korea. Both LCD modules for FPDs and thin film photovoltaics are regarded as technologies with high growth potential. The worldwide indium demand is expected to increase from around 230 tonnes in 2013 to 360 tonnes in 2035. Marschweider-Weidemann et al. predicts still a large supply surplus in 2035, while the ad hoc group of the EC assumes a small supply deficit up to 2020.[7,18]

2.4 Neodymium

Neodymium belongs to the light REE. In elementary form, it is a soft silvery, reactive and ignoble metal with a strong paramagnetism.[7]

REE occur only together in nature and thus are extracted together. Most REE are relatively abundant in the earth crust. The content of neodymium in the earth crust lies between 33 ppm and 42 ppm.[8,9,11] However, REE are rarely concentrated enough for economically viable extraction. In most deposits, lanthanum, cerium, praseodymium and neodymium form over 90% of the total occurrence of REE. [7]

Neodymium is a REE used in small permanent magnets since the 1990s, of which between 8% and 35% are applied in hard disk drives (HDDs), optical drives and loudspeakers in computers, loudspeakers and vibration alarms in conventional mobile phones and smartphones as well as loudspeakers in headphones.[1,19] Neodymium is also found in small quantities in printed wiring boards (PWBs).[5,16,20] The exact source of the detected neodymium in PWBs is, however, unknown.

Until today, there is no systematic manual disassembly of hard disks and loudspeakers up to the level of magnets. Hard disks are often removed from computers, but they are not further decomposed. The neodymium content in magnets ranges from 8 to 31%,[1,21–24] with neodymium partly associated with praseodymium and dysprosium. Figure 2.3 depicts a hard disk with neodymium-iron-boron magnets.

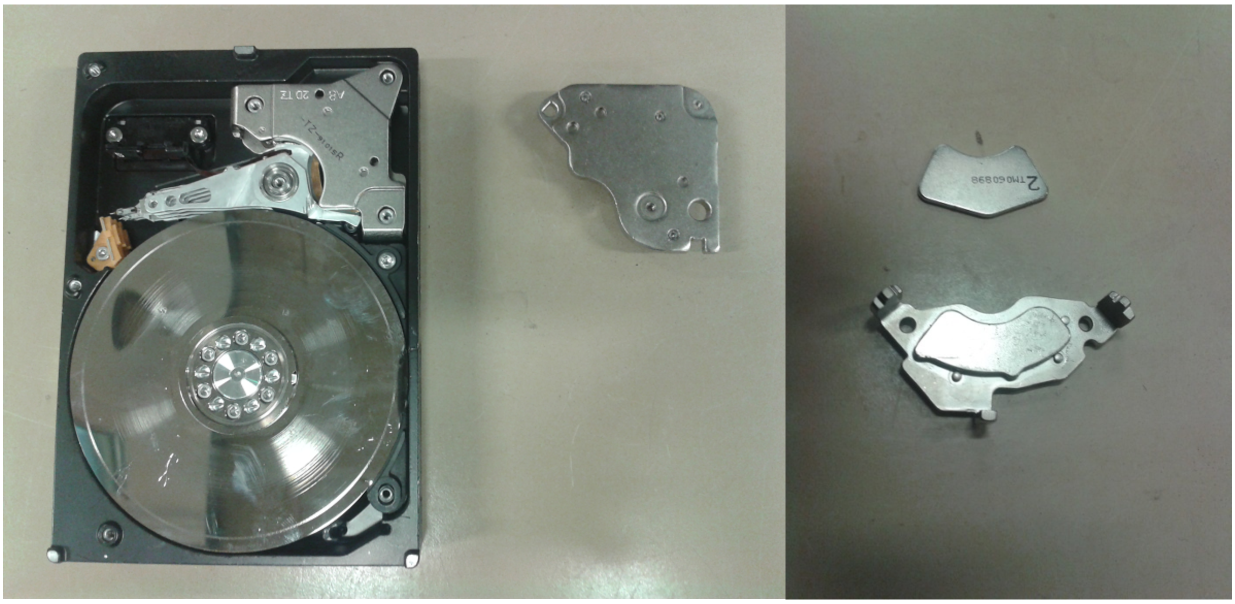

Figure 2.3: Hard disk with neodymium-iron-boron magnets[1]

The most important remaining applications of neodymium are also permanent magnets. These are used in cars, in electric two-wheelers, in wind turbines, in elevators and air conditioning systems, in mechanical sorting and processing systems and in magnetic resonance tomography. Further, neodymium is used in nickel metal hydride (NiMH) batteries, in various alloys, as an additive in glasses and ceramics and in lasers. According to Böni et al.,[1] neodymium in EE is present in much larger quantities both in sold products and the in-use stock, compared to other applications. EE are therefore the most relevant input and stock of neodymium in Switzerland.

The production of neodymium in 2013 is estimated at 29 000 tonnes. Wind turbines, car motors, and high-performance permanent magnets are regarded as technologies with highest growth potential. The demand is thus expected to increase to 62 000 tonnes in 2035, which implies 174% of the production of permanent magnets in 2013.[7]

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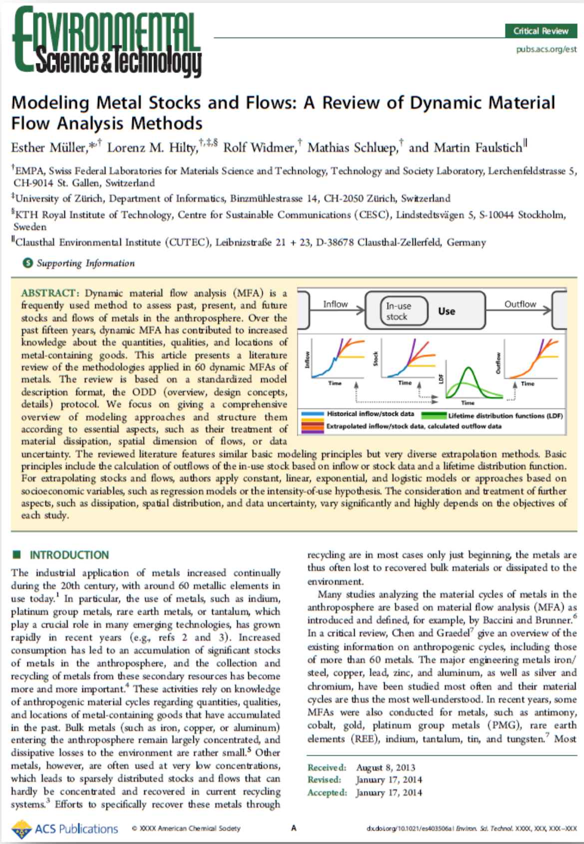
Chapter 3

Modeling Metal Stocks and Flows: A Review of Dynamic Material Flow Analysis Methods

Esther Müller, Lorenz M. Hilty, Rolf Widmer, Mathias Schluep, Martin Faulstich



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Modeling Metal Stocks and Flows: A Review of Dynamic Material Flow Analysis Methods

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Supporting Information

ABSTRACT: Dynamic material flow analysis (MFA) is a frequently used method to assess past, present, and future stocks and flows of metals in the anthroposphere. Over the past fifteen years, dynamic MFA has contributed to increased knowledge about the quantities, qualities, and locations of metal-containing goods. This article presents a literature review of the methodologies applied in 60 dynamic MFAs of metals. The review is based on a standardized model description format, the ODD (overview, design concepts, details) protocol. We focus on giving a comprehensive overview of modeling approaches and structure them according to essential aspects, such as their treatment of material dissipation, spatial dimension of flows, or data uncertainty. The reviewed literature features similar basic modeling principles but very diverse extrapolation methods. Basic principles include the calculation of outflows of the in-use stock based on inflow or stock data and a lifetime distribution function. For extrapolating stocks and flows, authors apply constant, linear, exponential, and logistic models or approaches based on socioeconomic variables, such as regression models or the intensity-of-use hypothesis. The consideration and treatment of further aspects, such as dissipation, spatial distribution, and data uncertainty, vary significantly and highly depends on the objectives of each study.

■ INTRODUCTION

The industrial application of metals increased continually during the 20th century, with around 60 metallic elements in use today.[1] In particular, the use of metals, such as indium, platinum group metals, rare earth metals, or tantalum, which play a crucial role in many emerging technologies, has grown rapidly in recent years (e.g., refs 2 and 3). Increased consumption has led to an accumulation of significant stocks of metals in the anthroposphere, and the collection and recycling of metals from these secondary resources has become more and more important.[4] These activities rely on knowledge of anthropogenic material cycles regarding quantities, qualities, and locations of metal-containing goods that have accumulated in the past. Bulk metals (such as iron, copper, or aluminum) entering the anthroposphere remain largely concentrated, and dissipative losses to the environment are rather small.[5] Other metals, however, are often used at very low concentrations, which leads to sparsely distributed stocks and flows that can hardly be concentrated and recovered in current recycling systems.[3] Efforts to specifically recover these metals through recycling are in most cases only just beginning, the metals are thus often lost to recovered bulk materials or dissipated to the environment.

Many studies analyzing the material cycles of metals in the anthroposphere are based on material flow analysis (MFA) as introduced and defined, for example, by Baccini and Brunner.[6] In a critical review, Chen and Graedel[7] give an overview of the existing information on anthropogenic cycles, including those of more than 60 metals. The major engineering metals iron/steel, copper, lead, zinc, and aluminum, as well as silver and chromium, have been studied most often and their material cycles are thus the most well-understood. In recent years, some MFAs were also conducted for metals, such as antimony, cobalt, gold, platinum group metals (PMG), rare earth elements (REE), indium, tantalum, tin, and tungsten.[7] Most

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Abstract

Dynamic material flow analysis (MFA) is a frequently used method to assess past, present, and future stocks and flows of metals in the anthroposphere. Over the past fifteen years, dynamic MFA has contributed to increased knowledge about the quantities, qualities, and locations of metal-containing goods. This article presents a literature review of the methodologies applied in 50 dynamic MFAs of metals. The review is based on a standardized model description format, the ODD (overview, design concepts, details) protocol. We focus on giving a comprehensive overview of modeling approaches and structure them according to essential aspects, such as their treatment of material dissipation, spatial dimension of flows, or data uncertainty. The reviewed literature features similar basic modeling principles but very diverse extrapolation methods. Basic principles include the calculation of outflows of the in-use stock based on inflow or stock data and a lifetime distribution function. For extrapolating stocks and flows, authors apply constant, linear, exponential, and logistic models or approaches based on socioeconomic variables, such as regression models or the intensity-of-use hypothesis. The consideration and treatment of further aspects, such as dissipation, spatial distribution, and data uncertainty, vary significantly and highly depends on the objectives of each study.

Keywords: material flow analysis (MFA), dissipation, data uncertainty, spatial dimension, metals

3.1 Introduction

The industrial application of metals increased continually during the 20th century, with around 60 metallic elements in use today.[1] In particular, the use of metals, such as indium, platinum group metals, rare earth metals, or tantalum, which play a crucial role in many emerging technologies, has grown rapidly in recent years (e.g refs 2 and 3). Increased consumption has led to an accumulation of significant stocks of metals in the anthropo-sphere, and the collection and recycling of metals from these secondary resources has become more and more important.[4] These activities rely on knowledge of anthropogenic material cycles regarding quantities, qualities, and locations of metal-containing goods that have accumulated in the past. Bulk metals (such as iron, copper, or aluminum) entering the anthroposphere remain largely concentrated, and dissipative losses to the environment are rather small.[5] Other metals, however, are often used at very low contents, which leads to sparsely distributed stocks and flows that can hardly be concentrated and recovered in current recycling systems.[3] Efforts to specifically recover these metals through recycling are in most cases only just beginning, the metals are thus often lost to recovered bulk materials or dissipated to the environment.

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In this article, we present a Critical Review of the methodologies applied in the literature on dynamic MFAs of metals. We focus on giving a comprehensive selection of modeling approaches that can be used as a basis for future studies on the dynamics of metal cycles. We further identify distinguishing aspects of MFAs, such as the treatment of material dissipation, the spatial dimension of flows, or data uncertainty. The review covers the published literature in English on dynamic MFAs of metals.

One German reference[12] is included because it provides background data used by Bader et al.[13] Literature with only rudimentary or incomplete model descriptions is not included. We thus compile information from 60 studies published between 1999 and 2013 and covering a total of 34 metallic elements.

3.2 Method

The review is structured based on the standardized description ODD (overview, design concepts, details) protocol that was originally developed for the documentation of individual-based and agent-based models.[14,15] Although the studies we review use a fundamentally different type of models, ODD has proven to be useful for structuring them. The main objective of the ODD protocol is to provide a complete, understandable, and reproducible description of the models to make their complexity manageable for the human reader.[14,15] MFAs are in general less complex than agent-based models, we therefore simplified the ODD protocol slightly to adapt it to the field of MFA. The adapted structure is provided in Table 3.1. Each element of the protocol is further specified with one or more questions.

The protocol is grouped into three parts. The first part gives an overview of the study, including the purpose, the scope, the system boundaries, and the structure of the MFA. The second part describes the generic concepts and modeling approaches of the research. The third part provides the details necessary to ensure the reproducibility of the study.

MFA-specific terms in the ODD are used as defined by Brunner and Rechberger.[16] In the following, we will further clarify some terms: static versus dynamic MFA, top-down versus bottom-up approach to MFA, prospective versus retrospective MFA, endogenous versus exogenous model variable, and material dissipation.

An MFA is *static* if it describes a “snapshot” of a system in time. An MFA is *dynamic* if it describes the behavior of a system over a time interval.[7]

The material stock of a process can be measured by two different methods. The first method, usually referred to as the *top-down approach*, derives the stock from the net flow: the difference between inflows (consumption) and outflows (discard). The second method, the *bottom-up approach,* directly estimates the stock by summing up the material in question present within the system boundary at a certain time.[17] Most authors define stock as the in-use stock and do not include “hibernating” materials, that is, those that have been retired and remain somewhere in storage. Hibernating or obsolete stock is explicitly included only by Daigo et al.[18]

Table 3.1: Elements of the ODD (Overview, Design concepts, Details) protocol for MFA

Overview	**Purpose**	What is the purpose and general framework of the model?
	Materials (goods, substances)	What materials (goods/substances) are included? Are materials further divided into material categories (and subcategories)?
	Processes	What processes are included? Do they transform, transport, or store materials? Are processes further divided into process categories (and subcategories)?
	Spatial and temporal scale and extent	What is the spatial and temporal scale and extent of the study?

	System overview	What is the structure of the system regarding processes, stocks, and flows?
Design concepts	**Basic principles**	Static or dynamic, top-down or bottom-up, retrospective or prospective?
	Static or dynamic modeling approaches	For example, how are stocks and flows modeled? What are the extrapolation methods for exogenous variables?
	Dissipation	How is dissipation accounted for?
	Spatial dimension	How is the spatial distribution of stocks and flows accounted for?
	Uncertainty	How is data and model uncertainty accounted for?
Details	**Initial condition**	How is the initial state (e.g., the initial stocks and flows) of the model set?
	Model input data	What data is used as input to the model?
	Model output data	What data is generated as model output?
	Evaluation	What methods (e.g., for data aggregation and visualization) are used to evaluate the results?
	Detailed model description	What, in detail, is the formal description (e.g., equations) of the system and what are the algorithms (e.g., solution procedures) used for the calculations? What are exogenous and endogenous model variables? What are the model parameters, their dimensions, and reference values?

An MFA can be either *retrospective*, analyzing past stocks and flows based on historical data, or *prospective*, looking into the future using data extrapolation, or a combination of both approaches.

An *endogenous model variable* is a variable whose value is determined by one of the functional relationships in the model, for example, the outflow of a given product as waste, determined by the inflow and lifetime of that product. An *exogenous model variable* is an independent variable that affects endogenous model variables without being affected by any of them. It represents a quantity that exists outside the chosen system boundary. For simulation, an exogenous variable needs input data, for example, data of the inflow of a certain product or socioeconomic data such as time series of the gross domestic product (GDP).

According to Ayres,[19] who was one of the first to use the concept of *dissipation* associated with material flows,[5] “there are only two possible long-run fates for materials—dissipative loss and recycling or reuse.” He argues that materials are recycled or reused if economically and technologically feasible, otherwise they are eventually dissipated. In Ayres et al.,[20] he later specifies four categories of metal stocks: long-lived goods in use, short-lived goods in use, landfill and identifiable mine waste dumps, and finally metals that have been irrecoverably dissipated into soil, groundwater, or surface water. In this definition, only the last category accounts for material dissipation.

3.3 Results

3.3.1 Purpose

Most of the reviewed studies (43 of 60) aim at understanding the pathways of metals in the anthroposphere, the magnitudes of their stocks and flows, and how they evolve as a function of time. This involves quantifying and visualizing the dynamics of relevant stocks and flows of metals and their use in specific product groups or end-use sectors. Additional purposes include to specifically examine the recycling potential of metals, including recycling efficiency[21,22] and future recycling flows,[23–25] to evaluate future scenarios of resource availability,[20,26–28] to assess changes in environmental impacts related to changes in material flows,[29–31] and to compare different methodological approaches.[11,32–36]

3.3.2 Materials

The studies assessed cover 34 metallic elements as summarized in Table 3.2, with iron, aluminum, and copper being the most frequently investigated elements. Dynamic MFAs are still lacking for more than 30 metals.[7]

Table 3.2: Metallic elements covered in the reviewed literature

Element (Alloy)	Covered in # of studies
Fe/steel	17
Al	12
Cu	11
Pb	6
Zn	4
Cr, Ni	3
Cd, Ce, Dy, Eu, Gd, In, La, Nd, Pt, Pr, Sm, Te, Tm, Y	2
Ag, Co, Er, Hg, Ho, Lu, Pd, Rh, Se, Sn, Tb, W, Yb,	1

Thirty-four studies consider metal use in some or all of the following end-use sector categories (sometimes disaggregated into subcategories): transportation, buildings and construction, infrastructure and telecommunication, machinery, electric appliances, and consumer goods, and containers and packaging.

Instead of end-use categories, 20 studies assess metal use in products. Among these, 13 studies cover the most relevant products containing the investigated metal and 7 studies cover the metal use only of individual products: CRT screens,[33] vehicles,[37,38] catalytic converters in automobiles[31], photovoltaic systems,[23,27] and products containing indium tin oxide (ITO).[39] Three further studies include both end-use sectors and products,[24,34,40] and three studies do not categorize metal use.[20,41,42]

The metal content of stocks and flows is calculated either directly by computing metal stocks and flows from input data or indirectly by computing material stocks of end-use sectors or products and then calculating metal quantities based on the assumed metal share in an end-use sector or content in a product. In the indirect case, the metal share or content is usually considered time-variant.

3.3.3 Processes

The processes most commonly included in MFAs of metals cover the whole life cycle of a metal, from primary mining to raw material production to product manufacturing to use and finally waste management. In most of the models, the use phase is the only process that stores materials, while the other phases transform them without accumulating stocks. Potential stocks outside the use phase are neglected because they are assumed to be stationary over the smallest time interval considered, usually the period of one year (this assumption may be challenged by stocks of very valuable metals created for speculation). Additional processes such as landfill, environment, or other repositories are often included to illustrate the final sink of the assessed metals. Table A.1 in the Supporting Information (SI) gives an overview of the processes covered by the studies.

3.3.4 Spatial and Temporal Scale and Extent

The spatial extent ranges from urban to global system boundaries, though most literature (38 studies) assesses metal stocks and flows of a specific country. Figure A.1 in the SI shows the percentage distribution of the reviewed studies by spatial extent. Regional or national studies exist mainly for industrial countries. Global studies often extrapolate data from industrial countries because of the lack of domestic data in developing countries.[7]

Thirty-one studies model both retrospective and prospective flows, examining temporal extents in the time frame from 1700 to 2100. Twenty-six studies analyze only past flows and three studies include only prospective flows. The temporal scale of input and output data is usually one year. Hence, discrete-time calculations are also carried out with time steps of one year.

3.3.5 System Overview

The structure of most studies is based on a generic system with processes graphically represented in a sequence or a loop.[7] Although some studies include subprocesses containing more details than the top-level processes listed, the general structure remains the same (see Figure 3.1). The topology of flows between the processes depends on the purpose, the characteristics of a considered metal (e.g., potential toxicity), and the complexity of the study; for example, some studies consider metal emissions of all processes (e.g., refs 20 and 33), some only of the use phase (e.g., refs 13 and 43), and other studies do not take emissions into account at all (e.g., refs 11, 41, and 42).

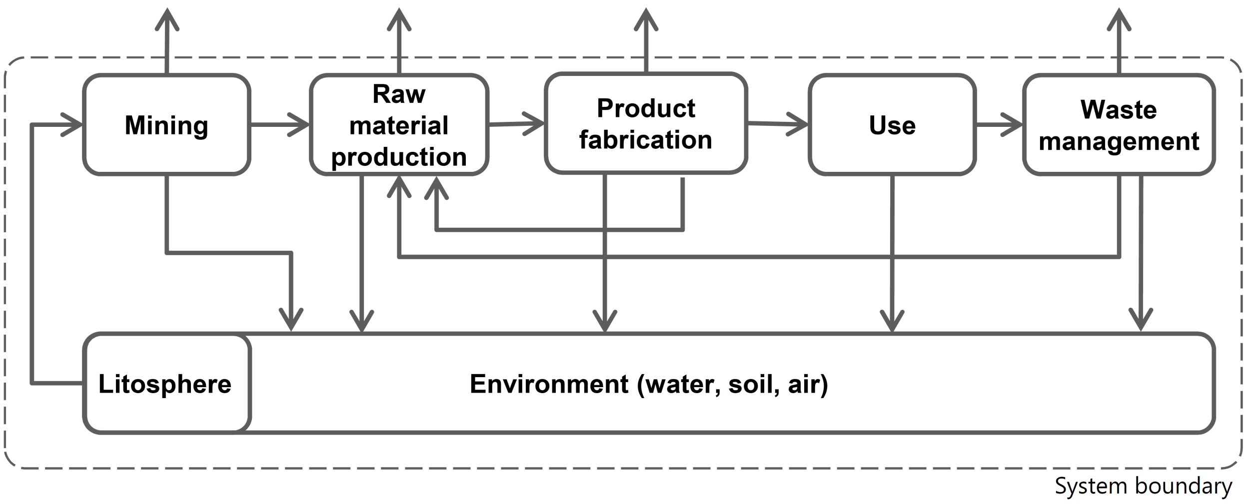

Figure **3.1**: System overview of a generic dynamic material flow model of metals**.**

3.3.6 Basic Principles

The dynamic MFAs of metals generally assume that in the production, manufacturing, and waste management processes, no material is stored or the net flow during the sample time is zero, that is, that this part of the system can be treated as static. Hence, the dynamic modeling approaches focus on the use phase (which has nonzero net flows) and the resulting in-use stock changes. Stocks and flows are often modeled as time series with a constant sampling rate T, that is, f[n] = f (nT), typically with T = 1 year.

The in-use stocks are quantified by one of the following two methods. The top-down approach derives the in-use stock S from the net flow by using the balance of masses as shown in eqs (1), (1a), and (1b).[17]

$$dS(t) = \big(inflow(t) - outflow(t)\big) \cdot dt = netflow(t) \cdot dt \tag{1}$$

$$S[n] = (inflow[n] - outflow[n]) \cdot T + S[n-1] \tag{1a}$$

$$S[N] = S[0] + T \cdot \sum_{n=1}^{N} (inflow[n] - outflow[n]) \tag{1b}$$

The second method, referred to as the bottom-up approach, derives the in-use stock S[n] at a time n by summing all the metals contents c_i in their respective products or end-use sectors P_i according to eq (2)[17]

$$S[n] = \sum_{i=1}^{I} P_i[n] \cdot c_i[n] \tag{2}$$

where I is the total number of products or end-use sectors considered. To construct time series of in-use stock, S[n] is computed for every requested year n. If required, net flow can be calculated by introducing eq (2) into eq (1a). Almost 90% of the reviewed literature applies the top-down approach and 10% the bottom-up approach.

For inflows, historical data (e.g., on long-term consumption) is often accessible, but outflows are rarely measured. Most authors use and adapt methods developed in the field of system reliability to quantify the outflow of discarded items. The frequently used quantitative measures to describe this process[44] are listed in the SI.

Most authors choose to quantify outflows by assigning lifetime distribution functions to specific products or end-use sectors, with the relationship between inflows and outflows corresponding to a convolution (eq (3) with ’*’ denoting the convolution; this approach is also called the residence time model or population balance model[11,45–47]). Since it is rarely possible to solve this convolution analytically, it is integrated numerically according to eq (4).

$$Outflow(t) = (Inflow * f)(t) = \int_{-\infty}^{\infty} Inflow(t-u) \cdot f(u)du \tag{3}$$

$$Outflow[n] = \sum_{m=-\infty}^{\infty} Inflow[n-m] \cdot f[m] \quad (4)$$

where f(t) and f[m] are the probability densities of the lifetime distribution function for the continuous and the time discrete case, respectively.

The lifetime distribution functions most frequently used are the Dirac delta distribution, which represents average and constant lifetime, and the Weibull distribution. Other distributions used are the normal, log-normal, beta, and gamma distributions. In 23 of the reviewed studies, authors use two or more distributions. They either choose this approach according to available lifetime data for their considered products or end-use sectors (e.g., refs 18, 48, and 49) or to explore the effect of applying different lifetime distributions on the model output.[11,21,50–55] Melo,[11] for example, uses the delta, Weibull, normal, and beta distributions for modeling scrap flows. He concludes that by applying the delta distribution, scrap flows are highly influenced by fluctuations of the inflows, which can lead to significant under- or overestimations of the scrap potential. All of the other distributions lead to a smooth progress of outflows, but compared to the normal distribution, the Weibull and beta distributions can assume a wide variety of shapes. The results thus show no significant difference between the two lifetime distributions. Dahlström et al.[50] compare the delta, Weibull, and log-normal distributions and reach similar conclusions, as the log-normal distribution can be adapted as well. Other authors tested the sensitivity of their models regarding different lifetime distributions, mean values of lifetimes, and deviations In addition to the findings described above, they find that their models are most sensitive to the mean values of lifetimes.[35,51,53,55]

Instead of using a lifetime distribution, Cheah et al.[37] choose a logistic survival rate function. In their review of methodologies for estimating lifetime distributions of commodities, Murakami et al.[56] and Oguchi et al.[57] give a comprehensive overview of how a lifetime distribution and a survival rate distribution, among others, are related. Only a few studies use time varying, nonparametric lifetime data, for example, for passenger vehicles and trucks in Japan[18,34,58,59] and lead-containing products in a global stock analysis.[60]

Table A.3 in the SI summarizes the characteristics and implementations of the different distributions.

3.3.7 Dynamic Modeling Approaches

The dynamic modeling approaches can be grouped according to their temporal extent and basic modeling principles as shown in Table 3.3.

Table 3.3: Dynamic modeling approaches implemented in the reviewed literature.

	Retrospective	Retrospective and prospective	Prospective
Top-down	*Historical data and lifetime distribution* [8,18,22,34,35,39–41,45,46,50,52–55,59,61–71]	*Historical data and lifetime distribution +* Constant consumption model[30,42,58,72,73] Linear consumption model[10,25,74] Exponential consumption model[11,32,37] Logistic consumption model[10,73] Regression model[33,43,73,75] Intensity of use[20,26] Consumption scenarios according to existing models[38] Individual consumption models for each product group[29]	Individual consumption scenarios [23,27,28]

		Logistic stock/capita model[24,47–49,76,77]
Bottom up	*Historical data and lifetime distribution*[34]	*Historical data and lifetime distribution +* Exponential stock model[78] Stock scenarios according to existing models[31,79] Individual stock models for each metal-containing technology[13,80]

The first dynamic MFAs of metals were modeled using retrospective and partly also prospective top-down approaches. With the exception of Van Beers and Graedel,[78] bottom-up approaches have been applied only since 2009. Likewise, solely prospective dynamic MFAs of metals based on scenario analyses have only been established recently. Figure 3.2 summarizes the development of modeling approaches over time.

3.3.7.1 Retrospective Top-Down Approach

Probably the simplest approach is the retrospective top-down dynamic MFA. It analyzes past stocks and flows based on time series of historical inflow data, such as trade, import, or consumption statistics. Given the past inflows, the outflows are calculated according to eq (4), and subsequently, stocks are calculated using eq (1). This approach is the most frequently chosen in the existing literature on dynamic MFA of metals (see Table 3.3), probably because of the better availability of inflow data compared to the stock data needed for bottom-up approaches.

In a recent study, Pauliuk et al. extended the top-down approach by calibrating its results based on the assumption that the old scrap supply equals the apparent old scrap demand, given a balanced scrap market, a homogeneous stock, and a perfectly closed steel cycle.

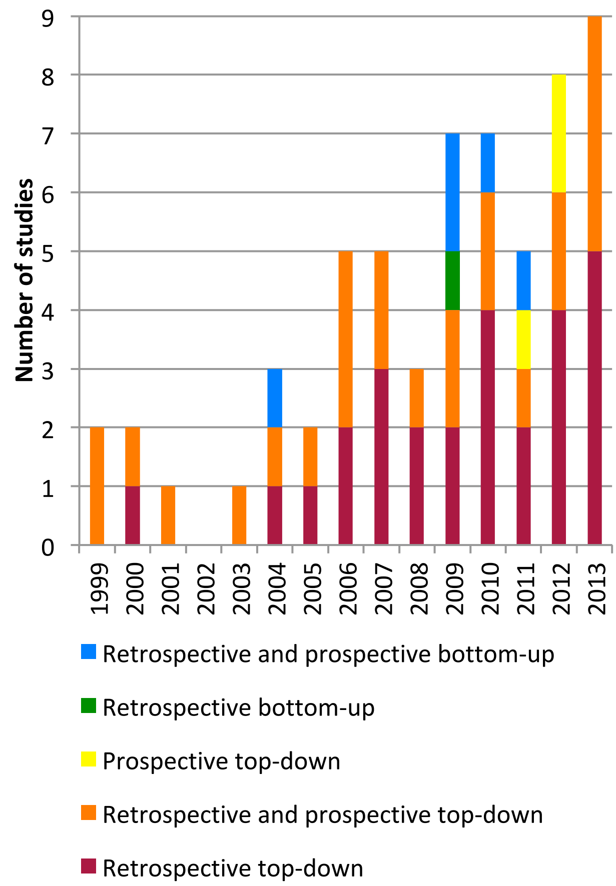

Figure 3.2: Development of modeling approaches used in dynamic MFAs of metals from 1999 to 2013.

3.3.7.2 Retrospective Bottom-Up Approach

A retrospective bottom-up model produces time series of historical stock data based on eq (2). If lifetime distributions and an initial stock value S[0] are known, past inflows and outflows can be calculated iteratively by applying eq (4) and eq (1). These latter calculation steps overlap with the retrospective top-down approach, so both approaches can be used to calculate the missing time series given the inflow and a lifetime distribution, the outflow and stock are calculated (thus an input-driven model) and given the stock and a lifetime distribution, the outflow and inflow are calculated (thus a stock-driven model). Hirato et al.[34] use both retrospective top-down and bottom-up models for automobiles in Japan and compare the results of the two approaches.

3.3.7.3 Retrospective and Prospective Top-Down Approach

Given that retrospective dynamic MFAs provide insights only on resource use in the past, the top-down approach is often combined with extrapolating time series of historical inflow data by fitting an appropriate continuous time function. The same approach can be used for a stock-driven model, as first introduced by Müller,[8] who proposes using the service provided by the in-use stock as the main driver of a material cycle, especially for materials with a long service lifetime. Past stocks are first calculated using an input-driven model; then the stocks are extrapolated to finally calculate future inflows using a stock-driven model.

The models used by Michaelis and Jackson, Hatayama et al., Igarashi et al., and Oda et al.[30,42,58,72,73] assume that metal consumption is in a steady state at a level of a specific reference year t_0

$$Inflow(t) = Inflow(t_0) \tag{5}$$

According to Michaelis and Jackson[42] and Oda et al.,[58] this simplification is justified when the metal stock has already reached or is going to reach saturation.

Zeltner et al.,[10] Park et al.,[74] and Yan et al.[25] use a linear model to extrapolate total metal consumption

$$\begin{aligned} Inflow(t) &= p_1 \cdot t + p_0 \text{ (continuous time)} \\ Inflow[n] &= p_1 \cdot nT + p_0 \text{ (discrete time)} \end{aligned} \tag{6}$$

with p_0 = initial value [kg/s] (all dimensions are given in SI units) and p_1= gradient [kg/s^2].

Models with an exponential consumption rate use a constant consumption growth rate which can be based on, for example, market reports or expert judgments[11,32,37]

$$\begin{aligned} Inflow(t) &= Inflow\ (t_0) \cdot (1 + p_1)^{\frac{t}{\tau}} \\ Inflow[n] &= Inflow\ (n_0 T) \cdot (1 + p_1)^{\frac{nT}{\tau}} \\ \text{if } \tau = \text{T} \quad Inflow[n] &= Inflow\ (n_0 T) \cdot (1 + p_1)^{n} \end{aligned} \tag{7}$$

with p_1 = constant growth rate in one period τ [-]. Zeltner[10] and Igarashi et al.[73] also model future metal consumption with a logistic function that takes growth limits of a system into account

$$\begin{aligned} Inflow(t) &= \frac{p_1}{1 + \mathrm{e}^{\,-p_2(t - p_3)}} \\ Inflow[n] &= \frac{p_1}{1 + \mathrm{e}^{\,-p_2(nT - p_3)}} \end{aligned} \tag{8}$$

with p_1 = saturation value of inflow [kg/s], p_2 = steepness of the sigmoidal curve [-], and p_3 = midpoint of the growth trajectory [-]. The parameters are determined using fitting algorithms (e.g., the ordinary least square method).

Elshkaki et al.[33,75] and Yamaguchi and Ueta[43] model inflows of lead-containing products as a function of socioeconomic explanatory variables such as GDP, population size, product price etc.

$$Inflow(t) = p_r + \sum_{i=1}^{n} p_i \cdot X_i(t) + \varepsilon(t) \tag{9}$$

with p_r = regression parameter [kg/s], n = number of socioeconomic explanatory variables, p_i = regression parameter [kg/(s·U)] (U: unit of explanatory variable), $X_i(t)$ = time series of socioeconomic variables [U], and ε(t) = the model error [kg/s].

Through regression analysis, the most significant socioeconomic variables can be found and evaluated based on statistical tests, such as the adjusted coefficient of determination, t-test, and F-statistics.[33] The regression parameters are determined via fitting algorithms. With extrapolations of the explanatory variables, the regression model is then used to estimate future inflows. Igarashi et al.[73] apply linear and non-linear regression models in a similar approach.

Ayres et al.[20] and Kapur[26] use extrapolative scenarios based on the intensity-of-use hypothesis, which describes a metal's intensity of use (metal demand per unit GDP) as a function of per capita income with a general form of an inverse U-shaped curve

$$IU\big(y(t)\big) = \frac{p_1}{y(t) + \frac{p_2}{y(t)^{p_3}}} \cdot p_4^{\frac{(t-t_0)}{T}} \qquad (10)$$

with y(t) = GDP per capita [US$/p], p_1 = parameter [kg/p], p_2 = parameter [US$/p], p_3 = parameter [-], p_4 = factor that scales down the intensity of use with time) [-], and t_0 = first year [s]. The inflow of refined copper is then

$$Inflow(t) = IU(y(t)) \cdot w(t) \qquad (11)$$

with w(t) = GDP [US$].

The curve illustrates the development from an agricultural to an industrial, more resource-intensive economy, and eventually to a high-income, service-oriented economy, which in turn has lower resource use. The extrapolation of population and GDP is based on scenarios developed by the Intergovernmental Panel on Climate Change (IPCC).[20]

Yano et al.[38] model the current and future end-of-life vehicle flows and their lead content based on Japanese car registration statistics and forecasts.

In a study on platinum use for new technologies, Elshkaki and Van Der Voet[29] present individual production and consumption models for each product group, often as functions of the exogenous variables GDP and population. Future GDP and population data is also taken from existing IPCC scenarios.

Instead of extrapolating inflows, Hatayama et al. and Pauliuk et al.[47–49] use forecasts of the in-use stock for their prospective dynamic MFAs.

Pauliuk et al.[49] apply logistic stock per capita models with scenario-dependent saturation levels

$$Per\_capita\_stock(t) = \frac{p_1}{1 + \left(\frac{p_1}{p_0} - 1\right)\, \mathrm{e}^{-p_2 p_1 t}} \qquad (12)$$

with p_0 = initial value [kg], p_1 = saturation value of total stock per capita [kg], p_2 = constant [1/(kg·s)]. They choose the parameters by assuming that the future per capita stock is tangent to the actual development and will ultimately reach the saturation level. Future inflows can be iteratively calculated based on future stock data with an initial value for inflow ($t=t_0$) and eq (4) being introduced into eq (1).

The models applied by Hatayama et al.[24,47,48] are based on per capita GDP as the only exogenous variable, allowing for different growth rates between regions

$$Per\_capita\_stock(t) = \frac{p_1}{1 + e^{(p_2 - p_3 \cdot (GDP(t)/cap(t))}} \tag{13}$$

with p_1: saturation value of total stock/capita [kg], p_2: parameter [-], and p_3: parameter [1/(US$)]. The model parameters are determined using nonlinear regression on the historical relationship between the per capita stock and GDP.

In their most recent studies, Pauliuk et al.[76] and Liu et al.[77] use a generalized five-parameter logistic function as a synthesis of the logistic curve and a Gompertz model to choose the saturation level and time independently for different world regions.

3.3.7.4 Retrospective and Prospective Bottom-Up Approach

Van Beers and Graedel[78] use a retrospective and prospective bottom-up approach to model zinc in-use stocks in Cape Town. They apply constant annual stock growth rates based on literature data for extrapolating past and future in-use stocks and flows.

$$Stock(t) = Stock\ (t_0) \cdot (1 + p_1)^{\frac{t}{T}} \tag{14}$$

with p_1 = constant growth rate in 1T [-]. Stock-driven copper flows were dynamically modeled by Bader et al.[13] The model comprises detailed analyses of historical copper stocks, including individual models for different end-use sectors and their copper-containing technologies.[12] For the extrapolation of stocks, Bader and his colleagues apply and fit logistic, linear-logistic, or double-logistic stock growth models, depending on the historical growth patterns. Gerst[80] also proposes a dynamic bottom-up model of global copper stocks. He derives his models from the historical stock of copper-containing technologies based on macrolevel socioeconomic variables such as GDP per capita, population, average household size, and level of urbanization. These time-dependent variables are then extrapolated, partly based on existing models and scenarios, to compute future stocks.

Saurat and Bringezu[31] model the use of platinum group metals in catalytic converters in automobiles in Europe based on a bottom-up simulation of the European fleet of passenger cars, including the expected evolution of the future fleet of fuel cell vehicles. For their dynamic analysis of iron and steel in Chinese residential buildings, Hu et al.[79] use an existing dynamic MFA model simulating the development of the floor area stocks in China's urban and rural housing systems.

3.3.7.5 Prospective Top-Down Approach

Zuser and Rechberger,[27] Marwede and Reller,[23] and Alonso et al.[28] model the future consumption of metals used in emerging technologies with a prospective top-down approach. Alonso et al.[28] model five demand scenarios for REE, basing the demand either on historical production or demand growth rates or on expected demand growth rates for emerging technologies according to expert knowledge or existing scenarios.

Zuser and Rechberger[27] analyze material demand for four different photovoltaic technologies according to three demand scenarios. In a similar approach, Marwede and Reller[23] develop three demand scenarios for tellurium in cadmium telluride photovoltaic cells.

3.3.8 Dissipation

The focus of the dynamic MFAs of metals reviewed is on bulk metal flows incorporated in durable goods and infrastructure. Metals, however, are also dissipated to the environment throughout their life cycle, with material dissipation being understood as defined by Ayres and colleagues[19,20] (see also Method section). In about half of the reviewed studies, dissipative flows are described as such or referred to as emissions, loss flows, stock leakage or specific flows to landfills or the environment. In 18 studies, the concept is an inherent part of the methodology, with 11 of these considering dissipative flows for all life phases and seven for only the use or disposal phase. Dissipative outflows of a specific process are calculated either from inflows and transfer coefficients or loss rates (e.g., refs 13, 20, and 53), from stocks and leaching/emission factors or corrosion coefficients (e.g., refs 13 and 75), from mass balances,[59] or based on historical data (e.g., slag sales[51]). Only three authors[13,20,71] consider time-variant coefficients; in all other studies, the share of dissipation remains constant over time.

Some literature specifically focuses on time-variant dissipative flows. In a recent study, Lifset et al.[5] assess dissipative copper flows in the United States based on historical data and individual models for the different copper flows. They further categorize these flows into "intentional and unintentional release", as well as "intentional and unintentional use". They also define a dissipation index that quantifies the ratio of dissipative flows to bulk flows as a measure of resource efficiency. Elshkaki et al.[81] model the nonintentional flows of lead in the Dutch economic system using a regression model approach, Sundset et al.[82] illustrate the mercury flows in the European Union, and Yamasue et al.[83] evaluate the potential amounts of dissipated rare metals from waste electrical and electronic equipment in Japan.

3.3.9 Spatial Dimension

It is important to know the location of a resource in addition to its quantity and quality to consider it for future mining.[78] Thus, some studies include the spatial distribution of in-use stocks, based on statistical or remote sensing data and usually processed in geographic information systems (GIS).[84] Van Beers and Graedel[78] link GIS data sets from a population census in Cape Town with zinc densities per area by applying weighting factors related to household income, dwelling type, or length of roads. In combination with annual stock growth rates, they also calculate the retrospective and prospective zinc distributions. Pauliuk et al.[70,76] and Liu and Müller[36] analyze steel and aluminum stocks and flows, respectively, for all countries in the world, based on statistical data. In addition, Pauliuk et al.[76] include capacity models to show how extensive trading of finished steel could prolong the lifetime of the steelmaking assets in different world regions, and Liu and Müller[85] develop a trade-linked multilevel MFA to map the global pathways of aluminum between countries. Remote sensing methods are used by Takahashi et al.,[86] who analyze in-use copper stocks using satellite nighttime light observation data. Other studies that include the spatial dimension of metal stocks are static MFAs.[87–89]

3.3.10 Uncertainty

Data included in an MFA of metals are acquired from many different sources with varying data reliability. If only individual values from measurements, expert interviews, or historical sources are available, it is often difficult to quantify the uncertainty of input data and parameters. The reviewed literature can be roughly divided into four groups according to how uncertainties are handled (see also Figure A.2 in the SI).

The first group, comprising approximately half of the studies, does not consider data uncertainty. The second group, 37% of all studies, applies sensitivity analysis. A sensitivity analysis helps to assess the relevance of uncertainties of the model parameters by providing knowledge of how the model output reacts to parameter changes. Many studies test the sensitivity of the model to different average lifetimes[22,35,45,51,67] or different lifetime distributions and standard deviations,[22,53,55,68,69] concluding that varying average lifetimes has a greater influence on model results than varying standard deviations or lifetime distributions. In addition to lifetime distributions, authors also carry out sensitivity analyses for other key parameters, such as steel intensity,[34,50,52,63,79] scrap recovery rate, [37,50,52,59] population size, stock saturation level, and saturation time.[76] In their most recent studies, Liu et al.[36,77] and Pauliuk et al.[70] carried out full sensitivity analyses for all model parameters according to their estimated data uncertainty. Besides the average lifetime, they found that the trade data estimation (based either on reported import or export data) and the metal content in commodities also have a high impact on resulting in-use stock calculations.

McMillan et al.[54] quantify the sensitivity of the lifetime distribution, recycling rate, and metallic recovery by using the Fourier Amplitude Sensitivity Test method, which provides a measure of input sensitivity defined as the fraction of total model variance.

The third group, 6% of all studies, uses uncertainty intervals. In particular, Kapur[26] assigns confidence levels to copper flows according to a confidence scale developed by Moss and Schneider.[90] He states that, as a general rule, the data quality decreases along the life cycle of a metal, that is, data for production, manufacturing, and the inflows into the use phase is more reliable than data for the waste management and recycling processes. Hedbrant and Sörme include uncertainty intervals as proposed in a comprehensive article on data uncertainty in urban heavy metal data collection. They assign uncertainty levels to sources of information, with associated uncertainty intervals based on factors (e.g., the value x could be as much as 3x or as little as 1/3x, annotated with "*/" analogue to "±"), which is especially useful for large uncertainties.[91] Van Beers and Graedel[78] apply asymmetrical uncertainty ranges for the zinc stocks per end-use sector.

Finally, the fourth group (5% of all studies) uses the Gaussian error propagation to calculate the standard deviation of stocks and flows based on standard deviations that were defined for each input variable and parameter.[10,13,36]

A different approach to handling uncertainty, which has not, however, been applied to MFAs of metals thus far, is probabilistic MFA, as proposed by Gottschalk et al.[92] They model inflows, transfer coefficients, and contents as probability distributions. The shape of the distributions (e.g., uniform, triangular, or log-normal) is chosen based on the characteristics of the available data. The dependent variables are calculated by means of Monte Carlo simulation and are therefore again provided as probability distributions. Bornhöft et al.[93] review existing modeling approaches and tools with regard to the requirements of probabilistic MFA.

3.3.11 Initial Condition

The initial condition of an MFA model depends primarily on the temporal extent chosen. If analyses go far back in time, initial stocks and flows are often considered zero at $t=t_0$. If the temporal extent is short or starts in the present, initial stocks, and flows are defined based on available data or the authors' assumptions.

3.3.12 Model Input Data

Model input data includes time series for exogenous model variables such as metal inflow and stock data, socioeconomic data, such as GDP or population, and model parameters such as the average lifetime of products or end-use categories. A detailed discussion of the data sources used by the studies reviewed is beyond the scope of this article.

3.3.13 Model Output Data

Model output data of the reviewed dynamic MFAs of metals comprise time series of those stocks and flows under investigation. Some studies quantify only the in-use stock (e.g., refs 68 and 80), while others provide information on all stocks and flows in their system from extraction to landfilling.[45] The resulting stocks and flows are often further divided into end-use sectors or products (most studies), disaggregated for different regions or countries,[40,48] or include details, such as the chemical composition of scrap flows or a breakdown into different alloy types (e.g., refs 59 and 72).

3.3.14 Evaluation

Besides a visualization and discussion of the output data, some studies include further evaluation. Various indicators can be applied that condense the results for better explication and communication.[16] Examples include the recycling rate, defined as the ratio between the actual scrap consumption and the scrap arising,[52] the scrap self-sufficiency ratio as the ratio of scrap recycling to scrap demand,[74] or other recycling indicators as applied by Glöser et al. or Yan et al.[25] Zeltner et al.[10] introduce the separation efficiency as the fraction of recycling in the total waste flow and Bader et al.[13] present the consumption loss as the sum of all metal flows to landfills or the soil/aquatic system.

Some evaluations are based on the relationship between material stocks and flows and socioeconomic indicators. McMillan et al.[54] analyze the relationship between the net addition to stock and GDP, and Mao and Graedel[66] and Liu and Müller[36] relate per capita stock to per capita GDP.

Comparisons of natural resources with anthropogenic stocks and flows are performed by Müller et al.,[51] Gerst[80] Alonso et al.,[28] and Liu and Müller.[36]

Some authors evaluate results by comparing them with the outcome of other studies (e.g., refs 13 and 80).

Further approaches evaluate the energy consumption or environmental impacts of the metal flows. Dahlström et al.[50] use value chain analysis to examine the material- and energy-related resource productivity and efficiency of the iron, steel, and aluminum industries in the United Kingdom, and Cheah et al.[37] analyze the embodied energy demand of automotive aluminum. For the United Kingdom steel sector, Michaelis and Jackson[41,42] calculate the consumption and development of exergy (available work). Hu et al.[79] assess resource depletion and global climate change by the accumulated net steel use and the net CO_2-equivalent emissions. The CO_2 emission volume reduction potential resulting from an enhanced collection of postconsumer steel was analyzed by Igarashi et al.[30] Saurat and Bringezu[31] model the SO_2 emissions related to PGM production and use in Europe. Liu et al.[35,77] analyze the energy use and greenhouse gas (GHG) emissions of the U.S. aluminum cycle and the GHG emission pathways of the global aluminum cycle.

Additional analyses include, for example, multimaterial pinch analyses to derive optimized recycling,[24] material intensity per service unit, life-cycle assessment, cost-benefit analysis, statistical entropy analysis,[16] and entropy analysis.[94–96]

3.3.15 Detailed Model Description

The detailed model description in the (adapted) ODD protocol comprises details about the model's formalisms (e.g., equations) and algorithms (e.g., solution procedures), the exogenous and endogenous variables, and the model parameters. Providing detailed model descriptions beyond the generic equations already discussed is beyond the scope of this article.

3.4 Discussion

We reviewed 60 studies of anthropogenic metal flows, comparing them with regard to their purpose, the materials, products, and sectors investigated, the coverage of processes, their spatial and temporal scale and extent, and the way they conceptualize and delimit the system under study. We extracted and summarized the basic concepts, principles, and methodological approaches underlying the models used and showed how the approaches developed over time.

The adapted ODD protocol proved to be beneficial for structuring the review process as well as this article. The literature fit well into the protocol's structure, which helped us to provide a one-to-one comparison of corresponding elements of the models despite their high diversity. The ODD protocol in our adapted form could hence provide a basis for the standardized description of MFA models in general, providing better orientation to the reader and supporting the completeness of model documentation and reproducibility of the results.

Most studies apply a top-down approach that could be used for any material. The required time series of inflow data are often provided by production, trade, or consumption statistics. Data is mainly available for bulk metals or, in the form of global production figures, also for less widely used metals. Because available inflow data is often highly aggregated, the top-down approach may be less suitable for specific products or smaller regions.

Only 10% of the studies apply a bottom-up approach. Bottom-up models are far less generic since the entire stock of a specific metal has to be assembled from all product groups containing that metal. These may all show different growth patterns, requiring a specific stock model for each product group as well as extensive data collection.[13,78,80] This approach is thus most suitable for analyzing metals that are only used in a few products, or for focusing on a specific product. Bottom-up models can also benefit from existing models that provide past and future time series of stock data that can be directly applied in an MFA.[31,79]

The bottom-up approach, although it has not been widely applied to date, could provide important insights on consumer behavior, which, for example, influences the product lifetime or the disposal pathways, sociocultural and spatial differences in patterns of metal use,[36] the split of metals to different end-use sectors, or the share of obsolete stock (e.g., stored products, abandoned infrastructure) in the in-use stock[70] by investigating in detail the in-use stock, for example, through consumer surveys.

Especially for studies with a long time horizon, the assumption of constant model parameters, for example, the lifetime distribution parameters, is a far-reaching simplification that could add a significant error to the results.[97,98] Sinha-Khetriwal et al.[99] also point out that forecasts of outflows can be improved by introducing product mass functions incorporating the changing weight of products over time as in Gregory et al.[100] Such data can often only be derived from bottom-up models. Detailed data generated by bottom-up models can also be used to calibrate and validate top-down models, an issue not yet often addressed in the literature. Future research should therefore investigate time-variant systems and how top-down and bottom-up models could be combined or complemented.

The diverse extrapolation methods reveal various challenges. Inflow data often fluctuates, depending on economic and technological developments, such as market crises or product substitutions, which are difficult to predict. Furthermore, linear and exponential consumption models, since they do not take into account resource scarcity and market saturation, are only valid within a short time frame. Extrapolation of inflow data is therefore prone to oversimplification. Stocks, however, are less affected by short-term market fluctuations and thus provide a more robust basis for forecasts.[8,77] For regression models, too, the forecasts based on socioeconomic variables are only valid as long as no unpredicted societal or economic changes occur.[33]

The majority of studies analyze bulk metals such as iron/steel, aluminum, copper, and zinc with increasingly detailed information on their stocks and flows. Literature on less widely used metals such as indium, tantalum or REE is still scarce. The use of these metals highly depends on new emerging technologies. At the end of product life, most of the metals are not recovered but lost from the system considered, since there is not yet any technically or economically feasible recycling option for many of them. It is thus highly advisable to further develop data and models with a special focus on metal dissipation. Earlier, dissipation, losses or emissions of metals to the environment were included in dynamic MFA models focusing on heavy metal pollution. In recent years, dissipation has also been addressed also from a resource point of view, but data is extremely scarce and many authors have still not taken up the issue. Moreover, metals may not only be dissipated, but also distributed in low contents to many products, and, within these products, scattered all over the globe. These metals are not irrecoverable, but great efforts are needed to concentrate them again. Rechberger and Graedel[101] developed statistical entropy analysis to measure the distribution pattern of a substance over its life cycle, that is, to describe how a system concentrates or distributes substances. It appears promising to apply statistical entropy analysis to metals in the anthroposphere for the measurement and illustration of their distribution as a basis for improved resource management. Future research should also investigate other, possibly new indicators of dissipation and material distribution.

The most recent literature[36,70,77] has shown that besides lifetime distribution parameters there might be many other parameters or variables with a strong influence on the model's output. Performing uncertainty analysis such as Gaussian Error Propagation, or, if the data uncer-tainty is unknown, a full sensitivity analysis, is therefore important to understand the effect of uncertain model input. Probabilistic MFA by Gottschalk et al.,[92] which models all data as probability distributions and thus accounts for the influence of the model input's uncertainty on the model output, is a comprehensive approach for dealing with uncertainty.

Dynamic MFA is a useful method for providing knowledge of metal stocks and flows in the anthroposphere in a simple and comprehensible way. Many studies also include further evaluations of their results or serve as a basis for further assessments (e.g., ref 102). However, with the exception of Kapur et al.[26], the results and conclusions of the reviewed literature do not directly support environmental policy making. Some studies indirectly give recommendations, for example, regarding how to increase recycling rates (e.g., refs 23, 39, 49, 54, 62, and 65), reduce environmental impacts (e.g., refs 31 and 67) or mitigate climate change (e.g., refs 41 and 77), but without a clear target audience. We suggest that future studies that intend to provide environmental policy recommendations, identify their target audience and their purpose from the beginning. MFA could also be recognized as a necessary element of impact assessments used for new regulations, such as the sustainability impact assessment used for new trade agreements by the European Commission.[103] The mid- and long-term impacts of political decisions on material stocks and flows may become one of the most important economic and environmental concerns in the coming decades. To fulfill the requirements of policy support, MFA models should be embedded in an environment of scenario definition and simulation that easily connects the models to socioeconomic data taken from statistical databases and geographic information systems.

3.5 Associated Content

Summary and overview of all reviewed studies, overview of the processes covered by the reviewed literature, figures showing the percentage distribution of the reviewed studies by spatial extent and by treatment of data uncertainty, more details on the basic modeling principles, a summary of lifetime distribution functions, and additional information on the relationship between different measures from the field of system reliability. This material is available free of charge at http://pubs.acs.org.

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Chapter 4

Service Lifetime, Storage Time, and Disposal Pathways of Electronic Equipment

A Swiss Case Study

Esther Thiébaud (-Müller), Lorenz M. Hilty, Mathias Schluep, Rolf Widmer, Martin Faulstich

Service Lifetime, Storage Time, and Disposal Pathways of Electronic Equipment

A Swiss Case Study

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Supporting information is linked to this article on the *JIE* website

Summary

Product lifetime is an essential aspect of dynamic material flow analyses and has been modeled using lifetime distribution functions, mostly average lifetimes. Existing data regarding the lifetime of electronic equipment (EE) are based on diverging definitions of lifetime as well as different temporal and regional scopes. After its active use, EE is often not disposed of immediately, but remains in storage for some time. Specific data on the share of EE that is stored and the time they remain in storage are scarce. This article investigates the service lifetime, storage time, and disposal pathways of ten electronic device types, based on data from an online survey complemented by structured interviews. We distinguish between new and secondhand devices and compute histograms, averages, and medians of the different lifetimes and their change over time. The average service lifetime varies from 3.3 years for mobile phones to 10.8 years for large loudspeakers, the average storage time from 0.8 years for flat panel display televisions to 3.6 years for large loudspeakers. Most service lifetime histograms are positively skewed and show substantial differences among device types. The storage time histograms, being more similar to one another, indicate that the storage behavior is similar for most device types. The data on disposal pathways show that a large proportion of devices are stored and reused before they reach the collection scheme.

Introduction

The fast pace of innovation cycles for electronic equipment (EE) and the falling prices for new devices lead to short product lifetimes and increasing sales (Prakash et al. 2015). EE contains important material resources, including bulk materials, precious metals, and critical raw materials (Buchert et al. 2012). Whereas the recycling of bulk materials and precious metals is

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Keywords: Service lifetime; storage time, disposal pathways, electronic waste, obsolescence, material flow analysis

4.1 Introduction

The fast pace of innovation cycles for electronic equipment (EE) and the falling prices for new devices lead to short product lifetimes and increasing sales (Prakash et al. 2015). EE contains important material resources, including bulk materials, precious metals, and critical raw materials (Buchert et al. 2012). While the recycling of bulk materials and precious metals is often well established, efforts to specifically recover critical raw materials from EE are only beginning. They are hampered by low content per device, low market prices that make recycling unattractive, limitations of recovery technologies as well as limited knowledge on stocks and flows of these devices. Efficient recycling thus relies on knowledge of anthropogenic material cycles regarding location, lifetime, disposal pathways, quantities, and qualities of EE (UNEP 2010; UNEP 2011; UNEP 2013; van Schaik and Reuter 2010; Reuter 2011) This information enables recycling systems to forecast future mass flows, provide for sufficient recycling capacities, and invest in appropriate recycling technologies (Prakash et al. 2015).

The temporal analysis of material cycles is often based on a dynamic material flow analysis (MFA) approach. The product lifetime or lifespan – these terms are used as synonyms in existing literature - is an essential part of dynamic MFAs, necessary to calculate the development of stocks and outflows from inflow data or inflows and outflows from stock data. It has mostly been modeled in dynamic MFAs so far by assuming lifetime distribution functions, mostly average lifetimes (Müller et al. 2014). In many MFA studies, it has been discussed that stocks and flows are sensitive to the chosen lifetime distribution functions and their parameters (Chen and Graedel 2012; Liu, Bangs, and Müller 2011; Müller et al. 2006). Data regarding the product lifetime of specific EE are relatively abundant for mobile phones, for example (Cooper 2005; Echegaray 2014; EPA 2004; J.D. Power and associates 2007; Murakami et al. 2010; Polák and Drápalová 2012; Stocker et al. 2013; Wang et al. 2013; Wieser and Tröger 2015), televisions (TVs), for example (Cooper 2005; Echegaray 2014; Gutiérrez et al. 2011; Oguchi et al. 2010; Prakash et al. 2015; Wang et al. 2013; Wieser and Tröger 2015), as well as laptop and desktop computers (Babbitt et al. 2009; Cooper and Mayers 2000; Echegaray 2014; Prakash et al. 2015; Prakash et al. 2012; Sabbaghi et al. 2015; Wang et al. 2013; Wieser and Tröger 2015; Williams and Hatanaka 2005). Data is more scarcely available for printers (Echegaray 2014; Stocker et al. 2013; Wang et al. 2013), radio and HiFi components (Cooper 2005; Gutiérrez et al. 2011; Wang et al. 2013), video equipment (Cooper 2005; Echegaray 2014; Wang et al. 2013), cameras (Wang et al. 2013; Wieser and Tröger 2015), telephones (Cooper 2005; Wang et al. 2013), Monitors (Wang et al. 2013), and speakers (Wang et al. 2013). Most of these studies provide average or median lifetimes. Using average lifetimes in dynamic MFAs, that is, a delta function as the lifetime distribution, leads to modeled outflows that are identical to the often fluctuating inflows and often significantly under- or overestimate measured outflows (Müller et al. 2014). The only studies presenting more information on distributions are Polák and Drápalová (2012) as well as Wang and colleagues (2013), which provide Weibull parameters for lifetime distributions and Babbit et al. (2009) as well as Stocker and colleagues (2013), which derive Gaussian lifetime distributions from lifetime histograms.

Studies of product lifetime use different definitions of the phase of the life cycle that is taken into account (Murakami et al. 2010; Oguchi et al. 2010; Babbitt et al. 2009). Many studies highlight that the product lifetime may change significantly over time (Babbitt et al. 2009; Echegaray 2014; Murakami et al. 2010; Oguchi et al. 2010; Prakash et al. 2015; Sabbaghi et al. 2015; Wang et al.

2013). The existing studies all refer to a specific system boundary, that is, a country or a region and a specific time frame. A compilation of available data for mobile phones and TVs (Wieser and Tröger 2015) as well as a study on desktop PCs (Müller et al. 2009) has shown that the product lifetime can vary considerably between countries or regions. All these factors make it difficult to compare studies and adopt data to different settings.

At the time of replacement, EE is often not disposed of immediately, but stored for some time. This leads to, for example, very few mobile phones collected for disposal despite high sales numbers. Comparisons of assumed product lifetimes with the product age at recycling facilities have shown that products are often older than expected (Stocker et al. 2013). Data showing how long the various EE is stored are scarce. Of the literature mentioned above, Polák and Drápalová (2012) report the average storage time of used mobile phones, including reuse, Sabbaghi and colleagues (2015) investigate the storage time of desktop and laptop computers by analyzing used hard disk drives, and Williams and Hatanaka (2005) indicate the storage time for personal computers from a survey. In addition, Milovantseva and Saphores (2013) and Saphores and colleagues (2009) analyze the amount of TVs and e-waste in general, respectively, stored in households of the United States. Instead of storage, sometimes, the term hibernation is used, for example, by Murakami and colleagies (2010) and Daigo and colleagues (2015), who analyze the hibernating behavior of material stocks of steel in Japan. For how long products or materials are stored (or 'hibernating'), however, is not addressed in these articles.

For every dynamic MFA that takes into account both the use and the disposal phase of a product, the knowledge of disposal pathways and the related transfer coefficients is crucial. The disposal pathways chosen in existing MFAs of EE vary from highly aggregated processes (e.g. recycling, landfilling, export) to very detailed breakdowns (e.g. collection, repair, recycling, second-hand sale, export etc.), for example (Kahhat and Williams 2012; Lam, Lim, and Schoenung 2013; Lau, Chung, and Zhang 2013; Leigh, Choi, and Hoelzel 2012; Steubing et al. 2010; Yoshida, Tasaki, and Terazono 2009).

In this article, we present the results of a survey on the service lifetime, the storage time, and the disposal pathways of EE that we conducted between 2014 and 2016 in Switzerland. The goal of the survey is to obtain detailed 'bottom-up' information of the service lifetime and the storage time (hibernating time) of EE in Switzerland. We further aim at gaining insights in the trigger events to transfer devices between the use, storage and disposal phase. The distinction between service lifetime and storage time as well as the more detailed knowledge on reuse, storage and disposal flows enables a more accurate MFA model of the actual stocks and flows of EE in Switzerland. Such a model is important, for example, to explain the discrepancy between low collection flows and high sales flows, predict the average product age of devices at recycling facilities or account for long phase out periods of technologies that are no longer sold (e.g. CRT TVs and monitors). It can further provide a basis for recycling system managers to understand how different device types are handled in the use phase, forecast future recycling flows based on assumed or extrapolated sales flows as well as provide appropriate and tailored recycling capacities and technologies for the expected composition of the recycling flows. In life cycle assessment (LCA), the total impact should be split into the different phases, such as manufacturing, service, storage and recycling. Thus clarifying the difference between service lifetime and storage time has a potential to recalibrate many LCA studies. Knowledge on the effective service lifetime versus the total lifetime (including storage) and the share of reused devices might also be useful for other stakeholders, in particular: product designers who aim at improving product longevity and product remanufacturers who need this information for their capacity planning.

4.2 Method

The initial data collection consisted of an online survey distributed via social networks and email in 2014. The survey covered 10 electronic device types with a high content of indium, neodymium or gold, so that they cumulatively cover over 90% of these three metals in private Swiss households (Böni et al. 2015). Table 4.1 lists the considered device types as well as the associated UNU-Keys for comparability with existing studies, e.g. Baldé et al. (2015).

Table 4.1: Electronic device types included in the survey

Device type	Description	UNU-Key
Desktop	Desktop computer (incl. all-in-one computer, without external peripherals)	0302
Laptop	Laptop/Notebook computer	0303
Monitor	Flat panel display (FPD) monitor	0309
Mobile phone	Mobile phone / Smartphone	0306
Headset	Headphones / Headset	0401
CRT TV	Cathode ray tube television	0407
FPD TV	Flat panel display television	0408
Loudspeaker small	Portable loudspeaker / Loudspeaker docking station	0403
Loudspeaker large	Loudspeaker set (of Hi-Fi and Home Cinema Systems)	0403
DVD player	DVD (digital video disk) player / Blu-ray player	0404

The survey included questions on the service lifetime, the storage time, and the disposal pathways, and targeted at devices that were still in use, devices that were stored, and already disposed of devices. We defined the service lifetime as the *time of active use of a device*. The storage time is defined as the *time between the active use of a device and its final disposal or its transfer to a different user*.

The original questionnaires were provided in two language versions, German and English. In the first part of the questionnaire, we asked the participants how many devices of the considered device types they are currently using, storing or have already disposed of (Table B.1 in the supporting information 1 available in the Journal's website).

In the second part, we asked detailed questions for each device the participant had indicated in the first part. For each device, we asked for

- the condition of the device when it was purchased by the current user (new/second-hand),
- the year of purchase and, for second-hand devices, the age of the devices when it was purchased,
- the service lifetime
    - for devices already stored or disposed of,

- for devices still in use including an estimation of the years the user intended to continue to use it,

- the storage time
    - for devices already disposed of,
    - for devices still stored including an estimation of the years the user intended to continue to store it,
- the disposal pathway for devices already disposed of.

The questions were the same for all device types with one exception: as headphones or headsets are often abundant, and users have no accurate overview of their usage time, we only inquired the average service lifetime, storage time and disposal pathway of all headsets in use, stored or disposed of (Table B.2 to B.7 in the supporting information 1 on the Web).

In the third part of the survey, we asked each respondent for her/his gender, age, and level of education as independent auxiliary variables (Table B.8 in the supporting information 1 on the Web).

In total, the online survey resulted in 441 valid responses from Switzerland and Liechtenstein. Liechtenstein forms part of the Swiss e-waste recycling system and its population structure is very similar to that of Switzerland. We therefore decided to include the survey results for Liechtenstein in the Swiss context. With a Swiss population of roughly 8'000'000, a confidence level of 95% and a margin of error of 5%, we needed a sample size of at least 385 people. However, data from online surveys are often biased due to under-coverage and self-selection. Under-coverage in online surveys happens when the possible survey sample population does not equal the target population, since only respondents with access to the Internet can participate. Self-selection means that the researcher does not control the selection process. Survey respondents are individuals with Internet access who by chance learn about the survey via social networks or email and decide to participate. This means that the survey sample is not randomly selected from the target population and thus the principles of probability sampling are not followed (Bethlehem 2009; Bethlehem 2010; Gosling et al. 2004). While a truly random sampling of the population, for example by telephone-based questionnaires, is clearly preferable, there were not sufficient resources available to do this.

In order to account for the above concerns, we included three independent auxiliary variables in our survey: age, gender and highest level of education (Table B.8 in the SI). The statistical relevance of these variables was tested using the rank-based nonparametric Kruskal-Wallis test or Mann-Whitney U test. Subsequently, we created 30 age-gender-education subgroups and assessed the respondents' distribution to these subgroups compared to the Swiss and Liechtenstein population distribution. For groups that were missing or severely under-represented in our survey we held 23 additional, structured interviews in 2015 and 2016. The interviewees were personally approached by the researchers according to their gender, age and education attributes, therefore the response rate was 100%. In total, the data collection resulted in 464 participants from Switzerland plus Liechtenstein. To correct for the under- or over-represented groups, we introduced weighting adjustments (Bethlehem 2010). Further information regarding the independent auxiliary variables, the age-gender-education subgroups (Table B.9) and the weighing adjustments Table (S10) are described in the supporting information 1 on the Web.

From the survey results, we calculated the year each device was sold as a new product. On this basis, we calculated the service lifetime and storage time for each device. For new devices, 'service lifetime' refers to the time of active use of the device. For second-hand devices, 'service lifetime' refers to the time of active use by the first user. We assume that the time of active use by the first user is equal to the age of the device at the time of resale, although this might include some storage time. The 'second service lifetime' refers to the active use of a second-hand device.

The 'storage time' relates to the time between the active use of a new device and its final disposal or its transfer to a different user. The 'second storage time' relates to the storage time after the active use of a second-hand device. The resulting equations and the references to the survey responses are listed in Table B.11 of the supporting information 1 on the Web.

The service lifetime for devices still in use includes an estimate of the time the user intends to continue to use the device. Literature shows that people tend to overestimate future service lifetimes of their devices (Cooper and Mayers 2000; Echegaray 2014; Wieser and Tröger 2015; Wilhelm, Yankov, and Magee 2011). We thus compared this data with the service lifetime of devices no longer in use, that is, already stored or disposed of, in order to see whether we could confirm this finding. However, the sample of devices no longer in use is biased due to right censoring. Right censoring refers to the fact that for more recent sales years, many devices are still in use and therefore don't appear in the sample. Because only cases of shorter lifetime are included, the sample is distorted. To exclude right censored data, we omitted data for more recent sales years of each device type.

The storage time of devices currently in storage includes an estimate of the time the user intends to continue to store the device. If we only considered the storage time of devices already disposed of, the sample would again be biased, in particular because for many device types, more devices are currently stored than already disposed of. However, for storage time and second storage time, no literature data is available against which we could test the user estimates. Therefore, we analyzed the total sample, including all data from all sales years, keeping in mind that they include rough user estimates.

Based on the above definitions, we calculated for each device type the corresponding weighted and normalized histograms, box plots as well as the weighted averages and standard deviations of the service lifetime, the second service lifetime, the storage time and the second storage time. To analyze the temporal change of the service lifetime and the storage time, we defined 2014 as reference year, as most data was collected in this year. The data was divided into sales year groups. For each sales year group and device type, we computed a weighted and normalized cumulative histogram of the service lifetime and storage time. For the service lifetime, we took into account all available observations, but only display the service lifetime of devices that were stored or disposed of before 2014, in order to exclude prospective user estimates of the service lifetime because they are probably biased. Therefore we display only partial cumulated histograms for more recent sales year groups. For the storage time, we used all available observations.

The disposal pathway results were represented as transfer coefficients. A transfer coefficient refers to the relative share of a specific flow in relation to the total outflow from a specific process. Transfer coefficients are expressed as numbers between 0 and 1 (or 0 and 100%). The sum of the transfer coefficients from a process to all disposal pathways equals 1 for each device type.

More details regarding the data evaluation can be found in supporting information 1 on the Web. The microdata of the survey results are provided in supporting information 2 on the Web

4.3 Results and Discussion

4.3.1 Service Lifetime

4.3.1.1 Median and Average Service Lifetime

Figure 4.1 shows the box plots of the service lifetime of devices still in use compared to the box plots of the service lifetime and the second service lifetime of devices no longer in use. Each box plot is based on data over all available sales years.

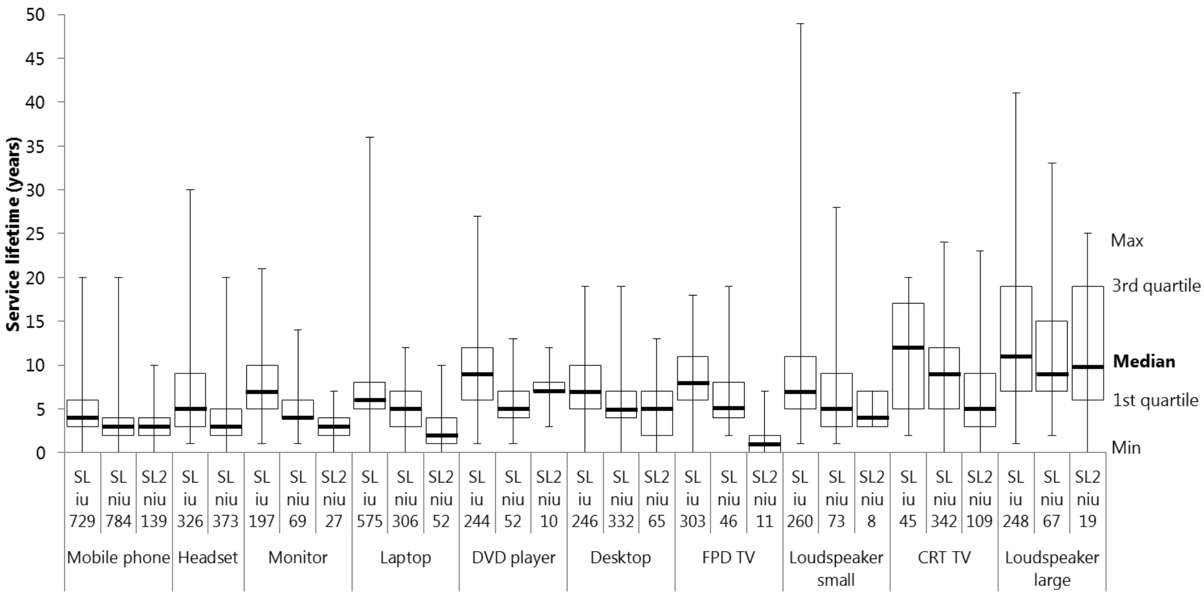

Figure 4.1: Comparison of the box plots of the service lifetime for 10 different electronic device types. SL iu: service lifetime of devices in use, SL niu: service lifetime of devices no longer in in use, i.e., already stored or disposed of, SL2 niu: second service lifetime of devices no longer in use. The number in the third line indicates the sample size. FPD TV = flat panel display television; CRT TV = cathode ray tube television; DVD = digital video disc.

The median service lifetime of devices still in use, including the estimated years the user intends to continue to use it, varies between 4 years for mobile phones and 12 years for CRT TVs. The average lifetime of devices still in use ranges between 4.3 years for mobile phones and 13.4 years for large loudspeakers. The median service lifetime of devices no longer in use varies between 3 years for mobile phones and headsets and 9 years for CRT TVs and large loudspeakers. The corresponding average service lifetimes are 3.3 years, 3.8 years, 9.2 years and 10.8 years, respectively. The median second service lifetime is equal to or shorter than the median (first) service lifetime, depending of the type of device and with the exception of DVD Players and large loudspeakers. The first and third quartiles as well as the minimum and maximum values illustrate the often large variance of the data.

For most device types, the service lifetime of devices still in use shows the highest variance. An overview of average values including standard deviations and standard errors of the average can be found in Table B.14 and B.15 of the supporting information 1 on the Web.

According to the results presented above, users declare intended service lifetimes that are higher than the service lifetimes of the products of the same type they used in the past, by 20% for laptops up to 80% for DVD players. The temporal analysis of the service lifetime has revealed either stable or decreasing service lifetimes for all device types (see below). If the intended service lifetimes declared by the respondents were taken for granted, this would thus imply a trend reversal, which doesn't seem very likely. In several studies users were explicitly asked for the expected and the measured lifetime of various consumer durables, including EE. Cooper and Mayers (2000) found that for mobile phones in the United Kingdom the expected service lifetime exceeds the measured service lifetime by 50%. In Brazil, users expect TVs to last 40%, computers 70% and mobile phones 80% longer than the service lifetime of their previously disposed devices (Echegaray 2014). Similar findings were made in a study on the lifetime of mobile phones in the US (Wilhelm, Yankov, and Magee 2011). In an Austrian study, the authors distinguished between desired, expected and actually measured service lifetime. They found that the expected lifetime is substantially lower compared to the desired lifetime, and the discrepancy between the desired and the actually measured lifetime amounts from 170% to 330% (Wieser and Tröger 2015). All these discrepancies show that longer lifetimes are expected by many users, but not achieved. The reasons may include fast innovation cycles, wear and tear, low product quality, or poor reparability. We thus regard the data for devices no longer in use as more reliable and use only this data for further analysis.

Mobile phones and headsets clearly show the shortest median and average service lifetimes. CRT TVs and large loudspeakers show the longest service lifetimes. The service lifetime of mobile phones is often connected to mobile phone contracts (J.D. Power and associates 2007), which in Switzerland normally last two years. This finding can be confirmed by the histogram of the service lifetime of mobile phones, with its mode at two years. Headsets are often included in mobile phone packages, which may explain that they have similar lifetimes as mobile phones. CRT TVs and large loudspeakers are both durable products that are rather replaced due to technical upgrade than failure. Using Fisher's classification of products (Fisher 1997), CRT TVs and large loudspeakers can be regarded as functional products, whereas the remaining device types are rather innovative products.

4.3.1.2 Histograms of Service Lifetime

Figure 4.2 shows the normalized histograms of the service lifetime of devices already stored or disposed of, for 10 electronic device types over all available sales years. The service lifetime is not normally distributed based on the Shapiro-Wilk test, but positively skewed. Headsets and mobile phones exhibit similar (and the narrowest) service lifetime distributions. The service lifetimes of desktops and laptops are also similarly distributed. CRT TVs and large Loudspeakers show the longest service lifetimes and the widest distributions. The positively skewed distributions show that for all device types, a small number of devices remain in use for a multiple of the average or median service lifetime. One potential explanation for this observation is that all devices of a given type could in principle be used for such a long time, as it seems to be technically feasible. However, as our survey did not investigate the reasons for obsolescence of devices, we have no positive evidence so far that most devices were replaced before failure.

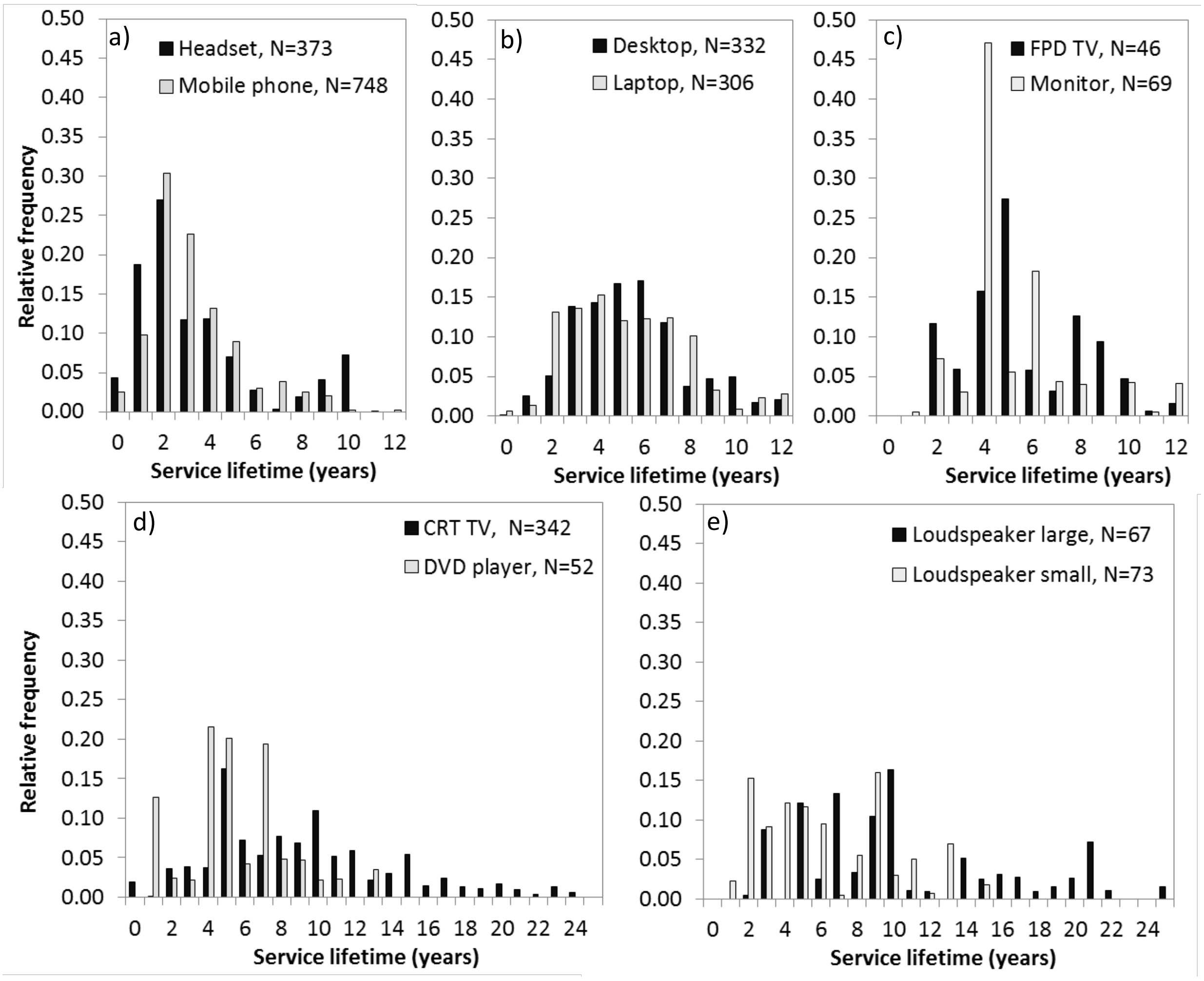

Figure 4.2: Normalized histograms of the service lifetime of devices no longer in use for 10 electronic device types: (a) headset and mobilephone; (b) desktop and laptop; (c) FPD TV and monitor; (d) CRT TV and DVD player; and (e) loudspeaker large and loudspeaker small. N denotes the sample size. FPD TV = flat panel display television; CRT TV = cathode ray tube television; DVD = digital video disc.

4.3.1.3 Temporal Change of Service Lifetime

Although various studies emphasize that the product lifetime may decrease significantly over time (Babbitt et al. 2009; Echegaray 2014; Murakami et al. 2010; Oguchi et al. 2010; Prakash et al. 2015; Sabbaghi et al. 2015; Wang et al. 2013), the cumulative histograms resulting from the analysis of the change over time of the service lifetime show no significant trend for the temporal change for desktops, laptops, FPD TVs and mobile phones. The service lifetimes of monitors, large and small loudspeakers and DVD players show a slight decrease, the more recently the devices were sold. For CRT TVs, after 10 years, a decrease of the further service lifetime as well as of the variance can be observed. Figure 4.3 shows two examples of the temporal change of the service lifetime. The remaining cumulative histograms are provided in Figures B.6 – B.9 in the supporting information 1 on the Web. Reasons for the decrease of product lifetime mentioned in literature, such as faster innovation cycles, falling prices (Prakash et al. 2015), increasingly sophisticated advertisement or the desire for social inclusion (Wieser and Tröger 2015) might have less influence than expected. It could therefore be justified to assume constant lifetimes for certain device types in a dynamic MFA model.

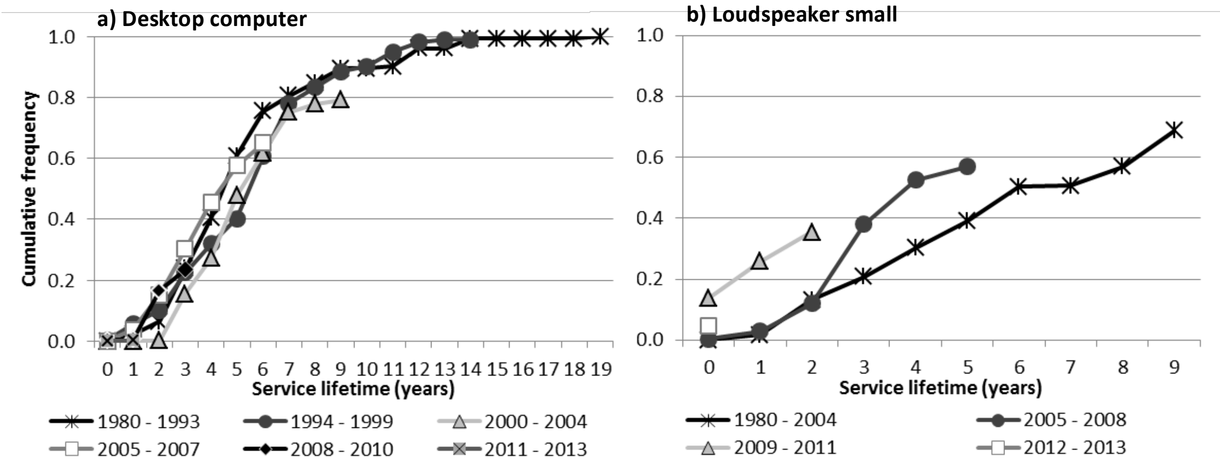

Figure 4.3: Example of the analysis of the temporal change of the service lifetime of a) desktop computers and b) small loudspeakers. Each line represents a sales years group of devices, i.e., the set of devices sold in the time period indicated in the legend.

4.3.2 Storage Time

4.3.2.1 Median and Average Storage Time

The box plots of the storage time and the second storage time are shown in Figure 4.4. Each box plot is based on data over all available sales years.

The median storage time, including devices with 0 storage time, is 0 years for most device types. Laptops and mobile phones have a median storage time of 1 year, headsets and large loudspeakers of 2 years. The average storage time ranges from 0.8 years for FPD TVs to 3.6 years for large loudspeakers. For FPD TVs, CRT TVs, Laptops and DVD players, the median second storage time is similar to or smaller than the median storage time. For all other device types, the median second storage time is larger by at least one year. With a few exceptions, the maximum storage time exceeds 10 years for most device types, which illustrates the large variance of the storage time. An overview of average values can be found in Table B.16 and S17 of the supporting information 1 on the Web.

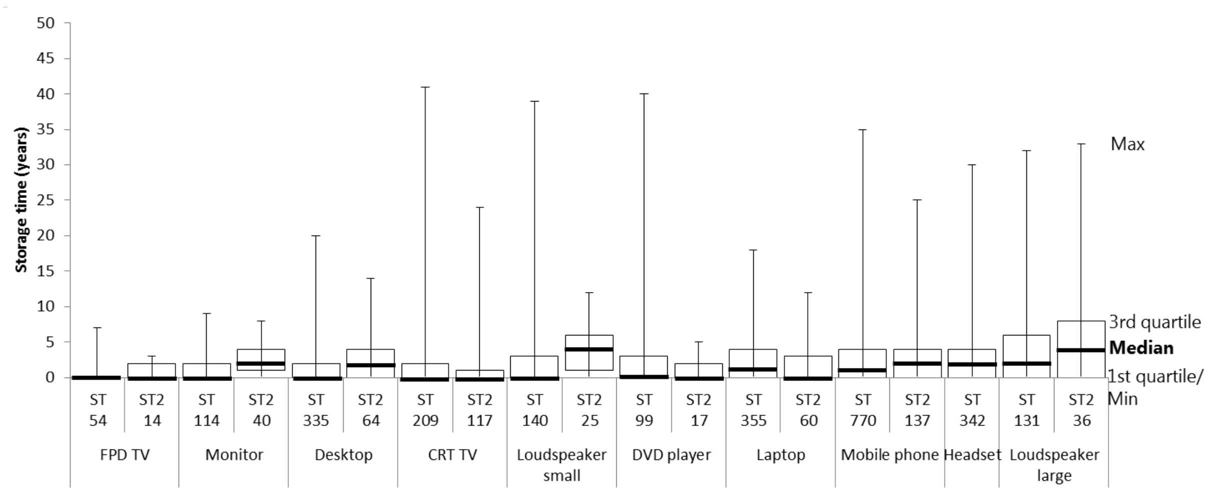

Figure 4.4: Comparison of box plots of the storage time for 10 different electronic device types. ST: storage time, ST2: second storage time. The number in the second line indicates the sample size. FPD TV = flat panel display television; CRT TV = cathode ray tube television; DVD = digital video disc.

The variation of the average storage time among device types is smaller than that of the service lifetime, with most device types having an average storage time of 1.5 to 2.8 years. Compared to the average service lifetime, the average storage time is similar for mobile phones and headsets, about a factor 6 to 7 smaller for CRT and FPD TVs and a factor 2 to 3 smaller for the remaining device types. Small devices such as mobile phones and headsets are thus stored for a longer time than large devices such as TVs, compared to the time they are actively used. Remy and Huang (2015) also found that the storage time is highly influenced by the personal attachment to a device. This could explain why laptops are longer stored than, for example, small loudspeakers.

4.3.2.2 Histograms of Storage Time

The normalized histograms of the storage time show similar distributions for most device types (Figure 4.5), indicating again a smaller variation of the storage time among device types, compared to the service lifetime. The share of devices that are not stored at all ranges from 30% for headsets up to 75% for FPD TVs.

4.3.2.3 Temporal Change of Storage Time

The analysis of the temporal change of the storage time illustrates that for most device types, more devices are stored, the more recently the devices are originally sold. Their storage time, however, is often shorter and the variance of the data is smaller. Exceptions are large and small loudspeakers, where more recently sold devices are less stored and have shorter storage times than devices with earlier sales years. Explanations for these observations are difficult to find. Evidence could only be provided by a survey that investigates more into the reasoning behind the users' decisions. The temporal change of the storage time is depicted in Figures B.10 – B.14 in the supporting information 1 on the Web.

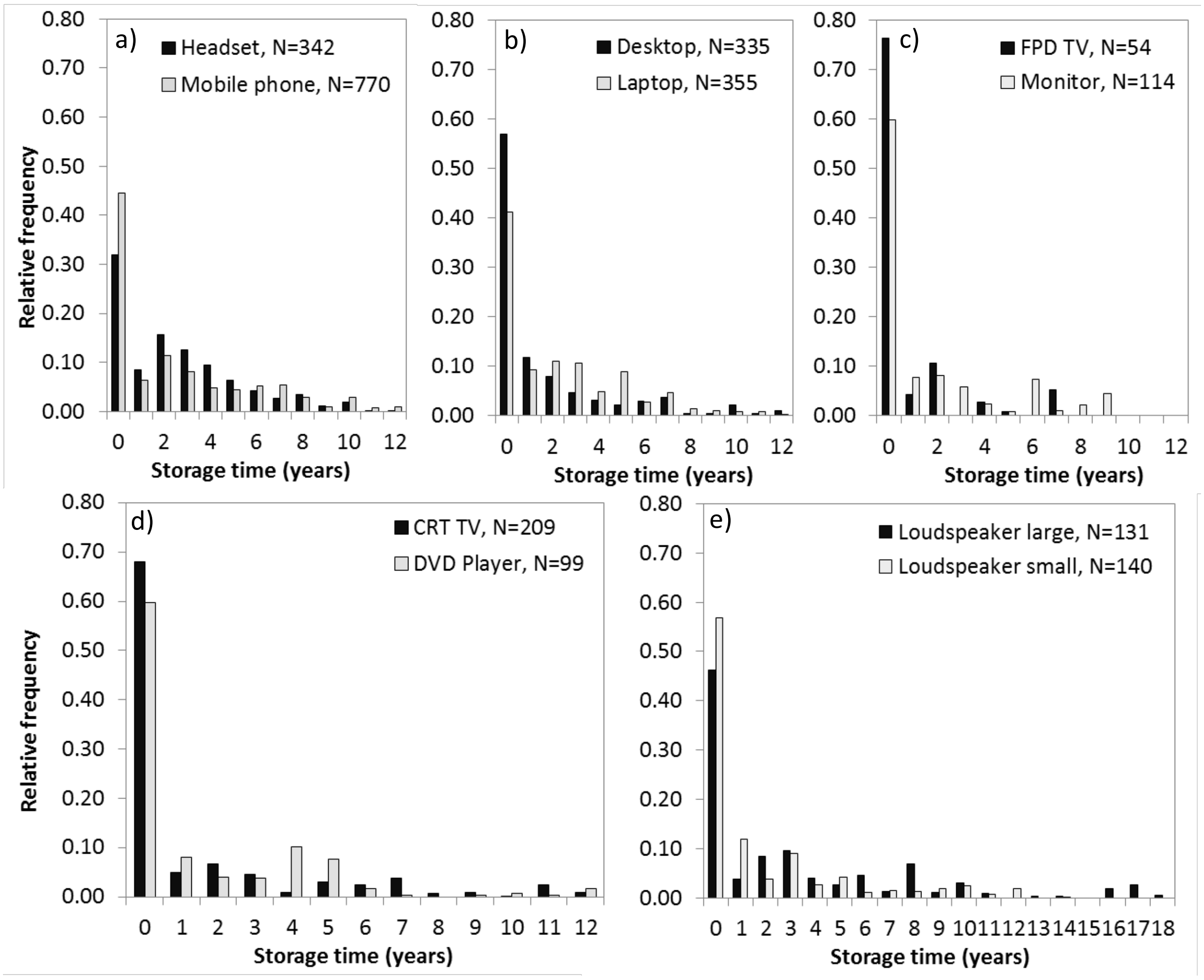

Figure 4.5: Normalized histograms of the storage time for 10 electronic device types: (a) headset and mobilephone; (b) desktop and laptop; (c) FPD TV and monitor; (d) CRT TV and DVD player; and (e) loudspeaker large and loudspeaker small. N denotes the sample size. FPD TV = flat panel display television; CRT TV = cathode ray tube television; DVD = digital video disc.

4.3.3 Reuse, Storage and Disposal Pathways

The transfer coefficients representing the different disposal pathways of the considered device types are listed in Table 4.2. The rate of reuse directly after active use is highest for FDP TVs (30%) and Monitors (20%). Headsets are the least likely to be directly reused with a rate of less than 1%. The share of devices that are stored after their first active use is highest for headsets with 70%. The lowest storage rates can be clearly attributed to CRT TVs and FPD TVs with 30%. Collection rates directly after first use are highest for CRT TVs (55%) and DVD players (42%). After first storage, reuse rates are higher for most device types than directly after active use. However, most devices are brought to the collection system. Exceptions are FPD TVs, whose reuse rates after first storage are higher than the collection rate.

Second-hand devices show similar or even higher storage rates for most device types compared to new devices, ranging from 28% for CRT TVs up to 83% for monitors. Collection rates directly after second use are highest again for CRT TVs (58%) and DVD players (47%), but also high for laptops (44%). After the storage of second-hand devices, again most devices are brought to the collection system. However, for some device types, still 18% to 40% of devices are reused as third-hand devices.

If a device is reused, its average service lifetime is extended by at least 30%. Taking into account the reuse rates up to 30% after the first active use (or up to 80% after the first storage), reuse can thus play a significant role in service lifetime extension.

The transfer coefficients from active use of new devices to storage show that headsets and mobile phones, but also laptops, are more often stored than, for example, desktops, monitors or TVs. This confirms again that size plays a role in storage decisions. The high storage rates after second use may indicate that users of second-hand devices still consider a third use for their devices and therefore rather store them instead of disposing of them immediately. As most devices are brought to the collection scheme after storage, the question arises why they were stored in the first place. The reason for storage was not explicitly sampled by our survey. In their comments, however, participants mention several reasons: usage as replacement devices, data storage, spare part storage, toys for kids, storage due to nostalgic reasons.

The disposal pathways 'donation', 'municipal waste' an 'unknown' do not play a significant role. Only headsets and small loudspeakers show a high disposal rate to the municipal waste directly after the active use as well as after storage.

Table 4.2: Transfer coefficients representing the different disposal pathways of the 10 device types.

From	To	Desktop	Laptop	Monitor	Mobile phone	Headset	CRT TV	FPD TV	Loudspeaker small	Loudspeaker large	DVD player
Active use	Storage	0.43	0.62	0.44	0.58	0.70	0.31	0.27	0.52	0.51	0.47
	Active use 2[a]	0.11	0.12	0.21	0.15	0.002	0.07	0.28	0.02	0.07	0.09
	Donation	0.03	0.02	0.01	0.01	-	0.01	0.02	0.00	0.01	0.00
	Collection	0.39	0.19	0.33	0.22	0.18	0.55	0.25	0.23	0.35	0.42
	Municipal waste	0.01	0.01	-	0.00	0.08	0.01	0.00	0.20	0.02	0.02
	Unknown	0.03	0.04	0.01	0.04	0.04	0.05	0.18	0.02	0.04	0.00
Storage	Active use 2[a]	0.09	0.26	0.11	0.24	0.13	0.21	0.80	0.30	0.16	0.29
	Donation	0.02	0.03	0.07	0.06	-	0.02	-	-	0.04	-
	Collection	0.82	0.67	0.68	0.59	0.51	0.73	0.10	0.38	0.69	0.63
	Municipal waste	0.00	-	0.03	0.02	0.30	-	0.10	0.27	0.07	0.08
	Unknown	0.06	0.04	0.11	0.09	0.07	0.04	-	0.05	0.04	-
Active use 2[a]	Storage 2[b]	0.63	0.40	0.83	0.69	-	0.28	0.53	0.81	0.65	0.45
	Active use 3[c]	0.05	0.08	0.05	0.14	-	0.12	0.10	0.01	0.15	0.08
	Donation	0.02	0.08	-	0.01	-	0.02	0.03	-	0.02	-
	Collection	0.26	0.44	0.11	0.14	-	0.58	0.26	0.18	0.18	0.47
	Municipal waste	-	-	-	0.00	-	0.00	0.06	-	-	-
	Unknown	0.04	-	-	0.02	-	0.01	0.01	-	-	-
Storage 2[b]	Active use 3[c]	0.38	0.34	0.20	0.18	-	0.21	-	-	0.40	-
	Donation	0.07	-	0.14	0.10	-	0.10	-	-	-	-
	Collection	0.51	0.53	0.62	0.54	-	0.62	-	1.00	0.60	1.00
	Municipal waste	0.04	-	-	0.10	-	0.04	-	-	-	-
	Unknown	-	0.13	0.03	0.07	-	0.04	-	-	-	-

- : no data available
[a]Active use 2: active use of second-hand devices
[b]Storage 2: storage of second-hand devices
[c] Active use 3: active use of third hand devices
FPD TV = flat panel display television; CRT TV = cathode ray tube television; DVD = digital video disc.

4.3.4 Comparison to Existing Literature

The comparison of our results to those of similar studies in literature should be interpreted with some caution due to different regional and temporal scopes. Moreover, most studies either focus on service lifetime or total lifetime, which in many cases includes possible storage time. To make comparisons with total lifetime, we thus sum up our service lifetime (of devices no longer in use) and storage time. Figure 4.6 shows our calculated total lifetime as an average over all available sales years, compared to available data from literature. Although our results contain participants' estimates of the expected storage time, they correspond well to existing data. An additional figure and a table with results from existing literature can be found in the supporting information 1 on the Web.

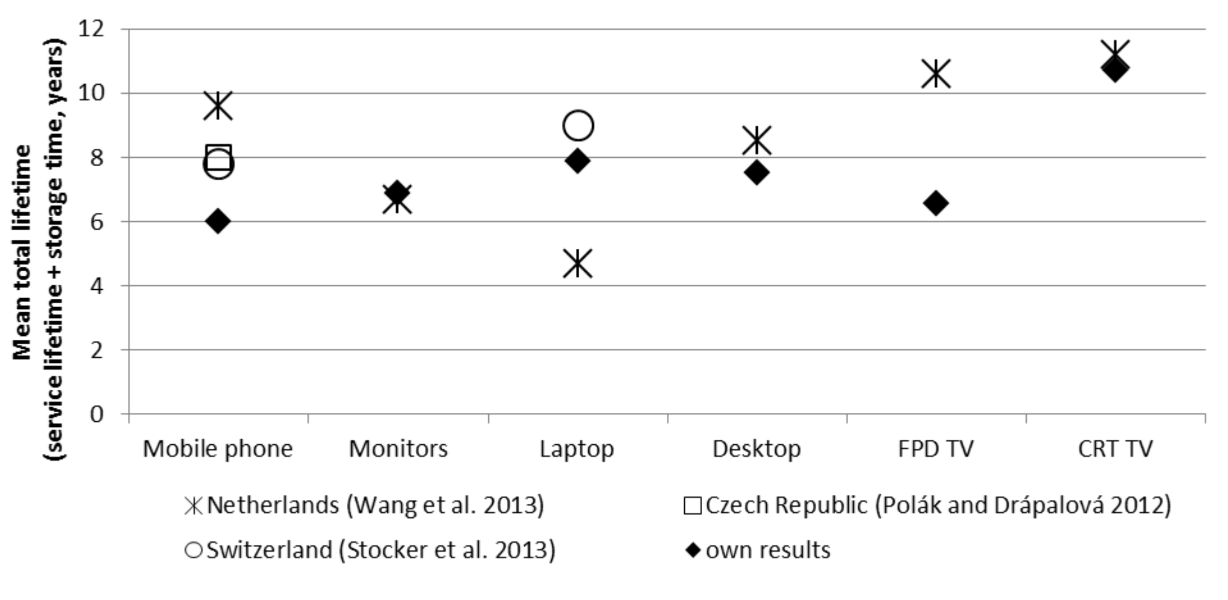

Figure 4.6: Comparison of our calculated total lifetime (on average over all available sales years) to average total lifetimes from existing literature.

4.4 Limitations and Uncertainty

The data on the service lifetime, storage time and disposal pathways were obtained from an online survey and complementary interviews. Survey participants indicated in which year they bought, put to storage or disposed of their devices. Thus, the uncertainty of their information is a maximum of plus or minus one year, depending on when exactly during the year a device was bought etc. Furthermore, as participants were asked not only about present devices, but also on the use, storage and disposal of past devices, the data may partly base on subjective estimates. The collection of data from online surveys has the advantage of fast data access and low resource requirements. The drawbacks are, however, that data quality is difficult to monitor and under-coverage and self-selection, among others factors, may lead to biased data, compared to a truly random sample of respondents of a given population (Bethlehem 2009). The introduction of weighting factors based on the 30 age-gender-education groups partly corrects the bias in our case; however, the weighting can also increase data uncertainty, for example by increasing already questionably high peaks in the survey results.

The number of observations for the second service lifetime and second storage time is quite low for most device types, so the uncertainty of these results is high. In order to avoid biased data due to right censoring, for some evaluations we had to omit data for more recent sales years of each device type sample, which decreases the sample size. Furthermore, as far as temporal trends in the service lifetime and storage time can be observed, the aggregation of all sales years increases the variance in the box-plots and histograms.

However, despite data uncertainty, our research provides an extensive data collection that can be used as a basis for improved future research. The microdata provided are valid for the Swiss context only if the proposed weighting adjustments are applied. The data could be used as a reference for other countries, provided that consumption and use patterns of EE are similar to Switzerland, and appropriate weighting adjustments are applied. For further data evaluation issues such as data uncertainty, overestimation of the remaining lifetime of devices in use, right censoring of data on devices no longer in use, or rough estimates of remaining storage time should be taken into account.

4.5 Conclusion and Outlook

Our results provide new and important insights on product lifetimes and the trigger events to transfer devices between active use, storage and disposal, despite the methodological limitations of our approach as discussed above. Our study is a first step towards a better understanding of the current stocks and flows of EE and improved forecasts of future stocks and flows. It also helps to identify knowledge gaps and potential for future research.

Our results suggest that the assumption of constant service lifetimes of EE over sales years, which is underlying many models in use, is a possible simplification for some device types. The distribution shape of the service lifetime varies significantly among device types and is often not normally distributed. Due to the lack of data, many MFAs of EE apply average service lifetime data. We thus recommend taking into account, if ever possible, device-type dependent and, if applicable, time variant distribution models to treat service lifetime of EE in MFAs adequately.

The storage time is a significant variable when considering the product lifetime of EE. Storage slows down the waste generation as well as the flows to the collection schemes and therefore increases the stock of material resources in households. This is substantiated by our observation that, depending on the device type, 27% to 70% of new devices and 28% to 83% of second-hand devices are stored after their active use and the average storage time accounts for 13% up to 80% of the service lifetime. If storage is more adequately considered in future MFA studies in the electronics sector, it will be possible to reduce the uncertainty about material resources in storage stocks at private households and about the delay of recycling streams.

The analysis of disposal pathways has shown that considerable quantities of devices are reused, which happens either directly after active use (which can be the first or second use) or after storage. The resource savings potentials connected to these pathways to reuse were not addressed in this study but could be the subject of future research.

The quantitative survey did not investigate the reasons for replacing a device, choosing a certain disposal pathway, keeping a device in storage, etc. Such information about the reasoning behind user decisions, however, would facilitate the interpretation of the existing quantitative data and is therefore considered an important area of future research.

Subsequent to this study, we intend to determine current and future return of indium, neodymium and gold contained in EE in Switzerland, based on past and assumed or extrapolated future sales flows of the different device types.

4.6 Acknowledgments

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Supporting Information

Supporting information is linked to this article on the JIE website:

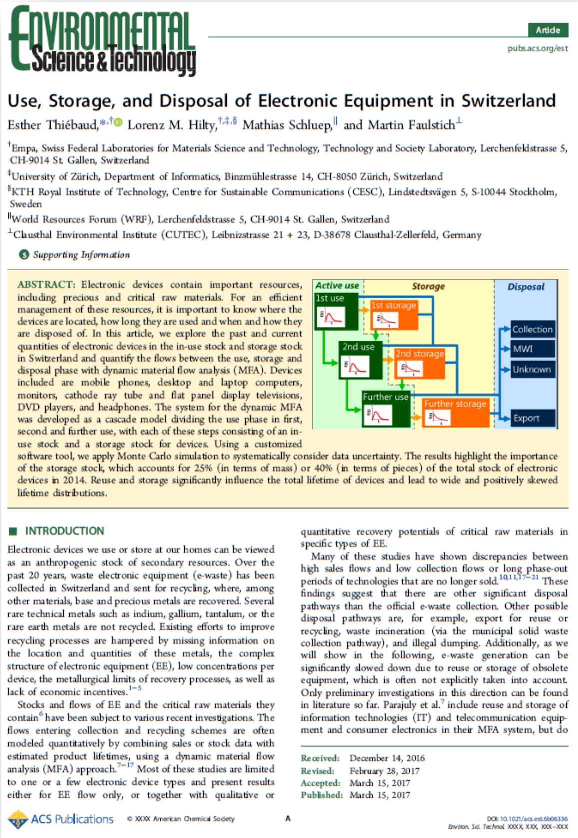
Supporting Information S1: This supporting information includes the online survey used to determine how long devices are used currently, how long they are stored, and how they have been disposed of. It also includes the basic demographic questions asked of each survey participant. The authors explain their methods of data evaluation, including the use of weighted adjustments to account for under- and over-represented subgroups in the study, data corrections to account for user estimates of how long they will use current devices, and methods for evaluating temporal change in device usage/storage and disposal pathways. Results are presented for average device service lifetime, storage lifetime, and temporal changes in both. Lifetime data are also compared to data from existing literature.

Supporting Information S2: This supporting information includes microdata from the study's online survey to determine service lifetimes, storage times, and disposal pathways of electronic equipment.

Chapter 5

Use, Storage, and Disposal of Electronic Equipment in Switzerland

Esther Thiébaud, Lorenz M. Hilty, Mathias Schluep, Martin Faulstich



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Use, Storage, and Disposal of Electronic Equipment in Switzerland

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Supporting Information

ABSTRACT: Electronic devices contain important resources, including precious and critical raw materials. For an efficient management of these resources, it is important to know where the devices are located, how long they are used and when and how they are disposed of. In this article, we explore the past and current quantities of electronic devices in the in-use stock and storage stock in Switzerland and quantify the flows between the use, storage and disposal phase with dynamic material flow analysis (MFA). Devices included are mobile phones, desktop and laptop computers, monitors, cathode ray tube and flat panel display televisions, DVD players, and headphones. The system for the dynamic MFA was developed as a cascade model dividing the use phase in first, second and further use, with each of these steps consisting of an in-use stock and a storage stock for devices. Using a customized software tool, we apply Monte Carlo simulation to systematically consider data uncertainty. The results highlight the importance of the storage stock, which accounts for 25% (in terms of mass) or 40% (in terms of pieces) of the total stock of electronic devices in 2014. Reuse and storage significantly influence the total lifetime of devices and lead to wide and positively skewed lifetime distributions.

■ INTRODUCTION

Electronic devices we use or store at our homes can be viewed as an anthropogenic stock of secondary resources. Over the past 20 years, waste electronic equipment (e-waste) has been collected in Switzerland and sent for recycling, where, among other materials, base and precious metals are recovered. Several rare technical metals such as indium, gallium, tantalum, or the rare earth metals are not recycled. Existing efforts to improve recycling processes are hampered by missing information on the location and quantities of these metals, the complex structure of electronic equipment (EE), low concentrations per device, the metallurgical limits of recovery processes, as well as lack of economic incentives.[1−5]

Stocks and flows of EE and the critical raw materials they contain[6] have been subject to various recent investigations. The flows entering collection and recycling schemes are often modeled quantitatively by combining sales or stock data with estimated product lifetimes, using a dynamic material flow analysis (MFA) approach.[7−17] Most of these studies are limited to one or a few electronic device types and present results either for EE flow only, or together with qualitative or quantitative recovery potentials of critical raw materials in specific types of EE.

Many of these studies have shown discrepancies between high sales flows and low collection flows or long phase-out periods of technologies that are no longer sold.[10,11,17−21] These findings suggest that there are other significant disposal pathways than the official e-waste collection. Other possible disposal pathways are, for example, export for reuse or recycling, waste incineration (via the municipal solid waste collection pathway), and illegal dumping. Additionally, as we will show in the following, e-waste generation can be significantly slowed down due to reuse or storage of obsolete equipment, which is often not explicitly taken into account. Only preliminary investigations in this direction can be found in literature so far. Parajuly et al.[7] include reuse and storage of information technologies (IT) and telecommunication equipment and consumer electronics in their MFA system, but do

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Abstract

Electronic devices contain important resources, including precious and critical raw materials. For an efficient management of these resources, it is important to know where the devices are located, how long they are used and when and how they are disposed of. In this article, we explore the past and current quantities of electronic devices in the in-use stock and storage stock in Switzerland and quantify the flows between the use, storage and disposal phase with dynamic material flow analysis (MFA). Devices included are mobile phones, desktop and laptop computers, monitors, cathode ray tube and flat panel display televisions, DVD players, and headphones. The system for the dynamic MFA was developed as a cascade model dividing the use phase in first, second and further use, with each of these steps consisting of an in-use stock and a storage stock for devices. Using a customized software tool, we apply Monte Carlo simulation to systematically consider data uncertainty. The results highlight the importance of the storage stock, which accounts for 25% (in terms of mass) or 40% (in terms of pieces) of the total stock of electronic devices in 2014. Reuse and storage significantly influence the total lifetime of devices and lead to wide and positively skewed lifetime distributions.

Keywords: In-use stock; storage stock, disposal pathways, lifetime, dynamic material flow analysis

5.1 Introduction

Electronic devices we use or store at our homes can be viewed as an anthropogenic stock of secondary resources. Over the past 20 years, waste electronic equipment (e-waste) has been collected in Switzerland and sent for recycling, where, among other materials, base and precious metals are recovered. Several rare technical metals such as indium, gallium, tantalum, or the rare earth metals are not recycled. Existing efforts to improve recycling processes are hampered by missing information on the location and quantities of these metals, the complex structure of electronic equipment (EE), low contents per device, the metallurgical limits of recovery processes, as well as lack of economic incentives.[1–5]

Stocks and flows of EE and the critical raw materials they contain[6] have been subject to various recent investigations. The flows entering collection and recycling schemes are often modeled quantitatively by combining sales or stock data with estimated product lifetimes, using a dynamic material flow analysis (MFA) approach.[7–17] Most of these studies are limited to one or a few electronic device types and present results either for EE flow only, or together with qualitative or quantitative recovery potentials of critical raw materials in specific types of EE.

Many of these studies have shown discrepancies between high sales flows and low collection flows or long phase-out periods of technologies that are no longer sold for certain device types.[10,11,17–21] These findings suggest that there are other significant disposal pathways than the official e-waste collection. Other possible disposal pathways are, for example, export for reuse or recycling, waste incineration (via the municipal solid waste collection pathway), and illegal dumping. Additionally, as we will show in the following, e-waste generation can be significantly slowed down due to reuse or storage of obsolete equipment, which is often not explicitly taken into account. Only preliminary investigations in this direction can be found in literature so far. Parajuly et al.[7] include reuse and storage of IT and telecommunication equipment and consumer electronics in their MFA system, but do not quantify related stocks and flows. Similarly, Yoshida et al.[16] mention both storage and reuse of computers, but only quantify domestic reuse flows. Chung et al.[22] evaluate the amount of stored televisions (TVs) and computers in China. The amount of stored TVs and e-waste in general in U.S. households is analyzed by Milovantseva et al.[23] and Saphores et al.[24]. Polák and Drápalová[14] include storage times for mobile phones and Sabagghi et al.[25], as well as Williams and Hatanaka[26] report storage times of personal computers in their studies. Steubing et al.[15] include reuse and storage times in their MFA of computers and monitors, and quantify related flows. To the best of the authors' knowledge, there is, however, no dynamic MFA research that includes reuse and storage times and differentiates between in-use, reuse and storage stocks and the related flows of EE.

In an attempt to close this research gap, we conducted a survey on the service lifetime, storage time, and disposal pathways of EE in Switzerland between 2014 and 2016. The findings highlight the influence of storage and reuse on the time a product remains in the use phase[27]. In this article, we present a dynamic MFA of nine different types of EE in Switzerland. We develop a model to identify past and current in-use stocks and storage stocks of EE and quantify in detail the flows between and from the use, storage and disposal phases. Based on the in-depth "bottom-up" data we are able to differentiate between new and (re)used devices. Such a model is a significant step toward a deeper understanding of the behavior of device types in the use phase, such as the above-mentioned discrepancy between high sales flows and low collection flows. It facilitates detecting leakages where devices and the incorporated resources leave the system, as well as determining their final sink. In combination with assumed or extrapolated sales flows, the model can also serve to forecast the composition of future recycling flows, a capability which enables recycling system managers to provide appropriate and tailored recycling capacities and technologies.[27]

To implement the model, we developed an open source software tool in Python 3, allowing for the calculation of dynamic MFAs in a generic and flexible way. The tool implements 'dynamic probabilistic material flow analysis', a combination of dynamic material flow modeling with probabilistic modeling, as proposed by Bornhöft.[28] The probabilistic approach allows to systematically dealing with data uncertainty in our model.

5.2 Materials and Method

This chapter is structured according to the ODD (overview, design concepts, details) protocol as proposed in Müller et al.[29]

5.2.1 Overview

The purpose of the dynamic MFA model is to quantify in-use stocks and storage stocks of nine types of EE with a high content of indium, neodymium or gold, so that they cumulatively cover around 90% of these three metals in private Swiss households[30] (see Table 5.1) and to map and evaluate in detail the flows between the use, storage and disposal phases.

The MFA system includes the process "use phase", divided into "active use" and "storage" (Figure 5.1). The "active use" and "storage" are further divided into the subprocesses "first use", "second use" and "further use" as well as "first storage", "second storage" and "further storage". Each subprocess includes an in-use stock or storage stock. The system boundary corresponds to the Swiss national border. The temporal extent is different for each device type. It starts when an electronic device type was first put on the market and ends with the year 2014. The temporal scale is one year.

Based on the empirical results of our survey, we developed a cascade model, with each step consisting of an in-use stock and storage stock. The first step includes the first use and storage of new devices, the second step the second use and storage of second-hand devices.

Table 5.1: Electronic device types included in the MFA

Device type	Description	Fraction containing			UNU-Key
		Indium	Neodymium	Gold	
Desktop	Desktop computer (incl. all-in-one computer, excl. peripherals)		Magnets in hard disk drive (HDD), optical drive[30,31] / printed wiring board (PWB)[32,33]	PWB[34]	0302
Laptop	Laptop computer/ Notebook	Liquid crystal display (LCD)[35]	Magnets in HDD, optical drive, loudspeaker[30,31] / PWB[32,33]	PWB[34]	0303
Monitor	Flat panel display (FPD) monitor	LCD[35]	PWB[32,33]	PWB[34]	0309
Mobile phone	Conventional mobile phone	LCD[35]	Magnets in loudspeaker, vibration alarm[30] / PWB[32]	PWB[34]	0306
Smartphone	Smart phone	LCD[35]	Magnets in loudspeaker, vibration alarm[30] / PWB[32]	PWB[34]	0306

Headset	Headphones / Headset		Magnets in Loudspeaker[30,36]		0401
CRT TV	Cathode ray tube (CRT) TV		PWB[32,33]	PWB[34]	0407
FPD TV	Flat panel display (FPD) TV	LCD[35]	PWB[32,33]	PWB[34]	0408
DVD player	DVD player / Blu-ray player		Magnets in optical drive[37] / PBW[32,33]	PWB[34]	0404

Unlike Thiébaud et al.[27], we do not include loudspeakers, as no sales data are available for such devices.

The third and final step summarizes all possible further uses and storages. The cascade model enables the representation of different service lifetimes, storage times and disposal pathways both for new and for used devices.

The system (see Figure 5.1) has one inflow corresponding to the sales of new devices and four outflows, one for each disposal option. Internal flows include "reuse" flows from in-use stock and storage stock to the next in-use stock and "storage" flows from in-use stock to storage.

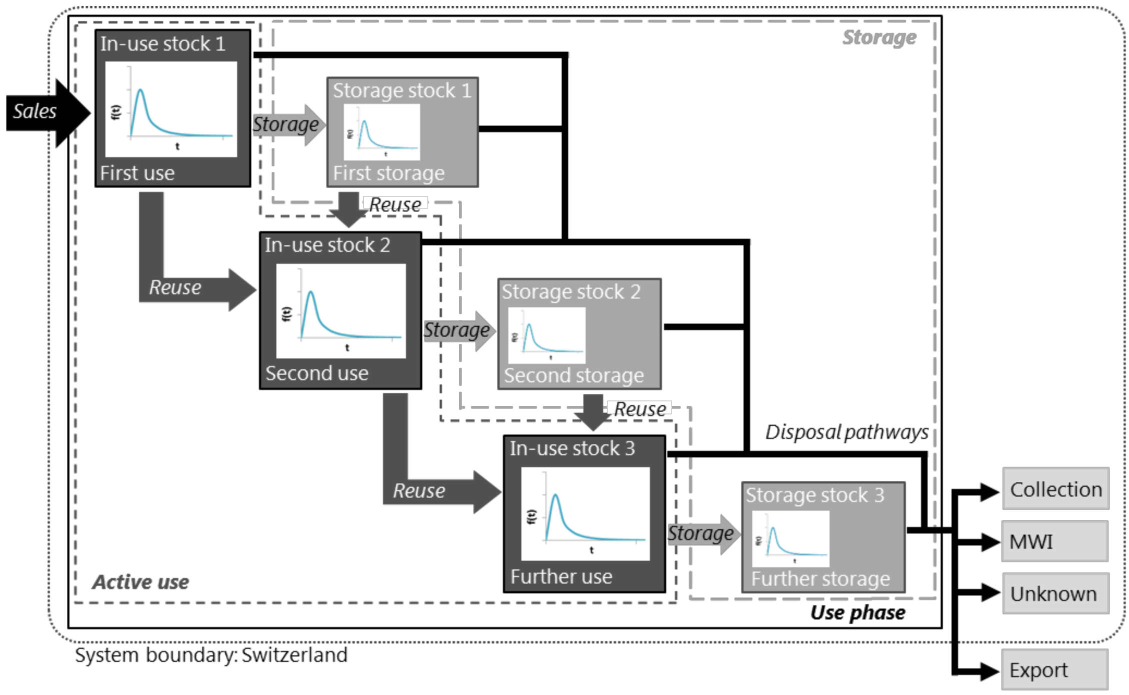

Figure 5.1: Cascade model of the process "use phase", divided into "active use" and "storage".

5.2.2 Design Concepts

The model employs a retrospective top-down approach, deriving the stock S[n] at a time n from the net flow by using the balance of masses (eq (1)), with the constant sampling rate T = 1 year.[29]

$$S[n] = (inflow[n] - outflow[n]) \cdot T + S[n-1] \quad (1)$$

The outflows of the in-use stock and the storage stock are calculated according to eq (2).[29]

$$outflow[n] = \sum_{m=0}^{\infty} inflow[n-m] \cdot f[m] \quad (2)$$

The model applies different lifetime distribution functions f[m] for the service lifetimes and storage times of new and used devices. The service lifetime is defined as *the time of active use of a device*.

The storage time is defined as *the time between the end of the active use of a device and its final disposal or transfer to a different user*.[27] The flows from the different stocks to the in-use stock, storage, or disposal pathways are determined by transfer coefficients. All data include changes over time provided that time series are available.

We accounted for data uncertainty by modeling inflows, transfer coefficients and mass of devices as probability distributions. We chose either normal or triangular distributions, based on the characteristics of the available data. The dependent variables are calculated by Monte Carlo simulation and again provided as probability distributions.[28,38] The uncertainty of the lifetime distribution parameter was taken into account by conducting a sensitivity analysis with a lower and upper lifetime distribution scenario.

5.2.3 Details

5.2.3.1 Initial Condition

In our model, stocks of EE are calculated from inflow data and lifetime distribution functions (eq (1) and (2)), starting from a stock = 0. For sales data, the model thus demands information back to the year when a device type was first brought onto the market. In cases where such data was not available, we extrapolated existing sales figures.

5.2.3.2 Model Input Data

Inflow data for desktops, laptops and flat screen monitors were taken from annual ICT market reports for Switzerland, which include both the business and the home segment.[39] Sales data of television sets (CRT and FPD TVs) and DVD players were collected by the Swiss Consumer Electronics Association[40] and the market research company GfK.[41] GfK additionally reports sales data for mobile phones and smartphones.[41,42] We further assumed that the number of sold headsets corresponds to the sum of all sold portable audio and video devices, smart phones and mobile phones. The uncertainties of inflow data were modeled as normal distributions. The standard deviation (SD) was estimated at 10% of the inflow value for time series with available data from Weissbuch, GfK or SCEA.[43] For extrapolated time series, we assumed a SD of 20%. As the inflow data for headsets are based on rough estimations, we assumed a SD of 30%.

Data regarding the service lifetime, the storage time, and the disposal pathways of EE were taken from Thiébaud et al.[27] They are provided as histograms of the service lifetimes and storage times of new and second hand devices of ten different EE types. The temporal change of the service lifetime of new devices was included where appropriate by dividing the histograms into sales year groups as defined in Thiébaud et al.[27] For the temporal change of the storage time of new devices, we calculated for each device the year it was put to storage and subsequently divided the histograms into storage year groups. For the third step of the cascade model, no data on the service lifetime or storage time are available, although the survey data confirms that for most device types, there are some second-hand devices that are further used.[27] Due to the lack of data, we assumed that the same service lifetimes and storage times as for the second use apply for all further uses in total (modeled as the third step of the cascade model). This assumption is tested by a sensitivity analysis.

In order to estimate the lifetime distribution functions for the dynamic MFA, we fitted Weibull distribution functions to the normalized histograms of the service lifetimes and storage times of new and second-hand devices. Modeling positively skewed distributions of product lifetimes is typically done with Weibull distributions, as they can adopt many different shapes.[44,45]

The uncertainty of the service lifetime and storage time is at least ±1 year for all histograms.[27] The tool developed for the simulation of the dynamic MFA supports probabilistic modeling of the inflows and transfer coefficients, but not of lifetime distribution parameters. In order to evaluate the sensitivity of our model to the Weibull distribution parameters as well, we shifted the histograms provided by Thiébaud et al.[27] by ±1 year and fitted the Weibull distribution functions again. The model was then run additionally with the resulting lower and upper lifetime distribution scenarios for each device type.

Thiébaud et al.[27] further provide transfer coefficients of the flows between the processes "active use" and "storage" as well as to the different disposal pathways: "donation", "collection", "municipal waste" and "unknown". We modeled the "donation" option as an export flow, as most donation projects for used electronics are situated abroad.[46,47] Due to the limited amount of available data sets, the temporal change of transfer coefficients was analyzed for CRT TVs, mobile phones, laptops and desktops only. For the third step of the cascade model, the transfer coefficients for second-hand devices were adapted by splitting the reuse flows proportionally to the other disposal pathways. As the uncertainty of the transfer coefficients increases with smaller sample sizes per process, we introduced a triangular distribution for each transfer coefficient, varying with $\pm\frac{1}{\sqrt{n}}$, with n being the total number of devices transferred from a specific process.

To simulate the distributions of total lifetime, that is, the time devices remain in the use phase from sales to disposal, we run an impulse response analysis. For each device type, we fed an impulse of 100 000 devices in one year into the cascade model and simulated the succeeding periods of 50 years. This includes all different service lifetimes and storage times, as well as the percentage of products that enter reuse and storage according to the transfer coefficients provided by Thiébaud et al.[27] The response at the outflow of the overall use phase, corresponding to the sum of all four disposal pathways and adding up to the 100 000 devices, is equivalent to the total lifetime distribution function.

The average mass per device of a given type was determined by own measurements and literature data.[9,17,34,48,49] We introduced a triangular distribution for the mass of each device type, determined by the weighted mean and the lowest and highest value. The lowest and highest values were extended by taking into account the SD of the SD, as suggested in JCGM, [50] so that the lower and upper limit of the triangular distribution should lie within the 95% confidence interval. For the mass per device type, available data did not show any significant temporal change.

More information on the inflow data, an overview of all fitted Weibull distribution functions including details on the sales year and storage year groups, the sensitivity analysis of the third step of the cascade model, the transfer coefficients, the impulse response analysis, and the uncertainty distributions of mass per device type can be found in the Supporting Information (SI).

5.2.3.3 Model Output Data and Evaluation

Model output data comprise stochastic time series of all stocks and flows occurring in Figure 1. The distribution of the time series is visualized by the 10th percentile, mean, and 90th percentile. Additionally, the lower, mean and upper lifetime distribution scenarios as described above are presented. With the exception of Sankey diagrams, the in-use stocks 1, 2 and 3 and storage stocks 1, 2, and 3 are aggregated to the total in-use stock and total storage stock.

The output can be compared to statistical data from various sources for model validation. From the Swiss Federal Statistical Office (FSO), time series data on the in-use stocks of desktops, laptops, CRT TVs, FPD TVs and DVD players are available from 2006 to 2013.[51] The Swiss Federal Office of Communications (OFCOM) provides in-use stock data for mobile phones from 1990 to 2014[52]. Swico Recycling, the collection system for EE in Switzerland, publishes numbers and tonnages of desktops, FPD monitors, laptops, mobile phones, CRT and FPD TVs they collected from 2010 to 2014.[48] The uncertainty of these statistical data is not quantified by their providers but it is assumed to be considerably high. These data are compared to the simulated stocks and outflows to collection.

5.2.3.4 Detailed Model Description

The conceptual model, as described in Figure 1, was implemented using our own MFA simulation tool called "pymfa" (written in Python 3, using the numpy, scipy, and matplotlib libraries), which supports the calculation of dynamic MFAs in a flexible way. The core of the tool can be used as a Python library and provides the necessary functionality to run analyses through a command line interface and an interactive web application that can also be run locally. Model descriptions are specified using a simple data-driven approach: the researcher creates a spreadsheet where each row represents a link in the MFA. Links can represent inflows, unit conversions, transfer rates, delays or the split of a fraction entering a process into multiple fractions leaving the process. For each link, time series of data including information on the probabilistic distribution have to be provided. As such, model descriptions can simply be stored and forwarded as spreadsheets or CSV files. Model description source files can be uploaded through the web interface, upon which the tool calculates the time series of resulting stocks and flows by means of Monte Carlo simulation, visualizes them and offers the resulting data as a CSV download. A screenshot of the web interface as well as an example of a model description source file and output plots can be found in the SI.

For each device type, the model was simulated for numbers of devices and for physical mass, producing results in pieces and in kg, respectively. In addition to the standard scenario, we run a lower and upper lifetime scenario and an impulse scenario for each device type. Each simulation run was repeated 10000 times, which we considered a sufficient sample size for the Monte Carlo simulation

5.3 Results and Discussion

5.3.1 Total Lifetime

Reuse and storage significantly extend the time a device remains in the use phase. Compared to the median service lifetime of new devices, the median total lifetime, derived from the impulse response analysis, increases, for example, from 3 to 7 years for conventional mobile phones and smartphones, from 5 to 8 and 9 years for desktops and laptops, respectively, and from 9 to 11 years for CRT TVs. Existing studies from The Netherlands[53] and Switzerland[54] show similar results regarding the average or median total lifetime. The median and average total service lifetimes as well as a comparison to existing studies are listed in Table C.6 in the SI.

The total lifetime distribution functions, compared to the service lifetime distribution functions of new devices, show a shifted mode toward longer lifetimes, and a wider and more positively skewed distribution. An example of four device types is illustrated in Figure 5.2 (for the remaining distributions see Figure C.22 in the SI). Only few sources present Weibull parameter for the total lifetime of a large range of EE.[17,53] As we assume that Dutch usage patterns are similar to those of the Swiss, we compare our results to Wang et al.[53] Their Weibull distribution functions for the year 2005 match well our findings for most device types. Exceptions are laptop computers, with a Weibull distribution function more similar to our service lifetime distribution, and mobile phones, with a sharply and monotonically decreasing Weibull distribution function. The blue and the red curve for mobile phones have similar means (9.6 and 8.5 years, respectively) but different medians (4.5 and 7.0 years, respectively). As both laptops and mobile phones are often stored and reused in Switzerland, our findings of wider and more positively skewed distributions seem to be more realistic.

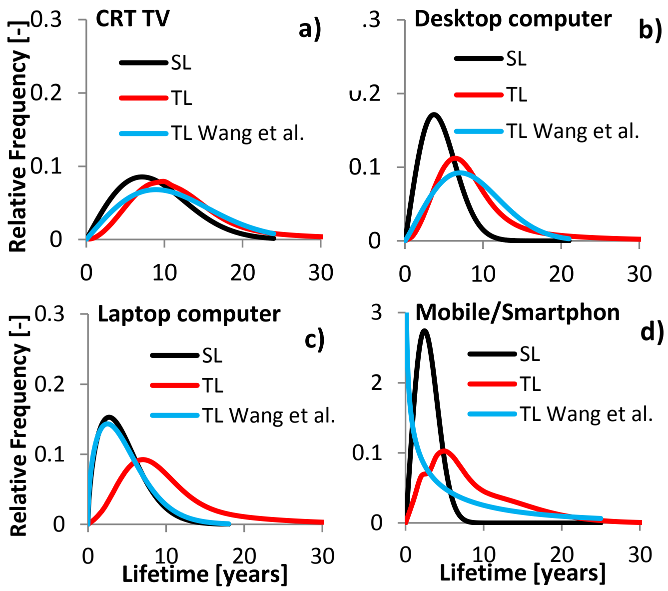

Figure 5.2: Comparison of own results of service lifetime (SL) with total lifetime (TL) and TL of Wang et al.,[53] for (a) cathode ray tube television (CRT TV), (b) desktop computer, (c) laptop computer and (d) mobile phone/smartphone.

The results show that data on mean or median lifetimes do not provide conclusive information on the actual lifetime distribution function. They further highlight the importance of including flexible lifetime distribution functions in dynamic MFA models of products that are reused and stored. Assuming average, median or normally distributed lifetimes may, in case of technology change, often result in underestimated phase-out times. Whether lifetime distributions functions are favorably divided into different service lifetime and storage time distribution functions or merged to total lifetime distribution functions depends on the question to be answered and the required level of detail.

5.3.2 Stocks and Flows

The cascade model produces a cumulated in-use stock and storage stock as illustrated in Figure 5.3 for nine device types between 1980 and 2014.

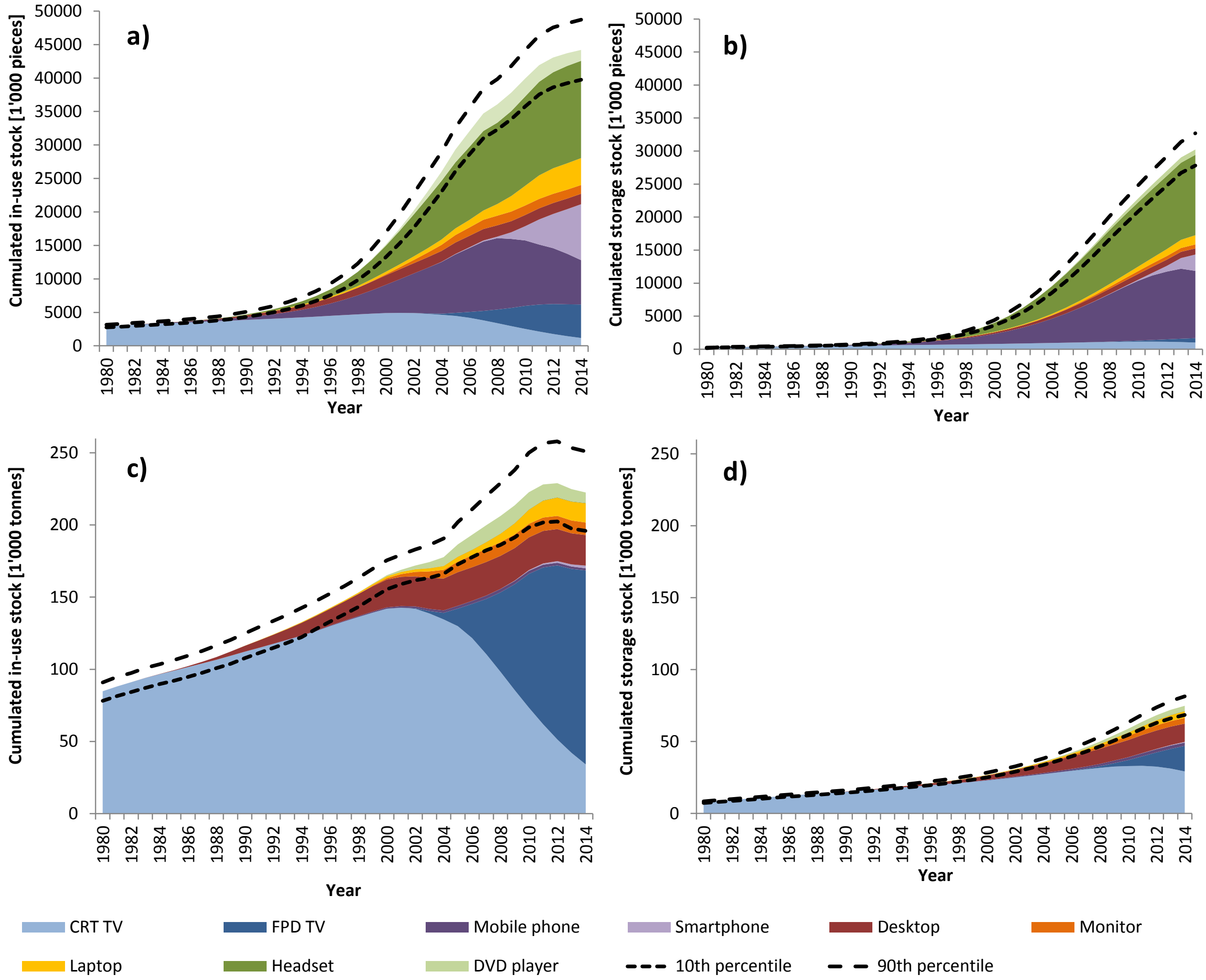

Figure 5.3: (a) Cumulated in-use stock in 1000 pieces, (b) cumulated storage stock in 1000 pieces, (c) cumulated in-use stock in 1000 tonnes and (d) cumulated storage stock in 1000 tonnes, for nine device types between 1980 and 2014

The cumulated in-use stock has grown in the past 20 years to around 44 million devices or 220 000 tonnes in 2014. In terms of devices, the 10th and 90th percentiles, resulting from the Monte Carlo simulation, account for ±10% or a range of roughly 9 million devices. As the mass of each device type increases uncertainty, the 10th and 90th percentile for the in-use stock in tonnes accounts for ±12%. The storage stock comprises 30 million ±8% devices or 75 000 ±9% tonnes in 2014. The storage stock thus accounts for 40% of the total stock in terms of number of devices and 25% in terms of mass. Both in-use stock and storage stock are dominated by small devices such as mobile phones, smartphones and headsets. Regarding mass, however, these devices types are insignificant and CRT as well as FPD TVs and desktop computers are prevalent. The in-use stock of CRT TVs and conventional mobile phones is declining as these device types are replaced by FPD TVs and smartphones, respectively. The stock of smartphones is increasing with a growth rate of 20 – 30%, while the in-use stocks of most other device types are slowly approaching or have already reached saturation. Thus, the overall growth rate of the in-use stock and storage stock in pieces as well as the storage stock's mass have been declining in the past 10 years. Mostly due to the replacement of CRT TVs with lighter FPD TVs, the in-use stock's mass is declining.

The simulated stocks and flows to, from and within the cascade model are depicted in Figure 5.4 for mobile phones and FPD TVs in 2014. For the sake of clarity, we refrain from including uncertainty ranges here. The Sankey diagrams for the remaining seven device types can be found in the SI (Figures C.23-C.29). The disaggregation of the total stock to the in-use stock and storage stock of new devices (cascade step 1), second-hand devices (cascade step 2) and all the rest (cascade step 3) shows large discrepancies among device types. Mobile phones, CRT TVs and headsets have a total storage stock that is larger than or close to the size of the total in-use stock. The smallest total storage stock, compared to the total in-use stock, have FPD TVs and smartphones. The in-use stock 2 of DVD players, CRT TVs and mobile phones accounts for over 15% of the total stock in 2014, while for smartphones and headsets, it is below 5%. Various trends can be derived from these findings: device types that are phased out, such as CRT TVs, mobile phones but also DVD players (replaced by streaming services) are more frequently stored or used as second-hand devices. New device types such as smartphones or FPD TVs are still located mostly in the in-use stock 1. Furthermore, small devices such as mobile phones and headsets are more frequently stored than large devices.27 The storage stock 2 compared to the total stock are largest for monitors, mobile phones and desktops. The decision to store a second-hand device therefore does not seem to be influenced by the size of the device, but more by the prospective utility. The small in-use stock 3 and storage stock 3 show that further uses occur, but are irrelevant.

Flows to collection are largest for all device types. Flows to municipal waste incineration (MWI) are only relevant for headsets. With 20-30% of the total outflow, the share of FPD TVs, mobile phones and smartphones entering unknown disposal pathways is high. Unknown flows include, for example, lost or stolen devices and devices disposed of by others than the survey respondents. It is thus possible that material reaching unknown disposal pathways still ends up in the collection system. Export flows are highest for laptops and mobile phones. Total outflows compared to sales flows are by a factor 3 to 7 higher for mobile phones and DVD players. For desktops, monitors and headsets, sales and outflows are of the same magnitude. For laptops, FPD TVs and smartphones, sales are still larger by a factor of 1.2, 2 and 5, respectively. Despite long delays due to reuse and storage, this comparison thus reveals for which device types the system is currently saturated. These delays, however, result in still growing outflows, with an average growth rate of 5% over the last 20 years. Figure C.30 in the SI shows the cumulated flows to collection between 1980 and 2014. In total, the nine device types result in 24 000 tonnes ±10% that reached the collection system in 2014, dominated by CRT TVs, FPD TVs and desktop computers.

From a resource point of view, it is not important how long products are delayed in the use phase, as long as they reach the collection system in the end, which is the case for 80% of all devices on average. However, if we would aim for more reuse or refurbishment, it would be important that devices are not stored for too long, as newer devices are more in demand on the second-hand market.

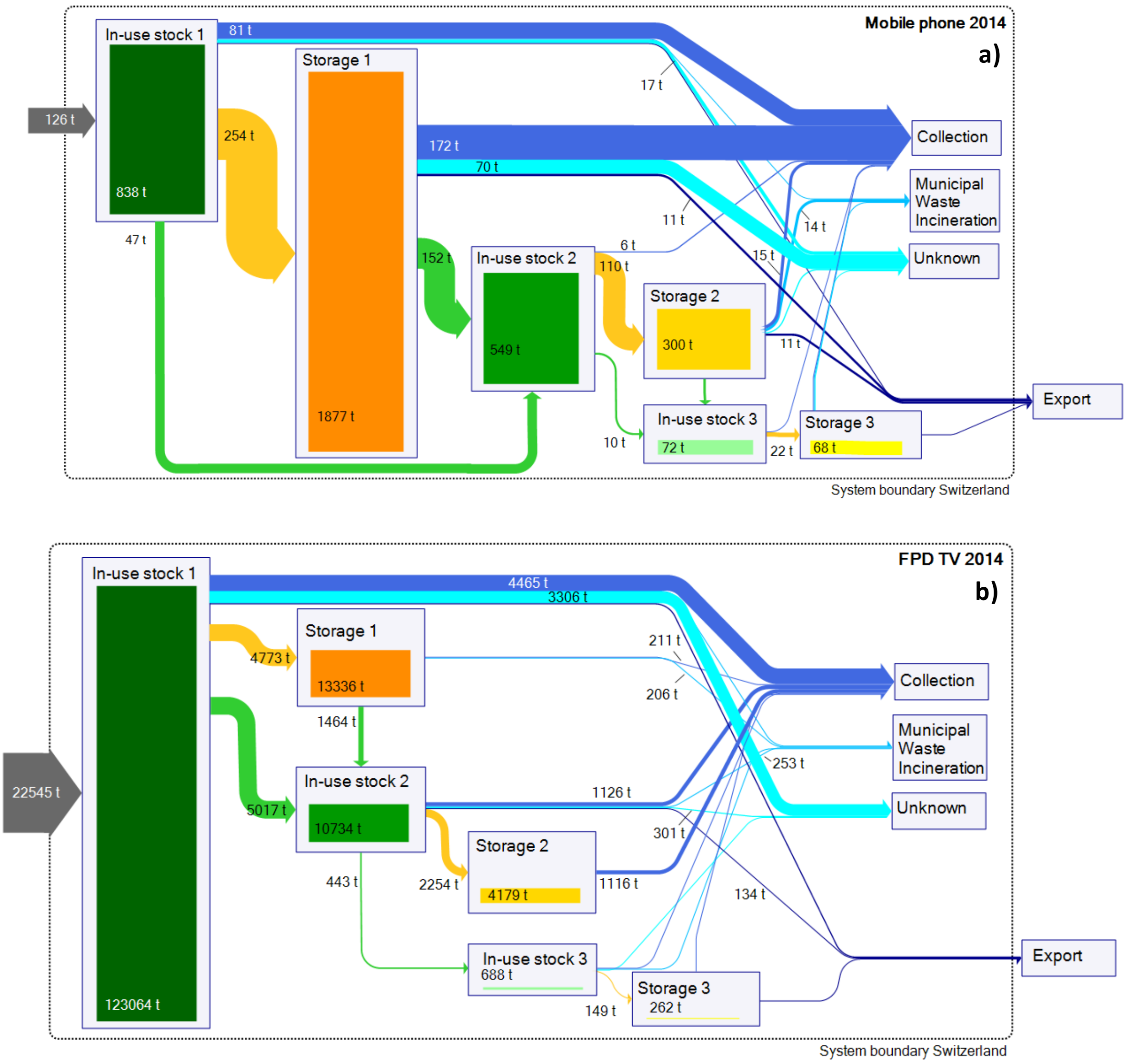

Figure 5.4: Stocks and flows of (a) mobile phones and (b) flat panel display televisions (FPD TVs) in 2014 in tonnes. Mobile phone flows below 5 t and FPD TV flows below 100 t are not labeled.

5.3.3 Model Sensitivity and Validation

The 10th and 90th percentiles of the in-use stock indicate an uncertainty of around ±10% for most device types (Figure 5.3). If the mass per product with its associated uncertainty is included in the model, the uncertainty range increases up to ±12%. The lower and upper lifetime distribution scenarios of the sensitivity analysis add a further ±10% deviation from the standard lifetime distribution scenario. Considering the 10th and 90th percentiles of the lower and upper scenario, the maximum deviation from the mean in-use stock of the standard scenario amounts to approximately ±30% (Figure 5.5). The storage stock is less sensitive with ca. ±20% maximum deviation.

The sensitivity of the flows is presented using the collection flows as an important example. As longer service lifetimes and storage times lead to smaller collection flows, the lower scenario accounts for the highest flows and vice versa. The deviations between the means of the standard, lower and upper lifetime distribution scenario are similar to the deviations of the 10th and 90th percentiles of the standard scenario (ca. ±10%). The maximum deviations between the mean of the standard scenario and the 10th and 90th percentiles of the lower and upper scenario account for ±20%.

By comparing the total in-use stock and the total collection flow with available data from FSO and Swico Recycling, we evaluated to what extent the cascade model is able to reproduce the behavior of the real system. As can be seen in Figure 5.5, the model seems to overestimate the total in-use stock. The sum of FSO and OFCOM data lies in the vicinity of the 10th percentile of our standard scenario and between the mean and the 90th percentile of our lower scenario. A potential explanation is that the service lifetimes and storage times we assumed in the model are longer than those in the real system. However, the comparison of collection flows shows that fewer devices are collected compared to the modeled collection flow, which could be, in contrast, an argument for longer service lifetimes and storage times. Again, the sum of devices collected by Swico Recycling lies in the vicinity of the 10th percentile of our standard scenario, but now also between the mean and the 90th percentile of our upper scenario. Breaking this down to single device types, the discrepancies between modeled and reported collection flows are largest for mobile phones, with 60% less collected than what the model calculates, even though we included long reuse and storage times. As our model for mobile phones overestimates the in-use stock by about 20%, we argue against extending the service lifetime in our model. Whether we assumed too few mobile phones being stored for a too short time, or a large amount of them is exported, could not be investigated within this study. Lange[19] or Huisman et al.[18], among others, report transboundary shipment of EE to Eastern Europe or Africa from Germany and The Netherlands. Although Switzerland has higher EE collection rates than most EU Member States, it might still be possible for small devices such as mobile phones to be formally or informally exported without our knowledge. It is well-known that mobile phones are the most difficult devices for collection schemes to get hold of, with already various studies investigating potential causes.[14,55,56]

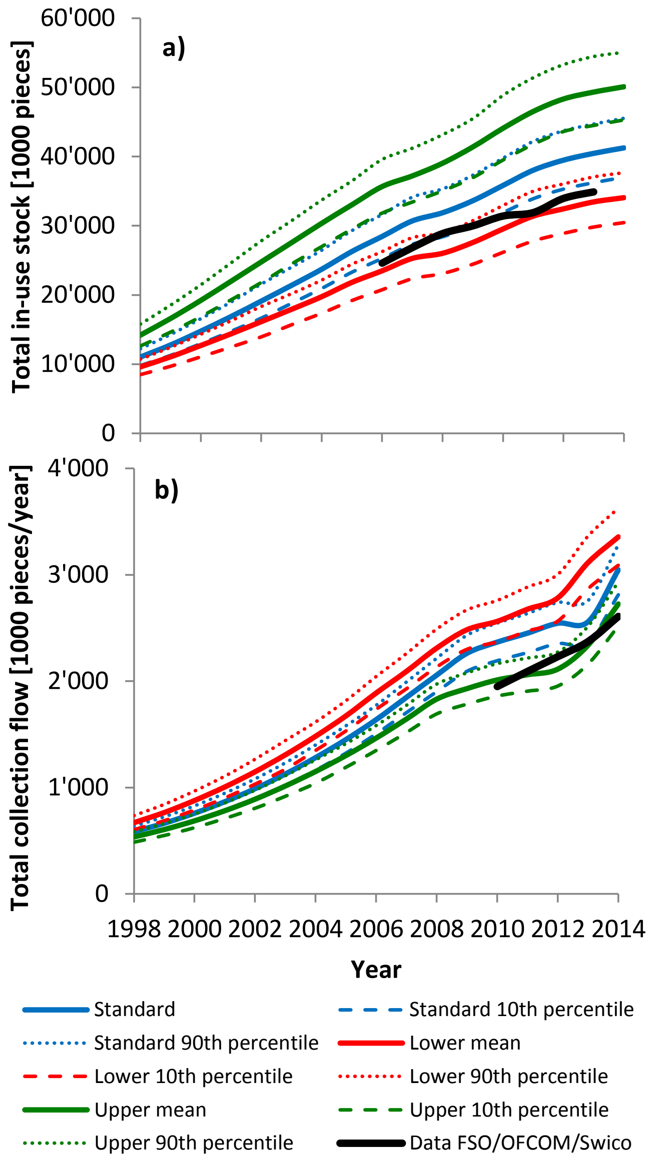

Figure 5.5: (a) Total in-use stock in 1000 pieces and (b) total collection flow in 1000 pieces/year based on the standard, lower and upper lifetime distribution scenarios for desktops, laptops, mobile phones, CRT TVs, FPD TVs, and DVD players (in-use stock) and FPD monitors, desktops, laptops, mobile phones, CRT TVs, and FPD TVs (collection flow), compared to statistical data available from FSO, OFCOM, and Swico Recycling. [48,51,52]

Taking into account the large model uncertainty of up to ±30%, the cascade model is able to map reasonably well the total in-use stock and the total collection flows. This shows that the mixture of detailed bottom-up information on service lifetimes, storage times and transfer coefficients combined with a top-down approach to calculate stocks and flows is an adequate approach to enhance dynamic MFAs. The cascade model provides important details regarding reuse and storage stocks and flows that have been often neglected in dynamic MFA studies. It further unveils different behavior of device types within the use phase, for example with regard to technology changes or phase out periods.

The model structure can be applied to any other product that is potentially reused and stored after its first service life. However, in many cases, data required for a cascade model will be difficult to collect. Furthermore, from a resource point of view, it is not important to know exactly the status of products in the use phase, as long as it can be safely assumed that they will reach the collection system in the end. The use phase could thus be modeled as a 'black box' as in earlier studies, but by adapting the total lifetime distribution function, eventual reuse and storage should be taken into account. Such a simplified model can still be useful to simulate outflows, their composition (mix of devices and materials) and the share reaching collection more precisely.

5.4 Associated Content

The Supporting Information is available free of charge on the ACS Publication website at DOI: 10.1021/acs.est.6b06336.

Detail about the inflow data, Weibull distribution functions, transfer coefficients, and the uncertainty distributions of mass per device type are available. More information on the software tool used to implement the model, run the simulations and visualize the output is available. Additional results on the total lifetime distributions as well as stocks and flows by device type are also provided (PDF)

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Notes

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Chapter 6

Where do our Resources go? Indium, Neodymium, and Gold Flows connected to the Use of Electronic Equipment in Switzerland

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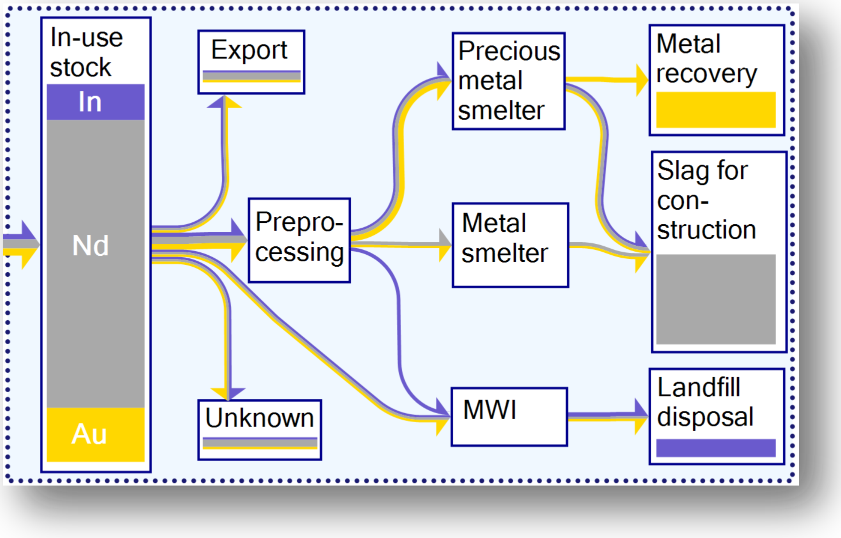

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Abstract

Electronic equipment (EE) contains important material resources, not only bulk material such as iron, aluminum, copper, and plastics, but also precious metals and critical metals. In contrast to bulk materials and precious metals, the recovery of most critical metals has not yet been established on a commercial scale. In this article, we use dynamic material flow analysis to explore the stocks and flows of indium, neodymium, and gold incorporated in EE. Our analysis covers the use, collection, recycling and disposal phases. This allows for the tracking the three metals from their entry into Switzerland as components of new devices until their recovery, disposal in landfill or dissipation to the environment. With statistical entropy analysis, we further analyze the dilution or concentration of the metals during their route through the current system. The largest quantities of all three metals are still found in the EE currently in use. The second largest stocks are disposed slags in landfills for indium, slags used for construction for neodymium, and the output of metal recovery processes for gold. The average metal quantities reaching recycling in 2014 were 90 kg for indium, 2800 kg for neodymium and 330 kg for gold. During preprocessing, not only gold, but also indium and neodymium are successfully concentrated, but subsequently lost in smelting and incineration processes.

Keywords: Indium, neodymium, gold, critical metals, dynamic material flow analysis, statistical entropy analysis

6.1 Introduction

Electronic equipment (EE) contains important material resources, not only bulk material such as iron, aluminum, copper, and plastics, but also precious metals and critical metals.[1] More than 20 years ago, "Swico Recycling", a take-back and recycling scheme for EE has been established in Switzerland. The system ensures the recovery of bulk materials and precious metals, as well as the environmentally sound disposal of pollutants.[2]

Many critical metals, such as indium, gallium, tantalum, or the rare earth elements (REE) are not recycled. Reasons are, for example, low contents of these materials within EE, low market prices that do not cover recycling costs, lack of recycling technologies on commercial scale, metallurgical limits to recovery processes, as well as limited knowledge on overall stocks, flows and disposal pathways of critical metals incorporated in EE.[3–7]

In Thiébaud et al.[8] we presented a dynamic material flow analysis (MFA) of nine different types of EE in Switzerland, identifying past and current in-use and storage stocks of EE as well as the flows between the use, storage, and disposal phases. In the present article, we derive the flows of critical metals incorporated in EE from these results. The detailed model of the use phase is extended with the collection, recycling, and disposal phase. This allows for the tracking of critical metals from their entry into Switzerland (as part of new devices) until their disposal in landfills or dissipation to the environment.

The flows of critical metals incorporated in EE are illustrated using the examples of indium and neodymium. Indium and neodymium were chosen as highly relevant based on a systematic multi-stage selection and evaluation process. Selection criteria took into account criticality, relevance regarding the application in EE, recycling potential and additional prioritization criteria such as ecological relevance, absolute quantities in the e-waste stream and availability of pilot recovery processes.[9] The most important application of indium in electronic devices is the use of indium tin oxide (ITO) as a transparent conducting layer in liquid crystal displays (LCDs).[10] Neodymium is a REE used in small permanent magnets since the 1990s, of which between 8% and 35% are applied in hard disk drives (HDDs), optical drives and loudspeakers in computers, vibration alarms and loudspeakers in mobile phones as well as headphones.[9,11] Neodymium is also found in small quantities in printed wiring boards (PWBs).[12–14] The main share of both metals is thus available in certain locatable components within EE, and recovery from the components in multilevel physical-chemical and metallurgical processes is technically feasible.[9] In addition to neodymium and indium, we include gold. The recycling of gold, mainly found in PWBs, is already well established and is an important economic driver in the recycling system.[15] In our MFA, gold stocks and flows serve as reference values to put the stocks and flows of indium and neodymium into the context of today's recycling system.

Many dynamic MFAs of indium and neodymium have been performed on a global scale. Yoshimura et al. mainly focused on the production process of ITO and LCD modules, with associated losses of indium. The waste management was modeled as a simple process with all indium lost to an unspecified sink.[16] Du and Graedel presented the past and current global demand and in-use stock of REE in general and of neodymium-iron-boron (NdFeB) permanent magnets in particular.[17,18] Alonso et al. modeled the future demand for REE and Halada et al. forecasted the consumption of gold, among other metals.[19,20] The latter four studies did not include waste management processes. When China – with a global market share of over 80% in the REE production – had implemented export quotas in 2010[11], research on the recycling potential of permanent magnets has increased, including dynamic MFAs of the future recycling potential for REE in general and of NdFeB magnets in HDDs, in particular.[11,21] Static MFAs include, for example, a study by Licht et al., who presented global indium flows with their dissipative losses. However, waste management processes of indium in non-dissipative consumer goods were not considered.[10] An MFA of REE in Europe estimated flows of neodymium, among other metals, into use, in-use stocks and waste streams.[22] An MFA of permanent magnets in Denmark explored the current and future potential of a secondary supply of neodymium and dysprosium from the recycling of NdFeB magnets,[23] and was complemented by a detailed MFA of the fate of REE in HDDs in a Danish recycling plant.[24] In a similar approach, Chancerel et al. presented flows and losses of precious metals during preprocessing of waste electrical and electronic equipment (WEEE).[15] Ueberschaar et al. extended their approach and analyzed recycling efficiencies of bulk metals, precious metals and critical metals in WEEE preprocessing.[25]

Ueberschaar et al. and Zhang et al. reviewed various methods available to recover indium from LCDs.[26,27] Binnemans et al. discussed different routes for REE separation from non-REE elements in recycled fractions containing NdFeB magnets.[28] In a recent study, Maât et al. presented a new process to recover REE from NdFeB magnets using water and sodium chloride.[29] Thus, various recycling technologies that are available for indium and neodymium, exist on a laboratory and pilot scale. But although indium in LCDs is viewed as a stock available for future recycling[10] and the recycling of HDDs is considered the currently most feasible way towards large-scale recycling of neodymium,[11] there are presently no commercial-scale recycling options available for indium and neodymium from EE.

In this article, we combine a dynamic MFA of the use phase with an MFA of the collection, recycling and disposal phase of indium, neodymium and gold in EE in Switzerland. With our dynamic MFA, we are able to track and quantify the pathways and losses of these three metals in the collection, recycling, and disposal phase over time, starting when the considered device types have been put on the market. We identify the most important sinks and compare them to current in-use and storage stocks in order to estimate the recycling potential for the different metals. With statistical entropy analysis (SEA), as proposed by Rechberger and Graedel[30], we further illustrate the dilution or concentration of each metal during its route through the current system. On this basis, we can detect the processes which are responsible for losses. As desktop and laptop computers have undergone a technology change from NdFeB magnets-containing HDDs to neodymium-free solid state drives (SSD), we include an analysis of the future recycling potential of NdFeB magnets, estimating the time NdFeB magnets from HDDs will still be found in the waste stream.

To implement the model, we used our own open source software tool in Python 3 applying a probabilistic approach, as described in Thiébaud et al.[8] This allows to systematically take into account data uncertainty in our model.

6.2 Materials and Method

This chapter is structured based on the standardized description ODD (overview, design concepts, details) protocol that was originally developed for the documentation of individual-based and agent-based models,[31,32] but has proven to be useful for structuring MFA models as well.[33]

6.2.1 Overview

The purpose of the dynamic MFA model is to quantify the stocks and flows of indium, neodymium, and gold in the use phase and in waste management processes in Switzerland, incorporated in nine EE device types that together cover around 90% of the stocks of these three metals in Swiss private households[9]. Device types included are: conventional mobile phones (referred to as “mobile phones” in this article), smartphones, desktop and laptop computers, monitors, cathode ray tube televisions (CRT TVs), flat panel display televisions (FPD TVs), digital video disk (DVD) players and headphones (for details, see Thiébaud et al.[8]). The dynamic MFA should provide insights regarding the past, current and future losses and sinks of critical metals and precious metals in the collection, recycling, and disposal phase.

The system under study includes the use phase as well as the collection, recycling, and disposal phase. For the use phase, we adopted the cascade model presented in Thiébaud et al.[8] The collection, recycling and disposal phase is divided into maximum 16 processes, depending on the considered metal, including all steps from collection to material recovery or slag disposal. In addition to the in-use and storage stocks, seven processes can potentially build up a stock. The system has one inflow, corresponding to the flow of metals incorporated in sold devices. Instead of outflows, we include various sinks within the system to show the final destination of critical metals and precious metals. The inner system boundary corresponds to the Swiss border, with exports to downstream processes in other countries (Figure 6.1). The temporal extent of the model depends on the device type and starts from the time when a device type was first put on the market up to the year 2014. In addition, for devices containing neodymium magnets, we included a prospective analysis up to the year 2050. The temporal scale is one year.

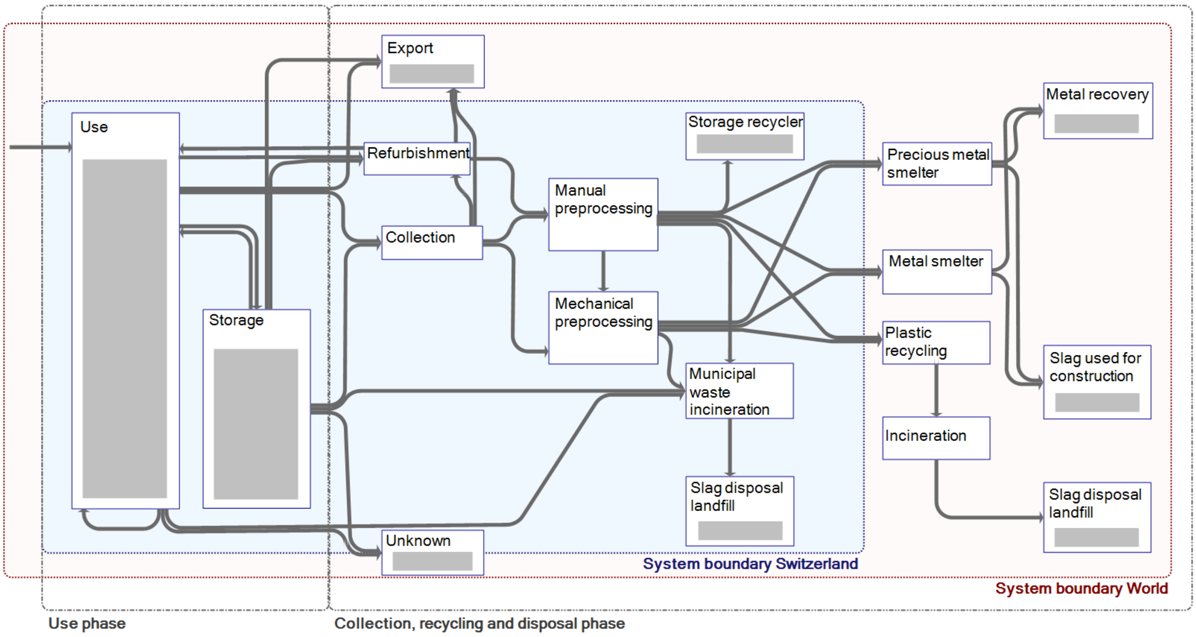

Figure 6.1: System overview

6.2.2 Design Concepts

6.2.2.1 Basic Principles and Modeling Approach

The model employs a retrospective and prospective top-down approach, deriving the stock S[n] at a time n from the net flow by using the balance of masses (equation (1)), with the constant sampling rate T = 1 year.[33]

$$S[n] = (inflow[n] - outflow[n]) \cdot T + S[n-1] \qquad (1)$$

The outflows of the in-use stock and the storage stock are calculated based on different lifetime distribution functions f[m] for the service lifetimes and storage times of new and used devices according to equation (2).[8,33]

$$outflow[n] = \sum_{m=0}^{\infty} inflow[n-m] \cdot f[m] \qquad (2)$$

To compute the resulting indium, neodymium and gold stocks and flows, the inflows of devices, taken from Thiébaud et al.[8] are multiplied with the respective indium, neodymium and gold content per device.

Desktop and laptop computers have undergone a technology change from neodymium-containing HDDs to neodymium-free solid-state drives (SSDs). This leads to decreasing neodymium inflows, which are computed by assuming a simple logistic diffusion model for the SSD technology and adding up a model for computers with HDDs and one for computers with SSDs. Long total lifetimes due to more storage and reuse will, however, delay the decrease of the amount of neodymium in the outflows of the use phase. In order to estimate the future quantities of neodymium magnets in the recycling system, we extrapolate the inflow, that is, sales data of desktop and laptop computers, headphones as well as mobile phones and smartphones with logistic functions up to the year 2050, based on current sales and stock data.

6.2.2.2 Dissipation

Dissipative flows of indium, neodymium, and gold are considered in the collection, recycling and disposal phase as specific flows to landfills or the environment, depending on process inflows and transfer coefficients.

6.2.2.3 Uncertainty

To account for data uncertainty, metal contents, as well as transfer coefficients, are modeled as probability distributions. Based on the data characteristics, we choose either normal or triangular distributions. The dependent variables are calculated by means of Monte Carlo simulation and are therefore again provided as probability distributions.[34,35]

6.2.3 Details

6.2.3.1 Initial Condition

The sales data of EE, on which the inflows of metals are based, go back to the year when a device type was first brought onto the market.[8] Thus, the simulation starts from a stock = 0.

6.2.3.2 Model Input Data

The indium, neodymium and gold content per device type is taken from own measurements[9,12] and literature.[1,13–15,18,23,26,36–42] As the data quality of these sources varies highly, we introduce data quality indicators similar to the pedigree matrix of Weidema and Wesnæs,[43] regarding the sample size, measurement or modeling approach and the analysis method. For metal contents with various observations available, the sum of the three indicators for each observation is translated to a weighting factor, and a weighted mean is calculated. We then introduce a triangular distribution for the metal content of each device type, including the weighted mean and the lowest and highest value. The lowest and highest values were extended by taking into account the standard deviation (SD) of the SD, as referred to in JCGM,[44] so that the lower and upper limit of the triangular distribution should lie within the 95% confidence interval of the metal content. As already stated above, the decreasing neodymium content in desktop and laptop computers is computed by adding up a model for computers with HDDs and one for computers with SSD. In addition to neo-dymium in magnets, neodymium is also found in PWBs, although its exact source is unknown.[12–14] According to Bangs et al.[42], the gold content in information and communi-cation technology hardware has been declining in the past 12 years by about 40%. This temporal change is accounted for in the triangular distributions of the gold content in desktop and laptop computers as well as in mobile and smartphones. For other device types and metals, no information on the temporal change of metal content was available.

Inflow data of EE, lifetime distribution functions for the service lifetime and the storage time, as well as the transfer coefficients for the flows within and out of the cascade model, are adopted from Thiébaud et al.[8,45]

For the prospective MFA of neodymium in magnets, the sales data of desktop computers, headphones, mobile phones, smartphones as well as DVD players are extrapolated with logistic functions up to the year 2050, based on current sales and stock data. For laptop computers, we assume future sales in a steady state at the level of 2014. For all sales flows, we assume an increasing uncertainty from 2014 up to 2050, modeled as normal distributions. All lifetime distribution functions and transfer coefficients are assumed to remain constant at the level of 2014.

In order to model the collection, recycling, and disposal phase, we needed data regarding transfer coefficients to refurbishment, preprocessing, downstream processes and disposal. CRT TVs, desktop computers, headphones and mobile phones already had significant outflows before the Swiss recycling system for EE had been established in 1994. We assume that before 1994, indium, neodymium, and gold incorporated in EE ended up in Swiss landfills, either directly or via municipal solid waste incineration. Data on transfer coefficients of mobile phone and smartphone flows are collected via interviews with two major Swiss telecommunication companies and a large Swiss EE retailer.[46–48] We further held interviews with four major Swiss e-waste recyclers, who together treat over 85% of the Swiss e-waste, to gather data on transfer coefficients to manual and mechanical preprocessing as well as downstream processes according to Figure 6.1.[49–52] Only in one recycling plant, measurements of indium, neodymium, and precious metals in output fractions have been possible.[12] For the other recyclers, data on the pathways of indium, neodymium and gold in their recycling process are partly based on estimations. The uncertainties of transfer coefficients in the collection, recycling, and disposal phase are modeled as triangular distributions, based on the information provided by the interviewees.

More information on the data quality indicators and the temporal change of metal contents, the extrapolation of sales data up to the year 2050, details on the flows of EE in the Swiss recycling system and the corresponding transfer coefficients as well as all related uncertainties is provided in the supporting information (SI) of this article.

6.2.3.3 Model Output Data and Evaluation

Model output data comprise stochastic time series of stocks and flows depicted in Figure 6.1, depending on the considered metal. The probability distributions of the time series are visualized by the 10th percentile, mean and 90th percentile. To further illustrate the dilution or concentration of metals during their route through the system and its change over time, we evaluated the model output data with SEA and calculated the relative statistical entropy (RSE) for each metal over all stages and years.[30] The RSE corresponds to the ratio between the statistical entropy H of each stage and the maximum statistical entropy H_{max}, calculated from the earth crust content of each respective metal. As this method requires information on material flows, in particular, metal contents for each flow shown in Figure 6.1, we extended the model described in Thiébaud et al.[8] with a simple MFA of the collection, recycling, and disposal phase for each device type, based on batch tests run by Swico Recycling.[53] More information on the SEA can be found in the SI.

6.2.3.4 Detailed Model Description

The conceptual model, as described in Figure 6.1, was implemented using our own MFA simulation tool called 'pymfa'. The tool is written in Python 3, using the numpy, scipy, and matplotlib libraries.[8]

As the SEA is connected to the MFA system, we extended the pymfa tool with a functionality to calculate the RSE over all system stages and years.

The model was simulated for each device type and metal, producing results in kg metal per product up to the year 2014. For device types containing neodymium, we simulated future flows up to the year 2050. The SEA was calculated for each metal from 1990 to 2014. Each simulation run was repeated 10 000 times, which we considered a sufficient number for the Monte Carlo simulation.

6.3 Results and Discussion

6.3.1 Development of Stocks

The nine device types represent a total indium stock of 1.7 tonnes ±19%, a neodymium stock of 39 tonnes ± 13% and a gold stock of 4.8 tonnes ± 20% in the use phase in Switzerland in 2014. The high uncertainty for the indium stock results from high variability in indium quantity per device due to variable display sizes and measured indium contents. The uncertainty for gold stocks origins from diverging information on gold content and few measured devices per data source. Figure 6.2 shows how the total stocks of indium, neodymium, and gold developed over time and how they are distributed among different device types. Indium is primarily stocked in FPD TVs. Neodymium is mainly stocked in desktops and laptops. With the exception of headphones, all device types include PWBs with some gold content. Hence, the gold stock is the most evenly distributed stock among device types, The growth rate of the indium stock is slowly decreasing, highly depending on FPD TVs sales, and amounts to 6% in 2014. The neodymium stock is growing at a 1% rate in 2014 and is expected to decrease in the near future due to the technology change from HDDs to SDDs. The growth rate of the gold stock accounts for only 2% in 2014 and will further decline due to decreasing gold contents in EE put on the market and the market saturation of most device types.

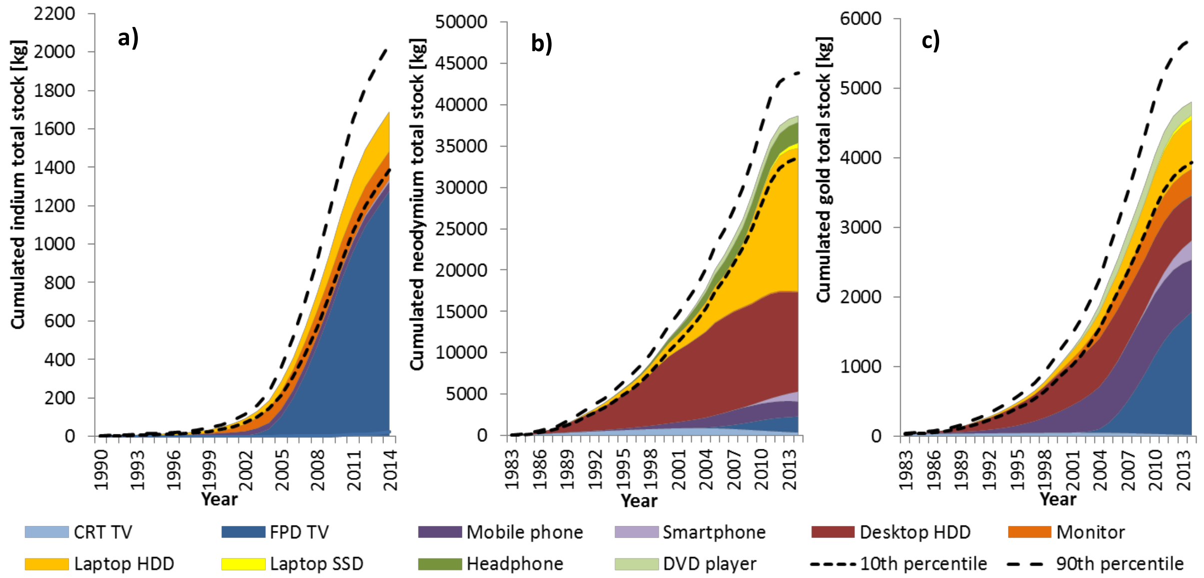

Figure 6.2: Cumulated total stock of a) indium, b) neodymium and c) gold in kg in the use phase. Desktops with SSDs are not included in the figure, as quantities are still too low in 2014.

6.3.2 Stocks and Flows in 2014

The simulated stocks and flows to and within the current system connected to the use of electronic equipment in Switzerland are depicted in Figure 6.3 for indium, neodymium, and gold in 2014. For the sake of clarity, we refrain from including uncertainty ranges for flows in the figure.

236 kg ±50% of indium is incorporated in EE sold in Switzerland in 2014. 135 kg ±20% leave the use phase through disposed of EE, whereof 45 kg ±20%, or 30%, do not reach recycling processes due to export, incineration of indium-containing devices or unknown disposal pathways. Currently, all indium-containing devices reaching recycling processes in Switzerland go through manual preprocessing, either due to possible mercury-containing backlights in FPDs or lithium-ion batteries in phones and laptops, which require depollution before further treatment.[54,55] 90% of the indium in dismantled LCD panels is sent to municipal waste incineration (MWI), where it ends up in the slag. 5% is stored for possible further treatment in the future and another 5% (in FPDs of mobile phones and smartphones) reach precious metal smelters (PMS), where indium ends up in the slag as well.

The inflow of neodymium in EE in 2014 amounts to 3900 kg ±35%. With 2800 kg ±14%, 80% of the outflows from the use phase reach the recycling processes. 70% of the collected neodymium reaches manual preprocessing. This is due to the need to depollute laptops and phones, as described above, or manually remove valuable components from desktops. HDDs and optical drives from laptops and desktops containing neodymium magnets are still sent to mechanical preprocessing after manual dismantling, while most neodymium in manually dismantled PWBs as well as in phones is directly sent to PMS. The pathways of neodymium from mechanical preprocessing to PMS and metal smelters are highly uncertain. According to the data available from a Swiss recycler, most neodymium reaches the finest fraction that is sent to PMS.[12] Estimations of other recyclers and literature data suggest, however, that neodymium is mostly transferred to the magnetic steel fraction[24,25,51]. Either way, neodymium in magnets and PWBs reaches a smelting process, where it ends up in the slag. With the exception of flows from manual preprocessing to PMS, neodymium in PWB accounts for less than 10% of the total neodymium flow. Considering that the source of neodymium in PWBs is unknown and neodymium recovery in PMS is currently not feasible, we recommend to further focus on neodymium in magnets.

520 kg ±50% of gold is found in EE sold in Switzerland in 2014. The outflow of the use phase amounts to 440 kg ±20%, whereof 25% is not reaching the recycling scheme due to export, incineration and unknown disposal pathways. 84% of the collected gold in EE is sent to manual preprocessing, either due to regulatory requirements as described above, or to remove high-value PWBs. Manually dismantled PWBs are directly sent to PMS without further treatment or losses. Only a small amount of PWBs from laptops (after displays have been removed) is further sent to mechanical preprocessing. From mechanical preprocessing, around 5% of the gold is lost in fractions going to MWI, metal smelters and plastic recycling. This result is in strong contrast to Chancerel et al., who found that only 25% of gold reaches fractions from which it may potentially be recovered.[15] Available data confirm significant amounts of precious metals reaching finely graded plastic fractions.[12] Therefore, some recyclers send finely graded plastic fractions also to PMS. Large amounts of precious metals in ferrous metal and aluminum fractions were not found.[12] Gold reaching PMS is recovered

with a rate of at least 95%. Overall, 70% of the gold leaving the use phase is recovered, with highest losses occurring directly after the use phase.

The total amount of indium, neodymium, and gold that entered Switzerland within the nine considered device types until 2014 is distributed among the respective stocks as shown in Figure 6.3. All three metals are mainly found in the use phase (indium: 60-90%, neodymium: 50-60%, gold: 50-70%, including uncertainty ranges). The second largest stocks are, for indium, disposed of slags in Swiss landfills (15-20%), for neodymium, slags used for construction (30-40%) and for gold, the recovered metal (30-35%). These three stocks represent the main fate of the considered metals. Although before Swico Recycling was established in 1994, all metals incorporated in EE are assumed to reach Swiss landfills, the share of neodymium (3%) and gold (2%) in Swiss landfills out of the total amount of these metals within the system is small. The large stocks in the use phase for all three metals indicate that most of the resources that have entered the system could theoretically still be recovered. The recycling potential is initially lowered by losses mostly due to unknown disposal pathways. They account for 10% to 24% of the use phase's outflows and lead to a reduction between 3% and 7% of the total amount of metals in the system. Although it is possible that material reaching unknown disposal pathways may still end up in a collection system,[8] it is more likely that a significant amount of resources is lost at the end of the use phase. It would be important for future research to break down and analyze disposal pathways in more detail in order to identify device types and incorporated materials that need special attention to improve collection rates.

The average metal quantities reaching recycling in 2014 are, with 90 kg for indium, 2800 kg for neodymium and 330 kg for gold, very low and correspond to only 40%, 70% and 60% of the indium, neodymium and gold inflow, respectively. Based on current metal prices, the value of the metal flows to recycling account for 36 000 US$ for indium, 200 000 US$ for neodymium and 13 600 000 US$ for gold.[56–58] Thus, although neodymium flows to recycling are nine times larger than gold flows, the revenues for gold are almost 70 times higher. The small quantities combined with low prices discourage Swiss recyclers to adapt their preprocessing and find appropriate downstream processes to recover indium and neodymium. These conditions also provide little incentive for downstream processors to establish commercial-scale recovery processes for indium and neodymium. With the exception of a few recyclers who store LCD modules, all indium and neodymium are currently irrecoverably dissipated to the slag of MWIs, PMS or metal smelters.

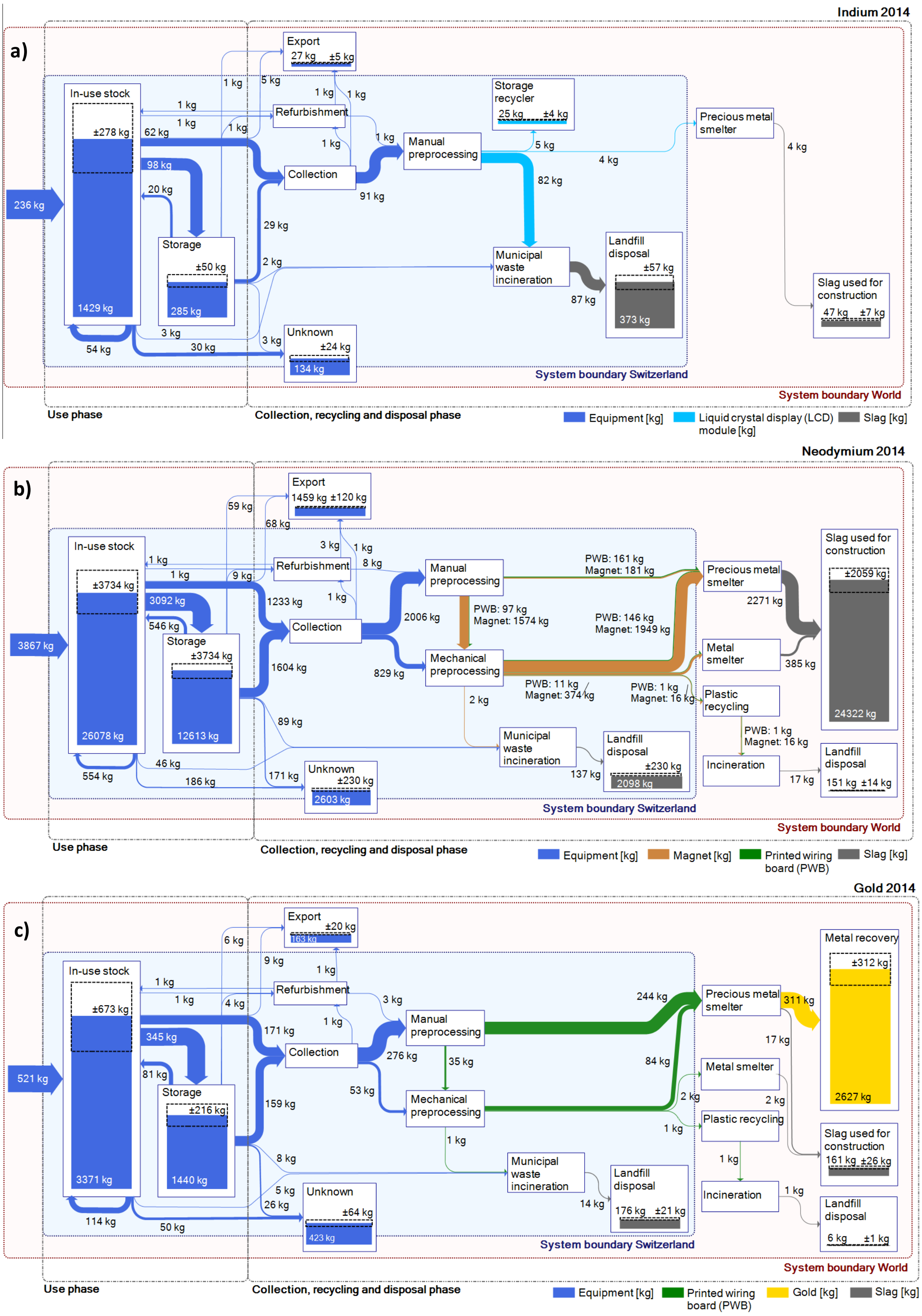

Figure 6.3: Indium, neodymium and gold flows in kg in 2014 connected to the use of electronic equipment in Switzerland.

6.3.3 Statistical Entropy Analysis

To measure the dilution of metals during their route through the system, we conducted a SEA and calculated the RSE for each metal over all system stages and years. The trends of the RSE for the total indium, neodymium and gold stocks and flows along the use, collection, recycling, and disposal phases are illustrated in Figure 6.4 for the years 1990, 2000, 2010 and 2014. The system under study (Figure 6.1) transferred to the respective stages is depicted in Figure D.8 in the SI.

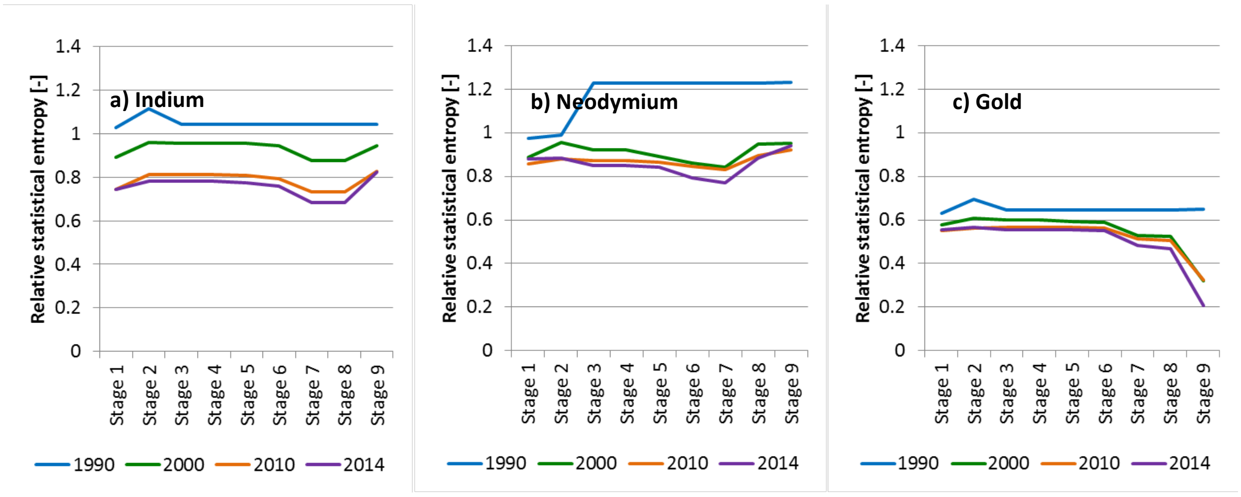

Figure 6.4: Relative statistical entropy for a) indium, b) neodymium and c) gold for the years 1990, 2000, 2010 and 2014.

The RSE values of 1990 correspond to the status of the system before an e-waste collection system was established. For the years 2000 to 2014, the differences in the RSE are due to changed composition of EE that is put on the market. Due to low contents of indium, neodymium and gold within EE, these metals are already highly diluted when the devices are put on the market (stage 1) and get even further diluted when they are in the stock (stage 2). Compared to indium and neodymium, gold is the least diluted metal in EE, with reference to the earth crust content. Dilution during stages 1 and 2 changes over time depending on the share of devices with higher metal contents. In 1990, EE is sent to MWIs and metals are dissipated to the slag which was disposed of in landfills. This results in low metal contents (for indium and neodymium below the earth crust content) and high RSE. The decreasing RSE within stages 3-7 for the years 2000 – 2014 shows the ability to concentrate the metals during the collection and recycling phase. Indium is concentrated to the level of dismantled LCD panels, neodymium to dismantled HDDs or optical drives and gold to dismantled PWBs or precious metal-containing fractions after mechanical preprocessing. However, the decrease in RSE is reversed for indium after stage 8, where LCD panels are incinerated and indium is dissipated to the slag. Within Switzerland, slags from MWIs are disposed of in specified landfill sites. According to data from a Swiss MWI, the content of indium in the slag is similar to very low-grade zinc deposits.[59,60] These data, however, are very uncertain and it is not known whether the recovery of indium from slags in landfills would be economically viable. For neodymium, after separation, HDDs and optical drives are sent to mechanical treatment with other EE (stage 7) with the resulting fractions treated in a smelter where neodymium is dissipated to the slag (stage 8/9). Slags from PMS and metal smelters are often used for construction, either within landfills or, for example, for the reinforcement of dams. Neodymium within smelter slag is thus further dissipated to the environment in contents well below primary mines (see SI for more details on slag contents). As gold is recovered, the RSE shows a sharp drop after stage 8. Due to losses in the system, the RSE stays at 0.2.

The SEA illustrates how the current collection and recycling system successfully concentrates indium and neodymium after the use phase, mostly because of manual dismantling that is either required due to depollution targets or done voluntarily due to valuable fractions in EE. The content of indium in LCD modules amounts to 130 to 230 ppm, which lies above contents in primary mines of 1 to 100 ppm.[9,26,60] A life cycle assessment of primary and secondary production of indium indicated that secondary production after manual preprocessing is favorable or at least equal to primary production.[9] The content of neodymium in dismantled HDDs and optical drives sent from manual to mechanical treatment (250 - 560 ppm) lies below contents in primary mines (1200 -17,600 ppm). This fraction should, therefore, be further physically separated and concentrated before it is sent to metallurgical extraction.[5] The secondary production of neodymium oxide from HDD is clearly preferable to primary production from an environmental point of view.[9] Thus, with small adaptations of today's recycling system, the recovery of indium and neodymium from the concentrated fractions should be possible, if commercial-scale separation and recovery processes for indium and neodymium were available.

6.3.4 Future Developments

Absolute quantities of indium from EE in the recycling process are low, and, according to Yoshimura et al., significantly lower than losses in the mining, smelting and refining processes.[16] Significant increases in indium quantities are not expected in the near future. Recycling of neodymium magnets, particularly from HDDs, seems more promising due to higher quantities.[11] However, the technology change from HDDs to SSDs has started in Switzerland already around 2010. The first decline of neodymium in magnets in the flow to recycling resulting from this change is expected in 2018 (Figure 6.5). In 2030, the flow will be more than halved from 2900 kg/year to 1100 kg/year. In 2050, unless some other technology changes will alter the use of neodymium magnets in EE, the flow to recycling will stabilize at 600 kg or around 20% of the flows in 2018, consisting mainly of neodymium magnets in speakers from smartphones, laptops, and headphones. Despite decreasing neodymium flows, most of the existing neodymium stock could be "mined" within the next 30 years if suitable recovery processes were available. Furthermore, neodymium magnets from speakers will persist most likely in future recycling flows.

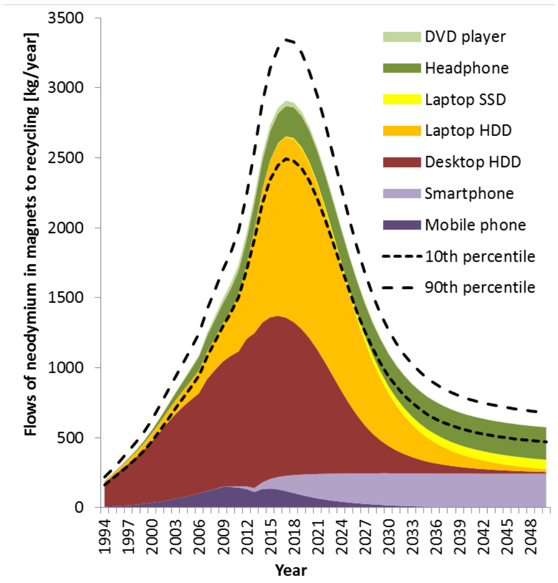

Figure 6.5: Flows of neodymium in magnets to recycling in kg/year from 1994 to 2050.

As quantities of indium and neodymium from EE will remain low or decrease in the future, and their prices currently do not match recycling costs, their recovery is not a priority among recyclers and downstream processors. However, an economic feasibility study investigating the Swiss recycling system has shown that indium recycling could be covered financially with a marginal increase of the advance recycling fee by maximum 0.2 US$/product.[9] The results indicate that small additional fees levied by recycling systems could provide sufficient funds to encourage recyclers and downstream processors to adapt and establish the necessary infrastructure for the recovery of indium and neodymium. Stricter regulations regarding metal-specific recycling rates could further encourage systems, recyclers and downstream processors to request and establish commercial-scale recovery options and close the material cycles.

The results for indium and neodymium cannot be generally transferred to other critical metals. Provided that a material, however, is locatable and available in small quantities in known components within EE, the situation is comparable.

6.4 Associated Content

The Supporting Information provides more detail regarding data quality indicators and the temporal change of metal contents, the extrapolation of sales data up to the year 2050, the flows of EE in the Swiss collection, recycling, and disposal system and the corresponding transfer coefficients as well as all related uncertainties. It further includes more information on the SEA and the extended software tool used to implement the SEA. Additional results include a table on the shares of stocks, losses, and sinks in the total amounts of indium, neodymium and gold in the current system. This material is available free of charge at http://pubs.acs.org.

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Notes

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Chapter 7

Final Discussion, Conclusion and Outlook

7.1 Final Discussion

The aim of this thesis was to provide insights regarding the guiding question:

What are the stocks, pathways, contents, and properties of critical metals in emerging end-user technologies (in particular, electronic equipment) within the anthroposphere and how can they be influenced in view of effective and efficient recycling systems?

In order to reach this aim, a twofold approach was chosen as illustrated in Figure 1.2: the empirical thread and the methodological thread. Both threads started with a literature review, of metals' criticality and composition of EE as well as of existing dynamic MFA methods (see Chapter 1, 2 and 3). The following three findings of the literature review could be derived as the most important research gaps for this thesis:

- As 'bottom-up' studies require extensive data collection, there is a lack of studies applying this approach and provide insights on **stocks, lifetimes and disposal flows** of metals. Especially for critical metals, consistent and reliable information is missing.
- Recovery rates for many critical metals are below 1% and they are either dissipated to recovered base metals or slags that are disposed of in landfills or used for construction. **Dissipation** of metals to the environment has only in recent years been addressed from a resource point of view and indicators as well as data are still scarce.
- Existing data on critical metals often do not offer information on **data uncertainty**. Without the analysis and communication of uncertainties, however, the effect of uncertain data input is difficult to understand.

7.1.1 Stocks, Lifetimes and Disposal Flows

Concerning the first of the above-mentioned research gaps, we conducted an extensive data collection regarding service lifetime, storage time and disposal pathways of new and secondhand EE through an online survey and structured interviews (see Chapter 4). The advantages of online surveys are fast data access and low resource requirements. A drawback is, however, that the collected data might be biased due to undercoverage and self-selection, among other factors.[1] A truly random sample of the population, in our case users of EE in Switzerland, would have been preferable but would have required resources that were not available. Besides biased data, that have been accounted for by additional structured interviews and weighting adjustments, data is also uncertain as users had to remember in which year they bought, put in storage or disposed of their current and past devices, which is often difficult. Nevertheless, surveys are an important method for 'bottom-up' data collection that give often more insights on consumer behavior than, for example, analyses of EE at collection points or recycling facilities.[2]

The results of the survey – histograms for the different lifetimes and transfer coefficients for the disposal pathways – provided in-depth 'bottom-up' information that was used as a basis for the development of the use phase's model. The results indicated that considerable quantities of devices are reused and stored, both after the first and second use. They showed positively skewed histograms for service lifetimes with wide distributions and substantial differences among device types. The assumption that the service lifetime decreases over time[3–9] could only be confirmed for monitors, loudspeakers, DVD players and CRT TVs. For conventional mobile phones and smartphones, desktops, laptops and FPD TVs, no decreasing trend was determined. It could, therefore, be justified to assume constant lifetimes for certain device types in a dynamic MFA model. The storage time histograms showed a more similar storage behavior for most device types.

The model of the use phase was developed as a cascade model (Figure 5.1), taking into account the above-mentioned findings regarding reuse, storage and disposal pathways (see Chapter 5). The model thus combined detailed 'bottom-up' information with a 'top-down' approach by calculating stocks and flows from inflows and lifetime distributions. The survey data allowed to fit Weibull lifetime distribution functions for the service lifetime and storage time of new, second-hand and further used devices for each device type. If applicable, also the distributions' changes over time were considered. The cascade model sheds light on the 'black box' use phase and breaks down the various stages a device passes during its stay.

Such a detailed model can be used for various purposes: from the inflow over time split into various device types, we calculated the development of the different stocks and flows by device types, and illustrated technology successions, for example from mobile phones to smartphones or CRT TVs to FPD TVs (Figure 5.3). Considering stocks and flows of a specific year, we highlighted the different internal and disposal flows and the resulting discrepancies of the distribution of the total stock to the different in-use stocks and storage stocks among device types (Figure 5.4). With an impulse response analysis, we simulated the total lifetime, that is, the time devices remain in the use phase from sales to disposal. We could show, for example, that the total lifetime distribution functions, compared to the service lifetime distribution functions of new devices, have a mode shifted towards longer lifetimes, and a wider and more positively skewed distribution (Figure 5.2). The median total lifetime increases, for example, from 3 to 7 years for mobile phones and smartphones and from 5 to 9 years for laptops. The cascade model could also serve as a basis for scenario analysis if inflows, lifetime distribution parameters or transfer coefficients are varied and resulting stocks and flows are calculated.

For calculating stocks and flows of indium, neodymium and gold incorporated in EE, the model of the use phase was extended with the collection, recycling, and disposal phase (see Chapter 6 and Figure 6.1). Instead of outflows, we included various sinks within the system to show the final destination of metals. This allowed to track the three metals from their entry into Switzerland as components of new devices until their recovery, disposal in landfill or dissipation to the environment and quantifying losses after the use phase. By comparing the sinks to current in-use and storage stocks, the recycling potential for the different metals was assessed.

The data collection for the collection, recycling, and disposal phase was based on interviews with two major Swiss telecommunication companies, a large Swiss EE retailer and 4 major Swiss e-waste recyclers. Collected quantities of specific device types are often not measured, but based on estimations. On the level of device input as well as base metals, plastics, and precious metals in the resulting output fractions, recyclers know their processes very well. However, they often do not know in which fractions they transfer critical metals, and data on the pathways of indium, neodymium and gold in their recycling process are partly based on estimations. Only a few studies have measured the content of critical metals in fractions after preprocessing.[10–12] Due to large discrepancies among recycling processes, for example, the share of manual dismantling versus mechanical preprocessing, the results of one preprocessing facility may not be representative for another one.

Both the cascade model and the extended model apply an inflow driven 'top-down' approach by calculating stocks and flows from inflows and lifetime distributions. As extrapolations of inflow data are prone to high uncertainty due to their fluctuations and dependence on economic and technological developments, many studies suggest that stocks would provide a more robust basis for forecasts.[13] As our simulation tool is currently not able to compute inflows and outflows from stock data, we applied an inflow driven approach also for the forecast of neodymium flows up to the year 2050. The extrapolated inflows, modeled with logistic functions, combined with time-invariant transfer coefficients and lifetime distributions, represent a strong simplification, especially regarding possible technology changes in the EE market. A more elaborate scenario analysis, including a stock-driven approach, would be useful to put future neodymium flows into a more solid context.

7.1.2 Dissipation

The second research gap addresses the quantification of dissipation of critical metals to the environment. We used two complementary approaches to meet this challenge (Chapter 6). First, in our dynamic MFA, we included dissipative flows of indium, neodymium, and gold in the collection, recycling and disposal phase as specific flows to landfills or the environment, depending on process inflows and transfer coefficients. As our system has no outflows, the sinks accumulated from these dissipative flows can be quantified. With literature data, if available, metal contents in slags can be compared to contents in primary mines or the average content in the earth crust. On this basis, it can be analyzed whether metals are irrecoverably dissipated or rather diluted.

As a complementary approach, we evaluated our MFA system with SEA to measure the ability of our system to dilute or concentrate a substance and illustrate which processes are responsible for it. The RSE, a ratio between the statistical entropy H of each stage and the maximum statistical entropy H_{max}, calculated from the earth crust content, can be used as a valuable indicator to determine the level of dilution or dissipation of a metal. Furthermore, the RSE visualizes in a simple way the dilution or concentration of a substance in a system and shows, at which stages processes should be adapted for a better substance recovery.

7.1.3 Data Uncertainty

The third research gap relates to the issue of data uncertainty. Data for MFAs often origin from many different data sources, including, for example, measured data, data from surveys or estimations by experts. As existing data of critical metals is already scarce, it is not always possible to collect various data points for a specific variable or parameter. Therefore, data uncertainty can often not be derived directly from available data.

We accounted for data uncertainty by modeling inflows, transfer coefficients, mass of devices and metal content per device type as probability distributions. We chose either normal or triangular distributions, based on the characteristics of the available data. As data for the metal content per devices type is taken from many different sources with variable data quality, we additionally introduced data quality indicators similar to the pedigree matrix of Weidema and Wesnæs,[14] regarding the sample size, measurement or modeling approach and the analysis method. As for most input parameters only single data points were available, the probability distributions were estimated based on expert opinion. Only for the mass per device type and metal contents, enough data was available to determine probability distributions based on statistical analyses. The dependent variables were calculated by Monte Carlo simulation and again provided as probability distributions. The uncertainty ranges of the dependent stocks and flows are represented by the 10^{th} and 90^{th} percentiles.

It would have required too much of an effort to treat the uncertainty of the lifetime distribution parameters in the same way because our tool did not provide this functionality. In many MFA studies, it has, however, been discussed that stocks and flows are sensitive to the chosen lifetime distribution functions and their parameters.[15–17] Therefore, their uncertainty was taken into account by conducting a sensitivity analysis with a lower and upper lifetime distribution scenario on the level of electronic equipment.

Within the cascade model, the 10th and 90th percentiles, resulting from the Monte Carlo simulation, account for deviations of ±10% from the mean total stock in 2014 in terms of devices. As the mass of each device type increases uncertainty, the uncertainty ranges between ±12% of the mean total stock in tonnes. For flows of devices in pieces or tonnes, the resulting uncertainty is of a similar order of magnitude. The 10th and 90th percentiles account for deviations from the mean total stock of ±19% for indium, ±13% for neodymium and ±20% for gold in 2014. The high uncertainty for the indium stock results from high variability in indium quantity per device due to variable display sizes and measured indium contents. The uncertainty for gold stocks origins from diverging information on gold content and very few measured devices per data source. Uncertainty regarding collection flow ranges between ±14% for neodymium and ±20% for indium and gold.

Considering the 10th and 90th percentiles of the lower and upper scenario of the sensitivity analysis, the maximum deviation from the mean stock of the standard scenario amounts to approximately ±30% (Figure 5.5). The sensitivity of the flows is presented using the collection flows as an example. As longer service lifetimes and storage times lead to smaller collection flows, the lower scenario accounts for the highest flows and vice versa. The maximum deviations between the mean of the standard scenario and the 10th and 90th percentiles of the lower and upper scenario account for ±20%. As the extended model on the level of indium, neodymium and gold adopts the use phase model for electronic equipment, no additional sensitivity analysis for different lifetime distributions was performed. If the uncertainty of the lifetime distributions were also accounted for, the uncertainty of collection flows could, therefore, increase to around ±30% for neodymium and ±45% for indium and gold.

Our approach to model input data as probability distributions does not account for model uncertainty. Models are always incomplete representations of real systems with some less important variables, processes, and interactions left out, either due to simplification purposes or lack of knowledge. In comparison to the uncertainty of input data, model uncertainty is even more difficult to quantify.[18] In order to evaluate to what extent our model is able to reproduce the behavior of the real system, we compared the total in-use stock and the total collection flows resulting from the cascade model with available data from FSO and Swico Recycling. As illustrated in Figure 5.5, the cascade model is able to map reasonably well the total in-use stock and the total collection flows, taking into account the large uncertainty of up to ±30% for stocks and ±20% for flows

7.1.4 Tool Development

The modeling of dynamic MFAs with a probabilistic approach to take into account data uncertainty, as well as the evaluation with SEA, required a powerful software tool. As no adequate tool was available, we developed an open source software tool in Python 3, allowing for a generic and flexible model design. The tool applies 'dynamic probabilistic material flow analysis', a combination of dynamic material flow modeling with probabilistic modeling, as proposed by Bornhöft et al[19] and SEA as proposed by Rechberger et al.[20]

The tool allows for simple handling of data input and output and requires no programming knowledge. Model descriptions are specified using a simple data-driven approach: the researcher implements the model via a spreadsheet where all necessary information regarding the system as well as the probabilistic input data are stored. Such model description source files can be uploaded through a web interface, upon which the tool calculates the time series of resulting stocks and flows by means of Monte Carlo simulation, visualizes them and offers the resulting data as a CSV download. With some additional information, the model can also be used to calculate SEA. Drawbacks are that in the input file, a large amount of information has to be stored in an inflexible grid structure, what might lead to large and complex spreadsheets. For large systems and simulations over long time periods, execution times can be considerable.

7.2 Conclusion

7.2.1 Methodological Thread

With our extensive data collection, we provided a basis for a better understanding of the past and current stocks and flows of EE and improved forecasts of future stocks and flows. The resulting data highlight the importance of storage and reuse within the use phase of EE and its associated prolongation of the product lifetime.

The cascade model, built upon the collected data, gives insights into the 'black box' of the use phase and provides important details about internal stocks and flows due to reuse and storage that are often neglected in dynamic MFA studies. The total lifetimes resulting from the cascade model are significantly longer than the service lifetime of new devices and positively skewed. The implementation of average or normally distributed lifetimes is thus a strong simplification which may, in the case of technology change, often result in underestimated phase-out times. As the temporal change of product lifetime could not be confirmed for all device types, the assumption of time-invariant lifetime distributions may be a permissible simplification, depending on the considered product.

The model further reveals the different behaviors of device types and their distribution within the use phase to different stocks and disposal flows. By including various device types, the changing composition of outflows due to technology changes can be accounted for, a capability which could enable recycling system managers to provide appropriate and tailored recycling capacities and technologies. As the model also includes other disposal flows, it facilitates to detect leakages or losses of devices and their incorporated resources that do not reach the collection flow. The validation of results of the cascade model by comparing its results with empirical data shows a better agreement concerning total in-use stocks and collection flows compared to simple models not taking into account reuse and storage.[21]

The cascade model is suitable for any end-user product that is potentially reused and stored after its first service life. However, the data collection for the cascade model is complex, and detailed knowledge on all internal processes is not always necessary. Also from a resource point of view, it is more important that products reach the collection system in the end than to know the exact status of products within the use phase. Therefore, the use phase could be modeled as a 'black box' as in earlier studies, but by adapting the total lifetime distribution function, potential reuse and storage should be taken into account.

By including sinks instead of outflows in the extended MFA system, we are able to show the final destinations of metals and quantify the related stocks and flows. This information allows for early identification of current or future problems regarding resource dissipation. The generic model of the collection, recycling and disposal phase can be customized for any end-user product or substance by simply adding or removing specific processes. The data to determine the flows between the processes, however, have to be assessed for each specific case.

The RSE, resulting from the SEA, proved to be a simple, comprehensible indicator to measure and illustrate the level of dilution or concentration of a metal in an MFA system. The SEA, as a tailor-made evaluation method for MFA, can be applied to any kind of MFA system. In literature, it has only been applied to static MFA. With stock and flow information available over time, the SEA can also be performed for each time step over a longer time period.

The inclusion of input data uncertainty in the form of probability distributions, the Monte-Carlo simulation and the analysis of the resulting probabilistic stocks and flows account for the variable data quality in MFAs in a comprehensive and flexible way. For recycling system managers, recyclers or downstream processors, uncertainties of collection flows could be important additional information to estimate in what ranges the expected flows could fluctuate, and to better plan financing mechanisms or dimension recycling capacities.

Finally, the developed tool provides a powerful package for the simulation and evaluation of probabilistic dynamic MFAs. Due to its generic structure, the tool can be used for the simulation of any kind of deterministic or probabilistic dynamic MFA. As an open source application, the tool can also be further developed and customized according to users' requirements.

7.2.2 Empirical Thread

In the following, the key findings concerning the empirical research questions are outlined for the case of indium, neodymium and gold in end-user EE in private Swiss households. The findings for gold are included as a reference and to facilitate the interpretation of results for indium and neodymium. More detailed results are provided in Chapter 6.

RQ1: Where are the largest stocks of critical metals in the anthroposphere?

For all three considered metals, the largest stocks in the anthroposphere are found in the use phase (indium: 60-90%, neodymium: 50-60%, gold: 50-70% of the totally accumulated metal within the system, including uncertainty ranges) (Figure 6.3). The mass of the stock amounts to 1.7 tonnes ±19% for indium, 39 tonnes ± 13% for neodymium and 4.8 tonnes ± 20% for gold in 2014 in Switzerland.

Indium is mainly stocked in FPD TVs. Neodymium is primarily stocked in desktops and laptops with HDDs. As all device types except headphones include PWBs with some gold content, the gold stock is the most evenly distributed stock among all device types (Figure 6.2). The metal stocks all recorded high growth until 2010 when growth rates slowly started to decrease. The growth rate of the indium stock highly depends on FPD TVs sales. The neodymium stock is expected to decrease in the near future due to the technology change from HDDs to SDDs. The growth rate of the gold stock will further decline due to decreasing gold contents in EE put on the market and the market saturation of most device types.

The stock in the use phase can be divided into the in-use stock and the storage stock. For indium, the storage stock accounts for 17% of the total use phase stock, for neodymium 33% and for gold 30% in 2014. These discrepancies result from different storage behavior regarding different device types. For example, neodymium-containing devices such as desktops and laptops are more frequently stored and kept in storage for a longer time, compared to indium containing FPD TVs. Within the in-use stock, most of the metals are incorporated in new devices.

The second largest stocks are, for indium, disposed slags in Swiss landfills (15-20%), for neodymium, slags from PMS and metal smelters (30-40%) and for gold, the recovered metal (30-35%). These three stocks represent the main fate of the considered metals within the collection, recycling, and disposal phase (Figure 6.3).

From these results, it can be derived that the largest stocks of critical metals in emerging technologies, which have been implemented in significant quantities only for the last two decades, are generally still found in the use phase. As other emerging technologies such as electric vehicles, solar cells or wind turbines are assumed to have longer lifetimes than EE, the share of critical metals found in the in-use stock should be even higher.

RQ2: How suitable are these stocks for urban mining?

The large stocks in the use phase for all three metals indicate that most of the resources that have entered the system could theoretically still be recovered. The recycling potential of the outflows of these stocks is initially lowered by losses of metals in devices disposed of to unknown disposal pathways, exported or sent to MWI. Devices reaching unknown disposal pathways and exported devices may still end up in a collection and recycling system. In addition, some MWIs in Switzerland include a dry bottom ash treatment with precious metal separation. Therefore, gold reaching these three disposal pathways might still be recovered. Indium and neodymium, however, are most likely lost.

Flows reaching the collection and recycling system in 2014 are with 91 kg ±20% for indium, 2800 kg ±14% for neodymium and 332 kg ±20% for gold rather small. As all indium-containing FPDs flow through manual preprocessing, indium-containing LCD modules are all separately dismantled. This is the best available fraction from preprocessing as an input material for indium recovery. The content of indium in LCD modules amounts to 130 to 230 ppm, which lies above contents in primary mines of 1 to 100 ppm.[22–24] A life cycle assessment of primary and secondary production of indium indicated that secondary production after manual preprocessing is favorable to or at least equal to primary production.[22] However, as indium recycling from LCD modules is economically unattractive, there are presently no commercial-scale recovery options available. Though the PMS of Umicore is able to recover indium, they only process indium contained in production waste. Therefore, 90% of all LCD modules are sent to MWI, where indium is irrecoverably dissipated to the slag. Only a few recyclers store indium-containing LCD modules for possible further treatment in the future.

More than 70% of the collected neodymium first reach manual preprocessing, mostly incorporated in desktops, laptops, and phones. The content of neodymium in dismantled HDDs and optical drives (250 - 560 ppm) lies below contents in primary mines (1200 -17600 ppm). This fraction should, therefore, be further physically separated and concentrated before it is sent to metallurgical extraction.[25] The secondary production of neodymium oxide from HDD is clearly preferable to primary production from an environmental point of view.[22] Nevertheless, as an economic neodymium recovery is not yet possible and again, commercial-scale recovery options are not yet established, neodymium in magnets is still sent to mechanical preprocessing after manual dismantling. After mechanical preprocessing, neodymium is transferred to the magnetic steel fraction or the fine fraction and therein reaches a smelting process where it is irrecoverably dissipated to the slag.

Around 84% of the gold incorporated in PWB is sent to manual preprocessing. Manual dismantled PWBs are directly sent to PMS without further treatment or losses. In the mechanical preprocessing, only around 5% of the gold is lost to fractions from where it is not recovered. According to Swiss recyclers, these losses are so low because even if gold is transferred to intermediate fractions that are sent to further treatment, and recyclers are not reimbursed for the gold content of these fractions, eventually gold will find its way to a PMS.[26–29] Gold reaching PMS is recovered with a rate of at least 95%. Overall, 70% of the gold outflow of the use phase is recovered, with highest losses occurring directly after the use phase.

Many factors are in favor of the suitability of the considered metals for urban mining. Compared to other emerging technologies, EE has a relatively short lifetime[30] which results in a small but still significant outflow. Losses directly after the use phase occur, but the majority of metals reach the recycling scheme. Thereof again most metals reach manual dismantling, where they are separated and concentrated without further losses. The large share of manual dismantling also fulfills the demand for a product-centric recycling approach, as manual processes are tailored to different products and can be easily adapted in case of technology changes. Recovery processes exist on laboratory or pilot scale and are favorable or at least equal to primary production in terms of environmental impact. The suitability for urban mining thus highly depends on economic factors. Based on current metal prices, the value of gold is around 600 times higher than the value of neodymium and around 100 times higher than the value of indium, which makes its recovery very attractive for all stakeholders in the recycling scheme. As long as economic incentives for the recovery of indium and neodymium are missing, they will further be irrecoverably dissipated.

These results do not apply to all critical metals in emerging technologies. Disposal pathways for different emerging technologies may vary greatly. Separation and concentration of critical metals may be more or less challenging, depending on whether critical metals are concentrated to a few components or spread across many components within a product. For example, neodymium magnets in wind turbines are easily separated and recovered, while a car contains between 60 and 200 magnets in many different applications.[30] Finally, not for all critical metals, recovery processes have yet been developed. However, for critical metals with a high economic value, for example, platinum group metals (PGM), separation, concentration and recovery processes are already well established.

RQ3: Where are the current sinks of critical metals that are not recovered?

The most important current sinks also form the second largest stocks and are, as already stated above, for indium, disposed of slags in Swiss landfills. A small amount of indium from displays in phones ends up in slags used for construction. For neodymium, the largest sinks are slags from PMS and metal smelters. Although before Swico Recycling was established in 1994, all metals incorporated in EE are assumed to reach Swiss landfills, the share of neodymium in Swiss landfills out of the total amount within the system is small. An even smaller amount of neodymium ends up in slags disposals after incineration abroad (Figure 6.3).

Within Switzerland, slags from MWIs are disposed of in specified landfill sites. According to data from a Swiss MWI, the content of indium in the slag is similar to very low-grade zinc deposits.[24,31] These data, however, are very uncertain and it is questionable whether the recovery of indium from slags in landfills would ever be economically viable. Slags from PMS and metal smelters are often used for construction, either within landfills or, for example, for the reinforcement of dams. Neodymium within smelter slag is thus further dissipated to the environment in contents well below primary mines.

The result, that critical metals either end up in Swiss landfill disposals or slags from different metal smelters abroad, depending on the disposal and recycling pathways of the products they are incorporated in, could also apply to other critical metals that are not yet recovered.

RQ4: How are critical metals diluted in the anthroposphere and how does this change during a critical metals life cycle?

The dilution of critical metals in the anthroposphere and its change over time was analyzed based on SEA and quantified with the RSE (Figure 6.4). Due to low contents of indium, neodymium and gold within EE, these metals are already highly diluted when the devices are put on the market and get even further diluted when they are in the stock. Compared to indium and neodymium, gold is the least diluted metal in EE, with reference to the earth crust content. In 1990, before an e-waste collection system was established, EE was sent to MWIs and metals were dissipated to the slag which was disposed of in landfills. This results in low metal contents (for indium and neodymium below the earth crust content) and high RSE. Between the years 2000 and 2014, within the collection, recycling, and disposal phase, metals are concentrated, as already described in *RQ2*. As within this time period, the system has not been significantly altered, the RSE changes over time only depend on the share of EE with higher metal contents. Indium is concentrated to the level of dismantled LCD panels, neodymium to dismantled HDDs or optical drives and gold to dismantled PWBs or precious metal-containing fractions after mechanical preprocessing.

However, the decrease in RSE is reversed for indium after the manual preprocessing, when LCD panels are sent to incineration and indium is dissipated to the slag. For neodymium, after separation, HDDs and optical drives are sent to mechanical treatment with other EE where the previously concentrated neodymium is again diluted and finally, after treatment in a smelter, dissipated to the slag. The Entropy of the slag is similar to the entropy of the earth crust. As gold is recovered, the RSE shows a sharp drop after the PMS. Due to gold losses in the system, the RSE does not fall to zero.

The SEA illustrates how the current collection and recycling system not only successfully concentrates gold but also indium and neodymium, mostly because of manual dismantling that is either required due to depollution targets or done voluntarily due to valuable fractions in EE. As mentioned in *RQ2,* due to a lack of economic incentives and missing commercial-scale recovery options, these concentration efforts are not taken advantage of for the recovery of indium and neodymium. O'Rourke et al. stated that in order to create closed material cycles, high-entropy wastes have to be refined back into low entropy or recovered material.[32] Regarding critical metals, this demand is not yet met in the Swiss EE recycling system.

For other critical metals in EE, similar results of a SEA could be expected, depending on the component other critical metals in EE are incorporated in and what pathway within the preprocessing they take. Regarding other emerging technologies, concentration efforts in the collection and recycling system may vary greatly as already stated above. Therefore, our results concerning the dilution of critical metals within the anthroposphere over their life cycle may only apply for technologies where critical metals are locatable and available in small quantities in known components.

RQ5: Where are possible starting points to improve the current recycling system?

Our results show, that the current recycling system in Switzerland with a large share of manual preprocessing offers favorable conditions for the recovery of critical metals. However, the small quantities of critical metals in EE combined with low prices discourage Swiss recyclers to adapt their preprocessing and find appropriate downstream processes to recover indium and neodymium. At the same time, these conditions also provide little incentives for downstream processors to establish commercial-scale recovery processes for indium and neodymium.

Starting points to improve the current system are technological, economic and regulatory adaptations as outlined in the following:

For indium, the manual dismantling of LCD modules already constitutes the best available preprocessing and no technological adaptations are necessary. Due to the technology change from mercury-containing cold cathode fluorescent lighting (CCFLs) to mercury-free light-emitting diodes (LEDs), in future, special treatment of FPDs might no longer be required and manual dismantling should, therefore, be fostered with other incentives. For neodymium, after the manual dismantling of HDDs and optical drives, further physical separation and concentration is necessary in order to provide suitable concentrates from which neodymium can be recovered.[25] Hence, also for EE containing neodymium magnets, manual dismantling should be fostered, and, for the resulting fractions, suitable separation and concentration technologies should be established. Until the establishment of commercial-scale recovery processes, storage of LCD modules and neodymium magnets or magnet containing fractions would also be a viable option to prevent resource losses. As metal quantities in fractions resulting from manual dismantling are very low, both further physical treatment and metallurgical recovery should take into account economy of scale in order to reach sufficient input quantities.

As their prices currently do not match recycling costs, the recovery of indium and neodymium should be fostered through economic incentives. The Swiss recycling system is financed by an advanced recycling fee, paid by the consumer upon purchase of a new device. The fee ranges from 0.2 CHF for smartphones to 28 CHF for large FPD TVs. An economic feasibility study investigating the Swiss recycling system has shown that indium recycling could be covered financially with a marginal increase of the advance recycling fee by maximum 0.2 CHF/product.[22] For neodymium, no such study exists, but it is assumed that a required increase would not be substantially higher. Thus, a slight increase of the advanced recycling fee could provide sufficient funds to encourage recyclers and downstream processors to adapt and establish the necessary infrastructure for the recovery of indium and neodymium.

Recycling systems, recyclers and downstream processes could also be forced to improve the current system by stricter regulations. To date, recovery targets are related to the total mass of EE of a certain WEEE category and are often met through the recycling of base metals and plastics. Specific recovery targets for individual materials have not yet been established but would play an important role to close the material cycles.

7.3 Outlook

The research presented in this thesis could be continued in various directions.

To improve the data basis for the cascade model, quantitative data collection regarding service lifetime, storage time and disposal pathways should be extended by including qualitative questions about the reasoning behind consumer decisions. This would include, for example, reasons for replacing, storing or disposing of a device to a certain disposal pathway. Such qualitative information would facilitate the interpretation of existing or newly collected quantitative data. As for metals that are recovered in the recycling phase, the largest losses occur after the use phase due to unknown disposal pathways, it would be important for future research to break down and analyze disposal pathways in more detail in order to identify device types and incorporated materials that need special attention to improve collection rates.

The data basis for modeling the collection, recycling, and disposal phase could be improved by more studies regarding the fate of critical metals within preprocessing. Existing studies and information provided by recyclers reveal large discrepancies among preprocessing facilities. With a harmonized approach as proposed by Ueberschaar et al.[12], results would be more comparable among recycling facilities, but also among different countries, where other prerequisites such as regulations etc. exist.

The dynamic MFA developed within this thesis provides a sound basis for scenario analyses of current and future stocks and flows. The presented analysis of future neodymium flows only provides one possible scenario. In addition to technology changes, the effects of new technological, financial or regulatory measurements could be tested with more elaborate scenario analyses including the possible time variance of lifetime distribution parameters and transfer coefficients. Future stocks and flows can be calculated either based on extrapolated inflows or stocks. As our simulation tool is currently not able to compute inflows and outflows from stock data, we recommend extensions in this regard for future work.

The overarching goal of this thesis was to provide a basis for improving current recycling systems and encourage the recycling and reintegration of critical metals into anthropogenic material cycles. In current recycling processes, most critical metals are irrecoverably dissipated into recovered base metals or slags. Future research should thus contribute to the improvement and upscaling of technological solutions to separate, concentrate and recover critical metals. Appropriate financing mechanisms should be developed to pay for economically unviable recovery processes. Last but not least, regulations should be adapted from mass related recovery targets to more resource specific recovery targets.

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Appendix A

Supporting Information to Chapter 3

This appendix was published as supporting information for the published paper:

Müller, E.; Hilty, L. M.; Widmer, R.; Schluep, M.; Faulstich, M. Modeling Metal Stocks and Flows: A Review of Dynamic Material Flow Analysis Methods. Environ. Sci. Technol. 2014, 48 (4), 2102–2113

A.1 Results

Table A.1: Processes covered by the reviewed literature

Reference	Covered Processes Primary Mining	Production/ Fabrication	Use	Waste management	Landfill/ Environment
Elshkaki and Van Der Voet, 2006; Glöser et al., 2013; Izard and Müller, 2010; Kavlak and Graedel, 2013; Liu and Müller, 2013; Liu et al., 2013, 2011; Müller et al., 2006; Pauliuk et al., 2012, 2013a, 2013b; Yan et al., 2013; Yoshimura et al., 2013; Zeltner et al., 1999)	X	X	X	X	X
Bader et al., 2011; Chen et al., 2010; Chen and Graedel, 2012; Dahlström et al., 2004; Daigo et al., 2010, 2009; Elshkaki et al., 2004; Harper and Graedel, 2008; Harper et al., 2012; Mao and Graedel, 2009; Matsuno et al., 2012; Müller et al., 2011; Spatari et al., 2005; Tabayashi et al., 2009; Yamaguchi and Ueta, 2006		X	X	X	X
Elshkaki et al., 2005	X	X	X	X	
Cheah et al., 2009; Davis et al., 2007; Du and Graedel, 2011; Geyer et al., 2007; Igarashi et al., 2007; Marwede and Reller, 2012; Oda et al., 2010; Park et al., 2011; Ruhrberg, 2006		X	X	X	
Michaelis and Jackson, 2000a, 2000b	X	X	X		
Hedbrant, 2001			X	X	X
Ayres et al., 2002		X	X		
Yano et al., 2013; Zuser and Rechberger, 2011			X	X	
Alonso et al., 2012; Daigo et al., 2007; Gerst, 2009; Hatayama et al., 2012, 2010, 2009, 2007; Hirato et al., 2009; Hu et al., 2010; Igarashi et al., 2008;			X		

Reference	Covered Processes				
	Primary Mining	Production/ Fabrication	Use	Waste management	Landfill/ Environment
Kapur, 2006; McMillan et al., 2010; Melo, 1999; Saurat and Bringezu, 2009					

Note: Green filling = covered in study

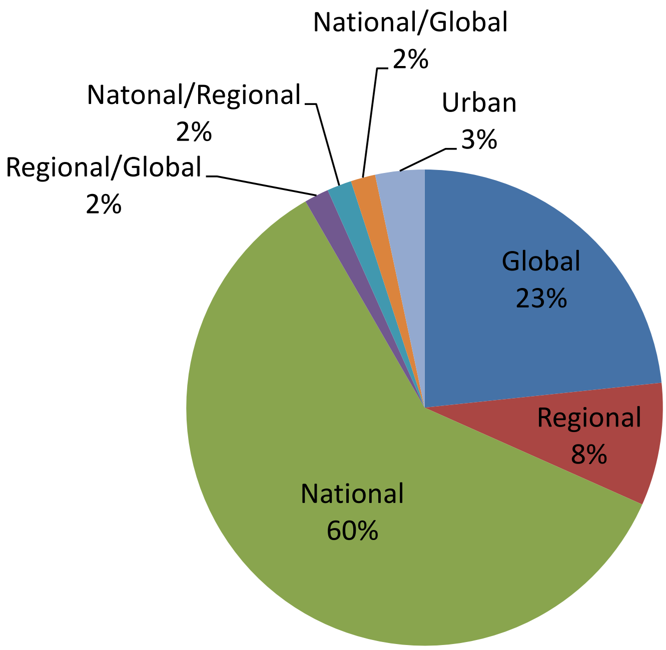


Figure A.1: Percentage distribution of the reviewed studies by spatial extent

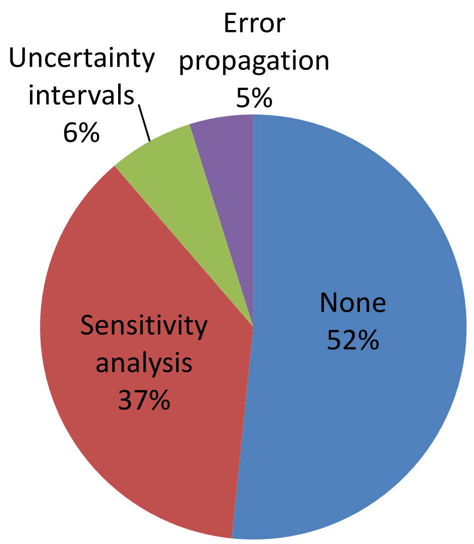


Figure A.2: Percentage distribution of the reviewed studies by treatment of data uncertainty.

A.2 Stock and Flow Model

The in-use stock $Stock(t)$ equals the netflow, that is, inflow minus outflow (equation (1) in the main article). In a time discrete form, where a time dependent variable $x(t)$ is only known for specific times and often given as a time series with a constant sampling time T, that is, $x[n] = x(nT)$, $Stock[n]$ at any time n equals the stock of the previous period $Stock[n\text{-}1]$ plus the netflow during that period of sample time T (equation (1a) in the main article). Or in an integral form according to equation (1b), the in-use stock $Stock[N]$ at a time N equals the initial value $Stock[0]$ plus the cumulated net flow over N sample times T (see Figure A.3).

$In\ (t)$, $In[n]$ →

$$dStock(t) = \big(In(t) - Out(t)\big) \cdot dt = Net(t) \cdot dt$$
$$Stock[n] + Stock[n-1] = (In[n] - Out[n]) \cdot T$$
$$Stock[N] = Stock[0] + T \cdot \sum_{n=0}^{N} (In[n] - Out[n])$$

→ $Out\ (t)$, $Out[n]$

Figure A.3: Graphical illustration of the generic stock-flow model (Sinha-Khetriwal et al., 2013). It assumes a causal system where outflows depend only on inflows at equal or earlier times and that inflows are zero for $t < 0$, thus $n \geq 0$.

Outflow $Out(t)$ of products from the $Stock(t)$ can be described as a function of the products inflow $In(t)$ and of their residence time in the stock, given as a survival or reliability function $R(t)$. Such a delay model assumes that the unique event triggering the release of products from stock into the outflow is its terminal failure, given as the probability density $f(t) = F'(t)$ of the life time distribution $F(t) = 1 - R(t)$ (see Table A.2).

The frequently used quantitative measures to describe the failure process are (Rausand and Arnljot, 2004):

- The reliability distribution function $R(t)$, also named survival or survivor function $S(t)$. The reliability function is the probability that the time of failure is later than some given time t.
- The lifetime distribution function $F(t)$ is the probability that the time of failure is sooner than some given time t, therefore $F(t) = 1 - R(t)$. $F(t)$ has the probability density $f(t) = F'(t)$ which is also called event density. Similarly, $R(t)$ has the probability density $R'(t) = r(t) = -f(t)$.
- The instantaneous failure rate $h(t)$, also known as hazard function, is the probability that the time of failure is at time t. It is defined as $f(t)/R(t)$ and is neither a density nor a probability, however, one could think of it as the probability of failure in the time interval $[t, t + dt]$, given that the item has survived until time t.

The term 'failure' is often used synonymously for 'discard' or 'obsolescence' implying that the reason for releasing an item from the stock is not necessarily its technical failure. The process describing the generation of outflow and the accumulation of stocks is shown in Figure A.4.

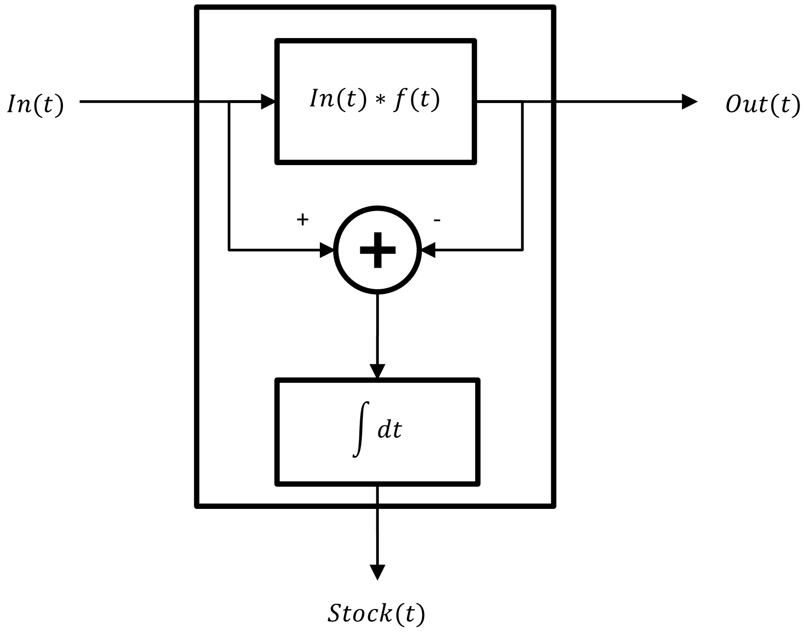
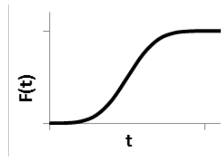
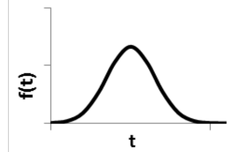
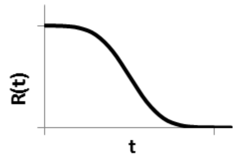
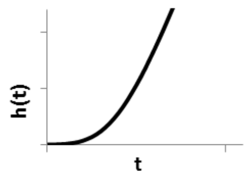


Figure A.4: Stock and flow diagram for the delay model (with '*' denoting a convolution). It is obvious that in this model the stock does not influence the outflow.

Table A.2: Relationship between the functions F(t), f(t), R(t) and h(t) (Rausand and Arnljot, 2004)

Expressed by	$\boldsymbol{F(t)}$	$\boldsymbol{f(t)}$	$\boldsymbol{R(t)}$	$\boldsymbol{h(t)}$	Graphical example
$\boldsymbol{F(t)} =$	-	$\int_0^t f(u)du$	$1 - R(t)$	$1 - exp\left(-\int_0^t h(u)du\right)$	
$\boldsymbol{f(t)} =$	$\frac{d}{dt}F(t)$	-	$-\frac{d}{dt}R(t)$	$\lambda(t) \cdot exp\left(-\int_0^t h(u)du\right)$	
$\boldsymbol{R(t)} =$	$1 - F(t)$	$\int_0^\infty f(u)du$	-	$exp\left(-\int_0^t h(u)du\right)$	
$\boldsymbol{h(t)} =$	$\frac{dF(t)/dt}{1 - F(t)}$	$\frac{f(t)}{\int_t^\infty f(u)du}$	$-\frac{d}{dt}lnR(t)$	-	

Note: F(t) = lifetime distribution function, f(t) = probability density of the lifetime distribution function or event density, R(t) = reliability distribution function, h(t) = instantaneous failure rate or hazard function